

GLOBAL MONITORING PLAN FOR PERSISTENT ORGANIC POLLUTANTS

UNDER THE STOCKHOLM CONVENTION ARTICLE 16
ON EFFECTIVENESS EVALUATION

3RD REGIONAL MONITORING REPORT WESTERN EUROPE AND OTHERS GROUP (WEOG) REGION

2021 (March 29, 2021_rev1)

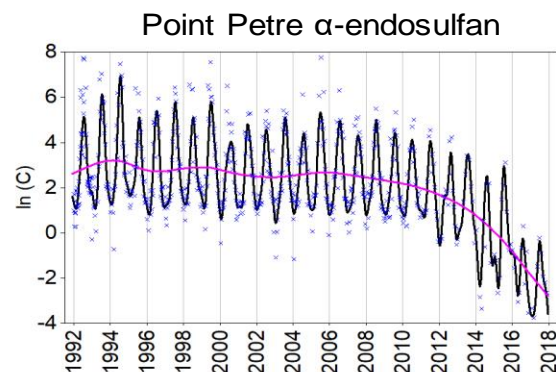
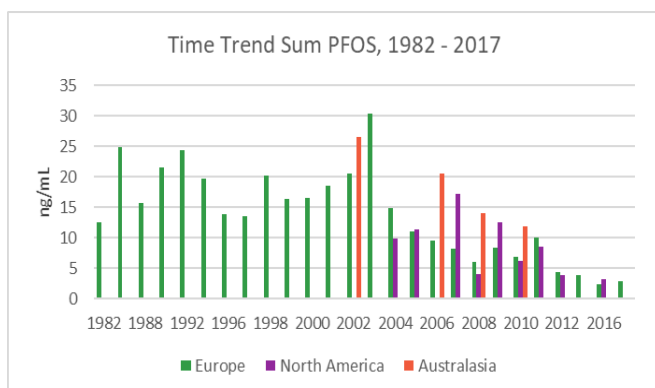
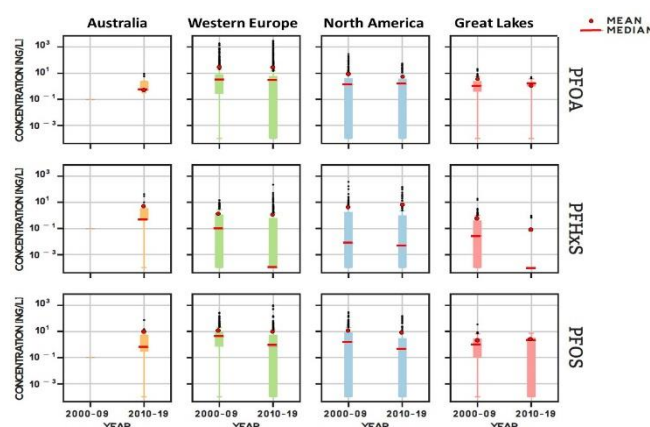
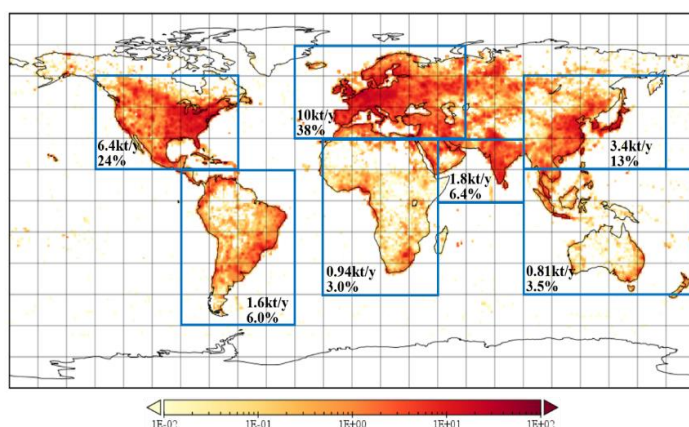


TABLE OF CONTENTS

ACKNOWLEDGEMENTS	3
PREFACE	6
ABBREVIATIONS AND ACRONYMS	7
GLOSSARY OF TERMS	12
EXECUTIVE SUMMARY	14
0.1 Contributing programs	14
0.2 Key messages from the data on baseline concentrations and temporal trends ...	14
0.3 Long range transport	16
0.4 Description of data gaps and capacity building needs	17
0.5 Future evaluations	17
0.6 Conclusions and recommendations	18
0.6.1 Media specific conclusions and recommendations	18
0.6.2 Generic conclusions and recommendations	21
1 Introduction	24
2 Description of the Region	25
2.1 Background	25
2.2 Political	26
2.3 Geographical	26
2.4 Social and economic considerations	27
2.5 Considerations in relation to sampling strategies	28
3 Organization of regional implementation	29
3.1 Organization of activities	29
3.2 Strategy to locate and collect information from existing programmes	29
3.3 Identified data gaps and strategies to fill them, including strategies and activities to generate monitoring data	30
3.4 Capacity strengthening needs	30
4 Methods for sampling, analysis and handling of data	31
4.1 Background	31
4.1.1 Key Message regarding “comparable data” from existing programmes and activities	31
4.1.2 Availability of information to allow data to be independently evaluated .	32
4.2 Description of national and international programmes and activities that have contributed information for the regional report	32
4.2.1 Programmes/activities related to air monitoring	34
4.2.2 Programmes/activities related to human tissues (milk and blood)	37
4.2.3 Programmes/activities related to water	38
4.2.4 Programmes/activities related to other media	38
4.3 Antarctica	39
4.4 Data handling and preparation for the regional monitoring report	39
4.5 Preparation of the monitoring reports	40
5 Results	41
5.1 The results in context	41
5.2 Review of concentrations and their changes over time in the regions	41
5.2.1 Ambient air	41

5.2.2	Human tissues (milk and/or blood)	86
5.2.3	Water	159
5.2.4	Other media / non-core GMP media	182
5.3	Long-Range Transport Modeling	215
6	Conclusions and recommendations.....	235
6.1	Findings and Conclusions	235
6.1.1	Summary of the baseline concentrations.....	235
6.1.2	Summary of evidence of temporal trends	238
6.1.3	Summary of evidence of long range transport, POPs modelling capabilities and the effects of climate change.....	245
6.1.4	Summary of gaps in data coverage	247
6.1.5	Summary of new developments in POPs monitoring	248
6.2	Recommendations for the future	249
7	References.....	256
	Database structure.....	Error! Bookmark not defined.
8	Annex	285

ACKNOWLEDGEMENTS

The WEOG regional organizational group (ROG) for the third evaluation was composed of the following six experts: Katrine Borgå (Norway); Sara Broomhall (Australia); Ramon Guardans (Spain); Tom Harner (Canada); Peter Korytar (representing the European Union) and Linda Linderholm (Sweden). Tom Harner acted as coordinator for the group and rapporteur for teleconferences.

The key drafting responsibilities were as follows and included additional regional/international experts: Chapter 5 – 5.2.1 Air (Hayley Hung, supported by Desmond Ng); 5.2.2 Human tissues (Linda Linderholm, supported by Staffan Åkerblom, Anna-Karin Dahlberg, Johan Fång, Minh Nguyen, Kerstin Putz, and Elin Stenfors); 5.2.3 Water (Derek Muir, supported by Luc Miaz); 5.2.4 Other Media (Katrine Borgå / Ramon Guardans, supported by Malin Røyset Aarønes); and 5.3 Long range transport (Matthew MacLeod, supported by Michael McLachlan, Li Li, Jing Li, Xianming Zhang, Elsie Sunderland, and John Hader); 6 Conclusions and Recommendations (Tom Harner, supported by WEOG chapter leads). Simon Wilson (AMAP) contributed to discussions of the WEOG experts group and coordination of AMAP data that were made available for the report.

This review could not have been completed without the full cooperation and assistance of the existing programmes that have contributed information and data. Although it is

difficult to draw attention to individual programmes, there is no doubt that a WEOG evaluation based upon existing monitoring arrangements would not have been feasible without the richness of the data sets afforded by the long-established international activities of AMAP, EMEP, and WHO and the generous cooperation provided by their organizers.

Additional thanks are due to the individuals who provided “Programme Summaries” on the activities and results from these programmes. These include:

For air: Fiona Wong and Hayley Hung (Canada), Simon Wilson (AMAP Secretariat) - AMAP air monitoring trends [with data input from Wenche Aas and Pernilla Bohlin-Nizzetto (Norway); Michelle Nerentorp Mastromonaco and Eva Brorström Lundén (Sweden); Kristín Ólafsdóttir and Árni Sigurðsson (Iceland); Katrin Vorkamp, Rossana Bossi and Henrik Skov (Denmark); Hannele Hakola (Finland); Hayley Hung, Fiona Wong and Helena Dryfhout-Clark (Canada)]; Susan Bengtson Nash (Australia) - Australia's Casey Station; Fisher Xianyu Wang, Chris Paxman, Jochen Mueller - Australia's National Passive Air Sampling and Archiving Program; Pernilla Bohlin-Nizzetto and Knut Breivik (Norway) - UNECE-EMEP; Tom Harner, Amandeep Saini, Jasmin Schuster - GAPS; Hayley Hung, Nick Alexandrou, Helena Dryfhout-Clark, Fiona Wong (Canada) - Great Lakes Basin (GLB) Monitoring and Surveillance Program; Marta Venier, Amina Salamova, Ron Hites (US) - Integrated Atmospheric Deposition Network (IADN); Roman Prokeš, Jiří Kalina, Jana Borůvková, Petra Přibyllová and Jana Klánová (Czech Republic) - MONET EU; Korbinian Freier, Gabriela Ratz, Wolfgang Körner, Ulrich Waller (Germany), Monika Denner, Peter Weiss, Wolfgang Moche (Austria), Manfred Kirchner, Gert Jakobi, Karl-Werner Schramm (Germany) - PureAlps; Pernilla Bohlin-Nizzetto, Knut Breivik (Norway) - Norwegian Troll Station; Ramon Guardans, Begoña Jiménez (Spain) - Spanish Monitoring Program on POPs, Linda Linderholm (Sweden) - Swedish National Monitoring Programme for Air; Andrew Sweetman (UK) - TOMPs; Carola Graf, Andrew Sweetman (UK) - UK/Norway SPMD Transect.

For human tissues: The authors would like to acknowledge the people who did the field work and the analytical work in labs that made the chapter on human exposure possible to write. We would also like to thank the managers of the regional and national monitoring programs for making the data available to us.

For water: The preparation of the water chapter was enabled by financial support from the King Carl XVI Gustaf Professorship in Environmental Science held at Stockholm University by DCGM in 2018-19. We thank Cynthia de Wit (Stockholm University) for coordinating the financial and technical support for the work. Thanks to the following persons for providing unpublished data: Karl Bowles (Australian Dept of Defence), Chad Furl and Callie Meredith (Washington State Dept of Ecology), Roland Kallenborn (Norwegian University of Life Sciences), Suzanne Vardy (Queensland Dept of Environment and Science). We also thank Dr Nobuyoshi Yamashita (National Institute of Advanced Industrial Science, and Technology, Tsukuba Japan) for seawater data. Special thanks to Kateřina Šebková.

For other media: We thank Dr. Simonetta Corsolini for making available her summary on work from Antarctica, Hayley Hung for help with the latest SOLEC report from the Great

Lakes, Luis Roscales (CSIC/IQOG, Spain) for review of data on the Mediterranean, and Frank Riget and Simon Wilson for help with updated AMAP time trends until 2018 for Greenland.

The worldwide implementation of the third phase of the Global Monitoring Plan was made possible thanks to the generous contributions to the Stockholm Convention Voluntary Trust Fund from the European Commission's the Global Public Goods and Challenges (GPGC) Thematic Programme, contributions by the monitoring programmes and engagement of experts within the regional organization groups and the global coordination group. Further, the contribution of the projects to support POPs monitoring activities in regions, funded through the Global Environment Fund (GEF) and implemented by the United Nations Environment Programme (UNEP) Chemicals and Health Branch is greatly acknowledged.

Air monitoring activities are implemented in the five UN regions in cooperation with strategic partners: the Arctic Monitoring and Assessment Programme (AMAP), the Global Atmospheric Passive Sampling (GAPS) Network, the East Asia Air Monitoring Program, the European Monitoring and Evaluation Programme (EMEP), Great Lakes Basin Monitoring and Surveillance program, the Integrated Atmospheric Deposition Network (IADN) and the MONET Programme of the Research Centre for Toxic Compounds in the Environment (RECETOX).

The human milk survey draws on the collaboration between the Secretariat of the Stockholm Convention, the United Nations Environment Programme (UNEP) Division of Technology, Industry and Economics (DTIE) Chemicals Branch and the World Health Organization (WHO). The State Institute for Chemical and Veterinary Analysis of Food (CVUA), Freiburg, Germany, is acknowledged for the analytical work related to human milk samples. The MTM Research Centre, Örebro University, Sweden, is acknowledged for the analysis and provision of data on perfluorinated chemicals in human milk. Thanks are also expressed to the national coordinators of the joint WHO/UNEP exposure study for the work to collect and process the human milk samples.

PREFACE

Persistent organic pollutants (POPs) are a group of chemicals that have toxic properties, resist degradation in the environment, bioaccumulate through food chains and are transported long distances through moving air masses, water currents and migratory species, within and across international boundaries. POPs belong to three main groups, however some of the chemicals fit into more than one of these three general categories:

- pesticides used in agricultural applications¹
- industrial chemicals used in various applications²
- chemicals generated unintentionally as a result of incomplete combustion and/or chemical reactions³.

Twelve POPs were initially listed in the Stockholm Convention (shown in bold font in footnotes 1-3). In general, these ‘legacy’ POPs were first produced and/or used several decades ago, their persistence, bioaccumulative properties and potential for long-range transport are well studied, and they have been globally banned or restricted since 2004. Eighteen additional chemicals have been listed in the Annexes of the Convention since, bringing the total number of POPs to thirty chemicals and/or groups of chemicals, as of January 2020 (the meetings of the Conference of the Parties at which the listing of the chemicals took place are indicated in parenthesis in footnotes 1-3).

Article 16 of the Stockholm Convention requires the Conference of the Parties to evaluate periodically whether the Convention is an effective tool in achieving the objective of protecting human health and the environment from persistent organic pollutants. This evaluation is based on comparable and consistent monitoring data on the presence of POPs in the environment and in humans, as well as information from the national reports under Article 15 and non-compliance information under Article 17. The global monitoring plan for POPs, which has been put in place under the Convention, is a key component of the effectiveness evaluation and provides a harmonized framework to

1

aldrin, chlordane, chlordane (COP-4, 2009), **dichlorodiphenyltrichloroethane (DDT)** (COP-4, 2009), **dieldrin**, endosulfan (COP-5, 2011), **endrin, heptachlor, hexachlorobenzene (HCB)**, gamma-hexachlorocyclohexane (γ -HCH, lindane) and by-products of lindane [α -hexachlorocyclohexane (α -HCH) and beta-hexachlorocyclohexane (β -HCH)] (COP-4, 2009), pentachlorophenol, its salts and esters (COP-7, 2015) **mirex, toxaphene**, dicofol (COP-9, 2019).

² tetra- and pentabromodiphenyl ethers (PBDEs) (COP-4, 2009), hexa- and heptabromodiphenyl ethers (PBDEs) (COP-4, 2009), decabromodiphenyl ether (COP-8, 2017), hexabromocyclododecane (HBCD) (COP-6, 2013), hexabromobiphenyl (COP-4, 2009), hexachlorobutadiene (COP-7, 2015), perfluorooctane sulfonic acid (PFOS), its salts and perfluorooctane sulfonyl fluoride (PFOS-F) (COP-4, 2009), perfluorooctanoic acid (PFOA), its salts and PFOA-related compounds (COP-9, 2019), pentachlorobenzene (PeCB) (COP-4, 2009), **polychlorinated biphenyls (PCBs)**, polychlorinated naphthalenes (PCN) (COP-7, 2015), short-chain chlorinated paraffins (SCCPs) (COP-8, 2017).

³ **hexachlorobenzene (HCB)**, hexachlorobutadiene (COP-8, 2017), pentachlorobenzene (PeCB) (COP-4, 2009), polychlorinated naphthalenes (PCN) (COP-7, 2015), **polychlorinated biphenyls (PCBs)** and **polychlorinated dibenzo-*p*-dioxins (PCDDs) and dibenzofurans (PCDFs)**.

identify changes in concentrations of POPs over time, as well as information on their regional and global environmental transport.

While monitoring activities are ongoing in the frame of the GMP, every sixth year the information generated is collected, compiled and analyzed in monitoring reports (regional and global). The first two phases of the GMP have been implemented during the period 2004-2017, with two sets of regional monitoring reports and global reports developed to date in the frame of the GMP and have informed the effectiveness evaluation under Article 16 of the Convention. The GMP Data Warehouse has been made operational during the second GMP phase and continued to support the regional organization groups in the work for the collection, processing, storing and presentation of monitoring data during the third phase of implementation of the GMP.

The present (third) monitoring report is synthesizing information from the first, the second and the third phase of the global monitoring plan and presents the most up-to-date findings on POPs concentrations in the Western Europe and Others Group (WEOG) Region. While the first and the second monitoring reports, presented at the fourth and seventh meeting of the Conference of the Parties respectively, provided information as to the changes in concentrations of the chemicals initially listed in the Convention, as well as baseline information on some of the newly listed POPs, this third report builds on the increasing information base of POPs monitoring data and provides a further in-depth assessment of the changes measured over time in POPs concentrations, including time trends where available, as well as recent baseline information on the more recently listed POPs.

ABBREVIATIONS AND ACRONYMS

ACP	Arctic Contamination Potential
ADI	Acceptable Daily Intake
AFN	Assembly of First Nations
AGAM	Australian Pilot Monitoring Programme
ALRT	Atmospheric Long Range Transport
AMAP	Arctic Monitoring and Assessment Programme
ANCOVA	Analysis of Covariance
ANOVA	Analysis of Variance
APEs	Alkylphenol Ethoxylates
BCF	Bioconcentration Factor
BFR	Brominated Flame Retardants
BHC	Benzenehexachloride
BPH	Benzo(a)pyrene oxidation
CBz	Chlorobenzenes
CCC	Chemical Coordinating Centre
CCEHIP	Coordinating Center for Environmental Health and Injury Prevention
CDC	Centers for Disease Control and prevention
CEE	Central and Eastern Europe
CEMP	Coordinated Environmental Monitoring Programme

CEP	Caspian Environment Programme
CHL	Chlordane-related Compounds
CHMS	Canadian Health Measures Survey
CLRTAP	Convention on Long Range Transboundary Air Pollution
CRM	Certified Reference Material
CIS	Commonwealth of Independent States
COP	Conference of the Parties
CTD	Characteristic Travel Distance
CUPs	Current Use Pesticides
CV	Coefficient of Variation
DDD /DDE	Metabolites of DDT
DDT	Dichlorodiphenyltrichloroethane
DLPCBs	Dioxin-like PCBs
EAC	Ecological Assessment Criteria
ECA	Environmental Contamination in Antarctica
ECF	Electrochemical Fluorination
EDCs	Endocrine Disrupting Chemicals
EMAN	Ecological Monitoring and Assessment Network
EMEP	Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe
EPA	(US) Environmental Protection Agency
EPER	European Pollutant Emission Register
ERL	Effects Range Low
ERM	Effects Range Median
EROD	7-ethoxyresorufin-O-deethylase
ESB	Environmental Specimen Bank
EtFOSA	Ethyl Perfluorooctane Sulfonamide (sulfluramid)
EtFOSE	Ethyl Perfluorooctane Sulfonamidoethanol
EUSES	European Union System for the Evaluation of Substances
FAO	Food and Agriculture Organisation of the United Nations
FERTIMEX	Fertilizantes Mexicianos, S.A.
FNBI	First Nations Biomonitoring Initiative
FOSA	Perfluoro-octanesulfonamide
FOSE	Perfluorooctane Sulfonamidoethanol
FTOH	Fluorotelmer Alcohol
GAPS	Global Atmospheric Passive Sampling (Network)
GAW	Global Atmospheric Watch
GEF	Global Environment Facility
GEMS	Global Environment Monitoring System
GerES	German Environmental Survey
GEOS	Global Earth Observation Systems
GLBTS	Great Lakes Bi-national Toxics Strategy
GLWQA	Great Lakes Water Quality Agreement
GLB	Great Lakes Basin Monitoring and Surveillance (Air Monitoring Program)
GMP	Global Monitoring Plan

HBB	Hexabromobiphenyl
HBCD	Hexabromocyclododecane
HCB	Hexachlorobenzene
HELCOM	Helsinki Commission/The Baltic Marine Environment Protection Commission
HCHs	Hexachlorocyclohexanes
HHAG	Human Health Assessment Group
HIPS	High Impact Polystyrene
HPLC	High Performance Liquid Chromatography
HRGC	High Resolution Gas Chromatography (capillary column)
HRMS	High Resolution Mass Spectrometer
HTAP	Hemispheric Transport of Air Pollutants
HxBB	Hexabromobiphenyl
IADN	Integrated Atmospheric Deposition Network
IARC	International Agency for Research on Cancer
ICES	International Council for the Exploration of the Sea
IFCS	Intergovernmental Forum on Chemical Safety
ILS	Interlaboratory Studies
IMO	International Maritime Organisation
INSPQ	Centre de Toxicologie du Québec
INFOCAP	Information Exchange Network on Capacity Building for the Sound Management of Chemicals
IPCC	Intergovernmental Panel on Climate Change
IPPC	Integrated Pollution Prevention and Control
I-TEQ	International Toxicity Equivalence
JGOFS	Joint Global Ocean Flux Study
KAW	Air/Water Partition Coefficient
KOA	Octanol/Air Partition Coefficient
Kow	Octanol/Water Partition Coefficient
LC50	Median Lethal Concentration
LC-MS/MS	Liquid Chromatography-tandem Mass Spectrometry
LD50	Median Lethal Dose
LOAEL	Lowest Observable Adverse Effect Level
LOD	Limit of Detection
LOQ	Limit of Quantification
LRT	Long Range Transport
LRTAP	Long Range Transboundary Air Pollution
L RTP	Long Range Transport Potential
MAP	Mediterranean Action Plan
MDL	Minimum Detectable Level
MEDPOL	Mediterranean Pollution Monitoring and Research Programme
MEA	Multi Lateral Environmental Agreements
MeFOSA	Methyl perfluorooctane sulfonamide
MeFOSE	Methyl perfluorooctane sulfonamidoethanol
MONARPOP	Monitoring Network in the Alpine Region for Persistent and other Organic Pollutants

MONET	MOonitoring NETwork (air monitoring network operated by RECETOX)
MORS	Monitoring Of Radioactive Substances
MRL	Maximum Residue Limit
MSCE-East	Meteorological Synthesizing Centre-East
MSFD	Marine Strategy Framework Directive
NAFTA	North American Free Trade Agreement
NAPS	National Air Pollution Surveillance (Canada)
NARAPs	North American Regional Action Plans
NAWQA	National Water-Quality Assessment Program
NCEH	National Center for Environmental Health
NCP	Northern Contaminants Program
ND	Not detected
NGOs	Non-Governmental Organisations
NHANES	National Health and Nutrition Examination Survey
NHATS	National Human Adipose Tissue Survey
NILU	Norwegian Institute of Air Research
NIS	Newly Independent States
NOAA	National Oceanic and Atmospheric Administration
NOAEL	No Observable Adverse Effect Level
NOEL	No Observable Effect Level
NS&T	National Status and Trends Program
NWT	Northwest Territories
OCs	Organochlorines
OCPs	Organochlorine Pesticides
OECD	Organisation for Economic Co-operation and Development
OPs	Organophosphates
OSPAR	Commission for the Protection of the Marine Environment of the North-East Atlantic
PAHs	Polycyclic aromatic hydrocarbons
PAS	Passive Air Sampler
PBDEs	Polybrominated diphenyl ethers
PBT	Polybutylene Terephthalate
PCBs	Polychlorinated biphenyls
PCDDs	Polychlorinated dibenzo- p-dioxins
PCDFs	Polychlorinated dibenzofurans
PCP	Pentachlorophenol
PeCB	Pentachlorobenzene
PFAA	Perfluoroalkyl acids
PFAS	Perfluoroalkyl sulfonates
PFCA	Perfluoroalkyl Carboxylic Acids
PFSA	Perfluoroalkyl Sulfonic Acids
PFOA	Perfluorooctanoic Acid
PFOS	Perfluorooctane sulfonate
PIC	Prior Informed Consent
PLC	Pollution Load Compilation
POPs	Persistent Organic Pollutants

PRTRs	Pollutant Release and Transfer Registers
PTS	Persistent Toxic Substances
PUF	Polyurethane Foam
PureAlps	Monitoring Network in the Alpine Region for POPs
PVC	Polyvinylchloride
QA/QC	Quality Assurance and Quality Control Regimes
QSAR	Quantitative Structure-Activity Relationship
QSPR	Quantitative Structure-Property Relationship
REACH	Registration, Evaluation and Authorisation of Chemicals
RECETOX	Research Centre for Environmental Chemistry and Ecotoxicology
RENPAF	Regional Network on Pesticide Production in Asia and Pacific
ROGs	Regional Organization Groups for the Global Monitoring Plan
ROPME	Regional Organisation for the Protection of the Marine Environment
ROWA	Regional Organisation of West Asia
SAICM	Strategic Approach to International Chemicals Management
SC	Stockholm Convention
SCAR	Scientific Committee of Antarctic Research
SCCPs	Short-chain chlorinated paraffins
SIP	Sorbent impregnated
SCOR	Scientific Committee of Oceanic Research
SOLEC	State of the Lakes Ecosystem Conferences
SOP	Standard Operating Procedure
SMP	Spanish Monitoring Program
SPM	Suspended particulate matter
SPMD	Semi-Permeable Membrane Device
SPREP	South Pacific Regional Environment Programme
SRES	Special Report: Emissions Scenarios
t	Tonnes
TBBPA	Tetrabromobisphenol A
TCDD	Tetrachlorodibenzo- <i>p</i> -dioxin
TEL	Tetraethyllead
TEQ	Toxicity Equivalents
TOMPs	Toxic Organic Micro Pollutants, Air Monitoring and Analysis Network
TPT	Triphenyltin
UNECE	United Nations Economic Commission for Europe
UNEP	United Nations Environment Programme
UNIDO	United Nations Industrial Development Organisation
USGS	U.S. Geological Survey
VOC	Volatile Organic Compound
WEOG	Western Europe and Others Group
WFD	Water Framework Directive
WHB	Western Hudson Bay
WHO	World Health Organisation
WMO	World Meteorological Organization
WWTP	Waste Water Treatment Plants
XAD	Styrene/divinylbenzene-co-polymer (resin)

GLOSSARY OF TERMS

Activity	Any programme or other activity or project that generates data or information on the levels of POPs in the environment or in humans that can contribute to the effectiveness evaluation under Article 16 of the Stockholm Convention Core matrices. These are the matrices identified by the Conference of the Parties to the Stockholm Convention at its second meeting as core for the first evaluation: A = ambient air; M = (human) mother's milk and/or B = human blood
CTD	The characteristic travel distance– defined as the “half-distance” for a substance present in a mobile phase
Hot spots	An area of high contamination
I L-1	Instrumentation level 1 capable to analyze PCDD/PCDF and dioxin-like PCB at ultra-trace concentrations: must be a high-resolution mass spectrometer in combination with a capillary column
I L-2	Instrumentation level capable to analyze all POPs: (capillary column and a mass-selective detector)
I L-3	Instrumentation level capable to analyze all POPs without PCDD/PCDF and dioxin like PCB (capillary column and an electron capture detector)
I L-4	Instrumentation level not capable to do congener-specific PCB analysis (no capillary column, no electron capture detector or mass selective detector)
Intercomparisons	Participation in national and international intercalibration activities such as ring-tests, laboratory performance testing schemes, etc
LOD	Limit of detection. Definition: The lowest concentration at which a compound can be detected; it is defined as that corresponding to a signal three times the noise.
<LOD	Result below the limit of detection
LOQ	Limit of quantification. Definition: The lowest concentration that can quantitatively be determined is three times higher than LOD.
<LOQ	Result below limit of quantification. Compounds found at levels between LOD and LOQ can be reported as present, or possibly as being present at an estimated concentration, but in the latter case the result has to be clearly marked as being below LOQ.
MDL	Method detection limit. The MDL considers the whole method including sampling, sample treatment and instrumental analysis. It is determined by the background amounts on field blanks.
Phase I	Activities to support the Article 16 effectiveness evaluation that will be conducted by the Conference of the Parties at its fourth meeting, information collected between 2000 and 2008.

- Phase II Activities to support the Article 16 effectiveness evaluation that will be conducted by the Conference of the Parties at its seventh meeting, information collected between 2009 and 2014.
- Phase III Activities to support the Article 16 effectiveness evaluation that will be conducted by the Conference of the Parties at its tenth meeting, information collected between 2015 and 2018.

EXECUTIVE SUMMARY

This third monitoring report presents the current findings on concentrations of persistent organic pollutants in the Western Europe and Others Group (WEOG) Region, and synthesizes information from the first (2000 to 2008), the second (2009 to 2014), and the third (2015-2020) phase of the global monitoring plan (GMP).

Because of the number of new POPs listed since the second phase and hence incorporated in to monitoring programmes, the scope of the GMP has increased for this report. In addition to trends, the work in the third phase was also directed at further enhancing comparability within and across monitoring programmes through new intercalibration studies, harmonising data handling, and continuing to ensure support to the collection, processing, storing and presentation of monitoring data in regions through the global monitoring plan data warehouse. A focus on data analysis and process modelling in ecosystems enables us to better interpret trends in POPs.

0.1 Contributing programmes

Key message: It is critical that the small number of international and national monitoring programmes continue in order to determine trends and therefore effectiveness of actions.

The data in this report and from the previous two GMP reports come from a relatively small number of long-term national and international programmes. Some new programmes have been implemented in the past several years, which add to the information on POPs available in the WEOG region.

0.2 Key messages from the data on baseline concentrations and temporal trends

Key messages: There are sufficient data to determine trends for many of the listed chemicals but not others. In general, concentrations are declining and are starting to level off where regulatory action was taken decades ago. It is noted however, that in some cases such as HCB there are slight increases, likely due to release from secondary sources and the effects of climate change.

The patterns for chemicals listed from 2009 onwards are complex and variable across chemicals, media and area. For example, certain chemicals showed mostly declining or no change in trends, while others showed increasing trends followed by decreasing trends, or consistent decreasing trends depending on location. There are insufficient data to detect trends for many of the most recently listed chemicals, inter alia, dicofol and SCCPs.

It is important that data and samples be maintained in a coordinated and sustainable way, such as through environmental specimen banks, and that programmes operate efficiently and collaboratively to address challenges.

Adequacy of information: Existing and new programmes in the WEOG region provide adequate monitoring data for POPs in core and other media, which includes baseline information on many of the new POPs. For many of the new POPs, however, temporal trends are not yet established.

Measurements in air: Most older and previously banned POPs are now showing slower rates of decline (e.g. DDTs, the drins, PCBs and chlordanes) or slight increases in some isolated cases (e.g. HCB), as secondary sources and effects of climate change and warming become increasingly important. Time trends for many of the new POPs are starting to become established. Endosulfan showed accelerated decline after listing, listed PBDEs show mostly declining or no change in trends, and PFOS and PFOA showed increasing followed by decreasing trends, or consistent decreasing trends depending on location. There are insufficient data to detect trends for HBCDD, SCCPs, PCP/PCA, HCB, PeCB, toxaphene, HBB, chlordecone, dicofol, and PCNs.

Measurements in human media: The levels of several of the initial POPs, such as PCBs, DDTs and dioxins, seem to decrease over time in human milk and/or blood, depending on area. This shows that regulations and banning of production and use of these chemicals combined with regulations on food to test for and restrict dioxin content, and changes in diet, have led to declining levels in humans. Some of the newer POPs (e.g. PBDEs, HBCD, PFOS and PFOA) show an increase over time followed by a decrease, depending on area. This further confirms that adequate risk management measures lead to declining levels in humans. The rising and then decreasing trend is likely because the risk management measures for these newer POPs were taken later than for the initial POPs. No clear trend over time could be seen for some of the POPs, such as HCHs, and the reason for this needs to be investigated further. For some of the POPs, e.g. SCCPs, there are not enough data to determine a trend.

Measurements in water: Water is a tier 2 core medium for reporting of PFOS, PFOA and PFHxS [if listed]. Over 11,200 individual measurements of PFOS, PFHxS and PFOA from approximately 3000 locations within the WEOG region, were found, including 2035 analyses for surface water samples collected from 2015-2019. The information available provides knowledge of spatial trends for PFOS, PFHxS and PFOA across western Europe, Canada/USA and southern/eastern Australia, the North Sea, the Baltic, the Great Lakes, and in the North Atlantic, Arctic Ocean, the Northeast Pacific, and Antarctica. No specific studies of temporal trends of each substance in water were found, therefore median concentrations for combined data from specific regions with the periods 2000–2009, 2010-2014 and 2015–2019 were compared. A challenge with these comparisons is that variation could be introduced due to differences in sampling locations and effects of seasonality such as river flow, snow melt, and thermal stratification (in lakes). Comparison of medians over the three time periods indicated that concentrations of PFOS and PFOA had declined from 2000-09 to 2015-19 for Western European rivers (including the Rhine), the North Sea, and the Baltic Sea. Higher median concentrations of PFOS were seen for 2015-19 vs 2000-09 for the Mediterranean, the North and Central Atlantic,

North Pacific and the Great Lakes. PFHxS increased in all oceans, while declining in European rivers and in the Great Lakes.

Measurements in Other Media: There is a wealth of information on POPs levels and temporal trends in other media across the WEOG region, reported under a variety of international programs, *inter alia*, AMAP, HELCOM, OSPAR, Great Lakes of NA, and MEDPOL. A few regions and species are well studied for most POPs. For other regions and newly listed POPs the information is sparse. Older POPs are present in the environment at low concentrations compared to the pre-2000 period. However, their concentrations have been rather stable, decreasing by only very low percentages since the last GMP report in 2015. Levels of some of these older POPs are still high and at levels of concern in some species and regions (e.g. PCBs in polar bears and whales). Due to the low and relatively unchanging concentrations, some of these older POPs, like DDT and Mirex, are not or will not continue to be monitored further under some of the monitoring programmes named above. In some regions, older POPs show increasing trends, such as HCB in the Great Lakes and the Baltic. POPs listed later in the Stockholm Convention are of more concern, as some show increasing concentrations over time, or stable high concentrations, or there is yet no good method for including them in the monitoring.

Meeting future challenges of additional new POPs: In response to the growing future demand for information on POPs it is important that data and samples be maintained in a coordinated and sustainable way, such as through environmental specimen banks, and that programmes operate efficiently and collaboratively.

0.3 Long range transport

Integrated assessment of POPs involves using models as a platform to quantitatively link emissions of POPs to the levels and trends observed in the environment. It is the most effective way to quantify the extent of long range transport of POPs and forecast the effectiveness of emission reductions. A variety of modeling tools are available to support integrated assessment of emissions, transport in the environment, and levels and trends of POPs observed at monitoring stations. When applied at local, regional and global scales, integrated assessments can be used to reduce uncertainties in emissions and transport profiles of POPs.

Some integrated assessment studies have used models to isolate the impacts of climate change on concentrations of POPs in the environment from other factors. The direct influence of climate change on levels and trends of POPs in the global environment on the time-scale of decades is likely to be much smaller than the influence of reductions in primary emissions that can be achieved through policy action. For example, for PCBs emission reductions that are forecast to occur from 2010 to 2100 are forecasted to drive declines in concentrations in Arctic air and seawater by several orders of magnitude, compared to changes in concentration of less than a factor of 4 due to climate change. However, indirect effects of climate change on chemical cycling, including changes in

food web structure, are not yet fully understood and are not described in these model scenarios.

Case studies of modeling emissions and fate of POPs presented in this report highlight: 1) linking emissions to human body burdens of PCBs at the global scale, 2) conducting integrated assessment of POP candidates, 3) estimating emissions of POPs and POP candidates from monitoring data, and 4) modeling global emissions, fate and transport of PFOS in the ocean. Experience with modeling existing POPs has beneficial synergies with identification and risk profiling of candidate POPs. Priorities for improving POP modeling capabilities include better descriptions of air/condensed phase partitioning, and reducing uncertainties in emissions and degradation rates of POPs in air, water and soil with targeted laboratory and field studies.

0.4 Description of data gaps and capacity building needs

Although baseline information for most POPs is adequate, temporal trends are not yet established for some of the newer POPs in the core media.

Recommendation: It is important that monitoring programmes are maintained and continue to establish analytical methods and time series for the newer POPs. Strategic partnerships with other expert labs/programmes may help to ameliorate the increased demands due to the growing number of listed POPs and candidate substances, the complexities of sample analysis for the newer POPs (e.g. SCCPs), and potential detection limit issues (e.g. air monitoring at background sites).

0.5 Future evaluations

As time series for newer POPs are developed within existing programmes, future evaluations will provide temporal trend information to inform the evaluation of the effectiveness of the Convention.

Recommendation: Monitoring programmes should continue to identify and target priority POP-like chemicals as this information is useful to the evaluation and assessment of candidate POPs and would support the work of the POP Review Committee. Efforts should continue to be made to integrate monitoring programmes and activities across regions for future reporting of comparable data under the GMP. Areas for cooperation include *inter alia*, continued monitoring across the Arctic region (e.g. AMAP), WHO human milk surveys, measurement of listed PFASs and their precursors in global oceans, and regional and global-scale passive air monitoring (e.g. GAPS, MONET).

Recommendation: Cooperative work, perhaps under the auspices of the effectiveness evaluation, to integrate GMP data with numerical models to estimate spatial and temporal trends in POP emissions, and link to human and environmental burdens, would greatly enhance understanding of the effectiveness of actions to protect human health and the environment.

0.6 Conclusions and recommendations

Cross-cutting conclusions across all media are that:

- The levels of many POPs, even those that have been regulated and managed, remain of concern.*
 - existing programmes need to continue, as well as certain ad hoc programmes such as those for water, in order to determine trends.*
 - large scale repeated programmes and the ability to share metadata would enable comparison and enhance the ability to determine long range transport.*
 - coordinating with other programmes such as ad hoc surveillance work on indoor air and urban and industrial emissions would enable more comprehensive understanding of exposure and effectiveness of actions to protect human health and the environment.*
-

0.6.1 Media specific conclusions and recommendations

Air:

Passive air samplers should continue to be implemented as a cost-effective approach for improving spatial information for POPs in air and their performance for measuring particle-associated POPs should be further assessed.

Several long-term air monitoring programmes, including EMEP, GLB and IADN, also conduct POP measurements in precipitation at some locations. It is recommended that future GMP reports include information on precipitation trends, which provides further insights on atmospheric loadings as an entry pathway to foodwebs affecting human health. Trend information for PFOS, PFOA, PFHxS and other PFASs is limited for air because these chemicals tend to partition to water. This data gap could be addressed in the future with available long-term precipitation data and historical records found in ice cores which solely reflects atmospheric deposition over time.

Recommendations beyond the current scope of the GMP, but relevant to future work on effectiveness of the Convention towards the protection of human health and the environment from POPs, include the development of strategies for linking concentration information for air with toxicity. In vitro methods such as transcriptomics and bioassays to assess various toxicity end-points are now available with the required sensitivity to be applied to air sample extracts.

Human media:

By gathering data from both national and regional monitoring as well as scientific literature it was possible to get an overview of the situation in the WEOG region. Differences and similarities between countries and continents could be identified, as well as changes over time. It is of great importance that these regional and national programmes evaluating time trends of both POPs and other environmental pollutants in blood and/or milk continues since human exposure data is important for following up the

effectiveness of the Stockholm Convention and to see that regulations and other actions taken in order to reduce the exposure to POPs are working.

Very little data could be found for several of the POPs listed after 2009 (e.g. SCCPs). In order to be able to follow up on these substances over time, it is important to start monitoring these substances now. In addition to measuring the POPs listed before 2009 that are already regulated, it is important to also monitor possible substitution substances that have POPs-like characteristics.

In order to be able to use monitoring data to evaluate and compare levels and trends between countries, regions and continents it is crucial to be able to retrieve as much meta data and background information as possible. To store monitoring data in searchable and open databases are preferable. Biobanking samples for later analysis is also very useful since it makes it possible to go back and produce time trends for new pollutants in the future.

Water:

Seawater monitoring sites for PFAS have not been established so far, but if they are, then they should be located within a region where spatial variability is relatively small.

Continued monitoring of rivers offers the best opportunity to assess temporal trends. However, there is always the need for consideration of waste water and tributary inputs and thus multiple sampling points preferably at sites that are well characterized in terms of flow and proximity to sources are needed.

Lower detection limits would help with future assessments of temporal trends in all locations but especially in open oceans, coastal seas and in the Great Lakes. Detection limits in the range of 1-5 pg/L for PFOS, PFHxS and PFOA have been demonstrated by several monitoring programmes and should be universally adopted.

Efforts should be made for more widespread measurement of precursors of PFOS, PFHxS and PFOA in waters. This can be through targeting specific known precursor compounds or using “total” methods such as total extractable organic fluorine or total oxidizable precursor (TOP) assay.

Other Media:

The monitoring work in other media across the WEOG region has been steadily growing since the second phase. Vast collections of high quality of data are available, however the diversity of monitoring efforts makes it difficult to compile those scattered data for statistical analyses, modelling and large-scale comparison. In consequence, regular, consistent and rigorous large-scale data compilations would greatly facilitate and enhance the ability to determine trends, LRT and enable ecosystem modeling. To achieve this goal, the archiving of data and samples, QA/QC and data flow between data providers and users should be considered a priority.

Current results seem to indicate that POPs regulated in source regions decades ago have significantly decreased but are still present at low levels that have not changed since the

previous GMP report in 2015, and some are still of concern in some regions. PCBs continue to be present in some regions and species, with levels exceeding thresholds for effects, which is of concern. Some substances listed later in the Stockholm Convention with extensive exemptions for continued use show increasing levels (e.g. HBCD, HCBd, PFOS and PCN in some areas).

Climate variability and climate change effects on the environment and ecosystems structure and function are more likely to affect food web accumulation and toxicity to a larger degree than long range transport of POPs. It is therefore important to increase the efforts in understanding how climate change induced changes in the ecosystems affect the time trends of POPs in other media including biota, e.g. through changes in diet, in order to separate ecosystem changes effects from changes due to global regulation and bans through the Stockholm Convention.

Efforts should be made to make use of the vast body of information that is available for POPs in other media (including wet deposition, water and soils), in collaboration with modelers, to develop a better understanding of the fate of POPs in the multi-media environment under different conditions.

0.6.2 Generic conclusions and recommendations

Key message: Sixteen years after the Stockholm Convention coming into force, the Global Monitoring Plan has shown that primary sources of POPs listed before 2009 have been substantially reduced and current low levels can be attributed to effectiveness of policies and regulations. Baseline concentrations of many but not all POPs listed after 2009 are becoming available through some of the monitoring programmes for core and other media. Additional data in the future will enable the evaluation of trends to inform effectiveness of management actions.

The Convention and its Global Monitoring Plan have raised awareness, triggered action and provided a framework for collaboration and integration at the national, regional and global scales

Key recommendations:

Challenges with POPs listed after 2009: Certain high molecular weight POPs including polar / ionizable chemicals (e.g. PFOS) exist in air primarily on the particle-phase. To improve understanding of regional and global transport of particle-associated POPs, improve the performance of fate models and help to interpret temporal trend information from monitoring programmes, better information is needed on gas-particle partitioning properties of POPs, as well as studies of particle-associated transport and fate.

Chemical Mixtures: Human health and the environment are impacted by exposure to chemical mixtures which includes a multitude of known and unknown POP-like chemicals, including their transformation products. Research in this area is advancing rapidly due to increased accessibility of high resolution analytical equipment (e.g. gas- or liquid chromatography - high resolution mass spectroscopy). Future reporting under the GMP would benefit from including information, where available, regarding levels of all listed POPs and potentially candidate POPs in the same samples, as this can enhance understanding about co-occurrences and thus about co-exposure and environmental loads. Similarly, reporting of the concentrations of precursors for listed PFASs, such as through total methods, also enhances understanding about environmental loads.

Efficiencies for Monitoring Programmes: In order to deal with analysis pressures for new POPs, monitoring programmes may need to adjust their protocols and resources to better align with new priorities. Possible strategies include reduced analysis frequency for legacy POPs (e.g. PCBs, organochlorine pesticides), optimized analytical methods, and partnerships among laboratories to address specialized analytical needs. It is not necessary for every laboratory to be an expert for every class of POP. In addition, many programmes are incorporating the latest advances in the field of science such as high resolution and non-targeted analysis (and associated data archiving for future reference). Future reporting may be able to draw on data generated through these advances.

Sample Archiving / Sample Banks: Investing resources to ensure the integrity and sustainability of sample banks will provide a highly cost effective tool for understanding the effectiveness of regulatory and policy intervention, as they enable retrospective analysis on current and future POPs, as well as for screening studies on candidate POPs.

Data archiving and accessibility: Databases and compilations such as the GMP reports play an important role in identifying, documenting and making accessible reliable and comparable data sets on POPs. These data are needed for assessing environmental levels and trends for POPs and for combining with emission estimates and models in integrated assessments of sources, fate and transport of POPs.

Recommended areas of future work include:

1. further efforts to make databases easily searchable, openly accessible (e.g. data downloadable) and to integrate / link databases.
2. further development of simple visualization tools such as those implemented under the GMP database would be useful.
3. Enhancing the ability to archive large data files due to the advancement of high resolution and non-target analysis methods. These approaches generate large data files that require sufficient and secure storage and provide an opportunity for retrospective analysis of these data files for targeting new POPs and related chemicals (e.g. precursors and transformation products).

Recommendations beyond the scope of the GMP:

Suggested future work on the effectiveness of the Convention towards the protection of human health and the environment from POPs, includes:

- Linking the GMP work with data from monitoring and research programmes aiming to understand current exposure levels and emissions to the broader environment including urban areas and waste sectors - given that newer POPs can be in domestic, household and commercial products. In addition, declines in environmental background concentrations are likely to be slower where listed chemicals have exemptions for continued use or presence in recycled materials.
- Accordingly, co-operative work to integrate GMP data and other data with numerical models to estimate spatial and temporal trends in POP emissions, and link to human and environmental burdens, would greatly enhance understanding of the effectiveness of actions to protect human health and the environment.
- Enhancing cooperation with other monitoring efforts in the frame of the Basel, Rotterdam and Stockholm Conventions, the Minamata Convention, the United Nations Framework Convention on Climate Change and the Convention on Biological Diversity.

The ROG wishes to emphasise that the information reviewed to provide baselines and to determine current trends in the WEOG region is predominantly drawn from a relatively small number of existing international programmes - which can in turn depend on contributing national programmes - as well as ad hoc programmes. The ability to compare POPs levels over time makes the long-term viability of international and ongoing national programmes of utmost importance.

1 INTRODUCTION

The first phase of the GMP has been implemented during the period 2004-2009 and the second phase during 2010-2017, providing information on changes in concentrations of the 12 POPs initially listed in the Stockholm Convention and information on baseline concentrations of the 11 substances newly listed in the annexes to the Convention in 2009, 2011 and 2013. Two sets of regional monitoring reports and global reports have been developed to date in the frame of the GMP and have informed the effectiveness evaluation under Article 16 of the Convention.

The present (third) monitoring report synthesizes information from the first, the second, and the third phase of the global monitoring plan and presents the current findings on POPs concentrations in the Western Europe and Others Group (WEOG) Region. While the first and second monitoring reports, presented at the fourth and seventh meeting of the Conference of the Parties, respectively, provided information as to the changes in concentrations of the chemicals initially listed in the Convention, as well as baseline information on some of the newly listed POPs, this third report builds on the increasing information base of POPs monitoring data and provides a further in-depth assessment of the changes measured over time in POPs concentrations, including time trends where available, as well as recent baseline information on the more recently listed POPs.

At its sixth meeting in May 2013, the Conference of the Parties, by decision SC-6/23 on the global monitoring plan for effectiveness evaluation, adopted the amended global monitoring plan for persistent organic pollutants (UNEP/POPS/COP.6/INF/31/Add.1) and the amended implementation plan for the global monitoring plan (UNEP/POPS/COP.6/INF/31/Add.2).

At its seventh meeting held in May 2015, the Conference of the Parties, by decision SC-7/25, welcomed the second regional monitoring reports, and, at its eighth meeting held in May 2017, by decision SC-8/19, it welcomed the second global monitoring report which marked the end of the second phase of implementation of the GMP. COP-8 requested the Secretariat to continue to support the work on the GMP to provide relevant input to the process of effectiveness evaluation under Article 16 of the Stockholm Convention and ensure sustainability of POPs monitoring toward the third GMP phase.

Monitoring activities have been ongoing in the five UN regions to support POPs monitoring data generation for the third GMP phase. The global coordination group met four times over the period 2015-2018 in order to oversee and guide implementation of the third phase of the global monitoring plan, with particular emphasis on addressing the sampling and analysis of the newly listed POPs, harmonizing data collection, storage and handling, addressing the needs for ensuring sustainability of ongoing monitoring activities and for further capacity strengthening to fill the existing data gaps, as well as improving data comparability within and across monitoring programmes.

Long term viability of existing monitoring programmes (air and human bio-monitoring) is essential to ensure that changes in concentrations over time can be investigated. National air monitoring activities having contributed data to the first and second

monitoring reports continued during the third phase, and new programmes have been identified to support the development of the third reports. Likewise, the continued operation of global and regional air monitoring programmes was a major pillar in the third phase. For the new monitoring activities, collaboration with strategic partners has ensured cost-effective generation of data and use of harmonized protocols for POPs monitoring. The implementation of the UNEP/WHO human milk survey is another important pillar of the global monitoring plan, providing useful long-term results showing how human exposure to POPs changed over time as measures are implemented to enforce the Convention.

Enhanced comparability within and across monitoring programmes to evaluate changes in levels over time and the regional and global transport of POPs was an equally important milestone. QA/QC practices have been and continue to be essential for ensuring comparability, along with inter-laboratory exercises and intercalibration studies. Efforts continue to be directed at ensuring comparability within and across programmes, providing for evaluation of changes in concentrations of POPs over time and enabling regional comparisons.

The GMP Data Warehouse has been made operational during the second GMP phase, supporting the regional organization groups in the work for the collection, processing, storing and presentation of monitoring data. The global monitoring plan data warehouse also constitutes a publicly available repository of valuable information that can serve as a useful resource for policy makers and researchers worldwide. The data warehouse was further enhanced and kept up-to-date to provide on-line access to the GMP monitoring data and enable data collection and processing during the third GMP phase and support the development of the third monitoring reports.

The process for updating the GMP guidance document has continued; information relevant to the POPs listed more recently in annexes to the Convention and on the chemicals recommended for listing or those in the process of review by the POPs Review Committee has been included in the guidance. The Guidance on the Global Monitoring Plan for Persistent Organic Pollutants has been streamlined and updated in 2019 (UNEP/POPS/COP.9/INF/36) and provided a useful basis as the reference document for POPs monitoring in the third phase of the GMP, as well as for harmonized data collection, storage and handling.

2 DESCRIPTION OF THE REGION

2.1 Background

The Western European and Others Group (WEOG) is composed of twenty-eight nations in Western Europe, North America, and Australasia.

A comprehensive summary of the characteristics of the region can be found in the reports of the 2002 UNEP/GEF: Regionally Based Assessment of Persistent Toxic Substances (PTS). This study used a different division of regions as was adopted by the Conference

of the Parties to the Stockholm Convention (COP) for the purposes of effectiveness evaluation. Therefore, the WEOG countries can be found in six different volumes of the PTS study. These are volumes on: the Arctic; North America; Europe; the Mediterranean; South East Asia and South Pacific; and the Antarctic. These reports are recommended for providing a synthesis of geographical, physical, and biological characterization of these areas, as it relates to the sources and fates of POPs released to the environment. They provided the basis for the following brief political, geographical, and social and economic overview.

2.2 Political

Specific membership is as follows: Australasia - Australia and New Zealand.

North America - Canada and United States. Western Europe – Andorra, Austria, Belgium, Denmark, Finland, France, Germany, Greece, Iceland, Italy, Liechtenstein, Luxembourg, Malta, Monaco, Netherlands, Norway, Portugal, Republic of Ireland, San Marino, Spain, Sweden, Switzerland, Turkey and United Kingdom.

2.3 Geographical

The region is not a coherent geographical unit. Its membership is divided across three continents, and two hemispheres. However, Canada and the United States collectively occupy similar latitudinal zones as those occupied by Western Europe while Australia and New Zealand occupy generally the similar corresponding latitudes in the southern hemisphere.

The climate in Australia is mainly arid or semi arid. It is temperate in the south and tropical in the north. New Zealand is temperate with some regional contrasts.

In North America, climatic variation is large extending from the Arctic in the north to a sub-tropical climate in the south. Within this pattern there is much variation. For example, within the Arctic, there is considerable moderating maritime influence on the coastal strip of Alaska but intense cold in the central and high eastern Canadian sectors.

Western Europe includes four climatic zones – A sector of the circumpolar Arctic in the north, alpine climate in the main mountain ranges such as the Alps and Pyrenees, a temperate zone in the central area and the Mediterranean zone to the south. The latter is generally characterised by mild wet winters and hot dry summers with more than 90% of annual precipitation falling in winter. Superimposed on this pattern is the moderating and wet influence of the Atlantic to the west, with drier and more extreme temperature ranges to the east.

2.4 Social and economic considerations

Australasia: Australia and New Zealand are developed countries with mixed economies and substantial agricultural sectors. Available information suggests that environmental levels and human exposure to POPs in both countries has always been low in relation to most regions elsewhere. Regulations are well established concerning the environmental release of the original twelve POPs.

North America: The USA and Canada are developed, industrialised countries. Of particular interest in terms of historical and potential source characterization is the Great Lakes Basin. It covers an area of approximately 774,000 square kilometers including much of Ontario, and the eight Great Lakes States: Illinois, Indiana, Michigan, Minnesota, New York, Ohio, Pennsylvania and Wisconsin. The Great Lakes basin is home to a total of about 33 million people, more than one-tenth of the population of the United States and one-quarter of the population of Canada. It is also a focus of both the U.S. and Canadian industrial capacity, while agriculture in Ontario and Quebec accounts for the largest single use of Canadian land in the basin, contributing about 40 percent of the value of agricultural output in the Canadian economy. Nearly 7 percent of the American agricultural production is located in the basin. The emissions and environmental fate of POPs in the Great Lakes and St Lawrence basins have been extensively studied and have provided much of the information for North America in the present report. Comprehensive regulations are well established concerning the environmental release of POPs.

Western Europe: In Western Europe the chemical industry, metal production and processing and agriculture are all significant parts of the economy. There is a range from highly industrialised economies to other countries with greater reliance on agriculture and/or a more developing economic structure.

Generally in Western Europe there is considerable information relating to industrial point source emissions to the atmosphere. Sources to air of well studied compounds such as PCBs and PCDDs/Fs are generally well characterised and inventories have been calculated and updated regularly via EMEP. A number of European countries such as the UK and Germany also have a long industrial history, involving combustion activity in the form of wood and coal burning. The smelting of metals, cement kilns, and the production of iron and steel also have a long history in Europe, activities known to produce significant PCDDs/Fs emissions.

Most of the Western Europe region is today part of the European Union and comprehensive regulations concerning the environmental release of POPs are well established with progressive development of harmonized regulation.

The Arctic: North America and Western Europe collectively include a large segment of the Arctic. This area is of particular interest in terms of the fate of POPs in the environment because it is lightly populated and with a generally low intensity of industrialization. However historical uses of POPs have occurred in the Arctic and waste disposal is continuing. The eight Arctic countries that are members of the Arctic Council contribute monitoring information on POPs to the Arctic Monitoring and Assessment Programme (AMAP) which produces extensive periodic assessment reports.

The Antarctic: The Antarctic is not defined by national boundaries. Because a number of WEOG region countries are parties to the Antarctic Treaty and are maintaining research activities in the area, the Antarctic has been given brief attention in this report (see section 4). The Antarctic is completely surrounded by ocean and is largely snow and ice covered. It is therefore remote from any significant primary source of the POPs listed in the Convention.

2.5 Considerations in relation to sampling strategies

The WEOG ROG report is based mainly upon existing programmes. Although it is difficult to provide a good categorization, the following illustrates the range of objectives observed from the programmes contributing to the WEOG report:

- ❖ Studies aimed to investigate levels and/or processes in areas close to historical local sources of POPs (e.g. HELCOM and studies in the Great Lakes region of North America such as IADN and GLB) and national initiatives such as in Australia and Spain.
- ❖ Studies aimed to investigate levels and/or processes in areas distant from local sources of POPs where long-range transport and other processes are believed to be important in determining environmental levels and/or human exposure (e.g. AMAP, ARCRISK, MONARPOP and NCP)
- ❖ Studies aimed at understanding regional and/or global transport (e.g., ARCRISK, EMEP and GAPS)
- ❖ Studies aimed at providing a wide survey of levels (e.g., WHO coordinated human milk surveys; NHANES)

It is important to be aware of the historical objectives of the different programmes that have been the source of information used by the ROG. This information is contained in the programme summaries that are provided in Annex 1 and 2 of this report.

3 ORGANIZATION OF REGIONAL IMPLEMENTATION

The WEOG regional Organizational Group (ROG) for this third evaluation was composed of six experts from each of the following countries: Australia, Spain, Canada, Sweden, Norway and one representing the European Commission.

3.1 Organization of activities

The ROG met exclusively by teleconference calls supplemented by internet communication. A record of each meeting was kept which documented key decisions and work responsibilities until the next meeting when progress would be reviewed. Sixteen such teleconferences were held before the draft report was sent to the WEOG focal points for their review. The chair rotated with each meeting. Early decisions were taken upon general responsibilities for coordination and drafting of the regional report (see acknowledgements section). A critical path was developed of work to be performed in sequence leading up to the finalization of the report and which included the elements detailed below. In general, this plan was followed throughout the data gathering and drafting process.

3.2 Strategy to locate and collect information from existing programmes

At its initial meetings, the ROG reviewed information from programmes that contributed to the first WEOG region report. These programs were initially identified based upon survey responses obtained by the Secretariat. The basis of the selection was the application of the criteria contained in the “Implementation Plan for the First Evaluation Reports”. For this third WEOG regional report, new programs were included if they also met these criteria.

For obtaining POPs data for Air and Human Tissue samples, programmes were contacted and invited to provide “programme summaries” according to a template developed by the ROG. In a few cases, the programme summaries were provided by ROG members, or by arrangements established by ROG members. This usually occurred when a single organization is host to the data bases for several programmes as is the case of the Norwegian Institute of Air Research (NILU) with the air information of both EMEP and AMAP. In cases where updated programmes information was available in reports or on-line, chapter leads summarized the most relevant information in their chapters.

Water was included for the second time in the WEOG report as a second tier core medium for PFOS, PFOA and PFHxS. Information was compiled by a WEOG expert based on a literature review of published papers and reports. The Other Media chapter was compiled from available reports from the programmes.

The drafting of the WEOG region report benefitted from ongoing and recent efforts under AMAP to assess temporal trends for POPs in Arctic datasets (AMAP Summary is included in Annex 1).

3.3 Identified data gaps and strategies to fill them, including strategies and activities to generate monitoring data

Following the compilation of the first WEOG region report, the ROG concluded that information from existing programmes in the WEOG region provides an adequate overview of the status of POPs in the core media for the first evaluation and therefore decided not to attempt to initiate any new monitoring activities. However, some new activities were implemented by some countries to further enhance data coverage across the WEOG region and data from these programmes are included in this report. In addition, the current report relies more heavily on data from the scientific literature, compared to the previous WEOG reports.

Although most of the data included in the regional report originates from existing long-term programmes, data from smaller scale programmes and even “snapshot” data were included in some cases to improve geographic resolution and completeness of reporting for POPs. The ROG decided on the strategy to include such data if it otherwise was consistent with the criteria specified in the implementation plan for the first evaluation. Information on PFOS, PFHxS and PFOA in water originates primarily from these types of data sets.

3.4 Capacity strengthening needs

The sustainability of the contributing programs and their capacity to expand the analytical list and assess mixtures in support of chemical assessment and management have been identified as ongoing and future challenges. Also there is a need to increase the capacity to monitor at known sources, e.g. urban/industrial environments for commercial chemicals.

4 METHODS FOR SAMPLING, ANALYSIS AND HANDLING OF DATA

4.1 Background

This 3rd report for the WEOG region builds upon activities and arrangements that contributed baseline and trends information from the WEOG region for the 1st and 2nd GMP reports. At that time and when initiating its activities to obtain monitoring information, the ROG took careful note of two concepts outlined in Article 16 of the Convention. First, it is stated that Parties shall make arrangements to obtain comparable monitoring data. The operational procedure to achieve comparability is the application of the criteria for programme selection outlined in the Implementation Plan for the first evaluation and the measures listed in the “Guidance Document”. Second, Article 16 further states that the arrangements to gather data should be implemented using existing programmes and mechanisms to the extent possible.

As noted in section 3 of this report, the ROG decided that for the first effectiveness evaluation, it would base its review of POPs levels in the region using information derived entirely from existing programmes and activities. These same programs have again contributed to this 3rd report and have been supplemented by newly initiated programmes and information on water for the purpose of reporting for PFOS, PFOA and PFHxS (see Ch. 5.2 and Ch. 5.3). The ROG has not initiated any sampling, analytical, or data handling activities and therefore has nothing to report on such matters. However, the practices to conduct these elements were carefully considered when the ROG reviewed information on existing programmes based upon survey responses obtained by the Secretariat, and selected candidate programmes to provide the basis for the first and second evaluation reports. The selection was performed by application of the above noted criteria which resulted in the identification of more than 16 established international and national programmes to be the main “comparable” information sources for the first evaluation. At least one member of the ROG and/or a designated WEOG expert (according to the expertise of that individual) then carefully examined the sampling, analytical, data quality, and data storage arrangements of each of the programmes. This enabled the ROG to satisfy itself that such arrangements are being maintained and will enable comparable data from the identified programmes to be used by the COP now and in the future to look for changes in POPs levels over time within those programmes.

4.1.1 Key Message regarding “comparable data” from existing programmes and activities

Although the ROG believed that it is practical and realistic to expect intra-programme (internal) comparability, it noted that each of the established programmes has its own procedures for conducting its work, usually including constraints on the use of different analytical laboratories within each programme. It is important to note that very few programmes share the same analytical laboratory with other programmes. Since the use of different analytical laboratories is a major source of variance, the ROG concluded that it would be very difficult to achieve levels of comparability between programmes

necessary for effectiveness evaluation. Therefore in preparing this report, the ROG focused on using information from programmes where measures and procedures are expected to provide intra-programme comparability over time for present and future effectiveness evaluations. While this conclusion generally means that there will be very limited direct comparability between programmes and regions, significant exceptions are evident, such as when a programme operating in several regions has maintained a centralized analytical facility servicing all regions, such as with the WHO coordinated human milk programme. The extensive use of such measures as utilization of common analytical laboratories and data centres has demonstrated the possibility of achieving adequate comparability between well established programmes in the WEOG region. An example is the collaborative practices of AMAP, EMEP, OSPAR, and HELCOM.

4.1.2 Availability of information to allow data to be independently evaluated

This report provides the COP with the information requested in a concise fashion but in an easily accessible form. The information is provided in four tiers:

- 1 The short “Executive Summary” and “Chapter 6 Summary” elements of this report which inform the COP of the essential features of the levels and temporal trends of POPs in the region;
- 2 The concise synthesis of information derived from the contributing existing monitoring programmes (Chapter 5 of this report);
- 3 More detailed information on the nature of operation and data used from each of the contributing programmes. These are termed “programme summaries” and are provided in Annex 1 and 2 of the WEOG report. The Water and Other Media chapters include detailed information in Annexes 3 and 4. They were chiefly prepared by experts working in the existing contributing programmes; and,
- 4 Ensuring that full details on any aspect of an existing programme can be accessed, usually through direct contact with the management or secretariats of each contributing programme.

Therefore if an individual would like to obtain more information for example on the analytical methodologies, quality assurance and control, data handling, and data availability practices of a contributing programme, that individual has a choice of the degree of detail that can be accessed.

4.2 Description of national and international programmes and activities that have contributed information for the regional report

The following tables (**Table 4.2a** and **Table 4.2b**) briefly summarize the nature of each of the main sources of information on environmental levels of the core media (air and human milk / blood) used by the ROG for this report. More details can be found in the programme summaries provided in Annex 1 and 2 including in many cases maps to

indicate geographical coverage, while a collective synthesis of the programme results is provided in chapter 5. The ROG found it difficult to develop a summary map of the geographic coverage due in part to the evolving nature of programmes.

4.2.1 Programmes/activities related to air monitoring

Table 4.2a Monitoring programmes which contributed to the assessment of POP temporal trends and levels (23 priority substances) in the WEOG region.

Monitoring programme	Abbrev.	Region of interest	Current # of monitoring sites in WEOG	Monitoring period	Monitored compounds (Stockholm conv. POPs)
Arctic Monitoring and Assessment Programme ^a	AMAP	Arctic	8 long-term 1 satellite	1992 – present (start dates vary among stations)	Chlordanes, PBDEs, HCB, α - and γ -HCH, DDTs at all stations In addition: Alert reports: aldrin, dieldrin, endrin, heptachlor, HBCDD, β -HCH, HCB, mirex, PeCB, PCA, PCBs, PFOA and precursors, PFOS and precursors, α - and β -endosulfan, endosulfan sulfate (screening of SCCPs) Stórhöfði reports: dieldrin, β -HCH, PCBs, toxaphene Zeppelin reports: HBCDD, β -HCH, PCBs, PFOA and precursors, PFOS and precursors, SCCPs Andoya reports: HBCDD, β -HCH, PCBs, PFOA and precursors, PFOS and precursors Pallas reports: PCBs, α -endosulfan Station Nord reports: aldrin, dieldrin, endrin, heptachlor, β -HCH, PFAS precursors, α - and β - endosulfan Tiksi and Amderma report: aldrin, dieldrin, endrin, heptachlor, mirex, PCA, PCBs Little Fox Lake (satellite station) reports: aldrin, dieldrin, endrin, α - and β -endosulfan, HBCDD, HCB, HCHs, heptachlor, PBDEs (Aug 2011 – Dec 2014)
Australia's Casey Station	Casey	Antarctica	1	2010-2015	PeCB, HCB, dieldrin, aldrin, endrin, heptachlor, chlordanes, toxaphene, endosulfan, mirex, HCHs, DDTs, PCBs
Australia's National Passive Air Sampling and Archiving Program	Australia-PAS	Australia	44	2011-present	PCBs, PeCB, HCB, HCHs, heptachlor, aldrin, dieldrin, endrin, chlordanes, DDTs, mirex (2012 at 15 sites only), α - and β -endosulfan, SCCPs (2016 at 15 sites only) Samples from other years and sites are archived (not analysed).
European Monitoring and Evaluation Programme	UNECE-EMEP	Europe	12	1993 - present	PCBs, DDTs, chlordanes, HCB, HCHs, PBDEs Note that AMAP sites Zeppelin, Pallas, Stórhöfði, Station Nord/Villum Research station, Amderma are also EMEP sites. Please see additional analytical compounds above under AMAP.

Table 4.2a continued...

Monitoring programme	Abbrev.	Region of interest	Current # of monitoring sites in WEOG	Monitoring period	Monitored compounds (Stockholm conv. POPs)
Global Atmospheric Passive Sampling network	GAPS	Global	46	2004 - present	dieldrin, chlordane, heptachlor, DDTs, α - and β -endosulfan, endosulfan sulfate, PCB, chlordanes, HBCDD, HCB, PeCB, HCHs, PBDEs, PFOS and precursors, PCDD/Fs (2011, 2012 only)
Great Lakes Basin Monitoring and Surveillance Program	GLB	Great Lakes (Canada)	2 master stations; 1 satellite station	1992 - present	aldrin, endrin, dieldrin, chlordanes, DDTs, heptachlor, HBCDD, mirex, PCBs, HCHs, α - and β -endosulfan, endosulfan sulfate, PBDEs, SCCPs
Integrated Atmospheric Deposition Network	IADN	Great Lakes (US)	3 master stations; 2 urban stations	1990 - present	aldrin, chlordanes, DDTs, dieldrin, heptachlor, α - and β -endosulfan, endosulfan sulfate, HCHs, PCBs, chlordanes, HCB, PBDEs, HBCDD, endrin, dieldrin
MONitoring NETwork (Europe)	MONET (EU)	Europe	23	2009-present	PCBs, HCHs, DDTs, HCB, PeCB, chlordanes, aldrin, dieldrin, endrin, heptachlor. Analysed but not yet reported: PCDDs/Fs and PBDEs
Monitoring Network in the Alpine Region for Persistent and other Organic Pollutants	PureAlps (formerly MONARPOP, POPAlp, EMPOP, VAO II)	European Alpine regions	2	2005 - present	PCB, chlordanes, HCB, PCDD/F, HCB, α - and β -endosulfan, aldrin, dieldrin, endrin, DDTs, HCHs, PeCB, DBDPE, heptachlor, mirex, PBDEs, HBB, HBCDD, endosulfan sulfate
National Air Pollution Surveillance	NAPS	Canada (Urban)	29 urban, 8 rural and 1 suburban sites	1989-2009	PCDD/Fs, co-planar PCBs (2004-2009)
Northern Contaminants Program (Canada) ^a	NCP	Arctic	1 long-term; 1 satellite	1992 - present	Alert reports: aldrin, dieldrin, endrin, heptachlor, HBCDD, HCB, β -HCH, HCB, mirex, PeCB, PCA, PCBs, PFOA and precursors, PFOS and precursors, α - and β -endosulfan, endosulfan sulfate (screening of SCCPs) Little Fox Lake (satellite station) reports: aldrin, dieldrin, endrin, α - and β -endosulfan, HBCDD, HCB, HCHs, heptachlor, heptachlor, chlordanes, DDTs, PBDEs (Aug 2011 – Dec 2014)
Norwegian Troll Station	Troll	Antarctica	1	2007-present	PCBs, HCB, HCHs, DDTs, chlordanes, PBDEs (periodically), SCCPs (periodically)
Spanish Monitoring Program on POPs	SMP-POPs	Spain	23	2008-present	PCDD/Fs, PCBs, PBDEs, HCHs, HCB, DDTs, PeCB, α - and β -endosulfan

Table 4.2a continued...

Monitoring programme	Abbrev.	Region of interest	Current # of monitoring sites in WEOG	Monitoring period	Monitored compounds (Stockholm conv. POPs)
Swedish National Monitoring programme for Air ^b		Sweden	3	1996-present	PCBs, HCB, HCHs, chlordanes, DDTs, PFOS, PFOA, PFOSA, PBDEs, endosulfan, HBCDD, Dioxin and furans, SCCPs (2 sites) Aldrin, dieldrin, heptachlor (1 site)
Toxic Organic Micro Pollutants	TOMPs	UK	6	1991 - present	PCDD/F, co-planar PCB, PCBs, PBDEs (2010-present)
UK/Norway SPMD Transect		UK and Norway	13	1994 - present	PCBs, PBDEs

^aThe Northern Contaminants Program (NCP) is the Canadian National Implementation Plan of the Arctic Monitoring and Assessment Programme (AMAP). Data from 9 AMAP stations are presented in the current assessment, within which 1 long-term (Alert, NU Canada) and 1 satellite (Little Fox Lake, YK Canada) stations are operated under NCP.

^bThe Swedish Monitoring Programme operates 3 long-term stations, Råö, Aspvreten/Norunda, Pallas, for the monitoring of POPs. These stations are also part of EMEP and Pallas is part of AMAP as well.

In total 16 atmospheric monitoring programmes contributed results to this assessment.

4.2.2 Programmes/activities related to human tissues (milk and blood)

Table 4.2b Programmes and activities related to human media (human milk and /or blood) that have contributed to the regional report.

Programme	Media	Geographic coverage	Time period	POPs measured
UNEP/WHO milk survey	Human milk	WEOG countries	1987 – 2014	Aldrin, Chlordanes, DDT-complexes, PCDD/PCDF, endosulfan, HBCD, HCB, HCH, heptachlor, mirex, HBB, PBDE, PCB, PeCB, toxaphene
Swedish health-related environmental monitoring	Human milk	Sweden	1972 – 2017	Chlordanes, DDT-complexes, PCDD/PCDF, HBCD, HCB, HCH, PAC, PBDE, PCB, PFAS
Swedish health-related environmental monitoring	Blood	Sweden	1987 – 2017	dioxins/furans, FRs, HBB, HCB, HCH, PBDE, PCB, PeCB, PFAS
Peer reviewed literature	Human milk	WEOG countries	1972 – 2017	Aldrin, Chlordanes, chlordecone, Chlorinated Paraffins (CPs), DDT-complexes, PCDD/PCDF, endosulfan, HBB, HBCD, HCB, HCH, heptachlor, mirex, OCP, HBB, PBDE, PCB, PeCB, toxaphene
Arctic Monitoring and Assessment Program (AMAP)	Blood	Alaska, Canada, Denmark, Faroe Island, Greenland, Iceland, Norway, Russia, Sweden	1992 – 2013	Chlordane, DDT-complexes, HCB, HCH, mirex, PBDE, PCB, Pentachlorophenol, PFAS
Australia HBM	Blood	Australia	2002 – 2012	Chlordane, DDT-complexes, dioxins/furans, HCB, HCH, mirex, PBDE, PCB, PFAS
The Canadian Health Measures Survey (CHMS)	Blood	Canada	2008 – 2016	Chlordane, DDT-complexes, HCH, mirex, PBDE, PCB, PFAS
National Health and Nutrition Examination Survey (NHANES)	Blood	USA	2005 – 2015	Chlordane, DDT-complexes, dioxins/furans, HCB, HCH, mirex, HBB, PBDE, PCB, PFAS
Biomonitorización de Contaminantes en la Población Española (BIOAMBIENT.ES)	Blood	Spain	2009 – 2010	Aldrin, dieldrin and endrin, DDT-complexes, HCB, HCH, Heptachlor, PCB, PFAS
German Environmental Survey (German ESB)	Blood	Germany	1982 – 2010	HCB, PCB, Pentachlorophenol, PFAS

It is important to note that the results from national programmes could be reported individually or through their pre-existing participation in an international programme (such as AMAP).

4.2.3 Programmes/activities related to water

While the majority of the available data for this assessment was from the peer reviewed scientific literature (see search criteria in Section 5.2.3.2), additional results were obtained from government laboratories that are involved in monitoring and surveillance of contaminants in water. PFAS results that were available on the NORMAN EMPODAT database website (<https://www.norman-network.com/nds/empodat/>) were also downloaded. Results for rivers in Germany, Italy, The Netherlands and Spain previously uploaded to the database from several agencies (Landesamt für Umwelt, Landwirtschaft und Geologie and the Federal Environment Agency (UBA-DE) in German, Association of Rhine Water Works (The Netherlands)), and the European Commission Joint Research Centre, Institute for Environment and Sustainability were included. Data for PFASs in water were also available from national programs in Canada, Finland, and Australia. Regional monitoring and assessment programs within the WEOG such as HELCOM, OSPAR, MEDPOL and AMAP have generally not focused on measurements of PFASs in water. However, early measurements in lake and seawaters were made in Scandinavia under the Nordic Council of Ministers (Kallenborn et al., 2004). The International Council for Results for PFAS from the North Sea and the Baltic from 2005 to 2017 from the Exploration of the Sea (ICES) database were included (ICES, 2020).

4.2.4 Programmes/activities related to other media

Following the same logic that was used in the 2009 and 2015 GMP reports, the information relating to data concerning POPs monitored in other media is based on a brief overview of the published outcomes from a number of well established long term monitoring programmes (AMAP, NCP, Great Lakes, HELCOM, OSPAR and MEDPOL) and published data from Antarctica. The data reported cover legacy POPs: PBDEs, PFOS, endosulfan, HBCD, PCNs, SCCPs and PCDD/Fs.

These programmes have well established QA/QC procedures and publish regular updates on the results in the regions of interest. The uninterrupted work of these monitoring programmes since the early 1990s has yielded an extraordinary corpus of knowledge concerning the presence and pathways of POPs in the abiotic and biotic environments. To a large extent the collections of consistent monitoring data reported by the programmes and briefly summarized here have been deposited in recent years in publicly accessible repositories (www.amap.no, www.helcom.fi, and others) opening up and enabling a vast new field of research for environmental scientists in years to come.

The continued existence of these long term programmes and data collections is indispensable to improve accuracy of assessments and the effectiveness of policies. AMAP has contributed to this GMP report an extensive study on trends in POPs monitored in biota.

4.3 Antarctica

The Norwegian Antarctica Station of Troll continues to report results since its establishment in 2007 (Chapter 5.1) and measurements are ongoing. Air monitoring information from 2010-2015 is available at the Australian station of Casey. Baseline data and time trends of air concentrations of POPs in Antarctica reported in recent scientific literature at various locations are also included to complement results reported by the Norwegian and Australian programmes.

Because human exposure of POPs is primarily derived from diet and all provisions at Antarctic field stations will be imported, human exposure studies would not have value in the context of effectiveness evaluation.

There is currently no comprehensive reporting on POPs in other media from the Antarctic Region, and information provided in this report is based on a literature review. As the Scientific Committee on Antarctic Research programme in Input pathways of Persistent Organic Pollutants to Antarctica (SCAR ImPACT) is aiming for establishing an Antarctic Monitoring and Assessment Programme (AnMAP), the situation on reporting and data availability is hopefully improved by the next GMP report.

The ROG believes that in future evaluations the engagement of the Antarctic Treaty and the Scientific Committee of Antarctic Research (SCAR) would be very beneficial.

4.4 Data handling and preparation for the regional monitoring report

Most of the measurements of POPs that are available across the WEOG region originate from a small number of existing programs. In many cases these programs have been operating for more than a decade and exercise well established QA/QC protocols for sample collection and analyses. These methods are consistent with the standards outlined in the GMP guidance document. Scientists and researchers involved with these programs are at the forefront of the field and employ what are deemed to be the most appropriate data analysis methods based on the latest scientific expert opinion.

The WEOG ROG has therefore relied on these programme experts for summarizing information on temporal trends of POPs in their respective monitoring programs. Additional details on data analysis and interpretation is provided in the Annexes to this report and/or is referenced to program reports and publications.

Many of the WEOG region programs continue to deliver data to long term databases (e.g. EBAS). These databases are integral for making the monitoring data available to modelers for assessing regional and global transport of POPs, making connections between emissions and environmental burdens, and for exploring climate change and climate variability effects on POPs.

The GMP data warehouse is recognized as a useful tool for archiving GMP data and for comparing data and data availability across different regions. WEOG monitoring programs that are not already contributing data to existing databases have been encouraged to submit data to the GMP warehouse.

4.5 Preparation of the monitoring reports

The ROG drafted its own report, with the assistance of several chapter leads: Hayley Hung, Linda Linderholm, Derek Muir, Katrine Borgå, Ramon Guardans, Matthew Macleod and Tom Harner. The drafting responsibilities were as follows: Executive Summary (Tom Harner/Sara Broomhall and all) , and Chapters 1-4 updates (Tom Harner and all); Chapter 5 – 5.2.1 Air (Hayley Hung); 5.2.2 Human tissue (Linda Linderholm); 5.2.3 Water (Derek Muir, supported by Luc Miaz); 5.2.4 Other Media (Katrine Borgå/Ramon Guardans, supported by Malin Røyset Aarønes); 5.3 Long range transport (Matthew Macleod, Michael McLachlan, Li Li, Jing Li, Xianming Zhang, Elsie Sunderland, and John Hader); and Chapter 6 Conclusions and Recommendations (Tom Harner and all). ROG members Sara Broomhall and Peter Korytar assisted with the review of the report. The WEOG report also benefitted from the contributions of Simon Wilson (AMAP).

5 RESULTS

5.1 The results in context

This 3rd GMP report for the WEOG region builds on earlier reports completed in 2009 and 2015. The 1st GMP report in 2009 established baseline information for the original POPs in the core media of air and human milk and/or blood. It was concluded that geographic coverage for the WEOG region as presented in the 2009 report was adequate for reporting baselines for most POPs. It was also recognized that some sub-regions would benefit from supplemental monitoring efforts.

The 2nd report in 2015 reported temporal trends for listed POPs, where available, as well as baseline data for newly listed POPs. A few new programs that were implemented in the WEOG region since 2009 have been helping to improve spatial resolution for POPs reporting across the region.

The 2nd report also included a summary of PFOS levels in water. Water had been included as a second tier medium for the GMP, for the first time.

For this 3rd GMP report (and for subsequent monitoring reports), the WEOG region continues to rely on the long-term programs for deriving temporal trends for the older POPs and for baseline measurements for some of the newly listed POPs.

This 3rd report takes further advantage of the wealth of data from existing programs that report POPs in non-core media (“Other Media”) for generating additional information on baselines and temporal trends in concentrations for older and newly listed POPs and changes in the exposure pathways driven by changes in ecosystem structure and function.

5.2 Review of concentrations and their changes over time in the regions

5.2.1 Ambient air

5.2.1.1 Background

This chapter summarizes temporal trends and levels of POPs measured in air in the WEOG region. For the past two decades, multiple air monitoring networks provided air measurement data for various POPs. The results from the networks listed in **Table 5.2.1a** are presented in this chapter.

Table 5.2.1a Monitoring programmes which contributed to the assessment of POP temporal trends and levels (23 priority substances) in the WEOG region.

Monitoring programme	Abbrev.	Region of interest	Current # of monitoring sites in WEOG	Monitoring period	Monitored compounds (Stockholm conv. POPs)
Arctic Monitoring and Assessment Programme ^a	AMAP	Arctic	8 long-term 1 satellite	1992 – present (start dates vary among stations)	Chlordanes, PBDEs, HCB, α - and γ -HCH, DDTs at all stations In addition: Alert reports: aldrin, dieldrin, endrin, heptachlor, HBCDD, β -HCH, HCB, mirex, PeCB, PCA, PCBs, PFOA and precursors, PFOS and precursors, α - and β -endosulfan, endosulfan sulfate (screening of SCCPs) Stórhöfði reports: dieldrin, β -HCH, PCBs, toxaphene Zeppelin reports: HBCDD, β -HCH, PCBs, PFOA and precursors, PFOS and precursors, SCCPs Andoya reports: HBCDD, β -HCH, PCBs, PFOA and precursors, PFOS and precursors Pallas reports: PCBs, α -endosulfan Station Nord reports: aldrin, dieldrin, endrin, heptachlor, β -HCH, PFAS precursors, α - and β - endosulfan Tiksi and Amderma report: aldrin, dieldrin, endrin, heptachlor, mirex, PCA, PCBs Little Fox Lake (satellite station) reports: aldrin, dieldrin, endrin, α - and β -endosulfan, HBCDD, HCB, HCHs, heptachlor, PBDEs (Aug 2011 – Dec 2014)
Australia's Casey Station	Casey	Antarctica	1	2010-2015	PeCB, HCB, dieldrin, aldrin, endrin, heptachlor, chlordanes, toxaphene, endosulfan, mirex, HCHs, DDTs, PCBs
Australia's National Passive Air Sampling and Archiving Program	Australia-PAS	Australia	44	2011-present	PCBs, PeCB, HCB, HCHs, heptachlor, aldrin, dieldrin, endrin, chlordanes, DDTs, mirex (2012 at 15 sites only), α - and β -endosulfan, SCCPs (2016 at 15 sites only) Samples from other years and sites are archived (not analysed).
European Monitoring and Evaluation Programme	UNECE-EMEP	Europe	12	1993 - present	PCBs, DDTs, chlordanes, HCB, HCHs, PBDEs Note that AMAP sites Zeppelin, Pallas, Stórhöfði, Station Nord/Villum Research station, Amderma are also EMEP sites. Please see additional analytical compounds above under AMAP.

Table 5.2.1b continued...

Monitoring programme	Abbrev.	Region of interest	Current # of monitoring sites in WEOG	Monitoring period	Monitored compounds (Stockholm conv. POPs)
Global Atmospheric Passive Sampling network	GAPS	Global	46	2004 - present	dieldrin, chlordane, heptachlor, DDTs, α - and β -endosulfan, endosulfan sulfate, PCB, chlordanes, HBCDD, HCB, PeCB, HCHs, PBDEs, PFOS and precursors, PCDD/Fs (2011, 2012 only)
Great Lakes Basin Monitoring and Surveillance Program	GLB	Great Lakes (Canada)	2 master stations; 1 satellite station	1992 - present	aldrin, endrin, dieldrin, chlordanes, DDTs, heptachlor, HBCDD, mirex, PCBs, HCHs, α - and β -endosulfan, endosulfan sulfate, PBDEs, SCCPs
Integrated Atmospheric Deposition Network	IADN	Great Lakes (US)	3 master stations; 2 urban stations	1990 - present	aldrin, chlordanes, DDTs, dieldrin, heptachlor, α - and β -endosulfan, endosulfan sulfate, HCHs, PCBs, chlordanes, HCB, PBDEs, HBCDD, endrin, dieldrin
MONitoring NETwork (Europe)	MONET (EU)	Europe	23	2009-present	PCBs, HCHs, DDTs, HCB, PeCB, chlordanes, aldrin, dieldrin, endrin, heptachlor. Analysed but not yet reported: PCDDs/Fs and PBDEs
Monitoring Network in the Alpine Region for Persistent and other Organic Pollutants	PureAlps (formerly MONARPOP, POPAlp, EMPOP, VAO II)	European Alpine regions	2	2005 - present	PCB, chlordanes, HCB, PCDD/F, HCB, α - and β -endosulfan, aldrin, dieldrin, endrin, DDTs, HCHs, PeCB, DBDPE, heptachlor, mirex, PBDEs, HBB, HBCDD, endosulfan sulfate
National Air Pollution Surveillance	NAPS	Canada (Urban)	29 urban, 8 rural and 1 suburban sites	1989-2009	PCDD/Fs, co-planar PCBs (2004-2009)
Northern Contaminants Program (Canada) ^a	NCP	Arctic	1 long-term; 1 satellite	1992 - present	Alert reports: aldrin, dieldrin, endrin, heptachlor, HBCDD, HCB, β -HCH, HCB, mirex, PeCB, PCA, PCBs, PFOA and precursors, PFOS and precursors, α - and β -endosulfan, endosulfan sulfate (screening of SCCPs) Little Fox Lake (satellite station) reports: aldrin, dieldrin, endrin, α - and β -endosulfan, HBCDD, HCB, HCHs, heptachlor, heptachlor, chlordanes, DDTs, PBDEs (Aug 2011 – Dec 2014)
Norwegian Troll Station	Troll	Antarctica	1	2007-present	PCBs, HCB, HCHs, DDTs, chlordanes, PBDEs (periodically), SCCPs (periodically)
Spanish Monitoring Program on POPs	SMP-POPs	Spain	23	2008-present	PCDD/Fs, PCBs, PBDEs, HCHs, HCB, DDTs, PeCB, α - and β -endosulfan

Table 5.2.1c continued...

Monitoring programme	Abbrev.	Region of interest	Current # of monitoring sites in WEOG	Monitoring period	Monitored compounds (Stockholm conv. POPs)
Swedish National Monitoring Programme for Air ^b		Sweden	3	1996-present	PCBs, HCB, HCHs, chlordanes, DDTs, PFOS, PFOA, PFOSA, PBDEs, endosulfan, HBCDD, Dioxin and furans, SCCPs (2 sites) Aldrin, dieldrin, heptachlor (1 site)
Toxic Organic Micro-Pollutants Air Monitoring and Analysis Network	TOMPs	UK	6	1991 - present	PCDD/F, co-planar PCB, PCBs, PBDEs (2010-present)
UK/Norway SPMD Transect		UK and Norway	13	1994 - present	PCBs, PBDEs

^a The Northern Contaminants Program (NCP) is the Canadian National Implementation Plan of Arctic Monitoring and Assessment Programme (AMAP). Data from 9 AMAP stations are presented in the current assessment, within which 1 long-term (Alert, NU Canada) and 1 satellite (Little Fox Lake, YK Canada) stations are operated under NCP.

^bThe Swedish Monitoring Programme operates 3 long-term stations, Råö, Aspvreten/Norunda, Pallas, for the monitoring of POPs. These stations are also part of EMEP and Pallas is part of AMAP as well.

In total 16 atmospheric monitoring programmes contributed results to this assessment.

5.2.1.2 Contributing Air Monitoring Programmes

This section provides a brief overview of the key programmes that have contributed data and information to the air chapter of the present report. **Figure 5.2.1** shows the locations of currently-operating sites where measurements were made under these programmes. Details on the objectives, sampling methods, operation and analytical information and spatial coverage of the various monitoring programmes are included in the programme executive summaries given in Annex 1.

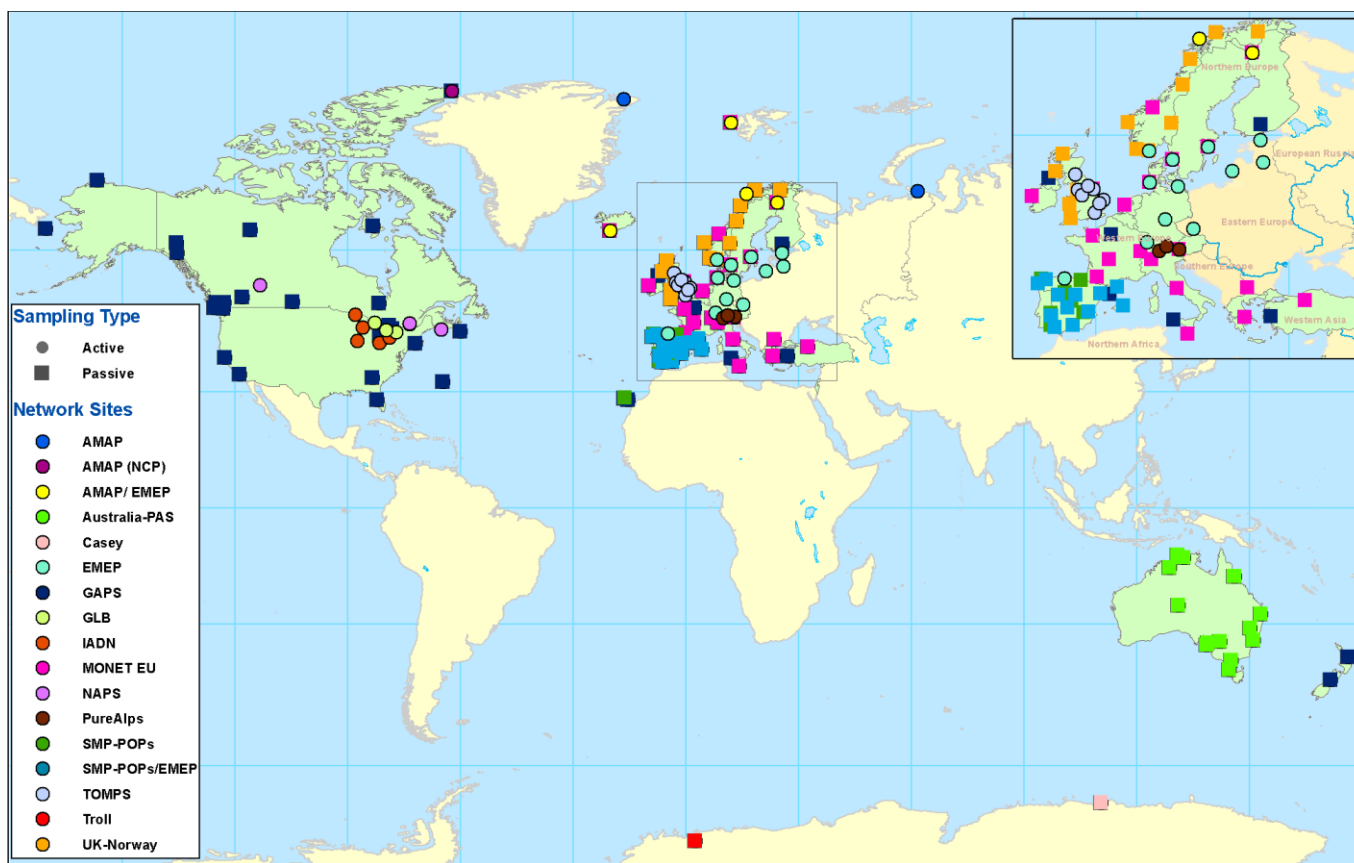


Figure 5.2.1 Site map for air monitoring stations of POPs that reported results for this monitoring period in the WEOG region. WEOG region is shaded in pale green. See **Table 5.2.1a** for definition of programme acronyms. Symbol shapes represent sampling types (passive ■, active ●) and symbol colors represent programme/network.

Arctic Monitoring and Assessment Programme (AMAP) and the Northern Contaminants Program (NCP)

The Arctic Monitoring and Assessment Programme (AMAP) is a Working Group of the Arctic Council, with a mandate to:

- monitor and assess the status of the Arctic region with respect to pollution and climate change issues;
- document levels and trends, pathways and processes, and effects on ecosystems and humans, and propose actions to reduce associated threats for consideration by governments;
- produce sound science-based, policy-relevant assessments and public outreach products to inform policy and decision-making processes.

AMAP is based largely on ongoing national monitoring and research activities of eight Arctic countries on pollutant issues. AMAP coordinates these activities and works to ensure harmonization, to promote quality assurance activities, and to compile results for use in circum-Arctic assessment activities.

The NCP is Canada's National Implementation Plan for AMAP. It features the longest time series for atmospheric POPs at the North American Arctic station of Alert (Canada).

An executive summary of results from AMAP for air, biota and human tissues for this reporting period is given in Annex 1.

Australia's Casey Station (Antarctica)

POPs were monitored at the Australian Antarctica station of Casey from December 2009 to November 2014. A flowthrough air sampler developed for sampling of POPs in cold, remote environments (Xiao et al., 2007) was used at the abandoned Wilkes Station, 3 km north of Australia's Casey Station (66°16'56"S, 110°31'32"E) (Bengston Nash et al., 2017). The easterly prevailing wind direction ensured that there was minimal influence from the local Casey station. A total of 70 POPs were analyzed in 33 sets of samples over 5 years and results contribute to this assessment.

Australia's National Passive Air Sampling and Archiving Program

Australia's National Passive Air Sampling and Archiving Program deploys XAD-based passive air samplers (XAD-PASs) once a year since 2011 to collect air samples at 44 sites across Australia, including 15 background, 14 agricultural and 15 urban/industrial sites over tropical, subtropical, arid/semiarid, and temperate climate zones. Samples are being archived and were analyzed for specific compounds during selected years. Results from measurements of SCCPs in ambient air samples collected in 2016 at 15 (5 remote, 6 rural and 4 urban) sites (Annex 1), and PCBs and OCPs at 15 (4 remote, 5 agricultural, 1 semi-urban and 4 urban) sites (Wang et al., 2015) are included in this assessment. Trend data can become available in the future when additional archived samples are analysed to assess changes in concentrations with time.

European Monitoring and Evaluation Programme (EMEP)

EMEP is a scientifically based and policy driven programme under the Convention on Long-range Transboundary Air Pollution (CLRTAP) for international co-operation to solve transboundary air pollution problems. The Chemical Coordinating Centre EMEP (EMEP-CCC) was established at NILU (Norwegian Institute for Air Research) in 1977 to harmonize previous international monitoring efforts. EMEP monitors POPs but also heavy metals, ozone, particulate matter, polycyclic aromatic hydrocarbons (PAHs) (as part of POPs monitoring), volatile organic compounds (VOCs) etc. In total, 52 sampling sites have been involved in POPs monitoring in air or precipitation samples, for short, or longer periods, as summarized in <http://www.nilu.no/projects/ccc/emepdata.html>. In total, about 120 discrete POP substances (excluding the PAHs) are reported to EMEP, although the number differ from site to site.

Global atmospheric passive sampling (GAPS) network for POPs

The GAPS network conducts global-scale measurements of POPs in air at more than 50 sites since 2005. Its objectives are to i) demonstrate the feasibility of passive air samplers (PAS) for POPs; ii) determine spatial and temporal trends for POPs in air; and iii) contribute useful data for assessing regional and global long-range atmospheric transport of POPs. Results from 46 stations under this network in the WEOG region are reported here.

The Great Lakes Basin (GLB) Monitoring and Surveillance Program

Air and precipitation monitoring of POPs in the Canadian Great Lakes Basin (GLB) was established in 1990, in collaboration with United States' Integrated Atmospheric Deposition Network (IADN), and supports the Canada/US Great Lakes Water Quality Agreement. The core objectives of GLB are to: a) acquire quality-assured air and precipitation concentration measurements of POPs; b) determine atmospheric loadings of POPs to the Great Lakes System and define the temporal and spatial trends of such atmospheric deposition; c) determine the sources of continuing inputs of these pollutants. Measurement results from two regionally-representative Master Stations, namely Point Petre on Lake Ontario and Burnt Island on Lake Huron (discontinued after March 2013); and a rural/suburban station Egbert on Lake Huron are included in this assessment.

Integrated Atmospheric Deposition Network (IADN)

The United States' Integrated Atmospheric Deposition Network (IADN) collaborates with Canada's GLB Monitoring and Surveillance Program to conduct air and precipitation measurements in the Great Lakes region. The long-term data set has allowed the determination

of POP loadings; development of reliable spatial and temporal trends; identification of sources and/or source regions; and tracking of overall chemical management efforts. Measurement results from 3 US Master Stations (Eagle Harbor on Lake Superior, Sturgeon Point on Lake Erie and Sleeping Bear Dunes on Lake Michigan) and 2 urban stations (Chicago and Cleveland) obtained between 1990 and 2015 are included in the current assessment. Additional data are available at the IADN Data Visualization Tool (<https://iadnviz.iu.edu/>).

The MOnitoring NETwork (Europe) (MONET-EU)

MONET EU was established in 2009 and is a passive air sampling network with a total of 34 stations in 26 European countries. Measurement results from 2013 to 2016 are reported here. For this reporting period, the WEOG region was represented by up to 18 sampling sites from 13 countries; of which, 17 stations were co-located at EMEP sites. Concentrations of PCDDs/Fs and PBDEs were only measured at 5 sampling sites in 2015 and 4 sites in 2016.

Norwegian Troll Station

Air monitoring of POPs started at the Norwegian Antarctic station of Troll, located on Dronning Maud Land, in 2007. High volume active air sampling method was used. The sampling site was moved in 2014 to minimize potential impacts of human activities. Also, in 2016/17, the sampling duration has been extended to two weeks (previously one week samples were taken). Also, the sampling material for the gas phase has been changed from a polyurethane foam (PUF) to a PUF/XAD/PUF sandwich at this time to improve detection and reduce breakthrough which may affect the concentrations and trends measured for more volatile compounds (e.g. HCB). Data were reported from 2007 to 2018 for this reporting period. Trends were reported for two periods: (i) 2007-2018 and (ii) 2007-2015 when the sampling duration and gas-phase sampling medium were changed. These changes have resulted in higher concentrations detected and affected the long-term trends and half-lives.

Monitoring Network in the Alpine Region for Persistent and Other Organic Pollutants (PureAlps)

PureAlps and its predecessor programmes starting in 2005 with MONARPOP aim to provide spatial and altitudinal monitoring information of POPs and other organic pollutants to decision makers in the European Alpine regions. The objective of the programme is to assess the exposure of the alpine environment to globally transported POPs. Key features of the programme are ambient air measurements of POPs and bulk deposition measurements that cover the years 2005 to 2017/2018. The project PureAlps (2016-2020) extended the data coverage on accumulation of POPs in the alpine biosphere (mammal herbivores, mammal predators, birds of prey, fish, and

insects). Air and bulk deposition monitoring were conducted using active sampling methods at the Environmental Research Station Schneefernerhaus at Mt. Zugspitze (“UFS”, Bavaria/Germany, 2650 m a.s.l.; coordinates: 10.9850 East, 47.4211 North) and the Sonnblick Observatory at the Hoher Sonnblick (“SBO”, Austria, 3106 m a.s.l.; coordinates: 12.9575 East, 47.0539 North). A third sampling site at Weissfluhjoch/Switzerland contributed samples until 2015. Temporal trends from 2005 to 2018 in air for Stockholm Convention-listed POPs PCDD/Fs, PCBs, several OCPs, PBDEs (2012-2018) and HBCD (2012-2018) are included in this reporting period.

Spanish monitoring Program on POPs (SMP-POPs)

The Spanish Monitoring Program on POPs (SMP-POPs) was established in 2008 and started with air monitoring using the passive sampling method at 12 remote sampling sites co-locating at EMEP stations. After two expansions in 2009 and 2011, the programme collected passive air samples at 23 locations (14 remote EMEP sites, 9 urban sites) in Spain between 2008 and 2019. Measurements are ongoing. Quarterly samples are taken and were analysed for 69 analytes.

The Swedish National Monitoring Programme for Air

The Swedish National Monitoring Programme for Air is operated by the Swedish Environmental Protection Agency. This programme operates 3 rural background long-term stations for POPs, Råö, Aspvreten and Pallas (Finland). These stations are also part of EMEP and Pallas is part of AMAP.

UK Toxic Organic Micro Pollutants (TOMPs) programme

TOMPs (Toxic Organic Micro Pollutants) is a monitoring network which measures the atmospheric concentrations of PCBs, co-PCBs, PAHs, PBDEs and PCDD/Fs within the U.K. since 1991 using the active sampling method. PBDEs were added to the target chemical list since 2010. Currently, the monitoring sites include 2 urban (London and Manchester) and 4 rural stations (Hazelrigg, Auchencorth, Weybourne and High Muffles). Two stations (Middlesbrough and Stoke Ferry) have ceased operation. Data are reported quarterly and temporal trends were reported for all stations since the early 1990s.

The UK – Norwegian transect programme

In 1994, a latitudinal ambient air sampling transect from the south of England to the north of Norway (UK-Norway transect) was established with eleven sampling sites, mainly in remote

locations, using a passive air sampling method. The network, now consisting of 13 sites, provides continuous, long-term ambient air trend data for a range of POPs, including PCBs and PBDEs

Other than these major programmes, additional concentration and trend data are obtained from peer-reviewed scientific literature or technical reports to compliment results reported.

Please note that several air monitoring programmes, e.g. EMEP, GLB, IADN and Swedish National Monitoring Programme for Air, also include POP measurements in precipitation. The Great Lakes programmes, namely GLB and IADN, conduct estimations of atmospheric loadings via wet and dry deposition to the lakes periodically (e.g. Guo et al., 2018). These datasets are not included in this report but can be useful in future assessments if air concentration data are not available or there is a need to determine changes in atmospheric loadings to certain sensitive ecosystems, e.g. the Great Lakes.

5.2.1.3 Levels and Trends of POPs in the WEOG Region

For evaluation of long-term temporal trends, a decline in air concentrations over time is often quantified by an apparent first order half-life or halving-time, $t_{1/2}$, which refers to the time required for the air concentration of a chemical to decline to half its original value. $t_{1/2}$ is estimated by dividing $\ln 2$ by the negative value of the linear regression slope of the time trend between the natural log of air concentrations, C (pg/m^3), and time (year). Similarly, for increasing time trends, doubling times, t_2 (or negative $t_{1/2}$) can be estimated. Note that half-life in this context assumes that air concentrations decline exponentially as a result of atmospheric clearance processes (e.g. photodegradation and deposition) which differs from radioactive decay that refers to the unstable decay of radioactive atoms themselves. Many compounds do not necessarily decline/increase linearly or consistently in the first order manner throughout the monitoring periods. Half-lives or doubling times are only presented here when the trends are declining or increasing more or less linearly and consistently. These values are only used to compare the relative rates of decline between stations or regions. Readers are advised to use the absolute values of these half-lives or doubling times with caution. **Table 5.2.1ab –Table 5.2.1e** summarize $t_{1/2}$ and t_2 (represented as negative $t_{1/2}$) observed for different chemicals and regions. Some programmes only reported whether the trend of a certain chemical is increasing, decreasing or not changing without providing a quantitative rate of change, such trends are indicated only by color in **Tables 5.2.1b-Table 5.2.1e** (green = decreasing; red = increasing; blue = not increasing or decreasing). The focus of the current assessment is on the temporal trends of POPs. Only rarely would actual concentration values be presented here. Actual

measured values reported by various programs can be found in the programme executive summaries given in Annex 1.

Aldrin, Endrin and Dieldrin

Few monitoring programmes report levels of aldrin, endrin and dieldrin in air. Dieldrin was declining in air at $t_{1/2}$ of 10 y at both Great Lakes stations of Point Petre on Lake Ontario (1992-2017) and Burnt Island on Lake Huron (1992-2012). A slower decline for dieldrin was observed at the satellite station of Egbert on Lake Ontario (1995-2006) ($t_{1/2}$ = 18 y). Declines were also observed for aldrin at all 3 sites with slightly shorter $t_{1/2}$ of 6.2-7.6 y. The Arctic stations of Alert (Canada) and Stórhöfði (Iceland) reported time trends of dieldrin since 1992 and 1995, respectively, and have shown declining trends with $t_{1/2}$ of 17 and 12 y, respectively. At Station Nord, its half-life was extremely long (>100 y). The long half-lives of dieldrin at these stations indicate that its level will likely remain consistent for an extended time. No trend was reported for aldrin as it is detected in less than 10 % of the samples at Alert and Station Nord. Air concentrations tended to be lower in the colder months and higher in the warmer months for both Great Lakes and Arctic sites. Slight decreases in the warmest months were observed for dieldrin in all years at Alert which may be related to greater photodegradation during Arctic summer under 24 hour sunlight. Dieldrin in air probably results from re-emission from historical sources and the use of aldrin, which can be converted to dieldrin in the environment. This indicates that dieldrin in air has probably reached some sort of steady state with other environmental media as it has been banned under various national and international initiatives worldwide for more than 30 years.

In Australia (2012), dieldrin showed relatively high concentrations at some sites (ND-160 pg/m³, median concentration at 15 sites 12 pg/m³) measured with passive air samplers. The highest concentrations were found at urban sites and, spatially, a clear increasing trend was found from background to agricultural and semi-urban to urban sites (Wang et al., 2015).

Chlordanes, Nonachlor, Methoxychlor and Oxychlordane

Declining or non-changing trends of *trans*- and *cis*-chlordane were observed at all Arctic stations, with $t_{1/2}$ ranging from about 7.9-25 y for *cis*-chlordane and 3.2-13 y for *trans*-chlordane (excluding that of Station Nord, Greenland, which showed non-changing trends). The *trans*-chlordane/*cis*-chlordane ratio at the Arctic sites Alert, Zeppelin, Stórhöfði, and Pallas were lower than 1.6, implying that chlordanes measured at these sites were from aged sources.

The concentrations of *trans*-nonachlor exhibited long declining rates at all Arctic stations with half-lives ranging from 10 to 36 y. Similarly, air concentrations of *cis*-nonachlor were slowly

declining with half-lives ranging from 8.5 to 12 y. *Cis*-nonachlor in air at Station Nord was climbing with a doubling time of 6.7 y. At Station Nord, it appeared that *cis*-nonachlor began to increase in 2013 with unknown reasons; but concentrations were mostly below detection limits in 2017 and 2018, except for a few high concentration episodes. Measurements need to continue at this site to better understand this trend. The slow declines of *trans*- and *cis*-nonachlor are likely reflections of their persistence in the Arctic and tendencies towards equilibrium with surface media.

At the 5 Great Lake background sites representing each of the lakes, $t_{1/2}$ ranged from 9.4-52 y for individual isomers on Lakes Ontario and Huron; and half-lives for Σ chlordanes (*trans*- and *cis*-chlordanes and *trans*-nonachlor) on Lakes Erie, Michigan and Superior in the vapour phase ranged from 8.7 to 11.7 y (Salamova et al. 2015). At the urban Great Lakes sites of Chicago and Cleveland, Σ chlordanes $t_{1/2}$ in the vapour phase ranged from 9-11.6 y similar to those found at remote sites. Slow declines for *cis*- and *trans*- chlordanes have been recorded also in the Swedish-operated stations of Råö and Pallas (annual decrease of 3-4 %) (Anttila et al., 2016; Fredricsson et al., 2018). Methoxychlor seems to show a declining tendency in air in the Great Lakes but few detectable data were available after 1995 to develop a reasonable trend.

Oxychlordanes, as the major metabolite of the chlordanes and nonachlors, showed consistent declining trends on Lakes Ontario and Huron with $t_{1/2}$ ranging from 8.5-71 y. Globally, the GAPS network reported no consistent increasing or decreasing tendencies for chlordanes or heptachlor from 2005 to 2017 and noted that their concentrations were steady and declining less rapidly over time at the majority of the sites. Similar to the drins, slow declines in the air with $t_{1/2}$ mostly longer than 10 years for all chlordanes and nonachlor-related isomers reflects the fact that chlordanes have been banned in the WEOG region since the 1980s and time trends seem to indicate a tendency towards equilibrium with other media.

DDTs

In Arctic air, DDTs at all sites were dominated by the degradation product, *p,p'*-DDE, which is indicative of old DDT residues. Concentrations of *p,p'*-DDE were decreasing or not changing at most sites with half-lives of 8.4 to 42 y. *o,p'*-DDE showed decreasing trends at most sites with half-lives ranging from 7.6 to 17 y. At Station Nord, *o,p'*-DDE increased between 2009 and 2012 but then stabilized from 2012-2017. Air concentration of *p,p'*-DDT at all Arctic stations, except at Station Nord, were declining with half-lives ranging from 3.4 to 20 y. *p,p'*-DDT at Station Nord was slightly increasing but with a very long doubling time of 20 y. *o,p'*-DDT was declining with half-lives of 7.0 to >100 y. DDT air concentrations at most Arctic sites seem to have reached steady state.

For the Great Lakes sites, consistent declining trends can be found for almost all isomers from 1992 to 2017. The half-lives of all DDT isomers on Lake Ontario and Lake Huron ranged from 7.2-38 y. On Lakes Michigan, Erie and Superior for both background and urban sites, the half-lives ranged from 9.1-12.6 y for Σ DDTs in the vapour phase (Salamova et al., 2015).

The global programme of GAPS reported no consistent increasing or decreasing tendency for DDTs from 2005 to 2017 and noted that DDTs show different trends depending on sampling region with some examples of recent use.

Under the MONET EU programme (2013-2016), the highest levels of DDTs were detected in the Netherlands and Malta and the lowest concentrations were measured in Pallas (Finland) and Le Montfranc (France). DDTs were found to be significantly decreasing at most MONET EU stations (Kalina et al., 2019). Under the EMEP programme, all sites have experienced a decline in concentrations in air over the entire monitoring period, with half-lives ranging from 5.5 to 131 y. However, from the start of the programme to 2004 (when the Stockholm Convention entered into force) and 2004-2017 (after regulation), there were sites experiencing both increasing and decreasing concentrations of p,p'-DDT. Notably, while most sites showed declining trends from 2004 to 2017 with half-lives ranging from 6.1-11 y, Stórhöfði (Iceland) showed a slight increasing trend with a doubling time of -21 y. It is possible that there are other contributions of DDTs to the site, such as re-emissions of formerly deposited DDTs from melting glaciers, but the detection frequency of the DDTs at this site was low since 2015.

At the Swedish-operated stations of Råö and Pallas, downward trends of p,p'-DDE and p,p'-DDT were observed in air with a reduction of about 2-5 % per year (1996-2011) (Anttila et al., 2016). With more data up to 2017, it was found that DDE concentrations were higher than those of DDT and DDD at the Swedish stations, and the trends of p,p'-DDT can no longer be derived (Fredricsson et al., 2018).

At the Antarctic station of Troll, p,p'-DDE showed a consistent declining trend (Kallenborn et al., 2013) with a half-life of 8.4 y between 2007 and 2018 and 5.0 y between 2007 and 2015 (after which the sampling duration has been changed from weekly to biweekly and the gas-phase sampling medium changed from a PUF to PUF/XAD/PUF to increase sampling capacity). Declining trend of Σ DDTs (half-life of 2.4 years) was also observed at 7 passive sampling sites close to Chinese Great Wall Station (Dec 2010 - Jan 2018) in West Antarctica (Hao et al., 2019).

Heptachlor and Heptachlor Epoxide

Heptachlor and heptachlor epoxide in Arctic air were reported for Alert (Canada) and Station Nord (Greenland) and their time trends show that the concentrations of both compounds are remaining steady. At Alert, concentrations of these compounds become mostly non-detectable

after 2010 and, thus, their measurements were discontinued after 2012. Similarly, between 2009 and 2017, heptachlor has only been detectable in 2 air samples in 2010 at the Swedish station of Råö (Fredricsson et al., 2018). Under the MONET programme, the levels of heptachlor were found to be very low for all sampling sites (2013 – 2016) with highest concentrations determined in Mace Head (Ireland) (0.20-2.57 pg/m³) and Peyrusse Vieille (France) (0.05-1.82 pg/m³). For three Great Lakes sites on Lakes Huron and Ontario, continuous declining trends for both compounds were observed, with $t_{1/2}$ ranging from 4.7 to 12 y.

Hexachlorobenzene (HCB)

HCB in air was reported at all Arctic stations, however, no significantly increasing or decreasing trends were observed, except for Pallas (Finland) where there was a rapid increase between 2008 and 2010 with no specific reasons. All other Arctic sites showed extremely long half-lives or doubling times (>30 y) indicating that HCB levels remained stable over time. Although it is known that HCB has a tendency to breakthrough in air samples collected using polyurethane foam plugs (PUFs), using Bidleman and Tysklind's (2018) method to estimate the collection efficiency based on the frontal chromatography theory showed that the mean collection efficiency for HCB at Alert (Canada) is 90 % indicating that HCB is mostly captured. HCB was not reported for the Canadian Great Lakes stations because of potential breakthrough during sampling with a single PUF. It was also not reported in the US Great Lakes region during this reporting period.

As part of the EMEP programme, HCB has been monitored at 12 stations. It was noted that HCB was often observed at elevated concentrations in the Arctic compared to other southerly European sites. Most sites showed an increase in or a steady-state condition of HCB concentrations in air in the period after the Stockholm Convention entered into force (i.e. after 2004). However, prior to 2004, the three EMEP sites with available data (Birkenes, Zeppelin and Stórhöfði) experienced decreasing concentrations of HCB (Colette et al., 2016) (Figure 5.2.2 for Zeppelin and Stórhöfði). Both sites experienced a decline before the Stockholm Convention entered into force, and a minor increase after 2004 when the Stockholm Convention entered into force. HCB is an unintentional by-product from combustion, and the contribution from secondary emissions into air may also be important due to its high volatility and slow degradation in air.

The PureAlps network also found non-changing trends of HCB at their two high elevation sites on the European Alps with a relatively high median concentration of 94 pg/Nm³ (normalised air volume) at both sites (2005-2017). No trends can be developed at the 3 Swedish-operated monitoring stations of Råö, Aspöreten and Pallas either (Fredricsson et al., 2018).

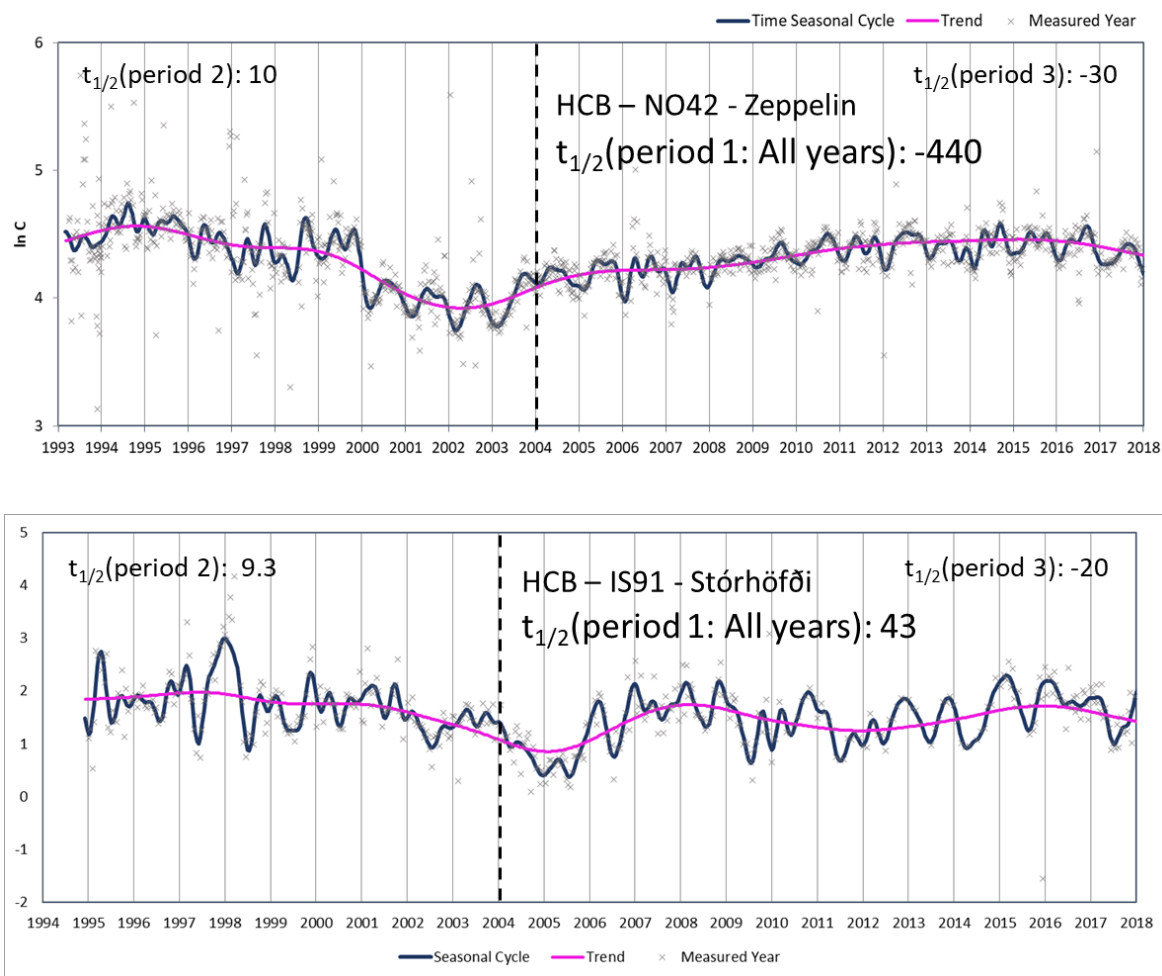


Figure 5.2.2 Temporal trends of HCB at two selected EMEP sites with similar duration of monitoring and long-term half-lives since the onset of measurements.

Globally, XAD passive air sampler measurements have shown reducing tendencies for HCB in air under GAPS from 2005 to 2008 (Shunthirasingham et al., 2010). A pilot study conducted under the GAPS network using sorbent impregnated PUF or SIP as sampling medium showed that HCB is fairly uniformly distributed globally with concentrations ranging from 37 to 240 pg/m^3 (Koblizkova et al., 2012).

The Spanish Monitoring Program on POPs did not find a difference between HCB concentrations measured at background versus urban sites, indicating that there is no point sources at the national level, and a significantly decreasing trend was observed from 2009 to 2017.

MONET reported HCB air concentrations at 13-16 sites in Europe between 2013 and 2016 with HCB concentrations showing highest concentrations in Austria, Iceland, Ireland and UK. The

high levels of HCB were also measured in Spitsbergen (Zeppelin, Svalbard, Arctic) in 2013 and 2014. Malta had the lowest levels of HCB and PeCB for all sampling periods in general. Kalina et al. (2019) found that there is a lack of consistency in time trends derived for HCB from collocated active and passive air samplers which may be due to ongoing emissions of HCB as a combustion by-product and the possibility of breakthrough in active sampling.

At the Australian Antarctic station of Casey (2010-2015), HCB dominated the atmospheric profiles with an average concentration of 12.6 pg/m^3 with higher concentrations in the austral summer, indicative of local, temperature-dependent volatilization. The concentration is similar to those previously measured under other campaigns and the collated dataset from multiple studies taken since the 1990s showed that HCB concentrations seemed to have plateaued (Bengston Nash et al., 2017). Hao et al. (2019) also reported HCB as the most dominant organochlorine found in air with no changes in trends from December 2010 to January 2018 for annual passive air sample measurements taken at 7 sites close to the Chinese Great Wall Station in West Antarctica. Pozo et al. (2017), who collected passive air samples along the Ross Sea in Antarctica (Dec 2010-Jan 2011), also noted that the air concentrations of HCB remained unchanged when comparing their results with historical measurements.

In contrast, an increasing trend for HCB was observed at the Norwegian Antarctic station of Troll, with a doubling time of 19 y (2007-2018) and 22 y (2007-2015) before changing the sampling duration to 2 weeks and the sampling medium from 1 PUF to a PUF/XAD/PUF sandwich.

Mirex and Photomirex

Air concentrations of mirex and its degradation product photomirex were reported at the Great Lakes stations of Burnt Island and Egbert on Lake Huron and Point Petre on Lake Ontario. Both compounds showed declining trends with $t_{1/2}$ ranging from 8 to 28 y for mirex, and 4.6 to 9.4 y for photomirex. At the two high elevation sites on the European Alps, the median concentration of mirex was found to be 0.08 pg/Nm^3 (normalised air volume) with non-changing trends. At the Australian Antarctic station of Casey, mirex was measured for the first time in air in Antarctica and the concentrations were not showing any consistent declining or increasing trend (Bengston Nash et al., 2017). Air concentrations seem to tend towards equilibrium with the environment at all measured locations in WEOG.

Polychlorinated Biphenyls (PCBs)

PCBs in air in the WEOG region tended to be generally declining in trends at all monitoring stations since the 1990's. The decline seems to have become less rapid in recent years as the concentrations became much lower resulting in longer half-lives (see Table 5.2.1b).

At the Arctic site of Stórhöfði (Iceland), individual PCB congeners in air showed a mix of increasing (PCB52, 101 and 118) and decreasing trends (PCB28, 138, 153, and 180). Nevertheless, their half-lives or doubling times were extremely long (ranged from 17 to >100 y) which indicate their levels in air were not changing. In fact, the trend plots showed that PCBs at Stórhöfði tended to increase since 2000s, which agrees with the previous report (Hung et al., 2016). Such elevated levels after 2000 could be due to the enhanced release of PCBs to the atmosphere from the melting ice caps in a warming climate around Iceland.

At the 2 summit sites in the European Alps, significant increases of ambient air concentrations at both sites can be observed for the entire spectrum of dioxin-like PCBs. However, these increases are only significant ($p < 0.1$) for PCB 105, PCB 114, PCB 118, and the sum of PCB (in TEQ) at Sonnblick (SBO) (Fig. 5.2.3). In addition, the indicator PCB 101 is also increasing significantly at SBO. These increases are inexplicable.

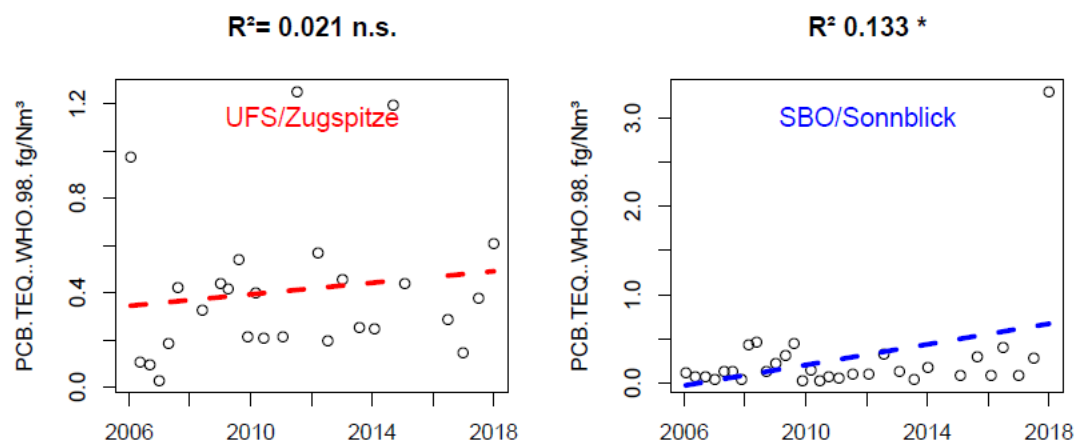


Figure 5.2.3 Ambient air concentrations for the sum of dioxin-like PCB in TEQ (WHO 1998)

Along the UK-Norwegian transect, PCB levels at all transect sites, which are mostly remote sites, decreased steadily since 1994 (Schuster et al., 2010). After 2008, no further decrease was observed. The overall concentrations declined from the south of England to the north of Norway and the highest concentrations were found at sites closer to population centres or construction work.

For temperate locations closer to urban centers, the rates of decline for PCBs at different TOMPs sites in the UK (TOMPs executive summary in Annex 1) (Graf et al., 2016) and those at the different Great Lakes sites in the US (Salamova et al., 2015) were not found to be statistically different from each other suggesting similar sources have been, and still are, contributing to the measurements. Primary sources from PCBs stocked in urban environments (e.g. in PCB transformers, electrical equipment and in landfills) still contribute to measured atmospheric levels. In the Great Lakes, declining or non-changing trends were observed at all US and Canadian sites with half-lives ranging from 8.3 to 160 y and were congener specific. The half-lives tended to be long (>10 y) for most congeners, implying the tendency towards equilibration with surrounding environmental surfaces. Co-planar PCBs were being monitored by Canada's National Air Pollution Surveillance (NAPS) (2004-2009) at 29 urban, 8 rural and 1 suburban sites which did not show an apparent decrease or increase in trends (CEC, 2014).

PCB 153 was monitored since 1993 at 11 sites under EMEP and mostly showed declining trends, except at Stórhöfði. Anttila et al. (2016) showed that PCB levels up to 2012 at the Swedish-operated sites of Pallas and Råö have on average decreased by 2-4 percent per year and are tending towards a steady state (Fredricsson et al., 2018).

Under the Spanish programme of SMP-POPs, among the PCB congeners, non-ortho-PCBs were found at the lowest concentrations, about one order of magnitude below mono-ortho and two orders of magnitude below indicator PCBs, across all 23 sites. Similar to the findings of the UK programme of TOMPs and the US IADN, spatial comparisons suggest urban populated areas as the main sources of PCBs. Concentrations significantly decreased from 2009 to 2017. Results suggest the presence of unknown sources of PCBs since their ban (Muñoz-Arnanz et al., 2018).

Under the MONET programme, the highest concentrations of the sum of 7 PCBs were observed at the EMEP station in The Netherlands (De Zilk) where a slow declining trend can be found in the period from 2013 to 2016. Kalina et al. (2019) report declining trends of PCBs at 6 MONET sites with collocated active and passive samplers.

Globally, under GAPS, measurements spanning 10 sampling years are available for 20 sites (Figure 5.2.4). Concentrations for legacy POPs in air between 2005-2014 showed that PCB concentrations in air were decreasing at most sites, though at differing rates.

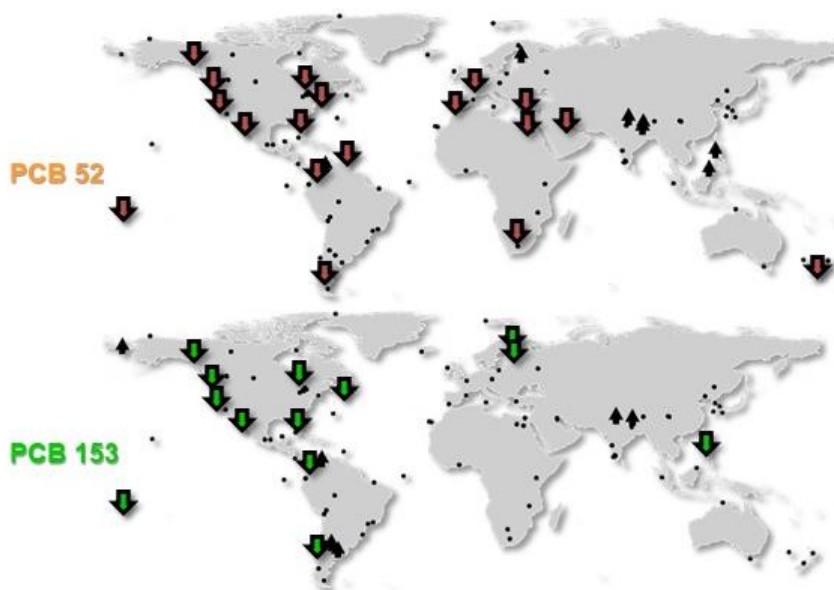
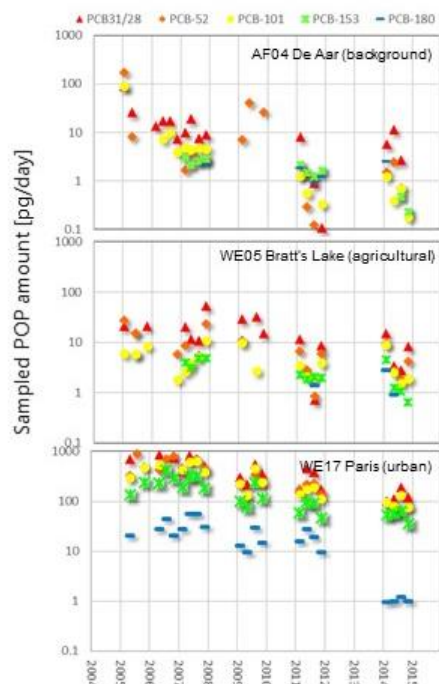


Figure 5.2.4. Global trends for PCB concentrations in air and local results for AF04, WE05 and WE17

At the Troll Antarctic station, PCB 52 showed declining trends with half-lives of 10 year between 2007 and 2018 and 7.2 year between 2007 and 2015 (when the sampling duration has changed to biweekly-integrated and the gas-phase sampling medium changed from PUF to PUF/XAD/PUF sandwich).

Hao et al. (2019) also reported declining trends of $\Sigma_{18}\text{PCBs}$ with a half-life of 2.0 years at 7 sites close to the Chinese Great Wall Station in West Antarctica measured using passive samplers from December 2011 to January 2018. A declining trend of PCBs in Antarctic air in the last 2 decades was also noted by Pozo et al. (2017) who compared their passive air sampling results along the Ross Sea (December 2010 – January 2011) with historical data from the same area. Several studies (Hao et al., 2019; Baek et al., 2011; Choi et al., 2008) reported high levels of PCB 11 (non-Aroclor congener) in Antarctic air which may have originated from local contamination from research stations.

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs)

Dioxin concentrations in air at two UK urban sites, namely London and Manchester, were found to be declining (1992-2016) at a $t_{1/2}$ of about 5 y and broadly correlated with annual atmospheric emission trends reported. Rural air concentrations, which are generally lower than urban concentrations, showed a slower decline with a half-life of about 5.8 years. Urban concentrations

are now similar to those in rural areas; thus, implying that most major, readily controllable sources were reduced and current levels in both rural and urban areas may remain similar unless there are major changes in energy requirements and generation options. Data collected during the winter periods show that concentrations are generally a factor of 2-3 higher than summer concentrations, implicating diffuse combustion sources (e.g., domestic space heating) (Graf et al., 2016).

Under SMP-POPs in Spain, OCDD was the congener found at the greatest concentrations followed by the OCDF and hepta- and hexa-substituted congeners. Similar to TOMPs, spatial variations indicate that urban areas are the main source of PCDD/Fs and higher concentrations were observed during the cold seasons, probably related to increased combustion sources in some locations during these times. PCDD/F concentrations showed significant decreases since 2008. This decline was sharper in urban places resulting in converging levels at urban and background sites for these pollutants (Muñoz-Arnanz et al., 2018).

Consistent with observations at urban sites in the U.K. and Spain, Canada's National Air Pollution Surveillance (NAPS) (1980-2009) also observed declining trends at 29 urban, 8 rural and 1 suburban sites after the early 1990s and during the first years in 2000s. After 2005, a clear trend was not evident. This decline was attributed to control measures taken in Canada for PCDD/F emission sources (CEC, 2014).

PCDD/Fs in air at 2 summit sites in the European Alps were also found to be significantly decreasing between 2005 and 2017. At the Swedish-operated monitoring stations of Råö and Aspöreten, PCDD/Fs were measured between 2009 and 2017. No trends were reported but the concentrations were generally lower in the summer (Fredricsson et al., 2018), similar to those observed in the UK and in Spain.

α -, β - and γ -hexachlorocyclohexane (HCH)

The HCHs were used globally as pesticides which were available in two formulae: (1) Technical HCH which is made up of a mixture of isomers, mainly γ -HCH (10–15%), α -HCH (60–70%) and β -HCH (5–12%); and (2) lindane which contains almost pure γ -HCH, the active ingredient. α - and γ -HCH were most widely measured in air, while the air concentrations of β -HCH were usually found to be mostly below the detection limits, with no discernible time trends or consistent seasonality.

In the WEOG region, both α - and γ -HCH concentrations were declining at most sites.

At the Spanish air monitoring stations under SMP-POPs, no significant temporal trends were detectable from 2009 to 2011, then followed by a constant decreasing trend from 2013 to 2017 in both urban and remote locations. These trends were consistent with those observed for the Great

Lakes and the Arctic for γ -HCH as noted in the last GMP report that the atmospheric decline rates have accelerated after its use has been restricted.

In the Great Lakes, α - and γ -HCH tend to maximize over the summer which is consistent with enhanced volatilization during warmer periods. The flows of the HCHs into and out of the lakes were about equal, indicating air-water equilibrium for these compounds. Both α - and γ -HCH showed half-lives of about 4 years in the Great Lakes region.

α - and γ -HCH declined in Arctic air at all stations, except γ -HCH at Station Nord. The half-lives of α -HCH ranged from 4.6 to 8.9 y, which were similar to the half-lives of γ -HCH at most Arctic stations (4.4 to 10 y). γ -HCH at Station Nord showed an exceptionally long doubling time of 28 y that implied its concentration was not changing over time. The reductions in atmospheric concentrations of α - and γ -HCH in air at all Arctic stations were likely due to the declining global emissions of technical HCHs and lindane.

Trend analysis of data from the Swedish-operated sites of Råö and Pallas between 1994 (1995) and 2011 show average decreases of α -HCH of 6 % and γ -HCH of 6-7 % per year for both stations (Anttila et al., 2016). Since 2005, the α -HCH air concentrations were similar at all three Swedish-operated stations, so there is no geographical variation between south and north. In contrast, the γ -HCH concentration was higher in the south (Råö and Aspvreten) compared with northern Finland (Pallas) (Fredricsson et al., 2018), probably because the southerly sites are closer to previous sources of lindane.

Globally, the GAPS network reported declining tendencies for α - and γ -HCH from 2005 to 2017 and no consistent trends for β -HCH.

Under the MONET programme, HCH isomers were reported between 2013 and 2016 at 13-16 sites. The highest mean concentration of α -HCH was measured at the Arctic site of Stórhöfði (Iceland) during the entire reporting period (median: 9.12 – 18.24 pg/m³). As part of the EMEP programme, α -HCH was measured at 12 stations and showed elevated concentrations in the Arctic compared to other southerly European sites as well (Colette et al. 2016). A likely contributing factor for the high concentrations observed in the Arctic is secondary emissions due to re-volatilization as a result of warming. Kalina et al. (2019) reported statistically significant declining trends of α - and γ -HCH at 6 MONET/EMEP stations.

At the Antarctic Station of Casey, the air concentrations of α - and γ -HCH ranged from 0.024 to 1.2 pg/m³ and 0.010 to 0.530 pg/m³, respectively. Levels are consistent with those detected in the Antarctic atmosphere over the past decade and have decreased dramatically over the past 30 years (Bengston Nash et al., 2017).

At the Norwegian Antarctic station of Troll, the air concentration of α -HCH showed a decreasing trend with a half-life of 12 y between 2007 and 2018 and 7.2 y between 2007 and 2015 before an implementation of biweekly-integrated sampling and inclusion of XAD in gas-phase sampling media which affected the trend. These findings are consistent with the decreasing trend of Σ HCHs (half-life of 2 years) observed at 7 sites close to Chinese Great Wall Station (Dec 2010- Jan 2018) in West Antarctica (Hao et al., 2019).

Perfluorooctane sulfonic acid (PFOS), its salts and PFOS-F and perfluorooctane sulfonyl and Perfluorooctanoic acid (PFOA), its salts and PFOA-related compounds

Per- and polyfluoroalkyl substances (PFASs) include a large number of substances, in which perfluoroalkyl acids (PFAAs), such as the perfluoroalkyl carboxylic acids (PFCAs) and perfluoroalkyl sulfonic acids (PFSAs), were found to have widespread presence in the environment, humans and wildlife, and tend to bioaccumulate through the food webs. PFAAs can be emitted directly into the environment but can also result from biotic and abiotic degradation of neutral precursors (Schenker et al., 2008). The focus of this review is on PFOS and PFOA, as well as their precursors which can transform or degrade to PFOS and PFOA in the environment.

In the Arctic, PFASs were analysed in air samples collected at Alert (Canada) (gas and particle phases) and at the Norwegian-operated sites of Zeppelin (Svalbard) and Andøya (Norway) (particle phase only). Telomer alcohols (PFOA precursors) were analysed at Alert (Canada) and Station Nord (Greenland). At Alert, air concentrations of PFOA increased from 2006 to 2013. After 2013, its concentration has steadily declined (Figure 5.2.5). At its peak, i.e. 2013 to 2017, the half-life of PFOA at Alert was 4.8 y. At Zeppelin and Andøya, PFOA showed gradually decreasing trends over the entire monitoring period. The half-lives of PFOA at Zeppelin and Andøya were 3.5 and 2.1 y, respectively.

At Alert, PFOS showed a similar trend as PFOA with a peak at 2013 (Figure 5.2.5). Its half-life from 2013 to 2017 was 2.8 y. Air concentrations of PFOS were slowly declining at Zeppelin and Andøya with half-lives of 10 and 13 y, respectively. At Zeppelin, both PFOA and PFOS showed an accelerated rate of decline up to 2017 as compared to those reported previously from 2006 to 2014 by Wong et al. (2018), probably as a result of the continuing global effort in the phase-out of PFOS and PFOA and related substances since the 2000s.

Atmospheric levels of the PFAS precursor compounds declined steadily in general. At Alert, air concentrations of 8:2 FTOH and 10:2 FTOH were increasing from 2006 until 2011, followed by a decreasing trend. Half-lives for the declining period (2011- 2017) were 4.0 y for 8:2-FTOH and 3.0 y for 10:2 FTOH. At Station Nord, 8:2 FTOH and 10:2 FTOH showed decreasing trends for the entire monitoring period (2008-2017). The half-life for 8:2 FTOH was 10 y and 10:2 FTOH

was 5.3 y. However, 6:2 FTOH remained stable at both Alert and Station Nord. These results were in line with findings of Rauert et al. (2018a) that stable concentrations of FTOHs were observed in sorbent impregnated passive air samplers (SIP-PAS) from GAPS sites deployed in the WEOG region from 2009-2015 (Figure 5.2.6). In global air, Gawor et al. (2014) reported that 8:2 and 10:2 FTOH showed decreasing trends in XAD-based passive air sampler (XAD-PAS) samples collected from GAPS from 2006 to 2011. Under GAPS, elevated concentrations of all the neutral PFAS were detected at the urban sites as compared to the polar/background sites. PFASs saw significant increases ($p < 0.001$) in concentrations from 2009 to 2015. PFCAs had elevated concentrations in 2015, however, the difference was not statistically significant ($p > 0.05$). Concentrations of the PFASs and the PFCAs were similar at all location types, showing the global reach of these persistent compounds.

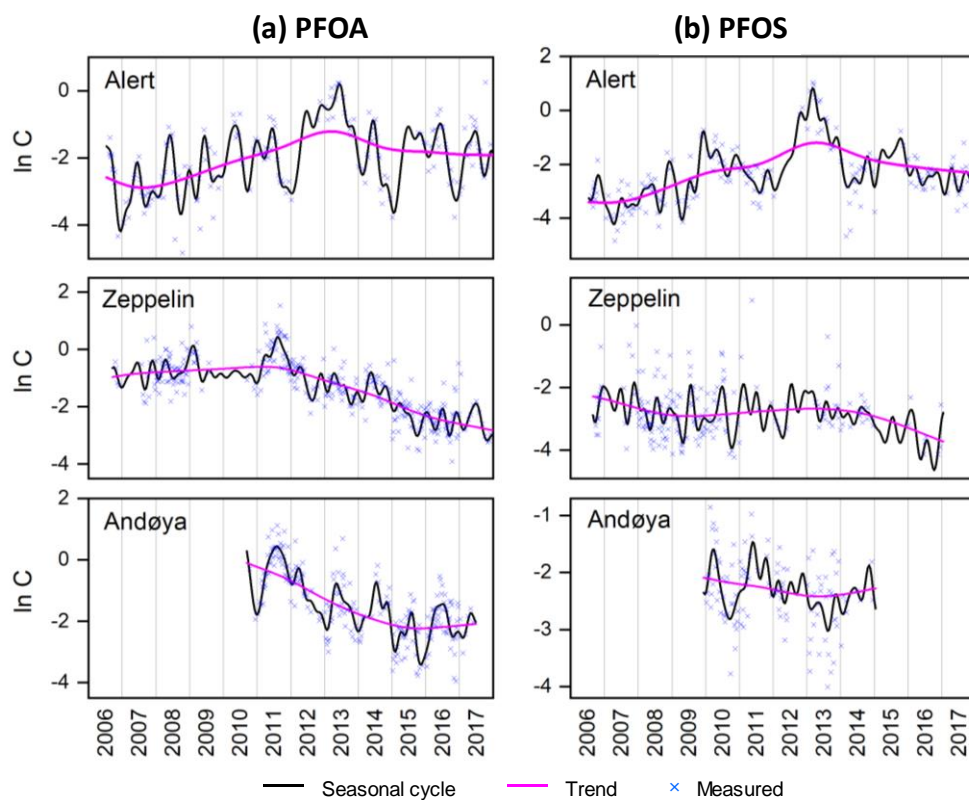


Figure 5.2.5 Time trends of PFOA and PFOS at the Arctic Stations of Alert, Zeppelin and Andøya from 2006-2017.

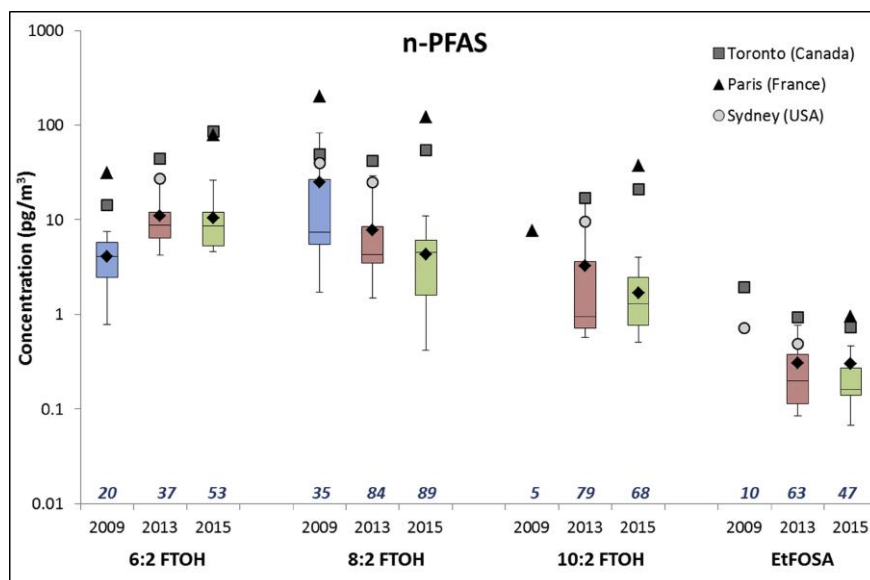


Figure 5.2.6 Box and whisker plots of concentrations (pg/m^3) of 6:2 FTOH, 8:2 FTOH, 10:2 FTOH and EtFOSA detected at background and polar sites in 2009, 2013 and 2015 under GAPS. The boxes show median concentrations and the 25th and 75th percentiles, minimum and maximum concentrations are indicated by the whiskers. Mean concentrations are indicated by the diamond. Where an analyte was below detection limits, the sample was not included, and detection frequencies are listed below the plots. Concentrations (pg/m^3) determined at the three urban sites are indicated separately by the triangle, square and circle icons. Note only Paris (France) saw detection of 10:2 FTOH above detection limits in 2009 and only Toronto (Canada) and Sydney (USA) saw detection of EtFOSA above detection limits in 2009.

The measurements of PFAS in air started in 2009 at the Swedish station of Råö (Fredricsson et al., 2018), and in 2006 at the Norwegian station of Birkenes (NILU, 2019). No trend was reported for PFAS in air at both sites. Similar to the observations at the Canadian Arctic site of Alert, the highest concentrations of PFOS were measured at Råö in 2011 and 2013, while PFOA peaked in 2015 and 2016.

Technical Endosulfan and its Related Isomers

Endosulfans and endosulfan sulphate concentrations in air measured in the WEOG region were generally declining during this reporting period. α -endosulfan air concentrations were usually found at higher concentrations than β -endosulfan at all monitoring sites.

Air concentrations of α -endosulfan at the Arctic stations of Alert, Pallas and Station Nord (1992-2017) were declining with half-lives of 10, 1.6 and 2.4 y respectively. At Alert, α -endosulfan was decreasing at a faster rate than previously reported for 1992-2012, which had a half-life of 19 y (Hung et al., 2016). This accelerated decline is probably related to the regulation of

endosulfan in the U.S. and Canada in 2010. The declining rates at Pallas and Station Nord are fast relative to other chemicals. β -endosulfan at all Arctic stations were mostly below detection limit and time trends were not derived.

An accelerated rate of decline of α -endosulfan was also apparent at the Lake Ontario station of Point Petre (Figure 5.2.7) after Canada and the US started the phase out of endosulfan. Salamova et al. (2015) also estimated the atmospheric degradation half-life for endosulfan to be approximately 4 y which is significantly less than other legacy OCPs, e.g. DDTs.

Significant reductions of the sum of α - and β -endosulfan and endosulfan sulphate were reported for the alpine stations of Zugspitze and Sonnblick (Kirchner et al., 2016).

At the Swedish-operated stations of Råö and Pallas, α - and β -endosulfan and endosulfan sulphate were measured in air for 2009-2017. α -endosulfan was measured at the highest levels compared to the others and ranged between <0.012 - 6.4 pg/m^3 at Råö and 0.096 - 23 pg/m^3 at Pallas, respectively. Decreasing trends of α -endosulfan were observed at both stations.

At the Australian Antarctic Station of Casey (2010-2015), endosulfan was found to be the second-most dominant POP (after HCB) with air concentrations ranging from 0.018 - 3.8 pg/m^3 and an average of 0.55 pg/m^3 . Endosulfan had only been reported in Antarctic air on King George Island (2005-2009) (Baek et al., 2011) with concentrations up to one order of magnitude higher than that found at Casey. One possible reason is that King George Island lies much closer to the South American continent where usages occurred. No interannual trend was discernable for endosulfan measured at Casey. On the other hand, Hao et al. (2019) observed decreasing concentrations of endosulfans at 7 sites close to Chinese Great Wall Station (Dec 2010- Jan 2018) in West Antarctica with a rapid half-life of 1.2 years.

The Spanish programme of SMP-POPs did not find any spatial differences in the total endosulfan concentrations measured in air.

The GAPS network reported declining tendencies for endosulfans and their metabolites globally.

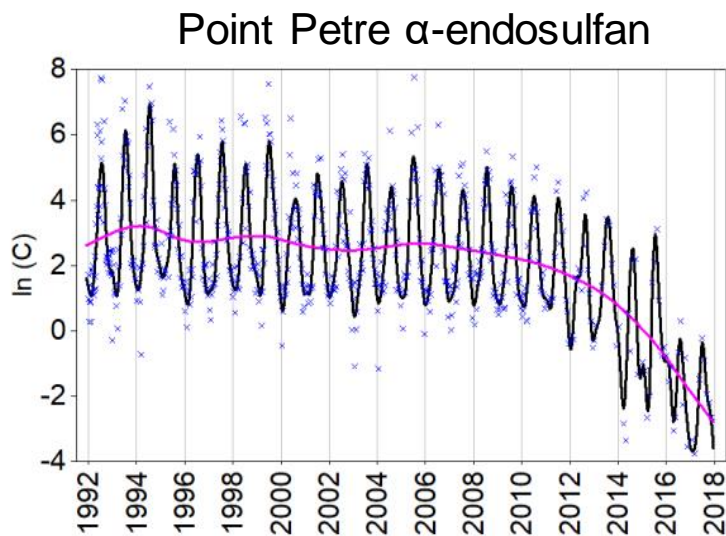


Figure 5.2.7 Temporal trend of α -endosulfan measured in air at Point Petre, Lake Ontario (Canada)

Polybrominated Diphenyl Ethers (PBDEs)

Air concentrations of individual PBDE congeners (BDE 47, 99, 100, 138) were declining in the Arctic stations. Exceptions were BDE 99 and BDE 100 at Stórhöfði that were slowly rising but PBDEs were mostly non-detectable at this site. In the previous report, PBDEs at Alert from 2002 to 2012 were stable (Hung et al., 2016). With 5 more years of data, decreasing trends of PBDEs were observable at Alert from 2002 to 2017, reflecting the effectiveness of the regulation of the penta- and octa-BDE mixtures in North America which started in 2005.

BDE 209 was added to the Stockholm Convention in 2017. At Alert, BDE 209 was slowly decreasing with a half-life of 4.8 y. This could be due to voluntary measures to reduce emissions initiated in mid-2000 in Europe in combination with the phase-out effort of BDE209 in Canada and U.S. in 2013 (de Wit, 2010; Commission Regulation (EU) 2017; USEPA, 2009 and 2013; EC, 2013).

At Zeppelin and Andøya, BDE 209 showed increasing trends with doubling times of 14 and 25 y, respectively (Figure 5.2.8). It is worth mentioning that at Zeppelin, the fraction of BDE 209 ranged from 19 to 88 %, from 2006 to 2016, and increased to 97 % in 2017. BDE 209 at Zeppelin became the dominant congener instead of the commonly detected BDE 47 or BDE 99. An increasing proportion of BDE 209 to Σ PBDEs was also observed at Andøya (Norway) in 2017. The elevated concentrations of BDE209 could be due to the phase-out of the commercial penta-PBDE.

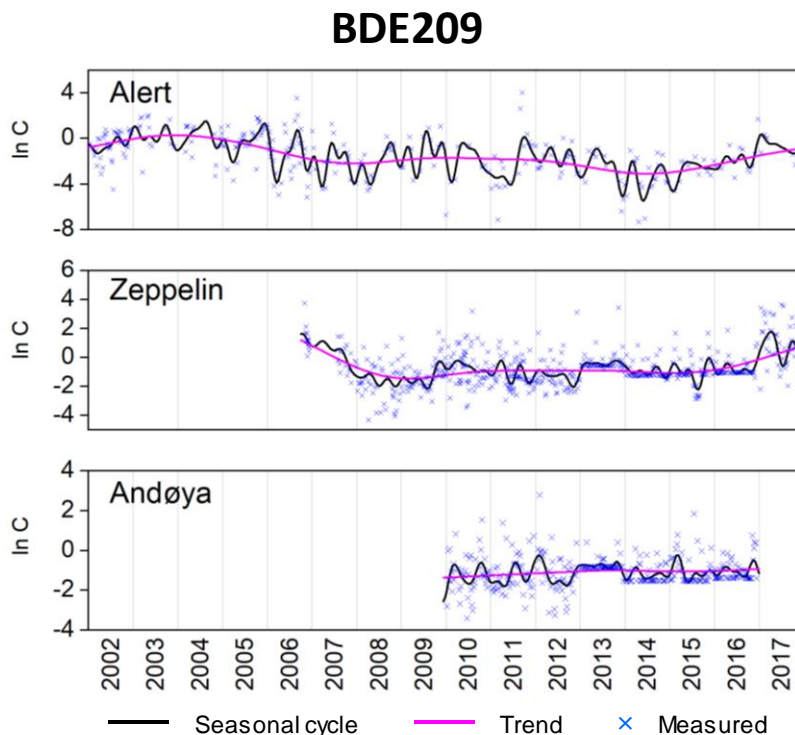


Figure 5.2.8 Time trends of BDE209 at Alert, Zeppelin and Andøya

PBDE levels have clearly decreased along the UK/Norway passive sampling transect from the first two-year integrated samples (2000-2002 and 2002-2004). Concentrations were elevated at sites where there was construction work or waste burning at the time.

PBDEs were measured in air in the UK from 2010 to 2018. Trend data for Σ_9 PBDE (sum of congeners BDE47, BDE49, BDE99, BDE100, BDE119, BDE154, BDE153, BDE138, BDE183) for the urban sites at London and Manchester showed evidence of a decline with an approximate half-life of 8.3 years. In contrast, the decline in Σ_9 PBDE atmospheric concentrations at the four rural/semi-rural sites was considerably slower with an estimated half-life of 22 years. At all UK sites, air concentrations of individual congener showed BDE-47 > BDE-99, and both dominated all calculated profiles. Given that these two congeners are the main components of the penta-BDE (PeBDE) technical mixture, with BDE-47 accounting for 38-42 % and BDE-99 accounting for 45-49 % of the Σ PBDEs, these results likely reflect the extensive use of that specific technical mixture.

Similar to the UK sites, the dominant congeners in Great Lakes air were also BDE 47 and 99, reflecting influence from the penta-technical mixture. At the US Great Lakes sites, the

concentrations of PBDEs were decreasing at the urban sites, Chicago and Cleveland, but were generally unchanging at the remote sites, Sleeping Bear Dunes and Eagle Harbor (2005-2013) (Liu et al. 2016). On the other hand, declining trends for BDE 47 and 99 were observed at the 2 Canadian Great Lakes sites, which are considered remote sites, with faster decline at Point Petre ($t_{1/2}$ of 8.5 y and 6.2 y, respectively) which is closer to urban development, probably reflecting the replacement of these substances in cities. However, the sum of 13 BDEs (excluding BDE209) showed no increasing or declining trend with t_2 of -44 y.

In Spain (2009-2017), BDE 209 was the prevalent congener found in air followed by BDE 47 and 99 in most cases regardless of the sampling year or location type (Roscales et al., 2018). Highly populated areas were the main sources of PBDEs in Spain with higher concentrations in the summer than the winter. No significant temporal trends were detected for total PBDE concentrations, neither at urban nor at remote sites. This observation may have reflected the difference in trends for BDE 209 (stable or increasing concentrations) and penta- or octa-technical mixtures (stable or significant decreasing concentrations) in 2009 to 2017 due to the timing differences in regulation introduction regarding the different technical mixtures.

Under PureAlps, BDE 28 and 154 showed declining trends at the alpine site of Zugspitze (Bavaria/Germany). No trends were found for the other congeners at this site or any PBDE congeners measured in air at the Sonnblick station (Austria). Under MONET EU, extremely high levels of PBDE100, PBDE153, PBDE154, PBDE47 and PBDE99 were found in Sonnblick compared to the other sampling sites in 2013 and 2014 in particular (see MONET Executive Summary in Annex 1). Unfortunately, the concentration of PBDEs was not measured in Sonnblick in the following years.

At the Swedish-operated air monitoring stations of Råö (Sweden), Aspöreten (Sweden) and Pallas (Finland), with the exception of Pallas in 2014, the levels of BDE 47, 99 and 100 in air have decreased since the start of measurements at all three stations. No trends could be discerned for BDE-209.

PBDEs were measured globally under GAPS; however, so far temporal trends cannot be elucidated due to high blank issues. For PBDEs detected in the 2014 GAPS samples, concentrations were similar to those previously reported from samples collected in 2005 at GAPS sites, suggesting global background atmospheric concentrations of PBDEs have not declined since regulatory measures were implemented (Rauert et al., 2018a).

Under MONET, passive air sampling at 6 EMEP stations showed generally insignificant and decreasing trends for the PBDE congeners, except for BDE 209 which is consistent with its known differences in use and emissions compared to the other PBDEs (Kalina et al., 2019).

Hexabromocyclododecane (HBCDD)

Zeppelin and Alert were the only Arctic stations that had HBCDD measurements. At Zeppelin, the three isomers of HBCDD (α -, β - and γ -) were only detected in 3.0 to 14 % in samples taken from 2006 to 2017. At Alert, HBCDD was reported as the sum of the three isomers, and the detection frequency is 29 % from 2002 to 2017. The concentrations of HBCDD at both sites were low, in which the median of α -HBCDD at Zeppelin is 0.11 pg/m³ and at Alert was 0.22 pg/m³. Given the low detectability of HBCDD in Arctic air, temporal trends cannot be derived.

In the Great Lakes region, HBCDD was found to be mostly non-detectable in air samples from 2009 and 2017 at the two Canadian master stations on Lakes Ontario and Huron. On the other hand, Olukunle et al. (2018) reported the concentrations of α -, β -, and γ - HBCDD in air-borne particles for 4 US IADN sites from January to December 2014. The median Σ HBCDD concentrations for the four sites were 2.0 pg/m³, 2.1 pg/m³, 1.7 pg/m³ and 5.2 pg/m³ for Chicago, Cleveland, Sturgeon Point and Sleeping Bear Dunes, respectively. Higher levels of Σ HBCDD were observed at the remote site of Sleeping Bear Dunes with comparable levels at the remaining three sites. Σ HBCDD were significantly lower than or similar in concentrations to those of PBDEs and other halogenated flame retardants at all sites, except for Sleeping Bear Dunes, and there were no significant correlations between Σ HBCDD and the other flame retardants, indicating different sources.

HBCDD in air at the Swedish-operated station of Råö (Sweden) (0.02-4.4 pg/m³) and Pallas (Finland) (0.05-0.092 pg/m³) (2009-2017) occurred only sporadically in detectable concentrations. No trends can be discerned for HBCDD at these sites.

HBCDD in air is usually found to be associated with particles. The first global scale distribution for HBCDD has been determined based on re-analysis of GAPS Network samples from 2005-2006 (Lee et al., 2010) which varied widely across different sites and different sampling periods. HBCDD diastereomers were only detected at 20 % of the GAPS sites in 2014. In the WEOG region, the highest concentrations of the sum of the three diastereomers were seen at Paris (France) with concentrations of 16– 58 pg/m³ (Rauert et al., 2018a). Future analysis of archived GAPS samples will help to resolve temporal tendencies for HBCDD in the global atmosphere.

Pentachlorobenzene (PeCB)

No time trend data are available for pentachlorobenzene because it has a tendency to breakthrough PUFs used in active air sampling programmes.

MONET reported air concentrations of PeCB measured with passive air samplers at 13-16 sampling sites measured from 2013 to 2016, with highest concentrations detected in Sonnblick

(Austria) (median: 14.04 – 48.83 pg/m³) and lowest concentrations found in Malta (3.55-8.50 pg/m³).

In the European Alps, PeCB showed a significant decrease but nonetheless high concentrations in ambient air (medians: 36-37 pg/Nm³, normalised air volume) (Kirchner et al., 2016).

In Spain, no significant spatial differences were detected for PeCB between urban and remote stations and concentrations showed lower variabilities among sites than those for other POPs. During 2015-2017, median concentrations remained highly stable in both remote and urban locations.

The GAPS network reported seasonal results for PeCB in a pilot study using SIP as sampling medium showing that PeCB has fairly uniform global distribution (Koblizkova et al., 2012).

Short-Chain Chlorinated Paraffins (SCCPs)

At the Canadian Arctic air monitoring station of Alert, some air samples collected from January to August 2011 were screened for SCCPs and the mean and median concentrations were 913 and 685 pg/m³, respectively, with a range of 206-2876 pg/m³. The same analytical method was used to measure SCCPs in September and December 1992 showing a mean SCCP concentration of 20 pg/m³ (range <1.7-67 pg/m³); and 4 air samples from January 1994 to January 1995 showing concentrations of 1.07-7.25 pg/m³. These data suggest that there was a significant increase in air concentrations of SCCPs from the 1990s to 2011 (Vorkamp et al., 2019).

SCCPs (C₁₀₋₁₃) have been monitored at the Norwegian-operated stations of Zeppelin (Svalbard, Arctic) since 2013, and at Birkenes (Norway) since 2017. The annual mean concentrations of SCCPs measured at Zeppelin in 2018 was 290 pg/m³ which is close to the lower end of the measured concentrations seen at Alert in 2011. The concentrations of SCCPs measured in air at Zeppelin showed no significant difference between the years (2013-2018) (NILU, 2019). SCCPs in Arctic air generally showed the predominance of the most volatile congener of C₁₀. Archived extracts from 2009 collected under the GAPS programme were analysed for SCCPs and those reported at 4 Arctic stations showed a concentration range of 81-1000 pg/m³ for ΣSCCPs. These results confirmed that SCCP concentrations exceeded those of PCBs in Arctic air (Vorkamp et al., 2019). It was noted that SCCP measurements at these sites are semi-quantitative as the contributions of possible contamination during sampling and analysis have not been validated.

The SCCP concentrations (2009-2017) varied between 7-220 pg/m³ at the Swedish stations of Råö and 6 - 2100 pg/m³ at Aspöreten (Fredricsson et al., 2018).

In Australia, XAD-passive air samples collected in 2016 at five remote, six rural and four urban sites were analyzed for SCCPs. SCCP concentrations were observed to be decreasing spatially from more densely to less densely populated areas ($p < 0.01$).

PCP/PCA

PCA was measured in air at 2 sites on the European Alps from 2013 to 2018, with median concentrations of 6.35 pg/m^3 for Zugspitze (Bavaria/Germany) and 6.79 pg/m^3 for Sonnblick (Austria). While no trend was deduced from Zugspitze, the concentrations were declining in Sonnblick.

PCA is also reported in air at the Arctic station of Alert, Canada, but no trend can be derived.

Hexachlorobutadiene (HCBd)

HCBd was monitored at the Canadian Arctic site of Alert since 2002 but high breakthrough was observed. Reported data were considered semi-quantitative and the concentrations were probably underestimated. Overall, the median concentration of HCBd at Alert from 2002-2017 was 1.8 pg/m^3 . The yearly average of HCBd from (2002-2017) ranged from 1.2 to 13 pg/m^3 . HCBd at Alert showed non-changing trend with a long half-life of 20 y.

Despite a decreasing trend in the European Alps, hexachlorobutadiene exhibits very high concentrations in air, which are a factor of 100 higher than for comparable compounds ($1,178 \text{ pg/Nm}^3$ (normalised air volume) at Zugspitze (Bavaria/Germany) and 804 pg/Nm^3 (normalised air volume) Sonnblick (Austria) (2014-2017).

Hexabromobiphenyl (HBB or BB-153)

No air measurement data were reported for HBB.

Toxaphene

No trend data were available for toxaphene.

Toxaphene was detectable in air samples collected at the Australian Antarctic station of Casey between 2010 to 2015 but temporal trends were not reported (Bengtson Nash et al., 2017).

Chlordecone

No air measurement data were reported for chlordecone.

Dicofol

No air measurement data were reported for dicofol. An analytical method for assessing dicofol in air samples exists under the GAPS Network (Eng et al., 2016) and has been applied to air samples from India and the GRULAC region (Rauert et al., 2018c). This method measures o,p'-dichlorobenzophenone (DCBP) which is a degradation product of dicofol that forms during instrumental analysis. In the future, this method will be applied to WEOG region samples from GAPS for future GMP reporting.

PCNs

No air measurement data were reported for PCNs. There are plans under GAPS to include results for PCNs in the future.

5.2.1.4 Summarised Compound Specific Trends

Tables 5.2.1b to d below summarize the general trends and half-lives/ doubling times reported by the different air monitoring programmes for all individual compounds or compound groups in air in the WEOG region. The color of each cell reflects the reported trend: **green** indicates decreasing trend, **red** indicates increasing trend and **blue** indicates a trend that is not increasing or decreasing (arbitrarily assigned when a reported half-life or doubling time is greater than ± 50 years). When a chemical is being measured in air, but no trend was reported or can be developed, the trend is labelled as “NA”. Please note that not all programmes reported half-lives/doubling times for all chemicals. The reported half-lives are given as positive values and doubling times are shown as negative values in years. Generally, the half-lives/doubling times were calculated over the entire period of monitoring for each compound/compound group and for different programs, the data time series span over different time frames, with some of them starting as early as 1990. Some programs have reported half-lives/doubling times over selected periods of time, e.g. EMEP reported time trends over the entire sampling period, before 2004 (when the Stockholm Convention came into force) and after 2004; and AMAP reported PFAS time trends for Alert (Canada) before and after a peak occurred.

Table 5.2.1b Apparent first order half-lives ($t_{1/2}$, year) of organochlorine pesticides and Σ PCB

Station	Period	Σ PCB	Aldrin	endrin	dieldrin	chlordanes	t-nonachlor	c-nonachlor	oxychlorodane	Σ DDTs
Antarctica										
Troll	2007-2015	7.2*	-	-	-	-	-	-	-	5**
Troll	2007-2018	10*	-	-	-	-	-	-	-	8.4**
Casey ^a	2010-2015	NA	NA	NA	NA	NA	NA	NA	NA	NA
Chinese Great Wall ^b	Dec 2010-Jan 2018	2	NA	NA	NA	NA	NA	NA	NA	2.4
King Sejong ^c	2005-2007, 2008-2009	NA	NA	-	NA	NA	NA	NA	-	NA
Station	Period	heptachlor	heptachlor epoxide	HCB	α -HCH	β -HCH	γ -HCH	Σ endosulfans	endosulfan sulfate	mirex
Troll	2007-2015	-	-	-22	7.2	-	-	-	-	-
Troll	2007-2018	-	-	-19	12	-	-	-	-	-
Casey ^a	2010-2015	NA	NA	NA	NA	NA	NA	NA	NA	NA
Chinese Great Wall ^b	Dec 2010-Jan 2018	NA	NA		2	-	-	1.2	-	NA
King Sejong ^c	2005-2007, 2008-2009	NA	-	-	NA	NA	NA	***	NA	-

^a Bengtson Nash et al. (2017); ^b Hao et al. (2019); ^c Baek et al. (2011); *PCB-52 only; ** p, p'-DDE only; *** endosulfan I only; NA = Compound analyzed, but half-lives not available.

Station	Period	PCB-153			chlordanes	<i>p,p'</i> -DDT			HCB			α -HCH	γ -HCH
		Entire Period*	Before 2004*	After 2004*		Entire Period*	Before 2004*	After 2004*	Entire Period*	Before 2004*	After 2004*		
Europe (EMEP)													
Birkenes	1998-2017	7.6 (2004-2017)	-	-	NA	(2010-2017)	-	(2010-2017)	36	6.8	28	NA	NA
Zeppelin	1993-2017	8.5	4.1	9.5 (2004-2017)	NA	5.6 (1994-2017)	3.9 (1994-2004)	7.5	-440	10	-30	NA	NA
Station Nord	2009-2017	-	-	-	NA	18	-	-	1100	-	-	NA	NA
Stórhöfði	1995-2017	30	57	-32	NA	11 (1996-2017)	2.9 (1996-2004)	-21 (2004-2017)	43	9.3	-20	NA	NA
Pallas	1996-2017	13	6.0	13	NA	11	13	7.7	-11 (2009-2017)	-	-	NA	NA
Aspvreten	1996-2017	13	12	17	NA	5.5 (2008-2017)	-	-	-14 (2009-2017)	-	-	NA	NA
Råö/Rörvik	1994-2017	20	9.1	20	NA	18 (1995-2017)	-4.4 (1995-2004)	11 (2004-2017)	-26 (2009-2017, Råö only)	-	-	NA	NA
Westerland	2007-2017	7.9	-	-	NA	131 (2009-2017)	-	-	8.0 (2007-2016)	-	-	NA	NA
Waldhof	2012-2017	-	-	-	NA	11	-	-	-	-	-	NA	NA
Zingst	2007-2017	4.3	-	-	NA	5.6 (2009-2017)	-	-	5.8 (2007-2016)	-	-	NA	NA
Kosetice	1999-2017	3.5	8.2	4.2	NA	7.2	-6.4	6.1	-22 (2006-2017)	-	-	NA	NA
Andøya	2009-2017	140 (2010-2017)	-	-	NA	-	-	-	-30	-	-	NA	NA
Schauinsland	2007-2015	9.5	-	-	NA	-	-	-	-	-	-	NA	NA

*For PCBs, DDTs, and HCB, half-lives are given based on the following timescales: • Entire Period: The whole monitoring period (either starting earlier than 2004 or later than 2004),

• Before 2004: From the starting year of the monitoring period until the Stockholm Convention entered into force for the selected compounds (2004)

• After 2004: From the Stockholm Convention entered into force for the selected POPs (2004) until the end of 2017.

The empty white cells mean that the available time-series are incomplete for this time period (i.e. did not start before 2004 or started later than 2004).

Years in () denote the period if it is different from the period in column 2 and other notes. NA = Compound analyzed, but half-lives not available.

Table 5.2.1b Continued

Station	Period	ΣPCB	dieldrin	t-chlordane	c-chlordane	t-nonachlor	heptachlor	heptachlor epoxide	α-HCH	γ-HCH	endosulfan I	endosulfan II
GAPS												
Alert, Nunavut	2004-2017											
Barrow, Alaska	2004-2017										NS	
St. Lawrence Island, Alaska	2005-2017											
Snare Rapids, Northwest Territories	2005	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Bratt's Lake, Saskatchewan	2004-2017											
Whistler, British Columbia	2004-2017								NS	NS		
Vancouver, British Columbia	2005	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Dorset, Ontario	2004-2005	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Downsview, Ontario	2004-2017		NS	NS	NS				NS	NS		NS
Simi Valley, California	2004-2005	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Athens, Georgia	2004-2006	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Tudor Hill	2004-2017			NS								
Ny-Ålesund	2004-2017		NS		NS			NS	NS	NS		
Stórhöfði	2004-2017				NS			NS		NS		
Hollola	2004-2006	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Malin Head	2004-2017		NS						NS	NS		
Paris	2005-2017		NS	NS				NS				
Barcelona	2004-2006	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Izmir	2004-2007	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Isola Marettimo	2004-2006	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Telde, Las Palmas	2005-2010		NS					NS	NS			NS
Darwin	2004-2017		NS	NS				NS				
Cape Grim	2004-2017											
Mario Zucchelli Station	2004-2005	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Little Fox Lake, Yukon	2006-2017	NS							NS		NS	NS
Dyea, Alaska	2006-2017	NS							NS			
Cortes Island, British Columbia	2006-2007	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Lasqueti Island, BC	2006-2012	NS			NS							
Saturna Island, British Columbia	2006-2007	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Pallas (Matorova station)	2005-2017		NS							NS		
North Island	2006	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Fraserdale, Ontario	2007-2017	NS	NS					NS	NS	NS		
Ucluelet, British Columbia	2007-2017											
Sable Island, Nova Scotia	2007-2017								NS	NS		
Point Reyes, California	2007-2017											
Sydney, Florida	2007-2017			NS						NS		
Mauna Loa Obs, Hilo, Hawaii	2007-2017	NS			NS				NS	NS		
Tula, American Samoa (Cape Matatula)	2007-2017											
Temple Basin, Arthur's Pass	2009-2017											
Groton, Connecticut	2009-2017	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Doñana National Park	2009-2017									NS	NS	NS
Coral Harbour, Nunavut	2009-2017		NS					NS	NS	NS		
Mount Revelstoke, British Columbia	2009-2017	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Izana	2013-2017	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Longwoods	2013-2017	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Warsaw Caves	2013-2017	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
Egbert / CARE Station	2013-2017	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA

NS = Not statistically significant following first-order kinetics

NA = Compound analyzed, but trend not available.

Table 5.2.1b Continued

Station	Period	ΣPCB	aldrin	endrin	dieldrin	chlordane	ΣDDTs	heptachlor	HCB	α-HCH	β-HCH	γ-HCH	endosulfans	mirex
GAPS	2005-2017		NA	NA										NA

NA = Compound analyzed, but trend not available.

Station	Period	ΣPCB	aldrin	dieldrin	t-chlordane	c-chlordane	t-nonachlor	methoxychlor	oxychlordane
GLB									
Point Petre (Lake Ontario)	1992-2017	17	6.2	10	9.4	9.8	13	NA	8.5
Burnt Island (Lake Huron)	1992-2012	65	7.6	10	12	11	13	NA	12
Egbert (Lake Huron)	1995-2006	74	7.1	18	31	34	52	NA	71
Station	Period	p, p'-DDT	p, p'-DDE	p, p'-DDD	o, p'-DDT	o, p'-DDE	heptachlor	heptachlor epoxide	HCB
Point Petre (Lake Ontario)	1992-2017	9.1	12	7.5	9	11	6.5	4.7	NA
Burnt Island (Lake Huron)	1992-2012	7.4	9.6	9.5	9.3	7.2	8.6	5.8	NA
Egbert (Lake Huron)	1995-2006	12	38	12	28	14	8.3	12	NA
Station	Period	α-HCH	γ-HCH	endosulfan I	endosulfan II	endosulfan sulfate	mirex	photomirex	
Point Petre (Lake Ontario)	1992-2017	4.8	4.3	4.8	5.2	8.1	13	9.4	
Burnt Island (Lake Huron)	1992-2012	4.9	4.7	13	11	10	28	6.1	
Egbert (Lake Huron)	1995-2006	4.2	4.7	38	28	4.5	8	4.6	

NA = Compound analyzed, but half-lives not available.

Station	Period	ΣPCB	chlordane	ΣDDTs	α-HCH	γ-HCH	endosulfan
IADN^d							
Chicago	1991-2017	11.9	11.6	11.9	3.9	4.3	8
Cleveland	1991-2017	18.6	9.0	9.1	4.1	4.2	7.6
Sturgeon Pt	1991-2017	15.3	8.7	10.4	3.5	3.6	8
Sleeping Bear Dunes	1991-2017	12.9	9.5	9.3	3.9	3.7	8.1
Eagle Harbor	1991-2017	13.2	10.2	10.5	4.3	4.5	8.9
Point Petre	2013-2017	16.3	11.7	12.6	4.4	3.9	7.1

^d Salamova et al. (2015)

Station	Period	ΣPCB	aldrin	endrin	dieldrin	chlordane	methoxychlor	oxychlordane	ΣDDTs	heptachlor
PureAlps										
Zugspitze	2005-2017									
Sonnblick	2005-2017									
Station	Period	heptachlor epoxide	HCB	α-HCH	β-HCH	γ-HCH	endosulfan	endosulfan sulfate	mirex	
Zugspitze	2005-2017				NA					
Sonnblick	2005-2017				NA	NA				

NA = Compound analyzed, but half-lives not available.

Table 5.2.1b Continued

Station	Period	HCB	α -HCH	γ -HCH	p, p' -DDE
MONET^e					
Košetice (active)	1996-2016	21	6	6	16
Košetice (passive)	1996-2016	5	3	3	7
Pallas (active)	1996-2016	-97	7	5	41
Pallas (passive)	1996-2016	7	3	2	5
Råö (active)	2002-2016	-55	7	6	36
Råö (passive)	2002-2016	9	3	3	8
Zeppelin (active)	1993-2016	-108	6	5	8
Zeppelin (passive)	1993-2016	8	20	4	-15
Birkenes (active)	2003-2016	83	8	7	-64
Birkenes (passive)	2003-2016	5	5	5	-27
Stórhöfði (active)	1995-2016	118	6	8	Infinity
Stórhöfði (passive)	1995-2016	6	3	2	4

^e Kalina et al. (2019)

Station	Period	Σ PCB	DDTs	HCB	α -HCH	β -HCH	γ -HCH	endosulfan
SMP-POPs	2008-2017							NA

NA = Compound analyzed, but half-lives not available.

Station	Period	Σ PCB
TOMPs		
London	Jan 1991-2017	~7.9
Manchester	Jan 1991-2017	~7.9
Auchencorth	Dec 2008-2017	~8.3
Middlesbrough	Apr 1993- July 2008	~8.3
Hazelrigg	Sept 1992-2017	~8.3
High Muffles	Jan 1997-2017	~8.3
Weybourne	Dec 2008-2017	~8.3
Stoke Ferry	Feb 1997-Dec 2007	~8.3

~ = values are approximate

Table 5.2.1b Continued

Station	Period	ΣPCB	endrin	dieldrin	Chlordane	t-chlordane	c-chlordane	t-nonachlor	c-nonachlor
AMAP									
Alert	1992-2001		-	-	-	-	-	-	-
Alert	2003-2017		-	-	-	-	-	-	-
Alert	1992-2017		11	17	-	10	13	12	12
Zeppelin	1993-2017	(1998-2017)	-	-	-	10	13	17	10
Stórhöfði	1995-2017		-	12	-	12	14	22	-
Pallas	1996-2017	(1997-2017)	-	-	-	13	12	18	-
Andøya	2010-2012	(2010-2017)	-	-	-	3.2	7.9	10	8.5
Station Nord	2008-2017	-	NA	420	-	-410	25	36	-6.7
Little Fox Lake	2003, 2008, 2013	-	-	-	-	-	-	-	-
		oxychlordane	p, p'-DDT	p, p'-DDE	p, p'-DDD	o, p'-DDT	o, p'-DDE	o, p'-DDD	heptachlor
Alert	1992-2017	19 (1992-2012)	20	29	-13	13	16	-57	28(1992-2012)
Zeppelin	1994-2017	-	5.4	8.4	9.3	7	7.6	9.5	-
Stórhöfði	1995-2017	-	10	21	11	4200 (1996-2017)	-	-	-
Pallas	1997-2017	-	11	42	-35	-	-	-	-
Andøya	2010-2016	-	3.4	28	-133	9.7	17	9.3	-
Station Nord	2008-2017	-	-20	-29	-5.6	-13	-1.9	-	-23
		heptachlor epoxide	HCB	α-HCH	γ-HCH	endosulfan I			
Alert	1992-2017	23 (1992-2012)	35	4.6	4.4	10			
Zeppelin	1993-2017	-	-2740	5.3	4.4	-			
Stórhöfði	1995-2017	-	54	6	7.3	-			
Pallas	1996-2017	-	-4.4 (2008-2017)	7.4	5.4	1.6 (2009-2016)			
Andøya	2010-2017	-	-33	8.3	10	-			
Station Nord	2008-2017	23	44	8.9	-28	2.4			
Little Fox Lake	2003, 2008, 2013	-							

Years in () denote the period if it is different from the period in column 2.

NA = Compound analyzed, but half-lives not available.

Station	Period	ΣPCB	chlordane	ΣDDTs	HCB	α-HCH	γ-HCH
<i>NILU</i>							
Birkenes	1991-2018	(2004-2017)	(2010-2016)	NA (2010-2017)	(1993-2017)		
Zeppelin	1993-2018	(2001-2017, older data may be contaminated)		NA (1994-2017)			
Andøya	2009-2018	(2009-2016)	-	NA (2010-2016)		(2010-2016)	(2010-2016)

Years in () denote the period if it is different from the period in column 2.

NA = Compound analyzed, but half-lives not available.

Table 5.2.1b Continued

Station	Period	ΣPCB
UK-Norway Transect		
North Wyke	1994-2008	
Aberystwyth	1994-2008	
Hazelrigg	1994-2008	
Colonsay	1994-2008	
Ullapool	1994-2008	
Birkenes	2010-2016	
Ualand	1994-2008	
Osen	1994-2008	
Fureneset	1994-2008	
Namsvatnet	1994-2008	
Bodø	1994-2008	
Tromsø	2010-2016	
Lakselv	1994-2008	

Station	Period	ΣPCB
NAPS	2004-2009	Co-planar PCBs only

Station	Period	ΣPCB	aldrin	dieldrin	chlordane	ΣDDTs	heptachlor	HCB	α-HCH	β-HCH	γ-HCH	endosulfan	endosulfan sulfate
Sweden^f													
Råö	1996-2017	No decrease in recent years	NA	NA			NA	NA		NA			NA
Pallas	1996-2017	No decrease in recent years	NA	NA			NA	NA		NA			NA
Aspvreten	1996-2017	No decrease in recent years	NA	NA	NA (2009-2017)	NA (2009-2017)	NA	NA		NA	NA	-	-

^fFredricsson et al. (2018)

NA = Compound analyzed, but half-lives not available.

Table 5.2.1c Apparent first order half-lives ($t_{1/2}$, year) of PBDEs and HBCDD

Station	Period	ΣPBDEs	HBCDD
GAPS	2005-2017		NA

NA = Compound analyzed, but half-lives not available.

Station	Period	ΣPBDEs	HBCDD
GLB			
Point Petre (Lake Ontario)	1992-2017	-44 (2002-2017)	NA
Burnt Island (Lake Huron)	1992-2012	-4.2 (2005-2013)	NA
Egbert (Lake Huron)	1995-2006	NA	-

NA = Compound analyzed, but half-lives not available.

Station	Period	ΣPBDEs
IADN^d		
Chicago	1991-2017	11
Cleveland	1991-2017	5.6
Sturgeon Pt	1991-2017	
Sleeping Bear Dunes	1991-2017	
Eagle Harbor	1991-2017	
Point Petre	2013-2017	

^d Salamova et al. (2015)

Station	Period	ΣPBDEs	HBCDD
PureAlps			
Zugspitze	2005-2017	Declining BDE 28 and 154	
Sonnblick	2005-2017	-	NA

NA = Compound analyzed, but half-lives not available.

Station	Period	ΣPBDEs
SMP-POPs	2008-2017	

Station	Period	ΣPBDEs
TOMPs		
London	Jan 1991-2017	~8.3
Manchester	Jan 1991-2017	~8.3
Auchencorth	Dec 2008-2017	~22
Middlesbrough	Apr 1993- July 2008	~22
Hazelrigg	Sept 1992-2017	~22
High Muffles	Jan 1997-2017	~22
Weybourne	Dec 2008-2017	~22
Stoke Ferry	Feb 1997-Dec 2007	~22

~ = values are approximate

Table 5.2.1c Continued...

Station	Period	ΣPBDEs	HBCDD
AMAP			
Alert	1992-2017	(2002-2017)	NA
Zeppelin	1993-2017	(2006-2017)	NA
Stórhöfði	2008-2017	*	-
Pallas	2003-2017		-
Andøya	2010-2016		-
Station Nord	2014-2017	**	-
Little Fox Lake	2012-2014		-

Years in () denote the period if it is different from the period in column 2.

NA = Compound analyzed, but half-lives not available.

*BDE 47 decreasing; BDE 99 and 100 increasing but mostly non-detectable.

** BDE 47 increasing with t_2 of 20 y and BDE 99 decreasing with $t_{1/2}$ of 4.2 y

Station	Period	ΣPBDEs	HBCDD
NILU			
Birkenes	2006-2018	(2008-2017)	NA
Zeppelin	1993-2018		NA (2006-2018)
Andøya	2009-2016		-

NA = Compound analyzed, but half-lives not available.

Station	Period	ΣPBDEs
UK-Norway Transect		
North Wyke	2000-2016	
Aberystwyth	2000-2016	
Hazelrigg	2000-2016	NA
Colonsay	2000-2016	
Ullapool	2000-2016	
Birkenes	2010-2016	
Ualand	2000-2016	
Osen	2000-2016	
Fureneset	2000-2016	
Namsvatnet	2000-2016	
Bodø	2000-2016	
Tromsø	2000-2016	
Lakselv	2000-2016	

NA = Compound analyzed, but half-lives not available.

Station	Period	ΣPBDEs	HBCDD
Sweden^f			
Råö	1996-2017	(47, 99, 100 only)	NA
Pallas	1996-2017	(47, 99, 100 only)	NA
Aspvreten	1996-2017	(47, 99, 100 only)	NA

^fFredricsson et al. (2018)

NA = Compound analyzed, but half-lives not available.

Table 5.2.1d Apparent first order half-lives ($t_{1/2}$, year) of PFOS, PFOA, and fluorinated compounds

Station	Period	PFOS	PFOA	6:2 FTOH	8:2 FTOH	10:2 FTOH	MeFOSE	EtFOSE	MeFOSA	EtFOSA
AMAP										
Alert	Before peak year	-5.5 (2006-2013)	-6.6	ND (2006-2011)	-2.6 (2006-2011)	-3.3 (2006-2011)	-	-	-	-
Alert	After peak year	2.8 (2013-2017)	4.8	ND (2011-2017)	4.0 (2011-2017)	3.0 (2011-2017)	-	-	-	-
Alert	Entire period	NA (2006-2017)	NA (2006-2017)	13 (2010-2017)	NA (2010-2017)	NA (2010-2017)	NA (2006-2017)	NA (2006-2017)	NA (2006-2017)	NA (2006-2017)
Zeppelin	2006-2017	10	3.5	-	-	-	-	-	-	-
Andøya	2010-2017	13	2.3	-	-	-	-	-	-	-
Station Nord	2008-2017	-	-	-14	10	5.3	NA (2008-2017)	NA (2008-2017)	NA (2008-2017)	NA (2008-2017)

NA = Compound analyzed, but half-lives not available.

Station	Period	PFOS and related chemicals	PFOA and related chemicals
Sweden^f			
Rådö	2009-2017	No clear trend	No clear trend
Pallas	1996-2017	P	P

^fFredricsson et al. (2018)

P = Planned for future analysis

Table 5.2.1d Apparent first order half-lives ($t_{1/2}$, year) of PCDD/Fs, HCBd, SCCPs, PCP/PCA

Station	Period	SCCPs
Australia	2016	NA
EMEP	1993-2017	P

P = Planned for future analysis

Station	Period	SCCPs
GLB		
Point Petre (Lake Ontario)	1992-2017	P
Burnt Island (Lake Huron)	1992-2012	-
Egbert (Lake Huron)	1995-2006	-

NA = Compound analyzed, but half-lives not available.

P = Planned for future analysis

Station	Period	PCDD/Fs	HCBd	PCP/PCA
PureAlps				
Zugspitze	2005-2017			
Sonnblick	2005-2017			

Station	Period	PCDD/Fs
SMP-POPs	2008-2017	

NA = Compound analyzed, but half-lives not available.

Station	Period	PCDD/Fs
TOMPs		
London	Jan 1991-2017	~5
Manchester	Jan 1991-2017	~5
Auchencorth	Dec 2008-2017	~5.8
Middlesbrough	Apr 1993- July 2008	~5.8
Hazelrigg	Sept 1992-2017	~5.8
High Muffles	Jan 1997-2017	~5.8
Weybourne	Dec 2008-2017	~5.8
Stoke Ferry	Feb 1997-Dec 2007	~5.8

~ = values are approximate

Station	Period	HCBd	SCCPs
AMAP			
Alert	1992-2017	20	(1992, 1994-1995, 2011)
Zeppelin	2013-2017	-	

Station	Period	SCCPs
NILU		
Birkenes	2017-2018	
Zeppelin	2013-2018	

Station	Period	PCDD/Fs
NAPS	1989-2009	

5.2.1.5 Summary and Conclusions

- ❖ Most POPs that have been regulated for extended periods of time (>30 years) in the WEOG region, e.g. DDTs, the drins, PCBs and chlordanes, are now either showing slower rates of decline with longer half-lives or no further decreases are observable in recent years, indicating that they are approaching steady state with other environmental media.
- ❖ Measurements at locations closer to urban centers indicate that primary sources of PCBs stocked in cities (e.g. old transformers and electrical equipment and in waste streams) still continue to contribute to measured air concentrations resulting in relatively slow decreases with long half-lives (>10 years). Non-Aroclor related PCBs are found at some locations, reflecting unintentional release (e.g. PCBs in paints, combustion-related PCBs).
- ❖ HCB showed slightly increasing trends at some Arctic, European and Antarctic stations which may be related to secondary emissions due to warming and emissions as unintentional by-products from combustion; reflecting its high volatility and slow degradation in air.
- ❖ In the last reporting period, PBDEs related to the penta- and octa-BDE mixtures were not declining in North American air as was observed in Europe. With additional data in this reporting period, decreasing trends of these PBDEs are now observed reflecting the effectiveness of the regulation of these technical mixtures. On the other hand, decaBDE (BDE 209) was found to be increasing or not changing in concentrations over time which is probably because it was not yet regulated during this reporting period and its use as a replacement for the penta- and octa-BDE mixtures which were regulated earlier.
- ❖ Accelerated decline of endosulfan was observed in air as its usage was phased out domestically and globally under the Stockholm Convention.
- ❖ PFOS, PFOA and their precursor compounds are either showing accelerated declining trends or their concentrations have peaked (after earlier increasing trends) as a result of the global regulation of these chemicals.
- ❖ Time trends of some POPs (HBCDD, SCCPs, PCP/PCA, HCBd, PeCB, and toxaphene) and air concentration data of HBB, chlordecone, dicofol, and PCNs, are lacking; probably due to difficulties in chemical analysis, low or non-detectable concentrations in air, or sampling artefacts (e.g. breakthrough in active air sampling or high blank levels).
- ❖ Air monitoring programmes have started or are planning to measure the newer POPs (e.g. SCCPs, HBCDD, PFOA and related chemicals) to determine baseline concentrations. Additional data in the next reporting period will enable the assessment of temporal trends.

5.2.1.6 Recommendations, Challenges and Future Outlook

Many new POPs and emerging chemicals of concern have been in use for many years before their discovery. Air monitoring programmes should maintain sample or extract archives to be retrospectively analysed for assessing how levels of emerging contaminants have changed over time. The inclusion of suspected chemicals of concern in long-term monitoring programmes also helps to establish trends to inform policies and to monitor the effectiveness of regulations.

New POPs, such as HCBd, PBDEs, PFOS, PFOA and related chemicals, differ from legacy POPs in many aspects, including their physical-chemical properties, transport and transformation mechanisms and form of emission. Sampling strategies may need to change in order to capture these substances, e.g. include sorbents that can minimize breakthrough in active sampling and increase sampling capacity in passive sampling. Also, employ clean field and laboratory practices to constrain blank levels.

Some POPs (e.g. HBCDD, precursors of PFOS and PFOA) generally have very low concentrations in air, making the development of atmospheric trends difficult. Assessments of their historical atmospheric deposition through analysis in precipitation over time, as well as in dated ice/snow/sediment cores, can complement air measurements in the evaluation of regulatory effectiveness.

While the occurrence of a chemical in air at a remote location provides evidence for its long-range transport potential, the air concentrations of some POPs are very low and mostly non-detectable at some remote or background sites, e.g. consumer-product-related chemicals PBDEs, HBCDD, PFOS, PFOA and related compounds. Monitoring programmes should consider increasing monitoring at locations closer to known sources, e.g. in urban and/or industrial areas, to assess emission changes over time in response to regulation.

The increasing number of POPs continue to add pressure on monitoring programmes and analytical laboratories in terms of funding, resources and need for development of new sampling and analytical methodologies. Monitoring strategies need to be reviewed periodically to ensure the sustainability of the programmes. It is notable that some programmes have reduced the number of sampling sites and/or sampling frequencies since the last reporting period. Some programmes have taken samples but have only analysed a few selected POPs in selected years, without any reportable temporal trends. It is recommendable to develop a monitoring strategy such that the long-term temporal trends of POPs can still be assessed with a reduced sampling or analytical schedule. For instance, the GAPS program compared concentrations of PFOS, PFOA and related compounds measured in 2013 and 2015 with those reported from 2009, to assess trends over 7 years of monitoring (Rauert et al., 2018b). Archived air samples or extracts can also be used to retrospectively analyse for POPs to rebuild time trends when funding and resources become available in the future.

While the concentrations of most POPs are declining in air, the number of chemicals that have been identified as POPs and their related chemicals (precursors, transformation products), as well as chemicals used as their substitutes after regulation, form complex mixtures in the environment that may result in various toxicity end points. Some monitoring programmes have developed

strategies to screen air samples for new chemicals of concern and to assess the potential hazard of chemical mixtures. Such techniques include database screening of global commercial chemical inventories for substances with POP-like properties, a combination of non-target and targeted analysis to verify the presence of suspect chemicals and their transformation products, and sensitive in vitro techniques to assess different toxicity endpoints, such as transcriptomic studies for toxicity profiling. There is increasing need for better connections between air monitoring and long-term effects of human and environmental exposure to mixtures of chemicals.

Observed changes in temporal trends of many POPs indicate influence of climate and social/economic changes. Environmental fate and transport modelling is useful in linking chemical emissions to observed changes of POPs in air and other media and can help to better understand and interpret observed trends. On the other hand, inverse modelling using observed measurements in the environment can help constrain changes in chemical emissions; further informing the Convention on the effectiveness in chemical management strategies.

5.2.2 Human tissues (milk and/or blood)

5.2.2.1 Background

The analysis of trends of POPs in human tissues is based on data that have been collected with the aim to evaluate international, and when possible national, trends of pollutants in breast milk and human blood. Data have been collected from several sources and cover several years (Table 4.2b). Datasets from national and international coordinated biomonitoring programmes constitute a majority of the observations in the dataset. These datasets are complemented with data on breast milk retrieved from peer reviewed scientific papers.

Variation in concentrations of POPs in human tissues is the result of several underlying factors that contribute to the overall variation. Non-time dependent variations (such as regional differences and sampled population) thus influence the robustness of trends presented in this chapter. The data sources used cover a broad range of monitoring and survey strategies focusing on targeted populations (such as students in the German ESB and socio-cultural cohorts in AMAP and peer reviewed papers) or using a multistage, probability sampling design that obtain nationally representative concentrations (NHANES). It is important to note that differences in data collection strategies between the data sources decrease the potential for evaluation of general trends and patterns across the WEOG region when data are jointly evaluated.

To facilitate the analysis of trends and its interpretation, metadata are included for each source of data used in this chapter. Age of the mother and the proportion of primipara (woman who has had one pregnancy) are two such factors that are commonly used for POPs data on breast milk that are important for interpretation of concentration of POPs. Variation in POP concentration levels in blood is affected by age, gender, life history and socio-cultural factors as well.

5.2.2.2 Quality assurance and quality control and data treatment

During the work to assemble data on POPs in human tissue, a data treatment procedure has been conducted to harmonize data. This was achieved by setting up a process of quality assurance and quality control (QA/QC) practices. The overall aim of these processes was to get a common standard for the data to ensure comparability within and between data sources. All data collected were assembled in a primary dataset that contained all raw data from each data source. Separate primary datasets were created for breast milk and human blood data using variables presented in Table 5.2.2 (human milk) and Table 5.2.3 (human blood). After a series of data treatment steps and selection of data to improve comparability of data, final datasets for breastmilk and human blood were produced. These final datasets were then used for analysis of trends and patterns of POPs in human tissue. A more extensive description of the QA/QC and data treatment can be found in Annex 8.2.

Table 5.2.2 Variables included in the primary dataset for trend analysis in POPs in human milk.

Variable name	Description
Substance	Compound name
Substance group under Stockholm Convention	Substances as defined by the Stockholm Convention
Concentration	Value of POP concentration in breast milk
Unit	Unit of concentration, DL and QL
Year	Year of sampled data
Country	WEOG country origin of sample
Continent	Continental group of WEOG country
Matrix	Sampling matrix (breast milk)
Age	Age of mother at sampling
Primipara	Percentage of <i>primipara</i> ^a in sample
Detection	Proportion of data below DL or QL
LD	Limit of detection
LQ	Limit of quantification
TEQ	WHO toxic equivalent scheme used for data on dioxins, furans and dl-PCBs: TEQ 1990, TEQ 1998 (LB and UB), TEQ 2005 (LB and UB)
Data info	Type of data: single measurement or aggregated data (mean, median, min, max, percentiles)
Sample info	Information if data is based on pooled sample or individual samples
Reference	Reference for collected data

^a a woman who has had one pregnancy.

Table 5.2.3 Variables included in the dataset for trend analysis in POPs in human blood.

Variable name	Description
Substance	Compound name
Substance group under Stockholm convention	Substances as defined by the Stockholm Convention
Concentration	Value of POP concentration in human blood
Unit	Unit of concentration, DL and QL
Year	Year of sampled data
Country	WEOG country origin of sample
Region	Region within country that have been sampled
Continent	Continental group of WEOG country
Matrix	Sampling matrix (serum, plasma, blood)
Age	Age of sampled cohort
Gender	Information on gender in sampled cohort
Detection	Proportion of data below DL or QL
Quantity	Number of samples used in summary statistics
Number BDL	Number of samples below DL or QL
LD	Limit of detection
LQ	Limit of quantification
Statistical measure	Type of data: single measurement or aggregated data (mean, median, min, max, percentiles)
Cohort information	Metadata about sampling cohort based on stratification of sampled population (gender, age group, lifestyle)
Source	Sampling or monitoring programme

5.2.2.3 Human milk

Primary dataset

All collected data for human milk were compiled into the primary human milk data set. It contains 18 593 observations covering a period ranging between 1972 and 2017 (Figure 5.2.9). To address factors that can be considered important for interpreting trends in POPs in breast milk, metadata were added for comparison between data sources. Such factors are the type of data that have been retrieved that may represent primary non-aggregated (raw data) or aggregated (mean, median, min, max, percentile) data as well as primipara and age. Data sources that contributed to the primary dataset and summaries of data retrieved from each data source are specified below.

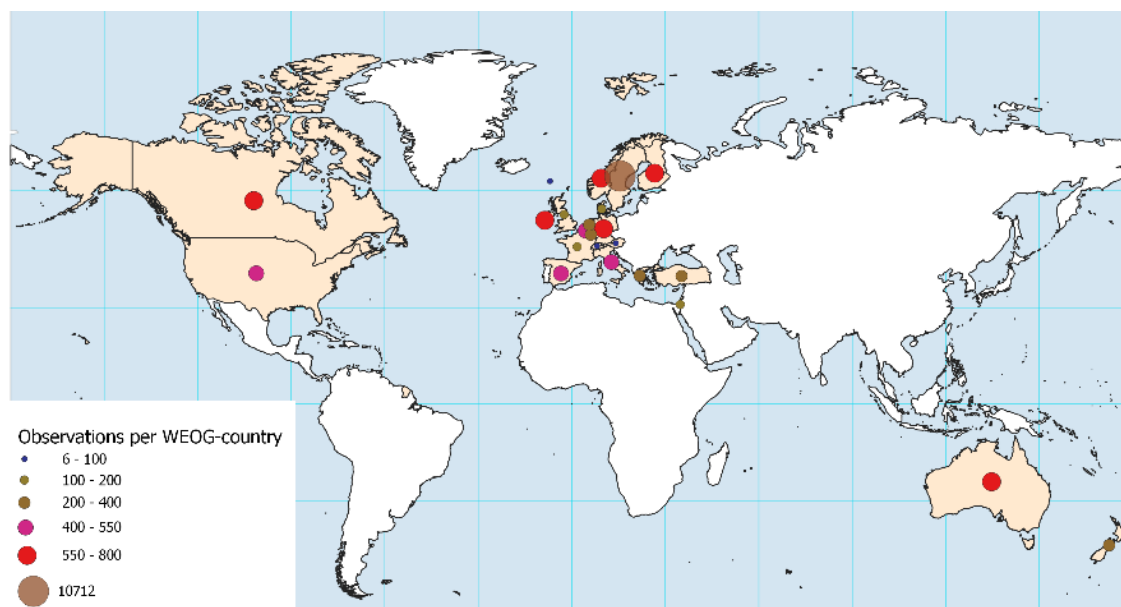


Figure 5.2.9. Distribution of observations per country in the WEOG region in primary human milk dataset used for trend analysis of POPs.

WHO/UNEP milk survey

Human milk data from WEOG countries from the Global Monitoring Plan Data Warehouse (GMP DWH) were used to include a comparable and harmonized data set on POP concentrations in breast milk across several countries (Figure 5.2.10). Data from the latest sampling, in 2019, was not included in this report since the data was not accessible at the time when the data was

collected for this report. The GMP DWH encompasses all relevant documentation on data collection, analysis as well as data storage and data that are provided through their web portal. This documentation can be found in Hulek et al. (online: www.pops-gmp.org/visualization-2014) and background documentation. Breast milk data were downloaded for breast milk matrix for WEOG countries.

Criteria for selecting donors for the WHO/UNEP milk survey population are specified in Guidelines for developing a national protocol in the Fourth WHO-Coordinated Survey of Human Milk for Persistent Organic Pollutants in Cooperation with UNEP (2007). This document gives a general starting point for selection of donors for this survey that they should be primipara and should be under 30 years of age. The primary dataset from WHO milk survey contains 4 369 observations over a period from 1987 to 2014 (Table 5.2.4).

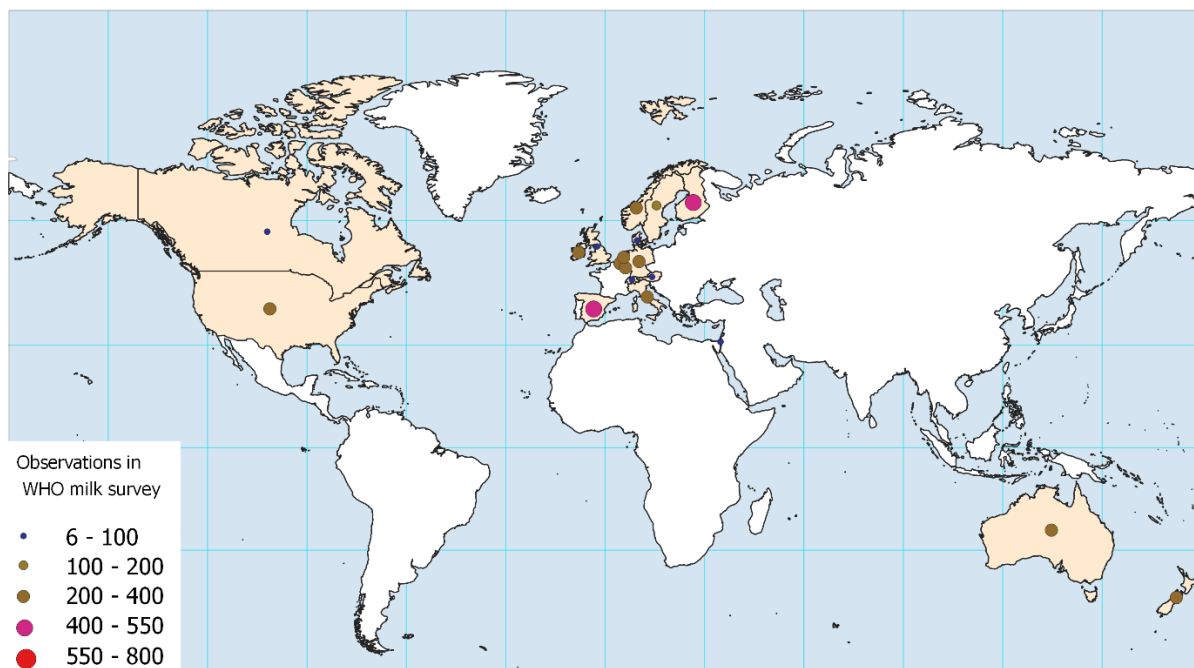


Figure 5.2.10. Distribution of observations per country in the WEOG region in data retrieved from the WHO/UNEP milk survey.

Table 5.2.4. Summary of data retrieved from the UNEP/WHO milk survey.

Stockholm Convention substances	N	Range year	N statistical measure					Proportion below DL / QL (%)
			non-aggregated	mean	median	min/max	percentiles	
Aldrin, dieldrin and endrin	65	2001 – 2012	36	2	5	22	0	0 / 60
Chlordane	78	2001 – 2012	39	3	6	30	0	0 / 67
Chlordecone	0							
Chlorinated Paraffins	0							
DDT complex	208	2001 – 2012	104	8	16	80	0	0 / 45
dioxins/furans	2105	1987 – 2014	889	64	256	896	0	0.14 / 2.14
Endosulfan	78	2001 – 2012	39	3	6	30	0	0 / 100
Flame retardants	0							
HBB	0							
HBCD	15	2006 – 2012	9	0	0	6	0	0 / 67
HCB	26	2001 – 2012	13	1	2	10	0	
HCH	78	2001 – 2012	39	3	6	30	0	0 / 47
Heptachlor	69	2001 – 2012	45	1	5	18	0	0 / 48
Mirex	13	2006 – 2012	10	0	1	2	0	0 / 100
OCP	0							
PAC	0							
PBB	6	2009 – 2012	6	0	0	0	0	0 / 83
PBDE	182	2001 – 2014	91	7	14	70	0	0.6 / 7.7
PCB	1 362	1987 – 2014	457	48	185	672	0	0.5 / 3.2
Pentachlorophenol	0							
PeCB	6	2009 – 2012	6	0	0	0	0	0 / 100
PFAS	0							
Toxaphene	78	2001 – 2012	39	3	6	30	0	0 / 64
Summary	4 369	1987 - 2014	1 822	143	508	1 896	0	0.25 / 12

Swedish national monitoring program

The Swedish Environmental Protection Agency has the overall responsibility for coordinating national health related environmental monitoring in Sweden. This monitoring involves long-term monitoring of environmental pollutants that can affect human health. Data used in this report are non-aggregated data meaning that data represent individual measured concentrations from persons that have been sampled in different regional campaigns across Sweden.

Table 5.2.5. Summary of data retrieved from national monitoring in Sweden.

Stockholm Convention substances	N	Range year	N statistical measure ^a					Proportion below DL / QL (%)
			non-aggregated	mean	median	min/max	percentiles	
Aldrin, dieldrin and endrin	0							
Chlordane	334	2002 – 2016	334	0	0	0	0	
Chlordecone	0							
Chlorinated Paraffins	0							
DDT complex	719	2002 – 2016	719	0	0	0	0	0 / 50
dioxins/furans	1 989	1972 – 2016	1989	0	0	0	0	0 / 9
Endosulfan	0							
Flame retardants	0							
HBB	0							
HBCD	213	2002 – 2016	213	0	0	0	0	0 / 26
HCB	184	2002 – 2016	184	0	0	0	0	
HCH	218	2002 – 2016	218	0	0	0	0	0 / 16
Heptachlor	0							
Mirex	0							
OCP	0							
PAC	36	2012 – 2017	36	0	0	0	0	0 / 33
PBB	0							
PBDE	1 930	2002 – 2016	1 930	0	0	0	0	0 / 57
PCB	3 665	1972 – 2016	3 665	0	0	0	0	0 / 19
Pentachlorophenol	0							
PeCB	0							
PFAS	650	1972 – 2015	650	0	0	0	0	0 / 19
Toxaphene	0							
Summary	9 938	1972 – 2017	9 990	0	0	0	0	0 / 26

^a number of observations being a = non-aggregated data, b = mean, c = median, d = min/max, e = percentiles.

Peer reviewed literature

Data from peer-reviewed literature represent a compilation based on scientific publications between 1972 and 2017 (Figure 5.2.11). The data are based on results that previously have been published by Fång et al. (2015) (n= 1723) that covered a period between 1995 and 2011. To cover data that represent more recent years this dataset was complemented with data in literature published after 2012 (n = 2511).

The population of mothers in the peer review literature had a mean age of 30 years (min/max: 24.5 / 35) and were 53 ± 45 % (mean ± SD) primapara. The primary dataset originating from literature contains 4 294 observations over a period from 1972 to 2017 (Table 5.2.6).

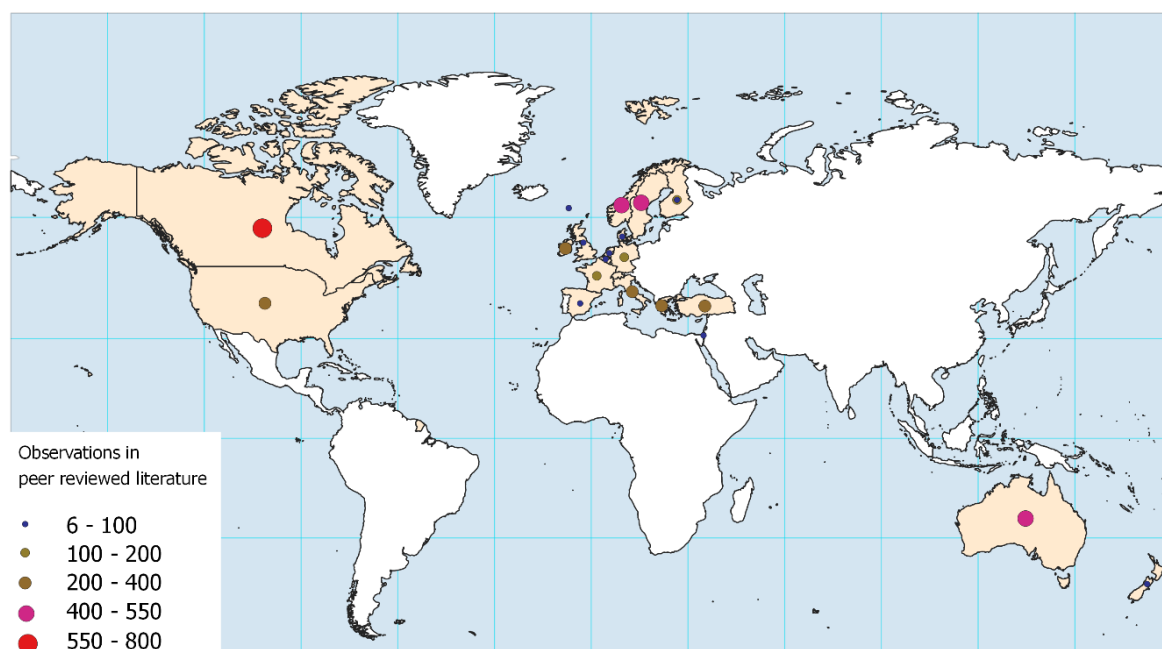


Figure 5.2.11. Distribution of observations per country in the WEOG region in data on POPs in human milk retrieved from peer review literature.

Table 5.2.6. Summary of data on POPs in human milk retrieved from peer review literature.

Stockholm Convention substances	N	Range year	N statistical measure					Proportion below DL / QL (%)
			non-aggreg.	mean	median	min/max	percentiles	
Aldrin, dieldrin and endrin	53	1988 – 2013	1	22	30	0	0	0 / 1
Chlordane	101	1992 – 2013	0	46	45	0	10	0 / 0
Chlordecone	1	2006 – 2006	0	1	0	0	0	
Chlorinated Paraffins (CPs)	60	1996 - 2016	0	60	0	0	0	5 / 0
DDT complex	432	1972 – 2015	3	266	146	0	17	0 / 0.23
Dioxins /furans	811	1972 – 2017	319	349	141	0	2	0 / 0.74
Endosulfan	19	1999 – 2009	0	12	7	0	0	0 / 0
Flame retardants	0							
HBB	3	2005 – 2013	0	2	1	0	0	0 / 0
HBCD	140	1980 – 2014	48	55	29	0	8	8 / 14
HCB	160	1972 – 2013	0	96	56	0	8	0 / 0
HCH	299	1990 – 2014	1	194	95	0	9	0 / 2
Heptachlor	80	1988 – 2013	0	57	22	0	1	0 / 1.25
Mirex	46	1988 – 2013	0	14	32	0	0	
OCP	9	2006 – 2013	0	3	3	0	3	
PAC	0							
PBB	16	1999 – 2008	0	4	3	0	9	12.5 / 37.5
PBDE	766	1972 – 2015	110	274	242	0	140	7.4 / 6.8
PCB	1 159	1972 – 2015	116	552	341	0	107	0 / 0
Pentachlorophenol	0							
PeCB	28	1992 – 2009	0	6	22	0	0	
PFAS	108	1972 – 2014	0	72	21	0	15	0 / 1.85
Toxaphene	3	1996 - 1998	0	2	1	0	0	0 / 1
Summary	4 294	1972 – 2017	598	2 027	1 237	0	329	2 / 2.2

Final dataset human milk

Final dataset was created by treatment of data from primary dataset. In the primary dataset, both aggregated data, presented as central tendencies (mean and median) and other summary statistics (percentiles, and min/max), as well as non-aggregated individual data were present. The combination of both aggregated data and non-aggregated data in the primary dataset lead to a potential reporting bias since observations of individual data will become more influential in the analysis compared to aggregated data. Aggregated data also represent populations from spatial or age stratification or other grouping factors that have been used in a study. To derive a final dataset with concentration data that are comparable across different sources of data, the primary dataset was harmonized before trend analysis. Harmonization of data was done by first excluding data presented as min/max or percentiles. The harmonization process was further done by calculating medians from individual concentrations from the primary dataset by groups defined by 1) year, 2) country, 3) region, 4) reference, and when applicable 5) TEQ. For studies that presented several measures of central tendencies, median values were used preferentially to mean values. In cases where only mean values were presented, the mean value was used as a measure for central tendency. LD and LQ were also calculated as a mean for the reported LD and LQ for each aggregate.

The final breast milk dataset contained 7 295 observations (Table 5.2.7). The highest number of observations in the final dataset originated from Scandinavia (N: Sweden = 606, Norway = 470), Canada (N = 592) and Australia (N = 410) followed by USA (N = 330), Ireland (N = 285), Turkey (N = 242), Italy (N = 217) and Greece (N = 207) (Figure 5.2.12).

Some substance groups were reported both as sums as well as concentration of individual substances, (*i.e.* PBDEs, dioxins/furans and PCBs). It is important to note that there may be cases in the summary statistics where concentrations of individual substances were higher compared to summed concentrations for comparable substance groups in a region. This is due to an effect that several different data sources have contributed with data for a specific region and sampled populations differed between these data sources as well as due to the differences in the substance groups that have been targeted in their respective analysis.

Table 5.2.7. Summary of data in final human milk dataset for POPs.

Stockholm Convention substances	N	N per data source			Range year	N statistical measure		Proportion below DL / QL (%)
		WHO/UNEP milk survey	National monitoring programs	Peer review data		mean	median	
Aldrin, dieldrin and endrin	75	36	0	39	1988 – 2013	9	66	0 / 30.7
Chlordane	175	39	72	64	1992 – 2016	20	155	0 / 15.4
Chlordecone	1	0	0	1	2006 – 2006	1	0	0 / 0
Chlorinated Paraffins (CPs)	60	0	0	60	1996 - 2016	60	0	5 / 0
DDT complex	558	104	171	283	1972 – 2016	139	419	0.2 / 23
Dioxins /furans	2 217	889	711	617	1972 – 2017	235	1 982	0 / 6
Endosulfan	54	39	0	15	1999 – 2012	8	46	0 / 72
Flame retardants	0							
HBB	2	0	0	2	2005 – 2013	1	1	0 / 0
HBCD	158	9	45	104	1980 – 2016	30	128	7 / 22
HCB	162	13	45	104	1972 – 2016	50	112	0 / 0
HCH	293	39	63	191	1990 – 2016	98	195	0 / 16
Heptachlor	105	45	0	60	1988 – 2013	39	66	0 / 38
Mirex	45	10	0	35	1988 – 2013	10	35	0 / 22
OCP	4	0	0	4	2006 – 2013	1	3	0 / 0
PAC	36	0	36	0	2012 – 2017	0	36	0 / 33
PBB	13	6	0	7	1999 – 2012	4	9	8 / 54
PBDE	925	91	385	449	1972 – 2016	106	819	3 / 24
PCB	2 128	457	1 021	650	1972 – 2016	267	1 861	0 / 11
Pentachlorophenol	0							
PeCB	22	6	0	16	1992 – 2012	3	19	0 / 27
PFAS	251	0	171	80	1972 – 2015	59	192	1 / 17
Toxaphene	41	39	0	2	1996 - 2012	1	40	0 / 51
Summary	7 295	1 822	2 720	2 783	1972 – 2017	1 141	6 184	1 / 14

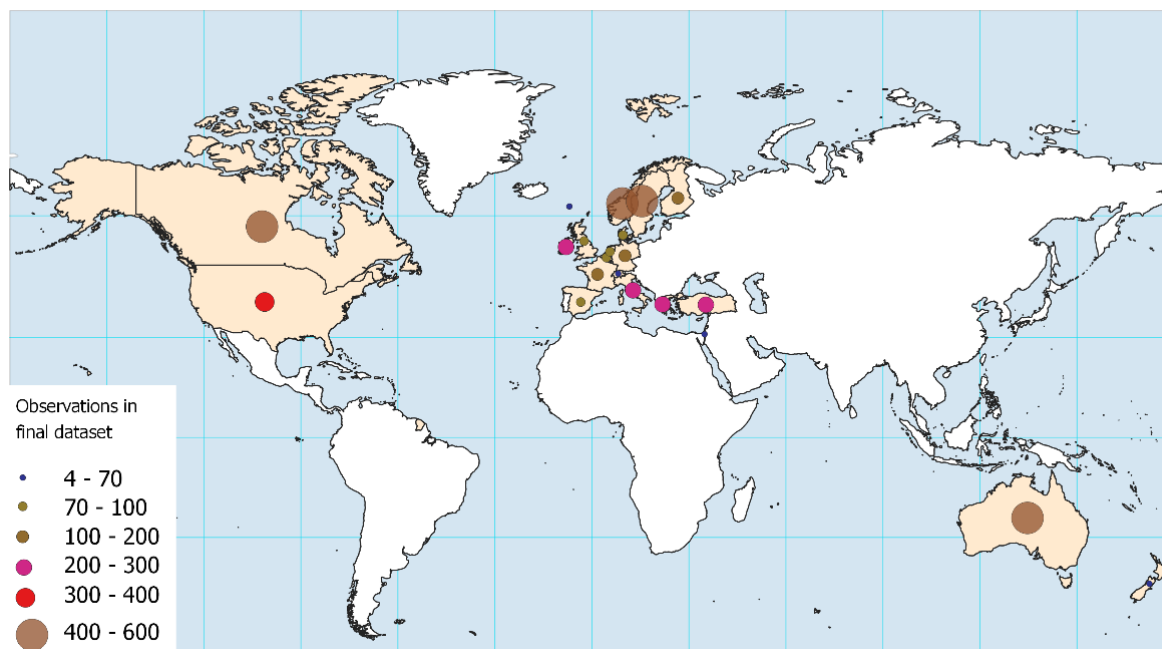


Figure 5.2.12. Distribution of observations per country in the WEOG region in final dataset used for trend analysis of POPs in human milk.

5.2.2.4 Human blood

Primary dataset

All collected data for human blood were compiled into the primary human blood data set. It contains 24 563 observations covering a period ranging between 1982 and 2017 (Table 5.2.8). Majority of data in the primary dataset are aggregated (geometric/arithmetic means and medians) with the exception of Spanish data (BIOAMBIENT.ES) for which individual data are available. Data sources that contributed to the primary dataset and summaries of data retrieved from each data source are specified below.

Table 5.2.8. Summary of observations from data sources in primary blood dataset.

Stockholm Convention substances	N							
	Total	AMAP	Australia	Canada	Germany	USA	Spain	Sweden
Aldrin, dieldrin and endrin	2 817			24			2 793	
Chlordane	687	138	364	120		36		29
Chlordecone	0							
Chlorinated Paraffins	0							
DDT complexes	2 501	177	546	48		54	1 640	36
Dioxins /furans	1 017		864			153		
Endosulfan	0							
Flame retardants	8							8
HBB	2							2
HBCD	0							
HCB	1 226	79	182		214	18	709	24
HCH	3 323	58	364	48		36	2 793	24
Heptachlor	931						931	
Mirex	258	34	182	24		18		
OCP	0							
PAC	0							
PBB	36			24		12		
PBDE	2 077	121	1 404	216		204		132
PCB	14 228	328	432	600	558	693	11 203	414
PeBEB	2							2
Pentachlorophenol	217	4			213			
PeCB	0							
PFAS	6 900	136	633	288	615	192	4 494	542
Toxaphene	68	20		48				
Summary	36 368	1 095	4 971	1 440	1 600	1 416	24 563	1 283

Arctic Monitoring and Assessment Program (AMAP)

Data on POPs in blood were retrieved from results presented in the AMAP Assessment report on human health in the Arctic. These data originated from biomonitoring activities across eight Arctic countries (USA (Alaska), Canada, Denmark (Greenland), Faroe Islands, Iceland, Norway, Russia, and Sweden) and were presented as aggregated data (geometric and arithmetic means). The AMAP Human Health Assessment Group established a QA/QC programme (the AMAP Ring Test) for ensuring quality of data produced by laboratories that have delivered data of POPs in human tissue to the AMAP report. Data from AMAP used in the primary dataset covered a period between 1986 and 2013 and contained 1 095 observations of aggregated data (arithmetic mean) (Table 5.2.8).

Several of the monitoring data in the AMAP Assessment Report were accompanied by information on human population cohorts. This information has been used by AMAP to track effects from contaminant exposure that were related to socio-cultural conditions and diet. Cohort information were not used in the evaluation of trends and patterns in POPs blood data in this report but were noted in case this influenced the observed trends.

Australia

The National Research Centre for Environmental Toxicology and the University of Queensland in Australia have coordinated biomonitoring activities of the Australian population for POPs. The data collection in the Australian human biomonitoring has used a pooling strategy where individual specimen has been pooled based on gender, age strata and regions across Australia. 4 971 observations were used in the primary blood dataset and covered a period between 2002 and 2016 (Table 5.2.8).

The Canadian Health Measures Survey (CHMS)

The CHMS collect information that help the evaluation and the extent of health problems among Canadians that, among other objectives, relates to environmental exposures, such as POPs. The biomonitoring of CHMS is an ongoing national survey with data collected in two-year cycles and POPs are measured pre individual. Aggregated data were downloaded from the CHMS Human Biomonitoring Data for Environmental Chemicals dataset. Biomonitoring data from CHMS were presented as aggregated data (median) for each substance summarized by biological matrix (blood), biomonitoring cycle with information of gender and age strata. The primary CHMS dataset contained 1 440 observations and covers a period between 2008 and 2016 (Table 5.2.8).

USA, National Health and Nutrition Examination Survey (NHANES)

The National Health and Nutrition Examination Survey (NHANES) is a program of studies designed to assess the health and nutritional status of adults and children in the United States. NHANES collects and releases data on POPs in blood in two-year cycles using a complex multi-stage sampling design. In order to obtain nationally representative data sample weights are assigned to each sampled person. The sample weights are a measure of the number of people in the population represented by that sample person in NHANES.

Data on concentrations of POPs and weights in blood were downloaded from the NHANES dataset and covered the period 2005 – 2015. Sample weights were applied to NHANES POPs blood concentrations according to a standard procedure as advised by the NHANES tutorial for environmental chemical data. The primary blood dataset contained 1 416 aggregated data (geometric means) from NHANES (Table 5.2.8).

Biomonitorización de Contaminantes en la Población Española (BIOAMBIENT.ES)

Data for a primary dataset from Spain (BIOAMBIENT.ES) were delivered by the Instituto de Salud Carlos III (ISCIII) and Spanish Ministry of Environment (MITECO). The human biomonitoring programme BIOAMBIENT.ES was a Spanish survey designed to collect samples and analyse POPs blood data from the years 2009 – 2010. The study had a stratified cluster sampling design that covered all geographical areas, sex and ages. The aim of the monitoring programme was to obtain a representative sample of the Spanish workforce. The primary dataset for blood from BIOAMBIENT.ES contained 24 563 non-aggregated data (Table 5.2.8).

German Environmental Specimen Bank (German ESB)

The German Environmental Specimen Bank (German ESB) serves to investigate, document and archive environmental and human exposure to pollutants in Germany at a specific point in time. Data from German ESB are owned by the German Environment Agency (UBA). Data from the German ESB cover a period between 1982 and 2010 with 1 600 observations of aggregated data (geometric means) (Table 5.2.8). Samples for the German ESB blood monitoring represent populations of students (even number of female and male, aged 20-29 years) from different regions in Germany (Münster, Greifswald, Halle/Saale and Ulm).

Swedish National Monitoring

The blood data contained by the Swedish Health-Related Monitoring covers a period 1987 – 2017 with a total number of 1 275 observations (Table 5.2.8). Observations in the dataset were aggregated (arithmetic means) and contained information on gender, age strata and population cohorts (i.e. fish consumers, pregnancy, cord blood).

Final dataset blood

Final dataset was created by treatment and harmonisation of data from primary dataset. The primary blood dataset contained blood data from all ages, including cord blood and data for children (<18 yrs). During the data harmonisation only data from adults (>18 yrs) were used in the final dataset. The data harmonisation was also done by selecting a subset of substances from the primary blood dataset based on their abundance and to what extent they complemented substances used in the final breast milk dataset.

Data from Spain (BIOAMBIENT.ES) were introduced in the primary dataset as non-aggregated POPs concentration data and were accompanied by information on age and gender. Before being included into the final blood dataset, median POP concentrations were calculated from the BIOAMBIENT.ES data for groups based on gender and age strata (18 – 25, 26 -30, 31 – 35, 36 – 40, 41 – 45, 46 – 50, 51 – 55, 56 – 60, 60 – 70 yrs.).

The final blood dataset contained 4 387 observations (Table 5.2.9) originating from Western Europe and others (N: Denmark = 16, Faraoe Islands = 24, Germany = 729, Greenland = 268, Iceland = 31, Norway = 12, Russia = 54, Spain = 304, Sweden= 484), North America (Canada = 391, USA = 316) and Australasia (Australia = 1 758) (Figure 5.2.13).

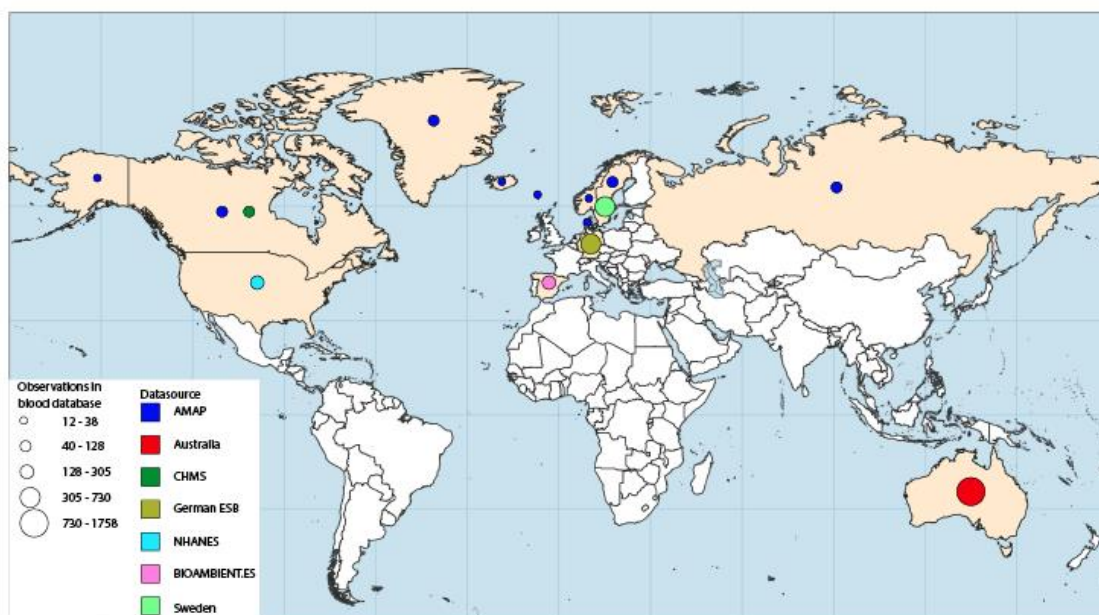


Figure 5.2.13. Distribution of observations per country in the WEOG region in dataset used for trend analysis of POPs in human blood.

Table 5.2.9. Summary of final dataset for POPs in human blood.

Stockholm Convention substances	N	N per data source							Year	
		AMAP	Australia	Canada	Germany	USA	Spain	Sweden	min	max
Aldrin, dieldrin and endrin	32	0	0	0	0	0	32	0	2009	2010
Chlordane	174	61	54	24	0	12	0	23	1992	2015
Chlordecone	0									
Chlorinated Paraffins	0									
DDT complexes	372	129	163	24	0	24	32	0	1992	2015
Dioxins /furans	681	0	522	0	0	102	0	57	1987	2012
Endosulfan	0									
Flame retardants	8	0	0	0	0	0	0	8	2011	2017
HBB	2	0	0	0	0	0	0	2	2016	2017
HBCD	0									
HCB	431	68	82	0	213	12	32	24	1985	2015
HCH	206	49	80	24	0	12	32	9	1992	2015
Heptachlor	32	0	0	0	0	0	32	0	2009	2010
Mirex	69	29	16	12	0	12	0	0	1992	2015
OCP	0									
PAC	0									
PBB	12	0	0	0	0	12	0	0	2005	2015
PBDE	815	103	557	24	0	54	0	77	2002	2017
PCB	553	131	80	48	186	24	36	48	1991	2015
PeBEB	2	0	0	0	0	0	0	2	2016	2017
Pentachlorophenol	215	2	0	0	213	0	0	0	1985	2012
PeCB	0									
PFAS	783	76	204	107	117	14	108	157	1982	2017
Toxaphene	0									
Summary	4 387	648	1 758	263	729	278	304	407	1982	2017

5.2.2.5 Time trends

Aldrin, dieldrin and endrin

Human milk

The three compounds aldrin, dieldrin and endrin have been measured in breast milk on one or more occasions in 21 countries within the WEOG region during the period 1990 - 2013. Most of the reported mean and median values for aldrin and endrin were below the quantification limit except for one aldrin mean value from Turkey and one from Australia. Both the quantifiable concentrations were however lower than the set quantification limit for the other studies which results in an average concentration that is lower than the limit of quantification in these regions. One data point from a study in Turkey was eliminated from the data analysis as the samples were collected from women with expected high exposure (seasonal agriculture workers). One data point from the Netherlands, collected in 1988, was eliminated due to high detection limits at the time of analysis. Out of the three substances, dieldrin was the most reported (46 mean and median values) with highest average of mean and median value in Australasia region (24 ng/g lw, n=6) followed by North America (8.6 ng/g lw, n=11) and Europe (4.2 ng/g lw, n=29), see Table 5.2.10.

A relative time trend for dieldrin within the WEOG during period 1990 to 2013 can be seen in Figure 5.2.14. Dieldrin has a decreasing trend in the Australasia region during the two decades while only small variations of the average median level of dieldrin has been observed in Europe (<6 ng/g lw). As has been reported in the previous global monitoring report for the WEOG region as well as in literature (9), the aldrin and endrin levels are below detection or quantification limits in the majority of the reported median values. The decreasing trend for dieldrin that was observed in the previous report is maintained but has been stabilizing since 2010. Due to the long time ban of these substances, the concentrations are expected to be low. As aldrin degrades into dieldrin, a higher detection frequency for dieldrin compared to aldrin is expected.

A decrease in the detection limits for dieldrin has occurred during the measuring period, where detection limits of 5 ng/g lw were reported in the studies from the early 1990s. In the most recent reported median value, the detection limit for dieldrin was 0.034 ng/g lw.

Table 5.2.10. Summary statistics of concentrations in human milk concentrations for aldrin, dieldrin and endrin in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
Aldrin	Europe	ng/g lw	9	0.22	0.25 ³	0.083	0.002 - 0.25	8	1, 20, 33
	Australasia	ng/g lw	3	0.18	0.25 ³	0.12	0.05 - 0.25	2	1, 9
	WEOG	ng/g lw	12	0.21	0.25³	0.088	0.002 - 0.25	10	
Dieldrin	Europe	ng/g lw	29	4.2	2.8	3.5	0.018 - 14	0	1, 9, 14, 20, 29, 30, 49, 59
	North America	ng/g lw	11	8.6	6.3	7.5	1.09 - 30	0	9
	Australasia	ng/g lw	6	24	0	13	9.3625 - 40	0	1, 9
	WEOG	ng/g lw	46	7.8	4.9	9.1	0.018 - 40	0	
Endrin	Europe	ng/g lw	12	0.23	0.25 ¹	0.058	0.048 - 0.25 ¹	11	1, 20
	Australasia	ng/g lw	2	0.25 ¹	0.25 ¹	0	0.25 ¹ - 0.25 ¹	2	1
	WEOG	ng/g lw	14	0.24	0.25³	0.054	0.048 - 0.25³	13	

¹ Data below LOD or LOQ

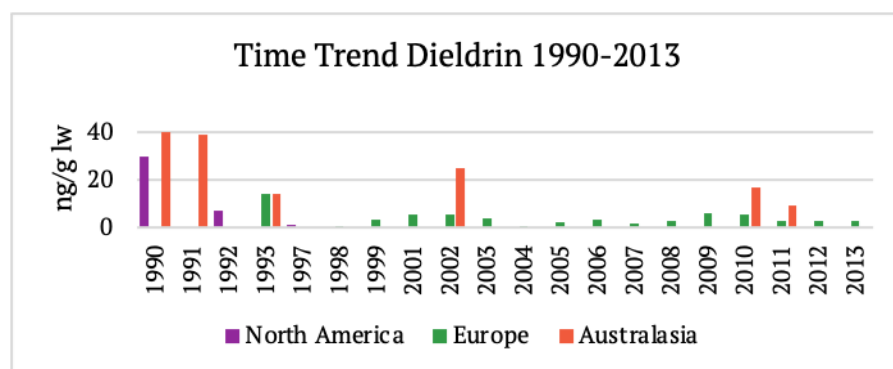


Figure 5.2.14. Time trend (1990 – 2013) for reported human milk concentrations for dieldrin in North America, Europe and Australasia.

Human blood

Aldrin, dieldrin and endrin have only been analyzed in human serum from Spain (in year 2009 and 2010) within the WEOG region (Table 5.2.11). The concentrations of Aldrin and Endrin were below the limit of quantification whereas for Dieldrin the ratio of measured data above LOQ ranged from 0-62%.

Table 5.2.11. Summary statistics of human blood (serum) concentrations on wet weight basis for Dieldrin in WEOG region (Spain) (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
Dieldrin	Europe (Spain)	ng/g ww serum	32	0.031	0.020	0.022	0.015 -0.082	32	65

Polychlorinated biphenyls (PCBs)

Human milk

Different congeners of PCBs in breast milk were measured in 23 countries within WEOG regions since 1972. Among 209 PCB congeners, only 130 could be found in commercial mixtures that could be divided into 2 subgroups, dioxin-like PCBs (dl-PCBs) and non-dioxin-like PCBs. The members in dl-PCBs group including CB 77, 81, 126, 169, 105, 118, 156, 157, 167, 114, 123, and 189 have co-planar structures and consequently similar structures and effects as the dioxins.

CB118 and CB153 were the two most common congeners and thus these two PCBs were usually used as indicator for the occurrence of PCBs. Both CB118 and CB153 indicated similar pattern of average values in breast milk with highest value in Europe 11 ng/g lw (N=34) and 64 ng/g lw (N=64), respectively (Table 5.2.12). As for sum PCBs, North America showed highest value for average of median values (114 ng/g lw, N=25), followed by Europe (75.5 ng/g lw, N=163) and Australasia (4.5 ng/g lw, N=18).

There was a clear decreasing trend of PCBs for indicators including CB153 (period 1987-2015) (Figure 5.2.15), CB118 (period 1992-2015) (Figure 5.2.16), and sum (6) PCB (period 1987-2014) (Figure 5.2.17) as well as sum PCBs (period 1987-2014) (Figure 5.2.18) in WEOG regions. Sweden started to monitor PCBs in breast milk in 1972. Since then a decreasing trend could be observed clearly until the last collected data point in this report. This decreasing trend showed agreement with expectation after the ban of producing and using PCBs in commercial products.

Table 5.2.12. Summary statistics of human milk concentrations for Σ PCBs, Σ indicator PCBs (6), CB118 and CB153 in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ ($N < DL/QL$))

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
Σ PCBs	Europe	ng/g lw	357	110	110	220	0.0010 - 2200	0	9, 20, 27, 33, 38, 46, 59
	North America	ng/g lw	36	130	140	120	0.0013 - 620	0	8, 9, 12, 27, 55
	Australasia	ng/g lw	20	48	0.0022	120	0.0013 - 480	0	1, 9, 27
	WEOG	ng/g lw	413	110	110	210	0.001 - 2200	0	
Σ Indicator PCBs (6)	Europe	ng/g lw	72	160	110	160	0.049 – 770	0	1, 9, 14, 30, 31, 45
	North America	ng/g lw	5	120	140	79	34 – 200	0	1, 55
	Australasia	ng/g lw	2	14	14	3.1	12 – 16	0	1
	WEOG	ng/g lw	79	160	110	160	0.049 - 770	0	
CB118	Europe	ng/g lw	111	9.5	6.2	11	0.00069 - 64	0	1, 2, 14, 20, 26, 29, 30, 31, 33, 38, 48, 61
	North America	ng/g lw	4	7.1	4.1	6.8	2.9 - 17	0	1, 12, 22, 60
	Australasia	ng/g lw	2	1.9	1.9	0.63	1.4 - 2.3	0	1
	WEOG	ng/g lw	117	9.3	5.9	11	0.00069 – 64	0	
CB153	Europe	ng/g lw	155	65	46	54	3.4 - 330	0	1, 2, 9, 14, 20, 26, 29, 30, 31, 33, 38, 42, 48, 59, 61
	North America	ng/g lw	16	31	22	28	1.1 - 85	0	1, 8, 9, 12, 13, 22, 60
	Australasia	ng/g lw	2	6.1	6.1	1.6	5.0 - 7.2	0	1
	WEOG	ng/g lw	173	61	43	53	1.1 - 330	0	

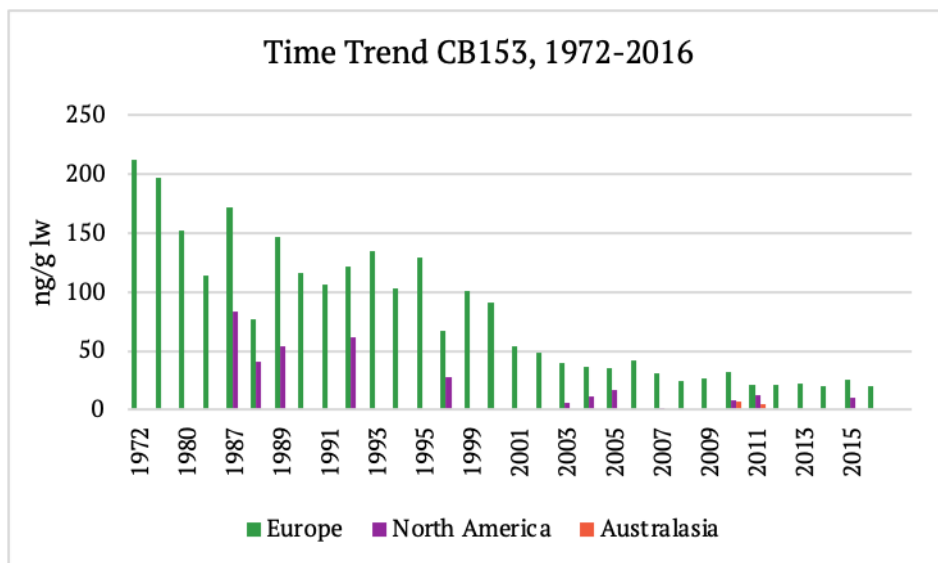


Figure 5.2.15. Time trend (1972 – 2016) for breast milk concentrations of CB153 in the WEOG region.

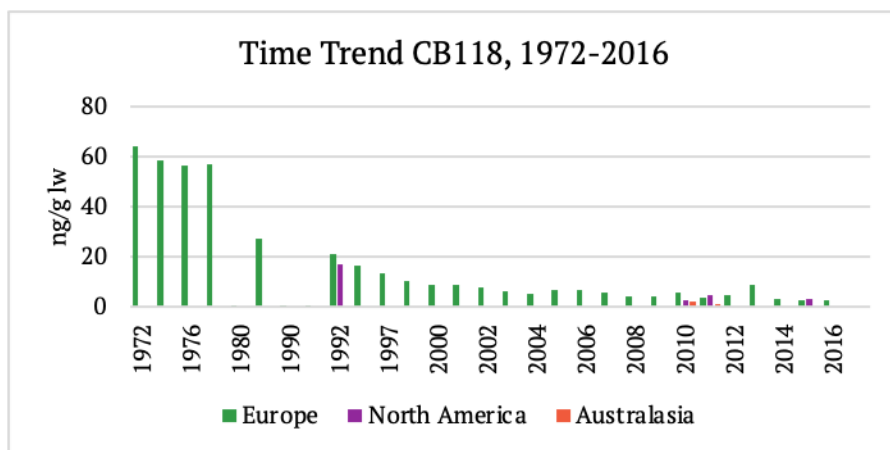


Figure 5.2.16. Time trend (1972 – 2016) for breast milk concentrations of CB118 in the WEOG region.

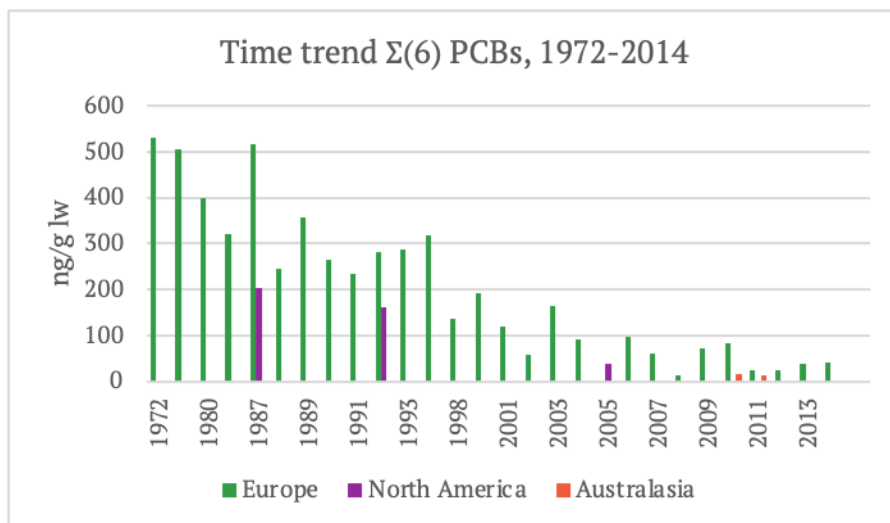


Figure 5.2.17. Time trend (1972 – 2014) for breast milk concentrations for $\Sigma(6)$ indicator PCBs in the WEOG region.

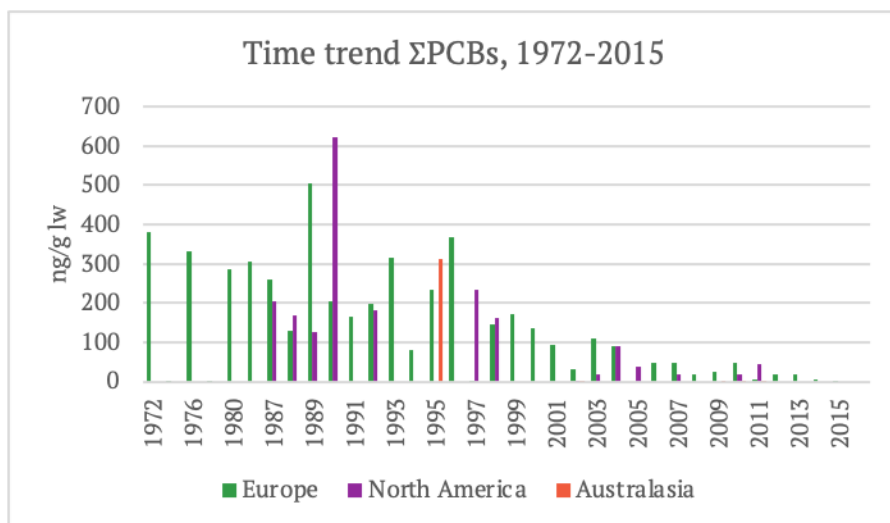


Figure 5.2.18. Time trend (1972 – 2014) for human milk concentrations for Σ PCBs in the WEOG region. The time trend is based on 410 reported PCB sums (containing 2 or more congeners), out of which 66 values originate from the Swedish national monitoring program, 253 values from scientific articles and 90 values from GMP UNEP.

Human blood

PCBs were measured in blood or serum samples in 11 countries for PCB 118 and in 12 countries for CB153 within WEOG regions since 1994 and 1991, respectively. To not exclude any

monitoring programmes, the PCB data are shown both on a lipid weight (Table 5.2.13) and wet weight (Table 5.2.14) basis of the samples separately.

CB118 in blood on a lipid weight basis is on a long-time (1994-2015) downwards trend (Figure 5.2.19). The years 2007, 2009 and 2013 within the European dataset seem outlying, but these years included solely the AMAP programme, with partly highly exposed groups such as Inuit and people from Greenland with mostly fish and or whale based diets, whereas some of the other years solely include Swedish monitoring data.

Table 5.2.13. Summary statistics of human blood (plasma or serum) concentrations on lipid weight basis for CB118 and CB153 in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
CB118	Europe	ng/g lw plasma or serum	45	31.7	22	33.7	3.9 -160	0	2, 62
	North America	ng/g lw plasma or serum	24	6.0	5.5	2.7	2.3 -13	0	62, 66, 67
	Australasia	ng/g lw serum	80	5.0	3.5	3.7	0.80 -15	0	68
	WEOG	ng/g lw plasma or serum	149	13.2	6.1	22.2	0.80 -160	0	
CB153	Europe	ng/g lw plasma or serum	86	162.9	99	180.0	12 -1000	0	2, 62
	North America	ng/g lw plasma or serum	36	45.6	27	39.1	8.2 -170	0	62, 66, 67
	WEOG	ng/g lw plasma or serum	122	128.3	72	161.5	8.2 -1000	0	

Table 5.2.14. Summary statistics of human blood (plasma or serum) concentrations on wet weight basis for CB118 and CB153 in the WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
CB118	Europe	ng/g ww plasma or serum	11	0.064	0.054	0.1	0.017 -0.18	0	2
	North America	ng/g ww plasma or serum	18	0.036	0.035	0.0	0.0098 -0.068	0	66, 67
	WEOG	ng/g ww plasma or serum	29	0.37	0.038	0.0	0.0098 -0.18	0	
CB153	Europe	ng/g ww plasma or serum	235	0.15	0.33	0.2	0.11 -1.3	5	2, 65, 69
	North America	ng/g ww plasma or serum	18	0.36	0.14	0.1	0.049 -0.28	0	66, 67
	WEOG	ng/g ww plasma or serum	253	0.33	0.32	0.2	0.049 -1.3	5	

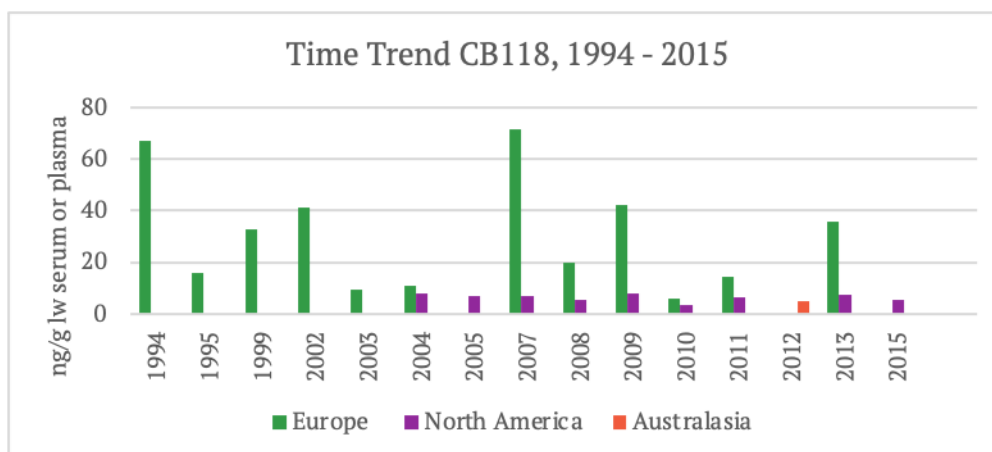


Figure 5.2.19. Time trend (1994 - 2015) of human blood (serum or plasma) concentrations **on lipid weight basis** for CB118 in the WEOG region during the sampling period 1994-2015.

CB153 related to the lipid weight in serum or plasma shows a declining trend since 1991 (Figure 5.2.20). The apparently outlying years (especially 2002 and 2007) in the dataset include several highly exposed groups, within AMAP and Sweden, such as inuits and fishermen, or heavy fish-eaters.

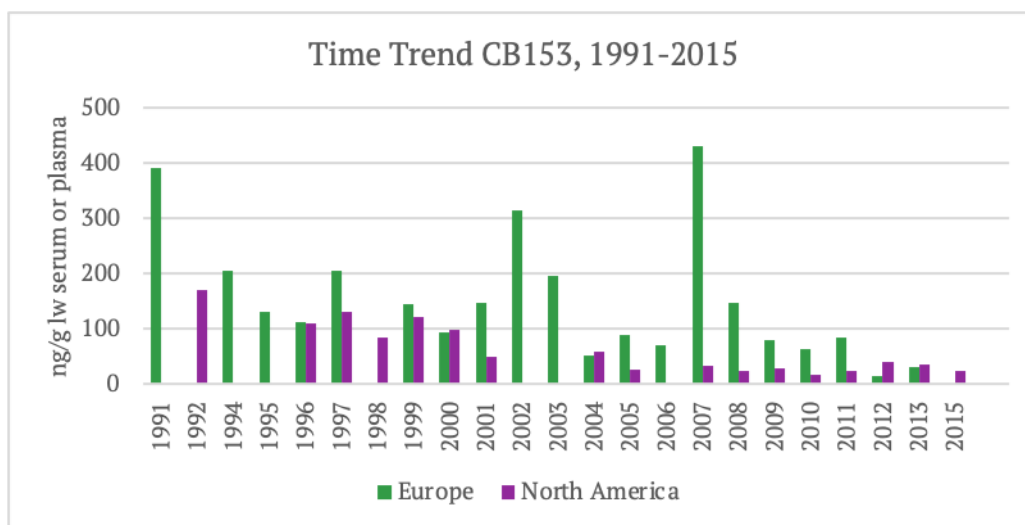


Figure 5.2.20. Time trend (1991 – 2015) of human blood (serum or plasma) CB153 concentrations **on lipid weight basis** in the WEOG region.

CB153 concentrations in serum or plasma based on wet-weight basis show a downwards trend (Figure 5.2.21). The higher concentrations in 2008-2011 are most likely influenced by the inclusion of partly exposed groups in Sweden in 2008 and 2011. Another reason might be the generally higher levels measured in the Spanish population (2009, 2010) including all ages, compared to the German monitoring programme investigating only students, which have a lower historical exposure due to the age difference.

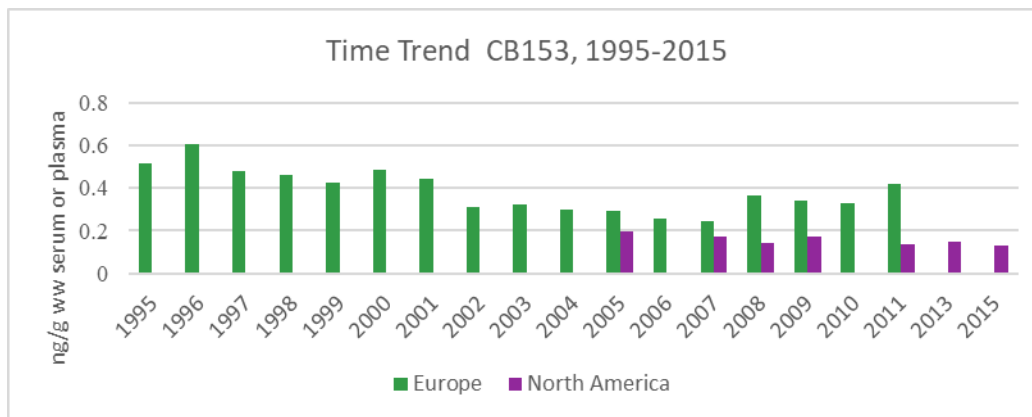


Figure 5.2.21. Time trend (1995 - 2015) of human blood (serum or plasma) CB153 concentrations **on a wet weight basis** in the WEOG region.

Chlordane

Human milk

Chlordane, including cis and trans configurations, have been measured in breast milk on at least one occasion in 20 countries within the WEOG region during the period 1999-2012 (Table 5.2.7). Approximately 79% of the reported median values for cis and trans chlordane were below the quantification limit and the highest reported value was 0.3 ng/g lw. The reason for the high number of non-detected values is due to the quick metabolism of these compounds inside human body which resulted in oxychlordane as metabolite. Moreover, cis- and trans-chlordane could also degrade to cis- and trans nonachlor. Therefore, oxychlordane, cis- and trans-nonachlor were used as indicators for the contamination of chlordane in breast milk. Out of the three indicators, oxychlordane was the most reported (N=47 median values) and it also had the highest average median value (8.9 ng/g lw) as compared to trans nonachlor (N=4 median values, 2.6 ng/g lw) and cis nonachlor (N=2 median values, 0.35 ng/g lw) (Table 5.2.15). It could also be seen that North America had the highest average median value (8.9 ng/g lw, N=12) followed by Australasia (4.1 ng/g lw, N=2) and Europe region (3.8 ng/g lw, N=33).

In general, there were decreasing trends of oxychlordane, cis- and trans nonachlor in breast milk in Europe region as well as in North America (Figure 5.2.22).

Table 5.2.15. Summary statistics of human milk concentrations for chlordane in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
Cis Chlordane	North America	ng/g lw	2.0	0.76	0.76	0.72	0.25 - 1.3	0	9
	Australasia	ng/g lw	2.0	0.25 ¹	0.25 ¹	0	0.25 ¹ - 0.25 ₁	2.0	1
	Europe	ng/g lw	13	0.23	0.25	0.072	0.001 - 0.3	11	1, 9, 20
	WEOG	ng/g lw	17	0.30	0.25	0.26	0.001 - 1.3	13	
Trans Chlordane	North America	ng/g lw	3.0	8.8	1.1	14	0.16 - 25	0	9
	Australasia	ng/g lw	2.0	0.25 ¹	0.25 ¹	0	0.25 ¹ - 0.25 ₁	2.0	1
	Europe	ng/g lw	17	0.17	0.25	0.11	0.0020 - 0.25	11	1, 9, 20
	WEOG	ng/g lw	22	1.4	0.25	5.3	0.002 - 25	13	
Sum Chlordane	Europe	ng/g lw	2.0	6.5	6.5	0.49	6.1 - 6.8	0	38
	WEOG	ng/g lw	2.0	6.5	6.5	0.49	6.1 - 6.8	0	
Oxychlordane	North America	ng/g lw	11	12	8.5	11	3.8 - 43	0	9
	Australasia	ng/g lw	2.0	4.1	4.1	5.0	0.56 - 7.6	0	1
	Europe	ng/g lw	77	3.7	3.3	1.8	1.0 - 11	1.0	1, 2, 9, 20, 26, 29, 31, 38, 48, 59, 61
	WEOG	ng/g lw	90	4.7	3.7	4.9	0.56 - 43	1.0	
Methoxychlor	Europe	ng/g lw	1	0.063	0.063		0.063 - 0.063	0	20
	WEOG	ng/g lw	1	0.063	0.063		0.063 - 0.063	0	
Cis Nonachlor	Europe	ng/g lw	2.0	0.35	0.35	0.049	0.31 - 0.38	0	38
	WEOG	ng/g lw	2.0	0.35	0.35	0.049	0.31 - 0.38	0	
Trans Nonachlor	Europe	ng/g lw	40	5.4	4.4	2.7	1.4 - 13	0	2, 29, 31, 38
	WEOG	ng/g lw	40	5.4	4.4	2.7	1.4 - 13	0	
Sum Chlordanes	Europe	ng/g lw	1	3.6	3.6		3.6 - 3.6	0	1
	WEOG	ng/g lw	1	3.6	3.6		3.6 - 3.6	0	

¹ Data below LOD or LOQ.

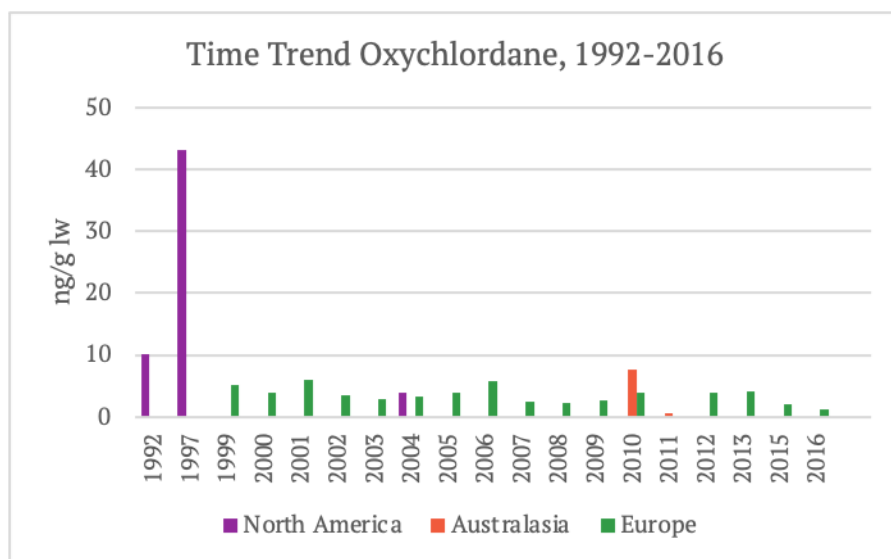


Figure 5.2.22. Time trend (1992 – 2016) of human milk oxychlordane concentrations in the WEOG region. The time trend is based on the average of 90 reported concentration values out of which 36 values originate from the Swedish monitoring program, 41 from scientific literature and 13 from WHO/UNEP.

Human blood

Oxychlordane has been measured in human serum and plasma on at least one occasion in 9 countries within the WEOG region (N=168). Lipid weight normalized concentrations of oxychlordane range between 0.20-488 ng/g lw serum or plasma within WEOG (N=141, Table 5.2.16). The highest average concentration (43 ng/g lw serum or plasma, N=51) was found in Europe. Oxychlordane shows a decreasing trend during the period 1992-2015 in the WEOG region (Figure 5.2.23). The high average value for Europe in 2007 includes mainly samples from Greenland.

Table 5.2.16. Summary statistics of human blood (serum or plasma) concentrations on lipid weight basis for Oxychlordane in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <LOQ	References
Oxychlordane	Europe	ng/g lw serum or plasma	51	43	18	93	0.20 -488	0	2, 62
	North America	ng/g lw serum or plasma	36	18	11	17	2.3 -77	0	62, 66, 67
	Australasia	ng/g lw serum or plasma	54	7.0	5.8	4.8	1.1 -19	0	68
	WEOG	ng/g lw serum or plasma	141	23	8.5	59	0.20 -488	0	

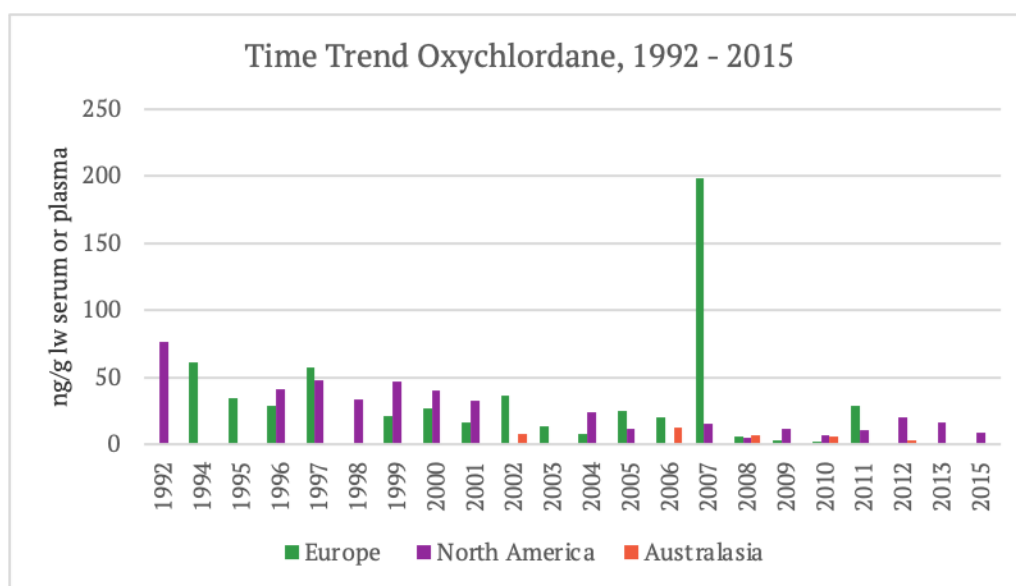


Figure 5.2.23. Time trend (1992 – 2015) of human blood oxychlordan concentrations in the WEOG region during the sampling period 1992-2015. This time trend is based on the average of 141 reported concentration values.

Wet weight normalized human blood concentration of Oxychlordan range between 0.0013–0.12 ng/ww serum or plasma within WEOG (N= 27, Table 5.2.17). This data is only based on data from two countries (Sweden and Canada) and no significant decreasing trend was observed for the period 2002-2011.

Table 5.2.17. Summary statistics of human blood (serum or plasma) concentrations on wet weight basis for Oxychlordan in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL).

Substance	Continent	Unit	N ²	Average	Median	SD	Range (min-max)	N <LOQ	References
Oxychlordan	Europe (Sweden)	ng/g ww serum or plasma	15	0.023	0.0076	0.036	0.0013 -0.12	0	2
	North America (Canada)	ng/g ww serum or plasma	12	0.032	0.029	0.018	0.0098 -0.059	0	66
	WEOG	ng/g ww serum or plasma	27	0.027	0.013	0.029	0.0013 -0.12	0	

Chlordecone

Chlordecone was included in the Stockholm convention in 2009. Just as in the previous global monitoring report (UNEP, 2015), no biomonitoring data of Chlordecone in human milk were found in the literature search or from the national biomonitoring programs.

Short-chained chlorinated paraffins (SCCPs)

Human milk

SCCPs were included in the Stockholm convention in 2017. Recent studies have examined concentrations and trends of short-, medium-, and long-chained chlorinated paraffins (SCCPs, MCCPs, and LCCPs, respectively) in breast milk in Asia and Europe and a temporal trend study in Sweden (Table 5.2.18). A consistent pattern in Asia and Europe was found with the highest concentrations found of SCCPs followed by MCCPs and LCCPs. In the Swedish study, no trend was found in concentrations of SCCPs or MCCPs between 1996 and 2009.

Table 5.2.18. Summary statistics of human milk concentrations for short-, medium-, and long-chained chlorinated paraffins (SCCPs, MCCPs, and LCCPs, respectively) in Europe and Asia (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	Reference
SCCPs	Europe	ng/g lw	5	81	68	83	10 – 180		63
MCCPs	Europe	ng/g lw	4	31	29	11	21 – 43		63
LCCPs	Europe	ng/g lw	3	4	4	1	3 - 4		63
SCCPs	Asia	ng/g lw	12	201	35	280	1 – 733	3	63
MCCPs	Asia	ng/g lw	7	66	63	27	36 – 121		63
LCCPs	Asia	ng/g lw	3	10	11	5	5 - 15		63
SCCPs	Europe (Sweden)	ng/g lw	13	107	112	31	45 – 157		64
MCCPs	Europe (Sweden)	ng/g lw	13	13	15	10	1 - 30		64

Human blood

No biomonitoring data of SCCPs in human blood were found in the literature search or in the national biomonitoring programs.

Dichloro-diphenyl-trichloroethane (DDT)

Human milk

DDTs, including ortho-para and para-para configurations of DDD, DDE and DDT, has been monitored in breast milk since 1972 within WEOG (Table 5.2.7). The p,p'-DDT is the major compound in the commercial product and p,p'-DDE, the main metabolite of DDT, have been measured intensively with an average of median levels at 5.7 ng/g lw (N=156) and 107 ng/g lw (N=170), respectively (Table 5.2.19). The highest median level of p,p'-DDE in breast milk was found in Australasia (280 ng/g lw, N=13), followed by North America (145 ng/g lw, N=23) and Europe (82 ng/g lw, N=134).

In general, there were clear trends of decreasing p,p'-DDE (Figure 5.2.24) and p,p'-DDT (Figure 5.2.25) in breast milk during period 1972-2014 for Europe region, except for a few unexpected occasions where sum p,p'-DDE level reached up to 2500 ng/g lw. Similar trends could be observed for p,p'-DDT and p,p'-DDE for combination of median values for all regions. However, no clear trend could be observed for DDT complex in North America as the average median levels for p,p'-DDE and p,p'-DDT varied during the almost two decades when it was measured (1988-2007) (Figure 5.2.24 and Figure 5.2.25).

Table 5.2.19. Summary statistics of human milk concentrations for DDTs in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
p,p'-DDE	North America	ng/g lw	23	200	150	190	32 - 960	0	9, 22
	Australasia	ng/g lw	13	400	280	280	52 - 1000	0	1, 9, 23
	Europe	ng/g lw	130	270	82	470	3.6 - 2500	0	1, 2, 9, 14, 20, 26, 29, 30, 31, 33, 38, 42, 48, 49, 59, 61
	WEOG	ng/g lw	170	270	120	430	3.6 - 2500	0	
p,p'-DDT	North America	ng/g lw	16	15	110	16	0.60 - 68	1	9, 22
	Australasia	ng/g lw	13	29	15	62	4.3 - 230	0	1, 9, 23
	Europe	ng/g lw	130	37	7.7	98	1 - 690	1	1, 2, 9, 14, 20, 26, 29, 30, 31, 33, 38, 48, 49, 59, 61
	WEOG	ng/g lw	160	34	5.0	91	0.6 - 690	2	
Sum DDT	North America	ng/g lw	4.0	88	52	40	41 - 140	0	9
	Australasia	ng/g lw	2.0	500	52	74	450 - 550	0	1
	Europe	ng/g lw	39	480	140	750	0.92 - 2700	0	1, 9, 20, 30, 33, 38, 50, 59
	WEOG	ng/g lw	45	450	500	710	0.92 - 2700	0	

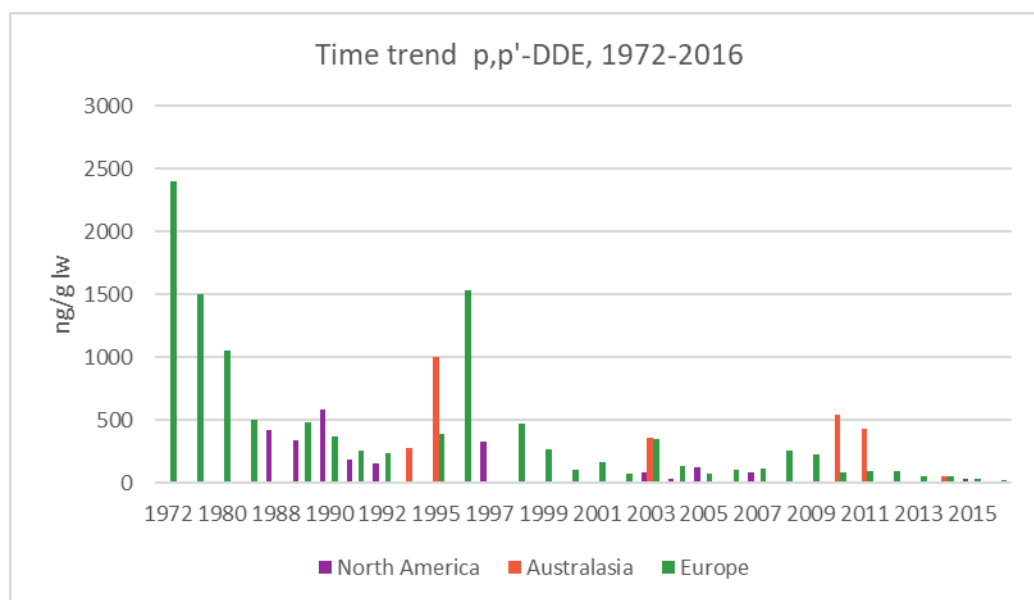


Figure 5.2.24. Time trend (1972 – 2016) for human milk concentrations of p,p'-DDE [ng/g lw] in the WEOG region. Time trend is based on 170 reported mean/median values, out of which 45 values originate from the Swedish monitoring program, 112 values from scientific articles and 13 from UNEP/WHO.

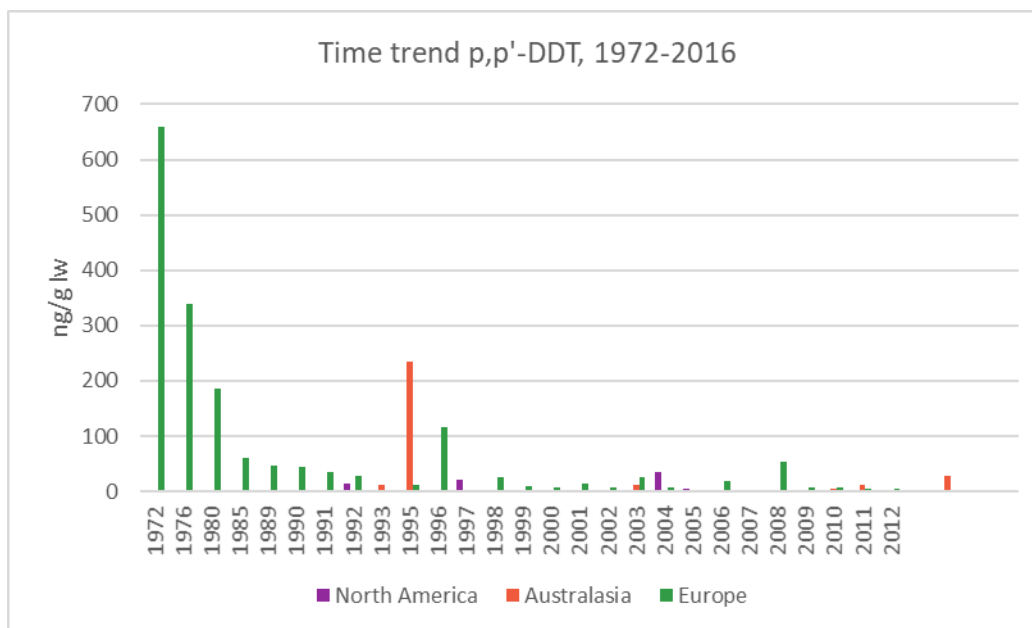


Figure 5.2.25. Time trend (1972 – 2016) for human milk concentrations of p,p'-DDT [ng/g lw] in the WEOG region. Time trend is based on 155 reported mean/median values, where 45 values are from Swedish monitoring program, 97 values from scientific articles and 13 values from UNEP/WHO.

Human blood

p,p'-DDT has been measured in human serum and plasma on at least one occasion in nine countries within the WEOG region (N=157). Lipid weight normalized concentrations of p,p'-DDT range between 0.059-40 ng/g lw serum or plasma within WEOG (N=125, Table 5.2.20). The highest average concentration was found in Europe (13 ng/g lw serum or plasma, N=23), followed by North America (8.6 ng/g lw serum or plasma, N=23) and Australasia (6.2 ng/g lw serum or plasma, N=79). The time trend shows a decrease during the period 1992-2015 in the WEOG region (Figure 5.2.26). The high average value for Europe in 2007 and 2013 includes mainly samples from Greenland and Russia, respectively.

Table 5.2.20. Summary statistics of human blood (serum or plasma) concentrations on lipid weight basis for p,p'-DDT in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
p,p'-DDT	Europe	ng/g lw serum or plasma	23	13	7.7	13	0.059 -40	0	62
	North America	ng/g lw serum or plasma	23	8.6	6.7	5.5	2.5 -26	0	62, 67
	Australasia	ng/g lw serum or plasma	79	6.2	4.5	4.6	2.4 -26	0	68
	WEOG	ng/g lw serum or plasma	125	7.9	5.1	7.4	0.059 -40	0	

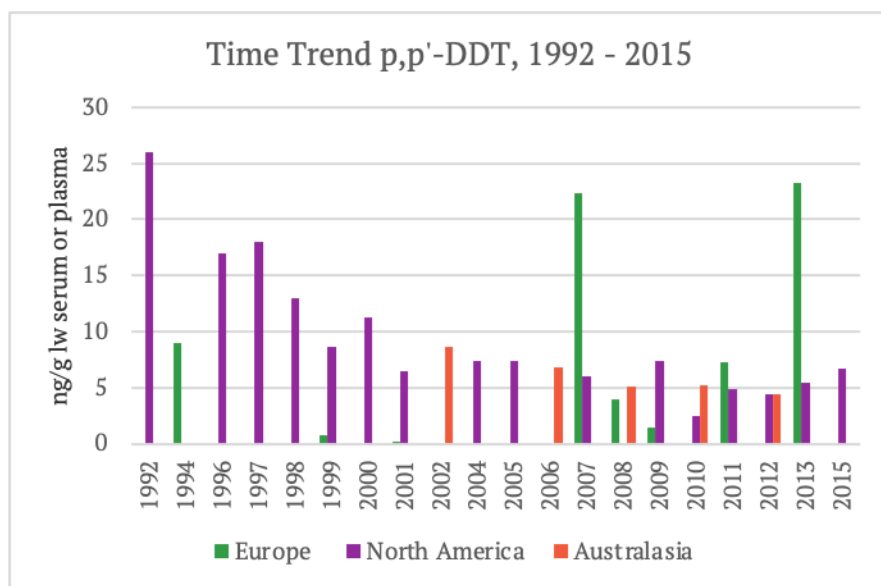


Figure 5.2.26. Time trend (1992 – 2015) of human blood (serum or plasma) p,p'-DDT concentrations in the WEOG region. This time trend is based on the average of 125 reported mean/median values.

Wet weight normalized concentrations of p,p'-DDT were only available from Spain (sampled in year 2009 and 2010) where the average concentration was 0.24 ng/g ww serum (N=32). However, the ratio of measured data above LOQ in this data set ranged from 0-50%.

p,p'-DDE has been measured in human serum and plasma on at least one occasion in ten countries within the WEOG region (N=203). Lipid weight normalized concentrations of p,p'-DDE range between 0.78-1847 ng/g lw serum or plasma within WEOG (N=191, Table 5.2.21).

The highest average concentration was found in Australasia (420 ng/g lw serum or plasma, N=84), followed by Europe (329 ng/g lw serum or plasma, N=71) and North America (224 ng/g lw serum or plasma, N=36). The time trend shows a decrease of p,p'-DDE during the period 1992-2015 in the WEOG region (Figure 5.2.27).

Table 5.2.21. Summary statistics of human blood (serum or plasma) concentrations on lipid weight basis for p,p'-DDE in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
p,p'-DDE	Europe	ng/g lw serum or plasma	71	329	201	369	0.78 -1847	0	62
	North America	ng/g lw serum or plasma	36	224	200	137	56 -640	0	62, 66, 67
	Australasia	ng/g lw serum or plasma	84	420	250	394	65 -1820	0	68
	WEOG	ng/g lw serum or plasma	191	349	230	356	0.78 -1847	0	

Figure 5.2.27. Time trend (1992 – 2015) of human blood (serum or plasma) p,p'-DDE concentrations in the WEOG region. This time trend is based on the average of 191 reported mean/median values.

Wet weight normalized concentrations of p,p'-DDE were only available from Canada (sampled in year 2008) where the average concentration was 0.90 ng/g ww plasma (N=12, Table 5.2.22).

Table 5.2.22. Summary statistics of human blood (plasma) concentrations on wet weight basis for p,p'-DDE in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL))

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
p,p'-DDE	North America (Canada)	ng/g ww plasma	12	0.90	0.79	0.46	0.41 -1.8	0	66

Endosulfan

Human milk

Endosulfan and endosulfan sulphate were measured in 14 countries within the WEOG region. Beta endosulfan was the major compound that could be found in breast milk with 71 ng/g lw (N=16) as compared to 5 ng/g lw (N=16) for alpha endosulfan (Table 5.2.23). Most of the measurements were from Europe with 20 median values for alpha endosulfan and 14 median values for beta endosulfan while the two median values in Australasia region were below LOD/LOQ. For endosulfan sulphate, 13 out of 14 median values were below LOD/LOQ. No trend could be observed for both isomers of endosulfan or endosulfan sulphate during period 1999-2012 within the WEOG region.

Table 5.2.23. Summary statistics of breast milk concentrations for endosulfan in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
Alpha Endosulfan	Europe	ng/g lw	20	5.5	0.25	12	0.028 - 50	11	1, 9, 18, 20
	Australasia	ng/g lw	2	0.25	0.25	0.00	0.25 - 0.25	2	1
	WEOG	ng/g lw	22	5.0	0.25	11	0.028 - 50	13	
Beta Endosulfan	Europe	ng/g lw	14	81	0.25	260	0.17 - 950	11	1, 9, 20
	Australasia	ng/g lw	2	0.25	0.25	0.00	0.25 - 0.25	2	1
	WEOG	ng/g lw	16	71	0.25	240	0.17 - 950	13	
Endosulfan sulphate	Europe	ng/g lw	12	11	0.25	36	0.25 - 125	11	1, 9
	Australasia	ng/g lw	2	0.25	0.25	0.00	0.25 - 0.25	2	1
	WEOG	ng/g lw	14	9.2	0.25	33	0.25 - 125	13	

Human blood

No biomonitoring data on endosulfan in blood were collected for this report.

Hexabromocyclododecane (HBCD)

Human milk

The commercial mixture of HBCD has been used since the 1980s and it has three major diastereomers; alpha-, beta-, and gamma- HBCD. In 2013, HBCD was listed in Stockholm convention annex A and thereafter the production of HBCD has decreased. The highest average of median values of sum HBCD is found in Australasia (11 ng/g lw, N=9) while Europe has the lowest values (0.37 ng/g lw, N=59) (Table 5.2.24). Only one study was found measuring HBCD in breast milk in North America and that was from 2004.

A trend of increasing HBCD concentrations in breast milk could be observed for the period 1980-2014 in 13 countries, where the highest average of median value of HBCD in milk was in year 2007 (Figure 5.2.28). After 2011 a decreasing trend can be observed (Figure 5.2.28).

Table 5.2.24. Summary statistics of human milk concentrations for hexabromocyclododecane (HBCD) in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
Alpha HBCD	Europe	ng/g lw	8	1.8	1.0	1.8	0.31 - 4.4	0	1, 9, 10, 19, 29
	North America	ng/g lw	1	0.50	0.50		0.5 - 0.5	0	7, 54
	Australasia	ng/g lw	13	3.0	2.8	2.7	0.17 - 7.8	4	1, 39
	WEOG	ng/g lw	22	2.4	1.2	2.4	0.17 - 7.8	4	
Beta HBCD	Europe	ng/g lw	4	0.24	0.25	0.15	0.045 - 0.4	2	1, 9, 10, 19
	North America	ng/g lw	1	0.50	0.080		0.50 - 0.5	0	7, 54
	Australasia	ng/g lw	13	1.1	0.50	0.96	0.050 - 3.6	12	1, 39
	WEOG	ng/g lw	18	0.91	0.50	0.90	0.045 - 3.6	14	
Gamma HBCD	Europe	ng/g lw	5	4.9	0.50	10	0.19 - 23	1	1, 9, 10, 19
	North America	ng/g lw	1	0.50	0.20		0.5 - 0.5	0	7, 54
	Australasia	ng/g lw	13	2.5	2.2	2.6	0.11 - 6.8	4	1, 39
	WEOG	ng/g lw	19	3.1	0.50	5.3	0.11 - 23	5	
Sum HBCD	Europe	ng/g lw	59	0.99	0.37	3.5	0.024 - 27	12	2, 9, 10, 17, 42
	North America	ng/g lw	1	0.70	0.86		0.7 - 0.7	0	7, 54
	Australasia	ng/g lw	9	10	11	5.2	2.5 - 19	0	39
	WEOG	ng/g lw	69	2.2	0.39	4.9	0.024 - 27	12	

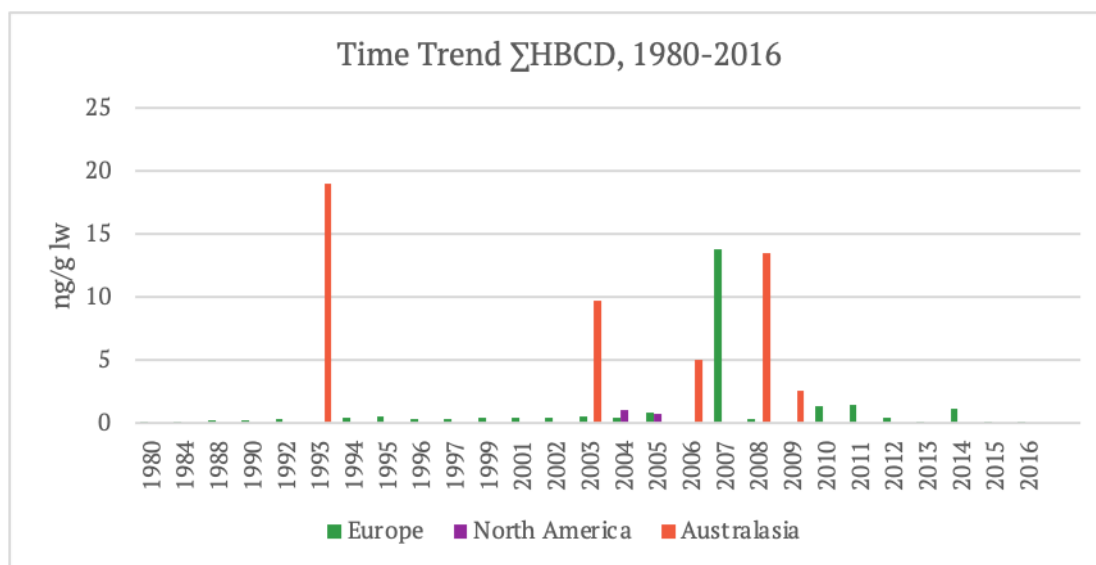


Figure 5.2.28. Time trend (1980 - 2016) for human milk Σ HBCD concentrations in the WEOG region. Time trend is based on 69 reported mean/median values, out of which 45 values originate from Swedish monitoring program and 24 values from scientific articles.

Human blood

HBCD was reported in human blood only in Sweden in three years; 2011 (men and women), 2016 and 2017 (first-time mothers) (Table 5.2.25).

Table 5.2.25. Summary statistics of human blood (plasma or serum) concentrations on lipid weight basis for HBCD in Europe region (Sweden) (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ ($N < DL/QL$)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
HBCD	Europe (Sweden) 2011	ng/g lw plasma or serum	6	0.7	0.49	0.6	0.33 -1.9	6	2
HBCD	Europe (Sweden) 2016, 2017	ng/g ww plasma or serum	2	0.00082	0.00082	0.00016	0.00071 -0.00093	2	2

Hexachlorobenzene (HCB)

Human milk

Since the introduction of HCB as a pesticide in 1945, it was used widely in agriculture. HCB has been measured in breast milk in 21 countries within the WEOG during the period 1972-2013. The average median level of HCB was highest in Australasia (98 ng/g lw, N=15), followed by Europe (33 ng/g lw, N=123) and North America (15 ng/g lw, N=22) (Table 5.2.26). All levels of HCB in measured breast milk samples were above the quantification limit (QL). There were three studies in Australia, Germany and Spain that showed exceptionally high levels of HCB in breast milk. This was because these samples were collected nearby either heavy industrial or agricultural areas. There is a decreasing trend of average of median value for HCB in Europe as well as in WEOG countries together during period 1972-2013 (Figure 5.2.29).

Table 5.2.26. Summary statistics of human milk concentrations for hexachlorobenzene (HCB) in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
HCB	Europe	ng/g lw	123	33	11	100	2.8 - 910	0	1, 2, 9, 14, 20, 26, 29, 31, 33, 38, 42, 48, 49, 52, 59, 61
	North America	ng/g lw	22	15	9.6	22	1.6 – 110	0	9
	Australasia	ng/g lw	15	98	19	160	5.1 - 460	0	1, 9
	WEOG	ng/g lw	160	37	11	100	1.6 - 910	0	

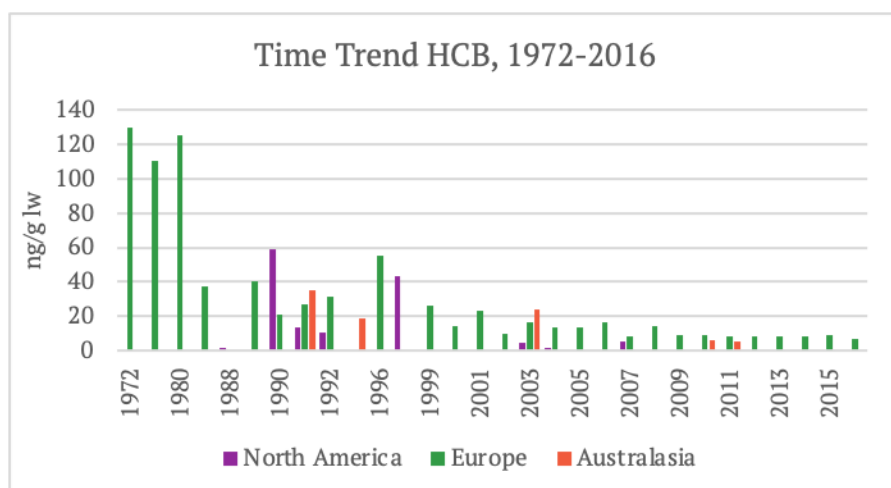


Figure 5.2.29. Time trend (1972-2016) for human milk Σ HCB concentrations in the WEOG region. Time trend is based on 366 reported mean/median values, where 184 values originate from the Swedish monitoring program, 156 values from scientific literature and 26 values from UNEP/WHO.

Human blood

HCB has been measured in human serum and plasma on at least one occasion in 12 countries within the WEOG region (N=425). Lipid weight normalized concentrations of HCB range between 1.7-391 ng/g lw serum or plasma within WEOG (N=169, Table 5.2.27). The highest average concentration (60 ng/g lw serum or plasma, N=63) was found in Europe. A decreasing trend was observed for HCB for the period 1991-2015 in the WEOG region (Figure 5.2.30). The high average values for Europe in 2007 and 2009 represent mainly samples from Greenland and Russia, respectively.

Table 5.2.27. Summary statistics of human blood (serum or plasma) concentrations on lipid weight basis for HCB in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
HCB	Europe	ng/g lw serum or plasma	63	60	36.7	72	1.7 -391	0	2, 62
	North America	ng/g lw serum or plasma	24	26	21.0	19	8.7 -95	0	62, 67
	Australasia	ng/g lw serum or plasma	82	26	8.6	39	2.9 -252	0	68
	WEOG	ng/g lw serum or plasma	169	39	18.9	55	1.7 -391	0	

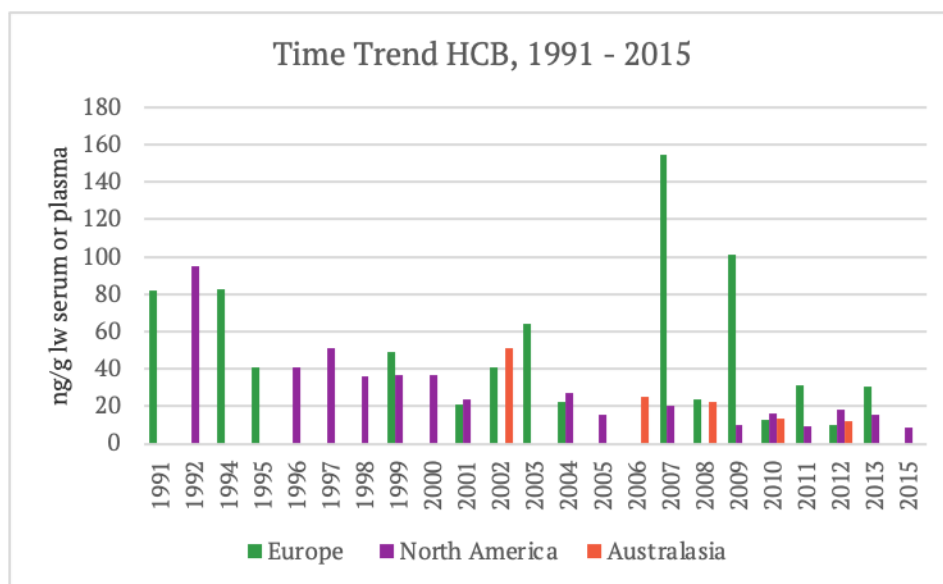


Figure 5.2.30. Time trend (1991 – 2015) of human blood (serum or plasma) HCB concentrations on lipid weight basis in the WEOG region. This time trend is based on the average of 169 reported mean/median values.

Human blood (serum or plasma) concentrations of HCB on wet weight basis range between 0.033-3.4 ng/ww within Europe (N= 256) Table 5.2.28). This data is only based on three countries (Germany, Spain and Sweden) but shows a decreasing trend for HCB during the period 1985-2009 (Figure 5.2.31).

Table 5.2.28. Summary statistics of human blood (serum or plasma) concentrations on wet weight basis for HCB in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
HCB	Europe	ng/g ww serum or plasma	256	0.30	0.16	0.46	0.033 -3.4	27	2, 65, 69

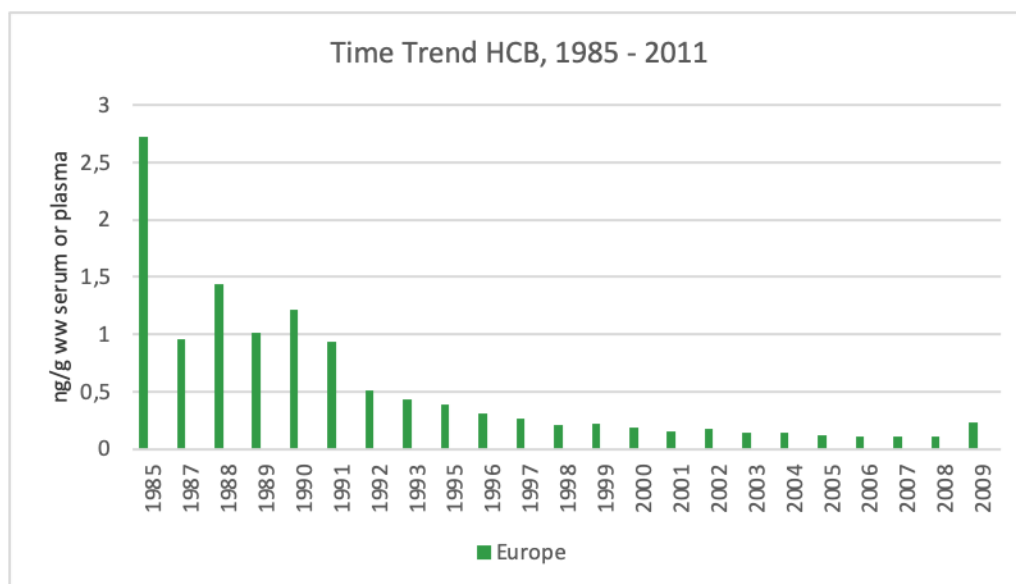


Figure 5.2.31. Time trend (1985 – 2009) of human blood (serum or plasma) HCB concentrations on wet weight basis in the WEOG region. This time trend is based on the average of 256 reported mean/median values.

Hexachlorobutadiene (HCBD)

HCBD was included in the Stockholm convention in 2015. No biomonitoring data of HCBD in breast milk or human blood was found in the literature search or in the national biomonitoring programs.

Hexachlorocyclohexanes (HCH)

Human milk

The technical mixture of HCH includes three main isomers; alpha, beta and gamma hexachlorocyclohexane, which were listed in Stockholm convention in 2009. These three isomers have been measured in breast milk in 21 countries within the WEOG during the period 2001-2016. Beta HCH is more persistent in the environment than alpha and gamma isomers. Beta has therefore a higher potential to bioaccumulate and biomagnify in food chain and transfer into breast milk over time. As a result, beta HCH has the highest average of median values in breast milk with 9.4 ng/g lw (N=137), followed by gamma HCH (0.54, N=65) and alpha HCH (0.25, N=58) in WEOG (Table 5.2.29). Within WEOG, Australasia has the highest average of median values for all three isomers with 15 ng/g lw for alpha HCH, 134 ng/g lw for beta HCH, and 26 ng/g lw for gamma HCH. North America has the lowest average of median values for

alpha, beta and gamma HCH with 1.1 ng/g lw (N=3), 13 ng/g lw (N=13), and 2.3 (N=13), respectively.

Table 5.2.29. Summary statistics of human milk concentrations for hexachlorocyclohexane (HCH) in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
Alpha HCH	Europe	ng/g lw	41	6.1	0.26	16	0.12 - 67	23	1, 2, 9, 20, 33, 38, 59
	North America	ng/g lw	3	1.1	1.4	0.68	0.31 - 1.6	0	9
	Australasia	ng/g lw	14	15	0.14	30	0.030 - 85	2	1, 9
	WEOG	ng/g lw	58	8.1	0.25	20	0.030 - 85	25	
Beta HCH	Europe	ng/g lw	107	28	8.0	71	1.7 – 420	1	1, 2, 9, 20, 26, 29, 30, 31, 33, 38, 48, 49, 52, 59, 61
	North America	ng/g lw	13	13	14	4.5	4.4 - 21	0	9
	Australasia	ng/g lw	17	130	21	200	5.0 - 660	1	1, 9, 23
	WEOG	ng/g lw	137	40	9.4	101	1.7 - 660	2	
Gamma HCH	Europe	ng/g lw	48	3.6	0.61	6.0	0.12 – 23	18	1, 2, 9, 14, 20, 33, 38, 49, 59
	North America	ng/g lw	3	2.3	1.0	2.5	0.66 - 5.1	0	9
	Australasia	ng/g lw	14	26	0.27	52	0.09 – 130	2	1, 9
	WEOG	ng/g lw	65	8.5	0.54	26	0.09 – 130	20	
Sum HCH	Europe	ng/g lw	14	270	170	250	32 - 750	0	9, 20, 33, 38
	North America	ng/g lw	1	13	13		13 – 13	0	9
	WEOG	ng/g lw	15	260	160	250	13 - 750	0	

Since beta HCH has the highest potential to be transferred into breast milk, the temporal trend of beta HCH in breast milk can be used as an indicator for the exposure of HCH. There is a decreasing trend of beta HCH in breast milk during the period 2001-2016 (Figure 5.2.32). The decrease started before HCH was listed in Stockholm convention in 2009.

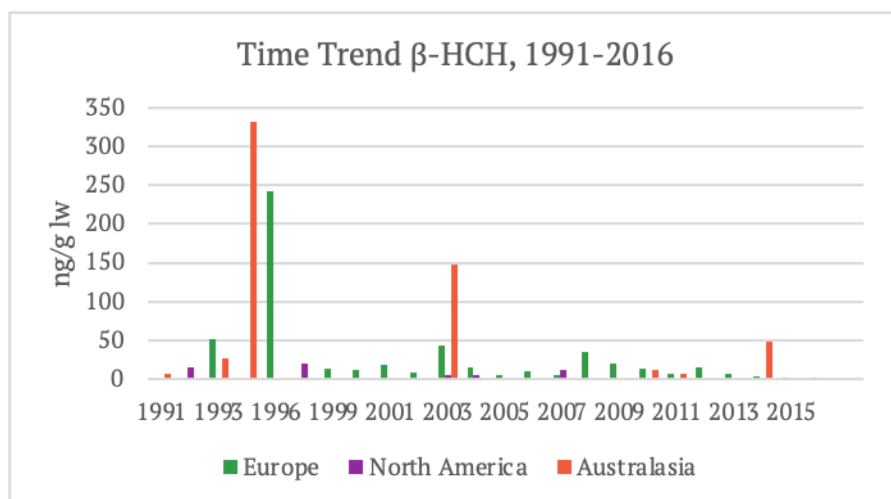


Figure 5.2.32. Time trend (1991 - 2016) for human milk beta HCH concentrations in the WEOG region. Time trend is based on 136 reported mean/median values, where 45 samples are from the Swedish monitoring programme, 78 values from scientific literature and 13 values from UNEP/WHO.

Human blood

Beta-HCH has been measured in human serum and plasma on at least one occasion in ten countries within the WEOG region (N=200). Lipid weight normalized concentrations of beta-HCH range between 1.1-71 ng/g lw serum or plasma within WEOG (N= 150, Table 5.2.30). The highest average concentration (15 ng/g lw serum or plasma, N=35) was found in Australasia, followed by Europe (13 ng/g lw serum or plasma) and North America (7.0 ng/g lw serum or plasma). No time trend was observed for beta-HCH during the period 1992-2015 in the WEOG region (Figure 5.2.33).

Table 5.2.30. Summary statistics of human blood (serum or plasma) concentrations on lipid weight basis for beta-HCH in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
Beta-HCH	Europe	ng/g lw serum or plasma	35	13	5.9	16	1.2 -56	0	62
	North America	ng/g lw serum or plasma	35	7.0	5.5	4.4	1.6 -19	0	62, 66, 67
	Australasia	ng/g lw serum or plasma	80	15	8.7	16	1.1 -71	0	68
	WEOG	ng/g lw serum or plasma	150	13	6.9	14	1.1 -71	0	

¹Mean value of concentrations in final dataset.

²Number of reported mean/median values in dataset

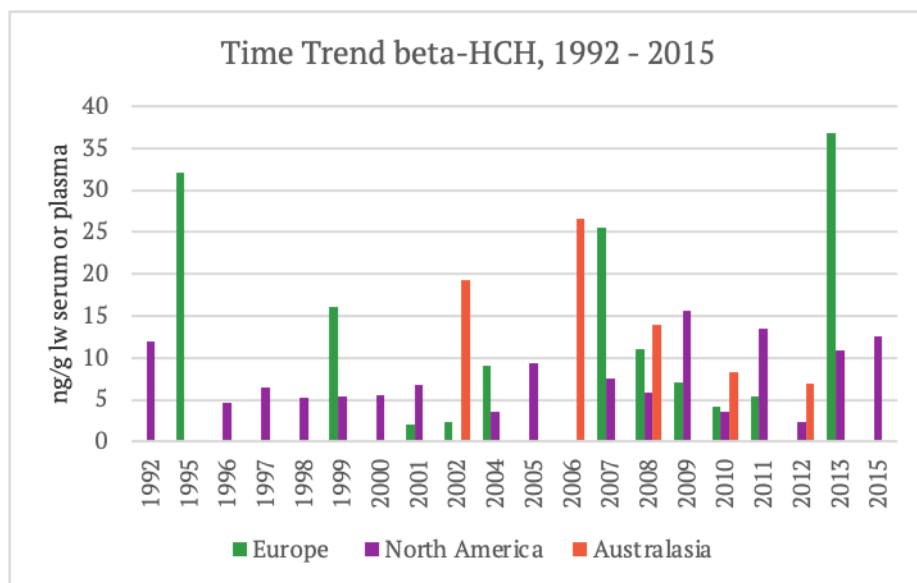


Figure 5.2.33. Time trend (1992 – 2015) of human blood (serum or plasma) beta-HCH concentrations in the WEOG region. This time trend is based on the average of 150 reported mean/median values.

For wet weight normalized data the concentration of beta-HCH ranged between 0.0096-0.43 ng/ww serum or plasma within WEOG (N= 50, Table 5.2.31). This data is only based on three countries (Canada, Spain and Sweden) and no time trend was observed for the period 2008-2011.

Table 5.2.31. Summary statistics of human blood (serum or plasma) concentrations on wet weight basis for beta-HCH in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
Beta-HCH	Europe	ng/g ww serum or plasma	38	0.15	0.13	0.082	0.0096 -0.43	32	2, 65
	North America	ng/g ww serum or plasma	12	0.037	0.034	0.016	0.020 -0.068	0	66
	WEOG	ng/g ww serum or plasma	50	0.12	0.12	0.086	0.0096 -0.43	32	

Heptachlor

Human milk

Heptachlor and its degradation product heptachlor epoxide were measured in 19 countries within the WEOG during the period 1988-2013 (Table 5.2.7). In Australasia the average level of Heptachlor is 6.7 ng/g lw (N=17) as compared to 3.3 ng/g lw (N=14) in Europe (Table 5.2.32). For heptachlor epoxide, the highest average of median values can also be seen in Australasia with 15 ng/g lw (N=15), followed by Europe with 8.8 ng/g lw (N=19) and North America with 2.1 ng/g lw (N=10). There was no clear trend of reported mean and median values for heptachlor during period 1988-2013.

Table 5.2.32. Summary statistics of human milk concentrations for heptachlor concentrations in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
Cis Heptachlor epoxide	Europe	ng/g lw	16	2.9	2.4	1.9	0.6 - 7.3	0	1, 9, 20, 29
	Australasia	ng/g lw	2	2.1	2.1	2.6	0.25 - 3.9667	1	1
	WEOG	ng/g lw	18	2.8	2.4	1.9	0.25 - 7.3	1	
Heptachlor	Europe	ng/g lw	14	3.3	0.25	7.7	0.001 - 22	10	1, 9, 20
	Australasia	ng/g lw	17	6.7	3.0	7.6	0.25 - 24	17	1, 9
	WEOG	ng/g lw	31	5.1	0.9	7.7	0.001 - 24	27	
Heptachlor epoxide	Europe	ng/g lw	19	8.8	2.7	16	0.11 - 61	1	1, 9, 59
	North America	ng/g lw	10	2.1	2.0	0.79	0.75 - 3.25	0	9
	Australasia	ng/g lw	15	15	7.3	21	0.25 - 78	1	1, 9
	WEOG	ng/g lw	44	9.5	3.0	17	0.11 - 78	2	
Trans Heptachlor epoxide	Europe	ng/g lw	9	0.22	0.25	0.082	0.0050 - 0.25	8	1, 20
	Australasia	ng/g lw	2	0.25	0.25	0.00	0.25 - 0.25	2	1
	WEOG	ng/g lw	11	0.23	0.25	0.074	0.005 - 0.25	10	

Human Blood

Heptachlor was reported in human blood in 2 years (2009 and 2010) in Spain with all measured data below LOQ (65).

Mirex

Human milk

The insecticide Mirex is one of the initial 12 POPs included in the Stockholm Convention when it came into force in 2004. In the previous report, Mirex was reported to be below the limit of detection in all reporting countries (UNEP, 2015). In this survey, 44 reported mean or median values were found out of which 10 values were below the limit of detection or quantification. Mirex has been analysed in 16 countries with samples from 1988 to 2013. Most samples are from North America and Europe, where North American values are from the 1980's and 1990's. The highest average of median values was found in North America (2.5 ng/g lw, N=21), followed by Europe with 0.5 ng/g lw (N=19) and Australasia with 0.22 ng/g lw (N=4) (Table 5.2.33). There was no clear trend of reported mean and median values for Mirex during the period 1988 to 2013.

Table 5.2.33. Summary statistics of human milk concentrations for Mirex in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
Mirex	Europe	ng/g lw	19	0.50	0.25	0.61	0.073 - 2.3	8	1, 9, 14, 20, 49
	North America	ng/g lw	21	2.5	1.8	2.9	0.75 - 14	0	9
	Australasia	ng/g lw	4	0.22	0.23	0.034	0.18 - 0.25	2	1, 9
	WEOG	ng/g lw	44	1.4	0.8	2.2	0.073 - 14	10	

Human blood

Mirex has been measured in human serum and plasma on at least one occasion in 6 countries within the WEOG region (N=63). Lipid weight normalized concentrations of Mirex range between 0.078-91 ng/g lw serum or plasma within WEOG (N=57, Table 5.2.34). The highest average concentration (18 ng/g lw serum or plasma, N=15) was found in Europe, followed by North America (5.6 ng/g lw serum or plasma, N=26) and Australasia (1.1 ng/g lw serum or plasma, N=16). Mirex shows decreasing trend for the period 1992-2015 in the WEOG region

(Figure 5.2.34). The high average value for Europe in 2007 includes mainly samples from Greenland.

Table 5.2.34. Summary statistics of human blood (serum or plasma) concentrations on lipid weight basis for Mirex in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
Mirex	Europe	ng/g lw serum or plasma	15	18	5.8	27	0.078 -91	0	62
	North America	ng/g lw serum or plasma	26	5.6	3.3	4.9	2.0 -24	0	62, 67
	Australasia	ng/g lw serum or plasma	16	1.1	1.1	0.094	0.95 -1,3	0	68
	WEOG	ng/g lw serum or plasma	57	7.5	3.0	15	0.078 -91	0	

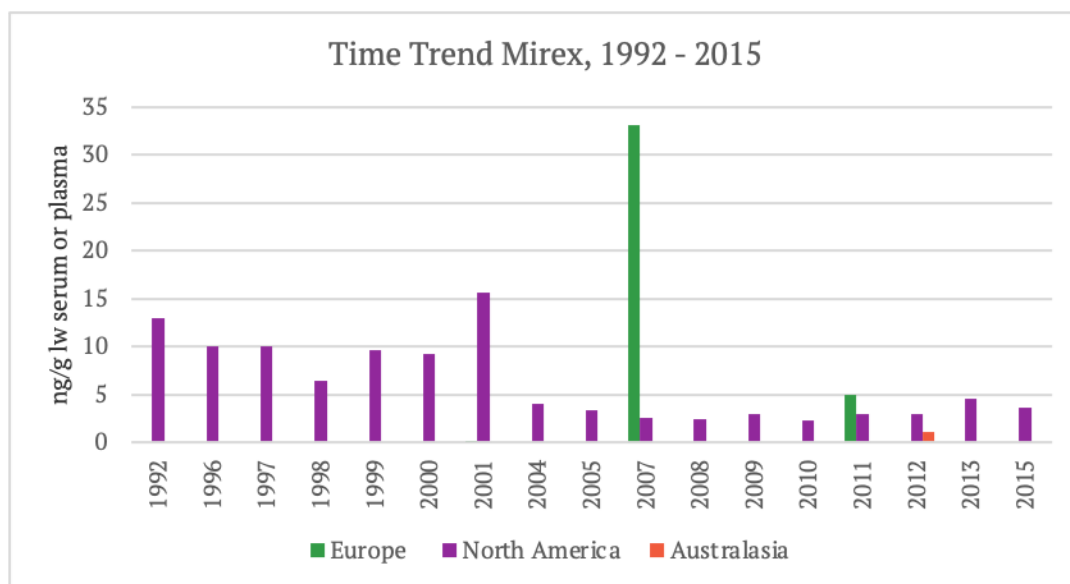


Figure 5.2.34. Time trend (1992 - 2015) of human blood (serum or plasma) Mirex concentrations in the WEOG region. This time trend is based on the average of 57 reported mean/median values.

Wet weight normalized concentrations of Mirex were only available from Canada (sampled in year 2008) where the average concentration was 0.015 ng/g ww plasma (N=6, Table 5.2.35).

Table 5.2.35. Summary statistics of breast milk wet weight concentrations for Mirex in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
Mirex	North America (Canada)	ng/g ww plasma	6	0.015	0.015	0.0053	0.0098 -0.020	0	66

Polychlorinated naphthalenes PCNs

Human milk

No biomonitoring data of PCNs in human milk were found in the literature search or in the national biomonitoring programs.

Human blood

PCNs was included in the Stockholm convention in 2015. Measurements of PCNs in human blood has only been found from Sweden during the period 2012-2017 (Table 5.2.9). In total, 5 compounds of PCNs including penta-, hexa-, hepta-, octa-, and tetra- chlorinated naphthalenes were measured with the highest average of median values for pentachloronaphthalene with 14 ng/g lw (N=6) and tetrachloronaphthalene with 12 ng/g lw (N=12) (Table 5.2.36). No trend of PCNs could be observed during this observed period.

Table 5.2.36. Summary statistics of human blood concentrations for PCNs in Europe (Sweden) (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
Tetrachloro-naphthalene	Europe (Sweden)	ng/g lw	12	12	9.0	8.2	3.5 - 27	0	2
Pentachloro-naphthalene	Europe (Sweden)	ng/g lw	6	14	14	2.1	11 – 17	0	2
Hexachloro-naphthalene	Europe (Sweden)	ng/g lw	6	6.1	6.0	1.0	4.6 - 7.7	0	2
Heptachloro-naphthalene	Europe (Sweden)	ng/g lw	6	0.82 ³	0.84 ³	0.091	0.7 - 0.94	6	2
Octachloro-naphthalene	Europe (Sweden)	ng/g lw	6	0.23 ³	0.23 ³	0.029	0.21 - 0.28	6	2

Pentachlorophenol (PCP)

Human milk

PCP was included in the Stockholm convention in 2015. No biomonitoring data of PCP in breast milk was found in the literature search or in the biomonitoring programs.

Human blood

Data for PCP in human plasma was only found from two countries, Germany (N=213) and Canada (N= 2) (Table 5.2.37). PCP show decreasing trend for the period 1985-2012 (Figure 5.2.35).

Table 5.2.37. Summary statistics of human blood (plasma) concentrations on wet weight basis for PCP in Europe (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
PCP	Europe (Germany)	ng/g ww plasma	213	2.8	1.5	4.1	0.33 -26	0	69
	North America (Canada)	ng/g ww plasma	2	0.39	0.39	0.14	0.29 -0.49	0	62
	WEOG	ng/g ww plasma	215	2.8	1.5	4.1	0.29 -26	0	

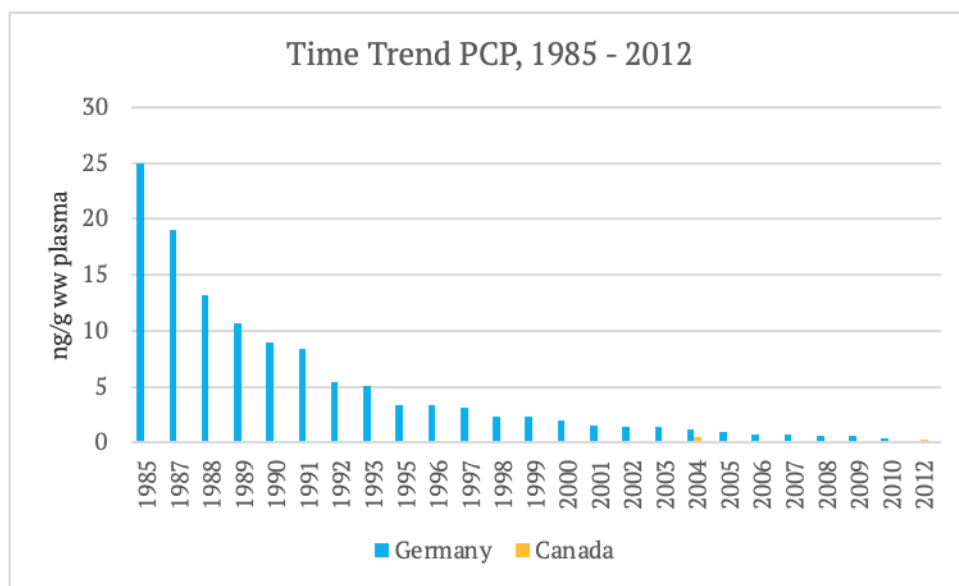


Figure 5.2.35. Time trend (1985 – 2012) of human blood (plasma) PCP concentrations in the WEOG region (Germany and Canada). This time trend is based on the average of 215 reported mean/median values.

Hexabromobiphenyl ethers (HBB)

Human milk

Polybromobiphenyls (PBBs) have been measured in ten countries within the WEOG but most of the measured data, 8 out of 11 median values, were below LOD (Table 5.2.38). Moreover, the number of measurements in the investigated period 1999-2012 was low. Consequently, no trend could be identified for PBB in breast milk within the WEOG region.

Table 5.2.38. Summary statistics of human milk concentrations for PBB in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
BB153	Europe	ng/g lw	4	0.25	0.25	0.00	0.25 - 0.25	4	1
	North America	ng/g lw	5	0.67	0.62	0.48	0.14 - 1.3	3	56, 57
	Australasia	ng/g lw	2	0.88	0.88	0.88	0.25 - 1.5	1	1
	WEOG	ng/g lw	11	0.56	0.25	0.48	0.14 - 1.5	8	
Sum PBB	Europe	ng/g lw	2	0.22	0.22	0.064	0.17 - 0.26	0	9
	WEOG	ng/g lw	2	0.22	0.22	0.064	0.17 - 0.26	0	

Human blood

In serum, polybromodiphenyl ethers (i.e. BB153) was measured within the NHANES program in the USA every second year between 2005 and 2015. Except for 2013, there seems to be a downwards trend in the data (Figure 5.2.36), though data of the latest years or future years are needed to confirm this statistically.

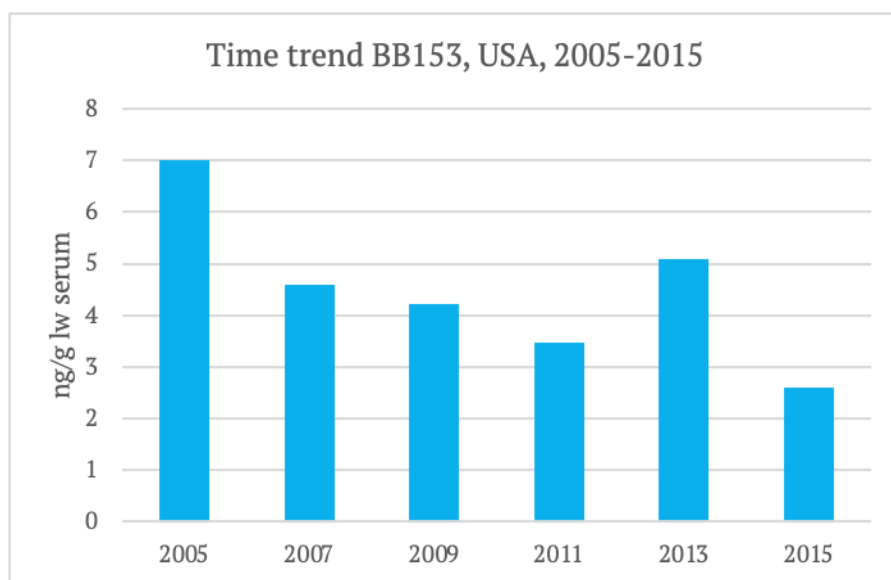


Figure 5.2.36: Time trend (2005 – 2015) for reported geometric mean values for BB153 in North America (USA, NHANES).

Pentachlorobenzene (PeCB)

Human milk

PeCB was measured in ten countries within WEOG with the highest average of median values in North America (1.1 ng/g lw, N=10) followed by Europe (0.28 ng/g lw, N=10) (Table 5.2.39). No trend could be observed for PeCB within WEOG region during the period 1992-2012.

Table 5.2.39. Summary statistics of human milk concentrations for PeCB in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
PeCB	Europe	ng/g lw	10	0.28	0.26	0.038	0.25 - 0.36	4	1, 9, 20
	North America	ng/g lw	10	1.1	1.1	0.29	0.5 - 1.5	0	9
	Australasia	ng/g lw	2	0.25	0.25	0.00	0.25 - 0.25	2	1
	WEOG	ng/g lw	22	0.63	0.34	0.45	0.25 - 1.5	6	

Human blood

No biomonitoring data of PeCB in human blood was found in the literature search or in the national biomonitoring programs.

Perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA)

Human milk

Per- and polyfluoroalkyl substances (PFAS) has been used intensively as dirt and water repellant as well as in fire-fighting foams since the 1950s but recently got attention due to their unique physical and chemical properties. In 2009, PFOS, its salts and perfluorooctane sulfonyl fluoride (PFOSF) were included in Stockholm convention and in 2019 PFOA, its salts and PFOA-related compounds were listed. These chemicals have been measured in breast milk in eight countries within the WEOG during the period 1972-2011 with average of median values of 0.32 ng/g lw (N=33) for sum linear and branched PFOA and 0.15 ng/g lw (N=52) for sum of linear and branched PFOS (Table 5.2.40).

Sweden has measurements of PFOA and PFOS in biobanked breast milk from 1972. Since linear PFOA and PFOS are the major components as compared to their branched isomers, observation for these linear isomers could be used as indicator for changing in trends of PFOS and PFOA in breast milk. Since 1972, there is an increasing trend of PFOS and PFOA in breast milk in Sweden until the beginning of 2000s (Figure 5.2.37, Figure 5.2.38, Figure 5.2.39, Figure 5.2.40) when PFAS as a group received more attention due to reports on their persistence, toxicity and presence in humans and the environment. Since then, the trend of linear PFOA and PFOS has decreased as a result of the ban/restriction from use in commercial as well as industrial products and fire-fighting foams. Measurements of Extractable organic fluorine (EOF) shows that target

PFASs explains 11-75% of the total PFAS in Swedish breast milk and that the proportion of unknown PFASs increases over time.

Table 5.2.40. Summary statistics of human milk concentrations for target PFASs in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
PFOA	Europe	ng/ml	29	0.084	0.10	0.033	0.025 - 0.16	15	2, 3, 25, 28, 31, 42, 44, 48, 51, 59, 61
PFOA br	Europe	ng/ml	23	0.0010	0.0010	0	0.0010 - 0.0010	23	2
PFOA L	Europe	ng/ml	23	0.068	0.067	0.025	0.029 - 0.13	0	2
PFOS	Europe	ng/ml	50	0.14	0.20	0.071	0.023 - 0.26	4	2, 3, 9, 25, 28, 31, 42, 48, 51, 59, 61
	North America	ng/ml	1	0.11	0.11		0.11 - 0.11	0	9
	WEOG	ng/ml	51	0.14	0.20	0.070	0.023 - 0.26	4	
PFOS br	Europe	ng/ml	23	0.0070	0.0047	0.0061	0.00025 - 0.024	1	2
PFOS L	Europe	ng/ml	27	0.064	0.053	0.041	0.0073 - 0.17	0	2

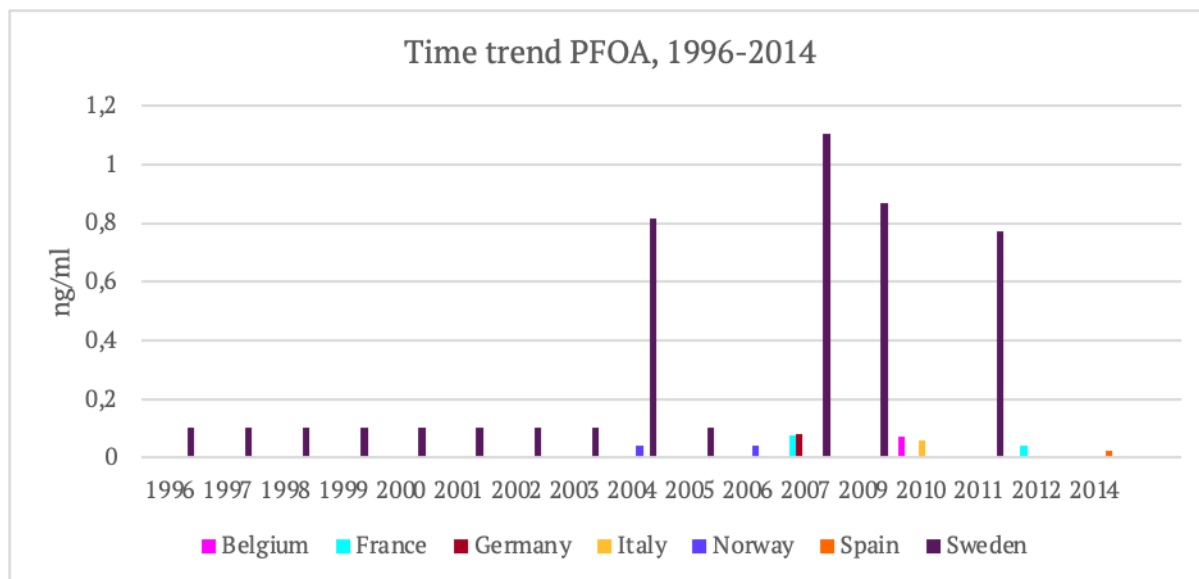


Figure 5.2.37. Time trend (1996 – 2014) for human milk PFOA concentrations in European countries. Time trend is based on 33 reported mean/median values, where 21 samples are from the Swedish national monitoring program and 13 from scientific literature.

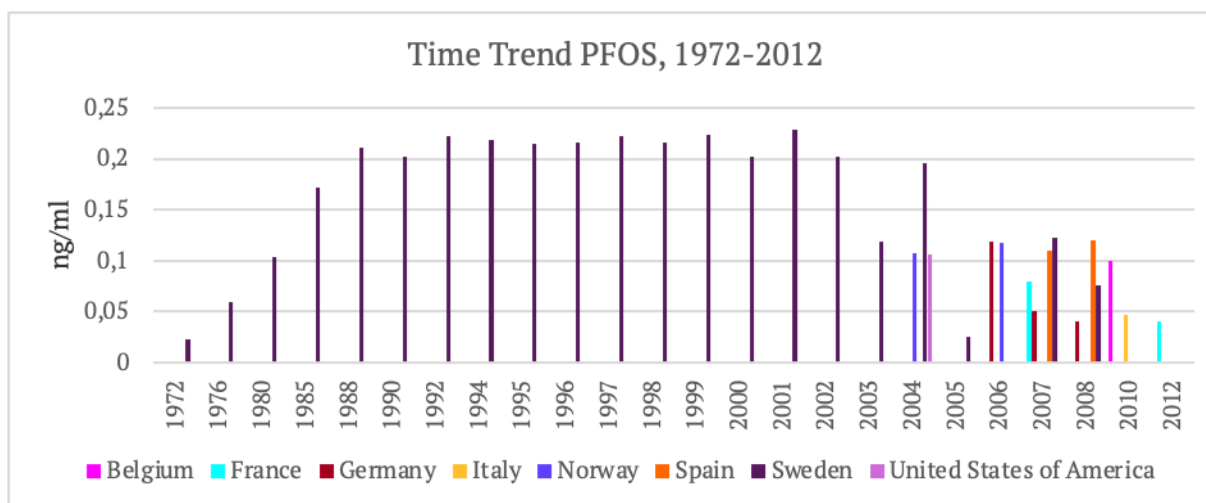


Figure 5.2.38. Time trend (1972 – 2012) for reported human milk PFOS concentrations in the WEOG region. Time trend is based on 52 reported mean/median values, where 4 samples are from the Swedish monitoring program and 48 from scientific literature.

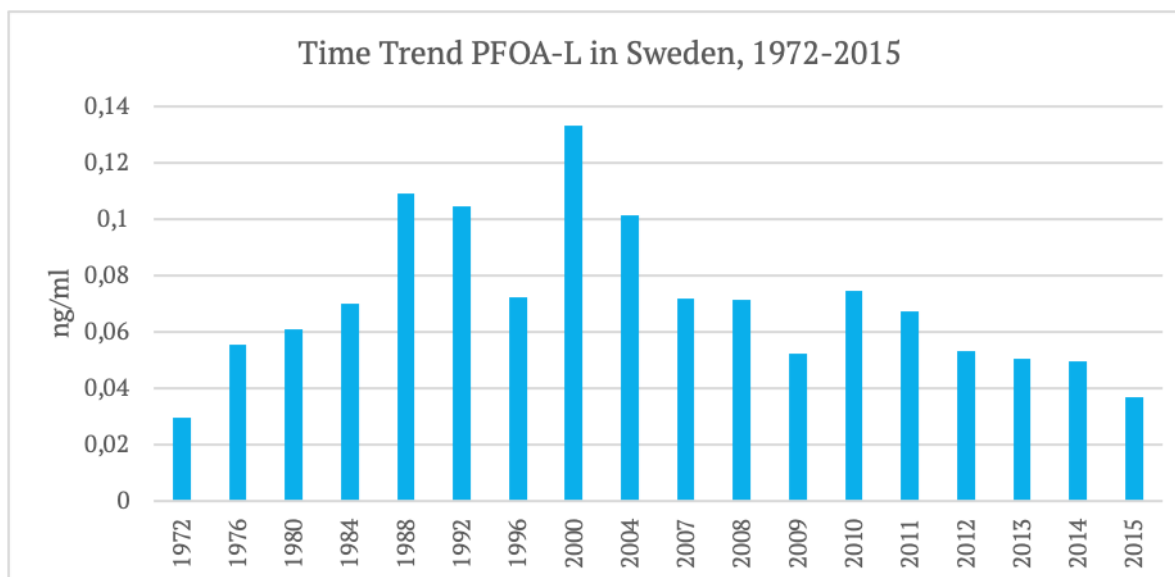


Figure 5.2.39. Time trend (1972 – 2015) for reported concentrations of PFOA-L in Swedish breast milk. The time trend is based on 67 single samples originating from the Swedish monitoring program.

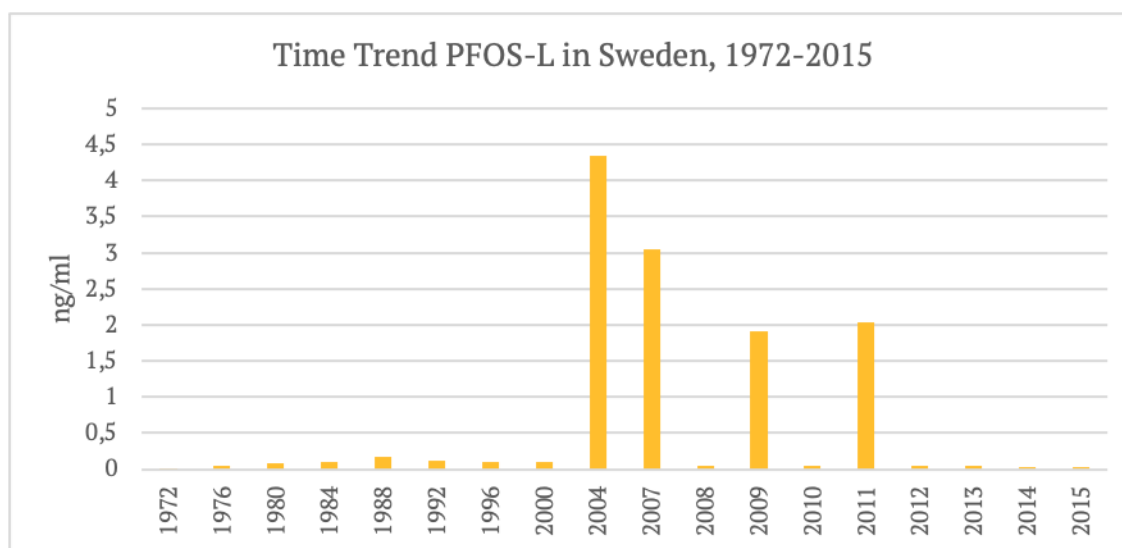


Figure 5.2.40. Time trend (1972-2015) for breast milk PFOS-L concentrations in Sweden. The time trend is based on 86 single samples and 31 pooled samples originating from the Swedish national monitoring program.

Human Blood

During the period 1982-2017, PFOA and PFOS in blood have been reported in ten countries within the WEOG. Summary statistics of PFOA and PFOS in human blood can be found in Table 5.2.41 with data reported from Europe, North America and Australasia regions. The highest average concentration for PFOA and PFOS is in Australasia region with 6.6 ng/mL (N=68) and 19 ng/mL (N=68), respectively. The lowest average concentration could be observed in North America with 2.2 ng/mL (N=43) for PFOA and 5.7 ng/mL (N=43) for PFOS.

Table 5.2.41. Summary statistics of human blood (plasma or serum) concentrations for PFOA and PFOS in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
PFOA	Europe	ng/mL	142	3.0	2.6	1.5	0.079 - 7.8	115	2, 62, 65, 69
PFOA	North America	ng/mL	43	2.2	2.3	1.0	0.30 - 4.9		62, 66, 67
PFOA	Australasia	ng/mL	68	6.6	5.9	2.4	2.8 – 12		68
PFOA Total	WEOG	ng/mL	253	3.8	3.0	2.4	0.079 – 12	115	
PFOS	Europe	ng/mL	162	13	9.9	9.6	1.4 - 61	135	2, 62, 65, 69
PFOS	North America	ng/mL	43	5.7	6.0	4.5	0.10 – 22		62, 66, 67
PFOS	Australasia	ng/mL	68	19	18	7.3	7.1 – 36		68
PFOS Total	WEOG	ng/mL	273	13	10.7	9.3	0.10 - 61	135	

Since 2009, the trend of PFOA and PFOS in human blood has decreased as a result of the ban/restriction of these compounds in commercial use as well as industrial products. Levels of PFOA and PFOS in human blood since 1982 until 2009 did not show any significant trend but rather kept stable (Figure 5.2.41 and Figure 5.2.42).

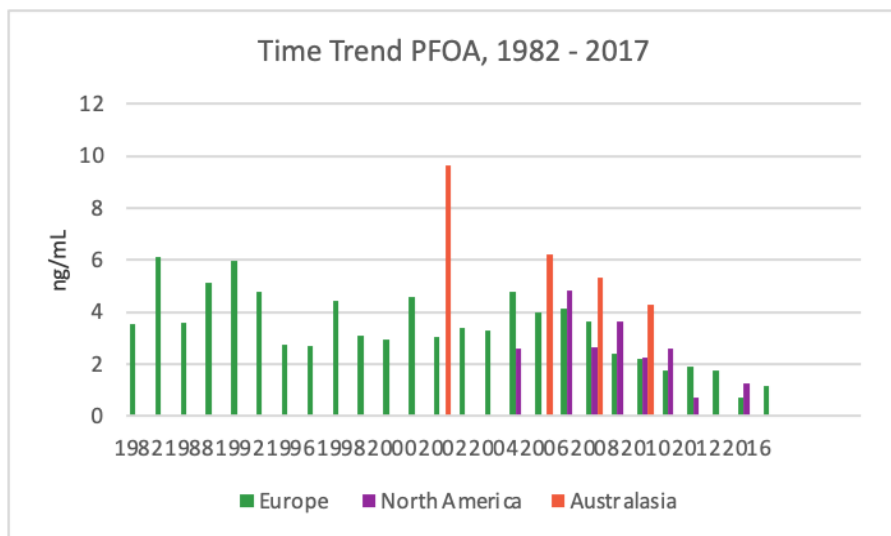


Figure 5.2.41. Time trend (1982 – 2017) for human blood PFOA concentrations in North America, Australasia and Europe.

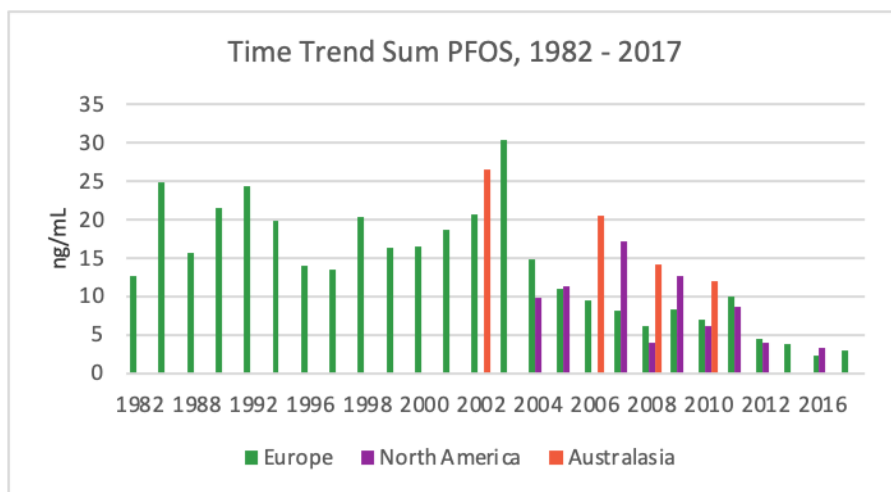


Figure 5.2.42. Time trend (1982 – 2017) for human blood PFOS concentrations in North America, Australasia and Europe.

Interestingly, a slightly increasing trend of Perfluorohexane sulfonic acid (PFHxS) could be observed in human blood (Figure 5.2.43). This might be due to the increasing use of PFHxS as a replacement for PFOA and PFOS since the usage of these compounds started to be phased out in the 2000s. PFHxS has been nominated for listing to the Convention and it will be considered by the COP at COP10.

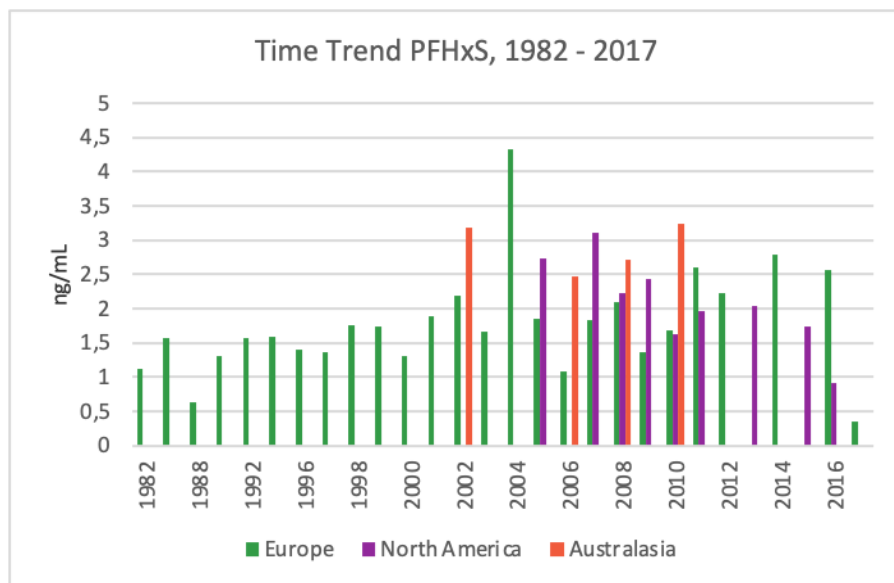


Figure 5.2.43. Time trend (1982 – 2017) for human blood PFHxS concentrations in North America, Australasia and Europe.

Toxaphene

Human milk

The three indicator congeners (Parlar 26, 50 and 62) of the insecticide Toxaphene have been measured in breast milk within the WEOG region. The congeners have been measured in 13 different countries in Australasia and Europe (Table 5.2.42). One reported value of “Toxaphene” could be found in Canada, sampled in 1999. The three congeners are regarded as indicator congeners as they generally comprise a major part of the toxaphene residues found in biota.

Out of the three congeners, Parlar 62 appears in lower concentrations, where all reported data were below the quantification limit. Both Parlar 26 and 50 are decreasing over time, as can be seen in Figure 5.2.44, where the two last sampling years (2011 in New Zealand and 2012 in Israel) yielded values below the quantification limit.

Table 5.2.42. Summary statistics of human milk concentrations for toxaphene in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	Reference
Parlar 26	Europe	ng/g lw	11	0.83	0.50	0.91	0.25 - 3.3	4	1
	Australasia	ng/g lw	2	0.32	0.32	0.099	0.25 - 0.39	1	1
	WEOG	ng/g lw	13	0.75	0.45	0.85	0.25 - 3.3	5	
Parlar 50	Europe	ng/g lw	11	2.0	1.8	1.7	0.25 - 6.7	2	1
	Australasia	ng/g lw	2	0.70	0.70	0.64	0.25 - 1.2	1	1
	WEOG	ng/g lw	13	1.8	1.5	1.7	0.25¹ - 6.7	3	
Parlar 62	Europe	ng/g lw	11	0.25 ¹	0.25 ¹	0	0.25 ¹ - 0.25 ¹	11	1
	Australasia	ng/g lw	2	0.25 ¹	0.25 ¹	0	0.25 ¹ - 0.25 ¹	2	1
	WEOG	ng/g lw	13	0.25¹	0.25¹	0	0.25¹ - 0.25¹	13	
Toxaphene	North America (Canada)	ng/g lw	1	56	56	-	56 - 56	0	

¹ All values below LOD/LOQ.

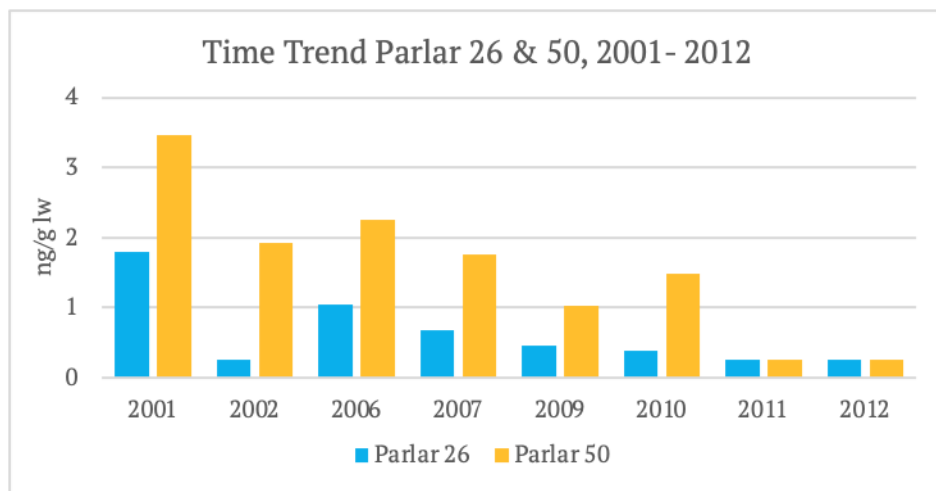


Figure 5.2.44. Time trend (2001 – 2012) for breast milk concentrations of Parlar 26 and Parlar 50 in the WEOG region (values reported from Europe and Australasia).

Human blood

No biomonitoring data of Toxaphene in human blood were found in the biomonitoring programs.

Polychlorinated dibenzo-p-dioxins (dioxins) and Polychlorinated dibenzofurans (furans)

Human milk

The polychlorinated dibenzo-p-dioxin/polychlorinated dibenzofuran (PCDD/PCDF) concentrations are reported as toxic equivalents (TEQs) based on WHO TEF values from 1998 (Van den Berg et al. 1998) and 2005 (Van den Berg et al. 2006). Within the region, mean and median values for Σ PCDD/F have been reported in 23 countries during the period 1972-2017. The reported breast milk concentrations vary between 0.0033-0.049 ng/g lw (WHO TEQ 1998) and 0.0019-0.036 ng/g lw (WHO TEQ 2005) (Table 5.2.43). A decreasing trend can be observed for Σ PCDD/F during the sampling period, as can be seen in Figure 5.2.45 (WHO TEQ1998) and Figure 5.2.46 (WHO TEQ2005).

In total, 58 and 35 reported mean/median values for Total TEQ were reported in WHO TEQ 1998 and WHO TEQ 2005 respectively. The median/mean values ranged from 0.0065 - 0.10 ng/g lw (WHO TEQ 1998) and 0.0063 - 0.072 ng/g lw (WHO TEQ 2005) (Table 5.2.43). When examining regional differences in concentrations, Europe exhibits slightly higher concentrations than North America and Australasia during the sampling period. A decreasing trend can be observed for total TEQ, during the sampling period 1972-2010 (Figure 5.2.47).

Table 5.2.43. Summary statistics of human milk concentrations for dioxins and furans in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	TEQ	Continent	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
ΣPCDD/F	WHO TEQ 1998	Europe	63	0.012	0.0089	0.0087	0.0045 - 0.049	0	4, 9, 11, 21, 24, 45
[ng/g lw]		North America	5	0.0070	0.0049	0.0045	0.0033 - 0.015	0	9, 55
		WEOG	68	0.012	0.0087	0.0086	0.0033 - 0.049	0	
	WHO TEQ 1998 LB/(UB)	Europe	60	0.0093	0.0061	0.0073	0.0020 - 0.033	0	1
		North America	5	0.014	0.015	0.0050	0.0054 - 0.018	0	1
		Australasia	7	0.0032	0.0035	0.0017	0.0011 - 0.0058	0	1
		WEOG	72	0.0090	0.0059	0.0071	0.0011 - 0.033	0	
	WHO TEQ 2005	Europe	48	0.010	0.0091	0.0068	0.0023 - 0.036	0	4, 9, 24, 27, 29, 46, 47
		North America	9	0.0045	0.0047	0.0021	0.0019 - 0.0083	0	9, 12, 26, 54
		Australasia	3	0.0046	0.0049	0.00096	0.0035 - 0.0054	0	5, 26
		WEOG	60	0.0092	0.0077	0.0066	0.0019 - 0.036	0	
	WHO TEQ 2005 LB/(UB)	Europe	60	0.0084	0.0056	0.0071	0.0014 - 0.033	0	1
		North America	5	0.013	0.015	0.0054	0.0038 - 0.017	0	1
		Australasia	7	0.0030	0.0032	0.0018	0.00075 - 0.0058	0	1
		WEOG	72	0.0082	0.0053	0.0070	0.00075 - 0.033	0	
Total TEQ	WHO TEQ 1998	Europe	55	0.023	0.016	0.018	0.0068 - 0.10	0	9, 16, 21, 45
[ng/g lw]		North America	3	0.012	0.012	0.0059	0.0065 - 0.018	0	9, 55
		WEOG	58	0.023	0.015	0.018	0.0065 - 0.10	0	
	WHO TEQ 2005	Europe	30	0.023	0.019	0.015	0.0072 - 0.072	0	9, 11, 27
		North America	3	0.0080	0.0088	0.0015	0.0063 - 0.0090	0	12, 27, 55
		Australasia	2	0.0070	0.0070	0.00013	0.0069 - 0.0071	0	27
		WEOG	35	0.021	0.014	0.015	0.0063 - 0.072	0	

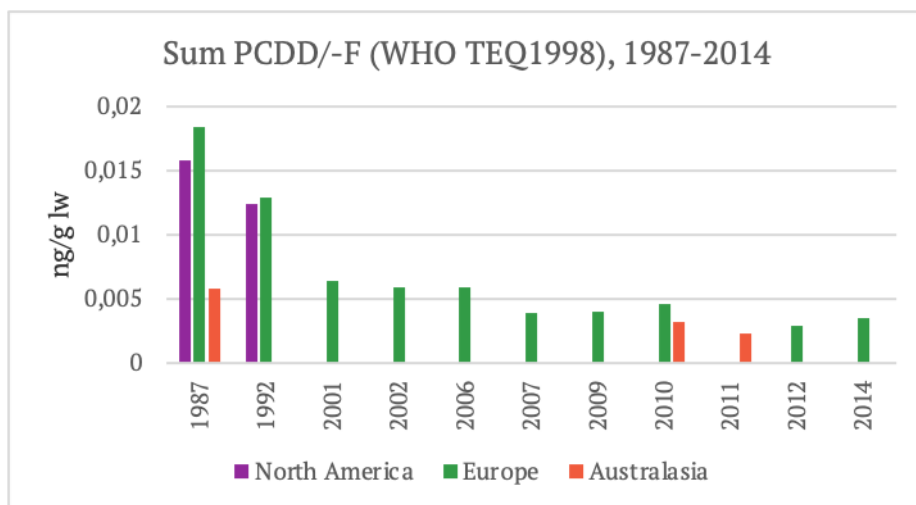


Figure 5.2.45. Time trend (1987 - 2014) for human milk concentrations of sum PCDD/-F WHO TEQ1998 in North America, Europe and Australasia.

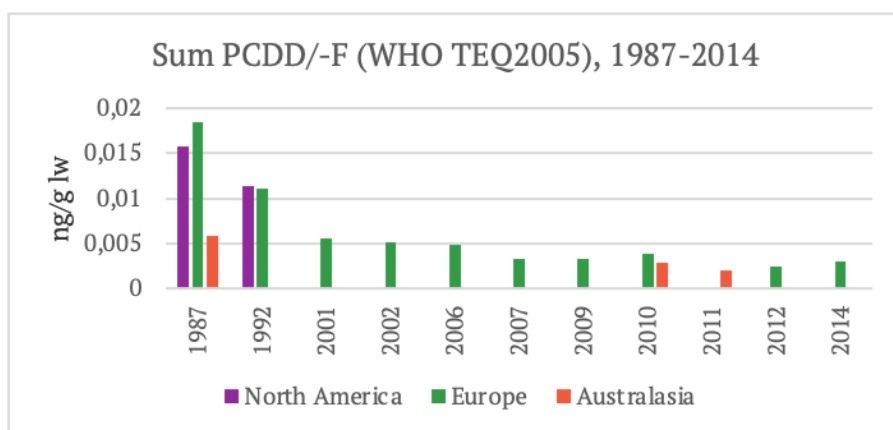


Figure 5.2.46. Time trend (1987 - 2014) for human milk concentrations of sum PCDD/-F WHO TEQ2005.

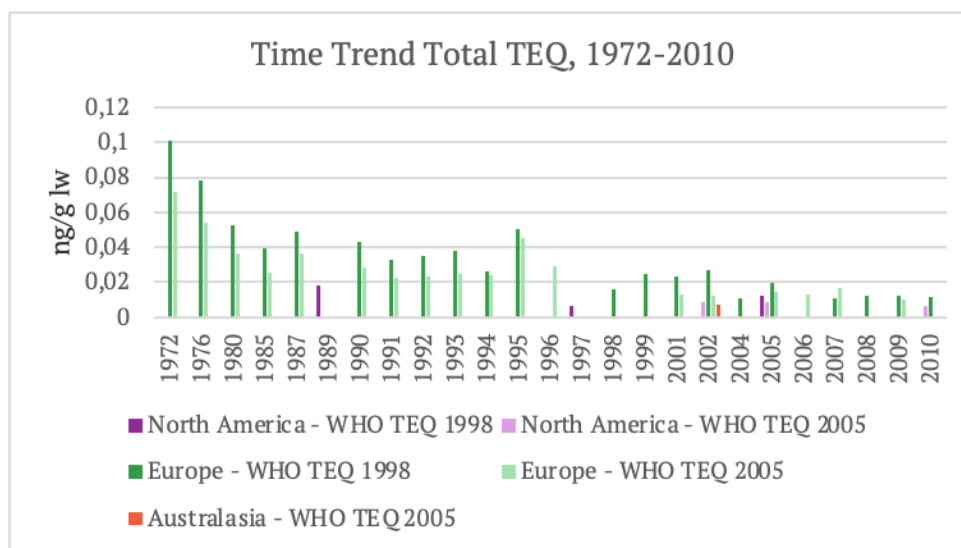


Figure 5.2.47. Time trend (1972 – 2010) for reported human milk concentrations of total TEQ in North America, Europe and Australasia.

Human blood

TEQ-values for PCDDs or PCDFs in blood, serum or plasma were hardly available, except for within the Australian dataset for 2010 and 2012, as well as for the Swedish national monitoring (for highly exposed fish-consumers in 2002).

The dataset from Australasia was chosen to indicate the different PCDD and PCDF levels compared to each other, as this dataset included the most current year (2012). For this, an average per year and over all pooled sample groups was taken (Figure 5.2.48). Highest average levels in 2012 are detected for OctaCDD (the concentration refers to right axis), followed by 1,2,3,4,6,7,8-HpCDD and 1,2,3,6,7,8-HxCDD (Figure 5.2.48). It cannot be ruled out, that this congener pattern might be continent specific though.

Figure 5.2.48. Levels of different PCDDs and PCDFs in serum; data from Australia, average values calculated of the 2012 data. Note that the OctaCDD bar relates to values of the right axis.

Several of the dioxins and furans showed a decreasing trend between 1987 and 2012 as illustrated in Figures 5.2.49 – 5.2.54 and Tables 5.2.44 – 5.2.46 for 2,3,7,8-TeCDD, 1,2,3,7,8-PeCDD, 1,2,3,4,7,8-HxCDD, 1,2,3,4,7,8-HxCDF, 1,2,3,4,6,7,8-HpCDD and OctaCDD.

Generally, the dioxin and furan levels of this dataset are higher in Europeans than in North Americans and Australians, and often Australian data are lowest. However, one should be cautious in concluding that this observation represents the real differences among the continents. The observed differences may originate to large extent from the variation in timing and scope of the monitoring programs that provided data for this dataset. The European data are the oldest one, followed by data from North America while the Australian data are for the latest period. This means that the decrease in concentrations achieved as a result of the Stockholm Convention provisions are most visible in the Australian data, while the European data shows concentrations from the earlier period, even before the Stockholm Convention. In addition, European data shows results for highly or higher exposed people, because the underlying monitoring program focused on people such as fish-consumers, or men over 45, as well as people from known contaminated areas.

OctaCDD is the only compound among all the PCDD/Fs, for which data of different continents are overlapping in time (Figure 5.2.54). The 2008 data from Europe (Sweden) are within the time span of the NHANES campaign. Considering that the European data are for humans living in a locally contaminated area (therefore the average is probably higher than the average of the timely closest NHANES data) and the levels were related to the lipid weight in plasma (instead of lw in serum), the difference between European and North American data is not big (Figure 5.2.54).



Figures 5.2.49-5.2.54. Time trend of human blood serum concentrations (on lipid weight basis) of 2,3,7,8-TeCDD (Figure 5.2.49), 1,2,3,7,8-PeCDD (Figure 5.2.50), 1,2,3,4,7,8-HxCDD (Figure 5.2.51), 1,2,3,4,7,8-HxCDF (Figure 5.2.52), 1,2,3,4,6,7,8-HpCDD (Figure 5.2.53) and OctaCDD (Figure 5.2.54) in the WEOG region for the period 1987 – 2012.

Table 5.2.44. Summary statistics of human blood (serum) concentrations *on lipid weight basis* for the TCDD/Fs and PeCDD/Fs in the WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
2,3,7,8 - TeCDD	Europe	ng/g lw serum	3	0.0042	0.0037	0.0011	0.0033 -0.0054	0	2
	North America	ng/g lw serum	3	0.0011	0.0012	0.00015	0.00094 -0.0012	0	67
	Australasia	ng/g lw serum	27	0.00066	0.00036	0.00047	0.00022 -0.0016	0	68
	WEOG	ng/g lw serum	33	0.0010	0.00070	0.0011	0.000215 -0.0054	0	
2,3,7,8 - TeCDF	Europe	ng/g lw serum	3	0.0020	0.0017	0.00043	0.0017 -0.0025	0	2
	North America	ng/g lw serum	3	0.00026	0.00023	0.00012	0.00015-0.00038	0	67
	Australasia	ng/g lw serum	27	0.00032	0.00019	0.00030	0.00013 -0.0013	0	68
	WEOG	ng/g lw serum	33	0.00047	0.00020	0.00057	0.00013 -0.0025	0	
1,2,3,7,8- PeCDD	Europe	ng/g lw serum	3	0.011	0.012	0.0013	0.0097 -0.012	0	2
	North America	ng/g lw serum	3	0.0039	0.0040	0.00029	0.0035 -0.0041	0	67
	Australasia	ng/g lw serum	27	0.0021	0.0015	0.0013	0.00036 -0.0048	0	68
	WEOG	ng/g lw serum	33	0.0031	0.0025	0.0029	0.00036 -0.012	0	
1,2,3,7,8- PeCDF	Europe	ng/g lw serum	3	0.00085	0.0010	0.00038	0.00043 -0.0012	0	2
	North America	ng/g lw serum	3	0.00035	0.0004	0.000049	0.00029 -0.00038	0	67
	Australasia	ng/g lw serum	29	0.00032	0.0003	0.00012	0.00023 -0.00076	0	68
	WEOG	ng/g lw serum	35	0.00037	0.00032	0.00021	0.00023 -0.0012	0	
2,3,4,7,8- PeCDF	Europe	ng/g lw serum	3	0.040	0.042	0.0042	0.036 -0.044	0	2
	North America	ng/g lw serum	3	0.0045	0.0045	0.000046	0.0044 -0.0045	0	67
	Australasia	ng/g lw serum	29	0.0027	0.0026	0.0012	0.0010 -0.0055	0	68
	WEOG	ng/g lw serum	35	0.0061	0.0028	0.011	0.0010-0.044	0	

Table 5.2.45. Summary statistics of human blood (serum) concentrations *on fresh weight basis* for the HxCDD/Fs in the WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
1,2,3,4,7,8-HxCDD	Europe	ng/g lw serum	3	0.0035	0.003	0.00052	0.0030 -0.0041	0	2
	North America	ng/g lw serum	3	0.0027	0.003	0.00038	0.0024 -0.0031	0	67
	Australasia	ng/g lw serum	30	0.0012	0.001	0.00082	0.00008 -0.0028	0	68
	WEOG	ng/g lw serum	36	0.0015	0.001	0.0011	0.00008 -0.0041	0	
1,2,3,4,7,8-HxCDF	Europe	ng/g lw serum	3	0.0080	0.007	0.0018	0.0070 -0.010	0	2
	North America	ng/g lw serum	3	0.0032	0.0033	0.00027	0.0029 -0.0034	0	67
	Australasia	ng/g lw serum	29	0.0011	0.0010	0.00039	0.00017 -0.0020	0	68
	WEOG	ng/g lw serum	35	0.0019	0.0012	0.0021	0.00017 -0.010	0	
1,2,3,6,7,8-HxCDD	Europe	ng/g lw serum	3	0.042	0.042	0.0078	0.034 -0.050	0	2
	North America	ng/g lw serum	3	0.021	0.021	0.0023	0.019 -0.024	0	67
	Australasia	ng/g lw serum	30	0.0091	0.0078	0.0060	0.0022 -0.022	0	68
	WEOG	ng/g lw serum	36	0.013	0.010	0.011	0.0022 -0.050	0	
1,2,3,6,7,8-HxCDF	Europe	ng/g lw serum	3	0.0081	0.0077	0.0018	0.0066 -0.010	0	2
	North America	ng/g lw serum	3	0.0033	0.0033	0.00017	0.0032 -0.0035	0	67
	Australasia	ng/g lw serum	30	0.0014	0.0013	0.00061	0.000065 -0.0027	0	68
	WEOG	ng/g lw serum	36	0.0021	0.0015	0.0020	0.000065 -0.010	0	
1,2,3,7,8,9-HxCDD	Europe	ng/g lw serum	3	0.0058	0.0059	0.0018	0.0039 -0.0075	0	2
	North America	ng/g lw serum	3	0.0031	0.0031	0.00019	0.0030 -0.0033	0	67
	Australasia	ng/g lw serum	30	0.0022	0.0018	0.00104	0.0010 -0.0049	0	68
	WEOG	ng/g lw serum	36	0.0026	0.0022	0.0015	0.0010 -0.0075	0	
1,2,3,7,8,9-HxCDF	Europe	ng/g lw serum	3	0.0010	0.00050	0.00085	0.0005 -0.0020	0	2
	North America	ng/g lw serum	3	0.000088	0.000089	0.0000069	0.000081 -0.000094	0	67
	Australasia	ng/g lw serum	30	0.00014	0.00012	0.000082	0.000095 -0.00056	0	68
	WEOG	ng/g lw serum	36	0.00021	0.00012	0.00032	0.000081 -0.0020	0	

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
2,3,4,6,7,8-HxCDF	Europe	ng/g lw serum	3	0.0032	0.0023	0.0018	0.0021 -0.0053	0	2
	North America	ng/g lw serum	3	0.00086	0.00095	0.00018	0.00065 -0.00097	0	67
	Australasia	ng/g lw serum	30	0.00034	0.00037	0.00025	0.000055 -0.00076	0	68
	WEOG	ng/g lw serum	36	0.00062	0.00042	0.00094	0.000055 -0.0053	0	

Table 5.2.46. Summary statistics of human blood (serum) concentrations on lipid weight basis for the HpCDD/Fs and OctaCDD/Fs in the WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
1,2,3,4,6,7,8-HpCDD	Europe	ng/g lw serum	3	0.064	0.067	0.0	0.033 -0.093	0	2
	North America	ng/g lw serum	3	0.025	0.023	0.0	0.021 -0.030	0	67
	Australasia	ng/g lw serum	31	0.013	0.012	0.0	0.0055 -0.027	0	68
	WEOG	ng/g lw serum	37	0.018	0.013	0.0	0.0055 -0.093	0	
1,2,3,4,6,7,8-HpCDF	Europe	ng/g lw serum	3	0.015	0.017	0.0	0.012 -0.017	0	2
	North America	ng/g lw serum	3	0.0075	0.0078	0.0	0.0065 -0.0081	0	67
	Australasia	ng/g lw serum	31	0.0024	0.0024	0.0	0.00075 -0.00394	0	68
	WEOG	ng/g lw serum	37	0.0038	0.0027	0.0	0.00075 -0.017	0	
1,2,3,4,7,8,9-HpCDF	Europe	ng/g lw serum	3	0.00085	0.00050	0.00060	0.0005 -0.0015	0	2
	North America	ng/g lw serum	3	0.00012	0.00010	0.000047	0.000080 -0.00017	0	67
	Australasia	ng/g lw serum	19	0.00019	0.00011	0.00014	0.00009 -0.000484	0	68
	WEOG	ng/g lw serum	25	0.00026	0.00011	0.00031	0.000080 -0.0015	0	
OctaCDD	Europe	ng/g lw serum	5	0.3971	0.3996	0.1053	0.26 -0.52	0	2
	North America	ng/g lw serum	3	0.2252	0.2093	0.03256	0.20 -0.26	0	67
	Australasia	ng/g lw serum	31	0.17466	0.15990	0.06335	0.090 -0.33	0	68
	WEOG	ng/g lw serum	39	0.2071	0.18190	0.1001	0.090 -0.52	0	
OctaCDF	Europe	ng/g lw serum	3	0.0033	0.0013	0.00380	0.001 -0.0077	0	2
	North America	ng/g lw serum	3	0.00163	0.00189	0.00092	0.00060 -0.0024	0	67
	Australasia	ng/g lw serum	31	0.00085	0.00080	0.00015	0.00065 -0.0012	0	68
	WEOG	ng/g lw serum	37	0.00112	0.00085	0.00117	0.00060 -0.0077	0	

Polybrominated diphenyl ethers (PBDEs)

Human milk

Among 209 possible congeners, only BDE 28, 47, 99, 100, 153, 154, 183, and 209 are relevant when looking at the dietary exposure of PBDEs. BDE47 and BDE209 are usually used as indicators for exposure of humans to PBDEs. During the period 1972-2016, PBDEs in breast milk have been measured in 19 countries within the WEOG. The highest average of breast milk median values for either BDE47 (Figure 5.2.55), BDE209 (Figure 5.2.56) or sum PBDEs (Figure 5.2.57) are found in North America with 23 ng/g lw (N=14), 39 ng/g lw (N=6) and 31 ng/g lw (N=14), respectively (Table 5.2.47). A lower concentration in the sum PBDEs compared to concentrations of separate PBDE congeners (BDE47 and BDE209) was an effect from differences in populations used in the data sources from North America for the different congeners. Europe has the lowest average of median values with 1.4 ng/g lw (N=130) for BDE47, 0.75 ng/g lw (N=31) for BDE209, and 2.8 ng/g lw (N=81) for sum PBDEs (Table 5.2.47).

Tetra-, penta-, hexa-, and heptabromodiphenyl ether were included in the Stockholm Convention in 2009 and decabromodiphenyl ether were included in 2017. Before 2009, BDE47 showed a significant increasing trend during the period 1976-2009 while an opposite trend was observed during the period 2010-2016 where BDE47 showed decreasing tendency in breast milk. More data are required in order to show more conclusive trends for PBDEs in breast milk.

Table 5.2.47. Summary statistics of breast milk concentrations for BDE 209, BDE 47 and Σ PBDE in WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	Reference
BDE 209	Europe	ng/g lw	31	0.75	0.59	1.0	0.08 - 5.8	12	2, 6, 9, 15, 17, 19, 29, 31, 36, 37
	North America	ng/g lw	6	39	7.2	81	0.10 - 200	1	9, 58
	WEOG	ng/g lw	37	7.0	0.65	33	0.08 - 200	13	
BDE 47	Europe	ng/g lw	130	1.4	1.2	0.99	0.0041 - 6.8	4	1, 2, 6, 9, 15, 19, 26, 29, 30, 31, 35, 37, 38, 42, 43, 48, 61
	North America	ng/g lw	14	23	21	17	0.012 - 73	0	9, 22, 34, 54, 56, 58, 60
	Australasia	ng/g lw	14	5.7	6.0	2.9	1.4 - 12	0	1, 38
	WEOG	ng/g lw	160	3.6	1.4	7.9	0.0041 - 73	4	
Σ PBDE ³	Europe	ng/g lw	81	2.8	2.2	4.8	0.0097 - 43	0	9, 29, 16, 17, 30, 36, 37, 38, 43, 46, 61
	North America	ng/g lw	14	31	32	17	0.02 - 54	0	9, 54, 56, 58, 60
	Australasia	ng/g lw	12	7.4	8.0	4.1	2.5-16	0	38
	WEOG	ng/g lw	107	7.0	2.7	12	0.0097 - 54	0	

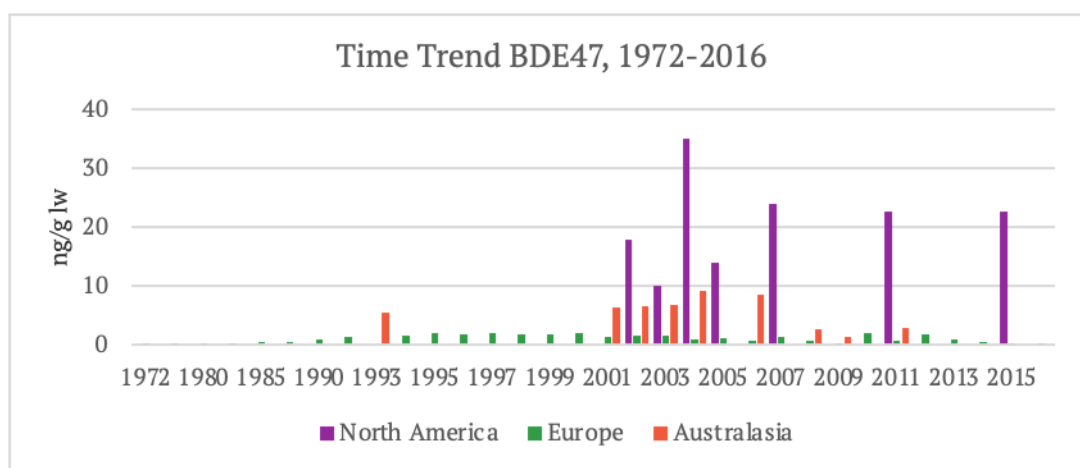


Figure 5.2.55. Time trend for mean and median values of BDE47 in human milk on a lipid weight basis in North America, Europe and Australasia during the sampling period 1972 to 2016.

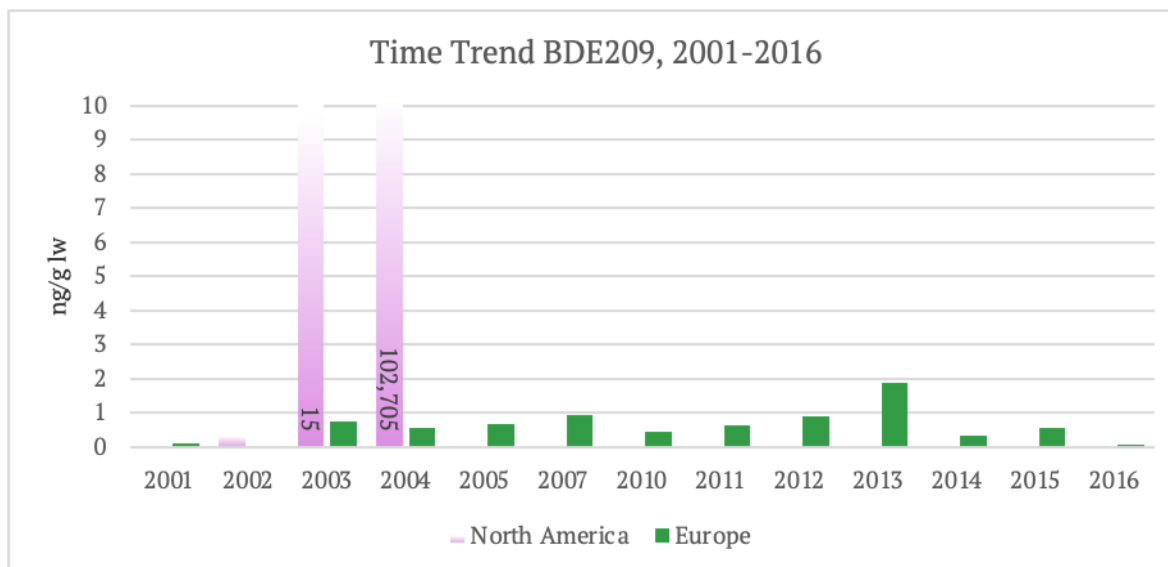


Figure 5.2.56. Time trend for mean and median values for BDE209 in human milk on a lipid weight basis in North America and Europe during the sampling period 2001-2016.

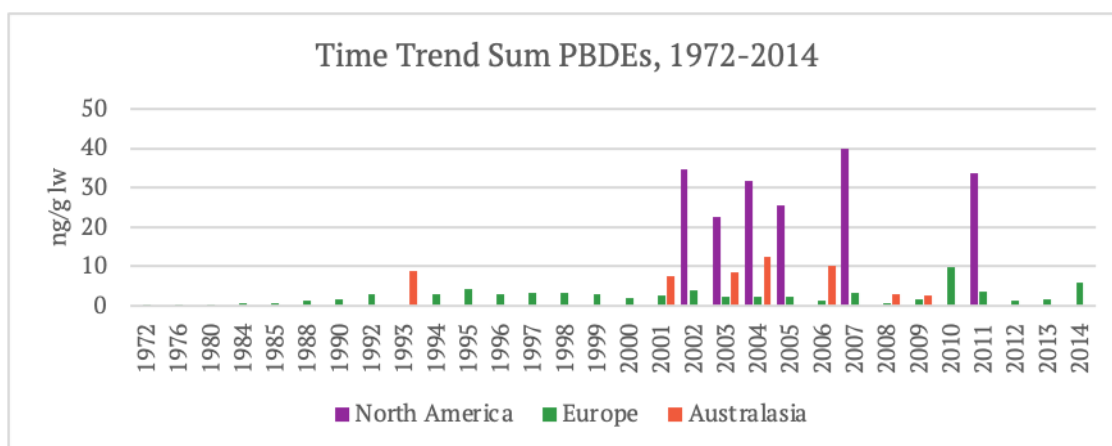


Figure 5.2.57. Time trend for mean and median values for the sum of PBDEs in human milk on a lipid weight basis in North America, Europe and Australasia during the sampling period 1972-2014.

Human Blood

During the period 2002-2017, concentrations of BDE47 and BDE209 in blood have been reported in seven countries within the WEOG (Table 5.2.9). Table 5.2.48 shows data reported on lipid-weight basis, while Table 5.2.49 shows data reported on wet-weight basis. Data on lipid-weight basis are available for years 2002-2015 (excluding the AMAP dataset in year 2010) and on wet-weight basis for years 2002-2017.

The highest average of lipid weight normalized concentration for either BDE47 or BDE209 is found in North America with 11 ng/g lw (N=20), 9.6 ng/g lw (N=4), respectively. Europe has the lowest average of lipid weight normalized concentration with 1.9 ng/g lw (N=27) for BDE47 and 2.1 ng/g lw (N=7) for BDE209 (Table 5.2.48).

The highest average of wet-weight normalized concentration for either BDE47 or BDE209 are found in North America with 0.094 ng/g ww (N=18), 0.019 ng/g ww (N=6), respectively. Europe has lower values with 0.010 ng/g ww (N=14) for BDE47 and 0.004 ng/g ww (N=3) for BDE209 (Table 5.2.49).

Table 5.2.48. Summary statistics of human blood (plasma or serum) concentrations on lipid weight basis concentrations for BDE209 and BDE47 from WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
BDE47	Europe	ng/g lw plasma or serum	27	1.9	1.7	1.1	0.63-6.6	14	2, 62
	North America	ng/g lw plasma or serum	20	11.0	9.7	4.8	6.6-26	0	62, 66, 67
	Australasia	ng/g lw plasma or serum	139	4.4	4	2.2	1.8-21	0	68
	WEOG	ng/g lw plasma or serum	186	4.7	4.0	3.4	0.63-26	14	
BDE209	Europe	ng/g lw plasma or serum	7	2.1	1.2	1.6	0.46-4.0	7	2
	North America	ng/g lw plasma or serum	4	9.6	10	6.4	1.9-16	0	62
	WEOG	ng/g lw plasma or serum	11	4.8	3.0	5.3	0.46-16	7	

Table 5.2.49. Summary statistics of human blood (plasma or serum) concentrations on wet weight basis concentrations for BDE209 and BDE47 from the WEOG region (Number (N) and average, median, standard deviation (SD), range, number of reported values below LOD/LOQ (N<DL/QL)).

Substance	Continent	Unit	N	Average	Median	SD	Range (min-max)	N <DL/QL	References
BDE47	Europe	ng/g ww plasma or serum	14	0.010	0.010	0.0	0.0027-0.020	14	2
	North America	ng/g ww plasma or serum	18	0.094	0.059	0.1	0.049-0.29	0	66, 67
	WEOG	ng/g ww plasma or serum	32	0.057	0.054	0.1	0.0027-0.29	14	
BDE209	Europe	ng/g ww plasma or serum	3	0.004	0.0029	0.0	0.0018-0.0059	3	2
	North America	ng/g ww plasma or serum	6	0.019	0.020	0.0	0.013-0.024	0	67
	WEOG	ng/g ww plasma or serum	9	0.014	0.014	0.0	0.0018-0.024	3	

No clear trend could be observed for BDE47 and BDE209 in human blood for either lipid basis during period 2002-2015 within WEOG region. Only a slightly decreasing trend could be seen for wet-weight basis for BDE47 (Figure 5.2.58) and BDE 209 (Figure 5.2.59) in the North America region, however there was no significant statistical value to confirm this seemingly decreasing trend.

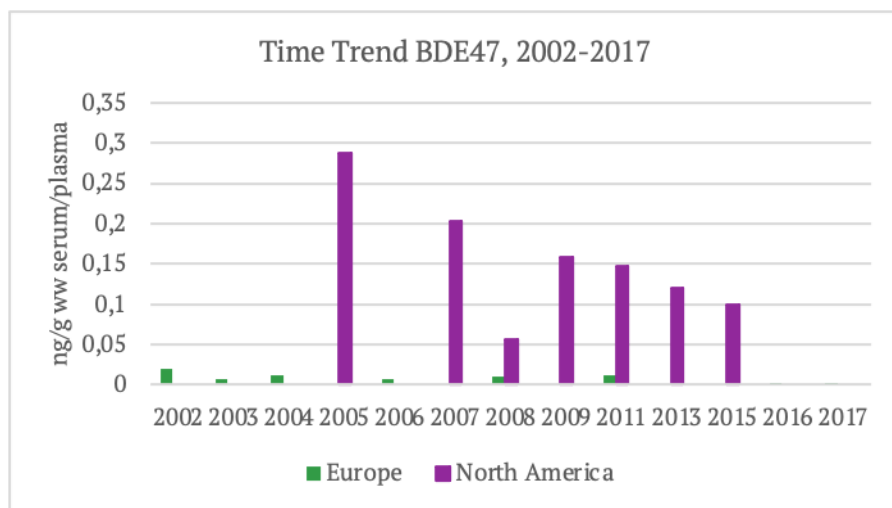


Figure 5.2.58. Time trend (2002 – 2017) for human blood (serum or plasma) BDE47 concentrations on wet weight basis in North America and Europe.

Figure 5.2.59. Time trend (2005 – 2017) for human blood (serum or plasma) BDE209 concentrations on wet weight basis in North America and Europe.

Conclusions

Comparing data from different studies is very challenging and should be performed carefully and any interpretations and analysis of such results should be done with caution. However, by gathering data from both national and regional monitoring as well as scientific literature it was possible to get an overview of the situation in Western Europe, North America and Australasia (the WEOG region). Differences and similarities between countries and continents could be identified, as well as changes over time.

- ❖ The levels of several of the initial Stockholm Convention POPs such as PCBs, DDTs, PCDDs/PCDFs, HCB, Toxaphene, Chlordanes and Dieldrin are decreasing over time in human milk and/or blood. PCP seem to decrease as well even though data are quite scarce. This shows that regulations and banning of production and use of these chemicals combined with regulations on food and recommendations and changes in diet have led to declining levels in humans.
- ❖ Some of the newer POPs show an increase over time followed by a decrease. This is the case of brominated flame retardants PBDEs and HBCD. Data for PFOS and PFOA also indicate an increasing trend accompanied by a decrease. This further confirms that adequate risk management measures lead to declining levels in humans. The rising and then decreasing trend is likely because the risk management measures for these newer POPs were taken later than for the initial POPs.
- ❖ No clear trend over time could be seen for HCHs, Heptachlor, PeCB and Mirex. And, due to lack of enough data, no clear trend could be determined for PBBs (including HBB), PCNs and Endosulfan. It is not clear why no clear trend can be observed for HCHs, Heptachlor and Mirex, one reason could be the variability among data sources, but this needs to be investigated further.
- ❖ Much more monitoring data is needed for the short-chained chlorinated paraffins, since the produce and use of these compounds are known to be large and information on human exposure in the WEOG region is substantially missing. No monitoring data could be found for Chlordecone and HCBd in human milk or blood.

Recommendations

It is of great importance that the regional and national programmes evaluating time trends of both POPs and other environmental pollutants in blood and/or milk continues. This data is inevitable to be able to follow up the effectiveness of the Stockholm Convention and to see that regulations and other actions taken in order to reduce the exposure to POPs are purposive and efficient. The timing of the WHO/UNEP milk survey could be better synchronized with the cycle of the effectiveness evaluation of the Stockholm Convention to allow that the Global Monitoring

Plan work can use the latest available data. Also, participation from more countries in this survey would increase its representativeness.

In order to be able to use monitoring data to evaluate and compare levels and trends between countries, regions and continents it is crucial to be able to retrieve as much meta data and background information as possible (*e.g.* sampling strategies, information on cohorts and participants, analytical methods and statistical parameters) and to have proper identification of substances. To store monitoring data in searchable and open databases are preferable. Biobanking samples for later analysis is also very useful since it makes it possible to go back and produce time trends for new pollutants also in the future.

Very little data could be found for several of the newer POPs. In order to be able to follow up on these substances over time, it is important to start monitoring these substances now. And, in addition to measuring the classic POPs, that are already regulated, it is important to also monitor possible substitution substances. Strategies to detect and identify exposure to new emerging pollutants i.e. “early warning systems” are also needed and this could preferably be done on both a national, regional and global level.

Acknowledgements

The authors would like to acknowledge the people who did the field work and the analytical work in labs that makes this chapter on human exposure possible to write. We would also like to thank the managers of the regional and national monitoring programs for making the data available to us. Finally, we would like to thank the Swedish Environmental Protection Agency for supporting the contribution of Linda Linderholm and financing the work of the co-authors from the Swedish Environmental Emissions Data (SMED).

5.2.3 Water

Introduction

The objective of this chapter is to summarize the current state of knowledge of the three poly-/perfluoroalkyl substances (PFAS) currently listed or proposed for listing on the Stockholm Convention in water within the WEOG countries. These substances are perfluorooctane sulfonic acid (PFOS), perfluorooctanoic acid (PFOA) and perfluorohexane sulfonic acid (PFHxS). PFHxS is proposed for listing (Stockholm Convention, 2020).

With the addition of PFOS to the Stockholm Convention, the need to have data for water became important because of the hydrophilic properties of this chemical. The inclusion of PFOA and PFHxS adds additional incentive to consider water as an important environmental compartment. Water is an important medium for regional and global transport of PFOS, PFOA and other perfluoro-alkyl acids due to their high water solubility and persistence (Yamashita et al., 2012). This is unlike the case for neutral chlorinated or brominated POPs which are hydrophobic and generally present at picogram per liter (parts per quadrillion) concentrations in background surface waters (Muir and Lohmann, 2013). Direct emissions to water from wastewater treatment plants are thought to be the main sources of PFOA, PFHxS and PFOS to surface waters globally as a result of past or continuing industrial production and ongoing use and disposal of products containing their precursors and residuals (Earnshaw et al., 2014; Wang et al., 2017; Wang et al., 2014). The emissions of these PFAS to surface waters can be directly linked to exposure of fish, fish eating birds and mammals via food-chain bioaccumulation and biomagnification. Hence knowing dissolved concentrations in the water enables prediction and validates modelling of fate and bioaccumulation of PFASs in aquatic environments.

In addition to biomagnification in wildlife, which can lead to human dietary exposure, human exposure via drinking water contamination with PFASs is a major concern in some regions (Domingo and Nadal, 2019; Post et al., 2017; US EPA, 2016). Surface waters serve as major sources for drinking water in WEOG countries and globally. Thus, while this assessment does not deal directly with drinking water, a review of available data for surface waters is timely, and provides information on potential human exposure.

This chapter reviews the results available, focusing on PFOS, PFOA and PFHxS in rivers, lakes, estuaries and oceans within the WEOG area (Western Europe, North America, Australia/New Zealand and Antarctica). Similar to the previous assessment of PFOS in water in this region (Chapter 5.2 in (UNEP, 2015b)) and in the global report (Chapter 6.3 in (UNEP, 2015a)), the focus in this report is mainly on background (non-urban) sites. Two major questions for this 2nd assessment of PFAS in water were addressed:

(1) Are concentrations of PFOS, PFOA and PFHxS declining as a result of national and international chemical management controls?

(2) What are prevailing concentrations of PFOS, PFOA and PFHxS in rivers, lakes, estuaries of WEOG countries as well as in neighboring seas (Baltic, North) and oceans (North Atlantic, Arctic Ocean, Southern Ocean)?

Sources of information and methodology

All the available peer reviewed literature which included data on PFAS in water were reviewed to June 2020. We selected results for PFOS, PFOA and PFHxS. Selection of articles was based on the inclusion of the literature previously reviewed to early 2015 (UNEP, 2015a, b) and from an extensive literature search on *EBSCO* and *Google Scholar* using combinations of milieu terms e.g. *water, river, ocean, sea* and target terms e.g. *PFAS, perfluoroalkyls, fluor*, PFOS*. Only the literature that included determination of PFAS in surface water samples were reviewed. As the focus was on background contamination levels, the articles that focused on contamination events or specifically on waste water treatment plants (WWTPs) were not retained. Nevertheless, the dataset does include sites that may be directly contaminated by industries, or close to a large urban population pool, and thus plausibly to urban WWTP effluents. A list of the articles is given in Appendix 1

While the majority of the available data for this assessment was from the peer reviewed scientific literature, additional results were obtained from government laboratories that are involved in monitoring and surveillance of contaminants in water. PFAS results that were available on the NORMAN EMPODAT database website (<https://www.norman-network.com/nds/empodat/>) were also downloaded. Results for rivers in Germany, Italy, The Netherlands, and Spain previously uploaded to the database from several agencies (Landesamt für Umwelt, Landwirtschaft und Geologie and the Federal Environment Agency (UBA-DE) in German, Association of Rhine Water Works (The Netherlands)), and the European Commission Joint Research Centre, Institute for Environment and Sustainability were included. Criteria for selection were that the studies had >60% results above reported MDLs for individual river basins. Data for PFASs in water were also available from national programs in Canada, Finland, and Australia. Regional monitoring and assessment programs within the WEOG such as HELCOM, OSPAR, MEDPOL, AMAP have generally not focused on measurements on PFASs in water. However, early measurements in lake and seawaters were made in Scandinavia under the Nordic Council of Ministers (Kallenborn et al., 2004). The International Council for Results for PFAS from the North Sea and the Baltic from 2005 to 2017 from the Exploration of the Sea (ICES) database were included (ICES, 2020).

The contaminated character of the sites was loosely assessed individually during inclusion of the reviewed articles based on the site description and on a conservative concentration threshold at 2000 ng/L for any PFASs at the site (122 sites excluded on post hoc). The remaining sites were labelled as urban/non-urban by computing the density of population in a 100 km radius (using

minimum geodesic distance from Postgis type *geography*) to the sites location and classifying with threshold 500 capita/km². The population size was taken from *Natural Earth* (naturalearthdata.com) and represented point-wise for each large city.

We also included information on analytical methodology and quality assurance in the database including method detection limits (MDLs) and brief descriptions of analytical methodology (instruments, columns, calibration, use of mass labelled standards). An important methodological aspect was the requirement that all studies would have used mass labelled internal standards of PFASs along with high performance liquid chromatography-tandem mass spectrometry (LC-MS/MS) instrumentation or high resolution MS. This had become standard methodology by the mid-2000s (Taniyasu et al., 2005; Yamashita et al., 2004) and is also described in an International Standards Organization method for PFOS and PFOA in water (ISO, 2009). Thus some early studies for surface waters were not included although it has to be acknowledged that they were important for identifying the issue of water contamination in the first place (eg Hansen et al., 2002; Moody et al., 2002).

Data from the selected literature were inserted in a Postgis database following closely the structure of the Global Monitoring Plan Data Warehouse of the Stockholm Convention on POPs (pops-gmp.org). Details on the database structure are given in the supplementary information. In total, there were 11270 individual PFAS measurements (6688 over method detection limits (MDL)) available from 3002 sites based on results from 110 peer reviewed papers and reports. These represented 3812 measurements (2598 detections over MDL) for PFOS, 2711 (1250) for PFHxS and 4747 (2840) for PFOA (Table 5.2.50). The period 2015-2019 saw 2035 results for PFOS, PFHxS and PFOA reported from urban and non-urban sites (Table 5.2.50) thus enabling a substantial update of results for PFAS in water relative to the previous report which was based on results to 2014 (UNEP, 2015b). Appendix 2 provides the list of individual samples, locations, DOIs for the articles, and concentrations of PFOS, PFHxS and PFOA.

Table 5.2.50. Summary of data available for PFAS in water within the WEOG region (to June 2020) including samples collected during 2015-2019

	Urban and non-urban sites (3002)			Non-urban sites (2435)		
	All 2000-2019	>MDL	2015-19	All measurements	>MDL	2015-19
PFOA	4747	2840	645	3485	2211	535
PFOS	3812	2598	808	2979	2040	697
PFHxS	2711	1250	582	2171	1087	472
Total	11270	6688	2035	8635	5338	1704

Medians and median absolute deviations (MAD) from the medians, were calculated for PFOS, PFHxS or PFOA for inland waters, the Great Lakes, coastal waters and open ocean waters. Median concentrations were used to compare spatial and temporal trends for samples collected during the periods 2000–2009, 2010–2014 and 2015–2019. It must be emphasized that the calculation of % declines or increases using medians provides only qualitative assessment of trends.

Concentrations reported as <MDL were substituted with $MDL \times \sqrt{2}/2$ as recommended by Antweiler (2015) rather than being omitted from the dataset. Due to the wide range of detection limits for PFOS, PFHxS and PFOA reported among the studies and the potential statistical biases of multiple MDLs, we decided to use single MDLs for each analyte taking into account the distribution of reported concentrations. The selected MDLs (PFOS and PFHxS = 0.002 ng/L; PFOA = 0.005 ng/L) were taken from studies on lake waters (De Silva et al., 2011) and seawater (Benskin et al., 2012; Yamashita et al., 2008) that reported MDLs in the low pg/L range. These MDLs are readily achievable using 1 L samples with LC-MS/MS analysis. The substituted values for $MDL \times \sqrt{2}/2$ are provided in Appendix 2.

The analytes of interest

The main focus is on PFOS, PFOA and PFHxS which are present as ionized species at prevailing pH of most natural waters. However there are many PFOS precursors (see air chapter) which are known to degrade to perfluoroalkyl acids in water, especially during wastewater treatment (Martin et al., 2010; Schultz et al., 2006). Several studies of PFASs in water have included perfluorooctane-sulfonamide (FOSA). A series of perfluoro-sulfonamides and -sulfamidoethanols have also been included in measurements of seawater (Xie et al., 2013). Thus this review also includes brief discussion of these precursors. PFOS isomers are also of interest as indicators of partitioning and transformation of precursors and are discussed briefly.

Results for rivers, lakes, estuaries and oceans

Inland waters (rivers and lakes)

Median concentrations, statistical dispersion (MAD divided by median) and sample numbers for PFOS, PFHxS and PFOA on rivers and lakes within Western Europe, Canada/USA and Australia are provided in Table 5.2.51. Ranges are shown with arithmetic means and medians in Figure 5.2.60. Maps showing all sampling sites and range of concentrations within major river watersheds are provided in Appendix 3. There are 1992 measurements for PFOS, 3277 for PFOA, and 1384 for PFHxS. In general median concentrations of PFOA, PFHxS and PFOS at combined urban and non-urban influenced sites were higher than at non-urban sites (Table 5.2.51A). MAD/median values (a measure of relative data dispersion around the median) were similar in both combined urban and non-urban results (medians of 0.45 and 0.47, respectively) indicating factors other than proximity to high population density were affecting the variation in

concentrations. Given the differences in concentrations the groupings of sites are discussed separately.

Considering all inland water sites, median concentrations of PFOS were 2.0-fold higher in Europe than Canada/USA for the period 2000-09 but for more recent measurements (2015-19) North American medians were 4.0 fold higher (Table 5.2.51A; Fig. 5.2.60). The same differences for PFOS were observed for non-urban sites with 3.3 fold higher concentrations for the period 2015-19 in Canada/USA vs Europe (Table 5.2.51B). Similarly median concentrations for PFOA were 2.9 fold higher in Western Europe waters than in Canada/USA for the 2000-09 period but 3-fold lower for 2015-19. PFHxS concentrations were distinctly different for all time periods with higher medians in Western Europe than North America in 2000-09 and in 2015-2019.

Median PFOS concentrations in non-urban Western Europe sites declined 70% between samples collected in 2000-09 and in 2015-19 but were 227% higher in Canada/USA waters over the same period. MAD/median values at non-urban sites for PFOS in 2000-09 and 2015-19 were similar for Western Europe (0.88-1.0) and for Canada/USA (0.87-1.0) indicating similar dispersion of < 2 fold for individual data for these time periods. Median PFOA in 2015-19 was 70% lower than in 2000-09 in Europe and but much higher (352%) in Canada/USA samples. Median PFHxS concentrations at non-urban sites in Europe increased 72% between 2000-09 and 2015-19 and had a huge increase in North American waters over those time periods (Table 5.2.51B). The increased PFOA, PFHxS and PFOS reflected reports from locations were not the same as those sampled in earlier time periods. For example they include surveys of relatively small water bodies in rural surface waters in New Jersey (Procopio et al., 2017), Rhode Island and New York (Zhang et al., 2016b), and in Washington State (Mathieu and McCall, 2017). The results from these rural US sites suggest continued significant PFAS emissions to the aquatic environments in both rural and urban regions.

Table 5.2.51. Summary of available data for PFOA, PFHxS and PFOS in inland waters of Western Europe, Canada/USA and Australia (ng/L). A. All sites and B. Non-urban (background) sites. Comparison of medians, data dispersion (MAD/median) and number of measurements [#] by region, pre 2010, 2010 to 2014, and 2015-2019, where available.

		PFOA			PFHxS			PFOS		
	period	median	MAD/ Median	#	median	MAD/ Med	#	median	MAD/ Med	#
A. All sites within a region										
W. Europe	2000 - 2009	2.67	0.99	1053	0.07	0.98	378	1.82	1.00	888
	2010 - 2014	3.00	1.00	1537	0.001	0.00	451	0.90	1.00	534
	2015 - 2019	0.63	0.84	339	0.20	0.99	282	0.50	0.86	381
% change (2000-09 vs 2015-19)		-76%			184%			-72%		
Canada/USA	2000 - 2009	0.91	1.00	429	0.007	0.80	357	0.90	1.00	425

	2010 - 2014	0.95	1.00	263	0.001	0.00	199	0.03	0.94	263
	2015 - 2019	2.09	1.00	117	0.68	1.00	117	1.71	1.00	117
% change (2000-09 vs 2015-19)		130%			9577%			90%		
Australia	2010 - 2014	1.20	0.83	13	2.50	0.90	13	3.10	0.74	14
	2015 - 2019	0.50	0.00	14	0.50	0.00	14	0.001	0.00	134
% change (2010-14 vs 2015-19)		-58%			-80%			-100%		
B. Non-urban sites only										
W. Europe	2000 - 2009	2.00	0.98	685	0.12	0.99	312	1.17	0.97	624
	2010 - 2014	2.58	1.00	964	0.001	0.00	270	0.72	1.00	287
	2015 - 2019	0.60	0.83	334	0.20	0.99	277	0.48	0.88	376
% change (2000-09 vs 2015-19)		-70%			72%			-60%		
Canada/USA	2000 - 2009	0.51	0.99	340	0.008	0.83	293	0.52	1.00	336
	2010 - 2014	0.34	0.99	180	0.005	0.72	119	0.03	0.95	180
	2015 - 2019	2.29	0.89	66	0.93	0.76	66	1.70	0.87	66
% change (2000-09 vs 2015-19)		352%			11556%			227%		
C. Australia – all sites										
Australia	2010 - 2014	0.45	0.58	9	0.49	1.00	9	1.10	0.92	10
	2015 - 2019	0.50	0.00	14	0.50	0.00	14	0.001	0.00	134
% change (2010-14 vs 2015-19)		11%			2%			-100%		

Medians concentrations for surface waters in Australia for the period 2010-14 and 2015-20 are compared in Table 5.2.51C. Results for 2010-14 are based on three published studies which included urban rivers and creeks (Allinson et al., 2019; Gallen et al., 2014; Thompson et al., 2011b). Allinson et al. (2019) determined PFAS in water samples collected in 2012 from rivers and creeks in and around the city of Melbourne (Victoria). PFOS, PFHxS and PFOA were the most prominent PFAS. Gallen et al. (2014) studied the trends of PFAS in the Brisbane River system (Queensland) during a severe flooding event in 2011. PFOA was the most prominent PFAS at two remote sites upstream of the urban area (0.31-0.45 ng/L). However, PFOS and PFHxS were present at higher concentrations in flood water (medians, 4.8 and 2.8 ng/L, respectively) than PFOA (1.9 ng/L) within the urban area and also in the Moreland Bay estuary. Earlier studies by Thompson et al. (2011b) showed that PFOS, PFHxS and PFOA were the top 3 PFASs in waters of the upper Parramatta River, the main tributary of Sydney Harbour. Lower concentrations of PFOS, PFHxS and PFOA were reported in 2015-20 in a survey of surface waters near Melbourne by the Australian Department of Defence (Australia Defence, 2018). A study of legacy pollutants in water in the State of Victoria (Sardiña et al., 2019) found PFOS, PFOA and PFHxS at <MDL concentrations at background and agricultural sites (<1 ng/L). An extensive survey of surface waters of urban and rural sites in Queensland (Baddiley et al., 2020). PFOS was detected (<0.1 to 0.6 ng/L) in 18 of 29 rivers and near shore marine water sites that

were sampled every two months over a 12 month period (2019-20). Maps showing all sampling sites and range of concentrations within major river watersheds in Australia are shown in Appendix 3. Drinking water (potable water) was extensively surveyed by Thompson et al (2011a) at 34 locations across Australia, including capital cities and regional centers, indicating the presence of low ng/L concentrations of PFOS, PFHxS and PFOA, similar to other WEOG countries (Domingo and Nadal, 2019).

Information is much more limited for New Zealand. Recent studies of PFAS contamination near military bases in New Zealand have included limited background surface water sampling indicating contamination resulting from use of fire-fighting foams showing the presence of PFOS, PFHxS and PFOA at low ng/L concentrations (NZDF 2019a,b). Surface waters collected in two creeks flowing near the Woodburne base (near Blenheim NZ) had median concentrations for samples above the 1 ng/L reporting limit of 11, 19 and 2.6 ng/L for PFOS, PFHxS and PFOA, respectively (NZDF 2019b). Results from surface waters on the Ohakea base (Bulls NZ) had medians of 205, 103 and 69 ng/L for PFOS, PFHxS and PFOA, respectively, reflecting contribution from a plume of contaminated groundwater to originating on the base. Due to limited information on sampling locations these results from New Zealand have not been included in the current database for this report.

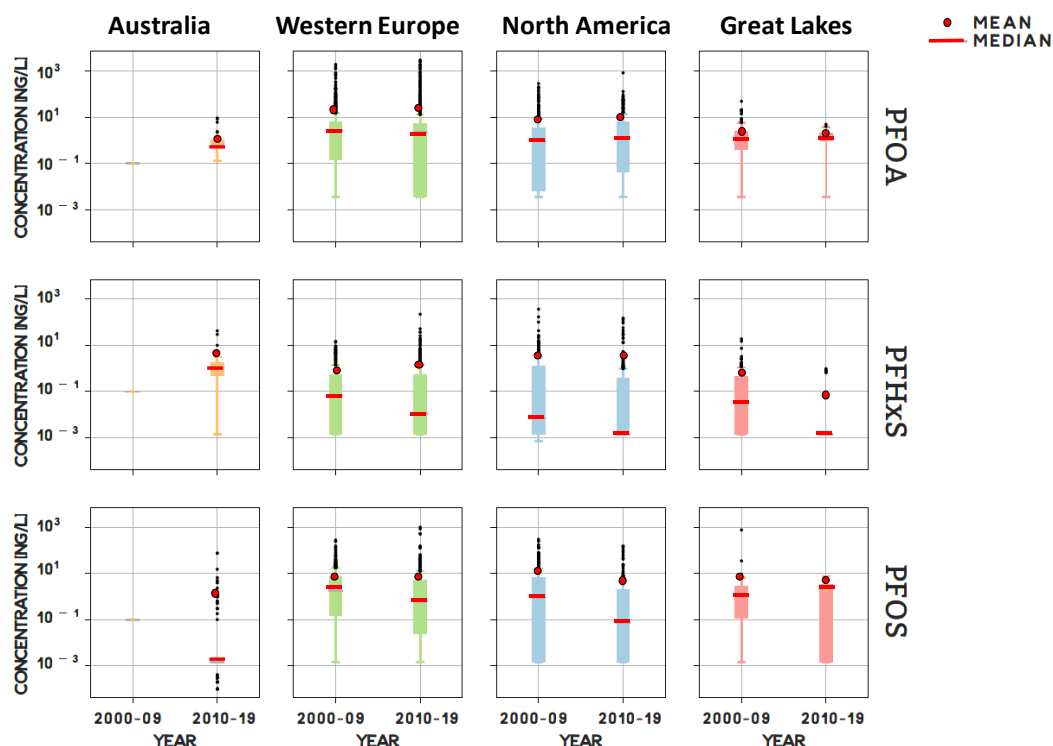


Figure 5.2.60. PFOA, PFHxS and PFOS in non-urban river and lake waters in Australia, Western Europe, North America (Canada/USA) and Great Lakes (ng/L) for 2000-09 and 2010-19. Vertical lines represent the range of concentrations. Red colored central bars and circles represent median and arithmetic means, respectively. The boxes comprise the interquartile range

(IQR); whiskers show 1.5 IQR.

The North American Great Lakes are a region with a large number of measurements of PFASs in water and one of the few regions with enough data to evaluate temporal trends. Thus they are discussed separately from other inland waters in Canada/USA. PFASs in approximately 256 surface water samples have been reported since 2002 with the majority from offshore locations. Median concentrations and MAD/median values for PFOA, PFHxS and PFOS for all sites in the Great Lakes, and also for non-urban sites, are provided in Table 5.2.52, while arithmetic means and medians are shown in Figure 5.2.60. With urban sites included, median concentrations of PFOA and PFOS in 2015-19 were 37% and 159% higher, respectively than in the period 2000-09, while PFHXS was 95% lower due to non-detect levels in 2015-19 (Table 5.2.52A). For non-urban sites median PFOA and PFHxS were lower in 2015-19 while PFOS was higher (Table 5.2.52A and Figure 5.2.60). However, comparison of only non-urban sites was problematic because of a high percentage of results that were less than MDLs (2000-09 =76%, 2010-2014 = 60% and 2015-2019 =51%), especially in Lakes Superior, Huron and Michigan. These < MDL results were substituted by $MDL \cdot \sqrt{2}/2$.

Table 5.2.52. Summary of available data for PFOA, PFHxS and PFOS from (A) all sites (open and coastal waters) and (B). Open water/non-urban sites within the Great Lakes. Comparison of medians, data dispersion (MAD/median), and number of measurements [#] by region, pre 2010, 2010 to 2014, and 2015-2019, where available.

		PFOA			PFHxS			PFOS		
	period	median	MAD/ Median	#	median	MAD/ Median	#	median	MAD/ Median	#
A. All sites within the region										
Great Lakes	2000 - 2009	1.19	0.77	167	0.028	0.95	164	1.00	1.00	165
	2010 - 2014	1.54	0.34	55	0.001	0.00	54	0.001	0.00	55
	2015 - 2019	1.63	0.54	36	0.001	0.00	36	2.58	0.66	36
% change (2000-09 vs 2015-19)		37%			-95%			159%		
B. Open lake/non-urban sites										
Great Lakes	2000 - 2009	0.696	0.73	130	0.028	0.95	127	0.34	1.00	127
	2010 - 2014	1.34	0.46	33	0.001	0.00	32	0.001	0.00	33
	2015 - 2019	0.004	0.00	21	0.001	0.00	21	2.06	1.00	21
% change (2000-09 vs 2015-19)		-99%			-95%			504%		

Values for MAD/median for PFOS, PFHxS and PFOA had overall smaller values (median 0.5) in Great Lakes waters of the data compared to river waters in Canada/USA (median 1.0) indicating a narrower range of dispersion (Table 5.2.52). Zero values for MAD (and

MAD/median) occurred due to high numbers of the same replacement values used for the <MDLs.

Lakes Superior, Huron and Michigan had the lowest concentrations of PFOA, PFHxS and PFOS of the five Great Lakes while Lakes Ontario and Erie had the highest (Figure 5.2.61). Trends are clearer in Lakes Erie and Ontario where the frequency of MDLs was <10% for PFOA and PFOS. Despite the overall decline over the 13 to 15 year period the downward trend is not smooth. For example, all three PFAS showed higher concentrations in Lake Erie in 2008 while declining to lower levels by 2012. PFOS concentrations were relatively uniform in surface waters within each lake within a given season (De Silva et al., 2011; Myers et al., 2012; Scott et al., 2010). Depth profiles in Lake Superior showed similar concentrations in bottom waters and at the surface (Scott et al., 2010).

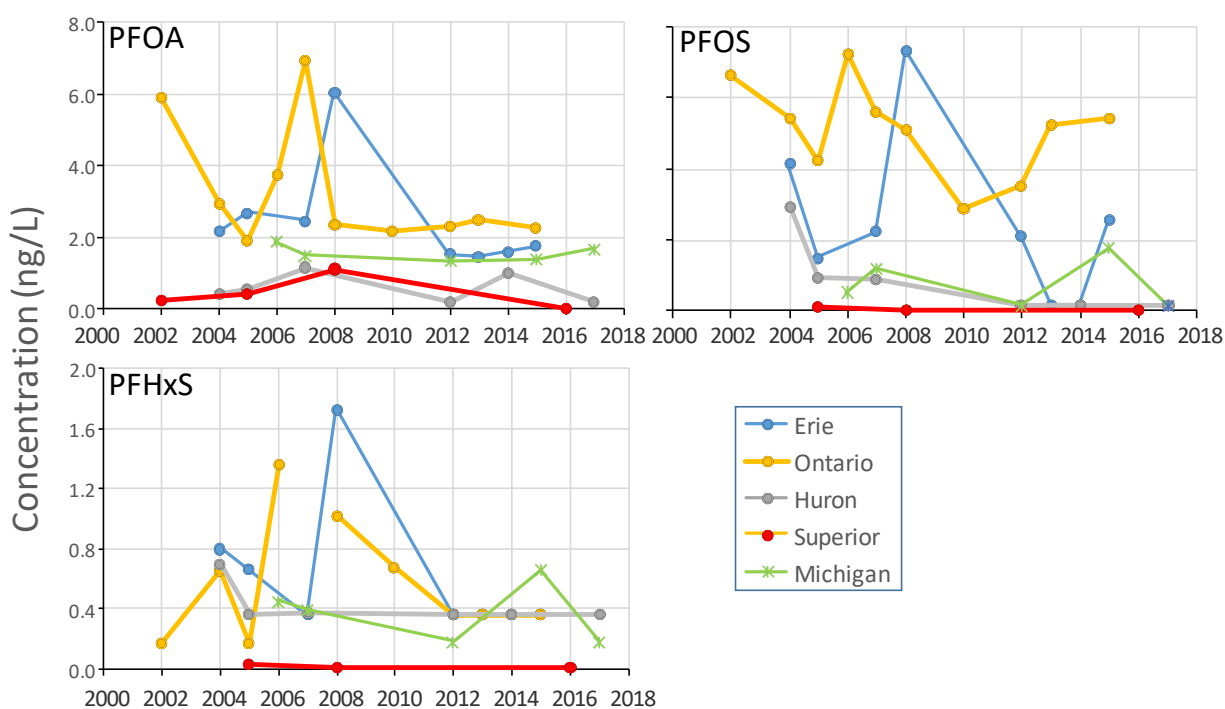


Figure 5.2.61. PFOS, PFHxS and PFOA concentrations (ng/L) from all locations sampled in the North American Great Lakes for 2002 to 2017. Symbols are annual median concentrations in surface waters (~1-4 m depth). The results are based on studies by Furdui et al. (2008), DeSilva et al. (2011), Sinclair et al. (2006), Scott et al. (2010), Myers et al. (2012) and Gewurtz et al. (2019).

The largest PFAS datasets for Western European rivers are for the Ebro, the Elbe, and the Rhine, and their tributaries. Detailed studies were conducted in the period 2005 to 2016 on all three including estimates of loadings in the Rhine and Elbe (Ahrens et al., 2009c; Paul et al., 2012; Zhao et al., 2015). The Elbe and Rhine rivers have had multiple detailed geospatial studies of PFOS and other PFASs (Ahrens et al., 2009b; Ahrens et al., 2009c) (Loos et al., 2008a; Möller et

al., 2010; Quinete et al., 2009; Skutlarek et al., 2006; Zhao et al., 2015) as well by monitoring agencies in Germany and the Netherlands (see methods section). Studies on the Ebro have included samples from 2007 to 2011 (Ericson et al., 2007; Llorca et al., 2012; Lorenzo et al., 2016). The medians and range of concentrations of PFOA and PFHxS were similar in all 3 rivers while PFOS median in the Rhine was about 3 fold higher than the Elbe (Figure 5.2.62A). The time interval of sampling for the non-urban sites ranged from 3 years on the Rhine to 10 years on the Elbe which is rather short for assessing temporal trends. PFOS concentrations declined in the Elbe as of about 2006, coinciding with phase out of most uses of PFOS-related products in Europe. A larger number of sampling years are available for the combined urban and non-urban sites on the Ebro, Elbe and Rhine (Figure 5.2.62B). A 4-fold decline of median concentrations

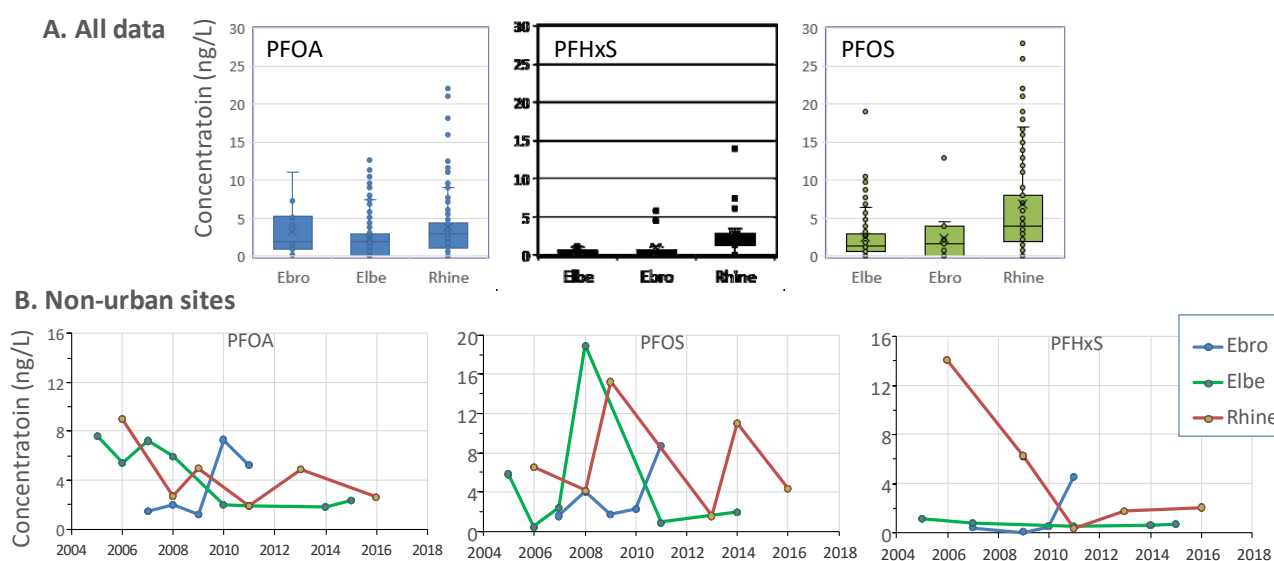


Figure 5.2.62. A. Box and whisker plots for PFOA, PFHxS and PFOS in the Ebro, Elbe and Rhine Rivers. Vertical lines represent the range of reported concentrations (ng/L). Central bars represent the median of the concentrations, the boxes comprise the interquartile range (IQR) while the whiskers show 1.5 IQR. B. Median concentrations at non-urban sites shown over time (years).

of PFOA and 7-fold decline of PFHxS from 2006 to 2016 occurred in the Rhine. Much lower concentrations of PFOA, PFHxS and PFOS were found in the Elbe in 2014/15 compared with 2007-08. However in the Ebro, higher median concentrations were found in 2011 compared to 2007-2009. No previous studies on temporal trends of PFASs in European river waters were found. However, Earnshaw et al. (2014) found no change in PFOS concentrations in the Aire and Calder Rivers in northeast England between 2010 and 2013, based on similar profiles in concentrations with distance downstream.

Reports from contamination of surface waters as a result of accidental or deliberate release of PFOS containing fire-fighting foams (Ahrens et al., 2015; Dauchy et al., 2017; De Solla et al., 2012; Moody et al., 2002; Rumsby et al., 2009) were not included in this assessment. However, understanding the fate of PFOS released in these events is important because these contamination incidents could influence prevailing concentrations in rivers and confound interpretation of loadings and of temporal trends.

Coastal waters

Detailed measurements have been made for PFAS in the Baltic Sea (Ahrens et al., 2010a; Kallenborn et al., 2004; Kirchgeorg et al., 2010), the North Sea (Ahrens et al., 2010a; ICES, 2020; Theobald et al., 2011) and the Mediterranean Sea (Brumovský et al., 2016; Schmidt et al., 2019; Yamazaki et al., 2019) as well as for coastal waters (<200 m depth) on both sides of the North Atlantic (Figure 5.2.63). In general median concentrations of PFOA, PFHxS and PFOS at combined urban and non-urban influenced sites and non-urban only sites were similar. Data dispersion was low (medians of MAD/median values for all sites vs non-urban sites of 0.37 and 0.32, respectively, compared with rivers in Western Europe and North America (Table 5.2.53A, B). Lower median concentrations of PFOA were found in non-urban samples from 2015-19 compared to 2010-14 in the Mediterranean (-95%) while PFHxS was at detection limits and PFOS was 110x higher due to non-detect levels being reported in earlier studies (Table 5.2.53B). Due to relatively high sample numbers for all 3 time periods comparisons of 2000-09 with 2015-19 were possible for the Baltic and the North Sea. PFOA and PFOS concentrations were 31% and 81% lower in 2015-19 than 2000-09 in the Baltic, however in the North Sea higher median PFOA was found (44%) while PFOS declined (-78%). PFHxS medians were higher in 2015-19 in the Baltic and the North Sea. Including urban influenced sea waters (Table 5.2.53A) showed similar differences for PFOA PFHxS and PFOS between earlier sampling and medians from 2015-19. In coastal Atlantic waters, for which comparisons were between 2000-09 and 2010-14 due to lack of results for 2015-19, all 3 PFAS showed modest declines (14-33%) (Table 5.2.53A). Also, as mentioned previously, most of the variation in medians can be explained by new geographic locations being sampled.

Less spatially comprehensive results are available from a large number of other studies of estuaries and included coastal waters. Lowest concentrations of PFOS were found in coastal waters of King Georg Is (Antarctica) and in several Arctic locations including Mackenzie Bay (Southern Beaufort Sea), northwestern Spitzbergen, and coastal East Greenland. Highest concentrations were found in urban influenced estuary and coastal waters e.g. in the Scheldt (Rhine River delta), near the Elbe, in the northern Adriatic, Puget Sound, and San Francisco Bay.

Table 5.2.53. Summary of available data for PFOA, PFHxS and PFOS in coastal seas of Europe (ng/L). A. All sites and B. Non-urban (background) sites. Comparison of medians, data dispersion (MAD/median), and number of measurements [#] by region, , pre 2010, 2010 to 2014, and 2015-2019, where available.

region	Period	PFOA median	MAD/ Median	#	PFHxS median	MAD/ Median	#	PFOS median	MAD/ Median	#
A. All sites										
Mediterr'n Sea	2010 - 2014	0.07	0.95	31	0.001	0.00	31	0.02	0.92	31
	2015 - 2019	0.004	0.00	12	0.001	0.00	12	0.16	0.23	12
% change (2000-09 vs 2015-19)		-95%			0%			756%		
Baltic Sea	2000 - 2009	0.39	0.15	89	0.09	0.56	89	0.20	0.30	89
	2010 - 2014	0.52	0.26	18	0.42	0.40	18	0.73	0.29	18
	2015 - 2019	0.27	0.15	45	0.21	0.33	45	0.04	0.21	45
% change (2000-09 vs 2015-19)		-31%			133%			-81%		
North Sea	2000 - 2009	0.31	0.82	19	0.06	0.97	19	0.19	0.75	19
	2010 - 2014	1.30	0.53	125	0.32	0.50	125	0.42	0.69	125
	2015 - 2019	0.45	0.33	35	0.14	0.21	35	0.04	0.42	35
% change (2000-09 vs 2015-19)		47%			155%			-80%		
North Atlantic coastal	2000-2009	0.11	0.64	104	0.03	0.67	64	0.07	0.43	115
	2010-2019	0.08	0.13	24	0.02	0.50	24	0.06	0.17	24
% change (2000-09 vs 2010-19)		-27%			-33%			-14%		
B. Non-urban sites										
Mediterr'n Sea	2010 - 2014	0.07	0.95	19	0.001	0.00	19	0.001	0.00	19
	2015 - 2019	0.00	0.00	12	0.001	0.00	12	0.16	0.23	12
% change (2000-09 vs 2015-19)		-95%			0%			10971%		
Baltic Sea	2000 - 2009	0.39	0.15	89	0.09	0.56	89	0.20	0.30	89
	2010 - 2014	0.52	0.26	18	0.42	0.40	18	0.73	0.29	18
	2015 - 2019	0.27	0.15	45	0.21	0.33	45	0.04	0.21	45
% change (2000-09 vs 2015-19)		-31%			133%			-81%		
North Sea	2000 - 2009	0.31	0.78	18	0.05	0.97	18	0.17	0.68	18
	2010 - 2014	1.30	0.53	125	0.32	0.50	125	0.42	0.69	125
	2015 - 2019	0.45	0.33	35	0.14	0.21	35	0.04	0.42	35
% change (2000-09 vs 2015-19)		44%			180%			-78%		

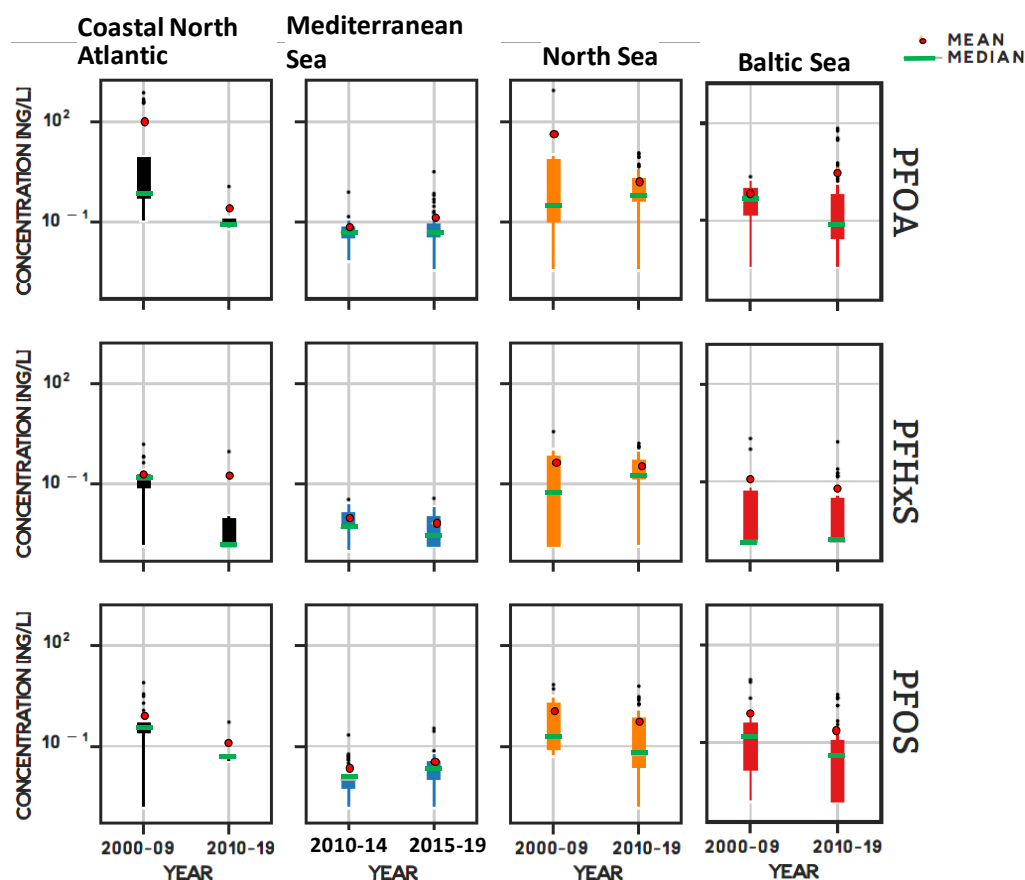


Figure 5.2.63. PFOA, PFHxS and PFOS in European coastal waters and seas (ng/L). Vertical lines represent the range of reported concentrations for 2000-09 and 2010-19. Vertical lines represent the range of reported concentrations. Green colored central bars and red circles represent median and arithmetic means, respectively. The boxes comprise the interquartile range (IQR) while the whiskers show 1.5 IQR.

Ocean waters adjacent to WEOG region

Detailed datasets for PFOA, PFHxS and PFOS are available for the North and Central Atlantic, the Eastern/ North Pacific Ocean, and the Arctic Ocean, while limited measurements have been made in Antarctic coastal waters. Compared to freshwaters and estuaries, PFAS concentrations are very low. Lowest mean concentrations (<10-20 pg/L) are reported for the Greenland Sea and the Labrador Sea, as well as in the Southern Ocean near Antarctica (Table 5.2.54). Most sampling in the North Atlantic and Arctic Ocean has been done prior to 2015 limiting the comparisons for the most recent time period for global monitoring. Thus the comparisons in Table 5.2.54 are limited to two time periods, 2000-09 and 2010-19. Median concentrations of PFOA in ocean waters showed little change (+5%) in the Arctic Ocean for the period 2010-19 compared to 2000-09 but had much large values in the North Pacific (1000%), Central Atlantic (42%) and Antarctica Ocean (16x) (Table 5.2.54). Median PFOS concentrations were lower in the Arctic Ocean and North Atlantic (35-57%, respectively) but higher in the Central Atlantic and North Pacific (164% and 25%, respectively) while lower values were reported in Antarctica for 2010-19. PFHxS concentrations in 2010-19 were higher in the Central Atlantic and North

Pacific but lower in the Arctic Ocean (-35%) and North Atlantic (-57%). Timing and locations of ocean cruises in polar waters is particularly critical as melting snow and riverine inputs have been shown to influence observed concentrations (Benskin et al., 2012; Li et al., 2018).

Table 5.2.54. Summary of median concentrations (ng/L) and available data for PFOA, PFHxS and PFOS in ocean sampling sites of the Arctic Ocean, North Atlantic, North Pacific, Central Atlantic, and Antarctic waters. Comparison of medians, data dispersion (MAD/median), and number of measurements [#] by region, pre 2010 and 2010 to 2019.

		PFOA			PFHxS			PFOS		
Region	period	median	MAD/ median	#	median	MAD/ median	#	median	MAD/ median	#
Arctic Ocean	2000 - 2009	0.05	0.40	51	0.005	0.71	49	0.010	0.50	51
	2010 - 2019	0.05	0.35	60	0.003	0.56	44	0.021	0.52	70
% change (2000-09 vs 2010-19)		5%			-35%			110%		
North Atlantic	2000 - 2009	0.10	0.66	105	0.028	0.86	65	0.027	0.95	116
	2010 - 2019	0.08	0.17	24	0.012	0.82	24	0.062	0.21	24
% change (2000-09 vs 2010-19)		-23%			-57%			130%		
Central Atlantic	2000 - 2009	0.023	0.84	76	0.004	0.36	44	0.021	0.93	84
	2010 - 2019	0.032	0.89	39	0.007	0.81	39	0.054	0.95	40
% change (2000-09 vs 2010-19)		42%			87%			164%		
North Pacific	2000 - 2009	0.051	0.39	27	0.003	0.33	27	0.016	0.19	27
	2010 - 2019	0.56	0.78	49	0.007	0.80	50	0.020	0.55	121
% change (2000-09 vs 2010-19)		1000%			133%			25%		
Antarctic seas	2000 - 2009	0.004	0.00	2	0.001	0.00	2	0.009	0.17	2
	2010 - 2019	0.061	0.94	12	0.001	0.00	5	0.001	0.00	8
% change (2000-09 vs 2010-19)		1631%			0%			-85%		

The transect from Western Europe (north western France) to West Africa (Figure 5.2.64) is the most thoroughly sampled (Ahrens et al., 2009a; Ahrens et al., 2010b; Benskin et al., 2012; Zhao et al., 2012). These waters are influenced by the south-flowing Canary Current which carries PFAS from continental Europe. The results from 2010-14 from this region could serve a baseline for future temporal trend monitoring along the same cruise transect.

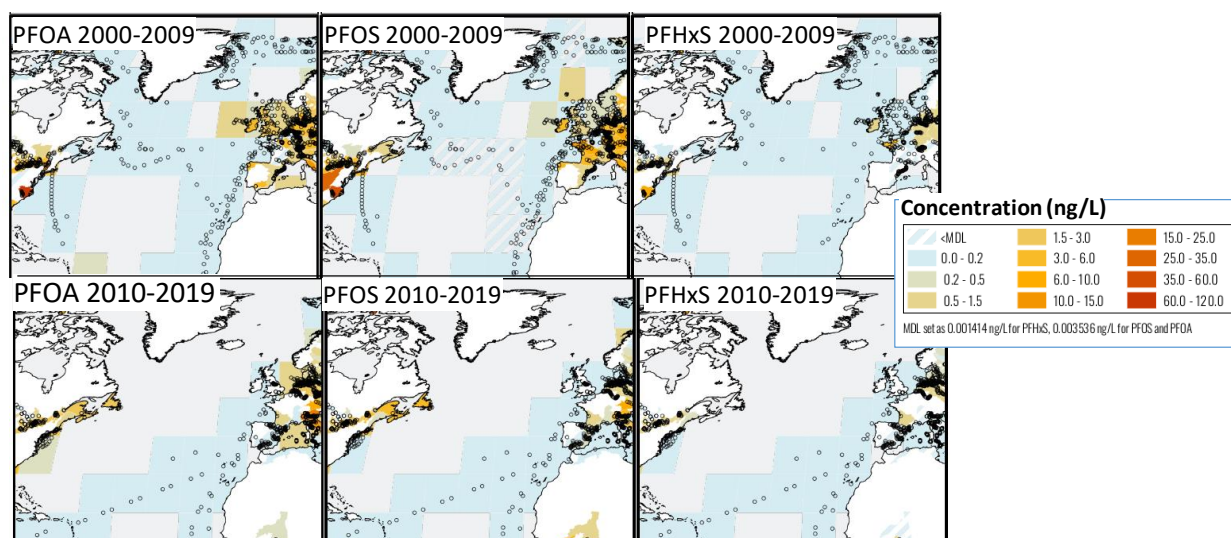


Figure 5.2.64. PFOA, PFOS and PFHxS in open ocean waters of the North Atlantic and Arctic Ocean as well as open waters of the North Sea and the Baltic (ng/L). Range of concentration for river basins of tributaries to the North Atlantic are also shown. Small circles represent sampling stations. Color shaded areas give the range of concentrations for surface waters in the oceans, seas and major watersheds. Median concentrations and MAD/median are given in Table 5.2.54.

Discussion of study design and analytes

PFOS, PFHxS and PFOA precursors

The majority of water measurements have been for total PFOS (usually analysed along with other perfluoroalkane sulfonates) and PFOA, usually with other perfluorocarboxylic acids (PFCAs). However a growing number of studies have included precursors ie substances that eventually break down to PFOS, PFHxS and PFOA in waste water treatment or in the natural environment. The perfluorooctanesulfonamide ethanols (FOSEs) and perfluoro-octanesulfonamide (FOSA), the amide derivative of PFOS, have been shown to degrade abiotically (D'Eon and Mabury, 2007; Martin et al., 2006) to PFOS and PFOA.

Median concentrations of FOSA in seawater for the period 2005-2014 were about 60% of those for PFOS (15 pg/L vs 20 pg/L, respectively) and there is also a much more variation in FOSA concentrations among studies along the same cruise transects. In freshwater and estuarine environments, FOSA appears to represent a smaller proportion of PFOS-related substances, with median values that are about 5 (rivers) to 12% (estuary) of PFOS medians. Of more than 1840 measurements of FOSA in Western European river waters in the NORMAN EMPODAT database only two were above reporting limits (0.2-7 ng/L)

Other potential PFOS and PFHxS precursors include the N-methyl and N-ethyl-perfluoroalkane sulfonamides (MeFOSA & EtFOSA), and N-methyl and N-ethyl perfluoroalkane

sulfonamidoethanols (MeFOSE & EtFOSE) as well as N-methyl perfluorooctane-sulfonamidoacetic acid (N-MeFOSAA), and N-ethyl perfluorooctanesulfonamidoacetic acid (N-EtFOSAA). Xie et al (2013) detected four PFOS precursors, MeFOSA, EtFOSA, MeFOSE, and EtFOSE in the North Sea at concentrations of <0.1-2.6, <0.1-5.1, <0.1-35 and <0.2-17 pg/L, respectively. The highest concentrations were in the estuary of the Weser and Elbe rivers and a decreasing concentration profile was found towards the central part of the North Sea. Total concentrations were, however, about 10-100 times lower than concentrations reported for FOSA and PFOS in the eastern North Sea near the Elbe. Möller et al. (2010) also determined MeFOSA, EtFOSA, MeFOSE, and EtFOSE in the Rhine River however concentrations were < 1 ng/L and not reported. Cai et al. (2012) also detected EtFOSA, MeFOSE, and EtFOSE in seawater in the Bering Sea at average concentrations ranging from 21-28 pg/L which were similar to PFOS concentrations although lower than FOSA but these compounds were < MDL of ~12 pg/L in the Beaufort Sea and Chukchi Sea in 2010. Zhang et al. (2016a) detected N-MeFOSAA and N-EtFOSAA in urban and rural waters of Rhode Island and the New York metropolitan area at concentrations generally <5% of PFOS and PFHxS. Overall, concentrations of these PFOS precursors in WEOG surface waters are not very significant from a mass balance perspective. This may explain the lack of measurements of the FOSAs and FOSEs for samples collected during the period 2015-19.

Many possible precursors of PFOA have been in commercial use such as the widely used polyfluoroalkyl phosphoric acid diesters (diPAPs), however, measurements in water are limited. Gebbink et al. (2016) detected the 6:2 diPAP in Baltic Sea water but not the 8:2-diPAP. Similarly Zheng et al. (2017) found that 6:2 diPAP was the predominant diPAP in water from the Yangtze River Delta in China while 8:2-diPAP was present at 100-fold lower levels than PFOA.

Analyses using the total oxidizable precursor (TOP) method (Houtz and Sedlak, 2012), total organic fluorine (Miyake et al., 2007) as well as ¹⁹F-NMR (Moody et al., 2001) suggest that there are other perfluorinated substances in surface waters and waste waters. Houtz and Sedlak (2012) showed that oxidative treatment of urban stormwater runoff samples produced considerable amounts of PFCAs including PFOA due to the presence of precursors including 8:2 diPAP and 8:2 fluorotelomer sulfonate. In a survey of Canadian surface waters D'Agostino and Mabury (2017) detected precursors of PFHxS (perfluorohexane sulfonamide and C6 perfluoroalkane sulfonamido amphoteric) and PFOA (fluorotelomer alkylbetaines, fluorotelomer betaines with 8 or 9 fluorocarbon chains). Using total organic fluorine analysis, D'Agostino and Mabury also noted that 36–99.7% of the total organofluorine in surface water samples was not measured in the targeted analysis. Some locations were influenced by past use of Aqueous Film Forming Foams (AFFFs) while others, mainly suburban and rural sites, had no known sources. The limited number of studies using total organic fluorine or TOP assays suggest that there are additional PFAS sources in surface waters and other aquatic media such as waste water effluents

and sediments which need to be characterized in order to get a full understanding of prevailing levels and trends of PFOS, PFHxS and PFOA.

PFOS isomer specific analysis

The production of PFOS by electrochemical fluorination (ECF) results in both linear and branched C₈F₁₇-containing sulfonic acid isomers (Benskin et al., 2010). Isomer profiling is particularly useful for PFCAs because production of PFOA and related compounds has been both from ECF and telomerization processes. However for PFOS, only ECF has been used so isomer ratios are of less interest for source profiling, but do provide information on the environmental fate of PFOS. Historically commercial PFOS reportedly had a consistent isomer composition of 70 ± 1.1% linear (L-PFOS) and 30 ± 0.8% branched (Reagen et al., 2007) and more recent studies of PFAS products in China have found the same proportions (Jiang et al., 2015). Schultz et al. (2020) have recently reviewed the literature on isomer compositions of PFOS, PFHxS and PFOA in environmental media including surface waters. They noted that branched PFOS and PFHxS can make up a significant portion of the total PFAS load in the environment, generally greater than what is reported in the technical products. However PFHxS has much less enrichment of branched isomers than PFOS. This enrichment is generally not observed or easily discerned for PFOA due to the ECF and fluorotelomer sources. Almost all monitoring and surveillance studies of PFOS in water have focused on total PFOS and total PFHxS, i.e. combined measurement of the linear and branched isomers, and in this review we have chosen to discuss geospatial and temporal trends of the total isomers. The enrichment in branched isomers in lakes may be indicative of greater sorption of L-PFOS, which is more hydrophobic than branched isomers, to sediments and sinking particles. Also biotransformation of PFOS precursor products proceeds more rapidly for branched isomers (Benskin et al., 2009), which could also lead to enrichment of branched PFOS content particularly near WWTPs. Branched and linear isomers may also transport differently in the oceans due to greater sorption of the linear isomers to the surface microlayer (Johansson et al., 2019). However Johansson et al. did not find differences between branched and linear isomers when comparing lab generated surface sea spray and bulk water.

River loading estimates and temporal trend modelling for PFOS and PFOA

Several studies have estimated mass flows of PFOS and PFOA in rivers in the WEOG region as well as discharge to estuaries and seas (Ahrens et al., 2009b; Earnshaw et al., 2014; Filipovic et al., 2013; Huset et al., 2008; Loos et al., 2008b; McLachlan et al., 2007; Möller et al., 2010; Scott et al., 2009). Mass flows (kg/time (day, year)) are of interest for understanding the fate of PFOS and for assessing the influence of seasonal trends due to variations in river flow, or to discontinuous emissions by WWTPs. Combined with population density they can be used to infer per capita emissions. However, most studies performed sampling only once, annual river

discharge can only be roughly estimated unless flows are constant. Pistocchi and Loos (2009) estimated emissions of PFOS and PFOA from Western Europe based on measured river water concentrations and climatologically averaged flows. They found that PFOS and PFOA loadings (t/yr) correlated well with river basin population, however, there was a greater scatter of PFOA loadings possibly due to multiple sources, ie manufacturing as well as waste water emissions from consumer use.

Temporal and spatial trends of PFOS in the North Atlantic Ocean and transport to the Arctic were studied by Zhang et al. (2017) using a 3D ocean general circulation model for the North Atlantic. The model results are presented as a case study in Chapter xx (MacLeod et al 2020) and are briefly described here in order to compare with observed levels in the North Atlantic. Inputs of PFOS to the North Atlantic were assumed to be from rivers only because the contribution from atmospheric deposition was thought to be small based on earlier modelling by Armitage et al. (2009b). Zhang et al utilized the per capita estimates for PFOS emissions developed by Pistocchi and Loos (2009). Zhang et al. (2017) predicted PFOS concentrations of 39 ± 14 pg/L in the North Atlantic in 2010, declining slowly to 35 pg/L by 2020, assuming negligible inputs after 2010 (see Figure 5.3.11 in Chapter 5.3). Median concentrations in the North Atlantic were 49 pg/L for 2000-09 (Table 5.2.54) thus very similar to modelled results, however, including coastal areas, median PFOS was much higher at 110 pg/L (Table 5.2.53). This spatial variability is in agreement with the model which predicted concentrations ranging from 100-500 pg/L in the coastal North Atlantic (Gulf of St Lawrence, North Sea). However, the assumption of negligible emissions after 2010 appears to have been overly conservative, as measurements suggest increasing PFOS in the Central Atlantic in 2010-14 compared to 2000-09 (Table 5.2.54). Stemmler and Lammel (2010) predicted seawater concentrations of PFOA using a global coupled atmosphere-ocean circulation model. Similar to Zhang et al. they noted that knowledge of ocean circulation and its variations was crucial to understanding PFOA's large-scale distribution and fate. Stemmler and Lammel (2010) predicted surface water concentrations of 50 pg/L in the western North Atlantic (Labrador Sea) in agreement with measurements by Yamashita et al. (2005). However their emissions scenario was based on fluoropolymer manufacturing sources (to air and water) in the USA, Japan, Belgium, and Italy, only rather than on riverine inputs, and thus not reflecting extensive measurements of PFOA for rivers in Europe, North America, as well as in East Asia.

Armitage et al.(2009b) modelled PFOS concentrations in the global oceans using the CliMoChem model which divided the global environment into a series of latitudinal bands spanning 18° of latitude. Model inputs included estimates of emissions of volatile precursors (FOSEs) as well as of PFOS from manufacturing and use. Predicted PFOS concentrations for the central North Atlantic in 2010 were about 60 pg/L declining to 45 pg/L by 2020. This range of concentrations is similar to observed medians for the Central Atlantic (Table 5.2.54). Temporal trends of PFOA were modelled with BETR Global, a multimedia environmental fate model that

describes the global environment as 288 regions based on a $15^{\circ} \times 15^{\circ}$ grid, and thus higher spatial resolution than the CliMoChem model (Armitage et al., 2009a). Estimated PFOA for 2005 ranged from about 60 to 250 pg/L in the central North Atlantic and higher concentrations (250-500 pg/L) near the US East Coast, the North Sea and the Mediterranean Sea. These estimates were 2-fold higher than median concentrations of PFOA in the coastal North Atlantic but in good agreement with the median for the Mediterranean for the period 2000-09 (Table 5.2.53). Wania (2007) modelled the time course of PFOA in the Arctic Ocean using the Globo-POP model which, like CliMoChem, includes five latitudinal bands, consisting of four atmospheric layers, terrestrial surfaces, and the surface ocean as a single, unstratified layer of 200 m depth. Assuming a scenario of direct emission of PFOA into the oceans beginning in the 1970s, using data from Prevedouros et al. (2006) as the main entry pathway, the Globo-POP model predicted maximum northward fluxes in 2007 followed by a 50% decline by mid-century. Predicted concentrations of PFOA increased from 50 to 70 pg/L between 2000 and 2005 for north flowing waters which is within the range observed in the North Atlantic for 2000-09 (Table 5.2.54).

Study design considerations

Guidance for water sampling for PFOS and other perfluoroalkyl acids has been published by the Stockholm Convention (UNEP, 2015c; Weiss et al., 2015) and recently updated to include PFHxS and PFOA (Vrana et al., 2019). These documents note that rivers offer perhaps the best opportunity to assess temporal trends due to reductions in emissions provided that sources e.g. WWTPs, tributaries, accidental spills etc, are well documented. However, actual sampling designs for rivers are not discussed in the guidance documents. Sampling procedure (depth integrated, flow normalized, bridges vs boats) may affect measured concentrations. Two sampling programs in the Rhine during 2007 and 2008 used different procedures and got somewhat different results. Samples analysed by Loos et al.(2008a) were from the river bank or bridges, while Möller et al. (2010) sampled the mainly by boat, taking a greater number of samples. Loos et al. (2008a) reported PFOS concentrations ranging between 15 and 32 ng/L, while Möller et al. (2010) report lower values ranging from 1.7 and 13 ng/L. Paul et al. (2012) used both datasets to validate a transport model for PFOS in the Rhine and they concluded that the greater variation shown in the Möller et al. dataset could be due to localized sources and dilution, highlighted by the greater number of sampling points. Huset et al (2008) used time-proportional composites at two collection sites on the Glatt River in Switzerland a 24 h, flow-normalized composite sample at a point just before the Glatt River joins the Rhine River, to estimate discharge and assess the mass balance compared with inputs from WWTP effluents. This approach provided a better estimate of average PFOS concentrations and discharge.

Particles vs dissolved concentrations

Most studies of PFOS in surface waters have using unfiltered samples and the ISO method for analysis of PFOS and PFOA in natural waters which is based on unfiltered samples (ISO, 2009). Ahrens et al (2009b; 2009c) filtered their Elbe River and estuary samples in a ship clean room. They found that PFOS was entirely in the dissolved phase in the North Sea and 70-85% dissolved in the Elbe River. However, EtFOSE was exclusively found in the particulate phase. Using a fate and transport model, Paul et al. (2012) estimated that <20% of the total PFOS entering the Rhine was bound to sediments or suspended particles. Ahrens (2011) has noted that during filtration, PFASs can be adsorbed to the filtration equipment, while the dissolved phase can also adsorb to the filter material and equipment and to the walls of the receiving flask. The filter and equipment may also be a source for blank contamination. Nevertheless filtration of river water samples (typically with 0.7 µm glass fiber filters) has become quite commonly used over the past 10 years, with only the filtered sample being analysed for anionic PFAS.

Summary and Conclusions

This assessment addressed the questions of temporal trends of PFOS, PFOA and PFHxS in water and the broad spatio-geographic trends within WEOG countries as well as in neighboring seas (Baltic, North, Mediterranean) and oceans (North Atlantic, Arctic Ocean, Northeast Pacific). Unlike air measurements of PFAS, surface water sampling has not been consistently or repeatedly sampled at the same locations. As well the dynamic nature of rivers, lakes, seas and oceans, along with seasonal differences in river flow, lake thermal stratification, and ice cover adds to variability of concentrations. As such, data dispersion measured by MAD/median, is relatively high, especially in rivers. Nevertheless with results from over 3000 locations and over 11,270 individual measurements within the WEOG region, including 2035 analyses for samples collected from 2015-2020 it is possible to qualitatively examine temporal and spatial trends using medians and estimates of data dispersion of the concentrations.

- The information available provides knowledge of spatial trends for PFOS, PFHxS and PFOA across western Europe, Canada/USA and southern/eastern Australia, the North Sea, the Baltic, the Great Lakes, and in the North Atlantic, Arctic Ocean and Northeast Pacific
- No specific studies of temporal trends of PFAS in water were found, therefore medians for combined data from specific regions with the periods 2000–2009, 2010-2014 and 2015–2019 were compared. A challenge with these comparisons is that all data are included but sampling sites varied for each study.
- The three PFAS compounds showed distinct differences in median concentrations between inland surface waters in Western Europe and Canada/USA. Median PFOS concentrations for Canada/USA were 3.3 fold higher at non-urban (mainly riverine) sites in 2015-2019 compared to Western Europe. Median PFOA were 3.0-fold while PFHxS had higher medians in Western Europe compared with Canada/US.

- Different temporal trends for PFOS, PFHxS and PFOA in non-urban waters were observed between Western Europe and Canada/USA based on comparison of median concentrations over 5 to 10 year time intervals. While median PFOS and PFOA concentrations in Western Europe declined 70% and 60%, respectively, between 2000-09 and in 2015-19, they were much higher in Canada/USA in the 2015-19 period (353% for PFOA and 227% for PFOS). Median PFHxS was also much higher in 2015-19 in Canada/US waters than in 2000-09 in the combined urban and non-urban dataset. The results suggest continued significant emissions of the 3 PFAS to the aquatic environments in North America relative to Western Europe.
- Data for PFOS, PFHxS and PFOA in Australian and New Zealand rivers and streams were available but too limited temporally to effectively evaluate temporal trends. Recent (2015-19) measurements show very low background concentrations of PFOS in Australian river waters compared to earlier measurements which were mainly from urban influenced areas.
- Lower median concentrations of PFOA were found in samples from 2015-19 compared to 2000-09 or 2010-14 in the non-urban sites in the Mediterranean, the Baltic, and the North Sea. PFOA was also lower in coastal North Atlantic Ocean sites 2010-19 compared to earlier years. PFOS was also lower in the Baltic and North seas in 2015-19 but had increased in the Mediterranean and was unchanged in the coastal North Atlantic. PFHxS concentrations were higher in the Baltic and the North Sea in 2010-2014 and 2015-19 compared to 2000-09. The lower medians in the North Sea for PFOA and PFOS in 2010-19 vs 2000-09, and higher PFHxS, were similar to trends for its inflowing rivers.
- Major declines in concentrations of PFOS, PFOA and PFHxS have occurred in the Rhine River from 2006 to 2016 based on a comparison of median values. These declines reflect phase out of these substances and their precursors in western Europe that began in the mid-2000s.
- In the North American Great Lakes median concentrations of PFOA and PFOS in 2015-19 were 37 % and 159% higher, respectively than in the period 2000-09, when considering all sampling sites. However for open (non-urban influenced) lake sites median PFOA was lower. PFHxS was lower in 2015-19 than earlier years for both urban and non-urban influenced sites. However, comparisons were complicated by detection limit issues with 51-76% of samples having <MDL values. MDLs varied widely in studies on the Great Lakes ranging from 0.8 ng/L to 0.001 ng/L despite use of essentially the same analytical methodology.
- Although overall median concentrations of PFOA and PFOS in the Great Lakes increased, comparisons over time showed considerable year to year variation with declining concentrations of the three PFAS in Lake Erie and Lake Ontario and little change in Michigan, Huron and Superior.
- In the oceans adjacent to the WEOG region PFOA median concentrations were generally higher for 2010-19 compared to 2000-09, except in the North Atlantic where slightly lower concentrations were observed (-23%). PFOS was higher in 2015-19 in all ocean water

measurements except in Antarctica where only very limited samples (N=20 were available from 2000-09 for comparison. PFHxS was at detection limits in Antarctica but was present at higher concentrations (87-133%) in the Central Atlantic and in the North Pacific from the 2000-09 period to 2010-19. A decline in PFOS concentrations in the North Atlantic as of 2010 has been predicted by 3D-modelling based on waste water emissions from Europe and Canada/USA (Zhang et al., 2017). However the modelling assumed negligible emissions after 2010. This assumption appears to be too conservative.

- A major knowledge gap is measurement of PFOS, PFHxS and PFOA precursors, both neutrals (MeFOSA, EtFOSA, MeFOSE, EtFOSE, fluorotelomer alcohols) as well as anionic and amphoteric chemicals. Recent studies have suggested they are widespread in surface waters (D'Agostino and Mabury, 2017; McGuire et al., 2014; Zhang et al., 2019).
- Studies using total organic fluorine or TOP assays suggest that there are additional PFAS sources in surface waters and other aquatic media such as waste water effluents and sediments which need to be characterized in order to get a full understanding of prevailing levels and trends of PFOS, PFHxS and PFOA
- Results of modelling of PFOS and PFOA in ocean waters have generally been quite successful in predicting the observed geospatial variation in concentrations in surface and deep waters of the North Atlantic (Armitage et al., 2009a; Stemmler and Lammel, 2010; Zhang et al., 2017) or in latitudinal zones equivalent to the Central North Atlantic (Armitage et al., 2009b; Wania, 2007). However, modelling results for temporal trends appear to overestimate declining trends, which have yet to be observed in the North Atlantic.

Recommendations

- As noted in Chapter 5.3 (MacLeod et al) seawater monitoring sites for PFAS have not been established so far, but if they are they should be located within a region where spatial variability is relatively small.
- Continued monitoring of rivers offers the best opportunity to assess temporal trends. However, there is always the need for consideration of waste water and tributary inputs and thus multiple sampling points preferably at sites that are well characterized in terms of flow and proximity to sources are needed.
- Lower detection limits would help with future assessments of temporal trends in all locations but especially in open oceans, coastal seas and in the Great Lakes. Detection limits in the range of 1-5 pg/L for PFOS, PFHxS and PFOA have been demonstrated by several monitoring programs and should be universally adopted.
- Efforts should be made for more widespread measurement of precursors of PFOS, PFHxS

and PFOA in waters with “total” methods (eg total oxidizable precursor (TOP) or total extractable organic fluorine) or targeting specific known precursor compounds.

Acknowledgements

The preparation of the water chapter was enabled by financial support from the King Carl XVI Gustaf Professorship in Environmental Science held at Stockholm University by DCGM in 2018-19. We thank Cynthia de Wit (Stockholm University) for coordinating the financial and technical support for the work. Thanks to the following persons for providing unpublished data: Karl Bowles (Australia Defence), Chad Furl and Callie Meredith (Washington State Dept of Ecology), Roland Kallenborn (Norwegian University of Life Sciences), Suzanne Vardy (Queensland Dept of Environment). We also thank Dr Nobuyoshi Yamashita (National Institute of Advanced Industrial Science, and Technology, Tsukuba Japan) for seawater data. Special thanks to Kateřina Šebková (RECETOX, Masaryk University) for downloading the NORMAN data for PFAS in water.

5.2.4 Other media / non-core GMP media

5.2.4.1 Introduction

When the GMP was established under article 16 on effectiveness evaluation at COP3 (Decision SC-3/19; UNEP, 2007) it was tasked to compile the best information on long term measurements of POPs in air and human media (milk and serum). Measurements in these media with good QA/QC are readily comparable on a global basis, air has the fastest response to changes in emissions and decreasing levels in humans is the core objective of the convention. The GMP for its second report in 2015 included water as a medium to monitor PFOS and PFAS. The reasoning behind this choice still stands. The importance of other than core media to understand and assess changes over space and time of the risk posed by POPs to humans and the environment is indisputable. The choice of core media is based on strategic long-term priorities in the context of a global exercise with very limited resources. However, good quality measurements of long-term changes over time in other media are central and indispensable to gauge the importance of biogeochemical and commercial pathways of POPs mixtures in the environment and the consequent exposure routes to humans and ecosystems.

The work delivered since the 2015 report by a number of long-term international POPs monitoring programmes in strategic partnership with the GMP will be briefly described. These programmes are measuring a number of POPs in aerosols, snow, ice, sea water, freshwater, sediments, soils, terrestrial and marine biota in the WEOG region and have been at work over the last few decades. We will focus in particular on information about long term trends in water, sediment, terrestrial and marine biota made available since the previous 2015 GMP report was released. Future assessments should include more details on precipitation and sediments, especially for chemicals of emerging concern.

1 Table 5.2.55 All listed POPs and all monitoring programs that contribute to the GMP. Grey cells indicate substance, media and region
2 for which long term trends are reported. The data on the Mediterranean relate to published literature, and the EU on objectives of the
3 WFD and MFD but no coherent EU wide POPs reporting is available at this point.

Programme	Media	Geographic coverage	Time period	1. Aldrin	2. α -HCH	3. β -HCH	4. Chlordane	5. Chlordane	6. DDT	7. Dieldrin	8. Endosulfan	9. Endrin	10. γ -HCH	11. Heptachlor	12. HBB	13. HBCD	14. Hexa-, hepta-, PBDE	15. HCB	16. Mirex	17. PeCBz	18. PFOS	19. PCB	20. PCDD	21. PCDF	22. Tetra-, penta-, PBDE	23. Toxaphene	24. HCBd	25. PCP	26. PCNs	27. decaBDE; BDE-209	28. SCCPs	29. Dicofoi;	30. PFOA, salts, related comp.	31. PFHxS, salts, related comp.
AMAP	Biota	Arctic region	1975-2014																															
OSPAR	Biota, sediments	North Atlantic	1995-2014																															
GLWQA	Biota, sediments, water	North America	1970-2017																															
HELCOM	Biota, sediments	The Baltic Sea	1978-2018																															
MAP		Mediterranean																																
W&M FD		EU																																
USA		USA																																
Antarctica		Antarctica																																

It has become apparent in recent years that a good understanding of ecosystem process and structure is central to interpret long term changes in POPs concentrations in organisms operating at different trophic levels (see e.g. Hebert et al., 2000; Braune et al., 2014; AMAP, 2021 (in prep.)). The fast changes in the last few decades of macroscopic covariables such as ice cover surface, air and sea water temperature, salinity and the resulting changes in species ranges and abundances has an impact on POPs' pathways, partitioning in the environment, degradation rates, transfer in the food web, and in particular in POP levels in top predators.

In this context, the interpretation of POPs monitoring data benefits from ecosystem modelling of changing POPs' pathways and shows the need and benefits of coherent long term and integrated monitoring strategies in abiotic media, marine, freshwater and terrestrial macroscopic and microbial ecosystems (Thompson et al., 2017). Research and interpretation of monitoring data on POPs must be interpreted in light of climate changes on ecosystems structure and function in the Arctic. (AMAP, 2021 (in prep.)).

Finally, we emphasize the interest and value of Environmental Specimen Banks (ESB) and dated sediments, to build the contour of the POPs landscape in ecosystems over the past century.

In this section we will first briefly review the main conclusions reported by monitoring programs on selected POPs, in different regions, then we will summarize by substances and propose conclusions and recommendations. Relevant text and results from reports from the main contributing programmes published in the last five years and which has been used for this chapter is copied in the Annex. The Annex therefore gives details on data and time series with references.

5.2.4.2 Monitoring results

Below we cite the most recent available reports of each programme and briefly describe some highlights. First, we describe what each program reports, then we consider what is reported by the programs for each group of substances.

The work of ROG WEOG on the media has been based on a number of long-term monitoring programs and strategic partners to the GMP. For non-core media the GMP ROG can only rely on published data compilations by the programs, consequently in this section we consider two groups:

1.) Regions with monitoring programmes reporting data systematically available for GMP

Arctic - Arctic Monitoring and Assessment Programme AMAP

North Atlantic - Oslo-Paris Convention for the Protection of the Marine Environment of the North-East Atlantic OSPAR

North America Great Lakes - Canada - United States Great Lakes Water Quality Agreement GLWQA

2.) Regions with monitoring results, but no systematic report available for the GMP

USA (NOAA, EPA)

Mediterranean (MAP - Mediterranean Assessment Programme)

European Union (EU - WFD/MSFD - Water Framework Directive/Marine Strategy Framework Directive)

Antarctica

1.) Regions with monitoring programmes reporting data systematically available for GMP

Arctic (Arctic Monitoring and Assessment Programme - AMAP)

AMAP has monitored and reported the most extensive list of POPs, which include data for 16 of 31 POPs listed under the Stockholm Convention. An executive summary of AMAP can be found in the Air-annex along with description of the other air programmes. The assessment report “Temporal trends in persistent organic pollutants in the Arctic” (AMAP, 2016) released in 2016 contains data on concentration trends of POPs until 2014, which were further summarized in the 2018 assessment “Biological effects of contaminants on Arctic wildlife and fish” (AMAP, 2018). Many of the time-series from AMAPs previous POPs temporal trend assessment (AMAP, 2010) were extended from 2006/7 to 2011/12, included at least six years of data, and had an average length of 12 years. The data have been divided into two groups, before year 2000 (pre-2000) and after year 2000 only (2000-2014), to aid visualize the impact of the Stockholm Convention. Only marine and freshwater species are included, and the POP time series were available from seven countries, which include locations in the following areas; Alaskan marine areas, Arctic Canada, East Greenland (Ittoqqortoormiit area), West Greenland (Disko Island area and Isortoq), marine areas around Iceland, the Faroe Islands, northern Norway, and lakes in Sweden. Time series from the Arctic areas of Russia and Finland has not been made available.

Of the included time series, only 12% are of adequate length to detect a 5% annual change with a statistical power of 80% (see Box 1 for AMAP time trend vocabulary). For the POPs where time trends were reported, the general trend is a decreasing trend. However, the decrease is stronger for the pre-2000 time-trends compared to the post-2000 time-trends, suggesting that the decrease has leveled out. This pattern is consistent with the voluntary bans and outphasing of several of the legacy POPs. Several time trends have a significant non-linear trend, where the common pattern is an initial increase and then a decrease in concentrations. Overall, the greatest annual decline is observed for “legacy” organochlorine pesticides, while newer POPs still exhibit increasing trends in some cases. Chlordane and DDT have some time-trends showing increasing concentrations due to influence by a local source (whale processing site).

Updated time trends from Greenland including data until 2018 were kindly made available by AMAP for selected substances to illustrate substances with decreasing trend with non-linear trend components (HCB in black guillemot eggs, Fig. 5.2.65), with decreasing trends (Sum PBDE in adult ringed seals, Fig. 5.2.66), with increasing trends (HBCD in young ringed seals, Fig. 5.2.67), and PFAS with opposing trends (PFNA increasing and PFOA decreasing in juvenile ringed seals, Fig. 5.2.68) (AMAP/Frank Riget, Aarhus University – personal communication, June 2020). These are shown under the respective compounds.

Box 1. AMAP Time trend vocabulary

Significant: significance of 5%.

Increasing trend: A statistically significant increasing log-linear trend.

Increasing trend with non-linear trend component: both the increasing log-linear and non-linear trend components are statistically significant.

Decreasing trend: A statistically significant decreasing log-linear trend.

Decreasing trend with non-linear trend component: Both the decreasing log-linear and non-linear trend components are statistically significant.

Non-linear trend component: A statistically significant non-linear (fluctuating) trend with no clear increasing or decreasing tendency.

No trend: The time-series did not exhibit a statistically significant trend.

Not evaluated: The time-series was unsuitable for trend analysis (for example it contained too many “less-than-detection-limit” values).

(Source/copied from - AMAP (2015) Table 3.3)

Time series were not systematically reported since the 2015 GMP report for Aldrin, Chlordecone, endosulfan, endrin, g-HCH, HBB, PBDE-153, -154, -175, -183, -209, PCDD, PCDF, hexachlorobutadiene, pentachlorophenol, PCN, SCCP, dicofol, PFOA, its salts and PFOA-related compounds, and PFHxS, its salts and PFHxS-related compounds. For the other compounds with reported time trends, a brief description follows:

Dieldrin: 22 dieldrin time-series starting before 2000: 19 (freshwater fish, seabird eggs, and marine mammals) from Arctic Canada, and 3 (marine mammals) from eastern Greenland. 10 showed significant decreasing trends or decreasing trends with non-linear trend components. The mean annual decrease for the pre-2000 time series was 3%. For the 13 time-series available post-2000, only one showed decreasing levels, where the mean annual decrease was close to zero, indicating little if any decrease in levels since 2000.

Chlordanes and nonachlor: close to half of the pre-2000 time-series for Σ CHL (sum of cis-chlordane, trans-chlordane, cis-nonachlor, trans-nonachlor, and oxychlordane (31)), trans-nonachlor (35), oxychlordane (20), cis-chlordane, and trans-chlordane showed decreasing trends with a linear or non-linear trend component. The trends for chlordane compounds from all Arctic sites ranged between annual decreases of 3.6% for trans-nonachlor and 9.7% for trans-chlordane. Only one site of local contamination due to whale processing in Hvalstod in Hvalfjörður southwestern Iceland, showed an increasing trend. The proportion of post-2000 time-series showing a decreasing trend is lower, and the proportion of no-trend series is higher. The mean annual concentration decreases post-2000 ranged from 0.6% for trans-nonachlor to 6.8% for trans-chlordane. The increasing ratio between oxychlordane, primary metabolite of chlordane, and Σ CHL in some species over time reflects that the levels of the metabolite are increasing compared to its precursors.

DDTs: The DDT time-series consists of Σ DDT (sum of p,p'-DDE, p,p'-DDD, and p,p'-DDT) and of individual o,p' and p,p' isomers. Of the pre-2000 time-series, 46% of Σ DDT series and 32% of p,p'-DDE series showed a significant decreasing trend. The mean annual decrease was 4.2% for Σ DDT, 4.1% for p,p'-DDE, and 9.1% for p,p'-DDT. The increasing ratio between p,p'-DDE to Σ p,p'-DDTs over the years indicate increasing levels of the metabolite compared to its precursor, indicate no new “fresh” DDT in the Arctic, however, long-range transport from current use and storages can happen. Lower proportions of the post-2000 time-series show a significant decreasing trend. For the post-2000 time-series the mean annual decreasing trends were 2.4% for Σ DDT and 3.6% for p,p'-DDE. There was one significantly increasing DDT trend time-series (pre-2000) due to whale handling sites (Hvalstod in Hvalfjörður southwestern Iceland).

Heptachlor and heptachlor epoxide: three time-series were available for heptachlor, however, with most values below detection limits in all years. There were 12 heptachlor epoxide pre-2000 time series, 2 with decreasing linear or non-linear trends, the rest with no statistically significant trend across the time series. None of the 6 post-2000 time-series showed a significant time trend, except one with a significant non-linear trend.

Hexachlorobenzene (HCB): 35% of pre-2000 time-series showed a significant decreasing trend or a significant decreasing trend with non-linear trend components. One HCB time-series showed an increasing trend and another time-series increasing trend with a non-linear trend component. The mean annual decrease for the pre-2000 time-series was 2.6%. Six post-2000 time-series showed decreasing trends, three showed increasing trends, and the remaining showed no trend or significant non-linear trend component. The mean annual change for post-2000 time series was 0.0%.

Updated time trends from Ittoqqortoormiit, Greenland including data until 2018 show decreasing HCB concentrations with non-linear trend components (HCB in black guillemot eggs, Fig. 5.2.65).

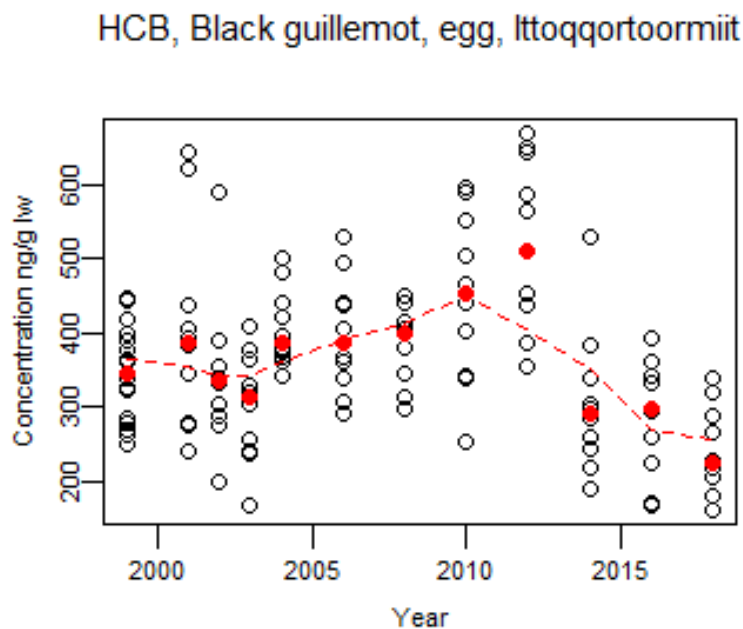


Figure 5.2.65. HCB in black guillemot egg (ng/g lipid weight) - updated time trends Greenland until 2018. Individual data points (open black), and mean (red) are shown. AMAP/Frank Riget, Aarhus University – personal communication, June 2020

Hexachlorocyclohexanes (HCHs): Most of the α -HCH time-series showed a decreasing linear or non-linear trend, 80% of the pre-2000 and 53% of post-2000 time series. Mean annual α -HCH decrease was 9% in the pre-2000 time-series, and 10% for the post-2000 time-series. 64% of the β -HCH pre-2000 time-series showed no significant trend or significant non-linear trend. Both decreasing and increasing trends were found, however, the mean annual decrease was only 1.5% for pre-2000 time-series. 83% of post-2000 time-series showed no trend or significant non-linear trend component. 68% of pre-2000 time-series for γ -HCH showed a decreasing trend or decreasing trend with non-linear components. 50% of the post-2000 time-series showed the same. Mean annual γ -HCH decrease was 7.6% for pre-2000 time-series and 6.2% for post-2000 time-series.

Mirex: There were 16 Mirex pre-2000 time-series, where 4 showed significant decreasing trends (one with non-linear trend component), the rest showed either non-linear trend or no trend. 10 post-2000 time-series, where one showed a significant decreasing trend, the rest showed either a non-linear trend or no trend. Mean annual decrease was 6.7% for pre-2000 time-series and 1.9% for post-2000 time-series.

Pentachlorobenzene (PeBz): Of the 12 pre-2000 time-series, 2 showed a decreasing trend or decreasing trend with non-linear components, and the rest showed a significant non-linear trend or no trend. Only one of the 6 post-2000 time-series showed a significant trend. Concentration in seabird eggs from Canada showed a significant non-linear trend with an initial increase followed by a decrease in recent years. The mean annual decrease for the pre-2000 time-series was 4.7%.

Polychlorinated biphenyls (PCBs): A total of 358 time-series of individual congeners starting before and after 2000 were analysed. The AMAP report focuses on the 214 time-series available for Σ PCB10 (CB28, CB31, CB52, CB101, CB105, CB118, CB138, CB153, CB156, CB180) and CB154. 38% of pre-2000 time-series for Σ PCB10 showed a decreasing trend, and 13% showed a decreasing trend with a non-linear trend component. For CB154, 34% of pre-2000 time-series had a decreasing trend, and 15% showed a decreasing trend with non-linear trend component. One pre-2000 time-series in blue mussels showed a significant increasing trend with non-linear trend components for both Σ PCB10 and CB154. Mean annual decrease was 3.7% for Σ PCB10 and 3.8% for CB154. For pre-2000 time-series, no significant difference in annual trend was found between eight PCB-congeners (CB28, CB52, CB101, CB105, CB118, CB138, CB153, CB180). 19% of post-2000 time-series for Σ PCB10 showed decrease and 18% for CB154 showed decrease. Mean annual decrease was 1.5% for Σ PCB10 and 2.5% for CB154. Two post-2000 time-series showed increasing trends for Σ PCB10, while one of them showed increased trend for CB154 and the other showed increased trend with non-linear trend component for CB154.

Toxaphene: 17 pre-2000 time-series (parlars 26 and 50). 5 parlar 26 and 4 parlar 50 time-series showed significant decreasing trends (two with significant non-linear trend component), whereas 3 (two parlar 26 and one parlar 50) time-series showed non-linear trends. The rest showed no trends. Annual decrease was 6% for parlar 23 and 0.8% for parlar 50. 20 post-2000 time-series available for parlar 26, where nine showed significant decreasing trends or decreasing trends with significant non-linear trend components. 22 post-2000 time-series available for parlar 50, where 8 showed a decreasing trend. The others showed either non-linear trends or no trend. Mean annual decrease of parlar 26 was 5.9% and for parlar 50 it was 0.8% increase.

Polybrominated diphenyl ethers (PBDEs): Several pre-2000 time-series available, however most included only BDE-47 and BDE-99. Of these, 9 BDE-47 and 2 BDE-99 pre-2000 time-series showed a significant increasing trend or a significant increasing trend with a non-linear trend component. The non-linear trend component significant in eight BDE-47 time-series showed an initial increase followed by a decrease in recent years. Post-2000 PBDE time-series showed no increasing trends for BDE-47, and only one increasing trend for BDE-99, however, 4 BDE47 and 3 BDE-99 post-2000 time-series showed a decreasing trend.

Updated time trends from Ittoqqortoormiit, Greenland including data until 2018 show decreasing trends for Sum PBDE in adult ringed seals (Fig. 5.2.66).

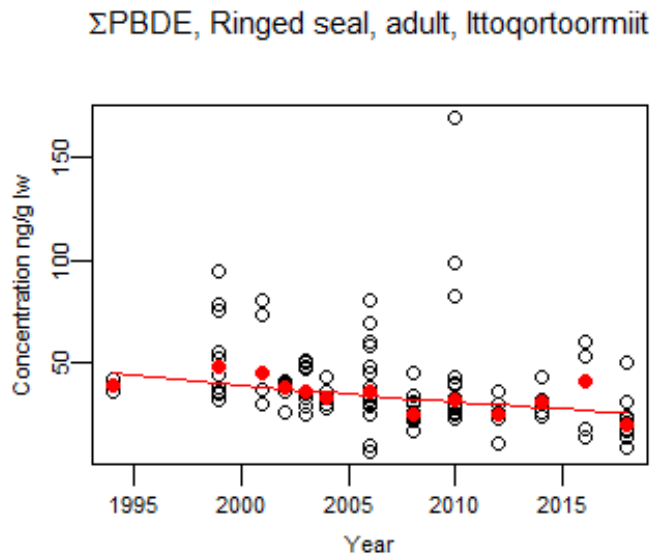


Figure 5.2.66. Sum PBDE (ng/g lipid weight) in adult Ringed Seal - updated time trends Greenland until 2018. Individual data points (open black), and mean (red) are shown. AMAP/Frank Riget, Aarhus University – personal communication, June 2020

Hexabromocyclododecane (HBCD): 7 pre-2000 time-series available for α -HBCDD, where 5 showed significant increasing trends. 2 time-series showed a significant non-linear trend or no trend. Mean annual increase of the 7 time-series was 7.6%. Post-2000 time-series showed a decreasing trend (one significant decreasing trend and two no-trend), with a mean annual decrease of 3.6%.

Updated time trends from Ittoqortoormiit, Greenland including data until 2018 show increasing HBCD trends in young ringed seals (Fig. 5.2.67).

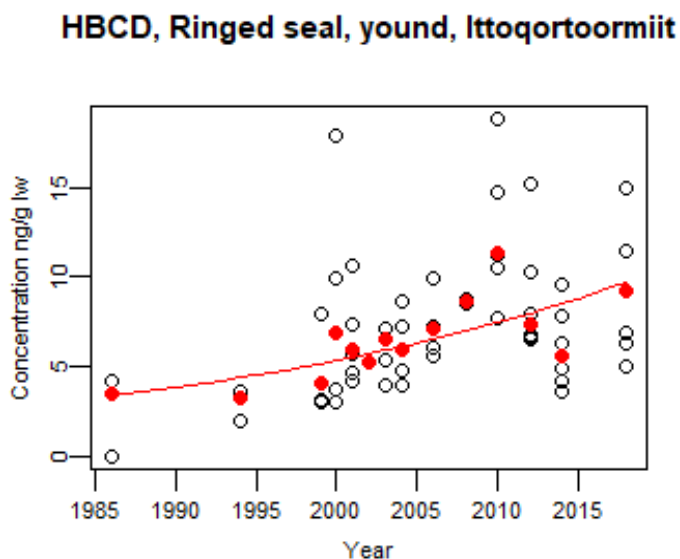
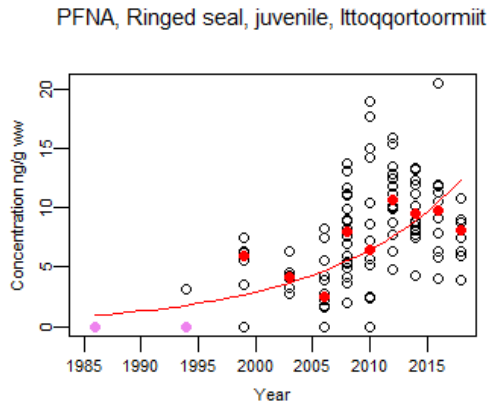


Figure 5.2.67. HBCD (ng/g lipid weight) in young Ringed Seal - updated time trends Greenland until 2018. Individual data points (open black), and mean (red) are shown. AMAP/Frank Riget, Aarhus University – personal communication, June 2020

Perfluorooctane sulfonic acids, its salts and perfluorooctane sulfonyl fluoride (PFOS): 8 of 16 pre-2000 time-series for PFOS showed a significant non-linear trend and 1 showed a significant increasing trend with a significant non-linear trend component. The others showed no trend. A common pattern was increasing concentrations until the mid-2000s, followed by decrease, which was described by the non-linear trend component. 11 post-2000 time-series for PFOS showed significant non-linear trend components or no trend.

Updated time trends from Ittoqortoormiit, Greenland including data until 2018 show that PFAS time trends are compound specific, with PFNA increasing and PFOA decreasing in juvenile ringed seals (Fig. 5.2.68). These data are not included in the tables of the present chapter.

a.) PFNA



b) PFOA

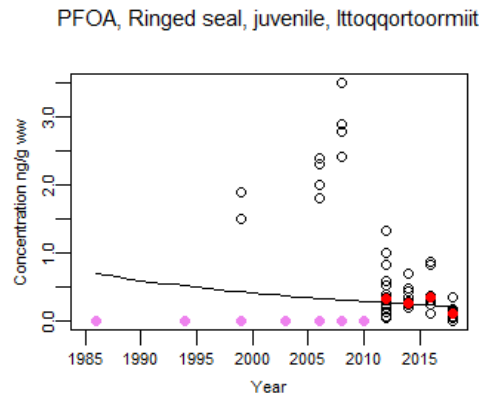


Figure 5.2.68. PFAS (ng/g wet weight) in juvenile Ringed Seal - updated time trends Greenland until 2018. Individual data points (open black), mean (red) and data below detection limit (pink) are shown. AMAP/Frank Riget, Aarhus University – personal communication, June 2020

North Atlantic (Oslo-Paris Convention for the Protection of the Marine Environment of the North-East Atlantic - OSPAR)

The Coordinated Environmental Monitoring Programme (CEMP) of the Oslo-Paris Convention for the Protection of the Marine Environment of the North-East Atlantic (the “OSPAR Convention”) releases annual assessment reports on the temporal trends of certain contaminants in sediments and biota (OSPAR, 2017e; 2018; 2019), some of which are part of the Stockholm Convention. There is, however, limited information describing the trends, such as the criteria for including or excluding a time series. The data are divided over five regions of the North-East Atlantic (<https://www.ospar.org/convention/the-north-east-atlantic>) (Fig. 5.2.69). Here we summarize the trends across the regions, as far as possible.



Figure 5.2.69. The five regions of the North Atlantic included in the OSPAR convention; I Arctic water, II Greater North Sea, III Celtic Seas, IV Bay of Biscay and Iberian Coast, V Wider Atlantic. Map copied from <https://www.ospar.org/convention/the-north-east-atlantic>

POPs assessed in biota were PCBs, PBDEs, HBCD, HCB, α -HCH, γ -HCH, and PFOS. POPs assessed in sediments included PCBs and PBDEs. Less than 50% of the time-series showed a significant trend, but for the ones that did, nearly all showed a decreasing trend. However, more than 20 time trends across all years showed increasing levels of PCBs in both biota and sediments. Time series not listed as either increasing or decreasing are assumed to show no trend.

Table 5.2.56. Summary of information given in tables in OSPAR Assessment reports 2016-2018 (OSPAR, 2017e; 2018; 2019). White areas indicate time trends assessed in biota, grey areas indicate trends assessed in sediments.

Contaminant	Assessment report year	Medium	Total time series	Decreasing time series	Increasing time series
PCBs	2016	Sediments	634	144	29
PBDEs	2016	Sediments	31	6	0
PCBs	2016	Biota	1851	911	21
PBDEs	2016	Biota	339	130	11
HCB	2016	Biota	97	26	3
Alpha-HCH	2016	Biota	78	32	1
Gamma-HCH	2016	Biota	131	88	0
PFOS	2016	Biota	10	8	1
PCBs	2017	Sediments	669	160	25
PBDEs	2017	Sediments	35	7	0
PCBs	2017	Biota	1536	858	24
PBDEs	2017	Biota	341	133	11
HCB	2017	Biota	105	27	3
Alpha-HCH	2017	Biota	80	34	1
Gamma-HCH	2017	Biota	115	62	0
PFOS	2017	Biota	5	4	0
PCBs	2018	Sediments	651	134	29
PBDEs	2018	Sediments	44	9	0
PCBs	2018	Biota	1756	739	31
PBDEs	2018	Biota	369	172	8
HBCD	2018	Biota	6	2	0
HCB	2018	Biota	81	21	1
Alpha-HCH	2018	Biota	45	24	0
Gamma-HCH	2018	Biota	85	41	1
PFOS	2018	Biota	8	8	0

In 2017, OSPAR released an intermediate status and trend assessment of PCBs and PBDEs in sediments (OSPAR 2017a,b), fish and shellfish (OSPAR 2017c,d). In biota, concentrations of PCBs were measured in fish liver and shellfish, while concentrations of PBDEs were measured in fish, mussels, and oysters. Which species used were not mentioned in the assessment, but it was noted that there is no homogenous choice of monitoring species across monitoring sites for PBDEs and that this may have affected the results as areas using fish show higher concentrations than areas using mussels.

PCBs: The assessment showed decreasing concentrations for PCBs in biota (1995-2014) in all areas except for in the Celtic Sea, where there is no statistically significant change. Estimated regional trends averaged over PCB congeners (CB28, CB52, CB101, CB118, CB138, and CB153) show yearly decreases ranging from -3.22% to -10.22%. All congeners have decreased to concentrations in biota to acceptable biological concentrations, the only exception being CB118, which is detected at levels which can cause unacceptable risk to the environment in 8 out of 11 areas assessed. PCB concentrations in sediments show a decreasing trend in three out of five areas (Northern North Sea, Southern North Sea, and Gulf of Cadiz), the two remaining areas show no statistically significant change (the Irish and Scottish West Coast and the Irish Sea). PCBs are found in all marine sediments, but all congeners except CB118 are below the EAC in all assessment areas. CB118 are close to or above EAC in three out of five assessment areas. Estimated regional trends averaged over PCB congeners (CB28, CB52, CB101, CB118, CB138, and CB153) show yearly decreases ranging from -2.38% to -9.21%. Time series show no statistically significant change in the Celtic Sea. Arctic Water, some parts of the Celtic Sea, Iberian Coast, and Bay of Biscay are some of the areas that lack monitoring data for PCBs.

PBDEs: For PBDEs, concentrations in biota have been decreasing in all assessment areas except Skagerrak and Kattegat (one area), where there was no statistically significant trend. Although 11 out of 339 monitoring sites show increasing mean concentrations, overall concentrations have decreased in biota after PBDE was regulated. Estimated regional trends in biota averaged over PBDEs (BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, and BDE-154) show a yearly change varying from +0.01% to -13.91%. Some time series measuring PBDE concentrations in sediments were not included due to concentrations being too low to measure accurately or the series being too short for analysis. Also, areas were only assessed if at least three monitoring stations had enough data, both in temporal (at least five years) and spatial spread. Therefore, only two areas had enough data to be included in the assessment, the Northern North Sea and the Irish Sea, from which only the latter show a statistically significant decreasing trend while the former showed no trend. BDE-209, the most common flame-retardant congener, occurs at the highest concentrations measured, while some congeners lack data as many measurements are below the limit of detection. Estimated regional trends averaged by PBDEs (BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, and BDE-154) show a yearly decrease varying from -6.49% to -12.80%.

Great Lakes of North America (Canada-United States Great Lakes Water Quality Agreement - GLWQA)

In the 1970s a vast cooperative effort between Canada and the USA to deal with environmental pollution and public health in the The Great Lakes of North America watersheds was established. Originally signed in 1972, the Canada-United States Great Lakes Water Quality Agreement (GLWQA) was recently amended in 2012 (<https://binational.net/glwqa-aqegl/>). Under GLWQA there are two different efforts related to monitoring fish: One, led by the USA monitors edible portions of fish in a context of public health using these fish, and the other, lead by Canada monitors whole fish with a wider ecosystem perspective. The most recent report of this endeavour summarizes the vast body of factual knowledge that has been compiled in the long-term cooperative effort (Environment and Climate Change Canada and the U.S. Environmental Protection Agency, 2019). Here we summarize both, but also emphasize the importance of internal consistency in measured matrices and inclusion of lipid content to allow estimation chemical activity by normalising for lipid differences between species and tissues (see e.g. Borgå & Ruus, 2019).

GLWQA monitors four sub-indicators (toxic chemicals in herring gull eggs, water, sediments, and whole fish) in the Great Lakes of North America. The lakes monitored include Lake Superior, Lake Michigan, Lake Huron (including St. Marys River), and Lake Erie (including Niagara River and International section of the St. Lawrence River). The programme focuses on Chemicals of Mutual Concern (CMCs), which is a list of chemicals agreed upon by both parties (Canada and USA) in the GLWQA in May 2016. The list includes Hexabromocyclododecane (HBCDD/HBCD), Long-Chain Perfluorinated carboxylic acids (LC-PFCAs), mercury (Hg), Perfluorooctanic acid (PFOA), Perfluorooctane sulfonate (PFOS), Polybrominated Diphenyl Ethers (PBDEs), Polychlorinated Biphenyls (PCBs), and Short-Chained Chlorinated Paraffins (SCCPs), where PFOA, PFOS, PBDEs, PCBs, and SCCPs are of interest to the GMP. GLWQA also monitors other chemicals of interest that may be included in the list of Chemicals of Mutual Concern or that can prove to be a problem in the future, but most of these chemicals are not monitored consistently across the sub-indicators.

The programme divides the time trends into two timelines: 10-year trends and long-term trends, assigning status of Fair, Good, Improving or Unchanging (Box 2). The 10-year trends started approximately at the time when the Stockholm Convention went into force, while the long-term trends started sometime in the mid-1970s. The years included in the time trends are different for different sub-indicators as some do not have updated monitoring results since 2014.

Box 2. GLWQA Time trend status vocabulary

Fair: The metrics show that the toxic chemical concentrations are not meeting the ecosystem objectives/guidelines, but they are exhibiting minimally acceptable conditions.

Good: The metrics show that the toxic chemical concentrations are meeting the ecosystem objectives/guidelines or they are otherwise in an acceptable condition.

Improving: Decrease in contaminant levels and other parameters of concern

Unchanging: No change in the level of contaminants and/or other parameters of concern

(Source/copied from - Environment and Climate Change Canada and the U.S. Environmental Protection Agency, 2019)

Overall, the status of the Great Lakes is **Fair**, with the exception of herring gull eggs which have the status of **Good**. The rationale behind the overall status is that the trends show declining concentrations, but the Chemicals of Mutual Concern are still found at concentrations exceeding ecosystem objectives. As seen in many other geographical areas, for example the Arctic, the long-term trends are **Improving**, while the 10-year trend often is **Unchanging**. Additional years of monitoring is needed for nearly all chemicals measured (both Chemicals of Mutual Concern and other chemicals of interest) to be able to provide status and trend information. There was a great decline of contaminant concentration found in herring gull eggs from the 1970s to 2013, but accounting for the gulls' change in diet (to consuming from lower trophic levels) happening in the same time period, the rates of decline were reduced.

PCBs: PCBs show declining long-term trends in all sub-indicators except for in water, where a temporal trend has not been observed since 2004. Concentrations have fallen dramatically since the voluntary stop in production in the 70s and the decline is the driving force for the overall assessment in many of the sub-indicators. However, the decline in concentrations measured in the environment has decreased in the last decade and the trends are not as strong. For example, the sum of 33 PCB congeners measured in herring gull colonies in 2002 showed a range between 0.96 and 11.27 µg/g, while in 2017, the same colonies showed a range between 0.9 and 7.81 µg/g. Also, despite PCB levels having decreased by 90% in fish fillet of some fish species, PCBs are still found at concentrations exceeding the ecosystem quality objectives in whole fish. The declining trend of PCBs is confounded by differences in analytical methodology between 1987 and 2009-2011.

PBDEs: PBDEs saw an increase in concentrations from 1982 to 2000, when production stopped, then no increase in the years 2000-2006, and finally declined by 2012 and also stabilised some places since 2012. Both long-term and 10-year trends show different directions in the different sub-indicators. The long-term trends in herring gull eggs are non-linear, following the pattern just explained, while the overall long-term trends are declining in sediments and whole fish

except for BDE-209 in sediments in all lakes and Hexa-BDE and Tetra-BDE in whole fish from Lake Erie, which are increasing in concentrations. The 10-year trends show no change in herring gull eggs, undetermined in water, are mixed in whole fish (TetraBDE decrease in all lakes, PeBDE increase in Lake Ontario, Huron, and Michigan, and decrease in Lake Erie and Superior, and HexaBDE increase in Lake Huron and Michigan, and decrease in Lake Ontario, Erie, and Superior) and are likely levelling off in sediments.

SCCPs: There is currently no new data available for short-chained chlorinated paraffins. Previous studies show that the contaminant is found at levels far below the ecosystem quality objectives in whole fish.

PFOA and PFOS: Both PFOA and PFOS have been increasing in concentration in herring gull eggs in Lake Superior and Lake Huron in the long-term trends. Long-term trends also show that PFOS has been increasing in whole fish in Lake Huron and Lake Ontario, but decreasing in concentration in whole fish in Lake Superior and Lake Erie, and decreased in sediments in Lake Ontario. 10-year trends in whole fish show a decline in concentration in Lake Erie and Lake Ontario, but an increase in Lake Superior and Lake Huron. The spatial variation found between the lakes is likely due to variations in analytical technique as the methodology is still under development. Concentrations are above the ecosystem quality objective lakewide. PFOA is no longer reported in whole fish as it is uncommon to find the contaminant in fish in the Great Lakes.

DDTs: DDTs show declining long-term trends in herring gull eggs (as DDE) and in sediments. The contaminant is no longer reported on as it has shown low and unchanging concentrations for a while.

HBBCD: The flame retardant shows the same long-term and 10-year trends in whole fish, with decreasing concentrations found in Lake Superior and Lake Ontario, and increasing concentrations found in Lake Michigan, Lake Erie, and Lake Huron. However, concentrations measured are still well below ecosystem quality objectives.

PCN: PCNs have in long-term trends declined in herring gull eggs in Lake Ontario and Lake Erie, and declined in whole fish in Lake Ontario. While 10-year trends show that they have declined in herring gull eggs in Lake Superior and continue to decline in whole fish in Lake Ontario. Historical PCB contamination is likely an important source of PCNs in whole fish in the Great Lakes. Lake Erie saw an increase in concentration of PCNs in herring gulls and whole fish between 1996 and 2005, after a decline from 1980 to 1995. There is also a spatial trend of higher concentrations found in lakes upstream and then decreasing concentrations as we move to lakes further downstream.

Organochlorine pesticides: Pesticides are not monitored regularly in the Great Lakes, but the programme report declining 10-year trends for α -HCH, γ -HCH/lindane, toxaphene, and dieldrin in water, and long-term trend of decline of toxaphene in sediments of Lake Superior. Toxaphene has also been found to have a declining long-term trend in whole fish with higher concentrations found in Lake Superior.

Dioxins and furans: PCDD/Fs and TCDD show declining long-term trends in herring gull eggs in all lakes and TCDD in whole fish in Lake Ontario. Concentrations of PCDD/Fs in herring gull eggs have fallen dramatically since the 1970s.

Other industrial byproducts: Increasing trends are found for HCB and HCBd in most lakes in water although the only significant trends are found for HCB in Lake Huron and Lake Erie and for HCBd in Lake Ontario. HCB are found to decline in concentration in sediments in long-term trends for Lake Erie and Lake Ontario.

The Baltic Sea (Baltic Marine Environment Protection Commission - Helsinki Commission HELCOM)

The Baltic Sea covers 420000 km² and has a drainage area about four times that surface and some 85 million people live in it. For a holistic management of the region, HELCOM releases assessments also including other topics than hazardous substances, such as eutrophication, biodiversity and maritime issues (<https://helcom.fi/>).

Since the previous GMP report, HELCOM has released an assessment in three parts of varying degree of details. State of the Baltic Sea – Second HELCOM holistic assessment 2011-2016 (HELCOM, 2018a) is the main publication, Thematic assessment of hazardous substances 2011-2016 (HELCOM, 2018b) is a supplementary report to the main publication, and finally, each of the indicators (HBCDD (HELCOM, 2018c), PBDEs (HELCOM, 2018d), PCBs (HELCOM, 2018e), and PFOS (HELCOM, 2018f)) are detailed in four HELCOM core indicator reports. New indicator specific reports were released in 2020 for PBDE (HELCOM, 2020a), PCB and Dioxin (HELCOM, 2020b) and PFOS and other PFASs (HELCOM, 2020c), too late for a detailed inclusion, however, the results up to 2018 are in line with the ones up to 2016, summarized below and in the Annex.

Here, 559 time series were assessed, where 311 showed no trend, 236 showed decreasing trends, and only 12 showed increasing trends. However, these numbers of time series include all hazardous substances monitored, not only POPs. For POPs only, most datasets showed no trend. The overall contamination status has not changed markedly since the previous holistic assessment (HELCOM 2010), though, it may be difficult to compare with the previous assessment because of differences in methodologies, for example, differences in substances or substance groups sampled and changes in threshold values. However, some comparisons can be made. PCBs were identified as one of the substance groups having the highest contamination ratios (concentration to threshold value) in the previous report, but do not seem to be a major contributor of contamination in the present report. Also, a number of substances previously assessed, such as γ -HCH and DDT and its metabolites, are no longer considered as of significant concern and are therefore not included in the present assessment. However, increasing HCB concentrations are reported in Swedish sediments (Apler & Josefsson, 2016).

Hexabromocyclododecane (HBCD): time series of levels in biota showed increasing concentrations from the 1970s and 1980s to the 2000s. Since the end of the 1990s, decreasing levels are seen at the Swedish west coast station Fladen, Utlängen in the southern Baltic Proper, and in two stations in the Bothnian Sea. Concentrations were measured in herring, cod, flounder,

dab, eelpout, and perch, where the concentrations were below the threshold value in all areas (EQS: 167 µg/kg WW with 5% lipid normalization). Concentrations were also below the threshold in sediments (QS from EQS dossier – 170 µg/kg DW with 5% organic carbon concentration (CORG) normalization). Monitoring in biota in general shows no detectable HBCD trends, with some exceptions showing decreasing trends. It should be noted that most of the Baltic monitoring stations are reference sites, and are not selected to reflect and assess the status of local primary and secondary sources.

Polybrominated diphenyl ethers (PBDEs): there is limited availability of long time series in biota in the Baltic Sea, and those available are concentrated to the western coast of the region. PBDEs were measured in herring, cod, flounder, dab, eelpout, and perch. Concentrations of single PBDE congeners (BDE28, BDE47, BDE99, BDE100, BDE153, BDE154) are declining but fail the threshold (EQS: 0.0085 µg/kg ww with 5% lipid normalization) as sumPBDEs in all areas monitored. However, the threshold is considered very precautionary and is due for review by the EU Chemicals Working Group. The PBDE concentrations in herring muscle in the Baltic are higher compared to the Swedish west coast in the North Sea, and there seems to be declining concentrations in the open sea, but the variation both between and within sites is larger in coastal areas. The threshold was achieved in sediments (QS from EQS dossier: 310 µg/kg dw with 5% organic carbon normalization), although the threshold was much higher in sediments compared to biota.

Polychlorinated biphenyls (PCBs): good status was achieved in biota in majority of coastal and open sea areas, but concentrations exceeded the threshold (EC: 75 µg/kg WW with 5% lipid normalization) along the coast of the Bothnian Bay, the Arkona Basin, Gulf of Finland, Kattegat, and in the Quark. PCBs (sum of CB28, CB52, CB101, CB138, CB153, CB180) were measured in flounder, common dab, herring, cod, European perch, European sprat, and eelpout, and showed either decreasing or no significant trend.

Perfluorooctate sulphonate (PFOS): Data is available from Denmark, Finland, Poland, and Sweden, but absent from some areas, for example, Gulf of Riga and the Estonian coast of the Gulf of Finland. A great deal of the data from monitoring stations are based on one or a few years. Levels of PFOS in biota show increasing concentrations since the 1970s and 1980s in the Baltic Proper and the Bothnian Sea, however, these levels have been decreasing in the most recent ten-year period. Concentrations of PFOS were measured in the muscle of herring, cod, perch, eelpout, and European flounder, and were below the threshold level (EQS: 9.1 µg/kg WW with conversion from liver to muscle) in all monitoring stations except one. However, monitoring stations are located at reference areas with no specific local pollution load, and there is reason to believe that PFOS may pose more severe contamination risks to the Baltic Sea than the current indicator evaluation would suggest.

There is limited data for PFOS in sediments, but most measurements show low concentrations.

2.) Regions with monitoring results but no systematic report available for the GMP

The United States of America (National Oceanic and Atmospheric Administration/ Environmental Protection Agency - NOAA/EPA)

The United States of America is, regrettably, not a party to the SC. It is well known that much relevant and high quality monitoring of POPs is ongoing in the USA under major federal efforts (NOAA, EPA) and regional coastal and land based work and is published in the scientific literature. Some of this is described below. However given the limitations of this work we are not in a position to review primary literature and in the absence of consolidated POPs trends reporting for the USA we do not include it in the overall summary and table.

The Center for Coastal Monitoring and Assessment (CCMA) addresses pollution through the National Status and Trends Program (NS&T), through which long-term monitoring of toxic chemicals and environmental conditions is conducted at more than 350 sites along U.S. coasts. The programme also documents the nature and severity of the biological effects associated with toxic chemicals in 25 coastal ecosystems. The programme started in 1984 and is the only nationwide source of long-term data on toxic contaminants in U.S. coastal waters and estuaries. Outcomes include a status of contaminant concentrations around the U.S. including Alaska, Hawaii, the Great Lakes, and Puerto Rico. The programme's data information products are available to the public via publications and the Internet.

The National Status and Trends Program consists of two programmes, Mussel Watch and Bioeffects. Parameters monitored in the Mussel Watch Program include sediment and bivalve tissue chemistry for over 100 organic and inorganic contaminants; bivalve histology; and *Clostridium perfringens* (pathogen) concentrations. The Mussel Watch programme regularly quantifies PAHs, PCBs, DDTs and its metabolites, TBT and its metabolites, chlorinated pesticides and toxic trace elements. Bioeffects Assessment Program identifies and assesses biological effects associated with contaminant exposure. Over forty intensive regional studies have been conducted since 1986 using the Sediment Quality Triad approach which utilizes a stratified random sampling method to determine the areal extent of contaminated sediments. The data include: sediment chemistry, toxicity, and species diversity and quantity for the same suite of organic contaminants and trace metals as the Mussel Watch Program (Law et al., 2010).

Kimbrough et al. (2008) presents an assessment of two decades of contaminant (Chlordanes, DDTs, Dieldrins, PCBs) monitoring in the US coastal zone. Sericano et al. (2014) describes in detail the temporal trends and spatial distribution of DDT in bivalves from the coastal marine environments of the continental United States, 1986–2009. Hartwell et al. (2018) assess the magnitude of effects of contaminants in coastal waters. See also the Southern California Coastal Water Research Project SCCWRP's 2018 Annual Report (www.sccwrp.org).

Mediterranean (Mediterranean Assessment Programme - MAP)

Unfortunately, there are no long term POPs monitoring and reporting strategy on a Mediterranean wide scale, despite many decades of effective cooperation on other issues under

Mediterranean Assessment Plan (MAP). The main reference for the Mediterranean Action Plan under the 1976 Barcelona Convention is the Integrated Monitoring and Assessment Programme of the Mediterranean Sea and Coast and Related Assessment Criteria (UNEP/MAP, 2016):

“Over 40 years ago, the Mediterranean Action Plan (MAP) was established as a framework of cooperation in addressing common challenges of marine environmental degradation, and in 1976 the Barcelona Convention was adopted by the Mediterranean countries. With an initial focus on pollution, which then expanded to further address biodiversity, coastal management and sustainable development, in 1995 the Convention was amended and renamed as the Convention for the Protection of the Marine Environment and the Coastal Region of the Mediterranean. In addition to the Barcelona Convention and seven protocols addressing specific aspects of Mediterranean environmental protection and conservation, since 2008 the Ecosystem Approach has been the guiding principle with the ultimate objective of achieving the Good Environmental Status (GES) of the Mediterranean Sea and Coast. An Integrated Monitoring and Assessment Programme (IMAP) was adopted by the 19th Meeting of Contracting Parties (COP 19) in 2016. The 2017 Quality Status Report is the first report based on the Ecological Objectives and Common Indicators of IMAP, with a view to assess the status of the Mediterranean in achieving GES.”

Further, the Indicator Assessment Factsheet (Code EO9CI17) (UNEP/MAP, 2017) states on the Ecological Objective 9 (EO9): Chemical pollution. Common Indicator 17. Concentration of key harmful contaminants measured in the relevant matrix:

“At present, there are still old threats and new pressures, although the trends and levels of the so-called legacy pollutants (e.g. heavy metals, persistent organic pollutants and pesticides), have decreased significantly in the most impacted areas in the Mediterranean Sea after the implementation of environmental measures (e.g. leaded-fuels ban, mercury regulations, anti-fouling paints ban), as observed in the Western Mediterranean Sea (UNEP/MAP/MEDPOL, 2011a).

Despite the implementation of the MED POL monitoring for chlorinated compounds during almost two decades, the availability of new data with sufficient spatial geographical coverage and quality assured impedes to further assess their occurrence in the Mediterranean Sea region, beyond known sources and hotspots in coastal areas. On the other hand, most of the recent datasets show non-detectable levels, mainly in biota matrices, which is in accordance with the earlier decreasing levels and trends observed in previous MAP reports (UNEP/MAP/MED POL 2011a, 2011b, 2012). However, there are still point and diffuse pollution sources releasing both priority and emerging chemical contaminants (e.g. pharmaceuticals, personal care products, flame retardants) in the Mediterranean Sea.

The scarcity of recent POPs quality assured datasets in the MED POL Database and the fact that most of these show non-detectable levels, mainly in biota matrices, is in accordance with the

earlier lowering levels and trends observed in previous reports (UNEP/MAP/MED POL 2011a, 2011b, 2012) and no further updates could be performed at present. “

This does not mean POPs are not an issue in Mediterranean ecosystems and populations, and there is a vast literature of very good QA/QC standards on presence of all POPs in all media, but unfortunately there is no coherent data flow that would enable official joint reporting.

Several recent publications indicate that levels of PCDD/F, dioxin-like PCBs and PBDEs in Mediterranean mammals are of concern, i.e. the levels of dioxin-like PCBs in the local population of sperm whales is ranked at the top of the reported values for this species worldwide (Bartalini et al., 2019). In a recent review of PCBs, DDTs, HCHs, HCB, PBDEs and PFOS/PFOA in common species of cetaceans in the Mediterranean Sea, including striped dolphin, common bottlenose dolphin, short beaked common dolphin, sperm whale and fin whale, the POPs exposure in Mediterranean cetaceans surpasses the estimated toxicity threshold value for several species/compounds (Marsili et al., 2018). For some legacy POPs, levels seem to decrease due to their ban and regulation, but emerging and/or not yet regulated compounds, for which levels are high, are showing increasing trends.

Also in avian biota, in gulls, local exposure through feeding near dump sites by yellow-legged gulls revealed the importance of dump sites as a source of POPs in Mediterranean seabirds (Roscales et al., 2016). In contrast, feeding from the marine food webs studies by gulls such as the Audouin's gull, leads to exposure and accumulation of low-chlorinated dioxins and furans, non-ortho substituted PCBs, which show the highest toxic equivalency factors (TEFs), led to a significantly greater toxicological concern in Audouin's gull as compared to yellow-legged gulls (Roscales et al., 2016). As the Audouin's gull feed predominantly pelagic, their exposure to high TEF POPs highlighting the need for further research given that this is the same source the fisheries target for human consumption.

In abiotic media, levels of POPs are of concern also in water (Berrojalbiz et al., 2011) and sediments (Gómez-Gutiérrez et al., 2007; Merhaby et al., 2019), and indicate that a coherent monitoring and reporting POPS in the Mediterranean would be possible and beneficial as also pointed out by Merhaby et al. (2019): the need to *“Enhance collaboration and exchange information, experiences and best practices between all Mediterranean countries by creating a large cooperation between institutions, and social actors to coordinate the implementation of Barcelona provisions including MAP and MED POL Program which should continue to play a key role in capacity building and developing quality criteria standards throughout the region for the benefit of all.”*

European Union - Water Framework Directive/Marine Strategy Framework Directive

The European Union became a party to the Stockholm Convention in 2004 and adopted a regulatory instrument to transpose the Stockholm Convention and the resulting obligations concerning POPs into EU legislation. This regulation was updated in 2019 (EU, 2019). For several decades, EU has developed normative frameworks to deal coherently with environmental protection, air, water quality and health. In the context of the present chapter on POPs monitoring in Stockholm Convention non-core, we focus on two major initiatives; the EU Water Framework Directive (WFD) and the Marine Strategy Framework Directive (MSFD).

On 23 October 2000, the "Directive 2000/60/EC of the European Parliament and of the Council establishing a framework for the Community action in the field of water policy" or, for short, the EU Water Framework Directive (WFD) was adopted (EC, 2000).

For a comprehensive and integrated approach to the protection of all European coasts and marine waters, EU has adopted two instruments, the 2002 Recommendation on Integrated Coastal Zone Management and the 2008 Marine Strategy Framework Directive. The regulation provides the legal framework to implement the Stockholm Convention and its engagements on production, trade and monitoring of POPs by its member states (EU, 2019).

Through these vast and comprehensive efforts, EU has established shared monitoring and assessment efforts compiling the monitoring results including a number of POPs in multiple media in terrestrial watersheds and coastal and marine environments from all the Member States. It would certainly be helpful for the GMP and the Stockholm Convention work under article 16 on Effectiveness Evaluation to have access to the actual monitoring data compiled by the EU under the WFD and MSFD, that are nominally available but dispersed and not systematically reported.

In 2008 the EU established a set of environmental quality standards (EQS) in the field of water policy, which also includes a list of priority substances (EC, 2008). EQS were established for the following POPs listed in the Stockholm Convention: aldrin, dieldrin, endrin, DDT, hexachlorocyclohexane (not divided into alpha, beta, gamma, so assumed that all are included), endosulfan, hexachlorobenzene (EQS only established for congener 28, 47, 99, 100, 153, 154), pentachlorobenzene, hexachlorobutadiene, and pentachlorophenol, where all are listed as priority hazardous substances except for pentachlorophenol, aldrin, dieldrin, endrin, and DDT. EQS has not been established for chlordane, chlordecone, heptachlor, hexabromocyclodecane, mirex, PFOS, PCBs, polychlorinated dibenzo-para-dioxins, polychlorinated dibenzofurans, toxaphene, polychlorinated naphthalenes, short-chain chlorinated paraffins, dicofol, PFOA, and PFHxS.

The MSFD provides a framework in which Member States must take the necessary measures to achieve or maintain "good environmental status" (GES) in all of the EU's marine waters by 2020. Achieving this objective means that the EU's seas are clean, healthy, and productive, and the use of the marine environment is sustainable. The MSFD includes eleven qualitative

descriptors describing what the environment should look like when good environmental status has been achieved. Commission Decision 2010/477/EU on criteria and methodological standards on good environmental status of marine water guides Member States on how this objective can be achieved (EC, 2010). In terms of monitoring contaminants, indicator 8 and 9 (contaminants are at a level not giving rise to pollution effects, and contaminants in fish and other seafood for human consumption do not exceed levels established by community legislation or other relevant standards) are of interest. However, not all Member States had monitoring programmes implemented by 2014, but stated that monitoring programmes will be in place by 2020, and so MSFD monitoring is expected to be fully implemented by that time (EC, 2017). The lack of monitoring programmes caused several gaps of knowledge in the 2018 assessment report, including lack of consistency and comparability between Member States. The States are aware of the gaps in their monitoring and what needs more work. There has been no update whether good environmental status has been achieved in 2020.

Antarctica

There are no monitoring programmes of other media in the Antarctic region, but several studies have analysed various POPs across species, regions matrices. Unfortunately, inconsistent reporting of wet, dry and lipid weight, several different tissues and matrices, and often without lipid content, makes the conversion between units impossible, and thus also a temporal trend analysis. Contrary to the Arctic where long-term monitoring data allow us to investigate the temporal trends, the POP temporal trend in the Antarctic is thus less clear.

Climate change is rapidly altering the global environment, with changes being most prominent in the Polar Regions. These climate changes affect both transport of contaminants, deposition and distribution in the abiotic environment, ecological and ecosystem changes, and uptake and accumulation in the food webs. Thus, temporal trends based on monitored contaminant concentrations should account for these changes, if the aim of the temporal trend analyses is to measure the effectiveness of the Stockholm convention and thus use and emission of POPs. Details on how the temporal contaminant trends relate to climate change are discussed in detail in the AMAP climate change and persistent organic pollutants report (AMAP, 2020 (in prep.)).

In general, declining POP concentrations during the last two decades have been observed in some Antarctic fish, although the trends appear weak and fluctuating. PCBs, DDTs, and PBDEs showed a concentration peak in 2001 and 2005 (Corsolini et al., 2006; Borghesi et al., 2008), that presumably was related to POP release after the iceberg B15 calved from the Ross Ice Shelf (NOAA, 2014), reported to be the possible source of POPs released into the seawater (Cincinelli et al., 2016).

There has been an increasing number of publication reporting data on the POP contamination in seabirds during recent years and most of them concern penguins, which are endemic to Antarctica. However, as most studies were carried out along the Ross Sea coasts and the Antarctic Peninsula and islands it is not possible to generalise across the vast Antarctic continent. POP time trend until 2011 in Antarctic seabirds reported that PCBs, HCB, and DDTs increased during the 1970s–1980s; and that they started to decreased in biota at the beginning of the 1990s,

followed by some POPs increasing again at the end of the 1990s, as some of them are still in use (<http://chm.pops.int/Default.aspx>), or are stored in legal or illegal depository, or can be released from their final sinks (e.g. deep sediments and waters) (Corsolini, 2009; 2012).

The temporal trends seem to differ between habitats; whereas no changes or slightly increasing POP levels are found in benthic organisms and fish (Corsolini, 2009; van den Brink et al., 2011), stable or decreasing concentrations are found in the more pelagic feeding fish and seabirds (Focardi et al., 1992; Corsolini et al., 2003; Weber and Goerke, 2003; Goerke et al., 2004; Corsolini, 2009; van den Brink et al., 2011; Corsolini, 2012). The contrasting trend between pelagic and benthic feeding organisms may ascribed to the transport of organic contaminants from the pelagic system to the benthic environments that is especially efficient in the Antarctic region because of its close relation to sea-ice dynamics (van den Brink et al., 2011; Cincinelli et al., 2016). The organic matter transport from the water-ice interface and pack ice may be affected by climate change: More ice will melt, contributing to the release of the entrapped organic matter to the water column from where it falls down to the benthic environment and community.

In most seabirds, the levels are decreasing, but it is not equally clear for DDTs, which increased in the same seabirds in the 2000s (Corsolini et al., 2011), and in Adélie penguin eggs from the Palmer Archipelago, the Σ DDT concentrations did not decrease from the 1970s to the 2000s (Geisz et al., 2008). The DDT increase in seabirds, along with the lack of decrease in the penguins despite low and decreasing global DDT emissions and deposition, may be caused by increased release of DDT to the marine environment due to melting glaciers and measurable DDT levels in meltwater (Chiuchiolo et al., 2004; Geisz et al., 2008).

In some species, like icefish collected around the Antarctic Peninsula, some POPs like PCBs and DDT have an increasing trend since the 1990s, while other POPs, e.g. HCB, showed a stable or slightly decreasing trend (Strobel et al., 2016).

POP concentrations are available in penguins from Antarctica since 1966, with samples collected from 1964 to 2011. A very first sample of emperor penguin fat was collected in 1911 and preserved in an igloo in the Ross Island was analyzed for DDT concentrations, but no residues were found in this sample (Sladen et al., 1966). This result assumes an historical meaning because the sample was collected more than 30 years before the beginning of the massive worldwide DDT use. Since the mid-1960s, the DDTs in Adélie penguins ranged from 0.128 ng/g lipid weight (eggs collected from the Ross Sea in 1967, Risebrough et al., 1967) to 132 ng/g lipid weight in 2008-09 (muscle sample collected from King George Island, Kim et al., 2015). The highest *p,p'*-DDE concentration, 820 ng/g lipid wt, was quantified in fat samples of Emperor penguin collected in 1981 in the Weddell Sea (Schneider et al., 1985).

5.2.4.3 Summary by substances

An overall summary of the general time trends for the individual substances across the different programmes is found in Table 5.2.57. Although Mediterranean and Antarctica do not report

monitoring results systematically to the GMP, some info can nevertheless be extracted from their reports concerning other themes/problems in their representative regions.

Across the regions, α - β - and γ -HCH, DDT, PCBs, tetra- to hepta-PBDEs show decreasing trends, and/or decreasing/no change. No substances show increasing trends across all regions, however in Great Lakes, HCB, HCBd, PCN, Deca-BDE in general show increasing trends, as well as DDT in Antarctica, and HCB in the Baltic region. Dieldrin and Toxaphene were reported in both AMAP and GLWQA, showing decreasing levels or nonlinear trends. PFOS was reported across several regions, with contrasting trends; increasing in the Arctic and the Great Lakes, and decreasing in the Baltic region and the North Atlantic.

Some compounds were only reported from one region, thus no general assessment across the regions can be made, such as for those only reported from the Arctic: Chlordane (decreasing), heptachlor (no linear trend), Mirex (decreasing/non linear trend), PeCBz (decreasing/non linear trend); and for those only reported from the Great lakes: PCDD (decreasing), PCDF (decreasing), HCBd (increasing), PCN (increasing) and PFOA (no linear trend).

For several substances, there is no new trend data reported since the last GMP report. New substances are probably not reported as they are not yet regularly measured and reported (PCP, Dicofol, SCCP, HBB, PFHxS), likely due to challenging analytical chemistry and lack of comparable methods. Some older substances are no longer being measured and reported due to low levels and no change over time (Aldrin, Chlordane, Endosulfan, Mirex, Endrin).

Table 5.2.57 Presentation of the direction of changes in concentrations over time included in the different programmes. Although Mediterranean and Antarctica do not report monitoring results systematically to the GMP, some info can be extracted from their reports concerning other themes/problems in their representative regions. The colours represent the direction of the changes in concentration over time according to the respective programmes; green: decreasing concentrations, yellow: non-linear changes in concentrations that are neither increasing nor decreasing, grey: no change, blue: no changes in concentrations/trend (statistically decided), red: increasing concentrations. Circles represent substances regulated earlier with low levels and no change in the last years and triangles represent newly listed substances of concern.

	AMAP (1975-2000)	AMAP (2000-2014)	OSPAR (1995-2014)	HELCOM (1978-2018)	Great Lakes (1970-2017)	Antarctica (NA)
Aldrin	○	○	○	○	○	○
α-HCH						
β-HCH						
Chlordane	1					
Chlordecone	○	○	○	○	○	○
DDT		1				
Dieldrin						
Endosulfan	○	○	○	○	○	○
Endrin	○	○	○	○	○	○
γ-HCH						
Heptachlor						
HBB	Δ	Δ	Δ	Δ	Δ	Δ
HBCD					3	
Hexa-, hepta-, PBDE			2	2	3	
HCB						
Mirex			○	○	○	○
PeCBz						
PFOS					3	
PCB						
PCDD						
PCDF						
Tetra-, penta-, PBDE					4	
Toxaphene		5				
HCBD						
PCP	Δ	Δ	Δ	Δ	Δ	Δ
PCNs						
DecaBDE						
SCCPs	Δ	Δ	Δ	Δ	Δ	Δ
Dicofol	Δ	Δ	Δ	Δ	Δ	Δ
PFOA, salts, related compounds						
PFHxS, salts, related compounds	Δ	Δ	Δ	Δ	Δ	Δ

1. One local source (whale processing site) causes an increasing trend in some species.

2. BDE-153 and BDE-154 only.
3. Increasing in some lakes, decreasing in other lakes.
4. Overall decreasing, increasing in whole fish in Lake Erie and in sediments in Lake Superior.
5. Parlar 26 decreases annually with 5.9%, parlar 50 increases annually with 0.8%.

Below is a summary of the substances with reported data, complementary to Table 5.2.57, with some selected examples, please refer to the specific regional programmes for details on specific matrices, time periods and substances.

HCHs are in general decreasing in all reported media and regions, however, still the levels are of concern in some species such as in cetaceans from the Mediterranean region.

DDTs are in general decreasing in all reported media and regions, however, the levels are increasing in Antarctic biota. Also the pattern of DDTs is changing, with an increasing ratio of the metabolite ppDDE compared to its precursor DDT, which indicates no new “fresh” DDT. In some areas like the Great Lakes, the average levels are well below the target threshold of 1.0 µg/g ww across the basin.

Dieldrin is in general decreasing or levelling off in all reported media and regions, however, it is still one of the most abundant organochlorines in waters of the Great lakes, second to α -HCH.

HCB is in general decreasing or no trends in most reported media and regions, however, still the levels are of concern in some species such as in cetaceans from the Mediterranean region. Some HCB time trends are increasing, such as in Great Lakes water and a few OSPAR trends for the North Atlantic.

Mirex levels are generally decreasing or showing low levels with no change. Mirex will no longer be reported for some of the programmes, such as the Great Lakes.

PCBs are in general decreasing or no annual trend is reported across media and regions. Most congeners have low and acceptable levels, whereas PCB-118 is close to or exceeds the EAC in three out of five assessment areas in the North-Atlantic (OSPAR), PCB concentrations exceeded the threshold in some areas of the Baltic Sea (EC: 75 µg/kg WW with 5% lipid normalization), and Sum PCBs is still above the target of 0.1 µg/g ww in whole fish in some fish species in some of the Great Lakes. Dioxin-like PCB levels in Mediterranean sperm-whales are the highest for this species worldwide, and PCB in Arctic top predators are still high and associated with negative effects.

PBDEs are in general decreasing or show a non-linear trend across regions for the tetra to hepta homologues. Deca-BDE, which is sparsely reported and only available as a separate congener for two regions, where Great Lakes show increasing trends, and Antarctica decreasing trends. In Great Lakes sediments, total PBDEs, and in particular the deca-substituted BDE 209 are

increasing across all five Great Lakes, with doubling times ranging from 4 years to 74 years. Although levels are declining in the Baltic region, SumPBDEs in all areas monitored in fish fail the threshold (EQS: 0.0085 µg/kg WW with 5% lipid normalization). The PBDE levels are still of concern in some species such as in cetaceans from the Mediterranean region, in particular prevalent high levels of PBDE 209.

Toxaphene is only reported in the Arctic and in the Great Lakes. In both the Arctic and the Great lakes, the levels are decreasing with a linear or non-linear trend, or no significant time trend. In the Great Lakes, current levels of toxaphene in fish for consumption is less of a concern for public health than other dominant compounds such as PCB and mercury, and thus will no longer be reported.

Hexabromocyclododecane (HBCD) trends post-2000 are decreasing across the regions that report data. In the Arctic and in the Baltic the levels were increasing pre-2000. HBCD concentrations in Baltic fish were below the threshold value in all areas (EQS: 167 µg/kg WW with 5% lipid normalization).

Hexachlorobutadiene (HCBd) is only reported from the Great Lakes, where there is an increasing trend in the water in most lakes.

PFOS and other PFAS. PFOS is generally decreasing or showing a non-linear trend across regions. In Great lakes whole fish, the declining trends observed at most locations are statistically significant only in Lakes Ontario (since 2002), Huron, and Michigan. Although the trends are decreasing, the levels are still high in the North Atlantic, and in edible portions of some Great lake fish. Average concentrations of PFOS in whole fish are generally above the FEQ guideline of 4.6 ng/g ww in all 5 Great Lakes. However, concentrations of PFOS in common fish species do not result in advisories that would be more restrictive than the consumption guidelines already set for PCB. There is a gradient toward increasing PFAS contamination from the upper Great Lakes (Superior and Huron) to the lower Great Lakes (Erie and Ontario) for both tributary and open-lake sediments. There are significant PFAS levels in cetaceans in the Mediterranean.

PCN is only reported from the Great Lakes, where there is an increasing trend in the water in most lakes.

PCDD and PCDF are surprisingly only reported from the Great Lakes, where there is a decreasing trend.

5.2.4.4 Data Interpretation and ecosystem modelling

Interpreting temporal changes from data and observations

As the monitoring programmes progress, the data richness increases. The thorough statistical analyses of temporal trends i.e. in regions with dense monitoring collections such as the Arctic, the Great Lakes, North Atlantic and Baltic Sea, combined with power analysis on how to best design the study in terms on numbers of samples needed to detect a given change, has greatly improved our understanding. This understanding is given a retrospective statistical model, analyzing the temporal trend in measured data.

In a recent analysis of Arctic data more than 1000 temporal contaminants data in Arctic biota, including all regions and biota from marine and freshwater ecosystems, the resulting conclusion was that legacy POPs with long range transport generally decline in the Arctic, whereas no significant change or even increase is found for contaminants in the Arctic or in local regions with additional local sources or remobilization due to altered habitat use and climate change (Rigét et al., 2019). This was a robust analysis of the measured contaminant levels over time, still only 12% of the long-time trend series in the Arctic compilation were of sufficient quality given the statistical requirements. The remaining data-series would require additional years of monitoring to adequately detect a temporal trend, some as long as 20 years.

The analyses did not include other factors that might affect the temporal trend, other than changes in emission, which is the target of the present report. As an example, changes in ecological interactions such as predator-prey relationships affecting diet and the resulting trophic level (Hebert & Weseloh, 2006; Braune et al., 2014; Hebert & Popp, 2018), affect the temporal trends in some areas to confound the emission-related temporal trends. Similarly, in areas with long time trends, climate parameters such as large scale or local climate indices (i.e. Arctic Oscillation and North Atlantic Oscillation), sea ice extent and distribution significantly explains temporal changes in POP levels in ringed seals (*Pusa hispida*) (Houde et al., 2019), and help understand the complexity of ecosystem responses, and exposure routes, that confound the observed declining trends linked to reduction in global emissions.

The above illustrates the important function of the Environmental Specimen Banks, in order to ensure sampling and storing of tissues under established QA/QC to enable retrospective monitoring and analysis of temporal trends in current and future POPs in the Stockholm Convention. It also illustrates the need to include supporting data in the temporal trends such as measures of dietary changes, to allocate the explained variance in the temporal trends to the correct explanatory variable, i.e. reduction in emissions vs food web changes. Although several regions have long term monitoring programmes for various media, changes in the study design during the run of the programme might hamper the information gained from the data. I.e. by changing location, matrix, reducing the frequency of sampling, halting the monitoring, the data become information poor resulting in weak statistical analyses. However, a strength of the monitoring data, despite single series being of low power, is that the understanding across the series, with data on different chemicals and media were taken together and run in parallel adds a huge value to the collection and use. The overall collection of the single time series is of high power and new machine learning tools will show this, as already illustrated in the Arctic when jointly evaluating the existing time trends (Rigét et al., 2019).

For some media, the temporal archive is present in the media it-self such as in sediment cores (e.g. Liber et al., 2019; Ontiveros-Caudras et al., 2019) and glacier cores (Hermanson et al., 2010), that can be dated and analysed for recent and part history of contamination to identify trends and sources. Also in biota, specific tissues may allow for a retrospective analysis to provide the temporal history of contaminants the individual has been exposed to, as well as identifying specific life stages of high exposure. An example of this is the opportunistic sampling of ear plugs from stranded and diseased migrating baleen whales, such as blue whale (*Balaenoptera musculus*) where the diet differs sufficient between seasons to use the lipid structure to determine the seasons and thus diet (Trumble et al., 2013). In the blue whale example, which was sampled after being accidentally hit by a passing boat, the ear plug functioned as a natural archive which preserved the accumulated contaminants. This approach will provide a longitudinal time trend, see below.

How can models help us understand temporal trends?

When addressing temporal contaminant levels in biota, not only changes in emissions come to play, but also changes in sources and exposure during different life stages, as well as species- and individual differences in elimination. By combining contaminant observations with mechanistic understanding of the processes involved in the contaminant distribution and enrichment of various media, mechanistic models can help us quantify and compare the different processes that are involved. In these models, the processes involved are chosen and defined by mathematical solutions, and the chemical-, environment-, and ecosystem is described by scaling the parameters defined in the processes such as octanol-water partitioning coefficients, temperature, species characteristics, dietary relations etc. When properly explaining observed data, the models can be used to identify which are the most sensitive parameters to affect the contaminant levels, and also to model prospective temporal trends given defined settings, i.e. effect of reduced emission.

Examples of mechanistic models used to understand contaminant behavior is the combination of a global multimedia model ([BETR Global](#), MacLeod et al., 2001), with a human exposure model (ACC-HUMAN, Czub & McLachlan 2004) to predict spatial PCB-153 concentration in human milk (McLachlan et al., 2018). This resulted in high correlation between modelled and measured concentrations, with identification of global regions with over or under-predictions where understanding is needed.

Similar approaches are used to understand the human exposure to contaminants in the Arctic, including contributors and drivers of the observed temporal trends, as reviewed by Wania et al. (2017). Here, the development of mechanistic models to understand Arctic environmental distribution, food web accumulation and human exposure and accumulation is summarized (Wania et al., 2017). In particular they identified how changes in diet from tradition to market affect and reduce the temporal trends, thereby confounding the observed temporal trends by reduced emissions (Quinn et al., 2012). Also, the human exposure models have been used to understand that PCB levels increase with age observed in cross-sectional monitoring studies, as a “ghost of the past” rather than increased levels with age *per se*, as the individual has experienced

past high exposure that remains in the body (Quinn and Wania, 2012). In fact, time since peak exposure best explains the relationship between body burden and age, and interpretation of body burden vs age relationships will differ between monitoring studies of repeated measures of an individual (longitudinal – concentration trends in individuals) and sampling a cross-section of the population over time (Ritter et al., 2009; Quinn and Wania, 2012; Nøst et al., 2016).

It is worth pointing out that the human temporal trend monitoring data in the GMP are cross-sectional data sampled multiple years, which are thus describing the contaminant level in different individuals of the population at one point of time- rather than following the temporal development within specific individuals. In wildlife it is therefore a mismatch between the model predictions of lifetime exposure and accumulation (longitudinal – e.g. PCBs in beluga whales (*Delphinapterus leucas*) (Hickie et al., 1999), and ringed seal (Hickie et al., 2005) and bowhead whales (*Balaena mysticetus*) (Binnington and Wania, 2014) and the observational data, which are mostly cross-sectional (i.e. summarized in Rigét et al. (2019)).

Global and ecosystem distribution of contaminants under climate change

Several studies summarize and report how contaminant global distribution and local ecosystem distribution occur, and how this is directly and indirectly affected by other factors such as climate change. This might lead to increased or decreased POP levels in the environment and biota, depending on which factor is considered, and often serves to show the complexity of the matter (i.e. Wang et al., 2016; AMAP, 2021 (in prep.)). An example is how increased temperature on the one hand leads to increased respiration and uptake of contaminants, increased water solubility, and increased primary production which decreased the bioavailable fraction of contaminants in water (Borgå et al., 2010). The investigation of the changing pathways to exposure of old and new POPs contributes valuable insights for a more accurate understanding of changes in ecosystem structure and function. The abundance of current and past monitoring data of POPs in abiotic and biotic matrices, including specimen banking, molecular, genetic and microbial probing coupled with the mighty new analytical tools and machine learning arts presents a landscape where much good and useful work can be done in science serving regulators and local communities.

5.2.4.5 Conclusions and recommendations

Conclusion. Long term stable monitoring of biotic and abiotic media, sharing environmental sample collections, analytical methods and curated data are central to evaluate the effect of actions undertaken and to enhance understanding of the changing structure of ecosystems and POPs pathways leading to exposure. Despite the relative abundance of monitoring data the statistical power to identify significant trends over time is limited, and efforts should be made in monitoring design and advanced data analysis to enhance accuracy of estimated time trends

Recommendation. *Facilitate cooperation and capacity to maintain long term monitoring plans/programmes and environmental sample banks integrating multiple media, robust QA/QC and reporting in a coherent transparent service to enhance accuracy of estimates of changes over time.*

Conclusion. Current results seem to indicate that POPs regulated in source regions decades ago have significantly decreased, but are still present at low levels that have not changed since the previous GMP report in 2015 and are still of concern in some regions. Substances listed later in the Stockholm Convention; PBDE, HBCD, HCBD, PFOS/PFOA, SCCP with ample exemptions for use, show increasing levels of concern.

Recommendation. *Report evidence of the success of past regulations in decreasing exposure to POPs to consolidate the need of further work with chronic low levels of old POPs (PCBs, PBDEs, α -HCH, HCB) and the growing threat of partially regulated POPs (PFOS/PFOA, BDCD, HCBD, SCCP) and new substances with POPs features sometimes used as alternative.*

Conclusion. The complex ecotoxicology of mixtures and the rapidly increasing quantities released to the environment of new substances with POPs features are a challenge and an opportunity to transform monitoring strategies. Integrated assessments should not bury primary data under indicators but develop tools to look at primary QA/QC controlled empirical data provided in public repositories by a diversity of fields, from meteorology to molecular biology. Machine learning, statistical and process modelling tools can enhance process understanding and operational capacity.

Recommendation. *Make best efforts to integrate POPs monitoring work and resulting data of abiotic media and macroscopic organisms with climate data and molecular biology and toxicology in curated and accessible repositories.*

Conclusion: Local, indigenous knowledge about ecosystem process and structure has been of great value for scientists in developing understanding of POPs pathways and exposure routes, and science should provide local agents with meaningful and operational tools to deal with contaminants and POPs.

Recommendation: *Make best efforts to establish and maintain conditions that facilitate common understandings and cooperation between local agents and scientists to develop effective strategies in public health and environmental policy on POPs*

5.2.4.6 Acknowledgements

The authors would like to acknowledge first and foremost the people who did the field work and the analytical work in labs that makes this possible, the managers of the programs that made the data available. We also thank Dr. Simonetta Corsolini for making available her summary on work from Antarctica, Hayley Hung for help with the latest SOLEC report from the Great Lakes, and Frank Riget and Simon Wilson for help with the updated AMAP time trends until 2018 for Greenland. We thank The Ministry of Ecological Transition and Demographic Challenge (MITECO) Madrid Spain for support to RG, the Norwegian Environmental Agency for financing the contribution by MA, and the University of Oslo for support for KB.

5.3 Long-Range Transport Modeling

5.3.1 Background

Measurements of POPs at locations far from sources provide direct empirical evidence of long-range transport at regional, continental, hemispheric, and global scales. However a complete assessment of long-range transport of POPs can only be achieved by integrating information and understanding about 1) the location, rate and time trend of emissions, 2) measured levels at different locations, and 3) rates of transport through the environment in the atmosphere, oceans, rivers, and migratory species. Mass balance chemical fate models provide a platform for synthesizing information about emissions, transport processes, and measured concentrations by quantitatively describing processes that control the fate of chemicals in the environment (MacLeod et al. 2010, Hung et al. 2013).

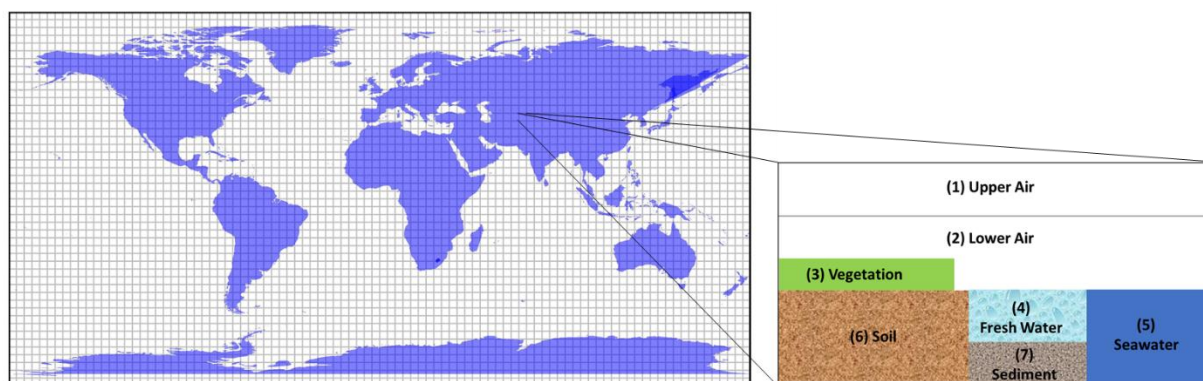


Figure 5.3.1. Conceptual structure of a global scale chemical fate and transport model that links the location, level and trend of POP emissions to measured concentrations. The BETR Global model (MacLeod et al. 2011) describes the global environment on a grid with variable spatial resolution. Within each grid cell chemical emissions and fate processes that distribute POPs between air, water, soil, sediment and vegetation are modelled, and grid cells are connected by flowing air and water.

Models link the concentrations of POPs in the environment to emissions that have occurred at specific locations and times and provide a conceptual basis for understanding the levels and trends that are observed at monitoring stations. Thus models provide a basis to assess how reductions in the emissions of POPs are reflected in reductions in levels in the environment. Environmental fate models linked to bioaccumulation models can extend the assessment to link emissions to POP levels and trends in wildlife and in the bodies of humans (Cowan-Ellsberry et al. 2009, McLachlan, 2018).

Models can also be applied to isolate the effect of emission reductions from other factors that may affect levels and trends of POPs observed at monitoring stations, such as variability in local conditions and long-term changes in climate (UNEP/AMAP, 2011, Carlsson et al. 2018).

Models are particularly useful when they support the formation of testable hypotheses that guide research (MacLeod et al. 2020). In recent years international working groups have reviewed POP modelling activities and made recommendations for research priorities, including the 2010 Assessment Report of the Task Force on Hemispheric Transport of Air Pollutants (UNECE 2010, Gusev et al. 2012), which was formed under the Convention on Long-range Transboundary Air Pollution in 2010, the Saltsjöbaden V workshop (Grennfelt et al. 2013), the workshop on Next Generation Air Quality Monitoring (Hung et al. 2013), and the outcomes of the ArcRisk EU FP7 Collaborative Project (Carlsson et al. 2018). These working groups have generally agreed that degradation rates of POPs are a driving source of uncertainty in model scenarios, and that improving emission inventories is among the most important research priorities for applying models to understand the long-range transport, levels, and trends of POPs in the environment and in human milk and blood.

The first two WEOG region and global effectiveness evaluation reports under the Stockholm Convention described POPs modeling studies that mostly focused on relatively well-studied POPs including PCBs and HCHs, and illustrated capabilities to link emission inventories to levels and trends. A role for models in identifying and composing risk profiles of new POPs and to screen for the most relevant transport pathways was illustrated in the second report.

5.3.2 Structure and Objectives of Section 5.3

This section of the third WEOG effectiveness evaluation report summarizes four recent case studies of applying the integrated approach of emission estimation, process-based modelling, and model evaluation against measurement data. These case studies have been selected to illustrate recent advances in integrated assessment modeling, specifically: 1) linking emissions to human body burdens of POPs at the global scale, 2) conducting integrated assessment of POP candidates, 3) estimating emissions of POPs and POP candidates from monitoring data, and 4) modeling global emissions, fate and transport of PFOS in the ocean.

Considered together, these four case studies demonstrate that cross-disciplinary collaboration with an integrated approach that brings together information from models, emission estimates and environmental monitoring provides a quantitative, global-scale accounting of POPs or POP candidates. Applying and iteratively improving POP assessments using the integrated approach has the potential to play a much more prominent role in implementation and effectiveness evaluation of the Stockholm Convention, and in the design and execution of the Global Monitoring Plan, for example by supporting development and critical evaluation of emission inventories, and by identifying priority research areas that target critical uncertainties within the {emissions - fate and transport - environmental concentration} chain.

5.3.3 Case Studies of the Integrated Approach to Understanding Long-range Transport of POPs

5.3.3.1. Predicting global scale exposure of humans to PCB153 from historical emissions

Most integrated approach studies of POPs have focused on understanding emission-to-concentration relationships for POPs in air, which is a core medium for monitoring, and in some cases also water, which is a core medium only for PFOS. The other core media for monitoring POPs for effectiveness evaluation of the Stockholm Convention are human milk and blood. The database of measured levels of POPs in human milk maintained under the Stockholm Convention UNEP/WHO Global Monitoring Plan is a valuable resource in this regard. This database was employed together with the global POP fate and transport model BETR Global (MacLeod et al., 2011) and the human exposure model ACC-HUMAN (Czub and McLachlan, 2004) to assess current capabilities for emission-to-human exposure modeling of a prototypical traditional POP, PCB-153. The concentration of PCB-153 in human milk for 56 countries around the world was modelled based on a global historical emissions scenario and compared with observations in the database.

Historical primary emissions of PCB-153 beginning in 1930 were obtained from Breivik et al. (2007 & 2016). This emission inventory considers emissions occurring throughout the entire lifecycle of PCB-containing products (including production), as well as emissions from the export and import of e-waste (see Breivik et al., 2016 for further details). The BETR Global model at a grid cell size of $3.75^{\circ} \times 3.75^{\circ}$ (lat/long) was used to simulate the global fate and transport of PCB-153, with transport process of chemical associated with particles from soil compartments to freshwater compartments removed because the parameterizations within the model were found to not apply to arid environments.

ACC-HUMAN (Czub and McLachlan, 2004) predicts the concentration of chemicals in foodstuffs (namely fish, beef, and dairy products) from concentrations in environmental media (namely air, water, and soil). Human exposure to the chemicals is further modelled using ingestion scenarios, ultimately leading to estimates of chemical concentrations in human tissue and milk. A range of uptake and elimination processes by organisms throughout the food web, such as ingestion of food and soil, inhalation, gill or root uptake, fecal egestion, childbirth, and nursing are considered. The lifetime exposure of representative individual humans was modeled based on variable concentrations in food and environmental media arising from the spatial and temporal trends in emissions.

Country-specific environmental concentrations of PCB-153 were derived by taking population-weighted averages of the overlying 3.75° grid cells of BETR Global. Concentrations of PCB-153 in human milk for first-time mothers were then modelled, using a model woman who was “born” 20-30 years before the year that breast milk measurements were made (the year of “birth” was selected based on the average age of primiparae mothers in the country of interest). Country-specific dietary patterns and animal lipid ingestion rates were employed using WHO Global Environment Modelling System cluster diets (UN, 2017) and food balance sheets from the UN’s Food and Agriculture Organization (FAO, 2017). All meat, dairy, and freshwater fish consumed in a country were assumed to be produced in that country, except marine fish which were assumed to be internationally-sourced and thus had chemical concentrations that were driven by modeled PCB-153 concentrations in BETR Global grid cells corresponding to regions with the highest global fish harvesting (UBC, 2014).

Modelled human milk concentrations were compared to measurements from the WHO/UNEP global monitoring plan for POPs (van den Berg et al., 2017; Hulek et al, 2014). This monitoring program under the Stockholm Convention measures country-specific PCB-153 human milk concentrations from first time mothers employing pooled milk samples from either 10 donors (until the year 2003) or 50 donors (after the year 2003) per country. The data employed covered the years 2000 to 2014 from 56 countries.

Measured and modelled concentrations of PCB-153 in human milk were strongly correlated when considering both log-transformed absolute values and rank orders of concentrations in different countries (Figure 5.3.2, $r = 0.76$ for Pearson illustrated in the left panel, $r_s = 0.74$ for Spearman illustrated in the right panel, $p < 0.0001$). The root mean square difference between the measured and modelled values was 0.65 log units (i.e., a factor of 4.5). Despite this overall good agreement, two particular clusters of bias were identified in the modelling results that showed a clear geographic pattern. The seven countries of West Africa (blue squares in Figure 5.3.2) were characterized by strong underprediction of PCB-153 concentrations in human milk, while concentrations in Hungary and Bulgaria (red triangles in Figure 5.3.2) were strongly overpredicted (see also Figure 5.3.3).

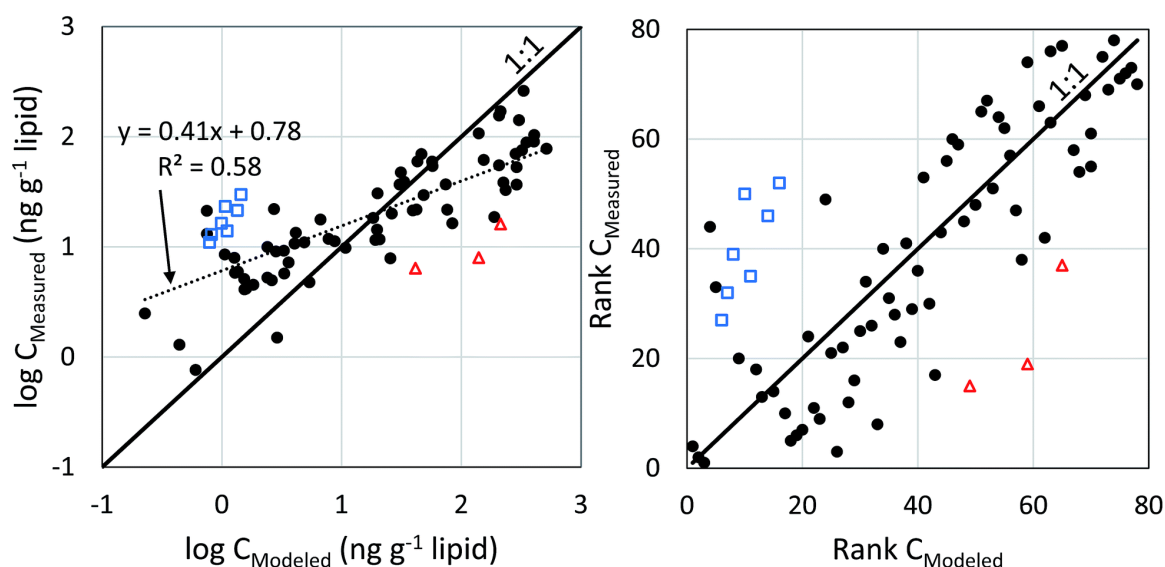


Figure 5.3.2. Comparison of measured and modelled concentrations of PCB-153 in human milk for 78 milk samples from 56 countries collected under the UNEP/WHO Global Monitoring Plan (modelling data as described in McLachlan et al., 2018). Left panel: Comparison between measured and modelled lipid-normalized values of the logarithm of PCB-153 concentrations. Right panel: Comparison between the measured and modelled rank order of the concentrations in the 78 samples. The blue squares represent samples from 7 countries in West Africa, and the red triangles represent data from Hungary (2001 and 2006) and

Bulgaria (2014). Figures 3 and 4 of McLachlan et al., 2018.

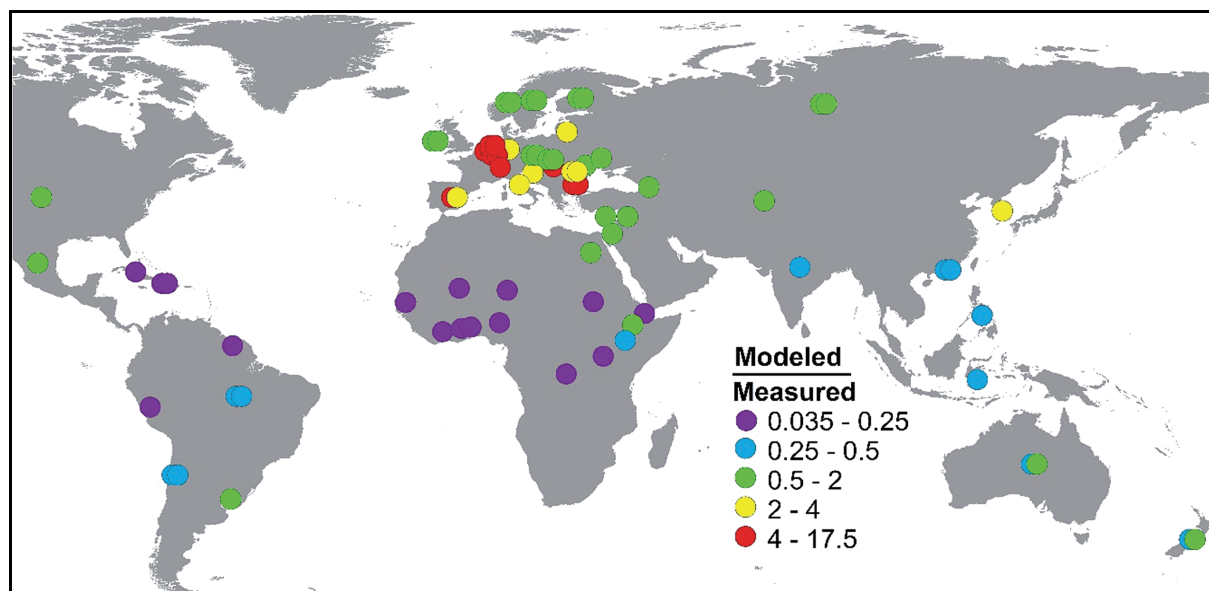


Figure 5.3.3. Quotient of the modelled/measured lipid normalized concentrations of PCB-153 in human milk in different countries. Measurement data collected under the UNEP/WHO Global Monitoring Plan; modelling data as described in McLachlan et al., 2018. Figure 5 of McLachlan et al., 2018.

The overprediction in Hungary and Bulgaria is an indication that the model may be predicting stronger gradients in concentrations across Europe than are measured (see also Figure 5.3.3). One possible explanation for this is the large amount of intra-European trade in agricultural goods that is not reflected in the modelling assumption that meat, dairy, and freshwater fish are sourced and consumed in the same country. A sensitivity analysis using European-wide averages did not improve the predictions for these outlying countries. Given the historically high emissions of PCB-153 in central Europe, strong gradients in PCB-153 would be expected, and this analysis highlights the need to use detailed and highly resolved data on food sourcing for regions with this characteristic.

In contrast to the overprediction of PCB-153 concentrations in human milk in European countries, underprediction of concentrations was seen for the seven West African countries (see Figure 5.3.3). One possible explanation is the under-estimation of recent emissions of PCB-153 in this region due to e-waste burning. A sensitivity analysis using a ‘worst-case’ scenario of the amount of e-waste that is burned in e-waste importing countries of 20% (compared to the default scenario of 5%) from the Breivik et al. (2016) emission inventory was conducted, and still resulted in underpredictions by a factor of roughly 10 or more of PCB-153 concentrations in milk. This indicates that other factors may be influencing the modelling framework’s ability to

capture the high levels of PCB contamination, such as contamination in imported food, processes occurring at a spatial scale finer than 3.75° X 3.75° or unrepresentativeness of the sampled population in regions with large spatial gradients in PCB concentrations.

In order to directly link POP emissions to toxic effects, one must understand the transfer from emissions to concentrations in vulnerable organisms, and in the case of human health outcomes to concentrations in particular people or groups of people. Modelling national averages and evaluating against the UNEP/WHO human milk database does not capture instances where specialized local or regional diets strongly influence the exposure potential to PCBs. Undeman et al. (2018) investigated the interplay of proximity to emission sources, sourcing of food, and food web structure on human exposure to PCB-153 employing a modelling framework similar to that used in the global case study by McLachlan et al. described above. They found that a diet composed of aquatic mammalian carnivores, particularly in Canadian Inuit communities, far outweighed the remoteness from PCB sources in determining exposure. PCB-153 concentrations for these Inuit communities were 6-8 times higher than a reference population in northeastern Europe that did not eat aquatic mammalian carnivores, despite having air and seawater concentrations that were 60 and 20 times lower, respectively. Such specialized diets are not explicitly considered in the approach taken by McLachlan et al. (2018) which (for the most part) considered exposure to PCBs for a representative population of each country.

This case study highlights the need for collecting detailed food sourcing data for regions with large gradients in environmental POP concentrations, as well as for better understanding of the environmental levels and exposure routes of POPs in Sub-Saharan African countries. Recent publications by Huang et al. (2020) and Moeckel et al. (2020) indicate that work on investigating exposure pathways for POPs in Sub-Saharan Africa is an area of active research.

5.3.3.2. Global Emission Inventory, Long-range Transport, and Environmental Distribution of the POP Dicofol

Dicofol is an organochlorine acaricide (USEPA, 1998; POPRC, 2013) that was a popular substitute for DDT due to the two chemicals' functional and structural similarity. Findings regarding dicofol's potential to harm wildlife and humans (Clark et al, 1990; Wiemeyer et al., 2001; Kojima et al., 2004; Reynolds et al., 2005; Hoekstra et al., 2006; Lesenger et al., 1991) led to the imposition of national and multinational restrictions (e.g., Weem, 2010; OSPAR, 2002), and to international regulation under Annex A of the Stockholm Convention in 2019 (POPRC, 2013, Decision SC-9/11). To support scientific assessment of dicofol's long-range transport, Li et al. (see Figure 5.3.4; see Li et al. 2015 for additional details) developed an inventory of global dicofol usage between 2000 and 2012 and used these estimates to conduct an investigation of the ability of dicofol to be transported from release regions in the lower and mid-latitudes to the Arctic and the subsequent fate characteristics of the chemical in the Arctic.

Depending on data availability, geographically-resolved estimates of dicofol usage were compiled using national reporting statistics, data on cultivated areas and dicofol application rates, and other regional or continental usage data, coupled with geographic constraints based on

dicofol-treated crops and agricultural intensities. Global usage data was generated for the years 2000-2012 and allocated to a 1° X 1° latitude/longitude grid (Figure 5.3.4 and Li et al. 2015 for additional details). A total of 28.2 kilotonnes (kt) of dicofol active ingredient were estimated to have been used globally between 2000 and 2012, with 77% of this in Asia and mainland China specifically contributing 69% of the total. Likely due to voluntary restriction and phase-outs at national and regional levels, as well as a decrease in its market share, annual global dicofol usage declined by a factor of 4.6 from 3.4 kt in 2000 to 0.73 kt in 2012. Areas of high dicofol emissions were identified in the low and mid-latitudes of the Northern Hemisphere and include central and eastern parts of China, India, countries on the Mediterranean coast, and California and Florida in the U.S.

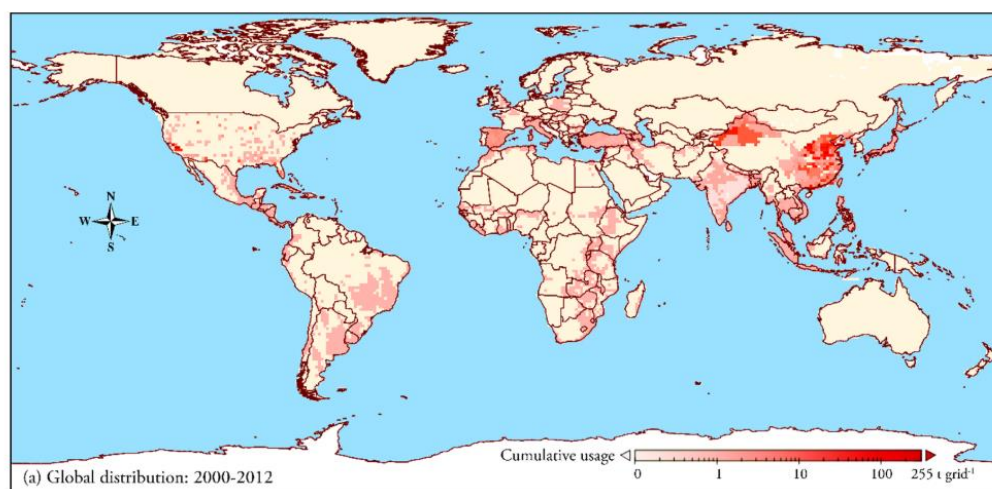


Figure 5.3.4. Estimated cumulative dicofol usage from 2000 to 2012 based on the emissions inventory compiled by Li et al., 2015. Grid spacing is 1° X 1° latitude/longitude. From Figure 2 of Li et al., 2015.

The gridded dicofol emission estimates were used to drive the BETR Global model at a 15° X 15° latitude/longitude grid spacing (MacLeod et al., 2011) to evaluate the spatio-temporal dynamics of the global transport and environmental distribution of dicofol between 2000 and 2012. The simulations indicated that dicofol can be transported northward, driven by atmospheric and oceanic advection, from its low- and mid-latitude source regions (see Figure 5.3.5). While the bulk of dicofol present in the atmosphere was distributed in low and mid-latitudes (panel B) — mirroring the geographic distribution of global emissions — seawater concentrations of dicofol were highest in Northern Hemisphere oceans north of 45° latitude (panel C). In some regions of the Arctic such as Alaska, Siberia, and northern Canada, modelled dicofol concentrations in soil were even comparable to those in tropical regions treated with dicofol (panel D). The excess contamination in northern oceans and soils relative to that in the atmosphere predicted by the BETR Global simulations suggests that dicofol has the potential to be enriched in the surface media of the Arctic.

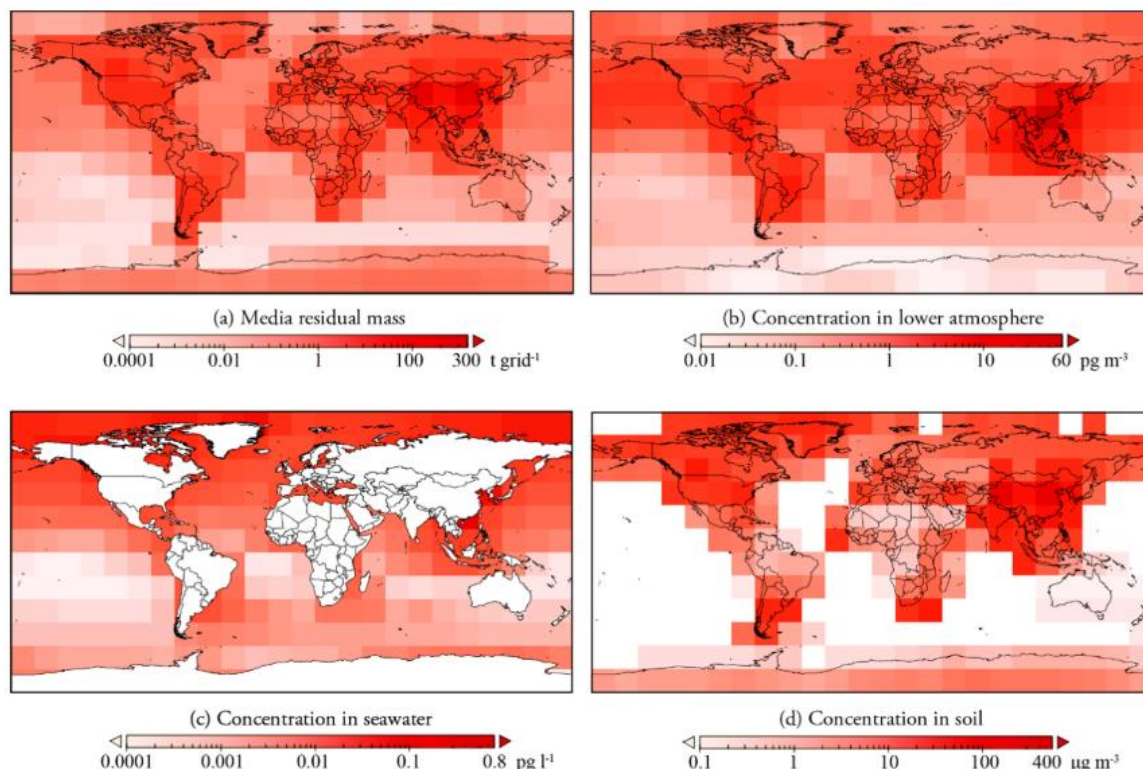


Figure 5.3.5. BETR Global modelling results of dicofol environmental distribution at the end of 2012 showing (a) total environmental burden, (b) lower atmosphere concentrations, (c) seawater concentrations, and (d) soil concentrations. Grid spacing is 15° X 15° latitude/longitude. Figure 3 of Li et al., 2015.

To assess the extent of the enrichment of dicofol in the Arctic surface media relative to legacy and candidate POPs, the authors further modelled the global fate and transport of dicofol using version 1.1 of the Globo-POP model (courtesy of F. Wania from the University of Toronto at Scarborough; see Wania and Mackay, 1995 & 2000). Globo-POP describes the global environment as 10 latitudinal climate regions. Results from this modelling indicated that with constant, continuous emissions of dicofol for a decade, 0.06% of the cumulative global dicofol emissions would be found in the Arctic surface media at the end of the tenth year (referred to as the absolute Arctic Contamination Potential (eACP)), and that 3.70% of the dicofol inventory in the global environment would be present in Arctic surface media at this time (referred to as the relative Arctic Contamination Potential (mACP)). Compared with 10 selected legacy and candidate POPs, dicofol has a moderate efficiency for long-range environmental transport given that it has an eACP value higher than three legacy POPs (heptachlor, aldrin, and hexabromocyclododecane) and has the strongest efficiency for accumulation in Arctic surfaces with an mACP value higher than all compared chemicals (see Li et al., 2015 for details).

Source region concentrations of dicofol were found to decrease more rapidly with concomitant decreases in emissions compared to environmental concentrations of dicofol in remote regions. For instance, despite a decline by a factor 4.6 in global dicofol emissions from 2000 to 2012, soil

and sediment concentrations in the Arctic only declined by factors of 3.9 and 3.5, respectively. This slower decline in surface media concentrations implies that once deposited on Arctic surfaces, dicofol has the potential to pose a long-lasting effect on the health of the ecosystem. The results of this modelling study also indicate that under the Global Monitoring Plan (GMP), monitoring atmospheric concentrations of dicofol in source regions would closely reflect the effectiveness of the Stockholm Convention in reducing or curbing the emissions of POPs. However, monitoring concentrations in surface media and biota in remote areas would provide a potentially different, but more complete, picture of the effectiveness of the Convention's implementation.

5.3.3.3. Global Gridded Emissions and Long-range Transport of Tris-(1-chloro-2-propyl) Phosphate (TCPP)

An expert working group recently noted that progress towards elimination of POPs is reflected in time trends in air, human milk and blood, but that it would be best assessed by tracking trends in emissions directly (<https://www.sciencedirect.com/science/article/pii/S1352231013004408>). Emission inventories for POPs are often compiled using a 'bottom-up' approach of collecting chemical production, usage, and emission factor data which is then extrapolated over space and time (e.g., see Section 5.3.3.2). An alternative is for modelers to tackle the problem of uncertain or lacking emission inventories more directly within the integrated approach by considering the problem "in reverse", i.e., by using monitoring data and models as a basis to propose scenarios that fill gaps in knowledge about the location, rate, and trend of emissions in a 'top-down' approach. The 'top-down' approach recognizes emissions as the most poorly constrained part of the integrated approach and makes predicting emissions, rather than environmental levels, the focus of modelling efforts. Emission scenarios that provide good agreement with monitoring data and available 'bottom-up' emission information provide a basis for formulating testable hypotheses that can be evaluated in further research work, which may include more focused work on 'bottom-up' estimates.

Tris-(1-chloro-2-propyl) phosphate (TCPP, CAS number 13674-84-5) is an organophosphate flame retardant (OPE) widely used in industrial products (van der Veen and de Boer, 2012). TCPP is not a Stockholm Convention POP, however it has been suggested as a POP candidate (<https://tema.miljodirektoratet.no/old/klif/publikasjoner/2871/ta2871.pdf>) and has been measured in remote regions globally (Sühling et al., 2016; Li et al., 2017; Castro-Jimenez et al., 2016; Salamova et al., 2014). TCPP is used as a replacement for flame retardants that have been more heavily regulated, including polybrominated diphenyl ethers (PBDEs) that have been banned under the Stockholm Convention (Marklund et al., 2005; Möller et al., 2012; Green et al., 2008). Estimates of the usage and emissions of TCPP to the environment are few and are poorly constrained (REF possibly their citations 15 and 19 as the only known emissions estimates). In addition, there are high uncertainties regarding physicochemical properties of TCPP, particularly degradation rates in air and water (<https://www.miljodirektoratet.no/globalassets/publikasjoner/klif2/publikasjoner/2871/ta2871.pdf>). Traditional 'bottom-up' emissions inventories of chemicals require knowledge of a chemical's production, usage, and emission factors from various use categories (e.g., Breivik et al., 2007), knowledge which is not currently available at the global scale for TCPP. Inspired by

previous modeling studies that aimed to estimate emissions at local and regional scales (Rodgers et al., 2018; Bogdal et al., 2014 A & B; Gasic et al., 2009; MacLeod et al., 2007; Buser et al., 2013; Schenker et al., 2009), a modified ‘top-down’ approach was taken by Li et al. (in press) to estimate global emissions of TCPP to air and water.

Measurements of TCPP in air (219 measurements from between 2006 and 2016) along with measurements in seawater (111 measurements from between 2010 and 2017) were compiled from literature sources (see Li et al., in press, for details). Measurement data from rural and remote areas, rather than urban areas, was selected as appropriate for comparison to the modeled concentrations. Measurements collected from the literature were averaged within BETR Global model grid cells, resulting in measurements in air for 130 grid cells and in water for 23 grid cells that could be used in the integrated assessment.

To simulate the fate of TCPP in the environment, the BETR Global model was employed with monthly time step and $3.75^\circ \times 3.75^\circ$ (lat/long) grid spacing (MacLeod et al., 2011; McLachlan et al., 2018). Scenarios using three potential gas phase atmospheric half-lives of TCPP were employed (12, 60, and 300 hours) to investigate the plausibility of the range of half-lives proposed by Li et al. (2017), and an analogous range of potential half-lives in water was also investigated (1440, 7200, and 36000 hours), with the value of 1440 hours being based on an estimate from EPI Suite 4.0 (see USEPA, 2017 for latest version). As a first step, unit emissions of TCPP to air were distributed globally to land-based emission grid cells proportional to artificial nighttime light intensities using the 2010 Defense Meteorological Satellite Program/Operational Linescan System (DMSP/OLS) dimensionless nighttime light index (NGDC, 20XX), as a proxy for intensity of industrialization (MacLeod et al., 2011; von Waldow et al., 2010; Huang et al., 2015; Jiang et al., 2017; Göktas et al., 2016). A range of scenarios for emissions to water were explored, including zero emissions to water and emissions to water equal to one-half and one times emissions to air. In total, 27 modelling scenarios were investigated employing all unique combinations of emission parameters and the degradation rates in air and water. The model was run assuming constant emissions to a state in which concentrations of TCPP during consecutive simulated years were repeated. Thus the assessment provides an estimate of the average emission rate of TCPP during the period when measurements were made, ie, 2006 to 2016.

The results of the unit emissions modelling simulations across the 27 scenarios were compared with the measurement data to derive scenario-specific global emissions scaling factors that provide the best agreement between measured and modelled concentrations in each scenario. The single scenario that was judged to provide the best agreement between model predictions and measurements was that with half-life in air, half-life in water, and emissions to water being 60h, 7,200h, and $E_w=0.5E_A$ (respectively), with total global emissions being 39.5 kt yr^{-1} . It was not possible to improve model performance by adjusting the relative emission rates of TCPP independently in seven (sub) continents indicated in Figure 5.3.7, meaning that the initial spatial allocation of TCPP emissions according to intensity of nighttime light intensities could not be refined. Figure 5.3.6 displays the regression between \log_{10} values of observed and modelled

concentrations of TCPP in air and water using the preferred modelling parameters, while Figure 5.3.7 shows the globally-gridded emissions to air under this preferred scenario.

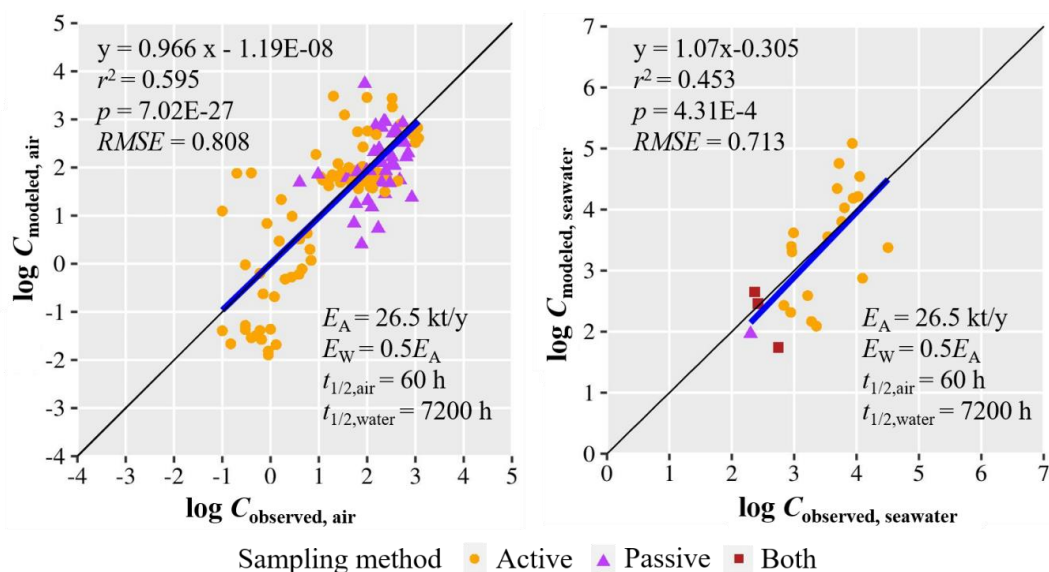


Figure 5.3.6. Comparison of the base-10 logarithm of modelled and observed concentrations of TCPP in global air (left panel) and seawater (right panel) based on the measurements and modelling described in Li et al. (in press). Modelling is based on the scenario with global TCPP emissions to air of 26.3 kt yr^{-1} , global emissions to water of 13.2 kt yr^{-1} , and half-lives for degradation of TCPP of 60 hours in air and 7200 hours in water. Black line indicates the 1:1 comparison line; blue line indicates the regression line. Figure 1 of Li et al. (in press).

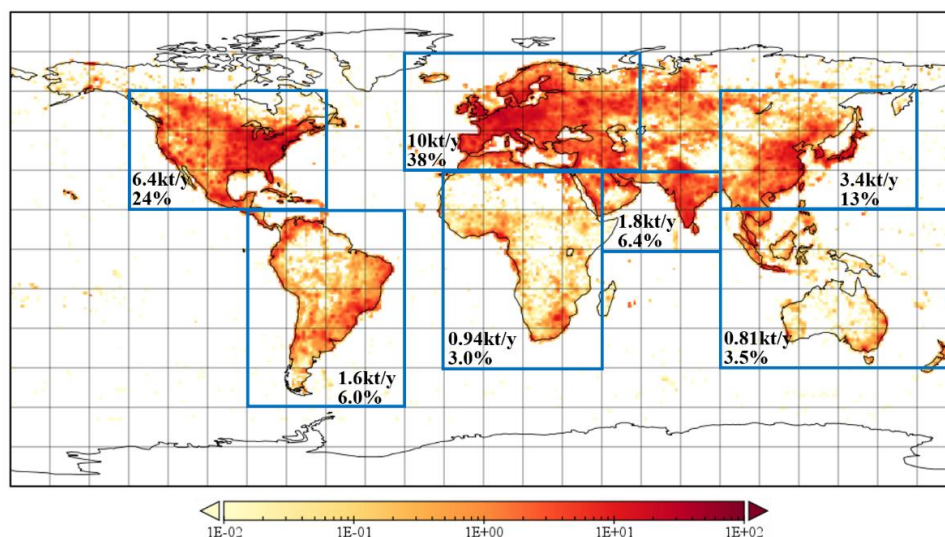


Figure 5.3.7. Global gridded inventory of TCPP emissions to air inferred from the integrated 'top-down' modelling approach described in Li et al. (in press) under the preferred scenario for TCPP emissions to air (i.e., 26.5 kt yr^{-1}) and environmental degradation half-lives (i.e., 60

hours in air and 7200 hours in water). Annual emission rates for different (sub) continents (blue rectangles) are indicated. Grid spacing is $1^\circ \times 1^\circ$ latitude/longitude. Figure 2 of Li et al. (in press).

Globally, the root mean square error (RMSE) of modeled atmospheric concentrations was 0.81 log units. Isolated occurrences of relatively large differences between modelled and measured values are found in several locations such as Barbados (0.03), the Great Lakes region (>20), Czechia (62), and Indonesia/Australia (200-400) (Figure 5.3.8, top panel). In seawater the RMSE was slightly lower than in air (0.71 log units), but TCPP measurements in seawater do not provide good global coverage, which results in higher uncertainties in the entire assessment than for atmospheric measurements (Figure 5.3.8, lower panel).

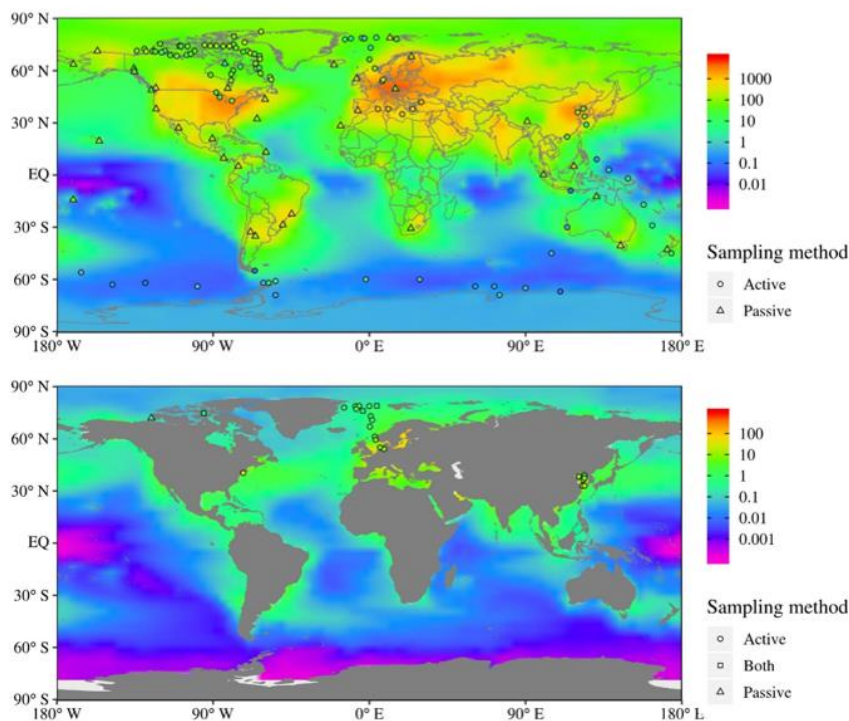


Figure 5.3.8. Modelled (color gradient in the background) and observed (colored symbols) concentrations of TCPP in air (pg m^{-3} ; top panel) and seawater (ng L^{-1} ; bottom panel) under the preferred scenario for TCPP emissions to air and water (i.e., 26.5 kt yr^{-1} and 13.3 kt yr^{-1} , respectively) and environmental degradation half-lives (i.e., 60 hours in air and 7200 hours in water). Modelled values are annual mean concentrations. Modelling data grid spacing is $3.75^\circ \times 3.75^\circ$ latitude/longitude. From Figures 3 and 4 of Li et al. (in press).

Limitations and sources of potential bias within the integrated assessment conducted by Li et al. (in press) include the lack of interannual variability in TCPP emissions, uncertainty in the

measurement data accuracy (both in terms of the absolute value as well as the geographic area covered by these measurements), and assumptions about physico-chemical properties of TCP that were not reflected in the scenario analysis. While this investigation of TCP should be considered preliminary, it provides valuable insights about the link between emissions and concentrations in the global environment. Plausible environmental fate parameters have been established for TCP in air and the ‘top-down’ emission inventory provided by this assessment serves as a reference point for future studies that could establish temporal and spatial trends.

5.3.3.4. Modelling Long-Range Transport of PFOS in the Ocean

Perfluorooctanesulfonate (PFOS) and other perfluoroalkyl acids (PFAAs) are part of a class of industrial chemicals, the perfluoroalkyl substances (PFASs), with large historical production volumes (Armitage et al., 2009b; Wang et al., 2014). PFAAs are extremely persistent in the environment and can adversely affect human and ecological health (Sunderland et al., 2019). PFOS (Annex B) and perfluorooctanoic acid (PFOA, Annex A) are regulated under the Stockholm Convention, and perfluorohexane sulfonate (PFHxS) is proposed for listing (Stockholm Convention, 2020). The ocean is recognized as the terminal sink for PFAAs released to the environment, and measurements of PFOS, PFOA and PFHxS in water are reviewed in Chapter 5.2 of this report. Zhang et al. (2017) conducted an emissions inventory and modelling case study of PFOS in the North Atlantic Ocean that illustrates application of the integrated approach to a PFAA with diffuse emission sources that provides valuable insights into current emission rates and fills gaps in available measurement data.

Spatially and temporally resolved oceanic discharges of PFOS were estimated from wastewater monitoring data at the river-catchment level in North America for the year 2010 (Figure 5.3.9, A) and from river monitoring in Europe for 2006-2007 (Figure 5.3.9, B). The estimated discharges from these discrete time periods were extrapolated to a range of initial estimates of total time-dependent PFOS inputs to the North Atlantic between 1958 and 2010 (Figure 5.3.9, C) based on global historical use and emission inventories (Armitage et al., 2009b; Wang et al., 2014). This initial emissions inventory was mapped onto a 1° x 1° latitude/longitude grid to drive an oceanic fate and transport model.

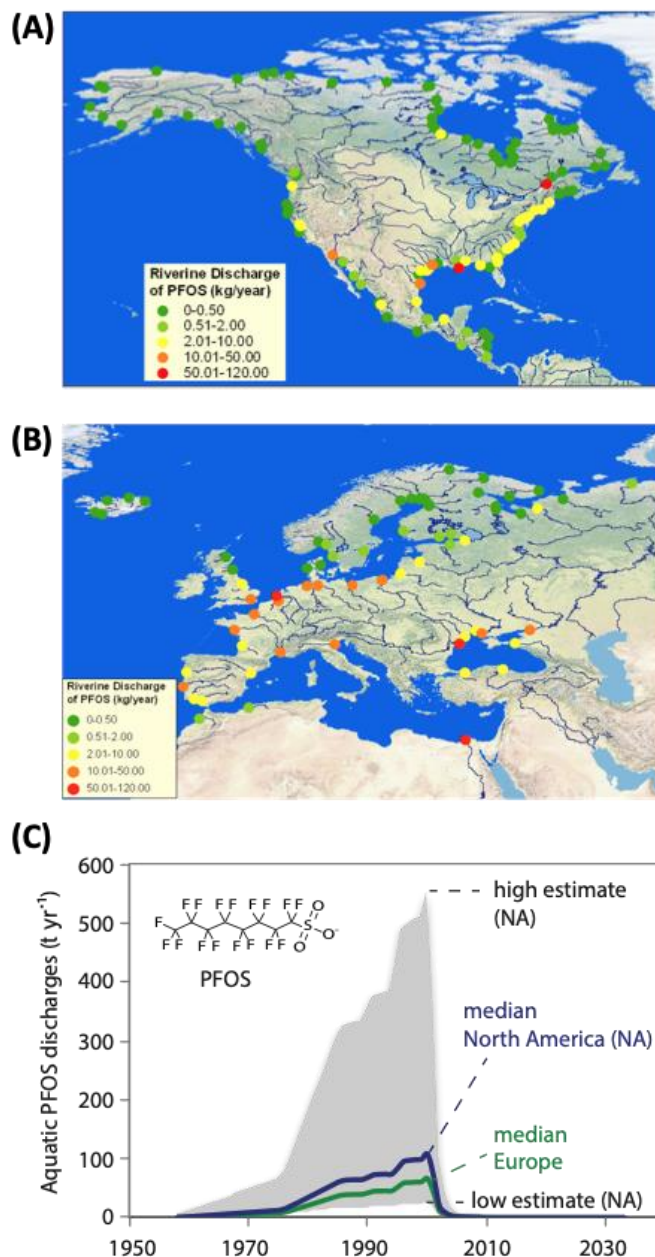


Figure 5.3.9. PFOS discharges (kg yr^{-1}) in 2010 from rivers in North America to the ocean (panel A) and from major rivers in Europe for 2006-2007 (panel B). North American discharges are based on the approach developed by Zhang et al. (2017), and the European discharges are based on the catchment population regression developed by Pistocchi and Loos (2009). Panel C displays extrapolated continental discharges of PFOS (t yr^{-1}) to the North Atlantic Ocean (20°N - 60°N) from European and North American (i.e., U.S. and Canada) wastewater and rivers from 1958 to 2010. Solid lines indicate median discharge estimates while the shaded grey region indicates the 95% confidence interval for North American discharges. Panel C is Figure 1 of Zhang et al., 2017.

The Massachusetts Institute of Technology general circulation model (MITgcm; <http://mitgcm.org>), a 3-D ocean circulation model, was used to simulate PFOS transport in the ocean through lateral and vertical oceanic circulation, mixing and particle settling. The model has a horizontal grid size of $1^\circ \times 1^\circ$ latitude/longitude and includes 23 vertical levels (see Zhang et al., 2017 for additional details on the model setup). For the years 1958 to 2010, the model was forced with $1^\circ \times 1^\circ$ gridded inputs to the North Atlantic Ocean between 20°N and 60°N from rivers and wastewater treatment plants in North America and Europe using the inventory developed here. Between 2010-2038 inputs are assumed to be zero to simulate elimination of PFOS production, use and emissions.

Modeled seawater concentrations are highly correlated with measurements ($R^2 = 0.90$, $p = 0.01$) but the model based on the median of the initial release estimation had a mean bias of 75% relative to observations for all regions. Adjusting the emissions produced mean modeled surface water (10 m) concentrations of $39 \pm 14 \text{ pg L}^{-1}$ in 2010 compared to an observed value across all offshore sampling locations of $43 \pm 21 \text{ pg L}^{-1}$ between 2009 and 2011. The adjusted releases represent a refined hypothesis about historical emissions that incorporates both ‘top-down’ and ‘bottom-up’ information from the integrated approach. According to these refined estimates, cumulative PFOS inputs to the North Atlantic Ocean were $2.4 \times 10^3 \text{ Mg}$. North American coastal releases accounted for 59% of total releases and the remaining 41% was from Europe (Figure 5.3.9).

The major surface currents (modeled at 10 m depth) in the Atlantic Ocean result in predominantly eastward and northward transport of coastal PFOS pollution toward the Subarctic (Figure 5.3.10). PFOS enriched seawater plumes from the St. Lawrence River, Florida Current, and North Sea are clear features of the modeled concentrations in 1980 and 2000, with concentrations more than an order of magnitude higher than the open North Atlantic. By 2020 these plumes have largely dissipated due to reductions in emissions (Figure 5.3.10).

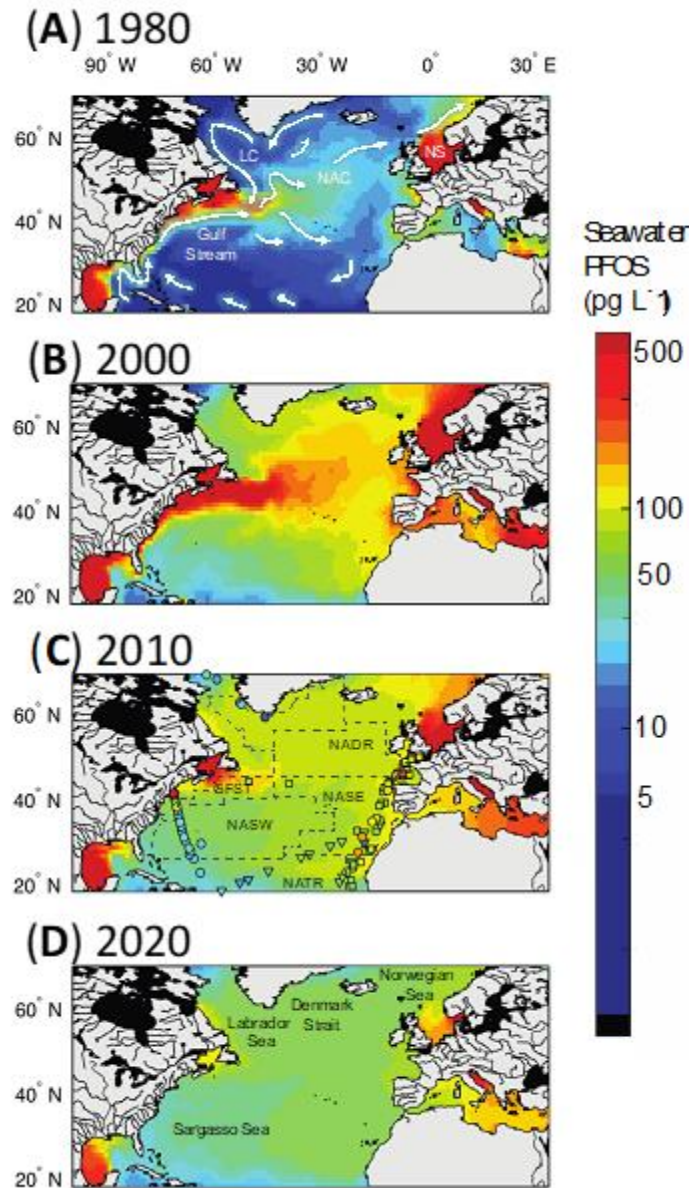


Figure 5.3.10. Modelled temporal evolution of surface water (10 m) PFOS concentrations (pg L^{-1}) in the North Atlantic Ocean from 1980 to 2020, as described in Zhang et al., 2017. Major surface currents are indicated by white arrows in Panel (A). Panel (C) shows modelled surface concentrations for 2010 compared to observations from 2009-2011 obtained from: Ahrens et al. (2009a; 2009b; 2010b); Benskin et al. (2012); Gonzalez-Gaya et al. (2014); Zhao et al. (2012). Current labels: NAC = North Atlantic Current, LC = Labrador Current, NS = North Sea. Longhurst Biogeographical Provinces (Longhurst, 2007) abbreviations: NADR = North Atlantic Drift Province, GFST = Gulf Stream Province, NASW = North Atlantic Subtropical Province - West, NASE = North Atlantic Subtropical Province - East, NATR = North Atlantic Tropical Gyral Province. From Figures 2 and 4 of Zhang et al., 2017.

The mass distribution of PFOS shifts from the surface ocean to the deeper ocean over time (Figure 5.3.11). Variability in the 95th percentile confidence interval of surface water PFOS concentrations (panel A) is quite large for the first two-thirds of the simulation, indicating large

spatial variability across the modelled domain, though this decreases over time (which is also evident in Figure 5.3.10). PFOS concentrations peak in 2001 in the surface layer (shortly after peak emissions around 2000), while the peak in subsurface concentrations lags by 2-3 years (roughly 2005, panel B). Peak PFOS concentrations lag even longer after peak emissions for seawater near the permanent thermocline (panel C), with concentrations not expected to plateau until approximately 2040.

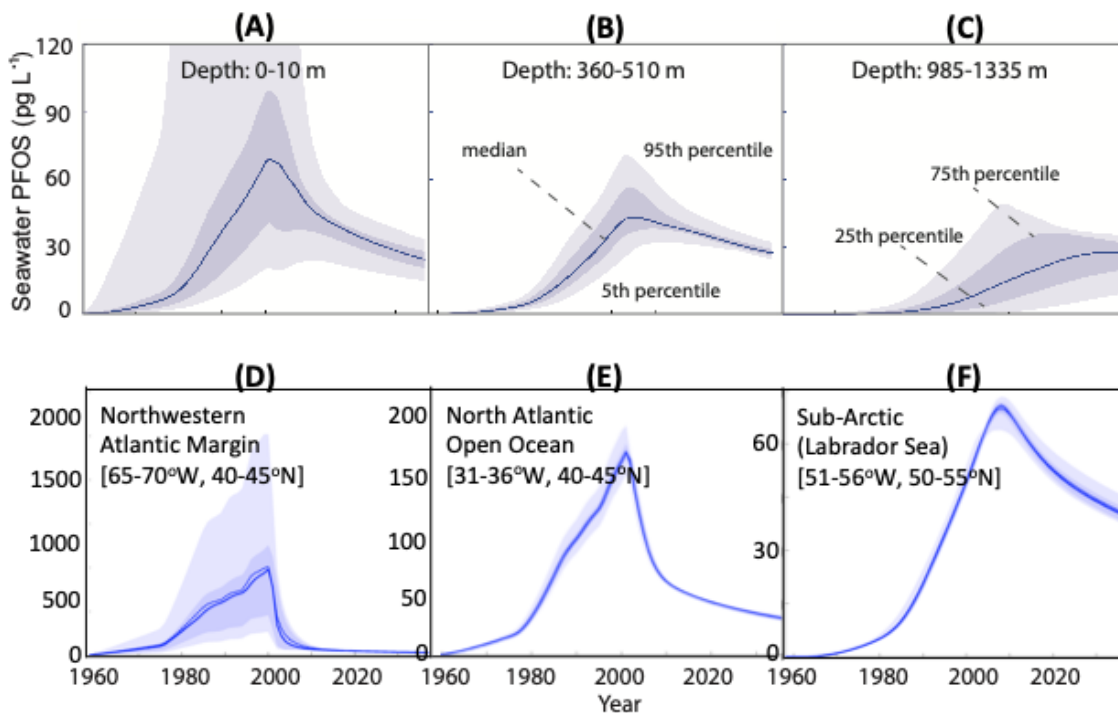


Figure 5.3.11. Modelled PFOS concentrations in North Atlantic seawater ($20^{\circ}\text{N} - 60^{\circ}\text{N}$; pg L^{-1}) at the surface mixed layer (A), the subsurface (B), near the permanent thermocline (C), and for the surface mixed layer within different $5^{\circ} \times 5^{\circ}$ latitude/longitude geospatial regions (D-F) based on the updated PFOS release scenario shown in Figure 5.3.9. Solid lines indicate the basin- or region-wide median concentration, darker shading indicates the 25th and 75th percentile concentrations, and lighter shading indicates the 5th and 95th percentile concentrations. Panels (A-C) are Figure 5 of Zhang et al., 2017.

Temporal trends in PFOS in the surface mixed layer (0-10 m) are highly location specific, which is evident in three $5^{\circ} \times 5^{\circ}$ latitude/longitude regions at different locations in the North Atlantic Ocean (Figure 5.3.11, panels D, E and F). For the region closest to sources (panel D), large spatial variability exists before PFOS source control in the early 2000s. After source controls are implemented, median seawater concentrations and spatial variability decrease rapidly in this $5^{\circ} \times 5^{\circ}$ region. For the open ocean (panel E), PFOS concentrations only decrease by one third over five years following source control. In the subarctic region (panel F), PFOS concentrations reach their maximum 8-9 years after source control due to the time lag for transport to this region. After the peak, and after dilution into the whole ocean occurs, concentrations decrease at a slow rate with a half time of over 20 years.

Figure 5.3.12 shows gross depth-specific PFOS flows into and out of the Arctic from the North Atlantic from 1958 to 2038. Most of the flux of PFOS into the Arctic occurred from mid-depth

inflow (20-1000 m; 1.1×10^3 Mg) relative to that from surface inflow (0-20 m; 60 Mg). Entrainment of PFOS into the Atlantic meridional overturning circulation (AMOC) is estimated to have prevented 5.3×10^2 Mg PFOS from entering the Arctic (see Zhang et al., 2017 for details). A weakening of the AMOC due to climate change (Fisher et al., 2010; Rahmstorf et al., 2015; Rhein et al., 2015; Yashayaev, 2007) could thus result in an increase in PFOS entering this region.

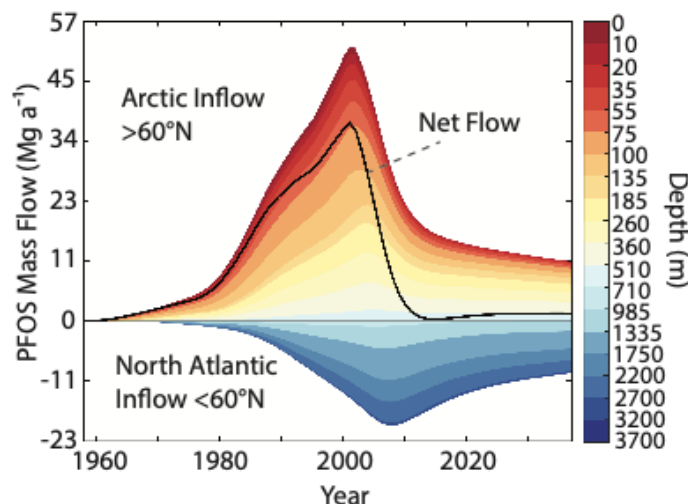


Figure 5.3.12. Modelled PFOS circulation above and below 60°N at various seawater depths from the years 1958 to 2038. Blue shading indicates deeper waters while red, orange, and yellow shading indicates surface or subsurface waters. Net flow is indicated by the solid black line, with positive numbers representing flow into the Arctic region (i.e., above 60°N) and negative numbers indicating flow into the North Atlantic (i.e., below 60°N).

The modelling conducted by Zhang et al. (2017) illustrates high spatial variability in PFOS ocean concentrations and lag times between elimination of emissions and declining trends in environmental concentrations that vary strongly as a function of geographic location and ocean depth. Long-term seawater monitoring sites for PFAAs have not been established so far, but if they are they should be located within a region where spatial variability is relatively small. Such monitoring sites could be at the ocean margin where PFAA concentrations respond quickly to changes in continental source strength and impact on the marine ecosystems occurs. Monitoring sites located where ocean currents deliver PFAAs to remote regions such as the Arctic would provide valuable information in integrated assessment of long-range transport. In addition to changes in source strength, the observed temporal trends from long-term monitoring can be influenced by other factors such as climate change. The modeling indicates that the AMOC has prevented substantial amounts ($\sim 5.3 \times 10^2$ Mg) of PFOS from entering the Arctic, and that the flux of other persistent chemicals similar to PFOS into the Arctic may increase with climate change-driven weakening of the AMOC.

5.3.4 Discussion and Recommendations

The four case studies presented in this section illustrate the value of applying models as a framework to support the integrated assessment of emissions, fate and transport, and

environmental concentrations of POPs. Using models to formalize the link between emissions and concentrations of POPs and POP candidates in air, water, soil, wildlife and humans has a wide range of benefits for supporting the development of the Global Monitoring Plan and for effectiveness evaluation of the Stockholm Convention. These include:

Spurring the development and refinement of emission inventories for POPs and POP candidates. By linking monitoring data back to emissions, the integrated approach focuses attention on evaluating the effectiveness of the Convention on encouraging emission reductions, and provides a more direct metric of effectiveness, ie, the rate of emission reduction. Parties to the Convention currently provide estimates of chemical production and/or consumption that can be compared to 'bottom-up' emission estimates based on usage and emission factors, and to 'top-down' emission estimates that lead to good model performance relative to monitoring data. Comparing these different types of emission estimates can help to close gaps in knowledge about fate and transport processes, and to identify unknown sources of POPs and improve the effectiveness of measures undertaken nationally and internationally to control releases.

Aiding in the design of monitoring programs and quality assurance of monitoring data. Effectiveness evaluation of the Stockholm Convention under the Global Monitoring Plan uses measured trends in levels of POPs in the environment and in human milk and blood as indicators of reductions in emissions to the environment. As more POPs, and POPs with more diverse properties and environmental fate profiles, are added to the Convention the costs for monitoring will increase and there will be increasing incentive to optimize monitoring strategies. Models provide information about locations and environmental media where monitoring will provide the highest leverage information for reducing uncertainty about levels and trends of POPs and POP candidates. Additionally, models provide a basis for evaluating monitoring data obtained from new locations or new analytical techniques for consistency and accuracy compared to the weight of evidence represented by the entire integrated assessment.

Understanding trends in POP levels in a changing climate. Climate change has the potential to affect POPs in a myriad of ways, some of which might affect temporal trends that are used for evaluating effectiveness of the convention. By providing mechanistic insights into the behavior of POPs in the environment and the emissions-to-concentration relationships, models can help to make an accurate assessment of the effectiveness of the convention in a changing global climate.

In order to fully realize these benefits the following research and monitoring priorities are recommended:

- 1) *Support the development of global scale modeled 'integrated assessments' of emissions, fate and transport processes and environmental levels and trends for all POPs and POP candidates.* Currently, global-scale case studies are only available for a select few POPs and POP candidates. In order to realize the benefits of these assessments it is recommended to compose a complete case study for each POP and POP candidate that

can be updated regularly and refined as new information is produced relevant to emissions, fate and transport properties and environmental levels and trends.

- 2) *Apply the principles of integrated assessment at local, regional and global levels to improve emission estimates.* High uncertainties and missing information in emission inventories is often the dominant source of uncertainty in integrated assessments of POPs. As reviewed in the 2nd Regional Monitoring Report of the WEOG Region, the integrated approach can be applied at local and regional scales and near suspected source areas to infer emission rates of POPs that can then be scaled using emission factors to compare with national estimates and to drive global scale models.
- 3) *Support the development and improvement of POP modeling capabilities and laboratory and field research to understand POP properties.* Important research needs to improve POP models include improving modeling of gas/particle partitioning in the atmosphere and of air/surface exchange in general. Uncertainties in assessment of individual POPs are often driven by uncertainties in physico-chemical properties and degradation rates in air, water and soil which could be reduced using targeted laboratory and field studies. Models can be evaluated and iteratively improved by identifying locations where observations would most effectively constrain model uncertainties and targeting field monitoring at those locations. Capabilities to model source-to-human body burden relationships for POPs would benefit from better resolving food sourcing patterns especially in poorly understood/high exposure areas.

6 CONCLUSIONS AND RECOMMENDATIONS

6.1 Findings and Conclusions

This section provides a brief synopsis of the major findings of the WEOG report and key messages for the Conference of the Parties.

6.1.1 Summary of the baseline concentrations

The concept of availability of ‘baseline information’ on POPs in this report has less to do with a specific measurement at a point in time and more to do with the state of knowledge for a particular substance. In this context, the geographic coverage of this information, across the WEOG region, is also an important requirement for an ‘adequate’ baseline. Many of the original POPs listed under the Stockholm Convention were already regulated or banned by countries long before 2004. Consequently the ‘baseline’ period for the GMP, may represent concentrations that have already decreased substantially from their peak levels or concentrations that are now levelling off. In some cases secondary sources have begun to dominate emissions (e.g. emissions from POP reservoirs such as oceans, soils and ice; see section 5.3). Therefore, for the ultimate purpose of attributing reductions in environmental levels of POPs to the ‘effectiveness’ of the Convention, it is more appropriate to consider baseline data and trends associated with the newer POPs. **Table 6.1a** summarizes the availability of ‘baseline’ information on POPs across the WEOG region.

It is important to note that with respect to POPs in human tissues (i.e. milk and blood) much of the WEOG region data comes from national programmes such as, *inter alia*, NHANES and ESB (or the Australian Pilot Monitoring Programme). Such programmes are important for assessing country-scale time trends however the levels are often not directly comparable to other countries/regions due to different study designs and populations. Similarly, individual research studies that are not a part of any programme can indicate whether the levels of pollutants are of concern in a certain area but cannot directly be used to evaluate differences between regions or over time. It is therefore challenging to set a “general” baseline concentration for the WEOG region for POPs in human blood and/or milk.

Table 6.1a Summary of available baseline information on POPs in core media in the WEOG region.

Chemical	Air	Human Tissues*
aldrin		ND
chlordane		
chlordecone		
Dicofol		Not included
DDT		
dieldrin		
endosulfan		
endrin		ND
HBB		
HCB		
HCB		
α HCH		
β HCH		
γ HCH		
Heptachlor		
mirex		
PBDEs		
PBDE-209 (Deca)		
Pentachlorophenol		
PCNs		Only Swedish data
Σ PCBs		
PCDD/Fs		
PFOS		
PFOA		
PFHxS		
PeCB		
SCCPs		
toxaphene		

	Adequate information on baselines
	Limited information on baselines
	No information on baselines

* these should be taken as general indicators since information varies between countries and subregions within WEOG.

ND – non detect (low levels often below detection limit)

Likewise for air, while air monitoring programmes provide the opportunity to derive time trends and region-specific spatial comparisons (e.g. the Arctic, the Great Lakes, the Alps and the UK),

concentrations reported by different air monitoring programmes are not directly comparable because of the differences in sampling methods (e.g. active versus passive sampling methods), analytical procedures, sampling frequencies, target compounds and programme QA/QC protocols. So it is also not possible to provide baseline concentrations of POPs in air that would apply to the entire WEOG region. Air monitoring programmes have started or are planning to measure the newer POPs (e.g. SCCPs, HBCDD, PFOA and related chemicals) to determine baseline concentrations, which will be available for the next reporting period of the GMP.

New air monitoring networks have been established to provide better spatial coverage of POPs, notably the Spanish Air Monitoring Program (SMP) (after 2008), the Australian National Passive Air Sampling and Archiving Program, and stations in Antarctica. There is also increased coverage in air measurements in the Arctic under AMAP with the Villum Research Station Nord (Greenland) initiated in 2008 and two Russian stations in 2015.

This assessment addressed the questions of temporal trends of PFOS, PFOA and PFHxS in water and the broad spatio-geographic trends within WEOG countries as well as in neighboring seas (Baltic, North, Mediterranean) and oceans (North Atlantic, Arctic Ocean, Northeast Pacific). Unlike air measurements of PFAS, surface water sampling has not been consistently or repeatedly sampled at the same locations. Additionally, the dynamic nature of rivers, lakes, seas and oceans, along with seasonal differences in river flow, lake thermal stratification, and ice cover adds to variability of concentrations. Nevertheless with results from over 3000 locations and over 10,000 individual measurements within the WEOG region, including 1300 analyses for samples collected from 2015-2019, it is possible to qualitatively examine temporal and spatial trends using medians and estimates of data dispersion (around the median) of the concentrations. The three PFAS compounds showed distinct differences in median concentrations between inland surface waters in Western Europe and Canada/USA. Median PFOS concentrations for Canada/USA were 2.5 fold higher at non-urban (mainly riverine) sites in 2015-2019 compared to Western Europe. Median PFOA and PFHxS were 3.0-fold and 1.9 fold higher, respectively, in Canada/USA in 2015-2019, compared with Western Europe. Early baseline trend indications for water in the WEOG region are summarized in Table 6.1d.

Current levels in non-core media of legacy POPs such as Dieldrin, Mirex, DDT, PCB, HCB and HCH are substantially below the levels in the 1970s, are now in the low ng/g range, and have not significantly decreased since 2000. Even when there are no new sources, the existing pool of pollutant can be of concern in some areas for decades. Levels of Endosulfan, PFOS, PBDE, and HBCD have increased over the past decades and are similar to the current levels of legacy pollutants. Although the levels in most biota are below the expected levels of effect (with the

exception of ‘hot spots’), the increasing load and increasing number of substances (including some newly listed POPs, POP candidates, and POP-like chemicals that have not yet been assessed) present reasons for concern and sustained attention.

6.1.2 Summary of evidence of temporal trends

In the first GMP report of the WEOG region, temporal trends were already available for many of the original POPs listed under the Convention. These trends established a baseline trend against which future trends could be compared. Many of the existing WEOG monitoring programmes and some of the new programmes have already included newly listed POPs in their target lists. In some cases, temporal information is already becoming available however, in most cases, the time series is not yet sufficient for assessing trends. It is challenging to summarize the availability of temporal trend information for POPs in human blood and/or milk since it varies between countries and subregions within the WEOG.

Table 6.1b summarizes the availability of trend information on POPs in the core media. Water is treated separately in Table 6.1d.

The levels of several of the initial Stockholm Convention POPs such as PCBs, DDTs, PCDDs/PCDFs, HCB, Toxaphene, Chlordanes and Aldrin, Dieldrin and Endrin are decreasing over time in human milk and/or blood. PCP levels seem to decrease as well even though data are quite scarce. This shows that banning of production and use of these man-made chemicals has lead to declining levels in humans. Some of the newer POPs show an increase over time followed by a decrease. This is the case for brominated flame retardants PBDEs and HBCD. Data for PFOS and PFOA also indicate an increasing trend accompanied by a decrease. This further confirms that adequate risk management measures lead to declining levels in humans. The rising and then decreasing trend is likely due to the fact that the risk management measures for these newer POPs were taken later than for the initial POPs. No clear trend over time could be seen for HCHs, Heptachlor, and Mirex. And, due to lack of enough data, no clear trend could be determined for PBBs (including HBB), PCNs and Endosulfan. It is not clear why no clear trend can be observed for HCHs, Heptachlor and Mirex, but it is likely due to the variability among data sources. Monitoring data is lacking for SCCPs in human milk/blood, despite the large scale production and use of this chemical. Monitoring data is also lacking for Chlordecone and HCBd.

Table 6.1b Summary of available temporal trend information on POPs in core media in the WEOG region.

Chemical	Air	Human Tissues
aldrin		ND
chlordane		
chlordecone		
Dicofol		Not included
DDT		
dieldrin		
endosulfan		
endrin		ND
HBB		Only US data
HCBD		
HBCD		
HCB		
α HCH		Trend not investigated
β HCH		
γ HCH		Trend not investigated
Heptachlor		
mirex		
PBDEs		
PBDE-209 (Deca)		
Pentachlorophenol		Only German data
PCNs		
Σ PCBs		
PCDD/Fs		
PFOS		
PFOA		
PFHxS		
PeCB		
SCCPs		
toxaphene		

	Adequate information on temporal trends
	Limited information on temporal trends
	No information on temporal trends

* these should be taken as general indicators since information varies between countries and subregions within WEOG.

ND – non detect (low levels often below detection limit)

National and international long-term monitoring programmes continued to provide time trends of POPs in air during this review period. Temporal trends for older POPs that were previously regulated continue to decline or have levelled off in air. There are some exceptions, for instance, HCB and PCBs in Arctic air are showing increasing trends at specific locations that may be related to warming and their unintentional release. Warming has led to enhanced revolatilization from oceans and other reservoirs of POPs (e.g. due to reduced sea-ice and melting glaciers). Some of the recently listed POPs (e.g. PBDEs, PFOS and precursors) have started to show declining trends across the WEOG region. For instance, most PBDEs are showing declining trends in air in most parts of the WEOG region. However, decaBDE is still increasing in air or not changing over time, likely reflecting the time-lag in regulating decaBDE compared to the other PBDEs. PFOS, PFOA and their precursors showed an increase in air concentrations over time at some locations, reaching a peak and have now started to decline. Time trends of some POPs (HBCDD, SCCPs, PCP/PCA, HCB, PeCB, and toxaphene) and air concentration data of HBB, chlordecone, dicofol, and PCNs, are lacking probably due to difficulties in chemical analysis, low or non-detectable concentrations in air, or sampling artefacts (e.g. breakthrough in active air sampling or high blank levels). The accelerated decline of endosulfan was observed in air. This is perhaps the best example of “effectiveness” of regulation for a widely used POP during the Stockholm Convention period.

Table 6.1c shows changes over time in POP concentrations measured in more media for the WEOG region. It is challenging to summarize the changes of temporal trend information for POPs in human blood and/or milk since it might vary between countries and subregions within WEOG.

The information available for water provides knowledge of spatial trends for PFOS, PFHxS and PFOA across western Europe, Canada/USA and southern/eastern Australia, the North Sea, the Baltic, the Great Lakes, and in the North Atlantic, Arctic Ocean and Northeast Pacific. Since no specific studies of temporal trends of PFAS in water were found, medians for combined data from specific regions with the periods 2000–2009, 2010–2014 and 2015–2019 were compared and are summarized in Table 6.1d. A challenge with these comparisons is that all data are included but sampling sites may vary for each study. A major knowledge gap is measurement of PFOS, PFHxS and PFOA precursors, both neutrals (MeFOSA, EtFOSA, MeFOSE, EtFOSE, fluorotelomer alcohols) as well as anionic and amphoteric chemicals. Recent studies have suggested they are widespread in surface waters (D'Agostino and Mabury, 2017; McGuire et al., 2014; Zhang et al., 2019).

Table 6.1c Summary of changes over time in POP concentrations measured in core media for the WEOG region.

Chemical	Air	Human Tissues**
aldrin		
chlordane		
chlordecone	No data	
Dicofol	No data	Not included
DDT		
dieldrin	No change at some sites	
endosulfan		
endrin	*	
HBB	No data	
HCBD	*	
HBCD	Few detections; decline at one site	
HCB	Slight increase at some polar sites	
α HCH		Not investigated
β HCH		Decrease in milk but not in blood
γ HCH		Not investigated
Heptachlor	Declining at some sites	
mirex		
PBDEs		
PBDE-209 (Deca)	Increasing at some sites	
Pentachlorophenol	* reported as PCA	*only German data
PCNs	No data	
Σ PCBs	Increase in the Alps	
PCDD/Fs		
PFOS	*	
PFOA	*	
PFHxS		
PeCB	Decrease in the Alps	
SCCPs	*	
toxaphene		*

	Generally decreasing trends
	Increasing trends
	No change or cannot establish trend
	Insufficient trend data

* warning to indicate limited data. ** these should be taken as general indicators since information varies between countries and subregions within WEOG.

Table 6.1d Trends for non-urban sites based on comparison of medians for 2000-09 vs 2015-2019 (unless otherwise noted).

Region or water body	PFOS	PFHxS	PFOA
Western Europe rivers			
North Sea			
Baltic Sea			
Rhine River			
Mediterranean Sea (2010-14 vs 2015-19)			
Central Atlantic (2000-09 vs 2010-19)			
North Atlantic (2000-09 vs 2010-19)			
Coastal North Atlantic (2000-09 vs 2010-19)			
North Pacific (2000-09 vs 2010-19)			
Arctic Ocean (2000-09 vs 2010-19)			
Great Lakes			
Canada/US rivers			
Australia			
New Zealand			

Increasing		No trend	
Decreasing		Insufficient data	

Indication of POP trends in non-core media, through long term monitoring by several international programs, has been presented in the chapter on Other Media (5.2.4). The derived trend information for listed POPs is summarized in Table 6.1e.

In brief, across the regions, α - β - and γ -HCH, DDT, PCBs, tetra- to hepta-PBDEs show decreasing trends, and/or decreasing/no change. No substances show increasing trends across all regions, however in Great Lakes, HCB, HCBd, PCN, Deca-BDE in general show increasing trends, as well as DDT in Antarctica, and HCB in the Baltic region. Dieldrin and Toxaphene were reported in both AMAP and GLWQA, showing decreasing levels or nonlinear trends. PFOS was reported across several regions, with contrasting trends; increasing in the Arctic and the Great Lakes, and decreasing in the Baltic region and the North Atlantic.

Some compounds were only reported from one region, thus no general assessment across the regions can be made, such as for those only reported from the Arctic: Chlordane (decreasing), heptachlor (no linear trend), Mirex (decreasing/non linear trend), PeCBz (decreasing/non linear trend); and for those only reported from the Great lakes: PCDD (decreasing), PCDF (decreasing), HCBd (increasing), PCN (increasing) and PFOA (no linear trend).

For several substances, there is no new trend data reported since the last GMP report. New substances are probably not reported as they are not yet regularly measured and reported (PCP, Dicofol, SCCP, HBB, PFHxS), likely due to challenging analytical chemistry and lack of comparable methods. Some older substances are no longer being measured and reported due to low levels and no change over time (Aldrin, Chlordane, Endosulfan, Mirex, Endrin).

Since climate variability and climate change affects the ecosystem structure and function and thus the POP food web accumulation and toxicity, it is important to understanding how climate change induced changes in the ecosystems affect the time trends of POPs in other media including biota, e.g. through changes in diet, in order to separate ecosystem changes effects of POP time trends from changes in POP time trends due to global regulation and bans of the POPs through the Stockholm Convention.

Table 6.1e Presentation of the direction of changes in concentrations over time included in the different programmes. Although Mediterranean and Antarctica do not report monitoring results systematically to the GMP, some info can be extracted from their reports concerning other themes/problems in their representative regions. The colours represent the direction of the changes in concentrations over time according to the respective programmes; **green**: decreasing concentrations, **yellow**: non-linear changes in concentrations that are neither increasing nor decreasing, **grey**: no change, **blue**: no changes in concentrations over time/trend (statistically decided), **red**: increasing changes in concentrations. Circles represent substances regulated earlier with low levels and no change in the last years and triangles represent newly listed substances of concern.

	AMAP (1975-2000)	AMAP (2000-2014)	OSPAR (1995-2014)	HELCOM (1978-2018)	Great Lakes (1970-2017)	Antarctica (NA)
Aldrin	○	○	○	○	○	○
α-HCH						
β-HCH						
Chlordane	1					
Chlordecone	○	○	○	○	○	○
DDT		1				
Dieldrin						
Endosulfan	○	○	○	○	○	○
Endrin	○	○	○	○	○	○
γ-HCH						
Heptachlor						
HBB	Δ	Δ	Δ	Δ	Δ	Δ
HBCD					3	
Hexa-, hepta-, PBDE			2	2	3	
HCB						
Mirex			○	○	○	○
PeCBz						
PFOS					3	
PCB						
PCDD						
PCDF						
Tetra-, penta-, PBDE					4	
Toxaphene		5				
HCBD						
PCP	Δ	Δ	Δ	Δ	Δ	Δ
PCNs						
DecaBDE						
SCCPs	Δ	Δ	Δ	Δ	Δ	Δ
Dicofol	Δ	Δ	Δ	Δ	Δ	Δ
PFOA, salts, related compounds						
PFHxS, salts, related compounds	Δ	Δ	Δ	Δ	Δ	Δ

1. One local source (whale processing site) causes an increasing trend in some species.
2. BDE-153 and BDE-154 only.
3. Increasing in some lakes, decreasing in other lakes.
4. Overall decreasing, increasing in whole fish in Lake Erie and in sediments in Lake Superior.
5. Parlar 26 decreases annually at 5.9%, parlar 50 increases annually at 0.8%.

6.1.3 Summary of evidence of long range transport, POPs modelling capabilities and the effects of climate change

The presence of POPs in regions of the world that are remote from sources, such as the Arctic, is compelling evidence of their long-range transport. People living in the Arctic and consuming a diet composed of local foods can even have higher exposure to certain POPs than people living in temperate regions.

A complete and quantitative picture of the location and strength of sources of POPs, their fate and long-range transport in the environment, and the levels that are present in remote regions can be assembled using mass balance chemical fate models as a unifying framework in an “integrated assessment approach”. Case studies that integrate emission inventories, chemical fate modelling, and monitoring data at the global scale have been carried out for some POPs, including PCBs, which have been modelled from source to presence in human milk, HCHs, DDT, dicofol and PFOS. Uncertainties in emission inventories and in the rates of degradation of POPs in air, water, soil and sediments often dominate uncertainties in the description of the global fate and long-range transport of POPs in these case studies.

Uncertainties in emission inventories for POPs can be reduced in a targeted way within an integrated assessment approach by applying models to estimate ranges of plausible emissions that are necessary to account for observed POP levels. Such “top-down” emission estimations can be applied at the global scale or at the regional scale to derive emission factors that can be used to constrain estimates of global emissions. More effective methods to identify and characterize emissions of POPs are needed to assess the impact of transport of goods and waste, in particular e-waste, all over the world, and “top-down” modeling could help address this data gap. The level of cooperation between scientists studying the sources of POPs, those engaged in monitoring and campaign-based measurements of POPs in the environment, and the contaminant fate and transport modeling community is growing and should be strongly encouraged.

The long-range transport of PFOS presents a new set of challenges relative to other POPs because of its distinct properties and potential for formation in the environment from precursor substances. Clear evidence for long-range transport in the atmosphere and deposition of PFOS and/or its precursors to remote waters (and ice cores) can be seen from detectable levels in

remote, uninhabited lakes in northern Canada and in Antarctica. Long-range transport in the oceans is also evident from detectable levels of PFOS thousands of kilometers from emission sources in surface and deep ocean waters that can be explained by ocean circulation modeling.

Improved modeling capabilities within the integrated assessment approach will enable the vast corpus of environmental monitoring data that has been established over the past decades to be leveraged to achieve a more quantitative and predictive understanding of the levels and trends of POPs and POP candidates in the environment. For example, total HCHs (Σ HCH) have been observed to decline in seals, beluga and polar bears due to rapid decline of the major isomer α -HCH (e.g. 12%/year in bears). However, β -HCH, the more bioaccumulative isomer, increased in the same species. The relative trends that are observed for α - and β -HCH in Arctic mammals are consistent with results of global-scale modeling which identify β -HCH to be more persistent and less rapidly transported to remote regions than α -HCH, and which forecast that β -HCH will eventually become the dominant HCH isomer in the environment (Wöhrnschimmel et al. 2013).

Levels of HCB and PCBs at certain monitoring stations in the Arctic have shown increasing time trends in recent years, which may be related to local revolatilization due to a warming environment. Global and regional-scale model scenarios that forecast the impact of climate change on levels of POPs generally find that changes of up to a factor of 2 in air and a factor of 4 in water are possible relative to base-case climate scenarios. However, in the same modeling scenarios, reductions in emission rates of POPs that are expected to occur simultaneously with climate change can reduce levels by orders of magnitude. Therefore, in the long term (i.e., decadal time scales) climate change is not expected to obscure or counteract reductions in levels of POPs that are possible due to phase-out of production, use and emissions.

Levels and trends of POPs at remote locations can also be affected by sources from materials used at the monitoring stations and emissions from local communities (i.e. short-range or regional transport). Many of the new POPs are commercial chemicals that are used in Arctic communities. Local emissions are expected to increase under climate change due to increased human activities in the North (resource development, shipping, mining, urbanization) and relatively poor waste management systems compared to the south. For example, stations in Antarctica have measured PBDEs in local air that are attributable to materials used at the station. However model scenarios that include enhanced emissions of chemicals in the Arctic in response to climate change demonstrate that when considering the Arctic as a whole, local emissions are a minor contributor to Arctic pollution by POPs compared to long-range transport in all scenarios (Wöhrnschimmel et al. 2013).

There is clear evidence that transport of POPs into remote regions occurs in the atmosphere, oceans and rivers, through biovectors, and in the technosphere by transport of materials. Models can be used to integrate information from emission inventories and monitoring programmes to quantitatively assess the relative weight of these routes in explaining the concentration for each pollutant, location and point in time. Enhancing modelling capabilities, especially by improving emission inventories and estimates of POP degradability, and by improving understanding of air-surface exchange, will help to reduce uncertainties about the extent of long-range transport of POPs and POP candidates. This is of great importance to interpret trends and ascertain the effectiveness of actions that have been undertaken, and to forecast impacts of current action or inaction.

6.1.4 Summary of gaps in data coverage

Table 6.1a to 6.1d reflect the availability of baseline monitoring and trend information for POPs in the WEOG region. While baseline data are becoming available for newer POPs, time trends are yet unavailable for most. Continued monitoring of these substances in a consistent manner is needed to provide temporal assessments in the future. A new Spanish air monitoring programme, one new station in Antarctica and one new station in Greenland have been established since 2008, 2007 and 2008, respectively, where additional baseline and trend data are now available. Temporal trend data are scarce in the southern hemisphere sub-regions of WEOG for this reporting period. Archived samples from Australia's national monitoring program can be analysed to provide additional time trend information for future reporting under the GMP. While the urban environment is a major source of new POPs that are associated with consumer products and household items, current urban air monitoring of POPs has been spatially limited. Better spatial coverage is being achieved through the use of passive air samplers, including studies targeting POP-like chemicals and health in major urban cities (e.g. GAPS Megacities pilot study) (Saini et al., 2020).

Trend information for PFASs (PFOS, PFHxS, and PFOA) is currently limited for air due to low concentrations of PFASs in the gas-phase. This gap in data could be addressed in the future using long term precipitation data, since PFASs partition strongly to water.

Temporal and spatial trend evaluations in the current report for POPs in human milk and/or blood are based on a small number of on-going international and national programmes, whereas results from individual research studies were considered unsuitable for the trend evaluation. Available information indicates that the levels of substances included in the Stockholm

Convention, where comparable data is available, are generally decreasing in most cases. It is noteworthy that although concentrations of some individual POPs are declining in some parts of the region (but not others), the number of POP-like chemicals to which humans and the environment are exposed are increasing.

Information on PFOS, PFHxS and PFOA in water from over 3000 locations and over 10,000 individual measurements within the WEOG region are included in this report, including 1300 analyses for samples collected from 2015-2019. These data allow trends to be derived for most sub-regions. However, data for PFOS, PFHxS and PFOA in Australian and New Zealand rivers and streams were available but too limited temporally and spatially to effectively evaluate temporal trends. Long term, systematic monitoring programs measuring PFASs in water in the WEOG region (i.e. in the same locations being sampled repeatedly over time by a dedicated program) do not exist.

Programs reporting POPs in other media have been updated and supplemented in the current report and provide an additional and broad perspective on data sources on POPs and their trends over time. The importance of ecosystem processing of POPs and challenges in interpretation of biotic data are also highlighted.

6.1.5 Summary of new developments in POPs monitoring

A review of the work carried out monitoring POPs in other media since 2009 in the WEOG region shows that the use of long time monitoring series has had an important role in shaping policy in the Arctic, the Great Lakes and the Baltic, North Atlantic and Mediterranean—and that the actions undertaken have worked. The environmental concentrations of the POP substances that were regulated decades ago and monitored since then have significantly decreased, and the majority of primary sources have stopped in these regions. There is a large corpus of high quality monitoring information on POPs in air, water, soil and organisms such as plankton, fish, birds and mammals in multiple sites regularly sampled over decades. The available data from these efforts should be studied in detail and in an integrated and interdisciplinary framework to develop better understanding of the rates of change in the environment and the time delays and modulations in concentration changes in different media. Improving the models of POPs pathways and fate is central to gauge the potential impact of action or inaction on a given process or substance.

While marine and freshwater ecosystems are relatively well documented, there is a notable gap of compiled information on terrestrial ecosystems, beyond mosses, lichens and a few large animals.

The practice of sample archiving is increasingly being adopted by long-term monitoring programmes for POPs. Sample banks allow retrospective analysis of environmental samples for determining concentrations of priority chemicals, candidate POPs and newly listed POPs. Retrospective analysis can be triggered by the availability of analytical standards, adequate and sensitive analytical methods being developed and by the demand for monitoring data for conducting risk assessments (e.g. needs of the POPRC or for national risk assessments) and/or risk management.

Lastly, a paradigm shift is underway in the field of POPs science and exposure assessment, where “real-world” exposures to chemical mixtures is being assessed. This has been made possible by new developments in high resolution screening of environmental samples (e.g. non-target analysis and suspect screening) and highly sensitive and cost effective toxicogenomic tools for assessing health impacts. In addition, transformation products of POPs and other previously unknown POP-like compounds are being investigated in environmental samples and in silico methods are being used to derive their physical chemical properties and to estimate toxicity. These advances are proceeding quickly and will have implications that will need to be considered for reporting under the 4th phase of the GMP.

6.2 Recommendations for the future

To complement the conclusions presented above, the following recommendations are offered to the COP to enhance future effectiveness evaluations.

Human Media: It is of great importance that the regional and national programmes evaluating time trends of both POPs and other environmental pollutants in blood and/or milk continues and expands. Future monitoring should address missing information for the newer POPs and improved spatial coverage for the WEOG regions. For instance, countries are urged to participate in the WHO/UNEP milk survey so that their data can be compared regionally and globally. The timing of the WHO/UNEP milk survey could be better synchronized with the cycle of the effectiveness evaluation of the Stockholm Convention to allow that the Global Monitoring Plan work can use the latest available data. Monitoring data for SCCPs in human milk and blood is urgently needed given the significant global production and use of this chemical mixture.

Data comparability of POPs in human tissues can be facilitated through the use of accessible and searchable databases, with sufficient background and metadata on samples and results (e.g., sampling strategies, information on cohorts and participants, and documenting analytical

methods and statistical parameters). Biobanks should also be exploited for retrospective analysis and deriving historical trends for POPs.

Air monitoring: As many of the legacy POPs are declining in air, and resources (funding and expertise) are becoming limited, programmes should consider strategically adapting their monitoring efforts for legacy compounds, as needed (e.g. reducing analytical frequencies and, in some cases, target chemical lists). These efficiencies will help to ensure that resources can be available for addressing the monitoring needs of newer POPs and future assessment of chemical mixtures. Passive air samplers should continue to be implemented as a cost-effective approach for improving spatial information for POPs in air.

One of the most relevant parameters that can be used to evaluate effectiveness of the Convention is the changes in emissions of POPs, regionally and globally. However, assembling emission inventories for POPs (i.e., the bottom-up approach; see Ch. 5.3) is challenging due to lack of information on use. Furthermore, many commercial products contain unknown quantities and types of POPs and this information is not adequately tracked. One way around this problem is through top-down approaches, where emissions are assessed from monitoring information and special studies (see Ch. 5.3).

With many of the newer POPs and candidate POPs being associated with commercial products and emissions from indoor air, urban areas may become more important as both sources and receptors of POPs; i.e. indoor to outdoor fate pathway. Monitoring programmes should consider expanding the spatial coverage of urban environments using passive sampling techniques. It may also be advantageous to include urban background sites (e.g. urban parks) and indoor surveys as part of air monitoring efforts as these sites are likely to respond relatively quickly to changes in the predominant emissions/usage of some ‘newer’ POPs that are used in commercial products (e.g. PBDEs, HBCDD and PFOS precursors).

There is growing impetus to make better connections between POPs monitoring and toxicity and long term effects of mixtures in air. Although the burden of individual POPs may be on the decline in some regions, the number of POP-like chemicals present in air is increasing. Chemicals associated with listed POPs, such as degradation products and precursors, may have POP-like qualities themselves, and these may exhibit temporal trends that differ from the listed POP. There is increasing need for better connections between air monitoring and long-term effects of human and environmental exposure to mixtures of chemicals.

Water: As noted in Chapter 5.3, seawater monitoring sites for PFAS have not been established so far, but if they are they should be located within a region where spatial variability is relatively small. Continued monitoring of rivers offers the best opportunity to assess temporal trends. However, there is always the need for consideration of waste water and tributary inputs and thus multiple sampling points preferably at sites that are well characterized in terms of flow and proximity to sources are needed. Lower detection limits would help with future assessments of

temporal trends in all locations but especially in open oceans, coastal seas and in the Great Lakes. Detection limits in the range of 1-5 pg/L for PFOS, PFHxS and PFOA have been demonstrated by several monitoring programs and should be universally adopted. Efforts should be made for more widespread measurement of precursors of PFOS, PFHxS and PFOA in waters with “total” methods (eg total oxidizable precursor (TOP) or total extractable organic fluorine) or targeting specific known precursor compounds.

Other Media:

Long term stable monitoring of biotic and abiotic media, sharing environmental sample collections, analytical methods and curated data are central to evaluate the effect of actions undertaken and to enhance understanding of the changing structure of ecosystems and POPs pathways leading to exposure. Despite the relative abundance of monitoring data the statistical power to identify significant trends over time is limited, and efforts should be made in monitoring design and advanced data analysis to enhance accuracy of estimated time trends. This can be facilitated through cooperation and capacity to maintain long term monitoring plans/programmes and environmental sample banks integrating multiple media, robust QA/QC and reporting in a coherent transparent service to enhance accuracy of estimates of changes over time.

Current results seem to indicate that POPs regulated in source regions decades ago have significantly decreased but are still present at low levels that have not changed since the previous GMP report in 2015 and are still of concern in some regions. Substances listed later in the Stockholm Convention; PBDE, HBCD, HCB, PFOS/PFOA, SCCP with ample exemptions for use, show increasing levels of concern. Therefore, it is important to report evidence of the success of past regulations in decreasing exposure to POPs to consolidate the need of further work with chronic low levels of old POPs (PCBs, PBDEs, α -HCH, HCB) and the growing threat of partially regulated POPs (PFOS/PFOA, BDCD, HCB, SCCP) and new substances with POPs features sometimes used as alternative.

The complex ecotoxicology of mixtures and the rapidly increasing quantities released to the environment of new substances with POPs features are a challenge and an opportunity to transform monitoring strategies. Integrated assessments should not bury primary data under indicators but develop tools to look at primary QA/QC controlled empirical data provided in public repositories by a diversity of fields, from meteorology to molecular biology. Machine learning, statistical and process modelling tools can enhance process understanding and operational capacity. Best efforts should be made to integrate POPs monitoring work and resulting data of abiotic media and macroscopic organisms with climate data and molecular biology and toxicology in curated and accessible repositories.

Local, indigenous knowledge about ecosystem process and structure has been of great value for scientists in developing understanding of POPs pathways and exposure routes, and science should provide local agents with meaningful and operational tools to deal with contaminants and POPs. There is a need to establish and maintain conditions that facilitate common understandings

and cooperation between local agents and scientists to develop effective strategies in public health and environmental policy on POPs.

Long range transport and effects of climate change and climate variability:

Using models to formalize the link between emissions and concentrations of POPs and POP candidates in air, water, soil, wildlife and humans has a wide range of benefits for supporting the development of the Global Monitoring Plan and for effectiveness evaluation of the Stockholm Convention. These include:

- i.) Spurring the development and refinement of emission inventories for POPs and POP candidates;*
- ii.) Aiding in the design of monitoring programs and quality assurance of monitoring data, and*
- iii.) Understanding trends in POP levels in a changing climate.*

In order to fully realize these benefits the following research and monitoring priorities are recommended:

1) *Support the development of global scale modeled 'integrated assessments' of emissions, fate and transport processes and environmental levels and trends for all POPs and POP candidates.* Currently global scale case studies are only available for a select few POPs and POP candidates. In order to realize the benefits of these assessments it is recommended to compose a complete case study for each POP and POP candidate that can be updated regularly and refined as new information is produced relevant to emissions, fate and transport properties and environmental levels and trends.

2) *Apply the principles of integrated assessment at local, regional and global levels to improve emission estimates.* High uncertainties and missing information in emission inventories is often the dominant source of uncertainty in integrated assessments of POPs. As reviewed in the 2nd Regional Monitoring Report of the WEOG Region, the integrated approach can be applied at local and regional scales and near suspected source areas to infer emission rates of POPs that can then be scaled using emission factors to compare with national estimates and to drive global scale models.

3) *Support the development and improvement of POP modeling capabilities and laboratory and field research to understand POP properties.* Important research needs to improve POPs models include improving modeling of gas/particle partitioning in the atmosphere and of air/surface exchange in general. Uncertainties in assessment of individual POPs are often driven by uncertainties in physico-chemical properties and degradation rates in air, water and soil which could be reduced using targeted laboratory and field studies. Models can be evaluated and iteratively improved by identifying locations where observations would most effectively constrain model uncertainties and targeting field monitoring at those locations. Capabilities to model source-to-human body burden relationships for POPs would benefit from better resolving food sourcing patterns especially in poorly understood/high exposure areas.

Climate-related effects can impact the emissions and cycling of POPs in the environment, and have the potential to obscure or modulate trends of POPs that are observed in air and other

media. The European research project ArcRisk concluded that the influence of climate change on levels and trends of POPs in the global environment on the time-scale of decades is likely much smaller than the influence of policies that would reduce emissions. However, shorter-term effects of climate change and climate variability that vary by region and subregion and by chemical should be taken into consideration when interpreting measurements of POPs in environmental media. Climate change influences foodweb structure and POP trends in biota, which will subsequently affect exposure to wildlife and humans. Climate variability and climate change effects on the environment and ecosystems structure and function more likely affect POP food web accumulation and toxicity to a larger degree than how climate change effects on long range transport of POPs. It is therefore important understand how climate change induced changes in the ecosystems affect the time trends of POPs in other media including biota, e.g. through changes in diet, in order to separate ecosystem changes effects of POP time trends from changes in POP time trends due to global regulation and bans of the POPs through the Stockholm Convention. However, there are still many uncertainties surrounding this complex topic which require additional research.

Sample Archiving / Sample Banks: Sample archiving is an important practice for all media and provides an opportunity to develop comparable data for spatial and temporal trends analysis. Sample banks are also useful for conducting retrospective analysis for screening studies and for assessing new POPs as these priorities are identified and/or as analytical methods become available. Long term monitoring programmes should invest resources to ensure the integrity and sustainability of sample banks.

Data archiving and accessibility: Databases and effective data flow between providers, institutional users and the public play an important role in identifying, documenting and making accessible reliable and comparable data sets on POPs. Scientists should be encouraged to share information on databases in a timely manner and should be given credit for datasets that have not yet been published through the peer review process. These data are needed for assessing environmental levels and trends of POPs and for developing and validating transport models. Effort should be invested to make databases easily accessible (e.g. open access and data downloadable) and integrate databases where appropriate and possible. Further development of simple visualization tools such as those implemented under the GMP database would be useful.

Challenges with new POPs: The growing number of listed POPs and candidate substances presents a resource pressure for existing monitoring programmes. Furthermore, some new POPs require modified sampling methods and/or new and sometimes complex analytical methods. Some new POPs do not behave in the conventional manner and may involve precursor chemicals (e.g. PFOS) or may generate stable degradation products which may also exhibit POP-like properties.

Research studies are needed as existing models are often not adequate for assessing new compounds (e.g. polar/ionizable POPs) that behave differently in the environment. For instance, studies of partitioning to particles and particle-associated transport and fate are needed for high

molecular weight POPs and polar/ionizable chemicals that exist in air primarily on the particle-phase. It was previously thought that chemicals that tend to attach to particles would deposit faster and are not subjected to atmospheric long-range transport. However, more recent studies have shown that such chemicals can be transported very far from sources because they tend to attach to very fine particles that do not deposit quickly. The chemicals may be protected from photodegradation and transformation during particle transport. It is thus important to better understand this transport process.

Emissions of POPs have shifted from mainly industrial / combustion / agricultural sources to a wide range of indoor sources associated with commercial products, where human exposure can be a significant concern. Emissions from the waste sector (e.g. waste water treatment plants and landfills) to water, air, and soil (e.g. through sludge application) is also relevant for the newer POPs, as a result of their commercial uses and waste disposal fate pathways. Monitoring strategies include assessment of these sources and attempts to link emissions of these chemicals to human and environmental burdens. Monitoring programmes need to be flexible and adaptable to deal with the challenges presented by new POPs.

Efficiencies for Monitoring Programmes: In order to deal with added analysis pressures of new POPs, monitoring programmes may need to adjust their protocols and resources to better align with new priorities. Possible strategies include reduced sampling frequency for legacy POPs for which long-term data series exhibiting declining trends already exist. Reducing or focusing analyte lists is also an option for complex technical mixtures (e.g. PCBs, Toxaphene and PBDEs). In some cases analytical methods can be improved or combined so that numerous target analytes can be determined in the same run.

Partnerships among labs and programmes at the regional and even international scale could be explored as an option for dealing with analytical challenges for some classes of new and candidate POPs. It is not feasible for every lab to be an expert for every class of POPs.

Improved integration between monitoring programmes and modelers may help to identify gaps in information and optimize future study designs and ongoing monitoring efforts.

Lastly, passive air samplers should continue to be implemented for air as a cost-effective approach for improving spatial resolution and for carrying out screening and source identification studies on new and candidate POPs.

Chemical Mixtures: With advances in non-target analysis, toxicogenomics, and greater capabilities and awareness of POP transformation products and unknown POP-like chemicals in core media, it is becoming possible to measure real-world exposures to chemical mixtures and predicted impacts to human health and the environment. Expert advice should be sought on how a chemicals mixtures approach can complement the current chemical-by-chemical reporting in core media under the GMP, in order to better serve the goals of the Convention.

Comparability of data: Each of the existing programmes that contributed information for the WEOG review has their own procedures for maintaining intra-programme comparability of information. However, in general, different programmes do not use the same analytical laboratories. Although there are procedures to evaluate variance between laboratories (such as blind inter-laboratory comparisons), the use of different analytical laboratories remains a potential major source of variance between programmes. Due to the large number of existing programmes used as information sources for the WEOG report, it was concluded that it would be unrealistic to expect comparability between programmes now and in the future. Therefore it is recommended that future efforts are focused to promote internal comparability within programmes over time. While this conclusion generally means that there will be very limited direct comparability between regions, significant exceptions are evident, such as the WHO coordinated human milk programme and the GAPS Network which use a single laboratory. The extensive use of such measures as utilization of common analytical laboratories and data centers has demonstrated the possibility of achieving adequate comparability between well established programmes in the WEOG region. An example is the collaborative practices of AMAP, EMEP, OSPAR, and HELCOM.

Mediterranean rim: The review found a significant improvement in the information on the Mediterranean rim since 2009. It is recommended that steps be explored for enhancing long term cooperation in POPs monitoring in the Mediterranean region to improve the information base for future effectiveness evaluations of the Convention.

Longevity of monitoring arrangements: Finally, this review has relied on information provided by a relatively small number of existing national and international monitoring programmes. The ability to compare POPs levels over time within these programmes makes their long term viability of utmost importance for future trend analysis to evaluate the effectiveness of the Convention. It is also important that such programmes do not alter their procedures in ways that compromise the data comparability that is necessary for the examination of temporal trends. The existing programmes should be commended for their outstanding work.

7 REFERENCES

Ambient Air (5.2.1)

- Baek, S., Choi, S., Chang, Y. (2011) Three-year atmospheric monitoring of organochlorine pesticides and polychlorinated biphenyls in polar regions and the south Pacific. *Environ. Sci. Technol.* 45: 4475–4482.
- Bengtson Nash, S. M., Wild, S. J., Hawker, D. W., Cropp, R. A., Hung, H., Wania, F., Xiao, H., Bohlin-Nizzetto, P., Bignert, A., Broomhall, S. (2017) Persistent organic pollutants in the east Antarctic atmosphere: Inter-annual observations from 2010-2015 using high-flow-through passive sampling. *Environmental Science and Technology*, 51(23): 13929-13937. DOI: 10.1021/acs.est.7b04224
- Bidleman, T. F., Tysklind, M. (2018) Breakthrough during air sampling with polyurethane foam: What do PUF 2/PUF 1 ratios mean? *Chemosphere*, 192: 267-271.
- Colette, A., Aas, W., Banin, L., Braban, C.F., Ferm, M., González Ortiz, A., Ilyin, I., Mar, K., Pandolfi, M., Putaud, J.-P., Shatalov, V., Solberg, S., Spindler, G., Tarasova, O., Vana, M., Adani, M., Almodovar, P., Berton, E., Bessagnet, B., Bohlin-Nizzetto, P., Boruvkova, J., Breivik, K., Briganti, G., Cappelletti, A., Cuvelier, K., Derwent, R., D'Isidoro, M., Fagerli, H., Funk, C., Garcia Vivanco, M., González Ortiz, A., Haeuber, R., Hueglin, C., Jenkins, S., Kerr, J., de Leeuw, F., Lynch, J., Manders, A., Mircea, M., Pay, M.T., Pritula, D., Putaud, J.-P., Querol, X., Raffort, V., Reiss, I., Roustan, Y., Sauvage, S., Scavo, K., Simpson, D., Smith, R.I., Tang, Y.S., Theobald, M., Tørseth, K., Tsyro, S., van Pul, A., Vidic, S., Wallasch, M., Wind, P. (2016). Air pollution trends in the EMEP region between 1990 and 2012. Joint Report of the EMEP Task Force on Measurements and Modelling (TFMM), Chemical Co-ordinating Centre (CCC), Meteorological Synthesizing Centre-East (MSC-E), Meteorological Synthesizing Centre-West (MSC-W). EMEP/CCC-Report 1/2016.
- Choi, S.-D.; Baek, S.-Y.; Chang, Y.-S.; Wania, F.; Ikonomou, M. G.; Yoon, Y.-J.; Park, B.-K.; Hong, S. Passive air sampling of polychlorinated biphenyls and organochlorine pesticides at the Korean Arctic and Antarctic research stations: Implications for long-range transport and local pollution. *Environ. Sci. Technol.* 2008, 42, 7125–7131.
- Commission for Environmental Cooperation (CEC) (2014) Atmospheric PCDD, PCDF and coplanar PCB data from North American ambient air monitoring networks Montreal, Canada: Commission for Environmental Cooperation. 256 pp.
- Commission Regulation (EU) 2017/227 of 9 February 2017 amending Annex XVII to Regulation (EC) No 1907/2006 of the European Parliament and of the Council concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) as regards bis(pentabromophenyl)ether. Official Journal of the European Union: pp.L35/6-L35/9. Available at: <https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32017 R0227&from=EN> (Accessed April 29, 2020).

- de Wit, C. A., Herzke, D., Vorkamp, K. (2010) Brominated flame retardants in the Arctic environment - trends and new candidates. *Sci. Tot. Environ.*, 408, 2885-2918.
- Environment Canada (EC) (2013) Canadian Environmental Protection Act, 1999. Federal Environmental Quality Guidelines. Polybrominated diphenyl ethers (PBDEs). <http://ec.gc.ca/ese-ees/default.asp?lang=En&n=05DF7A37-1#a3> (Accessed April 20, 2020).
- Fredricsson, M., Brorström-Lundén, E., Danielsson, H., Hansson, K., Karlsson, G. P., Nerentorp, M., Potter, A., Sjöberg, K., Kreuger, J., Nanos, T., Areskoug, H., Krejci, R., Wylde, H. A., Andersson, C., Andersson, S., Carlund, T., Josefsson, W., Leung, W (2018) Nationell luftövervakning - Sakrapport med data från övervakning inom Programområde Luft t.o.m. 2017. IVL Svenska Miljöinstitutet, Stockholm, Sweden, 150pp. ISBN 978-91-88787-99-6
- Gawor, A., Shunthirasingham, C., Hayward, S.J., Lei, Y.D., Gouin, T., Mmereki, B.T., Masamba, W., Ruepert, C., Castillo, L.E., Shoeib, M., Lee, S.C., Harner, T., Wania, F. (2014) Neutral and polyfluoroalkyl substances in the global atmosphere. *Environ. Sci. Processes Impacts*, 16, 404-413.
- Graf, C., Katsoyiannis, A., Jones, K.C. and Sweetman, A.J. (2016) The TOMP's Ambient Air Monitoring Network - continuous data on the UK air quality for 20 years. *Environmental Pollution*, 217: 42-51.
- Guo, J., Salamova, A., Venier, M., Dryfhout-Clark, H., Alexandrou, N., Backus, S., Bradley, L., Hung, H., Hites, R. A. (2018) Atmospheric flows of semi-volatile organic pollutants to the Great Lakes estimated by the United States' Integrated Atmospheric Deposition and Canada's Great Lakes Basin Monitoring and Surveillance Networks. *Journal of Great Lakes Research*, 44(4): 670-681.
- Hao, Y., Li, Y., Han, X., Wang, T., Yang, R., Wang, P., Xiao, K., Li, W., Lu, H., Fu, J., Wang, Y., Shi, J., Zhang, Q., Jiang, G. (2019) Air monitoring of polychlorinated biphenyls, diphenyl ethers and organochlorine pesticides in West Antarctica during 2011-2017: Concentrations, temporal trends and potential sources. *Environmental Pollution* 249: 381-389.
- Hung, H., Katsoyiannis, A., Brorström-Lund, E., Olafsdottir, K., Aas, W., Breivik, K., Bohlin-Nizzetto, P., Sigurdsson, A., Hakola, H., Bossi, R., Skov, H., Sverko, E., Barresi, E., Fellin, P., Wilson, S. (2016). Temporal trends of Persistent Organic Pollutants (POPs) in arctic air: 20 years of monitoring under the Arctic Monitoring and Assessment Programme (AMAP). *Environ. Pollut.*, 217: 52-61.
- Kalina, J., White, K. B., Scheringer, M., Přibyllová, P., Kukučka, P., Audy, O., Klánová, J. (2019) Comparability of long-term temporal trends of POPs from co-located active and passive air monitoring networks in Europe. *Environ. Sci.: Processes Impacts*, 21, 1132-1142.

- Kallenborn, R., Breivik, K., Eckhardt, S., Lunder, C.R., Mano, S., Schlabach, M., Stohl, A. (2013). Long-term monitoring of persistent organic pollutants (POPs) at the Norwegian Troll station in Dronning Maud Land, Antarctica. *Atmos. Chem. Phys.* 13: 6983-6992.
- Kirchner, Manfred; Jakobi, Gert; Körner, Wolfgang; Levy, Walkiria; Moche, Wolfgang; Niedermoser, Bernhard, Schaub, M., Ries, L., Weiss, P., Anritter, F., Fischer, N., Henkelmann, B., Schramm, K.-W. (2016). Ambient Air Levels of Organochlorine Pesticides at Three High Alpine Monitoring Stations. Trends and Dependencies on Geographical Origin. *Aerosol Air Qual. Res.* 16 (3): 738–751. DOI: 10.4209/aaqr.2015.04.0213.
- Koblizkova, M., Genualdi, S., Lee, S. C., Harner, T. (2012) Application of Sorbent Impregnated Polyurethane Foam (SIP) Disk Passive Air Samplers for Investigating Organochlorine Pesticides and Polybrominated Diphenyl Ethers at the Global Scale. *Environ. Sci. Technol.*, 46, 391-396.
- Lee, S. C., Sverko, E., Harner, T., Pozo, K., Barresi, E., Schachtschneider, J., Zaruk, D., DeJong, M., Narayan, J. (2016) Retrospective analysis of “new” flame retardants in the global atmosphere under the GAPS Network. *Environmental Pollution*. 217: 62-69.
- Liu, L. Y., Salamova, A., Venier, M., Hites, R. A. (2016) Trends in the levels of halogenated flame retardants in the Great Lakes atmosphere over the period 2005-2013, *Environ International*, 92-93: 442-449.
- Muñoz-Arnanz, J., Roscales, J.L., Vicente, A., Ros, M., Barrios, L., Morales, L., Abad, E., Jiménez, B. (2018). Assessment of POPs in air from Spain using passive sampling from 2008 to 2015. Part II: spatial and temporal observations of PCDD/Fs and dl-PCBs. *Sci. Total Environ.* 634: 1669-1676.
- Norwegian Institute for Air Research (NILU) (2019) Monitoring of environmental contaminants in air and precipitation. Annual report 2018. NILU report 11/2019. Norwegian Environment Agency, Norway. 125 pp.
- Olukunle, O. I., Venier, M., Hites, R. A. and Salamova, A. (2018) Atmospheric concentrations of hexabromocyclododecane (HBCDD) diastereomers in the Great Lakes region. *Chemosphere* 200: 464-470.
- Rauert, C., Schuster, J. K., Eng, A., and Harner, T. (2018a). Global Atmospheric Concentrations of Brominated and Chlorinated Flame Retardants and Organophosphate Esters. *Environ Sci Technol*, 52(5): 2777-2789. doi:10.1021/acs.est.7b06239
- Rauert, C., Shoieb, M., Schuster, J. K., Eng, A., and Harner, T. (2018b). Atmospheric concentrations and trends of poly- and perfluoroalkyl substances (PFAS) and volatile methyl siloxanes (VMS) over 7 years of sampling in the Global Atmospheric Passive Sampling (GAPS) network. *Environmental Pollution*, 238: 94-102. doi:10.1016/j.envpol.2018.03.017

- Roscales, J.L., Muñoz-Arnanz, J., Ros, M., Vicente, A., Barrios, L., Jiménez, B. (2018). Assessment of POPs in air from Spain using passive sampling from 2008 to 2015. Part I: Spatial and temporal observations of PBDEs, *Sci. Total Environ.* 634: 1657-1668.
- Salamova, A., Venier, M., Hites, R. A. (2015) Revised temporal trends of persistent organic pollutant concentrations in air around the Great Lakes. *Environmental Science and Technology Letter*, 2: 20-25.
- Schenker, U., Scheringer, M., MacLeod, M., Martin, J. W., Cousins, I. T. and Hungerbühler (2008). Contribution of volatile precursor substances to the flux of perfluorooctanoate to the Arctic. *Environ. Sci. Technol.*, 42: 3710-3716.
- Shunthirasingham, C., Oyiliagu, C.E., Cao, X., Gouin, T., Wania, F., Lee, S.C., Pozo, K., Harner, T., Muir, D.C.G. (2010) Spatial and temporal patterns of pesticides in the global atmosphere. *J. Environ. Mon.* 12: 1650-1657.
- Schuster, J. K., Gioia, R., Breivik, K., Steinnes, E., Scheringer, M., Jones, K. C. (2010) Trends in European Background Air Reflect Reductions in Primary Emissions of PCBs and PBDEs *Environ. Sci. Technol.* 44 (17): 6760-6766.
- United States Environmental Protection Agency (USEPA) (2009). "Polybrominated Diphenyl Ethers (PBDEs) Action Plan Summary." www.epa.gov/assessing-and-managing-chemicals-under-tsca/polybrominated-diphenyl-ethers-pbdes-action-plan. (Accessed April 29, 2020). USEPA, 2013. United States Environmental Protection Agency. "Polybrominated Diphenyl Ethers (PBDEs) Significant New Use Rules (SNUR)." www.epa.gov/assessing-and-managing-chemicals-under-tsca/polybrominated-diphenylethers-pbdes-significant-new-use. (Accessed April 29, 2020).
- Wang, X., Kennedy, K., Powell, J., Keywood, M., Gillett, R., Thai, P., Bridgen, P., Broomhall, S., Paxman, C., Wania, F., Mueller, J. F. (2015) Spatial distribution of selected persistent organic pollutants (POPs) in Australia's atmosphere. *Environmental science: Processes and Impacts*, 17:525-532.
- Xiao, H., Hung, H., Harner, T., Lei, Y.D., Johnston, G. W., Wania, F. (2007) A flow-through sampler for semi-volatile organic compounds in air. *Environ. Sci. Technol.* 41: 250-256.

Human Tissue (5.2.2)

1. R. Hulek, J. Boruvkova, J. Gregor, J. Kalina, Z. Bednarova, K. Sebkova, O. Melkes, M. Salko, R. Novak, J. Jarkovsky, L. Dusek, J. Klanova, *Global Monitoring Plan of the Stockholm Convention on Persistent Organic Pollutants: visualisation and on-line analysis of global levels of chemicals in air, water, breast milk and blood* [online]. Masaryk University, 2014. Available from: <http://www.pops-gmp.org/visualization-2014>.)
2. Health-related environmental monitoring (HÄMI). <https://ki.se/en/imm/health-related-environmental-monitoring-hami>
3. A. Barbarossa, R.M., T. Gazzotti, D. Zama, A. Astolfi, B. Veyrand, A. Pession and G. Pagliuca, *Perfluoroalkyl substances in human milk: A first survey in Italy*. Environment International, 2013. **51**: p. 27-30.
4. A. Giovannini, G.R., P. Carideo, R. Ceci, G. Diletti, C. Ippoliti, G. Migliorati, P. Piscitelli, A. Ripani, R. Salini and G. Scortichini, *Dioxins levels in breast milk of women living in Caserta and Naples : Assessment of environmental risk factors*. Chemosphere, 2014. **94**: p. 76-84.
5. A. t Mannetje, J.C., P. Bridgen, C. Brooks, S. Harrad, A. H. Smith, N. Pearce and J. Douwes, *Current concentrations, temporal trends and determinants of persistent organic pollutants in breast milk of New Zealand women*. Science of the Total Environment, 2013. **458**: p. 399-407.
6. Abdallah, S.H.a.M.A., *Concentrations of Polybrominated Diphenyl Ethers, Hexabromocyclododecanes and Tetrabromobisphenol-A in Breast Milk from United Kingdom Women Do Not Decrease over Twelve Months of Lactation*. Environmental Science & Technology, 2015. **49**(23): p. 13899-13903.
7. C. C. Carignan, M.A.E.A., N. Wu, W. Heiger-Bernays, M. D. McClean, S. Harrad and T. F. Webster, *Predictors of Tetrabromobisphenol-A (TBBP-A) and Hexabromocyclododecanes (HBCD) in Milk from Boston Mothers*. Environmental Science & Technology, 2012. **46**(21): p. 12146-12153.
8. C. D. Lynch, L.W.J., P. J. Kostyniak, B. M. McGuinness and G. M. B. Louis, *The effect of prenatal and postnatal exposure to polychlorinated biphenyls and child neurodevelopment at age twenty four months*. Reproductive Toxicology, 2012. **34**(3): p. 451-456.
9. J. Fång, E. Nyberg, Bignert, A., and Bergman. Å., *Spatial and temporal trends of the Stockholm Convention POPs in mothers' milk — a global review*. Environmental Science and Pollution Research, 2015. **22**: p. 8989-9041.
10. C. Inthavong, F.H., F. Bordet, V. Rigourd, T. Guerin and S. Dragacci, *Simultaneous liquid chromatography-tandem mass spectrometry analysis of brominated flame retardants (tetrabromobisphenol A and hexabromocyclododecane diastereoisomers) in French breast milk*. Chemosphere, 2017. **186**: p. 762-769.
11. D. Costopoulou, I.V.a.L.L., *Infant dietary exposure to dioxins and dioxin-like compounds in Greece*. Food and Chemical Toxicology, 2013. **59**: p. 316-324.
12. D. F. K. Rawn, A.R.S., V. A. Casey, F. Breton, W. F. Sun, T. E. Arbuckle and W. D. Fraser, *Dioxins/furans and PCBs in Canadian human milk: 2008-2011*. Science of the Total Environment, 2017. **595**: p. 269-278.
13. D. G. Hayward, J.C.A., S. Andrews, R. D. Fairchild, J. Gentry, R. Jenkins, M. McLain, U. Nasini and S. Shojaee, *Application of a High-Resolution Quadrupole/Orbital Trapping Mass Spectrometer Coupled to a Gas Chromatograph for the Determination of Persistent Organic Pollutants in Cow's and Human Milk*. Journal of Agricultural and Food Chemistry, 2018. **66**(44): p. 11823-11829.

14. E. Cechova, M.S., M. Seifertova, O. Mikes, K. Kroupova, J. Kuta, J. Forns, M. Eggesbo, I. Quaak, M. de Cock, M. van de Bor, H. Patayova, L. P. Murinova and A. Kocan, *Developmental neurotoxins in human milk: Comparison of levels and intakes in three European countries*. Science of the Total Environment, 2017. **579**: p. 637-645.
15. E. Cechova, S.V., P. Kukucka, A. Kocan, T. Trnovec, L. P. Murinova, M. de Cock, M. van de Bor, J. Askevold, M. Eggesbo and M. Scheringer, *Legacy and alternative halogenated flame retardants in human milk in Europe: Implications for children's health*. Environment International, 2017. **108**: p. 137-145.
16. E. De Felip, F.B., C. Bove, L. Cori, A. D'Argenzio, G. D'Orsi, M. Fusco, R. Miniero, R. Ortolani, R. Palombino, A. Parlato, M. G. Pelliccia, F. Peluso, G. Piscopo, R. Pizzuti, M. G. Porpora, D. Protano, O. Senofonte, S. R. Spena, A. Simonetti and A. di Domenico, *Priority persistent contaminants in people dwelling in critical areas of Campania Region, Italy (SEBIOREC biomonitoring study)*. Science of the Total Environment, 2014. **487**: p. 420-435.
17. F. Tao, M.A.A., D. C. Ashworth, P. Douglas, M. B. Toledano and S. Harrad, *Emerging and legacy flame retardants in UK human milk and food suggest slow response to restrictions on use of PBDEs and HBCDD*. Environment International, 2017. **105**: p. 95-104.
18. H. S. Canbay, S.O., M. Yilmazer and R. S. Unsal, *Pesticide Residues Analysis in Human Milk Samples in Isparta Region (Turkey)*. Asian Journal of Chemistry, 2013. **25**(7): p. 3931-3936.
19. Harrad, M.A.A.a.S., *Polybrominated diphenyl ethers in UK human milk: Implications for infant exposure and relationship to external exposure*. Environment International, 2014. **63**: p. 130-136.
20. I. Cok, B.M., M. A. Mazmanci, C. Turgut, B. Henkelmann and K. W. Schramm, *Analysis of human milk to assess exposure to PAHs, PCBs and organochlorine pesticides in the vicinity Mediterranean city Mersin, Turkey*. Environment International, 2012. **40**: p. 63-69.
21. I. Pratt, W.A., D. Crowley, S. Daly, R. Evans, A. Fernandes, M. Fitzgerald, M. Geary, D. Keane, J. J. Morrison, A. Reilly and C. Tlustos, *Brominated and fluorinated organic pollutants in the breast milk of first-time Irish mothers: is there a relationship to levels in food?* Food Additives and Contaminants Part a-Chemistry Analysis Control Exposure & Risk Assessment, 2013. **30**(10): p. 1788-1798.
22. J. C. Hartle, R.S.C., P. Sakamoto, D. B. Barr and S. L. Carmichael, *Chemical Contaminants in Raw and Pasteurized Human Milk*. Journal of Human Lactation, 2018. **34**(2): p. 340-349.
23. J. Du, Z.G., M. C. L. Gay, C. T. Lai, R. D. Trengove, P. E. Hartmann and D. T. Geddes, *Longitudinal study of pesticide residue levels in human milk from Western Australia during 12 months of lactation: Exposure assessment for infants*. Scientific Reports, 2016. **6**.
24. J. F. Focant, N.F., M. L. Bidondo, G. Eppe, G. Scholl, A. Saoudi, A. Oleko and S. Vandentorren, *Levels of polychlorinated dibenzo-p-dioxins, polychlorinated dibenzofurans and polychlorinated biphenyls in human milk from different regions of France*. Science of the Total Environment, 2013. **452**: p. 155-162.
25. J. Forns, N.I., R. A. White, S. Mandal, A. Sabaredzovic, M. Lamoree, C. Thomsen, L. S. Haug, H. Stigum and M. Eggesbo, *Perfluoroalkyl substances measured in breast milk and child neuropsychological development in a Norwegian birth cohort study*. Environment International, 2015. **83**: p. 176-182.
26. J. Forns, S.M., N. Iszatt, A. Polder, C. Thomsen, J. L. Lyche, H. Stigum, R. Vermeulen and M. Eggesbo, *Novel application of statistical methods for analysis of multiple toxicants identifies DDT as a risk factor for early child behavioral problems*. Environmental

- Research, 2016. **151**: p. 91-100.
27. J. Haedrich, C.S.a.M.S.D., *Ah-receptor based bioassay screening for PCDD/Fs and Dioxin-like PCBs in human milk samples from a WHO/UNEP global survey on halogenated POPs*. Deutsche Lebensmittel-Rundschau, 2018. **114**(9): p. 389-397.
 28. J. P. Antignac, B.V., H. Kadar, P. Marchand, A. Oleko, B. Le Bizec and S. Vandentorren, *Occurrence of perfluorinated alkylated substances in breast milk of French women and relation with socio-demographical and clinical parameters: Results of the ELFE pilot study*. Chemosphere, 2013. **91**(6): p. 802-808.
 29. J. P. Antignac, K.M.M., H. E. Virtanen, C. Y. Boquien, P. Marchand, A. VeniSseau, I. Guiffard, E. Bichon, C. Wohlfahrt-Veje, A. Legrand, C. Boscher, N. E. Skakkebaek, J. Toppari and B. Le Bizec, *Country-specific chemical signatures of persistent organic pollutants (POPs) in breast milk of French, Danish and Finnish women*. Environmental Pollution, 2016. **218**: p. 728-738.
 30. J. Wasser, T.B., L. Lerner-Geva, I. Grotto and L. Rubin, *Biological monitoring of Persistent Organic Pollutants in human milk in Israel*. Chemosphere, 2015. **137**: p. 185-191.
 31. K. Croes, A.C., G. Koppen, E. Govarts, L. Bruckers, E. Van de Mierop, V. Nelen, A. Covaci, A. C. Dirtu, C. Thomsen, L. S. Haug, G. Becher, M. Mampaey, G. Schoeters, N. Van Larebeke and W. Baeyens, *Persistent organic pollutants (POPs) in human milk: A biomonitoring study in rural areas of Flanders (Belgium)*. Chemosphere, 2012. **89**(8): p. 988-994.
 32. K. Croes, A.C., G. Koppen, S. De Galan, T. Vandermarken, E. Govarts, L. Bruckers, V. Nelen, G. Schoeters, N. Van Larebeke, M. S. Denison, M. Mampaey and W. Baeyens, *Determination of PCDD/Fs, PBDD/Fs and dioxin-like PCBs in human milk from mothers residing in the rural areas in Flanders, using the CALUX bioassay and GC-HRMS*. Talanta, 2013. **113**: p. 99-105.
 33. K. Eroglu, Y.K.D., Z. Golbasi, Z. Simsek, F. Koruk and E. Atmaca, *Persistent Organic Chlorinated Compound Residues in the Breast Milk of Female Seasonal Agricultural Workers*. Journal of Occupational and Environmental Medicine, 2018. **60**(9): p. 860-866.
 34. K. Hoffman, M.A., B. D. Goldman, A. Sjodin and J. L. Daniels, *Lactational Exposure to Polybrominated Diphenyl Ethers and Its Relation to Social and Emotional Development among Toddlers*. Environmental Health Perspectives, 2012. **120**(10): p. 1438-1442.
 35. K. Jakobsson, J.F., M. Athanasiadou, A. Rignell-Hydbom and A. Bergman, *Polybrominated diphenyl ethers in maternal serum, umbilical cord serum, colostrum and mature breast milk. Insights from a pilot study and the literature*. Environment International, 2012. **47**: p. 121-130.
 36. L. Bramwell, A.F., M. Rose, S. Harrad and T. Pless-Mulloli, *PBDEs and PBBs in human serum and breast milk from cohabiting UK couples*. Chemosphere, 2014. **116**: p. 67-74.
 37. L. Bramwell, S.H., M. A. Abdallah, C. Rauert, M. Rose, A. Fernandes and T. Pless-Mulloli, *Predictors of human PBDE body burdens for a UK cohort*. Chemosphere, 2017. **189**: p. 186-197.
 38. L. Dimitriadou, G.M., A. Covaci, E. Iossifidou, J. Tzafettas, V. Zournatzi-Koiou and O. I. Kalantzi, *Levels and profiles of brominated and chlorinated contaminants in human breast milk from Thessaloniki, Greece*. Science of the Total Environment, 2016. **539**: p. 350-358.
 39. L. M. L. Toms, P.G., E. Eljarrat, D. Barcelo, F. A. Harden, P. Hobson, A. Sjodin, E. Ryan and J. F. Mueller, *Brominated flame retardants in the Australian population: 1993-2009*. Chemosphere, 2012. **89**(4): p. 398-403.

40. M. de Cock, M.R.d.B., M. Lamoree, J. Legler and M. van de Bor, *First Year Growth in Relation to Prenatal Exposure to Endocrine Disruptors - A Dutch Prospective Cohort Study*. International Journal of Environmental Research and Public Health, 2014. **11**(7): p. 7001-7021.
41. M. de Cock, M.R.D.B., M. Lamoree, J. Legler and M. Van de Bor, *Prenatal exposure to endocrine disrupting chemicals and birth weight-A prospective cohort study*. Journal of Environmental Science and Health Part a-Toxic/Hazardous Substances & Environmental Engineering, 2016. **51**(2): p. 178-185.
42. M. de Cock, M.R.d.B., E. Govarts, N. Iszatt, L. Palkovicova, M. H. Lamoree, G. Schoeters, M. Eggesbo, T. Trnovec, J. Legler and M. van de Dor, *Thyroid-stimulating hormone levels in newborns and early life exposure to endocrine-disrupting chemicals: analysis of three European mother-p-child cohorts*. Pediatric Research, 2017. **82**(3): p. 429-437.
43. M. Hoopmann, U.V.A., E. Gierden, R. Huppmann and R. Suchenwirth, *Time trends and individual characteristics associated with polybrominated diphenyl ethers in breast milk samples 2006-2009 in Lower Saxony, Germany*. International Journal of Hygiene and Environmental Health, 2012. **215**(3): p. 352-359.
44. M. M. Guzman, C.C., M. D. Perez-Carceles, S. J. Rejon, A. Cascone, T. Martellini, C. Guerranti and A. Cincinelli, *Perfluorinated carboxylic acids in human breast milk from Spain and estimation of infant's daily intake*. Science of the Total Environment, 2016. **544**: p. 595-600.
45. M. Roberto, F.F., C. Valter, F. Silva, F. A. Rita, I. Nicola, I. A. Maria and D. Elena, *Chemical indicators of exposure to polychlorodibenzo-p-dioxins, polychlorodibenzofurans and polychlorobiphenyls in breast milk samples from mothers residing in Trento, Italy, and neighboring country municipalities*. Journal of Environmental Science and Health Part B-Pesticides Food Contaminants and Agricultural Wastes, 2018. **53**(8): p. 510-518.
46. M. Schuhmacher, H.K., P. Ruokojarvi, M. Nadal and J. L. Domingo, *Levels of PCDD/Fs, PCBs and PBDEs in breast milk of women living in the vicinity of a hazardous waste incinerator: Assessment of the temporal trend*. Chemosphere, 2013. **93**(8): p. 1533-1540.
47. M. Schuhmacher, M.M., M. Nadal and J. L. Domingo, *Concentrations of dioxins and furans in breast milk of women living near a hazardous waste incinerator in Catalonia, Spain*. Environment International, 2019. **125**: p. 334-341.
48. N. Iszatt, S.J., V. Lenters, C. Dahl, H. Stigum, R. Knight, S. Mandal, S. Peddada, A. Gonzalez, T. Midtvedt and M. Eggesbo, *Environmental toxicants in breast milk of Norwegian mothers and gut bacteria composition and metabolites in their infants at 1 month*. Microbiome, 2019. **7**.
49. O. P. Luzardo, N.R.-S., M. Almeida-Gonzalez, L. A. Henriquez-Hernandez, M. Zumbado and L. D. Boada, *Multi-residue method for the determination of 57 Persistent Organic Pollutants in human milk and colostrum using a QuEChERS-based extraction procedure*. Analytical and Bioanalytical Chemistry, 2013. **405**(29): p. 9523-9536.
50. O. Vall, M.G.-C., C. Puig, E. Rodriguez-Carrasco, A. G. Baltazar, L. Canchucaja, X. Joya and O. Garcia-Algar, *Prenatal and Postnatal Exposure to DDT by Breast Milk Analysis in Canary Islands*. Plos One, 2014. **9**(1).
51. R. Cariou, B.V., A. Yamada, A. Berrebi, D. Zalko, S. Durand, C. Pollono, P. Marchand, J. C. Leblanc, J. P. Antignac and B. Le Bizec, *Perfluoroalkyl acid (PFAA) levels and profiles in breast milk, maternal and cord serum of French women and their newborns*. Environment International, 2015. **84**: p. 71-81.

52. R. Criswell, V.L., S. Mandal, H. Stigum, N. Iszatt and M. Eggesbo, *Persistent Environmental Toxicants in Breast Milk and Rapid Infant Growth*. Annals of Nutrition and Metabolism, 2017. **70**(3): p. 210-216.
53. R. Dallaire, G.M., F. Rouget, P. Kadhel, H. Bataille, L. Guldner, S. Seurin, V. Chajes, C. Monfort, O. Boucher, J. P. Thome, S. W. Jacobson, L. Multigner and S. Cordier, *Cognitive, visual, and motor development of 7-month-old Guadeloupean infants exposed to chlordecone*. Environmental Research, 2012. **118**: p. 79-85.
54. J.J.R. Rawn, *Polychlorinated dioxins, furans (PCDD/Fs), and polychlorinated biphenyls (PCBs) and their trends in Canadian human milk from 1992 to 2005*. Chemosphere, 2014. **102**: p. 76-86.
55. J.J.R. Rawn "The brominated flame retardants, PBDEs and HBCD, in Canadian human milk samples collected from 1992 to 2005; concentrations and trends". Environment International, 2014. **70**: p. 43473.
56. S. A. Marchitti, S.E.F., P. Mendola, J. F. Kenneke and E. P. Hines, *Polybrominated Diphenyl Ethers in Human Milk and Serum from the US EPA MAMA Study: Modeled Predictions of Infant Exposure and Considerations for Risk Assessment*. Environmental Health Perspectives, 2017. **125**(4): p. 706-713.
57. S. N. Zhou, A.B., S. Siddique, L. Takser, N. Abdelouahab and J. P. Zhu, *Measurements of Selected Brominated Flame Retardants in Nursing Women: Implications for Human Exposure*. Environmental Science & Technology, 2014. **48**(15): p. 8873-8880.
58. S. Siddique, Q.M.X., N. Abdelouahab, L. Takser, S. P. Phillips, Y. L. Feng, B. Wang and J. P. Zhu, *Levels of dechlorane plus and polybrominated diphenylethers in human milk in two Canadian cities*. Environment International, 2012. **39**(1): p. 50-55.
59. U. Raab, M.A., U. Preiss, W. Volkel, U. Schwegler and H. Fromme, *Organochlorine compounds, nitro musks and perfluorinated substances in breast milk - Results from Bavarian Monitoring of Breast Milk 2007/8*. Chemosphere, 2013. **93**(3): p. 461-467.
60. W. H. Guo, A.H., S. C. Smith, R. Gephart, M. Petreas and J. S. Park, *PBDE levels in breast milk are decreasing in California*. Chemosphere, 2016. **150**: p. 505-513.
61. V. Lenters, N.I., J. Forn, E. Cechova, A. Kocan, J. Legler, P. Leonards, H. Stigum and M. Eggesbo, *Early-life exposure to persistent organic pollutants (OCPs, PBDEs, PCBs, PFASs) and attention-deficit/hyperactivity disorder: A multi-pollutant analysis of a Norwegian birth cohort*. Environment International, 2019. **125**: p. 33-42.
62. AMAP Assessment 2015: Human Health in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway. vii + 165 pp
63. Y. Zhou, B. Yuan, E. Nyberg, G. Yin, A. Bignert, A. Glynn, J. Ö. Odland, Y. Qiu, Y. Sun, Y. Wu, Q. Xiao, D. Yin, Z. Zhu, J. Zhao, Å. Bergman, *Chlorinated Paraffins in Human Milk from Urban Sites in China, Sweden, and Norway*. Environmental Science and Technology, 2020. **54** (7): p. 4356-4366.
64. P. O. Darnerud, M. Aune, A. Glynn, A. Borgen, *Chlorinated Paraffins in Swedish Breast Milk, Swedish Chemicals Agency PM 18/12 (22 pp)*, 2012.
<https://www.kemi.se/global/pm/2012/pm-18-12.pdf>
65. B. Pérez-Gómez, R. Pastor-Barriuso, M. Cervantes-Amat, E. Esteban, M. Ruiz-Moraga, N. Aragonés, M. Pollán, C. Navarro, E. Calvo, J. Román, G. López-Abente, A. Castano. *BIOAMBIENT.ES study protocol: rationale and design of a cross-sectional human biomonitoring survey in Spain*. Environmental Science and Pollution Research. 2013. **20**, p. 1193–1202.

66. D. A. Haines, G. Saravanabhavan, W. Kate and C. Khoury. *An overview of human biomonitoring of environmental chemicals in the Canadian Health Measures Survey: 2007–2019*. **International Journal of Hygiene and Environmental Health**. 2016. 220 (2). p. 13-28.
67. Centers for Disease Control and Prevention (CDC). National Center for Health Statistics (NCHS). National Health and Nutrition Examination Survey Data. Hyattsville, MD: U.S. Department of Health and Human Services, Centers for Disease Control and Prevention, 2019. <https://wwwn.cdc.gov/nchs/nhanes/Search/DataPage.aspx?Component=Laboratory>.
68. Australia. Pers. Comm. Leisa-Maree Toms (Queensland University of Technology) and Jochen Mueller (University of Queensland). April 2020
69. German Environmental Specimen Bank (German ESB).
<https://www.umweltprobenbank.de/de>. Data owned by German Environment Agency (UBA).

Water (5.2.3)

- Ahrens, L., 2011. Polyfluoroalkyl compounds in the aquatic environment: A review of their occurrence and fate. *Journal of Environmental Monitoring* 13, 20-31.
- Ahrens, L., Barber, J.L., Xie, Z., Ebinghaus, R., 2009a. Longitudinal and latitudinal distribution of perfluoroalkyl compounds in the surface water of the Atlantic Ocean. *Environmental Science and Technology* 43, 3122-3127.
- Ahrens, L., Felizeter, S., Sturm, R., Xie, Z., Ebinghaus, R., 2009b. Polyfluorinated compounds in waste water treatment plant effluents and surface waters along the River Elbe, Germany. *Marine Pollution Bulletin* 58, 1326-1333.
- Ahrens, L., Gerwinski, W., Theobald, N., Ebinghaus, R., 2010a. Sources of polyfluoroalkyl compounds in the North Sea, Baltic Sea and Norwegian Sea: Evidence from their spatial distribution in surface water. *Marine Pollution Bulletin* 60, 255-260.
- Ahrens, L., Norström, K., Viktor, T., Cousins, A.P., Josefsson, S., 2015. Stockholm Arlanda Airport as a source of per- and polyfluoroalkyl substances to water, sediment and fish. *Chemosphere* 129, 33-38.
- Ahrens, L., Plassmann, M., Xie, Z., Ebinghaus, R., 2009c. Determination of polyfluoroalkyl compounds in water and suspended particulate matter in the river Elbe and North Sea, Germany. *Frontiers of Environmental Science and Engineering in China* 3, 152-170.
- Ahrens, L., Xie, Z., Ebinghaus, R., 2010b. Distribution of perfluoroalkyl compounds in seawater from Northern Europe, Atlantic Ocean, and Southern Ocean. *Chemosphere* 78, 1011-1016.
- Allinson, M., Yamashita, N., Taniyasu, S., Yamazaki, E., Allinson, G., 2019. Occurrence of perfluoroalkyl substances in selected Victorian rivers and estuaries: An historical snapshot. *Heliyon* 5, e02472.
- Antweiler, R.C., 2015. Evaluation of Statistical Treatments of Left-Censored Environmental Data Using Coincident Uncensored Data Sets. II. Group Comparisons. *Environmental Science & Technology* 49, 13439-13446.

Armitage, J.M., Macleod, M., Cousins, I.T., 2009a. Modeling the global fate and transport of perfluorooctanoic acid (PFOA) and perfluorooctanoate (PFO) Emitted from direct sources using a multispecies mass balance model. *Environ. Sci. Technol.* 43, 1134-1140.

Armitage, J.M., Schenker, U., Scheringer, M., Martin, J.W., Macleod, M., Cousins, I.T., 2009b. Modeling the global fate and transport of perfluorooctane sulfonate (PFOS) and precursor compounds in relation to temporal trends in wildlife exposure. *Environmental Science and Technology* 43, 9274-9280.

Australia Defence, 2018. PFAS Detailed Site Investigation: Bandiana Military Area. <https://www.defence.gov.au/Environment/PFAS/docs/Bandiana/Reports/201809BandianaDetailedSiteInvestigationReportTablesFigures.pdf>, in: Golder Associates Pty LTD 1777738-081-R-RevB (Ed.).

Baddiley, B., Munns, T., Braun, C., Vardy, S., 2020. QLD Ambient PFAS Monitoring Program 2019-2020 Department of Environment and Science (DES), Queensland Government, Brisbane, Australia.

Benskin, J.P., De Silva, A.O., Martin, J.W., 2010. Isomer profiling of perfluorinated substances as a tool for source tracking: A review of early findings and future applications. *Reviews of Environmental Contamination and Toxicology* 208, 111-160.

Benskin, J.P., Holt, A., Martin, J.W., 2009. Isomer-specific biotransformation rates of a perfluorooctane sulfonate (PFOS)-precursor by cytochrome P450 isozymes and human liver microsomes. *Environmental Science and Technology* 43, 8566-8572.

Benskin, J.P., Muir, D.C.G., Scott, B.F., Spencer, C., De Silva, A.O., Kylin, H., Martin, J.W., Morris, A., Lohmann, R., Tomy, G., Rosenberg, B., Taniyasu, S., Yamashita, N., 2012. Perfluorinated compounds in the Arctic and Atlantic Oceans. *Environ. Sci. Technol.* 46, 5815-5823.

Brumovský, M., Karásková, P., Borghini, M., Nizzetto, L., 2016. Per- and polyfluoroalkyl substances in the Western Mediterranean Sea waters. *Chemosphere* 159, 308-316.

Cai, M., Zhao, Z., Yin, Z., Ahrens, L., Huang, P., Cai, M., Yang, H., He, J., Sturm, R., Ebinghaus, R., Xie, Z., 2012. Occurrence of perfluoroalkyl compounds in surface waters from the North Pacific to the Arctic Ocean. *Environ. Sci. Technol.* 46, 661-668.

D'Agostino, L.A., Mabury, S.A., 2017. Certain Perfluoroalkyl and Polyfluoroalkyl Substances Associated with Aqueous Film Forming Foam Are Widespread in Canadian Surface Waters. *Environmental Science and Technology* 51, 13603-13613.

D'Eon, J.C., Mabury, S.A., 2007. Production of perfluorinated carboxylic acids (PFCAs) from the biotransformation of polyfluoroalkyl phosphate surfactants (PAPS): Exploring routes of human contamination. *Environmental Science and Technology* 41, 4799-4805.

Dauchy, X., Boiteux, V., Bach, C., Rosin, C., Munoz, J.-F., 2017. Per- and polyfluoroalkyl substances in firefighting foam concentrates and water samples collected near sites impacted by the use of these foams. *Chemosphere* 183, 53-61.

De Silva, A.O., Spencer, C., Scott, B.F., Backus, S., Muir, D.C.G., 2011. Detection of a cyclic perfluorinated acid, perfluoroethylcyclohexane sulfonate, in the Great Lakes of North America. *Environ. Sci. Technol.* 45, 8060-8066.

- De Solla, S.R., De Silva, A.O., Letcher, R.J., 2012. Highly elevated levels of perfluorooctane sulfonate and other perfluorinated acids found in biota and surface water downstream of an international airport, Hamilton, Ontario, Canada. *Environment International* 39, 19-26.
- Domingo, J.L., Nadal, M., 2019. Human exposure to per- and polyfluoroalkyl substances (PFAS) through drinking water: A review of the recent scientific literature. *Environmental Research* 177.
- Earnshaw, M.R., Paul, A.G., Loos, R., Tavazzi, S., Paracchini, B., Scheringer, M., Hungerbühler, K., Jones, K.C., Sweetman, A.J., 2014. Comparing measured and modelled PFOS concentrations in a UK freshwater catchment and estimating emission rates. *Environment International* 70, 25-31.
- Ericson, I., Gómez, M., Nadal, M., van Bavel, B., Lindström, G., Domingo, J.L., 2007. Perfluorinated chemicals in blood of residents in Catalonia (Spain) in relation to age and gender: A pilot study. *Environment International* 33, 616-623.
- Filipovic, M., Berger, U., McLachlan, M.S., 2013. Mass balance of perfluoroalkyl acids in the Baltic sea. *Environmental Science and Technology* 47, 4088-4095.
- Furdui, V.I., Crozier, P.W., Reiner, E.J., Mabury, S.A., 2008. Trace level determination of perfluorinated compounds in water by direct injection. *Chemosphere* 73, S24-S30.
- Gallen, C., Baduel, C., Lai, F.Y., Thompson, K., Thompson, J., Warne, M., Mueller, J.F., 2014. Spatio-temporal assessment of perfluorinated compounds in the Brisbane River system, Australia: Impact of a major flood event. *Marine Pollution Bulletin* 85, 597-605.
- Gebbink, W.A., Bignert, A., Berger, U., 2016. Perfluoroalkyl Acids (PFAAs) and Selected Precursors in the Baltic Sea Environment: Do Precursors Play a Role in Food Web Accumulation of PFAAs? *Environmental Science & Technology* 50, 6354-6362.
- Gewurtz, S.B., Bradley, L.E., Backus, S., Dove, A., McGoldrick, D., Hung, H., Dryfhout-Clark, H., 2019. Perfluoroalkyl Acids in Great Lakes Precipitation and Surface Water (2006-2018) Indicate Response to Phase-outs, Regulatory Action, and Variability in Fate and Transport Processes. *Environmental Science and Technology* 53, 8543-8552.
- Hansen, K.J., Johnson, H.O., Eldridge, J.S., Butenhoff, J.L., Dick, L.A., 2002. Quantitative characterization of trace levels of PFOS and PFOA in the Tennessee river. *Environmental Science and Technology* 36, 1681-1685.
- Houtz, E.F., Sedlak, D.L., 2012. Oxidative conversion as a means of detecting precursors to perfluoroalkyl acids in urban runoff. *Environmental Science and Technology* 46, 9342-9349.
- Huset, C.A., Chiaia, A.C., Barofsky, D.F., Jonkers, N., Kohler, H.P.E., Ort, C., Giger, W., Field, J.A., 2008. Occurrence and mass flows of fluorochemicals in the Glatt Valley Watershed, Switzerland. *Environmental Science and Technology* 42, 6369-6377.
- ICES, 2020. Contaminants and biological effects of contaminants in seawater. Organofluorines. International Council for the Exploration of the Sea (ICES), Copenhagen Denmark.
- ISO, 2009. Water quality — Determination of perfluorooctanesulfonate (PFOS) and perfluorooctanoate (PFOA) — Method for unfiltered samples using solid phase extraction and liquid chromatography/mass spectrometry, ISO 25101:2009 International Organization for Standardization, Geneva, Switzerland, 24 pp.

- Jiang, W., Zhang, Y., Yang, L., Chu, X., Zhu, L., 2015. Perfluoroalkyl acids (PFAAs) with isomer analysis in the commercial PFOS and PFOA products in China. *Chemosphere* 127, 180-187.
- Johansson, J.H., Salter, M.E., Acosta Navarro, J.C., Leck, C., Nilsson, E.D., Cousins, I.T., 2019. Global transport of perfluoroalkyl acids via sea spray aerosol. *Environmental Science: Processes & Impacts* 21, 635-649.
- Kallenborn, R., Berger, U., Järnberg, U., 2004. Perfluorinated Alkylated Substances (PFAS) in the Nordic Environment. Nordic Council of Ministers, 107 pp.
- Kirchgeorg, T., Weinberg, I., Dreyer, A., Ebinghaus, R., 2010. Perfluorinated compounds in marine surface waters: Data from the Baltic Sea and methodological challenges for future studies. *Environmental Chemistry* 7, 429-434.
- Li, L., Zheng, H., Wang, T., Cai, M., Wang, P., 2018. Perfluoroalkyl acids in surface seawater from the North Pacific to the Arctic Ocean: Contamination, distribution and transportation. *Environmental Pollution* 238, 168-176.
- Llorca, M., Farré, M., Picó, Y., Müller, J., Knepper, T.P., Barceló, D., 2012. Analysis of perfluoroalkyl substances in waters from Germany and Spain. *Science of the Total Environment* 431, 139-150.
- Loos, R., Gawlik, B.M., Locoro, G., Rimaviciute, E., Contini, S., Bidoglio, G., 2008a. EU Wide Monitoring Survey of Polar Persistent Pollutants in European River Waters. European Commission, Joint Research Centre, Institute for Environment and Sustainability, 51 pp.
- Loos, R., Locoro, G., Huber, T., Wollgast, J., Christoph, E.H., de Jager, A., Manfred Gawlik, B., Hanke, G., Umlauf, G., Zaldívar, J.M., 2008b. Analysis of perfluorooctanoate (PFOA) and other perfluorinated compounds (PFCs) in the River Po watershed in N-Italy. *Chemosphere* 71, 306-313.
- Lorenzo, M., Campo, J., Farré, M., Pérez, F., Picó, Y., Barceló, D., 2016. Perfluoroalkyl substances in the Ebro and Guadalquivir river basins (Spain). *Science of the Total Environment* 540, 191-199.
- Martin, J.W., Asher, B.J., Beesoon, S., Benskin, J.P., Ross, M.S., 2010. PFOS or PreFOS? Are perfluorooctane sulfonate precursors (PreFOS) important determinants of human and environmental perfluorooctane sulfonate (PFOS) exposure? *Journal of Environmental Monitoring* 12, 1979-2004.
- Martin, J.W., Ellis, D.A., Mabury, S.A., Hurley, M.D., Wallington, T.J., 2006. Atmospheric chemistry of perfluoroalkanesulfonamides: Kinetic and product studies of the OH radical and Cl atom initiated oxidation of N-ethyl perfluorobutanesulfonamide. *Environmental Science and Technology* 40, 864-872.
- Mathieu, C., McCall, M., 2017. Survey of Per- and Poly-fluoroalkyl Substances (PFASs) in Rivers and Lakes, 2016. Washington State Department of Ecology, Olympia WA, 48 pp.
- McGuire, M.E., Schaefer, C., Richards, T., Backe, W.J., Field, J.A., Houtz, E., Sedlak, D.L., Guelfo, J.L., Wunsch, A., Higgins, C.P., 2014. Evidence of remediation-induced alteration of subsurface poly- and perfluoroalkyl substance distribution at a former firefighter training area. *Environmental Science and Technology* 48, 6644-6652.

- McLachlan, M.S., Holmström, K.E., Reth, M., Berger, U., 2007. Riverine Discharge of Perfluorinated Carboxylates from the European Continent. *Environmental Science & Technology* 41, 7260-7265.
- Miyake, Y., Yamashita, N., Rostkowski, P., So, M.K., Taniyasu, S., Lam, P.K.S., Kannan, K., 2007. Determination of trace levels of total fluorine in water using combustion ion chromatography for fluorine: A mass balance approach to determine individual perfluorinated chemicals in water. *Journal of Chromatography A* 1143, 98-104.
- Möller, A., Ahrens, L., Surm, R., Westerveld, J., Van Der Wielen, F., Ebinghaus, R., De Voogt, P., 2010. Distribution and sources of polyfluoroalkyl substances (PFAS) in the River Rhine watershed. *Environmental Pollution* 158, 3243-3250.
- Moody, C.A., Kwan, W.C., Martin, J.W., Muir, D.C., Mabury, S.A., 2001. Determination of perfluorinated surfactants in surface water samples by two independent analytical techniques: liquid chromatography/tandem mass spectrometry and ^{19}F NMR. *Anal Chem* 73, 2200-2206.
- Moody, C.A., Martin, J.W., Kwan, W.C., Muir, D.C.G., Mabury, S.A., 2002. Monitoring perfluorinated surfactants in biota and surface water samples following an accidental release of fire-fighting foam into Etobicoke Creek. *Environmental Science and Technology* 36, 545-551.
- Muir, D., Lohmann, R., 2013. Water as a new matrix for global assessment of hydrophilic POPs. *TrAC - Trends in Analytical Chemistry* 46, 162-172.
- Myers, A.L., Crozier, P.W., Helm, P.A., Brimacombe, C., Furdui, V.I., Reiner, E.J., Burniston, D., Marvin, C.H., 2012. Fate, distribution, and contrasting temporal trends of perfluoroalkyl substances (PFASs) in Lake Ontario, Canada. *Environment International* 44, 92-99.
- NZDF, 2019a. RNZAF Base Ohakea PFAS Investigation: Comprehensive site investigation report. <https://www.mfe.govt.nz/land/pfas-and-poly-fluoroalkyl-substances/latest-updates> Accessed June 2020.
- NZDF, 2019b. RNZAF Base Woodbourne PFAS Investigation: Comprehensive site investigation report (Woodbourne CSIR) - December 2019. <https://www.mfe.govt.nz/land/pfas-and-poly-fluoroalkyl-substances/latest-updates> Accessed June 2020.
- Paul, A.G., Scheringer, M., Hungerbühler, K., Loos, R., Jones, K.C., Sweetman, A.J., 2012. Estimating the aquatic emissions and fate of perfluorooctane sulfonate (PFOS) into the river Rhine. *Journal of Environmental Monitoring* 14, 524-530.
- Pistocchi, A., Loos, R., 2009. A Map of European Emissions and Concentrations of PFOS and PFOA. *Environmental Science & Technology* 43, 9237-9244.
- Post, G.B., Gleason, J.A., Cooper, K.R., 2017. Key scientific issues in developing drinking water guidelines for perfluoroalkyl acids: Contaminants of emerging concern. *PLOS Biology* 15, e2002855.
- Prevedouros, K., Cousins, I.T., Buck, R.C., Korzeniowski, S.H., 2006. Sources, fate and transport of perfluorocarboxylates. *Environ Sci Technol* 40, 32-44.
- Procopio, N.A., Karl, R., Goodrow, S.M., Maggio, J., Louis, J.B., Atherholt, T.B., 2017. Occurrence and source identification of perfluoroalkyl acids (PFAAs) in the Metedeconk River Watershed, New Jersey. *Environmental Science and Pollution Research* 24, 27125-27135.

- Quinete, N., Orara, F., Werres, F., Moreira, I., Wilken, R.D., 2009. Determination of perfluorooctane sulfonate and perfluorooctanoic acid in the Rhine river, Germany. *Fresenius Environmental Bulletin* 18, 1356-1362.
- Reagen, W.K., Lindstrom, K.R., Jacoby, C.B., Purcell, R.G., Kestner, T.A., Payfer, R.M., Miller, J.W., 2007. Environmental characterization of 3M electrochemical fluorination derived perfluorooctanoate and perfluorooctanesulfonate, Society of Environmental Toxicology and Chemistry 28th North American meeting, 11-15 Nov, 2007, Milwaukee, WI, USA.
- Rumsby, P.C., McLaughlin, C.L., Hall, T., 2009. Perfluorooctane sulphonate and perfluorooctanoic acid in drinking and environmental waters. *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences* 367, 4119-4136.
- Sardiña, P., Leahy, P., Metzeling, L., Stevenson, G., Hinwood, A., 2019. Emerging and legacy contaminants across land-use gradients and the risk to aquatic ecosystems. *Science of The Total Environment* 695, 133842.
- Schmidt, N., Fauvelle, V., Castro-Jiménez, J., Lajaunie-Salla, K., Pinazo, C., Yohia, C., Sempéré, R., 2019. Occurrence of perfluoroalkyl substances in the Bay of Marseille (NW Mediterranean Sea) and the Rhône River. *Marine Pollution Bulletin* 149, 110491.
- Schultz, M.M., Higgins, C.P., Huset, C.A., Luthy, R.G., Barofsky, D.F., Field, J.A., 2006. Fluorochemical Mass Flows in a Municipal Wastewater Treatment Facility. *Environ. Sci. Technol.* 40, 7350-7357.
- Schulz, K., Silva, M.R., Klaper, R., 2020. Distribution and effects of branched versus linear isomers of PFOA, PFOS, and PFHxS: A review of recent literature. *Science of The Total Environment* 733, 139186.
- Scott, B.F., De Silva, A.O., Spencer, C., Lopez, E., Backus, S.M., Muir, D.C.G., 2010. Perfluoroalkyl acids in Lake Superior water: Trends and sources. *Journal of Great Lakes Research* 36, 277-284.
- Scott, B.F., Spencer, C., Lopez, E., Muir, D.C.G., 2009. Perfluorinated alkyl acid concentrations in Canadian rivers and creeks. *Water Quality Research Journal of Canada* 44, 263-277.
- Sinclair, E., Mayack, D.T., Roblee, K., Yamashita, N., Kannan, K., 2006. Occurrence of perfluoroalkyl surfactants in water, fish, and birds from New York State. *Archives of Environmental Contamination and Toxicology* 50, 398-410.
- Skutlarek, D., Exner, M., Färber, H., 2006. Perfluorinated surfactants in surface and drinking waters. *Environmental Science and Pollution Research* 13, 299-307.
- Stemmler, I., Lammel, G., 2010. Pathways of PFOA to the arctic: Variabilities and contributions of oceanic currents and atmospheric transport and chemistry sources. *Atmospheric Chemistry and Physics Discussions* 10, 11577-11614.
- Stockholm Convention, 2020. Chemicals Proposed for Listing under the Convention <http://www.pops.int/TheConvention/ThePOPs/ChemicalsProposedforListing/tabid/2510/Default.aspx>. United Nations Environment Programme, Geneva, Switzerland, p. 12.
- Taniyasu, S., Kannan, K., Man, K.S., Gulkowska, A., Sinclair, E., Okazawa, T., Yamashita, N., 2005. Analysis of fluorotelomer alcohols, fluorotelomer acids, and short- and long-chain perfluorinated acids in water and biota. *Journal of Chromatography A* 1093, 89-97.

- Theobald, N., Caliebe, C., Gerwinski, W., Hühnerfuss, H., Lepom, P., 2011. Occurrence of perfluorinated organic acids in the North and Baltic seas. Part 1: distribution in sea water. *Environmental Science and Pollution Research* 18, 1057-1069.
- Thompson, J., Eaglesham, G., Mueller, J., 2011a. Concentrations of PFOS, PFOA and other perfluorinated alkyl acids in Australian drinking water. *Chemosphere* 83, 1320-1325.
- Thompson, J., Roach, A., Eaglesham, G., Bartkow, M.E., Edge, K., Mueller, J.F., 2011b. Perfluorinated alkyl acids in water, sediment and wildlife from Sydney Harbour and surroundings. *Marine Pollution Bulletin* 62, 2869-2875.
- UNEP, 2015a. Global Monitoring Plan for Persistent Organic Pollutants under The Stockholm Convention Article 16 on Effectiveness Evaluation. 2nd Global Monitoring Report. UNEP/POPS/COP.8/INF/38. United Nations Environment Program, Stockholm Convention, Geneva, Switzerland, 129 pp + Annexes.
- UNEP, 2015b. Global Monitoring Plan for Persistent Organic Pollutants under The Stockholm Convention Article 16 on Effectiveness Evaluation. 2nd Regional Monitoring Report, Western Europe and Others Group (WEOG) Region. United Nations Environment Program, Stockholm Convention, Geneva, Switzerland, 216 pp + Annexes.
- UNEP, 2015c. Guidance on the global monitoring plan for persistent organic pollutants. UNEP/POPS/COP.6/INF/31. United Nations Environment Programme, Stockholm Convention, Geneva, Switzerland, 172.
- US EPA, 2016. Fact Sheet. Drinking Water Health Advisories for PFOA and PFOS, EPA 800-F-16-003 (<https://www.epa.gov/ground-water-and-drinking-water/drinking-water-health-advisories-pfoa-and-pfos>). US Environmental Protection Agency, Washington DC.
- Vrana, B., Smedes, F., Klanova, J., Muir, D., 2019. Guidance on the Global Monitoring Plan for Persistent Organic Pollutants. 4. Sampling and Sampling Preparation Methodology. 4.1 Water. UNEP/POPS/COP.7/INF/39. United Nations Environment Programme, Stockholm Convention, Geneva, Switzerland, 5 pp.
- Wang, Z., Boucher, J.M., Scheringer, M., Cousins, I.T., Hungerbühler, K., 2017. Toward a Comprehensive Global Emission Inventory of C4–C10 Perfluoroalkanesulfonic Acids (PFASs) and Related Precursors: Focus on the Life Cycle of C8-Based Products and Ongoing Industrial Transition. *Environmental Science & Technology* 51, 4482-4493.
- Wang, Z., Cousins, I.T., Scheringer, M., Buck, R.C., Hungerbühler, K., 2014. Global emission inventories for C4-C14 perfluoroalkyl carboxylic acid (PFCA) homologues from 1951 to 2030, part II: The remaining pieces of the puzzle. *Environment International* 69, 166–176.
- Wania, F., 2007. A global mass balance analysis of the source of perfluorocarboxylic acids in the Arctic Ocean. *Environmental Science and Technology* 41, 4529-4535.
- Weiss, J., de Boer, J., Berger, U., Muir, D., Ruan, T., Torre, A., Smedes, F., Vrana, B., Clavien, F., Fiedler, H., 2015. PFAS analysis in water for the Global Monitoring Plan of the Stockholm Convention. Set-up and guidelines for monitoring. Chemicals Branch United Nations Environment Programme (UNEP) Division of Technology, Industry and Economics, Geneva, 35 pp.

- Xie, Z., Zhao, Z., Möller, A., Wolschke, H., Ahrens, L., Sturm, R., Ebinghaus, R., 2013. Neutral poly- and perfluoroalkyl substances in air and seawater of the North Sea. *Environ. Sci. Pollut. Res.* 20, 7988-8000.
- Yamashita, N., Kannan, K., Taniyasu, S., Horii, Y., Okazawa, T., Petrick, G., Gamo, T., 2004. Analysis of perfluorinated acids at parts-per-quadrillion levels in seawater using liquid chromatography-tandem mass spectrometry. *Environmental Science and Technology* 38, 5522-5528.
- Yamashita, N., Kannan, K., Taniyasu, S., Horii, Y., Petrick, G., Gamo, T., 2005. A global survey of perfluorinated acids in oceans. *Mar. Pollut. Bull.* 51, 658-668.
- Yamashita, N., Taniyasu, S., Petrick, G., Wei, S., Gamo, T., Lam, P.K.S., Kannan, K., 2008. Perfluorinated acids as novel chemical tracers of global circulation of ocean waters. *Chemosphere* 70, 1247-1255.
- Yamashita, N., Yeung, L.W.Y., Taniyasu, S., Kwok, K.Y., Petrick, G., Gamo, T., Guruge, K.S., Lam, P.K.S., Loganathan, B.G., 2012. Global Distribution of PFOS and Related Chemicals, in: Loganathan, B.G., Lam, P.K.S. (Eds.), *Global Contamination Trends of Persistent Organic Chemicals*. Taylor & Francis Group, pp. 593-628.
- Yamazaki, E., Taniyasu, S., Ruan, Y., Wang, Q., Petrick, G., Tanhua, T., Gamo, T., Wang, X., Lam, P.K.S., Yamashita, N., 2019. Vertical distribution of perfluoroalkyl substances in water columns around the Japan sea and the Mediterranean Sea. *Chemosphere* 231, 487-494.
- Zhang, C., Hopkins, Z.R., McCord, J., Strynar, M.J., Knappe, D.R.U., 2019. Fate of Per- And Polyfluoroalkyl Ether Acids in the Total Oxidizable Precursor Assay and Implications for the Analysis of Impacted Water. *Environmental Science and Technology Letters* 6, 662-668.
- Zhang, X., Lohmann, R., Dassuncao, C., Hu, X.C., Weber, A.K., Vecitis, C.D., Sunderland, E.M., 2016a. Source Attribution of Poly- and Perfluoroalkyl Substances (PFASs) in Surface Waters from Rhode Island and the New York Metropolitan Area. *Environmental Science & Technology Letters* 3, 316-321.
- Zhang, X., Lohmann, R., Dassuncao, C., Hu, X.C., Weber, A.K., Vecitis, C.D., Sunderland, E.M., 2016b. Source attribution of poly-and perfluoroalkyl substances (PFASs) in surface waters from Rhode Island and the New York Metropolitan Area. *Environmental science & technology letters* 3, 316-321.
- Zhang, X., Zhang, Y., Dassuncao, C., Lohmann, R., Sunderland, E.M., 2017. North Atlantic Deep Water formation inhibits high Arctic contamination by continental perfluorooctane sulfonate discharges. *Global Biogeochemical Cycles* 31, 1332-1343.
- Zhao, Z., Xie, Z., Möller, A., Sturm, R., Tang, J., Zhang, G., Ebinghaus, R., 2012. Distribution and long-range transport of polyfluoroalkyl substances in the Arctic, Atlantic Ocean and Antarctic coast. *Environmental Pollution* 170, 71-77.
- Zhao, Z., Xie, Z., Tang, J., Sturm, R., Chen, Y., Zhang, G., Ebinghaus, R., 2015. Seasonal variations and spatial distributions of perfluoroalkyl substances in the rivers Elbe and lower Weser and the North Sea. *Chemosphere* 129, 118-125.

Zheng, B., Liu, X., Guo, R., Fu, Q., Zhao, X., Wang, S., Chang, S., Wang, X., Geng, M., Yang, G., 2017. Distribution characteristics of poly- and perfluoroalkyl substances in the Yangtze River Delta. *Journal of Environmental Sciences* 61, 97-109.

Other Media (5.2.4)

- AMAP, 2010. AMAP Assessment 2009 – Persistent Organic Pollutants (POPs) in the Arctic. Science of the Total Environment Special Issue. 408:2851-3051- Elsevier, 2010.
- AMAP, 2016. AMAP Assessment 2015: Temporal Trends in Persistent Organic Pollutants in the Arctic. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway. In *AMAP Assessment Report*.
- AMAP, 2018. AMAP Assessment 2018: Biological Effects of Contaminants on Arctic Wildlife and Fish. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway.
- AMAP, 2021. AMAP assessment 2020: POPs and chemicals of emerging Arctic concern: influence of climate change. Arctic Monitoring and Assessment Programme (AMAP), Oslo, Norway. [In prep.].
- Apler, A., & Josefsson, S., 2016. Swedish Status and Monitoring Programme chemical contamination in offshore sediments 2003-2014. Swedish Status and Trend Monitoring Programme, Uppsala, Sweden. www.sgu.se.
- Bartalini, A., Muñoz-Arnanz, J., Marsili, L., Mazzariol, S., Fossi, M. C., & Jiménez, B., 2019. Evaluation of PCDD/Fs, dioxin-like PCBs and PBDEs in sperm whales from the Mediterranean Sea. *Science of Total Environment*, 653:1417-1425.
- Berrojalbiz, N., Dachs, J., Del Vento, S., Ojeda, M. J., Valle, M. C., Castro-Jiménez, J., Mariani, G., Wollgast, J., & Hanke, G., 2011. Persistent organic pollutants in Mediterranean seawater and processes affecting their accumulation in plankton. *Environmental Science and Technology*, 45(10):4315-4322.
- Binnington, M. J., & Wania, F., 2014. Clarifying relationships between persistent organic pollutant concentrations and age in wildlife biomonitoring: Individuals, cross-sections, and the roles of lifespan and sex. *Environmental Toxicology and Chemistry*, 33(6): 1415-1426.
- Borgå, K., & Ruus, A., 2019. Quantifying bioaccumulation in the Arctic environment. *Methods in Pharmacology and Toxicology*. Humana Press.
- Borgå, K., Saloranta, T. M., & Ruus, A., 2010. Simulating climate change-induced alterations in bioaccumulation of organic contaminants in an Arctic marine food web. *Environmental Toxicology and Chemistry*. 29(6): 1349-1357.
- Borghesi, N., Corsolini, S., & Focardi, S., 2008. Levels of polybrominated diphenyl ethers (PBDEs) and organochlorine pollutants in two species of Antarctic fish (*Chionodraco hamatus* and *Trematomus bernacchii*). *Chemosphere*, 73(2):155-160.
- Braune, B. M., Gason, A. J., Hobson, K. A., Gilchrist, H. G., & Mallory, M. L., 2014. Changes in food web structure after trend of mercury uptake at seabird colonies in the Canadian Arctic. *Environmental Science and Technology*, 48(22), 13246-13252.
- Chiuchiolo, A. L., Dickhut, R. M., Cochran, M. A., & Ducklow, H. W., 2004. Persistent organic pollutants at the base of the Antarctic marine food web. *Environmental Science and Technology*, 38(13): 3551-3557.
- Cincinelly, A., Martellini, T., Pozo, K., Kukučka, P., Audy, O., & Corsolini, S., 2016. *Trematomus bernacchii* as an indicator of POP temporal trend in the Antarctic seawaters. *Environmental Pollution*, 217:19-25.
- Corsolini, S., 2009. Industrial contaminants in the Antarctic biota. *Journal of Chromatography A*, 1216(3): 598-612. J Chromatogr A.
- Corsolini, S., 2012. Persistent organic chemicals in coastal, oceanic, Arctic, and Antarctic regions. In: Loganathan, B. G., Lam, P. K. S. (Eds.), *Global Contamination Trends of*

- Persistent Organic Chemicals*, CRC Press Taylor & Francis Group, Boca Raton, FL, p. 571-592.
- Corsolini, S., Ademollo, N., Romeo, T., Olmastroni, S., Focardi, S., 2003. Persistent organic pollutants in some species of the Ross Sea pelagic trophic web. *Antarctic Science*, 15: 95-104.
- Corsolini, S., Borghesi, N., Ademollo, N., Focardi, S., 2011. Chlorinated biphenyls and pesticides in migrating and resident seabirds from East and West Antarctica. *Environment International*, 37(8): 1329-1335.
- Corsolini, S., Covaci, A., Ademollo, N., Focardi, S., & Schepens, P., 2006. Occurrence of organochlorine pesticides (OPCs) and their enantiomeric signatures, and concentrations of polybrominated diphenyl ethers (PBDEs) in the Adélie penguin food web, Antarctica. *Environmental Pollution*, 140(2): 371-382.
- Czub, G. & McLachlan, M. S., 2004. A food chain model to predict the levels of lipophilic organic contaminants in humans. *Environmental Toxicology and Chemistry*, 10: 2356-2366.
- EC, 2000. Directive 2000/60/EC of the European Parliament and of the Council of 23 October 2000 establishing a framework for community action in the field of water policy. https://eur-lex.europa.eu/resource.html?uri=cellar:5c835afb-2ec6-4577-bdf8-756d3d694eeb.0004.02/DOC_1&format=PDF
- EC, 2008. Directive 2008/105/EC of the European Parliament and of the Council of 16 December 2008 on environmental quality standards in the field of water policy, amending and subsequently repealing Council Directives 82/176/EEC, 83/513/EEC, 84/156/EEC, 84/491/EEC, 86/280/EEC, and amending Directive 2000/60/EC of the European Parliament and of the Council. https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:32008L0105#ntr5-L_2008348EN.01009201-E0005
- EC, 2010. Directive 2008/105/EC of the European Parliament and of the Council of 16 December 2008 on environmental quality standards in the field of water policy, amending and subsequently repealing Council Directives 82/176/EEC, 83/513/EEC, 84/156/EEC, 84/491/EEC, 86/280/EEC, and amending Directive 2000/60/EC of the European Parliament and of the Council. Commission Decision of 1 September 2010 on criteria and methodological standards on good environmental status of marine waters. <https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A32010D0477%2801%29>
- EC, 2017. Commission staff working document. Accompanying the document. Report from the Commission to the European Parliament and the Council assessing Member States' monitoring programmes under the Marine Strategy Framework Directive {COM(2017) 3 final}.
- Environment and Climate Change Canada and the U.S. Environmental Protection Agency, 2019. State of the Great Lakes, 2019 Technical Report. Cat No. En161-3/1E-PDF. EPA 905-R-20-004. Available at binational.net [In prep.].
- EU, 2019. Regulation (EU) 2019/1021 of the European Parliament and of the Council of 20 June 2019 on persistent organic pollutants. <https://eur-lex.europa.eu/legal-content/en/TXT/?uri=CELEX:32019R1021>
- Focardi, S., Lari, L., & Marsili, L., 1992. PCB congeners, DDTs and hexachlorobenzene in Antarctic fish from Terra Nova Bay (Ross Sea). *Antarctic Science*, 4(2): 151-154.

- Geisz, H. N., Dickhut, R. M., Cochran, M. A., Fraser, W. R., & Ducklow, H. W., 2008. Melting glaciers: A probable source of DDT to the Antarctic marine ecosystem. *Environmental Science and Technology*, 42(11): 3958-3962.
- Goerke, H., Weber, K., Bornemann, H., Ramdohr, S., & Plötz, J., 2004. Increasing levels and biomagnification of persistent organic pollutants (POPs) in Antarctic biota. *Marine Pollution Bulletin*, 48(3-4): 295-302.
- Gómez-Gutiérrez, A., Garnacho, E., Bayona, J. M., & Albaigés, J., 2007. Screening ecological risk assessment of persistent organic pollutants in Mediterranean sea sediments. *Environment International*. 33(7): 867-876.
- Hartwell, S. I., Apeti, A. D., Pait, A. S., Radenbaugh, T., & Britton, R., 2018. Benthic habitat contaminant status and sediment toxicity in Bristol Bay, Alaska. *Regional Studies in Marine Science*, 24: 343-354.
- Hebert, C. E., Hobson, K. A., & Shutt, J. K., 2000. Changes in food web structure affect rate of PCB decline in herring gull (*Larus argentatus*) eggs. *Environmental Science and Technology*, 34(9): 1609-1614.
- Hebert, C. E., & Popp, B. N., 2018. Temporal trends in a biomagnifying contaminant: Application of amino acid compound-specific stable nitrogen isotope analysis to the interpretation of bird mercury levels. *Environmental Toxicology and Chemistry*, 37(5): 1458-1465.
- Hebert, C. E., & Weseloh, D. V. C., 2006. Adjusting for temporal change in trophic position results in reduced rates of contaminant decline. *Environmental Science and Technology*, 40(18): 5624-5628.
- HELCOM, 2018a. State of the Baltic Sea – Second HELCOM holistic assessment 2011-2016. *Baltic Sea Environmental Proceedings 155*. Baltic Marine Environment Protection Commission – Helsinki Commission (HELCOM), Helsinki, Finland.
- HELCOM, 2018b. HELCOM Thematic assessment of hazardous substances 2011-2016. Baltic Marine Environment Protection Commission – Helsinki Commission (HELCOM), Helsinki, Finland. Available at: <http://www.helcom.fi/baltic-sea-trends/holistic-assessments/state-of-the-baltic-sea2018/reports-and-materials/>
- HELCOM, 2018c. Hexabromocyclododecane (HBCDD). *HELCOM Core Indicator Report*. Baltic Marine Environment Protection Commission – Helsinki Commission (HELCOM), Helsinki, Finland. Online [Viewed 30.10.19], [<https://helcom.fi/wp-content/uploads/2019/08/Hexabromocyclododecane-HBCDD-HELCOM-core-indicator-2018.pdf>]
- HELCOM, 2018d. Polybrominated diphenyl ethers (PBDEs). *HELCOM Core Indicator Report*. Baltic Marine Environment Protection Commission – Helsinki Commission (HELCOM), Helsinki, Finland. Online [Viewed 30.10.19], [<https://helcom.fi/wp-content/uploads/2019/08/Polybrominated-diphenyl-ethers-PBDEs-HELCOM-core-indicator-2018.pdf>]
- HELCOM, 2018e. PCBs, dioxins, and furans. *HELCOM Core Indicator Report*. Baltic Marine Environment Protection Commission – Helsinki Commission (HELCOM), Helsinki, Finland. Online [Viewed 30.10.19], [<https://helcom.fi/wp-content/uploads/2019/08/Polychlorinated-biphenyls-PCBs-dioxin-and-furan-HELCOM-core-indicator-2018.pdf>]
- HELCOM, 2018f. Perfluorooctane sulphonate (PFOS). *HELCOM Core Indicator Report*. Baltic Marine Environment Protection Commission – Helsinki Commission (HELCOM),

- Helsinki, Finland. Online [Viewed 30.10.19], [<https://helcom.fi/wp-content/uploads/2019/08/Perfluorooctane-sulphonate-PFOS-HELCOM-core-indicator-2018.pdf>]
- HELCOM, 2020a. Undeman, E. and Johansson, J. 2020. Polybrominated diphenyl ethers (PBDEs) in the Baltic Sea – Sources, transport routes and trends. Helcom Baltic Sea Environment Proceedings n°172
- HELCOM, 2020b. McLachlan, M. & Undeman, E. 2020. Dioxins and PCBs in the Baltic Sea. Helcom Baltic Sea Environment Proceedings n°171
- HELCOM, 2020c. Johansson, J. and Undeman, E. 2020. Perfluorooctane sulfonate (PFOS) and other perfluorinated alkyl substances (PFASs) in the Baltic Sea – Sources, transport routes and trends. Helcom Baltic Sea Environment Proceedings n°173
- Hermanson, M. H., Isaksson, E., Forström, S., Teixeira, C., Muir, D. C. G., Pohjola, V. A., & Van De Wal, R. S. V., 2010. Deposition history of brominated flame retardant compounds in an ice core from Holtedahlfonna, Svalbard, Norway. *Environmental Science and Technology*, 44(19): 7405-7410.
- Hickie, B. E., Mackay, D., & De Koning, J., 1999, Lifetime pharmacokinetic model for hydrophobic contaminants in marine mammals. *Environmental Toxicology and Chemistry*, 18(11): 2622-2633.
- Hickie, B. E., Muir, D. C. G., Addison, R. F., & Hoekstra, P. F., 2005. Development and application of bioaccumulation models to assess persistent organic pollutant temporal trends in arctic ringed seal (*Phoca hispida*) populations. *Science of the Total Environment*, 351.352: 413-426.
- Houde, M., Wang X., Colson, T. L. L., Gagnon, P., Ferhuson, S. H., Ikonou, M. G., Dubetz, C., Addison, R. F., & Muir, D. C. G., 2019. Trends of persistent organic pollutants in ringed seals (*Phoca hispida*) from the Canadian Arctic. *Science of the Total Environment*, 665: 1135-1146.
- Kim, J. T., Son, M. H., Kang, J. H., Kim, J. H., Jung, J. W., & Chang, Y. S., 2015. Occurrence of legacy and new persistent organic pollutants in avian tissue from King George Island, Antarctica. *Environmental Science and Technology*. 49(22): 13628-13638.
- Kimbrough, K. L., Johnson, W. E., Lauensein, G. G., Christensen, J. D., & Apeti, D. A., 2008. An assessment of two decades of contaminant monitoring in the nation's coastal zone. *NOAA Technical Memorandum*, NOS NCCOS 74.
- Law, R., Hanke, G., Angelidis, M., Batty, J., Bignert, A., Dachs, J., Davies, I., Denga, Y., Duffek, A., Herut, B., Hylland, K., Lepom, P., Leonards, P., Mehtonen, J., Piha, H., Roose, P., Tronczynski, J., Velikova, V., & Vethaak, D., 2010. Marine Strategy Framework Directive – Task group 8 report contaminants and pollution effects. EUR 24335 EN - Joint Research Centre Scientific and Technical Reports. Luxembourg, Office for official Publications of the European Communities. *Scientific and Technical Research series*, 161pp, ISSN 978-92-79-15648-9.DOI10.2788/85887.
- Liber, Y., Mourier, B., Marchand, P., Bichon, E., Perrodin, Y., & Bedell, J. P., 2019. Past and recent state of sediment contamination by persistent organic pollutants (POPs) in the Rhône River: Overview of ecotoxicological implications. *Science of the Total Environment*, 646: 1037-1046.
- MacLeod, M., Woodfine, D. G., Mackay, D., McKone, T., Bennet, D., & Maddalena, R., 2001. BETR North America, A regionally segmented multimedia contaminant fate model for North America. *Environmental Science and Pollution Research*, 8(3): 156-163.

- Marsili, L., Jiménez, B., & Borrell, A., 2018. Persistent organic pollutants in cetaceans living in a hotspot area: The Mediterranean Sea. *Marine Mammal Ecotoxicology: Impacts of Multiple Stressors on Population Health*, 185-212. Academi Press.
- McLachlan, M. S., Undeman, E., Zhao, F., & MacLeod, M., 2018. Predicting global scale exposure of humans to PCB 153 from historical emissions. *Environmental Science: Processes and Impacts*, 20(5), 747-756.
- Merhaby, D., Radobonirina, S., Net, S., Ouddane, B., & Halwani, J., 2019. Overview of sediments pollution by PAHs and PCBs in Mediterranean basin: Transport, fate, occurrence, and distribution. *Marine Pollution Bulletin* (149).
- Muir, D. C. G., & de Wit, C. A., 2010. Trends of legacy and new persistent organic pollutants in the circumpolar arctic: Overview. Conclusions, and recommendations. *Science of the Total Environment*, 408(15): 3044-3051.
- NOAA, 2014. *World's largest ever recorded iceberg continues to break apart near Antarctica* [News release]. National Oceanic and Atmospheric Administration (NOAA), Silver Spring, USA.
- Nøst, T. H., Breivik, K., Wania, F., Rylander, C., Odland, J. Ø., & Sandanger, T. M., 2016. Estimating time-varying PCB exposures using person-specific predictions to supplement measured values: A comparison of observed and predicted values in two cohorts of Norwegian women. *Environmental Health Perspectives*, 124(3): 299-305.
- Ontiveros-Cuadras, J. F., Ruiz-Fernández, A. C., Sanchez-Cabeza, J. A., Sericano, J., Pérez-Bernal, L. H., Páez-Osuna, F., Dunbar, R. B., & Mucciarone, D. A., 2019. Recent history of persistent organic pollutants (PAHs, PCBs, PBDEs) in sediments from a large tropical lake. *Journal of Hazardous Materials*, 368: 264-273.
- OSPAR, 2017a. Status and trends of polychlorinated biphenyls (PCB) in sediments. *Intermediate Assessment 2017*. Oslo-Paris Convention for the Protection of the Marine Environment of the North-East Atlantic (OSPAR). Online [Viewed 19.11.19]. Available at: <https://oap.ospar.org/en/ospar-assessments/intermediate-assessment-2017/pressures-human-activities/contaminants/pcb-sediment/>
- OSPAR, 2017b. Status and trends of polybrominated diphenyl ethers (PBDEs) in sediments. *Intermediate Assessment 2017*. Oslo-Paris Convention for the Protection of the Marine Environment of the North-East Atlantic (OSPAR). Online [Viewed 19.11.19]. Available at: <https://oap.ospar.org/en/ospar-assessments/intermediate-assessment-2017/pressures-human-activities/contaminants/pbde-sediment/>
- OSPAR, 2017c. Status and trends of polychlorinated biphenyls (PCBs) in fish and shellfish. *Intermediate Assessment 2017*. Oslo-Paris Convention for the Protection of the Marine Environment of the North-East Atlantic (OSPAR). Online [Viewed 19.11.19]. Available at: <https://oap.ospar.org/en/ospar-assessments/intermediate-assessment-2017/pressures-human-activities/contaminants/pcb-fish-shellfish/>
- OSPAR, 2017d. Status and trends of polybrominated diphenyl ethers (PBDEs) in fish and shellfish. *Intermediate Assessment 2017*. Oslo-Paris Convention for the Protection of the Marine Environment of the North-East Atlantic (OSPAR). Online [Viewed 19.11.19]. Available at: <https://oap.ospar.org/en/ospar-assessments/intermediate-assessment-2017/pressures-human-activities/contaminants/pbde-fish-shellfish/>
- OSPAR, 2017e. Levels and trends in marine contaminants and their biological effects – CEMP Assessment report 2016. *Monitoring Assessment Series*, Publication number: 695/2017, ISBN: 978-1-911458-35-7.

- OSPAR, 2018. Levels and trends in marine contaminants and their biological effects – CEMP Assessment report 2017. *Monitoring Assessment Series*, Publication number: 712/2018, ISBN: 978-1-911458-52-4.
- OSPAR, 2019. Levels and trends in marine contaminants and their biological effects – CEMP Assessment report 2018. *Monitoring Assessment Series*, Publication number: 736/2019, ISBN: 978-1-911458-76-0.
- Quinn, C. L., Armitage, J. M., Berivik, K., & Wania, F., 2012. A methodology for evaluating the influence of diets and intergenerational dietary transitions on historic and future human exposure to persistent organic pollutants in the Arctic. *Environmental International*, 49: 83-91.
- Quinn, C. L., & Wania, F., 2012. Understanding differences in the body burden-age relationships of bioaccumulating contaminants based on populations cross sections versus individuals. *Environmental Health Perspectives*, 120(4): 554-559.
- Rigét, F., Bignert, A., Braune, B., Dam, M., Dietz, R., Evans, M., Green, N., Gunnlaussdóttir, H., Hoydal, K. S., Kucklick, J., Letcher, R., Muir, D., Schuur, S., Sonne, C., Stern, G., Tomy, G., Vorkamp, K., & Wilson, S., 2019. Temporal trends of persistent organic pollutants in Arctic marine and freshwater biota. *Science of the Total Environment*, 649: 99-110.
- Risebrough, R. W., Menzel, D. B., Martin, D. J., & Olcott, H. S., 1967. DDT residues in pacific sea birds: A persistent insecticide in marine food chains. *Nature*, 216(5115): 589-591.
- Ritter, R., Scheringer, M., MacLeod, M., Schenker, U., & Hungerbühler, K., 2009. A multi-individual pharmacokinetic model framework for interpreting time trends of persistent chemicals in human populations: Application to a postban situation. *Environmental Health Perspectives*, 117(8): 1280-1286.
- Roscales, J. L., Vicente, A., Muñoz-Arnanz, J., Morales, L., Abad, E., Aguirre, J. I., & Jiménez, B., 2016. Influence of trophic ecology on the accumulation of dioxins and furans (PCDD/Fs), non-ortho polychlorinated biphenyls (PCBs), and polybrominated diphenyl ethers (PBDEs) in Mediterranean gulls (*Larus michahellis* and *L. audouinii*): A three-isotope approach. *Environmental Pollution*, 212: 307-315.
- Schneider, R., Steinhagen-Schneider, G., & Drescher, H. E., 1985. Organochlorines and heavy metals in seals and birds from the Weddell Sea. *Antarctic Nutrient Cycles and Food Webs*, 652-655.
- Sericano, J. L., Wade, T. L., Sweet, S. T., Ramirez, J., & Lauenstein, G. G., 2014. Temporal trends and spatial distribution of DDT in bivalves from the coastal marine environments of the continental United States, 1986-2009. *Marine Pollution Bulletin*, 81(2): 303-316.
- Sladen, W. J. L., Menzie, C. M., & Reichel, W. L., 1966. DDT residues in Adelie penguins and a crabeater seal from Antarctica. *Nature*, 210(5037), 670-673.
- Strobel, A., Schmid, P., Segner, H., Burkhardt-Holm, P., & Zennegg, M., 2016. Persistent organic pollutants in tissues of the white-blooded Antarctic fish *Champsocephalus gunnari* and *Chaenocephalus aceratus*. *Chemosphere*, 161: 555-562.
- Thompson, L. R., Sanders, J. G., McDonald, D., Amir, A., Ladau, J., Locey, K. J., Prill, R. J., Tripathi, A., Gibbons, S. M., Ackermann, G., Navas-Molina, J. A., Janssen, S., Kopylova, E., Vázquez-Baeza, Y., González, A., Morton, J. T., Mirarab, S., Xu, Z. Z., Jiang, L., Haroon, M. F., Kanbar, J., Zhu, Q., Song, S. J., Kosciolk, T., Bokulich, N. A., Lefler, J., Brislawn, C. J., Humphrey, G., Owens, S. M., Hampton-Marcell, J., Berg-Lyons, D., McKenzie, V., Fierer, N., Fuhrman, J. A., Clauser, A., Stevens, R. L., Shade, A., Pollard, K. S., Goodwin, K. D., Jansson, J. K., Gilbert, J. A., Knight, R., & The Earth

- Microbiome Project Consortium, 2017. A communal catalogue reveals Earth's multiscale microbial diversity. *Nature*, 551: 457-463.
- Trumble, S. J., Robinson, E. M., Berman-Kowalewski, M., Potter, C. W., & Usenko, S., 2013. Blue whale earplug reveals lifetime contaminant exposure and hormone profiles. *Proceeding of the National Academy of Science of the United States of America*, 110(42), 16922-16926.
- UNEP, 2007. Report of the Conference of the Parties of the Stockholm Convention on persistent organic pollutants on the work of its third meeting. United Nations Environmental Programme (UNEP), Athens, Greece.
- UNEP/MAP, 2016. Integrated Monitoring and Assessment Programme of the Mediterranean Sea and coast and related assessment criteria. United Nations Environmental Programme/Mediterranean Action Plan (UNEP/MAP), Athens, Greece.
- UNEP/MAP, 2017. 2017 Mediterranean Quality Status Report. United Nations Environmental Programme/Mediterranean Action Plan (UNEP/MAP), Athens, Greece.
- van den Brink, N. W., Riddle, M. J., van den Heuvel-Grevem M., & van Franeker, J. A., 2011. Contrasting time trends of organic contaminants in Antarctic pelagic and benthic food webs. *Marine Pollution Bulletin*, 62(1): 128-132.
- Wania, F., Binnington, M. J., & Curren, M. S., 2017. Mechanistic modeling of persistent organic pollutant exposure among indigenous Arctic populations: Motivations, challenges, and benefits. *Environmental Reviews*, 25(4): 396-407.
- Weber, K., & Goerke, H., 2003. Persistent organic pollutants (POPs) in Antarctic fish: Levels, patterns, changes. *Chemosphere*, 53(6): 667-678.

Long Range Transport (5.3)

- Bogdal, C.; Wang, Z. Y.; Buser, A. M.; Scheringer, M.; Gerecke, A. C.; Schmid, P.; Muller, C. E.; MacLeod, M.; Hungerbuhler, K., Emissions of polybrominated diphenyl ethers (PBDEs) in Zurich, Switzerland, determined by a combination of measurements and modeling. *Chemosphere* 2014, 116, 15-23. (a)
- Bogdal, C.; Muller, C. E.; Buser, A. M.; Wang, Z. Y.; Scheringer, M.; Gerecke, A. C.; Schmid, P.; Zennegg, M.; MacLeod, M.; Hungerbuhler, K., Emissions of Polychlorinated Biphenyls, Polychlorinated Dibenzo-p-dioxins, and Polychlorinated Dibenzofurans during 2010 and 2011 in Zurich, Switzerland. *Environ. Sci. Technol.* 2014, 48, (1), 482-490. (b)
- Breivik, K. A. Sweetman, J. M. Pacyna and K. C. Jones. 2007. Towards a global historical mission inventory for selected PCB congeners—A mass balance approach 3. An update, *Sci. Total Environ.*, 377, 296–307.
- Breivik, K., J. M. Armitage, F. Wania, A. J. Sweetman & K. C. Jones. 2016. Tracking the global distribution of persistent organic pollutants accounting for e-waste exports to developing regions, *Environ. Sci. Technol.*, 50, 798–805.
- Buser, A. M.; Kierkegaard, A.; Bogdal, C.; MacLeod, M.; Scheringer, M.; Hungerbuhler, K., Concentrations in Ambient Air and Emissions of Cyclic Volatile Methylsiloxanes in Zurich, Switzerland. *Environ. Sci. Technol.* 2013, 47, (13), 7045-7051.
- Carlsson, P., Breivik, K., Brorström-Lundén, E., Cousins, I., Christensen, J., Grimalt, J.O., Halsall, C., Kallenborn, R., Abass, K., Lammel, G., Munthe, J., MacLeod, M., Odland, J.Ø., Pawlak, J., Rautio, A., Reiersen, L.-O., Schlabach, M., Stemmler, I., Wilson, S. & Wöhrnschimmel, H. 2018. Polychlorinated biphenyls (PCBs) as sentinels for the elucidation

- of Arctic environmental change processes: a comprehensive review combined with ArcRisk project results. *Environmental Science and Pollution Research*, 25(23), p.22499–22528.
- Castro-Jimenez, J.; Gonzalez-Gaya, B.; Pizarro, M.; Casal, P.; Pizarro-Alvarez, C.; Dachs, J., Organophosphate Ester Flame Retardants and Plasticizers in the Global Oceanic Atmosphere. *Environ. Sci. Technol.* 2016, 50, (23), 12831-12839.
- Clark, D. R.; Spann, J. W.; Bunck, C. M. Dicofol (kelthane®)- induced eggshell thinning in captive American kestrels. *Environ. Toxicol. Chem.* 1990, 9 (8), 1063–1069.
- Cowan-Ellsberry, C., McLachlan, M., Arnot, J.A., MacLeod, M., McKone, T.E. & Wania, F. 2009. Modeling exposure to persistent chemicals in hazard and risk assessment. *Integrated Environmental Assessment and Management*, 5(4), p.662–679.
- Czub, G., & McLachlan, M. S. (2004). A food chain model to predict the levels of lipophilic organic contaminants in humans. *Environmental Toxicology and Chemistry*, 23(10), 2356–2366.
- FAO 2017: FAO food balance sheets, <http://www.fao.org/faostat/en/#data/FBS>, accessed in September 2017.
- Gasic, B.; Moeckel, C.; MacLeod, M.; Brunner, J.; Scheringer, M.; Jones, K. C.; Hungerbühler, K., Measuring and Modeling Short-Term Variability of PCBs in Air and Characterization of Urban Source Strength in Zurich, Switzerland. *Environ. Sci. Technol.* 2009, 43, (3), 769–776.
- Göktas, R.K. and M. MacLeod, Remoteness from sources of persistent organic pollutants in the multi-media global environment, *Environ. Pollut.*, 2016, 217,33–41.
- Green, N.; Schlabach, M.; Bakke, T.; Brevik, E.; Dye, C.; Herzke, D.; Huber, S.; Plosz, B.; Remberger, M.; Schøyen, M., Screening of selected metals and new organic contaminants 2007. Phosphorous Flame Retardents, Polyfluorinated Organic Compounds, Nitro-PAHs, Silver, Platinum and Sucralose in Air, Wastewater, Treatment Facilities, and Freshwater and Marine Recipients, Report TA2367;. Norwegian Pollution Control Agency 2008.
- Grennfelt, P., Engleryd, A., Munthe, J., & Håård, U. Saltsjöbaden V – Taking international air pollution policies into the future. Nordic Council of Ministers 2013, <http://dx.doi.org/10.6027/TN2013-571>.
- Gusev, A., MacLeod, M. & Bartlett, P. 2012. Intercontinental transport of persistent organic pollutants: A review of key findings and recommendations of the Task Force on Hemispheric Transport of Air Pollutants and directions for future research. *Atmospheric Pollution Research*, 3, 463-465.
- Hoekstra, P. F.; Burnison, B. K.; Garrison, A. W.; Neheli, T.; Muir, D. C. Estrogenic activity of dicofol with the human estrogen receptor: Isomer-and enantiomer-specific implications. *Chemosphere* 2006, 64 (1), 174–177.
- Hulek, R., J. Boruvkova, J. Gregor, J. Kalina, Z. Bednarova, K. Sebkova, O. Melkes, R. Novak, J. Jarkovsky, L. Dusek and J. Klanova, Global Monitoring Plan of the Stockholm Convention on Persistent Organic Pollutants: visualisation and on-line analysis of global levels of chemicals in air, water, breast milk and blood, <http://www.pops-gmp.org/visualization-2014>, accessed in September 2014.
- Huang, T.; Tian, C. G.; Zhang, K.; Gao, H.; Li, Y. F.; Ma, J. M., Gridded atmospheric emission inventory of 2,3,7,8-TCDD in China. *Atmos. Environ.* 2015, 108, 41-48.

- Huang, T., Ling, Z., Ma, J., Macdonald, R. W., Gao, H., Tao, S., & Mao, X. (2020). Human exposure to polychlorinated biphenyls embodied in global fish trade. *Nature Food*, 1(5), 292–300. <https://doi.org/10.1038/s43016-020-0066-1>
- Hung, H., MacLeod, M., Guardans, R., Scheringer, M., Barra, R., Harner, T. & Zhang, G. 2013. Toward the next generation of air quality monitoring: Persistent organic pollutants. *Atmospheric Environment*, 80, p.591–598.
- Jiang, W. Y. H.; Huang, T.; Mao, X. X.; Wang, L.; Zhao, Y.; Jia, C. H.; Wang, Y. N.; Gao, H.; Ma, J. M., Gridded emission inventory of short-chain chlorinated paraffins and its validation in China. *Environ. Pollut.* 2017, 220, 132-141.
- Kojima, H.; Katsura, E.; Takeuchi, S.; Niiyama, K.; Kobayashi, K. Screening for estrogen and androgen receptor activities in 200 pesticides by in vitro reporter gene assays using Chinese hamster ovary cells. *Environ. Health Perspect.* 2004, 112 (5), 524.
- Lessenger, J. E.; Riley, N. Neurotoxicities and behavioral changes in a 12-year-old male exposed to dicofol, an organochloride pesticide. *J. Toxicol. Environ. Health A* 1991, 33 (3), 255–261.
- Li, C.; Chen, J. W.; Xie, H. B.; Zhao, Y. H.; Xia, D. M.; Xu, T.; Li, X. H.; Qiao, X. L., Effects of Atmospheric Water on ·OH-initiated Oxidation of Organophosphate Flame Retardants: A DFT Investigation on TCPP. *Environ. Sci. Technol.* 2017, 51, (9), 5043-5051.
- Li, L., Liu, J., & Hu, J. (2015). Global inventory, long-range transport and environmental distribution of dicofol. *Environmental Science and Technology*, 49(1), 212–222.
- Li, J.; Xie, Z.; Mi, W.; Lai, S.; Tian, C.; Emeis, K.-C.; Ebinghaus, R., Organophosphate Esters in Air, Snow and Seawater in the North Atlantic and the Arctic. *Environ. Sci. Technol.* 2017, 51, 6887–6896.
- Li, J.; Zhao, F.; Xie, Z.; Ebinghaus, R.; Emeis, K.-C.; Tian, C.; MacLeod, M. Global gridded emissions and atmospheric transport of tris-(1-chloro-2-propyl) phosphate (TCPP) (in press)
- MacLeod, M.; Scheringer, M.; Podey, H.; Jones, K. C.; Hungerbuhler, K., The origin and significance of short-term variability of semivolatile contaminants in air. *Environ. Sci. Technol.* 2007, 41, (9), 3249-3253.
- MacLeod, M., Scheringer, M., McKone, T.E. & Hungerbuhler, K. 2010. The state of multimedia mass-balance modeling in environmental science and decision-making. *Environmental Science & Technology*, 44(22), p.8360–8364.
- MacLeod, M., Von Waldow, H., Tay, P., Armitage, J. M., Wöhrnschimmel, H., Riley, W. J., McKone, T.E. & Hungerbuhler, K. 2011. BETR Global - A geographically-explicit global-scale multimedia contaminant fate model. *Environmental Pollution*, 159(5), 1442–1445.
- Marklund, A.; Andersson, B.; Haglund, P., Organophosphorus flame retardants and plasticizers in air from various indoor environments. *J. Environ. Monit.* 2005, 7, (8), 814-819.
- McLachlan, M. 2018. Can the Stockholm convention address the spectrum of chemicals currently under regulatory scrutiny? Advocating a more prominent role for modeling in POP screening assessment. *Environmental Science: Processes & Impacts*. 20, 32-37.
- McLachlan, M. S., Undeman, E., Zhao, F., & MacLeod, M. 2018. Predicting global scale exposure of humans to PCB 153 from historical emissions. *Environmental Science: Processes and Impacts*, 20(5), 747–756.
- Möller, A.; Sturm, R.; Xie, Z. Y.; Cai, M. H.; He, J. F.; Ebinghaus, R., Organophosphorus Flame Retardants and Plasticizers in Airborne Particles over the Northern Pacific and Indian Ocean toward the Polar Regions: Evidence for Global Occurrence. *Environ. Sci. Technol.* 2012, 46, (6), 3127-3134

- Moeckel, C., Breivik, K., Nøst, T. H., Sankoh, A., Jones, K. C., & Sweetman, A. (2020). Soil pollution at a major West African E-waste recycling site: Contamination pathways and implications for potential mitigation strategies. *Environment International*, 137, 105563.
- National Geophysical Data Center, Available at:
<https://ngdc.noaa.gov/eog/dmsp/downloadV4composites.html>
- OSPAR Commission. OSPAR Background Document on Dicofol; Convention for the Protection of the Marine Environment of the North-East Atlantic (OSPAR Convention), 2002.
- POPRC 2013: Persistent Organic Pollutants Review Committee (POPRC). Report of the Persistent Organic Pollutants Review Committee on the Work of Its Ninth Meeting, (UNEP/POPS/POPRC.9/13); Persistent Organic Pollutants Review Committee, the Stockholm Convention on Persistent Organic Pollutants: Rome, 2013.
- Reynolds, P.; Von Behren, J.; Gunier, R. B.; Goldberg, D. E.; Harnly, M.; Hertz, A. Agricultural pesticide use and childhood cancer in California. *Epidemiology* 2005, 16 (1), 93–100.
- Rodgers, T. F. M.; Truong, J. W.; Jantunen, L. M.; Helm, P. A.; Diamond, M. L., Organophosphate Ester Transport, Fate, and Emissions in Toronto, Canada, Estimated Using an Updated Multimedia Urban Model. *Environ. Sci. Technol.* 2018, 52, (21), 12465-12474.
- Salamova, A.; Hermanson, M. H.; Hites, R. A., Organophosphate and Halogenated Flame Retardants in Atmospheric Particles from a European Arctic Site. *Environ. Sci. Technol.* 2014, 48, (11), 6133-6140.
- Schenker, U.; Scheringer, M.; Sohn, M. D.; Maddalena, R. L.; McKone, T. E.; Hungerbühler, K., Using Information on Uncertainty to Improve Environmental Fate Modeling: A Case Study on DDT. *Environ. Sci. Technol.* 2009, 43, (1), 128-134.
- Sühring, R.; Diamond, M. L.; Scheringer, M.; Wong, F.; Pucko, M.; Stern, G.; Burt, A.; Hung, H.; Fellin, P.; Li, H.; Jantunen, L. M., Organophosphate Esters in Canadian Arctic Air: Occurrence, Levels and Trends. *Environ. Sci. Technol.* 2016, 50, (14), 7409-7415.
- UBC, 2014: Map of Global Fishing 2014, The Sea Around Us Research Initiative at The University of British Columbia, [http:// www.seaaroundus.org/data/#/spatial-catch](http://www.seaaroundus.org/data/#/spatial-catch), accessed in September 2017.
- UN 2017: GEMS cluster diets, [https://undatacatalog.org/dataset/ gemsfood-consumption-database](https://undatacatalog.org/dataset/gemsfood-consumption-database), accessed in September 2017.
- UNECE (United Nations Economic Commission for Europe). Hemispheric Transport of Air Pollutants 2010, Part C: Persistent Organic Pollutants. Air Pollution Studies No. 19, Edited by S Dutchak and A Zuber. EC/EB.AIR/102, United Nations Publication, New York. <http://www.htap.org/>
- UNEP/AMAP, 2011. Climate Change and POPs: Predicting the Impacts. Report of the UNEP/AMAP Expert Group. Secretariat of the Stockholm Convention, Geneva. 62 pp. <https://www.amap.no/documents/doc/climate-change-and-pops-predicting-the-impacts/753>
- USEPA 1998: United States Environmental Protection Agency (USEPA). Reregistration Eligibility Decision (RED), Dicofol, EPA 738-R-98-018; U.S. Environmental Protection Agency (USEPA): Washington D.C., 1998.
- USEPA, 2017. EPI Suite v4.11. Available online: <https://www.epa.gov/tsca-screening-tools/download-epi-suite-tm-estimation-program-interface-v411>
- Undeman, E., Brown, T. N., McLachlan, M. S., & Wania, F. (2018). Who in the world is most exposed to polychlorinated biphenyls? Using models to identify highly exposed populations. *Environmental Research Letters*, 13(6).

- van den Berg, M., K. Kypke, A. Kotz, A. Tritscher, S. Y. Lee, K. Magulova, H. Fiedler and R. Malisch, WHO/UNEP global surveys of PCDDs, PCDFs, PCBs and DDTs in human milk and bene?t – risk evaluation of breastfeeding, *Arch. Toxicol.*, 2017, 91,83–96.
- van der Veen, I.; de Boer, J., Phosphorus flame retardants: Properties, production, environmental occurrence, toxicity and analysis. *Chemosphere* 2012, 88, (10), 1119-1153.
- von Waldow, H.; MacLeod, M.; Scheringer, M.; Hungerbuhler, K., Quantifying Remoteness from Emission Sources of Persistent Organic Pollutants on a Global Scale. *Environ. Sci. Technol.* 2010, 44, (8), 2791-2796.
- Wania, F.; Mackay, D. A global distribution model for persistent organic chemicals. *Sci. Total Environ.* 1995, 160, 211–232
- Wania, F.; Mackay, D. The Global Distribution Model. A Nonsteady-State Multi-Compartmental Mass Balance Model of the Fate of Persistent Organic Pollutants in the Global Environment; Technical Report; 2000.
- Weem, A. P. Exploration of Management Options for Dicofol; VROM, Ministry of Environment: The Netherlands, 2010.
- Wiemeyer, S. N.; Clark, D. R.; Spann, J. W.; Belisle, A. A.; Bunck, C. M. Dicofol residues in eggs and carcasses of captive American kestrels. *Environ. Toxicol. Chem.* 2001, 20 (12), 2848–2851.

Conclusions (6)

Saini, A., Harner, T., Chinnadhurai, S., Schuster, J.K., Yates, A., Sweetman, A., Aristizabal-Zuluaga, B.H., Jimenez, B., Manzano, C.A., Gaga, E.O., Stevenson, G., Falandysz, J., Ma, J., Miglioranza, K.S.B., Kannan, K., Tominaga, M., Jariyasopit, N., Rojas, N.Y., Amador-Munoz, O., Sinha, R., Alani, R., Suresh, R., Nishino, T., Shoeib, T. GAPS-megacities: A new global platform for investigating persistent organic pollutants and chemicals of emerging concern in urban air. *Environ. Pollut.*, 2020, 267,115416

8 ANNEX

The following annexes to the WEOG report contain supplementary information and data. These annexes are available as separate documents.

8.1 Ambient Air (8.1a) and AMAP (8.1b)

8.2 Human Tissues

8.3 Water

8.4 Other Media