Annex 8.4: Other Media (March 2015)

In the following pages some of the relevant paragraphs of the reports that have been used in the Other media chapter, and in particular Table 5.2.4.1 are cited in detail below.

1 Northern Contaminants Program NCP

The text below is from NCP 2013, in particular chapters 3, 4 and 6.

Chapter 3 — Occurrence and Trends in the Physical Environment.
Coordinating authors: Hayley Hung and Perihan Kurt-Karakus Contributors: Lutz Ahrens, Terry Bidleman, Marlene Evans, Crispin Halsall, Tom Harner, Hayley Hung, Liisa Jantunen, Perihan Kurt-Karakus, Sum Chi Lee, Derek Muir, Mahiba Shoeib, Gary Stern, Ed Sverko, Yushan Su, Penny Vlahos, Hang Xiao

Chapter 4 — Occurrence and Trends in the Biological Environment.
Coordinating authors: Derek Muir, Perihan Kurt-Karakus and Jason Stow Contributors: Jules Blais, Birgit Braune, Craig Butt, Emily Choy, Amila De Silva, Marlene Evans, Barry Kelly, Perihan Kurt-Karakus, Nic Larter, Robert Letcher, Melissa McKinney, Adam Morris, Derek Muir, Gary Stern, Greg Tomy

Chapter 6 — Interlaboratory Quality Assurance for the Northern Contaminants Program.
Coordinating authors: Victoria Tkatcheva and Eric Reiner. Contributors: Mehran Alaee, Barry Ali, Rita Dawood, Cathy Doehler, Rania Farag, Sathi Selliah, Ed Sverko, Dan Toner

The Northern Contaminants Program (NCP) was established in 1991 in response to concerns about human exposure to elevated levels of contaminants in wildlife species that are important to the Yukon First Nations, Dene Nation, Inuit Circumpolar Conference, Inuit Tapirisat of Canada, and Métis Nation of the Northwest Territories.

The 2013 report (NCP 2013) is the third assessment of persistent organic pollutants (POPs) conducted by the NCP.

Previous assessments in 1997 and 2003 summarized results of Phase 1 (1991-1996) and Phase II (1997-2002) of the NCP and included both heavy metals and POPs. Under Phase III of the program which began in 2003-04, environmental monitoring focused on fewer sampling sites for both air and biological samples. The biological sampling programs were redesigned with the goal of being able to detect a 10% annual change in contaminant concentration over a period of 10-15 years with a power of 80% and confidence level of 95%.

The list of individual compounds analyzed was expanded in Phase III particularly for perfluorinated and polyfluorinated alkyl substances (PFASs), brominated flame retardants (BFRs) and current use pesticides (CUPs). About 35 chemicals or chemical groups that

were not previously reported, or for which only very limited measurements were available in the previous assessment, have been detected particularly in arctic air, snow and biota.

(NCP 2013 p vii) executive summary:

The declining trend in concentrations in biota is most apparent for OC pesticides and less evident for PCBs and chlorobenzenes (ΣCBz). In marine species, percent annual declines of ΣDDT ranged from 2.5%/year in thick-billed murre eggs (Lancaster Sound) to 11%/year in polar bear fat (western Hudson Bay, WHB). Declines of chlordane-related compounds (ΣCHL) ranged from 1.2%/year in murre eggs to 7.4%/year in blubber of ringed seals in Hudson Bay, while polar bears (WHB) showed no decline.

Total HCHs (ΣHCH) declined 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. This increase in β-HCH in seals varied regionally, with large increases in South Beaufort Sea seals (16% at Ulukhaktok) and a decline in Hudson Bay (2.5%/ year). The case of β-HCH highlights the importance of ocean water moving through the Arctic archipelago from the Pacific Ocean via the Bering Sea and possibly Russian freshwater inputs. No other POPs shows this trend although declines of PCBs, ΣDDT, ΣCHL were lower or non-existent in beluga, ringed seals and polar bears in the South Beaufort compared to Hudson Bay and East Baffin regions.

Declines of legacy POPs have generally been more rapid in freshwater fish than in marine animals. For example, PCBs in landlocked arctic char declined by 6.4% and 7.6%/year in Amituk Lake and Lake Hazen, respectively, versus 3.8% and 4.0%/year in thick-billed murres and northern fulmars, respectively. Declines of ≥ 5%/year were also seen for ΣHCH, ΣCHL, ΣDDT and toxaphene in lake trout from Lakes Laberge, Kusawa Lake and western basin of Great Slave Lake as well as in landlocked char in Lake Hazen, Char Lake and Amituk Lake. Declines for these OC pesticides were generally <5%/year in seabird eggs and marine mammals. A notable exception was the increase in concentrations of PCBs, ΣCHL, ΣDDTs, and toxaphene were over the period 2001 to 2009 in burbot liver sampled at Fort Good Hope on the Mackenzie River. However, as of 2010 concentrations of all four POPs had returned to levels found in the 1990s and early 2000s.

A major strength of the temporal trend programs conducted under the NCP is the availability of archived samples from specimen banks.

New POPs such as PBDEs and PFOS generally increased in seals, seabirds, beluga, and polar bear samples from the 1990s until the early 2000s and are now declining. Retrospective analysis of collections from specimen banks enabled measurements of the PBDEs, PFASs and other contaminants in samples from the 1970s, ‘80s and ‘90s, and annual sampling as of early 2000s enabled relatively rapid declines to be observed. For example, ΣPBDEs achieved maximum concentrations in northern fulmar and thick-billed

murre eggs in 2005 and 2006, respectively and declined to levels similar to those in the early 1990s within 3 years.

Reviews by Braune et al. (1999) and Evans et. al. (2005) summarize the state of knowledge on POPs in fish and freshwater food webs to late 2002. The major “new chemicals” analyzed in Arctic and subarctic fresh-water biota during the period 2003–2010 were the PBDEs and PFASs. Also on the list of analytes were other brominated flame retardants such as HBCDD, PBEB, BTBPE and DBDPE. (p286)

Prominent, relatively new POPs are PBDEs and PFOS, which are detectable in all biota although generally at much lower concentrations than PCBs or toxaphene. An exception is PFOS, which is at the part per million level in polar bear liver and on par with several chlorinated POPs in polar bear adipose tissue. (p403)

HBCDD, has been reported in freshwater and marine fishes, beluga, seals, seabirds and polar bears.

Perfluorinated compounds, PBDEs and HBCDD are higher in ringed seals in Hudson Bay than other locations, reflecting the closer proximity to source regions in southern Canada and northern US. (p403)

Beluga and narwhal have the dubious distinction of having the highest concentrations of most POPs compared to other marine mammals (seals, polar bear, and walrus). Polar bears have similar levels of PCBs (in subcutaneous fat) as beluga and narwhal blubber but much lower levels of other POPs. (p404)

Spatial trends of POPs in polar bears are generally similar to those in ringed seals with higher ΣHCH in the western and central Archipelago and higher DDT, dieldrin and chlordane related compounds in Hudson Bay and eastern arctic animals. (p404)

Limited data for POPs in caribou and moose confirms the much lower levels of most chemicals compared with marine mammals. However, in caribou and moose, PFASs were the major POPs with concentrations in liver ranking ahead of PCBs and PBDEs (ΣPFCAs > PFOS > ΣPCBs > ΣPBDEs). (p404)

Annual sampling for most biota since the early-2000s has revealed some surprising trends for legacy POPs such as increasing concentrations of PCBs in burbot liver, increased toxaphene in arctic char and ringed seals, as well as increased ΣCHL and PCDD/Fs in northern fulmar eggs. (p404)

These increases (and subsequent declines in some cases) are generally unexplained but presumably related to year-to-year differences in dietary exposure of sampled animals (percent lipid, size and sex of animals generally being similar).

Concentrations of legacy POPs are generally declining in Canadian arctic freshwater fishes

(burbot, lake trout, landlocked arctic char) and in sea-run char with some important exceptions.(p404)

The results for POPs in fish suggest that overall trends in remote sites (e.g., high arctic lakes, Kusawa Lake) are mirroring the gradual decline in atmospheric concentrations, however sites influenced by local or regional sources, which may be from local use or greater atmospheric deposition in the past, have more variable and sometimes opposite short term trends. (p405)

Concentrations of most of the legacy POPs (e.g. PCBs, DDE) have decreased in Canadian Arctic seabirds since 1975 and now appear to be levelling off.

Concentrations of PBDEs increased in northern fulmars and thick-billed murres between 1975 and 2003, and now appear to be decreasing in both species.

Legacy POPs (e.g., PCBs, DDTs and chlordanerelated compounds (CHLs) and polybrominateddiphenyl ether (PBDEs), as well as perfluorinated alkyl substances (PFASs) have been found in Arctic biota and humans. Of high concern are the potential biological effects of these contaminants in exposed arctic wildlife and fish. Prior to 1997, biological effects data were minimal and insufficient at any level of biological organization. However, post-2002, new POPs effects data have been generated for several high trophic level species, including seabirds (e.g., glaucous gull, polar bears, arctic fox, and arctic char) as well as semi-captive studies on sled dogs.

There remains minimal evidence that POPs are having widespread effects on the health of arctic organisms, with the possible exception of East Greenland and Svalbard polar bears and Svalbard glaucous gulls. However, the effects of POPs in arctic wildlife have to be placed in the context of other environmental, ecological and physiological stressors (both anthropogenic and natural) that create a highly complex picture. (p431)

There are a number of local sources in the Arctic, such as derelict installations and current settlements that can have an impact on the environment and wildlife. Some of these (e.g. PCBs in Saglek bay) have been intensively studied (p 435). Significant declines in PCB concentrations in the livers of guillemot chicks between 1999–2000 and 2007 indicate that the remediation of PCB-contaminated soil at Saglek Bay has been effective in reducing PCB levels in the marine ecosystem. (p446)

An extensive review of the health effects of longrange atmospheric transport (LRAT) contaminants in arctic top predators, including polar bears, was done by Sonne (2010). Polar bears inhabit a cold environment and therefore rely on energy-rich fatty and waxy tissues as their main energy source. Since the 1940s, large amounts of fat-soluble atmospheric and marine long-range transported persistent and toxic chemicals have biomagnified in the arctic marine food webs. The review by Sonne (2010) showed that hormone and vitamin concentrations, liver, kidney and thyroid gland morphology as well as reproductive and immune systems of polar bears are likely to be influenced by contaminant exposure. Bone density reduction, neurochemical disruption and DNA

hypomethylation of the brain stem were also observed health effects of contaminants on polar bears. (p444)

ΣPCB concentrations in beluga and polar bears exceed the toxicity reference value for immunotoxicity and endocrine disruption of 1.3 μg g−1 lw in harbor seals.(p450)

Mean ΣPCB concentrations in Canadian arctic seabird eggs are well below reported thresholds for egg mortality and hatching success in fisheating birds (except for glaucous gulls). (p450)

Between 2005 and 2012, six rounds of interlaboratory studies (ILS) were conducted to evaluate the analytical data provided to the Northern Contaminants Program (NCP).

From 2007 laboratories that participated in the Arctic Monitoring and Assessment Programme (AMAP) were included and also evaluated to ensure that the pre-established NCP data quality objectives were met (Selliah et al. 2008).

The NCP 2013 report presents a summary of the study design, evaluation and statistical treatment of the results covering the period from 2005–2012 of NCP-III Phase 1 to Phase 6 (NCP-III 1-6) study. It presents the performance of NCP laboratories compared to the AMAP and all other laboratories that participated in the ILS. (p463)

Thirty-four organizations with forty-three laboratories from thirteen countries participated in NCP-III 1-6 for persistent organic pollutant (POPs) studies.

The interlaboratory studies demonstrated that the NCP laboratories are capable of producing excellent and satisfactory results for POPS analysis in the standards (72–89%) and satisfactory results for PCBs, OCPs, PCDDs analysis in certified natural-matrix materials (65–71%). (p463)

2 HELCOM

HELCOM (2010) Hazardous substances in the Baltic Sea – An integrated thematic assessment of hazardous substances in the Baltic Sea. Balt. Sea Environ. Proc. No. 120B.

PCDD/Fs

Temporal and spatial trends in sediments: There are few historical sediment data (profiles) from the Baltic Sea and some data are from the late 1980s and thus unable to reveal very recent trends. All the cores, however, show a decline in surface PCDD/F concentrations compared with deeper sediments, with the highest concentrations generally dated back to the 1970s or 1960s in the northern basins, the Baltic Proper and the Kattegat – Danish straits.

Biota

HLCOM 2010a reports that “numerous recent papers have shown differences in PCDD/F and DL-PCB concentrations in Baltic herring, sprat and salmon between the Baltic Sea basins. Higher concentrations have been detected in the northern basins where dioxin and DL-PCB levels in herring exceed established maximum limit concentrations for human consumption. Regional variation within a sub-basin has been found in the Swedish coastal region of the Bothnian Sea. Since the atmospheric deposition pattern (lowest in the north) is different from concentrations in fish (generally highest in the north), other factors or sources are thus likely to be involved in determining concentrations in fish. The reasons remain unclear, but higher historical PCDD/F discharges from point sources in the northern basins have been suggested.

There is not much information about past or recent trends in PCDD/F concentrations in different fish species and generally the data do not cover past decades. The Swedish Museum of Natural History (NRM 2009) reported dioxin concentrations in the muscle of small herring collected from 1990 to 2005 at three stations on the Swedish coast that showed no indications of change during that period, but the guillemot egg data showed a major and significant decrease since 1970.

PCB

Concentrations of PCBs, including CB-153 and CB-180, show significant declining trends in herring, perch and blue mussels in several regions around the Baltic Sea. However, only a few available data sets have time series long enough to draw statistical conclusions regarding temporal trends. Decreasing trends for other PCB congeners, as well as for the sum of seven PCBs, have also been reported for some locations along the Baltic Sea. It is estimated that levels have been decreasing by approximately 5–10% per year since the end of the 1970s.

DDT

DDE levels are higher than DDT (and DDD) levels in fish and also in some cases in sediment.

DDE levels in blue mussel (*Mytilus sp*.) and the Baltic clam *Macoma balthica* occasionally exceeded (seven out of 40 sampling sites) the lower threshold of 5 μg kg^{-1} dw (EAC low, OSPAR 1997), but the higher threshold of 50 μg kg^{-1} dw (EAC high, OSPAR 1997) was exceeded only at one site on the German coast.

The temporal trend of DDE concentrations in herring muscle has been declining since the end of the 1970s in all Baltic Sea sub-basins. Helcom 2010 a p38 presents temporal trends for the Bothnian Bay, Bothnian Sea, Gulf of Finland and Arkona Basin.The DDE level has decreased significantly at a rate of 4–11% per year since the end of the 1970s and the beginning of 1980s in most matrices analyzed (herring, perch *Perca fluvialis* and eelpout *Zoarces viviparous* muscle, cod *Gadus morhua* liver, and blue mussel *Mytilus* sp.) at several Swedish coastal sites from the Kattegat to the Bothnian Bay. From 1969–

2007, the sum of DDTs in the eggs of common guillemot *Uria aalge* showed a significant decreasing trend of 10% per year on Stora Karlsö, an island in the Western Gotland Basin. DDT has generally decreased more rapidly than the sum of DDTs, which indicates that new DDT inputs to the Baltic Sea have not taken place (Bignert et al. 2009). The sum of DDTs in herring muscle at five Finnish stations (eastern and western Gulf of Finland, Åland Sea, southern and northern Gulf of Bothnia) has declined monotonically from 1986 to 2006 and the decline has been especially significant in the Gulf of Finland and the Åland Sea (Kankaanpää 2007). The sum of DDTs in the muscle of herring from the Estonian coastal area of the eastern and central Gulf of Finland and the Gulf of Riga decreased from 1995–1998, but thereafter the levels increased to some extent until the year 2002 for unknown reasons (Roose & Roots 2005).

HCH

HCH (sum of α-, β- and γ-isomers) levels in fish and mussels were lower than the threshold (16.7 µg kg^{-1} ww). HCH was not found at all in blue mussels. Lindane occurred in sediment at levels occasionally exceeding (eleven out of 38 sites) the lower threshold concentration (1.1 µg kg^{-1} dw) and at two sites (the Sound and offshore Northern Baltic Proper) the levels were of high concern, exceeding the higher threshold (3.3 µg kg^{-1} dw) in the sub-basins of the Baltic Sea. Similarly, HCH (sum of α-, β- and γ-isomers) concentrations in sea water occasionally exceeded (six out of 35 sites) the threshold concentration (2.0 ng l^{-1}) and at one site the level was of high concern exceeding the higher threshold (20 ng l^{-1}). Nevertheless, firm conclusions could not been drawn on some water samples, because the detection limit was higher than the threshold.

The trend of lindane concentrations in herring muscle has been declining since the end of the 1980s in the Bothnian Bay, Bothnian Sea, Northern Baltic Proper and Arkona Basin. The lindane level has decreased significantly since the 1980s in most matrices analyzed (herring and perch muscle, cod liver, eelpout and blue mussel) at several Swedish coastal sites from the Kattegat.

The α-HCH concentration in herring showed a similar, but even more pronounced downward trend. Lindane levels in blue mussels from the Mecklenburg-Vorpommern coast exhibited a decrease at all six stations inves- tigated between 1994 and 2001. Since 2002, lindane has been below the limit of determination (<0.5 µg kg^{-1} dw).

HCB

Several national monitoring reports indicate a downward trend of HCB levels in the Baltic marine environment. In Sweden, the HCB level has decreased significantly in almost all matrices analysed (herring, perch and eelpout muscle, cod liver and eggs of common guillemot) at several coastal sites from the Kattegat to the Bothnian Bay. The HCB level in blue mussels has been very low and since 2000 values have been at or below the detection limit (Bignert et al. 2009). In Finland, the HCB level in herring muscle has declined since the end of the 1990s at five sites (eastern and western Gulf of Finland, Åland Sea, southern and northern Gulf of Bothnia) (Kankaanpää

2007).However, Szlinder-Richert et al. (2008) found that HCB levels did not exhibit an obvious trend from 1995–2006 in coastal and off- shore waters of the southern Baltic Sea (e.g., Gulf of Gdansk, Eastern Baltic Proper and Bornholm Basin) in any of the fish species studied (herring, sprat, cod, flounder and salmon).

Endosulfan

Endosulfan adsorbs mainly onto suspended particulate matter in the aquatic environment and then deposits onto the sediment. However, a certain proportion is likely to remain in the water column due to its relatively high water solubility.

Endosulfan sulphate, which is as toxic as endosulfan, was found in almost all of the fi sh muscle samples in the eastern Baltic Sea and off southeastern Sweden (range <0.010–0.12 µg kg^{-1} ww) (Lilja et al. 2009). Herring had higher levels than perch or flounder. However, the observed concentrations were three magnitudes lower than the predicted no-effect concentration (PNEC) level. Endosulfan sulphate is an oxidation product found in technical endosulfan, but it is also the main microbial oxidation product of α- and β-endosulfan.

PFOS/PFA

Limited data exist for PFA concentrations in Baltic Sea sediments (Nordic Council of Ministers 2004, SEPA 2006, NERI 2007, Theobald et al. 2007). PFOS and/or PFOA were occasionally detected, but consistently at levels below 1 µg kg^{-1} dw or ww. The highest levels reported so far have been from the Gulf of Finland close to Helsinki (PFOS 0.9 µg kg−1 ww), close to Stockholm (PFOS 0.6 µg kg^{-1} ww) and along the coast of Poland (PFOS and PFOA both around 0.6 µg kg^{-1} dw)

PFAs have been analysed in blue mussels (*Mytilus spp*.), various fish species, eider duck (*Somateria mollissima*), common guillemot (*Uria aalge*) as well as grey, harbour and ringed seals (*Halichoerus grypus, Phoca vitulina*, *Phoca hispida botnica*) from the Baltic Sea. Due to the large interannual and interspecies variability of results, as well as the different analytical methods and tissue types (blood, liver, muscle, egg) applied in the studies, it is difficult to derive spatial or temporal trends from the diverse literature and screening data. Distinct case studies of spatial and temporal trends of PFAs in the Baltic are therefore presented in the HELCOM 2010a report.

In general, PFOS is the predominant PFA in biota, with the highest levels in marine predatory birds and mammals. Several hundreds to one thousand µg kg^{-1} ww of PFOS have been found in the livers of grey seals (Southern Baltic Proper and Bothnian Sea), harbour seals (Great Belt and the Sound) as well as ringed seals (Bothnian Bay). In the eggs of common guillemots (Western Gotland Basin), PFOS concentrations were greater than 1000 µg kg^{-1} ww (Holmström et al. 2005). Long-chain perfluorinated carboxylates (PFCAs) also tend to accumulate in the same tissues, however, at levels one to two orders of magnitude lower than PFOS.

Compared to marine mammals and seabirds, levels in fish are generally considerably lower with some exceptions. Despite the difficulties in comparing data from different studies, sample sizes, sampling years and analytical methods, the distribution of PFOS in herring liver was found to be quite homogeneous throughout the Baltic Sea (around 10 µg kg^{-1} ww), which probably is a result of the extraordinary persistence of the compound and its use for more than three decades. A somewhat higher level was found in the proximity of Stockholm. PFCA levels were typically one to two orders of magnitude lower, with a dominance of long-chain compounds with an odd number of carbon atoms (C9, C11, C13).

Temporal trends of PFAs in common guillemot eggs from the Western Gotland Basin PFAs have been analysed retrospectively in the eggs of common guillemot from the Western Gotland Basin on the island Stora Karlsö in a time series starting in 1968. A significant increasing trend was observed for PFOS in eggs, with an increase of 7–10% per year. This corresponds to 25–30 times higher levels in the early 2000s as compared to the late 1960s. Due to relatively high inter annual variations after 1996, the future trend for PFOS in the Baltic marine environment cannot be predicted. However, the running mean smoother suggests that concentrations may have started to level off after 1997. On the other hand, the trend lines for C –C PFCAs showed an exponential increase up to 2007 (Berger et al. 2008).

PBDE

To assess the penta-BDE contamination in marine organisms in the Baltic Sea region, BDE-47 was chosen as a representative for the lower-brominated BDE group. HELCOM 2010a has evaluated concentrations of BDE-47 in relation to a concentration at the lower end of the concentration gradient in the Baltic Sea, i.e., 0.005 mg kg^{-1} lw. Thus, exceedances of the threshold do not necessarily indicate ecotoxicological concern.

The concentrations of BDE-47 varied throughout the Baltic Sea. In the eastern part (Gulf of Finlandand Gulf of Riga), concentrations were below the threshold level. Levels were moderate in the Bothnian Bay and along the Swedish east coast. In the southern regions outside the coast of Poland, levels were more than threefold higher than the threshold level. Along the Danish coast and the Swedish west coast, concentrations were good to moderate.

In marine top predators, PBDE concentrations indicate a cause for concern. For example, white-tailed sea eagles in the Baltic Sea (Nordlöf et al. 2007) have BDE concentrations (sum of four BDEs with four to six bromines) up to four times higher than the reported effect levels in exposed American kes- trels, which were causing adverse effects (Fernie et al. 2005a, 2005b, 2009).

Relatively few studies have reported PBDEs in marine sediment from the HELCOM region ,the Swedish sediment monitoring programme, covering 16 stations in the coastal and offshore areas of the Baltic Sea, showed that concentrations of BDE-47, BDE-99 and BDE-100 were clearly the highest in the Kattegat (0.44, 0.62 and 0.08 µg kg^{-1} dw,

respectively, at station Fladen.

Several time series of BDE-47 concentrations in herring muscle tissue from the Bothnian Sea, the Baltic Proper and the Kattegat showed significant decreasing trends, with half-lives in the herring populations of about 6–8 years.

HELCOM 2009 : In general, the results show that BDE47 is the dominant congener in biota of the Baltic Sea. Ranking the BDE congeners according to the concentration in biota of the Baltic Sea gives the following order: BDE47 > BDE99 & BDE100. The levels in biota are, in general, low and always lower than the PNEC level . The high BDE209 levels found in roach muscle (Burreau et al. 2004) are alarming and more information on the BDE209 levels in biota of the Baltic Sea is needed. In general, BDE209 is the dominant BDE congener in the sediment of the Baltic Sea. Ranking the BDE congeners according to concentration in the Baltic Sea sediments gives the following order: BDE209 >> BDE99 > BDE47. In general, the levels in the sediment are low and do not exceed the PNEC level.

HBCDD

Time series of HBCDD from monitoring sites along the Swedish coasts showed no significant trends in herring muscle tissue, whereas a clear increasing trend of about 3% per year ($p<0.001$) was detected in eggs from common guillemot (*Uria aalge*) collected from Stora Karlsö in the Western Gotland Basin

3 MEDPOL

The analysis covered the full period of MED POL Phase III and Phase IV, until 2010, and incorporated additional scientific information included in the four sub-regional assessment reports, which were prepared by MAP in the framework of the gradual application of the Ecosystem Approach for the management of human activities in the Mediterranean.(UNEP/MAP 2010a,b,c,d)

The UNEP/MAP 2011 report includes the families of DDTs and PCBs, as the more representative persistent organic pollutants (POPs). The Drin’s (aldrin, endrin and dieldrin), HCB and lindane were also considered. In summary, after selection and harmonization of data, a total of 33,742 observations, corresponding to more than 400 stations monitored during the MED POL Phases III and IV (1999-2010) have been included in the assessment.

Chlorinated pesticides have been extensively analyzed in Mediterranean biota since the inception of MED POL (UNEP 1990). Mussels and mullets have been the most widely studied organisms in the whole basin as part of many case studies published in the literature and recently assessed on the occasion of the implementation of the Stockholm Convention (UNEP 2002).

Concentrations of aldrin, dieldrin, endrin, lindane and hexachlorobenzene in *Mytilus*

galloprovincialis are in the low ng g^{-1} range. Concentrations of DDTs were one order of magnitude higher, with p,p'-DDE being, in general, the predominant component, although recent inputs of DDT in some areas cannot be ruled out.

Concentrations are higher in the Aegean-Levantine region for aldrin and dieldrin, and in the Western Mediterranean for HCB and lindane. However, the highest values of HCB and lindane are found in Turkey and Albania, with a number of important hot spots. In the case of DDTs, their median values are similar for the four eco-regions, around 10ng g^{-1} dw, but again very high values are found in the Adriatic, corresponding to Albania. Despite the similarity of the mean DDT levels for the four eco-regions, available data indicate that contaminants are not uniformly distributed throughout the sub-regions.

PCBs occur in the vicinity of industrial and urban sites, as well as in major river mouths. The geographical distribution of concentrations (7 ICES PCB congeners) in the indicator organism *Mytilus galloprovincialis* is shown in UNEP/MAP 2011. The median values show the higher levels in the Adriatic where the Albania samples are well above the average, with values up to 1500 ng g^{-1} dw in one station of France.

Taking into account the limited data available, trends are just preliminarily assessed. The analysis has been mainly focused on *Mytilus galloprovincialis* and *Mullus barbatus*, with the exception of Israel, where the clam *Mactra corallina* has been considered. Moreover, a few available time series for sediments from Israel (Haifa Bay) and France (Gulf of Lions) have also been considered.

In the Western Mediterranean, the stations evidencing relatively high levels of PCBs are still exhibiting increasing trends whereas those more pristine seem to be stable or decrease.

In general, it was found that during the period 1979-1998 the decreasing trends were in the order: ΣDDT > HCHs > PCBs, which may reflect that the regulation of the use of these chemicals and, consequently, of the contaminant inputs to the sea was more efficient for DDT and lindane than for PCBs.

During the last decade, HCHs (α-, β-andγ-isomers) were found in significant amounts (1–30 ng L–1) in the marine wetlands of Amvrakikos and Thermaikos Gulfs in Greece and in the coastal waters of Alexandria. Along the Spanish coast the levels were of 1.3–2 ng l^{-1}. Very recently, concentrations of 30–1500 pg l^{-1} of lindane and 5–427 pg l^{-1} of HCB were found in NW Mediterranean coastal waters, off Barcelona, basically associated with the dissolved phase. In the same stations, the concentrations of particulate and dissolved PCBs in the subsurface waters averaged 0.57 and 1.2 ng l^{-1} (ΣICES-7 congeners), respectively, in the range of those reported 15 years ago (Albaiges 2005).

HCB has been widely distributed in Western Mediterranean fish species, particularly in coastal species. Measurable amounts of HCB were found in red mullet collected in the NW basin, with levels on the Spanish coast (0.24–2.80 ng g^{-1} ww) slightly higher than on the French one (0.42–0.97 ng g^{-1} ww) (Albaiges 2005).

Two mussel surveys were conducted along the Mediterranean coast of France and Italy, between Sete and Genova, in 1973/1974 and in 1988/1989 in order to assess the temporal contamination trend by organochlorine pesticides. The comparison of concentrations measured in both surveys showed that DDTs and PCBs decreased by a factor of approximately 5 in 15 years, from730 to 130 ng g^{-1} dw and 2430 to 527 ng g^{-1} dw Aroclor equivalents, respectively. This decrease is in agreement with the ban of DDT implemented in 1975 in western Europe and the gradual cessation of PCBs production in the 1970s and 1980s. A later survey along the French coast, in 1995–1999, gave a mean concentration of 17 ng g–1 dw (1.2–157 ng g^{-1}), pointing to a sustained reduction of levels. A similar trend was observed in mussels collected close to the Ebro River mouth, where a decrease by a factor of 3 in DDTs, with a concurrent increase of the DDE/DDT ratio was observed from1980 to 1990. The same survey revealed a decrease of HCB concentrations by a factor of 5 (from 1–2 to 0.2–0.3 ng g^{-1}). (Albaiges 2005).

As far as the monitoring of POPs is concerned, the RNO is composed of 23 shellfish sampling sites, visited four times a year. Contaminants measured are DDT, DDD, DDE, lindane (γ-HCH), α-HCH, PCBs (Congeners 28, 52, 101, 105, 118, 138, 153, 180) and PAHs.

Atmospheric circulation and deposition estimates of POPs in the Mediterranean region are scarce. First calculations were reported by GESAMP, based on very few data, but showing that 80–95% of total inputs of POPs (e.g. HCB, HCH, DDT, chlordane and PCBs) were atmospheric.

The extremely informative paper by (Gómez-Gutiérrez et al., 2007) presents a compilation of measurements in sediments and has also concluded that a decreasing trend is more evident for DDTs than for PCBs, indicating a steady input of the latter in the Mediterranean Sea and the need for an improved management of their potential sources.

A detailed analysis for the 4 subregions: Western Mediterranean , Ionian Sea and and Central Mediterranean subregion, Adriatic Sea, Eastern Mediterranean subregion can be found in UNEP/MAP 2012.

Toxicological studies have found that PCB levels in deepwater fishes (*Alepocephalus rostratus*, *Bathypterois mediterraneus*, *Coryphaenoides guentheri* and *Lepidion lepidion*) were lower than in coastal fishes, close to the pollution sources, but much higher than that of fish living on the continental shelf to upper slope (*Micromesistius poutassou*, *Phycis blennoides* and *Lepidorhombus boscii*). PCB levels recorded were within the same range as that of top predators like tuna (Solé et al., 2001).

Available data indicate that organic contaminants are not uniformly distributed throughout the Western Mediterranean. For example, concentrations of total DDT in sediments range from <0.25 to 885 ng / g and PCBs from 1.3 to 7274 ng / g, higher levels corresponding to the "hot spots" areas near outfalls waste water of major cities and at the mouths of large rivers (e.g. Rhône).

The PCB congeners 31, 52, 156 and 180 are present at low concentrations and in industrial areas or urban. The PCB 153 and 138 show maximum levels of accumulation along the French coast, particularly at Marseilles and his emissary (respectively 42.3 mg / kg and 27.6 mg / kg) and to a lesser extent al mouth of the Rhone basin of the Mediterranean (after Scarpato et al., 2010). Concentrations are also important along the Italian coast, in Napoli (28.0 mg / kg and 19.0 mg / kg) and Bagnoli (16.0 and 12.0 mg / kg), in Sardinia at La Maddalena (PCB 153: 26.0 mg / kg, PCB 138: 12.0 mg / kg), at the Llobregat mouth (18.1 and 14.4 mg / kg) and in Barcelona (11.0 and 8.2 mg / kg). In southern Mediterranean, significant values for PCB 153 and 138 (20.5 and 14.1 mg / kg) were demonstrated in Algiers.

Levels up to 400 mg / g fresh weight of DDT and 1400 mg / g wet weight of PCBs were found in the fat of marine mammals (dolphins), far greater than the equivalent data in the Atlantic. Some geographical areas are in situations of concern. These include estuaries (Rhône, Ebro), bays and gulfs (Fos Sea, Bay of Algiers and Tunis, Genoa, Naples, Algeciras) and areas affected by the landfill.

From the UNEP/MAP - MED POL database, it was shown concentrations of aldrin, dieldrin, endrin, hexachlorobenzene, lindane, as measured in wild *Mytilus galloprovincialis*, are low in the northwest Basin. Concentrations of DDT are higher, especially the degradation products as p, p'-DDE.

An analysis of dioxins in mussels *Mytilus galloprovincialis* in 33 stations from the whole occidental basin (Andral et al., 2004) shows highest values were recorded in Marseille (2.66 ng/kg) with significant inputs. On the basin scale, the distribution of dioxins was similar to the one of PCBs, with highest values at Barcelona, la Maddalena (Sardinia), Algiers and Napoli but also in Toulon (Munschy et al, 2008, Andral et al 2008, 2011) when Corsica and Northern Africa were the areas with lower concentrations.

In the North of the basin, total DDT concentrations in mussels *Mytillus galloprovincialis*, decreased. More recently, an analysis of concentrations of DDT in the fatty parts of bottlenose dolphins conducted between 1978 and 2002 different points on the coast of western basin showed that concentrations were divided by 23.7 during this period (Borrell and Aguilar, 2007).

According to UNEP/MAP-MED POL data, (UNEP/MAP - MED POL 2009a) the Central Mediterranean and Ionian Sea were relatively free of hotspots of chlorinated hydrocarbons in marine bivalves, at least according to the present availability of data. Lower median levels were also estimated for total DDTs, and for lindane in the bivalve Mytilus, when compared to median levels in other Mediterranean sub-regions.

Concentrations of aldrin, dieldrin, endrin, lindane and hexachlorobenzene in *Mytilus galloprovincialis* are in the low ng g^{-1} range, with the exception of some stations from Albania. Concentrations of DDTs were one order of magnitude higher, with p,p’-DDE being, in general, the predominant component, although recent inputs of DDT in some

areas cannot be ruled out. Concentrations up to 9,779 ng g^{-1} dw of total DDTs were found in mussels from the Albania coast, probably indicating the presence of stockpiles of DDT in the country, as well as of lindane.

While PCB levels in *Merluccius merluccius* from the Adriatic Sea have slightly decreased between 1993 and 2003 from 1,380 ng/g to 943 ng/g lipid weight. The remaining levels are still high and the declining trend was not statistically significant. Such values are in accordance to the PCB concentrations in other fish species from the same region (UNEP/MAP - MED POL, 2009).

The analysis covered the full period of MED POL Phase III and Phase IV, until 2010, and incorporated additional scientific information included in the four sub-regional assessment reports, which were prepared by MAP in the framework of the gradual application of the Ecosystem Approach for the management of human activities in the Mediterranean.(UNEP/MAP 2010a,b,c,d).

The UNEP/MAP 2011 report includes the families of DDTs and PCBs, as the more representative persistent organic pollutants (POPs). The Drin's (aldrin, endrin and dieldrin), HCB and lindane were also considered. In summary, after selection and harmonization of data, a total of 33,742 observations, corresponding to more than 400 stations monitored during the MED POL Phases III and IV (1999-2010) have been included in the assessment.

Chlorinated pesticides have been extensively analyzed in Mediterranean biota since the inception of MED POL (UNEP 1990). Mussels and mullets have been the most widely studied organisms in the whole basin as part of many case studies published in the literature and recently assessed on the occasion of the implementation of the Stockholm Convention (UNEP 2002).

Concentrations of aldrin, dieldrin, endrin, lindane and hexachlorobenzene in *Mytilus galloprovincialis* are in the low ng g^{-1} range. Concentrations of DDTs were one order of magnitude higher, with p,p'-DDE being, in general, the predominant component, although recent inputs of DDT in some areas cannot be ruled out.

Concentrations are higher in the Aegean-Levantine region for aldrin and dieldrin, and in the Western Mediterranean for HCB and lindane. However, the highest values of HCB and lindane are found in Turkey and Albania, with a number of important hot spots. In the case of DDTs, their median values are similar for the four eco-regions, around 10ng g^{-1} dw, but again very high values are found in the Adriatic, corresponding to Albania. Despite the similarity of the mean DDT levels for the four eco-regions, available data indicate that contaminants are not uniformly distributed throughout the sub-regions.

PCBs occur in the vicinity of industrial and urban sites, as well as in major river mouths. The geographical distribution of concentrations (7 ICES PCB congeners) in the indicator organism *Mytilus galloprovincialis* is shown in UNEP/MAP 2011. The median values show the higher levels in the Adriatic where the Albania samples are well above the

average, with values up to 1500 ng g^{-1} dw in one station of France.

Taking into account the limited data available, trends are just preliminarily assessed. The analysis has been mainly focused on *Mytilus galloprovincialis* and *Mullus barbatus*, with the exception of Israel, where the clam *Mactra corallina* has been considered. Moreover, a few available time series for sediments from Israel (Haifa Bay) and France (Gulf of Lions) have also been considered.

In the Western Mediterranean, the stations evidencing relatively high levels of PCBs are still exhibiting increasing trends whereas those more pristine seem to be stable or decrease.

In general, it was found that during the period 1979-1998 the decreasing trends were in the order: ΣDDT > HCHs > PCBs, which may reflect that the regulation of the use of these chemicals and, consequently, of the contaminant inputs to the sea was more efficient for DDT and lindane than for PCBs.

During the last decade, HCHs (α-, β-andγ-isomers) were found in significant amounts (1–30 ng L–1) in the marine wetlands of Amvrakikos and Thermaikos Gulfs in Greece and in the coastal waters of Alexandria. Along the Spanish coast the levels were of 1.3–2 ng l^{-1}. Very recently, concentrations of 30–1500 pg l^{-1} of lindane and 5–427 pg l^{-1} of HCB were found in NW Mediterranean coastal waters, off Barcelona, basically associated with the dissolved phase. In the same stations, the concentrations of particulate and dissolved PCBs in the subsurface waters averaged 0.57 and 1.2 ng l^{-1} (ΣICES-7 congeners), respectively, in the range of those reported 15 years ago (Albaiges 2005).

HCB has been widely distributed in Western Mediterranean fish species, particularly in coastal species. Measurable amounts of HCB were found in red mullet collected in the NW basin, with levels on the Spanish coast (0.24–2.80 ng g^{-1} ww) slightly higher than on the French one (0.42–0.97 ng g^{-1} ww) (Albaiges 2005).

Two mussel surveys were conducted along the Mediterranean coast of France and Italy, between Sete and Genova, in 1973/1974 and in 1988/1989 in order to assess the temporal contamination trend by organochlorine pesticides. The comparison of concentrations measured in both surveys showed that DDTs and PCBs decreased by a factor of approximately 5 in 15 years, from730 to 130 ng g^{-1} dw and 2430 to 527 ng g^{-1} dw Aroclor equivalents, respectively. This decrease is in agreement with the ban of DDT implemented in 1975 in western Europe and the gradual cessation of PCBs production in the 1970s and 1980s. A later survey along the French coast, in 1995–1999, gave a mean concentration of 17 ng g–1 dw (1.2–157 ng g^{-1}), pointing to a sustained reduction of levels. A similar trend was observed in mussels collected close to the Ebro River mouth, where a decrease by a factor of 3 in DDTs, with a concurrent increase of the DDE/DDT ratio was observed from1980 to 1990. The same survey revealed a decrease of HCB concentrations by a factor of 5 (from 1–2 to 0.2–0.3 ng g^{-1}). (Albaiges 2005).

As far as the monitoring of POPs is concerned, the RNO is composed of 23 shellfish

sampling sites, visited four times a year. Contaminants measured are DDT, DDD, DDE, lindane (γ-HCH), α-HCH, PCBs (Congeners 28, 52, 101, 105, 118, 138, 153, 180) and PAHs.

Atmospheric circulation and deposition estimates of POPs in the Mediterranean region are scarce. First calculations were reported by GESAMP, based on very few data, but showing that 80–95% of total inputs of POPs (e.g. HCB, HCH, DDT, chlordane and PCBs) were atmospheric.

The extremely informative paper by (Gómez-Gutiérrez et al., 2007) presents a compilation of measurements in sediments and has also concluded that a decreasing trend is more evident for DDTs than for PCBs, indicating a steady input of the latter in the Mediterranean Sea and the need for an improved management of their potential sources.

A detailed analysis for the 4 subregions: Western Mediterranean , Ionian Sea and and Central Mediterranean subregion, Adriatic Sea, Eastern Mediterranean subregion can be found in UNEP/MAP 2012.

Toxicological studies have found that PCB levels in deepwater fishes (*Alepocephalus rostratus*, *Bathypterois mediterraneus*, *Coryphaenoides guentheri* and *Lepidion lepidion*) were lower than in coastal fishes, close to the pollution sources, but much higher than that of fish living on the continental shelf to upper slope (*Micromesistius poutassou*, *Phycis blennoides* and *Lepidorhombus boscii*). PCB levels recorded were within the same range as that of top predators like tuna (Solé et al., 2001).

Available data indicate that organic contaminants are not uniformly distributed throughout the Western Mediterranean. For example, concentrations of total DDT in sediments range from <0.25 to 885 ng / g and PCBs from 1.3 to 7274 ng / g, higher levels corresponding to the "hot spots" areas near outfalls waste water of major cities and at the mouths of large rivers (e.g. Rhône).

The PCB congeners 31, 52, 156 and 180 are present at low concentrations and in industrial areas or urban. The PCB 153 and 138 show maximum levels of accumulation along the French coast, particularly at Marseilles and his emissary (respectively 42.3 mg / kg and 27.6 mg / kg) and to a lesser extent al mouth of the Rhone basin of the Mediterranean (after Scarpato et al., 2010). Concentrations are also important along the Italian coast, in Napoli (28.0 mg / kg and 19.0 mg / kg) and Bagnoli (16.0 and 12.0 mg / kg), in Sardinia at La Maddalena (PCB 153: 26.0 mg / kg, PCB 138: 12.0 mg / kg), at the Llobregat mouth (18.1 and 14.4 mg / kg) and in Barcelona (11.0 and 8.2 mg / kg). In southern Mediterranean, significant values for PCB 153 and 138 (20.5 and 14.1 mg / kg) were demonstrated in Algiers.

Levels up to 400 mg / g fresh weight of DDT and 1400 mg / g wet weight of PCBs were found in the fat of marine mammals (dolphins), far greater than the equivalent data in the Atlantic. Some geographical areas are in situations of concern. These include estuaries (Rhône, Ebro), bays and gulfs (Fos Sea, Bay of Algiers and Tunis, Genoa, Naples,

Algeciras) and areas affected by the landfill.

From the UNEP/MAP - MED POL database, it was shown concentrations of aldrin, dieldrin, endrin, hexachlorobenzene, lindane, as measured in wild *Mytilus galloprovincialis*, are low in the northwest Basin. Concentrations of DDT are higher, especially the degradation products as p, p'-DDE.

An analysis of dioxins in mussels *Mytilus galloprovincialis* in 33 stations from the whole occidental basin (Andral et al., 2004) shows highest values were recorded in Marseille (2.66 ng/kg) with significant inputs. On the basin scale, the distribution of dioxins was similar to the one of PCBs, with highest values at Barcelona, la Maddalena (Sardinia), Algiers and Napoli but also in Toulon (Munschy et al, 2008, Andral et al 2008, 2011) when Corsica and Northern Africa were the areas with lower concentrations.

In the North of the basin, total DDT concentrations in mussels *Mytillus galloprovincialis*, decreased. More recently, an analysis of concentrations of DDT in the fatty parts of bottlenose dolphins conducted between 1978 and 2002 different points on the coast of western basin showed that concentrations were divided by 23.7 during this period (Borrell and Aguilar, 2007).

According to UNEP/MAP-MED POL data, (UNEP/MAP - MED POL 2009a) the Central Mediterranean and Ionian Sea were relatively free of hotspots of chlorinated hydrocarbons in marine bivalves, at least according to the present availability of data. Lower median levels were also estimated for total DDTs, and for lindane in the bivalve Mytilus, when compared to median levels in other Mediterranean sub-regions.

Concentrations of aldrin, dieldrin, endrin, lindane and hexachlorobenzene in *Mytilus galloprovincialis* are in the low ng g^{-1} range, with the exception of some stations from Albania. Concentrations of DDTs were one order of magnitude higher, with p,p’-DDE being, in general, the predominant component, although recent inputs of DDT in some areas cannot be ruled out. Concentrations up to 9,779 ng g^{-1} dw of total DDTs were found in mussels from the Albania coast, probably indicating the presence of stockpiles of DDT in the country, as well as of lindane.

While PCB levels in *Merluccius merluccius* from the Adriatic Sea have slightly decreased between 1993 and 2003 from 1,380 ng/g to 943 ng/g lipid weight. The remaining levels are still high and the declining trend was not statistically significant. Such values are in accordance to the PCB concentrations in other fish species from the same region (UNEP/MAP - MED POL, 2009).

4 Great Lakes

State of the Great Lakes 2011: Environment Canada and the U.S. Environemntal Protection Agency. 2014. State of the Great Lakes 2011. Cat No. En161-3/1-2011E-PDF. EPA 950-R-13-002. Available at http://binational.net

Contaminants in birds: The long term trends (1974 to present) of virtually all legacy contaminants are declining. The short term trends, those over the last decade, are a mixture of some showing significant declines but others showing no significant change. (p195)

Lake Superior Status: Good Trend: Improving. Rationale: The traditional legacy contaminants, DDE, SUM PCBs and TCDD, have declined significantly in long term (1974-2009) and short term (2000-2009). SUM BDE has not declined significantly in the short term.

Lake Michigan Status: Good Trend: Improving. Rationale: The traditional legacy contaminants, DDE, SUM PCBs and TCDD, have declined significantly both since the 1970s (1974-2009) and in the last decade (2000-2009). SUM BDE has not declined significantly in the short term.

Lake Huron Status: Good Trend: Improving. Rationale: The traditional legacy contaminants, DDE, SUM PCBs and TCDD and Hg, have declined significantly both since the 1970s (1974-2009) and in the last decade (2000-2009). No significant change for SUM BDE in the short term.

Lake Erie Status: Fair Trend: Unchanging. Rationale: The legacy contaminants, DDE, SUM PCBs, TCDD and Hg, have all declined significantly since the 1970s (1974-2009). However, none of them, as well as SUM BDEs has declined significantly in the last decade (2000-2009).

Lake Ontario Status: Fair Trend: Unchanging. Rationale: The legacy contaminants, DDE, SUM PCBs, TCDD and Hg, have all declined significantly since the 1970s (1974-2009). However, none of them, as well as SUM BDEs has declined significantly in the last decade (2000-2009).

Although there are Great Lakes wildlife species that are more sensitive to contaminants than Herring Gulls, and colonial nesting waterbird species in general, there is no other species which has the historical dataset that the Herring Gull does. As contaminant levels continue to decline (if they do), the usefulness of the Herring Gull as a biological indicator species may lessen (due to its reduced sensitivity to low levels of contamination) but its value as a chemical indicator will remain and probably increase - as levels become harder and harder to measure in other media. It is an excellent accumulation tracker since many of the above biological measures are correlated with contaminant levels in their eggs. In other colonial waterbirds, there are similar correlations between contaminant levels in eggs and various biological measures. Contaminant levels in eggs of other colonial waterbirds are usually correlated with those in Herring Gulls. (p197)

Historical data on levels of chemical contamination in gull eggs are available, on an annual basis, for most sites in both the Canadian and U.S. Great Lakes dating back to the early 1970s. An immense database of chemical levels and biological measures from the

Great Lakes, as well as many off-Lakes sites, is available from the Ecotoxicology and Wildlife Health Division at Environment Canada. Data on temporal trends, portrayed as annual contaminant levels over time, for 1974-present in most instances, are available for each site and each compound. For example, DDE, from 1974-2008, is available for Toronto Harbour . Geographical patterns in contaminant levels, showing all sites relative to one another, are also available for most years from 1974-present and for most compounds. For example, PCBs, 2008, at 15 Great Lakes sites from Lake Superior to the St. Lawrence River (including U.S. Sites). (p197)

The size and distribution of the waterbird populations which breed on the Great Lakes is also an indicator of ecosystem health. Declining waterbird populations (number of breeding pairs or nests) and vital rates (hatching success, fledging success, mortality rates, etc.) can be indicators of local environmental stress. The Great Lakes- wide population of colonial waterbirds has been censused jointly, by the Canadian Wildlife Service and the U.S. Fish and Wildlife Service since the 1970s, approximately every 10 years; four “decadal” censuses have been conducted to date: in the 1970s, 1980s, 1990s and 2000s. Briefly, and in the long-term (from the 1970s to the 2000s), these censuses have shown that the breeding numbers of six species have increased: Double-crested Cormorants, Black-crowned Night-Herons, Great Egrets, Ring-billed Gulls, Great Black-backed Gulls, and Caspian Terns. Unfortunately, the numbers of three species, Great Blue Heron, Herring Gull and Common Tern, have declined. In the short-term (from the 1990s to 2000s), numbers of night-herons, the three gull species and Common Terns have declined. For Common Terns, which have declined continuously since the first census, the trend is alarming; numbers have declined from approximately 8,600 pairs to just 5,000 pairs (42%; Figure 3). The reasons for this decline are unclear but it is partially due to competition for nest sites with Ring-billed Gulls and habitat loss. Although the Herring Gull population is much more numerous (approximately 32,000 pairs), their decline should be monitored, especially in Lake Huron, where numbers have declined from approximately 33,500 pairs in the 1970s to 22,000 pairs in the 2000s (34%).

Long-term (greater than 25 years), basin-wide monitoring programs that measure whole body concentrations of contaminants in top predator fish (Lake Trout and/or Walleye) are conducted by both the U.S. Environmental Protection Agency (U.S. EPA) Great Lakes National Program Office through the Great Lakes Fish Monitoring and Surveillance Program, and Environment Canada’s (EC) Water Quality Monitoring Surveillance Division, through the Fish Contaminants Monitoring and Surveillance Program, to identify the risk of contaminants to wildlife consumers of fish and to monitor trends in time. (p204)

More information on the monitoring programs can be found at the following websites: http://www.epa.gov/glnpo/monitoring/fish/index.html and http://www.ec.gc.ca/scitech/default.asp?lang=en&n=828EB4D2-1

Contaminants in Whole Fish: Overall Assessment Status: Fair, Trend: Deteriorating Rationale: The assessment incorporates multiple contaminants and considers potential

effects of exposure to fish eating wildlife. Concentrations of PCBs and pentaPBDEs are currently above guidelines in Lake Trout and Walleye in all the Great Lakes; however concentrations of these contaminants are declining in most monitored fish.(p203)

Total polychlorinated biphenyls (PCBs) Basin Wide Status: Fair; Improving Total PCB concentrations in Great Lakes top predator fish have continuously declined since their phase-out in the 1970s. Median PCB concentrations in Lake Trout in Lakes Superior, Huron, and Ontario and Walleye in Lake Erie continue to decline; however, they are still above the target of 0.1 µg/g ww in the GLWQA.

Log-linear regression of Environment Canada data show the continued long-term annual declines of 5% in Lake Trout from Lake Superior and 7% in Lakes Huron and Ontario while PCBs in Lake Erie Walleye are declining by 3% per year. Similar analyses of U.S. EPA data show no significant annual declines of total PCB in Lake Trout from Lake Superior and 4%, 6%, 7%, and 4% annual declines in total PCB in Lake Trout from Lakes Huron, Michigan, Ontario, and Lake Erie Walleye, respectively. Data collected since the last SOLEC indicator report (2006-2009), show that total PCB concentrations in composited Rainbow Smelt measured by Environment Canada were all less than 0.1 µg/g ww in Lakes Superior and Huron.

In Lake Ontario, total PCB concentrations in Rainbow Smelt are declining by ~8% per year since monitoring began in 1977. Recent studies have suggested that rates of decline of PCB residues in fish are slowing or have stopped in some lakes in recent years.

Dichlorodiphenyltrichloroethane (DDT) and metabolites Basin Wide Status: Good; Improving the concentration of opDDT and its metabolites, opDDD and opDDE, (sumDDT) in Great Lakes top predator fish have continuously declined since the use of the chemical was banned in 1972. Concentrations measured since the last indicator report (2006-2009) remain well below the GLWQA target of 1.0 µg/g ww across the basin. Based on data collected at EC monitoring locations, annual rates of decline are 6.8% in L. Superior, 7.1% in L. Huron, 7.5% in L. Erie, and 7.3% in L. Ontario. Since the last indicator report, the rates of decline appear to be consistent with historical trends. Annual rates of decline determined using U.S. EPA data are slightly lower at 4.5% in L. Superior, 5.9% in L. Michigan, 5.9% in L. Huron, 6.0% in L. Erie, and 6.7% in L. Ontario. Rates of decline at the U.S. monitoring stations in the years since the last indicator report appear to be increasing (i.e. declining faster) in lakes Michigan, Huron, and Ontario compared to historical trends while rates remain consistent with historical trends in Lakes Superior and Erie. (p206)

Σα- & γ-Chlordane: Basin Wide Status: Good; Unchanging Concentrations of α- + γ-chlordane in whole Lake Trout and Walleye have consistently declined since the chemical was banned by the U.S. EPA in 1988. In recent years, the concentrations in fish appear to have reached a steady state with no significant increases or decreases. The highest observed median concentrations since the last indicator report (2006-2009) are in Lake Trout from Lake Michigan (0.018 µg/g ww), followed by Lake Ontario (0.012 µg/g ww). Median concentration in Lakes Superior, Huron, and Erie are all below 0.01 µg/g ww.

There is no target for chlordane in whole fish in the GLWQA. A report on the levels of chlordane in fish will not appear in future SOLEC indicator reports as focus is shifted to contaminants with established environmental quality guidelines or targets. (p206)

Mirex Basin Wide Status: Good; Improving Mirex is regularly detected only in fish from Lake Ontario due to historical releases in the Niagara River and other locations within the lake's watershed. Since the last indicator report (2006-09), median concentrations in Lake Trout were 0.061 µg/g ww (EC) and 0.041 µg/g ww (U.S. EPA). Declines in the concentration of mirex in Lake Trout from Lake Ontario are still declining at historical rates of between 4 and 12 % annually. According to the guidelines listed in the GLWQA, Mirex should be "substantially absent" from Great Lakes fish. (p207)

Dieldrin Basin Wide Status: Good; Improving The highest concentrations of dieldrin (and related compounds endrin and andrin) in top predator fish are observed in Lake Michigan (median = 0.034 µg/g ww) and Lake Ontario (median = 0.021 ug/g ww). Concentrations have declined substantially since monitoring began in the lakes and are still declining basin wide at rates ranging from 2 to 18% annually. There is no guideline for dieldrin in whole fish in the GLWQA. This will be the last report on the levels of dieldrin and related compounds SOLEC as focus is shifted to contaminants with established environmental quality guidelines or targets. (p207)

Toxaphene Basin Wide Status: Fair; Improving Decreases in toxaphene concentrations have been observed throughout the Great Lakes in all media following its ban in the mid-1980s. A recent study on toxaphene trends in Great Lakes fish show that concentrations remain the highest in Lake Superior (up to ~480 ng/g) and lowest in Lake Erie (up to ~50 ng/g) (Xia et al. 2012). Concentrations of toxaphene in Lake Trout and Walleye continue to exhibit exponential temporal declines in all of the Great Lakes; however, concentrations appear to level off starting in 2007 (Xia et al. 2012). Continued monitoring of toxaphene in top predator fish in the coming years should confirm whether toxaphene concentrations have reached a steady state in Great Lakes fish. (p207)

Polybrominated Diphenyl Ethers (PBDEs) Basin Wide Status: Fair; Improving The production and use of three popular commercial formulations of PBDE have or are being voluntarily phased out by industry in North America. The phase out of the more toxic penta- and octa-BDE compounds started in 2004 and by 2012, the use of deca-BDE will likely be reduced as a result of the voluntary withdrawal by industry (http:/www.bsef.com). In a national survey of PBDE concentrations in top predator fish from lakes across Canada, the highest concentrations were observed in fish from the Great Lakes and >95% of the PBDE compounds in the fish were tetra-, penta-, or hexa-BDEs (Gewurtz et al. 2011). Federal Environmental Quality Guidelines (FEQG) have been developed by Environment Canada for these three homologue groups which are meant to provide targets for acceptable environmental quality, assess the significance of observed concentrations, and to measure the success of risk management activities. The FEQGs to protect wildlife consumers of fish for tetra-, penta- and hexa-BDEs are 88, 1.0, and 420 ng/g ww respectively (Environment Canada 2010). Routine monitoring of PBDEs in whole top predator fish from the Great Lakes combined with retrospective

analyses of archived samples by the U.S. EPA and Environment Canada have provided a complete picture of PBDE contamination in Great Lakes fish from 1977 to the present day. Concentrations of PBDEs in Lake Trout and Walleye rose continuously through to the early 2000s then began to decline. Log-linear regression of PBDE concentrations in Lake Trout and Walleye (U.S. EPA; Lake Erie), show significant declining trends of 5.8%/year for tetra-BDEs, 6.4% for penta-BDEs, and 3.4% for hexa-BDEs in Lake Ontario and annual declines of 19% for tetra- BDEs and 17% for penta-BDEs from Lake Michigan. PBDE concentrations in Lakes Superior, Huron, and Erie also appear to be declining as the slopes of the regressions are all negative; however, the slopes are not significantly different from zero at $\alpha = 0.05$ with a power of 80%. The majority of tetra-BDE and all hexa-BDE concentrations reported for Lake Trout and Walleye in 2009 from all the Great Lakes are below Environment Canada's FEQGs; however, all measured penta-BDE concentrations are well above the FEQG of 1.0 ng/g ww "

Routine monitoring of PFOS in whole Lake Trout from the Great Lakes combined with retrospective analyses of archived samples from EC's National Aquatic Biological Specimen Bank have provided information on PFOS contamination in Lake Ontario Great Lakes fish from 1979 to 2008 (Figure 7). Concentrations of PFOS in Lake Trout rose continuously at a rate of 5.9%/year through to the late 1980s/early 1990s, after which no consistent change in time was observed. This contradicts trends observed in ringed seals in the Canadian Arctic, where significant PFOS declines were observed within the year following voluntary phase-outs (Butt et al. 2007). This contradiction may be due to continued inputs into Lake Ontario from the continued use of these substances. Perfluorooctanoic acid (PFOA) is another common fluorochemical and major manufacturers have voluntarily agreed to a 99% phase-out by 2015. However, PFOA is not highly bioaccumulative and time trends were not reliably measured in fish. Conversely, the concentration of two other fluorochemicals, perfluorodecane sulfonate (PFDS) and Perfluorooctane sulfonamide (PFOSA), have declined consistently in Lake Trout from Lake Ontario since 1992 at rates of 4.4% and 6.2% per year, respectively.(p207)

Contamination in Sediment Cores Overall Assessment Status: Fair Trend: Improving Rationale: Concentrations of legacy contaminants including PCBs and DDT are generally below guidelines in the Great Lakes and declining. Other contaminants such as the polybrominated diphenyl ethers (PBDEs) exhibit some exceedances of guidelines, particularly penta-BDE in Lake Ontario; however, temporal trends show recent declines as a result of management actions.(p220)

Bottom sediment contaminant surveys conducted in the Great Lakes from 1968 – 1974, from 1997 – 2002 and more recent surveys provide information on the spatial distribution of contaminants, the impacts of local historical sources and, in concert with sediment cores, the response to management initiatives. Contaminants across several chemical classes are measured in both surface sediment and sediment cores. The measured contaminants with the highest occurrences, causes of degradation of sediment quality and fish consumption restrictions are: Mercury • PCBs • Dioxins • HCB • Total DDT • Lead •

PAHs • Dioxins and Furans •(p221)

Comparisons of surficial sediment contaminant concentrations with sub-surface maximum concentrations indicate that contaminant concentrations have generally decreased by more than 35 per cent, and, in some cases, by as much as 80 per cent over the past four decades (p222).

The presence of new persistent toxic substances represents a potential threat to the health of the Great Lakes ecosystem. These compounds include perfluoroalklated compounds (PFCs) and brominated flame retardants (BFRs), the latter of which are heavily used globally in the manufacturing of a wide range of consumer products and building materials. The BFRs have been found to be bioaccumulating in Great Lakes fish and in breast milk of North American women. While end of the pipe discharges may not be responsible for ongoing contamination, modern urban/industrial centres can act as diffuse sources of current inputs. Sediment core profiles of brominated diphenyl ethers (BDEs) and PFCs in Lake Ontario suggest that accumulation of these chemicals has recently peaked, or continues to increase. The Lake Ontario BDE profile indicates a leveling off of accumulation in the past decade, presumably as a result of voluntary cessation of production of these compounds in North America. However, the deca-substituted BDE 209 is the predominant congener in sediment, and is still currently used. Despite these trends, maximum concentrations of many BFRs and PFCs remain well below maximum concentrations of contaminants such as DDT and PCBs observed in past decades.(p222)

Production of PFCs as stain repellents in carpets historically exceeded $1 billion annually. Two classes of PFCs, the perfluoroalkyl sulfonate acids (PFSAs), particularly perfluorooctane sulfonate (PFOS), and the perfluorocarboxylates, particulary perfluorooctanoic acid (PFOA), are the most commonly measured PFCs; these compounds are highly stable and persistent in the environment, and are potentially toxic.

Concentrations of PFCs in sediments of Great Lakes tributaries are highest in urbanized and/or industrialized watersheds. In general levels of perfluoroalkyl sulfonate acids and PFOS in tributaries and open waters of the Great Lakes are slightly higher than the perfluorocarboxylates with the highest levels of PFCs generally found in areas of Lake Ontario and the western end of Lake Erie and the Detroit River corridor. There is a gradient toward increasing PFC contamination from the upper Great Lakes (Superior and Huron) to the lower Great Lakes (Erie and Ontario) for both tributary and open-lake sediments (Figures 3 and 4). Concentrations of PFCs in open-lake sediments are driven not only by proximity to sources, but physical processes and bathymetry as well. The highest PFC concentrations in open-lake sediments were found in Lake Ontario. The spatial distributions of PFCs in Lake Ontario are fairly consistent across the lake, which is primarily due to lake currents that evenly distribute suspended particles and across the three major depositional basins.(p223).

Fish Consumption Restrictions Advisory Rating Scale
Overall Assessment Status: Fair Trend: Undetermined

The Fish Consumption Advisory Rating Scale Indicator was created to categorize the different levels of risk to sensitive populations (children under 15 and women of child bearing age) from consuming certain fish species in each of the Great Lakes. The Indicator involves a five-level, Consumption Advisory Rating Scale that corresponds to the current contaminant levels in Great Lakes fish. Protective measures associated with each consumption advisory rating scale allows a flexible, graduated and appropriate response to the level of risk from consumption. The information used to conduct this analysis demonstrates that there are consumption advisories in all of the Great Lakes for a variety of species of fish that are driven by PCBs, mercury, dioxin, chlordane, mirex and toxaphene.

On the methods used for sampling water:

"Background Water quality samples for the analysis of toxics have been collected from the Great Lakes since the mid 1980s as part of Environment Canada's Great Lakes Surveillance Program. Ship-based monitoring cruises are conducted to measure water quality in each of the lakes upon which Canada borders. Measuring organic contaminants in water is challenging, and it requires special equipment, techniques and knowledge. In the first years of monitoring for organic contaminants, whole water samples were collected. Special studies, conducted between 1992 and 1995, recommended collecting surface, dissolved phase samples during the spring only (Williams et al., 2001). With the exception of some in-use pesticides, maximum concentrations were observed during the spring, and therefore represent the worst-case situation and can be used to determine compliance with water quality objectives. Prior to 2004, samples for organic contaminants were centrifuged to separate the dissolved and particulate fractions, and the dissolved fraction was prepared for analysis immediately after collection, on board the ship, using a Goulden large volume extractor (Goulden and Anthony, 1985). Extracts were stored and returned to Environment Canada laboratory facilities in Burlington, Ontario, for analysis using gas chromatography/mass spectrometry. Since 2004, we have improved the technique and the 16 – 24 L samples are now stabilized in the field, and brought back to a specially constructed clean laboratory at Environment Canada for extraction. There appears to be less interference from extraneous contamination (presumably from ship-derived pollutants). Improvements in laboratory methods have resulted in much better (i.e., lower) detection limits for many compounds including PAHs and some organochlorines. For some parameters, the improvements mean that we have greater confidence in the more recent data compared to those obtained before 2004, but this also means that longer-term trends are difficult to determine. For example, detection limits for many polycyclic aromatic hydrocarbons (PAHs) have greatly improved. Measurable concentrations of some PAHs are now reported in Great Lakes waters for the first time; this does not necessarily mean that they were previously absent, but rather our ability to detect them has improved. The Canadian Council of Ministers of the Environment (CCME, 1999) has withdrawn the water quality guidelines for several of the organochlorine compounds (aldrin, chlordane, dieldrin, endrin, heptachlor and PCBs) and

a water quality guideline is no longer recommended. Exposure to these compounds for aquatic organisms is primarily via sediment, soil and/or tissue, therefore assessment of environmental quality relative to sediment and fish tissue guidelines is instead recommended. Indeed, these compounds are relatively hydrophobic and are difficult to measure in surface waters. Because of those difficulties, and because of the short time period of higher quality data that is available for assessing trends, it may be more useful to assess longer term trends using sediments or fish as environmental quality indicators for these compounds." (p426)

Levels in water

Lake Superior Concentrations of most organochlorine compounds are below detection limits or declining, although data are insufficient in most cases to quantify the rate of decline. Concentrations of a few organochlorines appear to be unchanging, such as HCB, heptachlor epoxide and dieldrin, although the latter shows some indication of a more recent decline (2005-2008). Increases are observed for the in-use pesticides atrazine and possibly metolachlor. The overall temporal trend for toxics is therefore mixed. Concentrations of most organic compounds are lowest in Lake Superior. This is likely because historic sources of most compounds were predominantly located in more industrial and agricultural regions. However, several compounds that are more susceptible to atmospheric transport and deposition are found at higher concentrations in the upper Great Lakes compared with the lower lakes. Compounds that are found at higher concentrations in Lake Superior include a-HCH, lindane, g-chlordane, a-endosulfan, endrin, and b-endosulfan (b-endosulfan was only found in trace quantities in Lake Superior). An example of the spatial distribution of one of these compounds, a-HCH, is shown using the most recent quality-assured data in Figure 1. No exceedences of Canadian federal water quality guidelines are observed for any parameter in Lake Superior.

Data are also available from the USEPA for Lake Michigan from 1994 to 1997 and from the mid-2000s, and these are used for comparison purposes. Samples were collected from six stations in Lake Michigan in 2006. Similar to Lakes Superior and Huron, concentrations of most compounds were low. However, certain compounds showed higher concentrations compared to the other Great Lakes, including dieldrin, heptachlor epoxide and a-chlordane. Although the Canadian water quality guidelines are not applicable to United States' waters, comparison with the benchmark CCME water quality guideline indicated no exceedences. Within Lake Michigan, higher values of certain compounds (some PAHs, g- chlordane, a-endosulfan) were found at sites in the southern basin compared to more offshore locations.(p427)

Lake Huron With inflows from both Lake Superior and Lake Michigan, the water quality of Lake Huron tends to reflect these other two Great Lakes. North Channel waters tend to reflect the outflow from Lake Superior, with very low values of many compounds (such as PAHs and organochlorines such as dieldrin), but higher concentrations of compounds that are deposited from atmospheric sources in Lake Superior, such as a-

HCH. The waters of Georgian Bay are similar to the main body of Lake Huron with respect to toxic chemicals (i.e., low concentrations). Slightly higher concentrations of some parameters (for example, HCB) have been observed in and near Saginaw Bay and the inflow from Lake Michigan, compared to the remainder of the lake.

The overall status for most toxic compounds is better in Lake Huron compared to the other Great Lakes. Temporal trends indicate little change over time. The ecosystem objective has not been achieved in Lake Huron because toxics are still measurable and because temporal trends are not demonstrating significant declines."(p428)

Lake Ontario. Many compounds, particularly those resulting from historical use in industry and agriculture, are found at highest levels in the lower Great Lakes (Ontario and Erie). These compounds include hexachlorobenzene (HCB), lindane, dieldrin, DDT and its metabolites and some PAHs. The spatial distribution of HCB is shown in Figure 2. Higher values of total PCBs are observed in Lake Ontario and along the southern shore and western basin of Lake Erie compared to the upper Great Lakes. The monitored current-use pesticides (atrazine and metolachlor) are observed in higher concentrations in Lake Ontario. However, no CCME water quality exceedences are observed."(p429)

Detection limit has improved from 0.8 ng/L to 0.044 ng/L. Field and laboratory blanks have improved as well, but background, extraneous PCB contamination remains problematic. Total PCBs are detected in all Great Lakes waters, but concentrations are significantly higher in sample water than in field blanks only in Lake Ontario and in the western basin of Lake Erie.

Temporal trends are difficult to discern because of improved detection limits and extraneous contamination as measured by laboratory and field blanks. The best record exists for Lake Ontario, where toxics were measured on five occasions between 2004 and 2010. The data indicate values in the offshore have been relatively constant over this time period (~190 pg /L). Studies conducted by the USEPA in spring 1993 indicated similar values (range 110 – 190 pg /L), indicating no change over the past 15 years. (p429)

Dieldrin is detected throughout the Great Lakes. Lakewide average concentrations are highest in Lake Michigan (184 pg/L) and lowest in Lake Huron and Georgian Bay (63 to 85 ng/L). Concentrations in most lakes are declining. In Lake Ontario, the rate is about 6.6 pg/L·yr ($p<0.001$), resulting in a half-fold time of approximately 16 years (starting from 1992). In Lake Erie the rate is about 8.9 pg/L·yr ($p=0.04$) and in Lake Superior the rate is about 3.3 pg/L·yr ($p=0.078$). In Lake Huron, dieldrin appears to be increasing at a rate of 5.9 pg/L·yr ($p=0.056$) but the data are relatively sparse and the trend in Lake Michigan is unknown.(p429)

Lindane (g-HCH) is detected in all of the Great Lakes. Concentrations are highest in Lake Superior and lowest in Lake Huron, Georgian Bay and Lake Michigan. The temporal trend shows that lindane is declining in all the lakes (no temporal information is available for Lake Michigan). The use of lindane in the US and Canada started to be restricted in the 1970s and in 2007 its major uses were banned entirely with the

exemption of its use for the treatment of head scabies and lice. The marked decline in the lakes reflects the success of usage restrictions. The high concentrations found in Lake Superior are likely due to atmospheric deposition and slower volatilization and breakdown at lower water temperatures.

For over 40 years, the Great Lakes Surveillance Program has monitored water quality in the Great Lakes, and since approximately 1986, toxic contaminants have comprised an important component of that program. Knowledge of the concentration of toxics dissolved in Great Lakes waters is important for comparison with other measurements in water (e.g., tributaries and precipitation), for the assessment of bioaccumulation and bioconcentration behaviours and rates, and for the calculation of water-atmosphere fluxes in order to assess atmospheric deposition and volatilization of contaminants. The long-range atmospheric transport of contaminants remains an important concern, particularly to more northern Great Lakes.(p431)

Concentrations of many substances are extremely low; in the part per quadrillion ($1 \times 10{-}15$) to part per trillion ($1 \times 10{-}12$) range. Routine monitoring for determining trends might be better accomplished, for some parameters, using sediment and fish samples. Contaminants in sediment can be used to indicate long-term changes in contaminant concentrations, as the settling of sediments represents a long- term sink for contaminants as they are gradually buried over time. Contaminants in fish are better indicative of the exposure of aquatic organisms to toxics in lake water and through their food chain. Because many of the legacy toxics are bioaccumulative and hydrophobic, higher concentrations can be measured in sediment and fish and these media are more appropriate for assessing ecosystem health. It remains important, however, to continue periodic monitoring of Great Lakes waters to verify concentrations and trends. Monitoring water concentrations is important for assessing compounds that are soluble in water such as certain in-use pesticides, selected legacy toxics as well as many of the compounds of emerging concern.(p431)

All water data from Great Lakes Surveillance Program, Water Quality Monitoring and Surveillance, Environment Canada, Burlington, Ontario. GLSP-PSGL@ec.gc.ca, Supplementary data for Lake Michigan from Great Lakes National Program Office, US EPA, Chicago Ill.(p432)

5 Antarctica

The ECA 2009 report includes the following information:

Studies to determine POPs in the aerosols during the last decade were carried out to define mechanisms and processes contributing to their presence in Antarctica and also to differentiate between local sources and long-range transport. Indeed there is evidence that aerosols are the main medium contributing to long-range transport in Antarctica. Comparable levels of PCBs content in seawater from north and south of the Antarctic Convergence were reported, indicating that the atmosphere, not the water, was the dominant pathway for the transport of PCB compounds to the Antarctic.

A study of temporal trends of OCP was carried out by measuring the content of HCB, heptachlor, α- and γ-HCH, heptachlor epoxide in air, seawater, sea ice, and snow samples from the Western Antarctic Peninsula. The results showed that HCB and HCH levels declined over the past 20 years, with a half-life of 3 years for ΣHCH in Antarctic air. However, heptachlor epoxide levels have not declined in Antarctic air over the past decade, possibly due to continued use of heptachlor in the southern hemisphere. Peak heptachlor concentrations in air were measured in coincidence with air masses moving into the region from lower latitudes. The ratio of α/γ-HCH in Antarctic air, sea ice and snow was <1, illustrative of a predominance of influx of lindane versus technical HCH to theregional environment. Water/air fugacity ratios for HCHs demonstrate continued atmospheric influx of HCHs to coastal Antarctic seas, particularly during late summer.

Few investigations have been carried out to determine the POP content of seawater. One recent investigation was carried out to study the vertical distribution of PCBs and PAHs in the coastal area of the Ross Sea during the Antarctic summer (from November to February). PCBs and PAHs showed a concentration range in the water column of 30–120 pg l^{-1} and 150–400 pg l^{-1}, respectively, and these values were strongly dependent on the suspended matter content. A nearly two-fold decrease in the pollutant concentration was also observed in the depth profile obtained in February, i.e. late summer, which might be correlated both with the increase of sedimentation due to the high content of suspended matter, and the reduction of the pollutant input.

Moreover, isomer ratios of PAHs, such as PHE/ANT and low molecular weight PAHs/high molecular weight PAHs (LMW/HMW) highlight that the main PAH source might be petrogenic in nature, whereas the pyrolytic source seems to be less important.

In consideration of the mechanism of cold condensation, the surface waters play an important role in the global distribution and in the long-range transport to cooler regions.

The sea surface micro-layer (SML) (0.1-0.001 mm) is the geographically widest environmental interface that can be accessed by sufficiently reproducible sampling methods. It is the site where many important processes occur, including the accumulation of pollutants and other chemical substances, atmospheric particles, and microorganisms. Most of the studies on POPs in the sea- surface micro-layer have been undertaken in coastal environments. Very few data are available from the open ocean, and there is a lack of data on the sea-surface micro-layer in remote areas, in particular on the presence of POPs in the Southern Ocean.

The surface water is normally described by a widely accepted conceptual model based on a multi-layer structure in which individual layers may have different properties and thickeness.

Sea surface micro-layer (SML) and sub-surface seawater (SSW) samples were gathered simultaneously. Sea surface micro-layer samples showed a total content of PCBs and PAHs in the range 400–450 pg l^{-1} and 2000–3000 pg l^{-1}, respectively, whereas the mean

contents in the sub-surface sea water samples were 48 pg l^{-1} and 325 pg l^{-1}, respectively. The mean enrichment factors of PCBs and PAHs in the sea-surface micro-layer were about 10 and 7, respectively.

A series of surveys in a large area of the Ross Sea and Victoria Land was performed in the period 1988-1992. The results showed a low and quite homogeneous distribution in surface water of PCBs with a mean concentration of 130 pg l^{-1}.

The concentration and the distribution of PCBs was also determined in marine sediments, and the mean content normalized for the relevant calculated specific surface area was 150 (pg g^{-1})/(m^2cm^{-3}).

More recently studies were carried out to determine emerging POPs, like polybominated dyphenil ethers (PBDEs), very commonly used as flame-retardants. Studies were carried out to assess the local sources of this class of POPs related to activity at bases. PBDE concentrations were determined in indoor dust and waste water sludge from the U.S. McMurdo and New Zealand- operated Scott Antarctic research bases. Levels tracked those in sludge and dust from their respective host countries. The major constituent in the commercial deca-PBDE (BDE-209) was the dominant congener in sludge and dust, as well as in aquatic sediments collected near the McMurdo wastewater outfall. The pattern and level of BDE-209 sediment concentrations, in conjunction with its limited environmental mobility, suggested to the authors inputs from local sources.

Soil and lake sediment samples have been included in several studies on the environmental contamination of Antarctic regions. Total PCB mean concentration was 0.12 (87%) ng g^{-1} dry wt for lake sediment samples and 0.06 (38%) ng g^{-1} dry wt for soil samples collected in a large area of Victoria Land. These values were similar to those reported in lake sediments of Arctic regions (0.12–0.60 ng g^{-1} dry wt).

In fact, in the lake core sediments with glacier melt water input, the accumulation flux of DDT shows an abnormal peak around the 1980s in addition to the expected one in the 1960s, which is most likely caused by the discharge of the DDT stored in the Antarctic ice cap into the lakes. 4,4'-DDE and 4,4'-DDT were also measured in soil samples from Victoria Land. The concentration range was 0.053–0.086 and <0.005–0.020 ng g^{-1} dry wt, respectively. The higher abundance of 4,4'-DDE over 4,4'-DDT precludes that their occurrence may be due to recent spillages. Moderate (2–7 ng g^{-1} dry wt) and high (90–157 ng g^{-1} dry wt) PCB concentrations, along with high level of HCHs and DDTs, were observed in soil samples from the Eastern coast of Antarctica. This local contamination was attributed to biotic focussing of pollutants, due to bird activities (nesting and excrement). High concentrations were also observed in soil samples from James Ross Island: PCBs 0.51-1.82 ng g^{-1} dry wt, HCHs 0.49-1.34 ng g^{-1} dry wt, DDTs 0.51-3.68 ng g^{-1} dry wt. Among soil HCH, only the isomer α-HCH was found above limit of detection between <0.01 and 0.026 ng g^{-1} dry wt. HCB ranged between 0.034 and 0.17 ng g^{-1} dry wt. DDT and HCH were also measured in two lake cores from King George Island, West Antarctica. All concentration ranges are similar to those reported in Arctic lake sediments.

PCBs were measured also in surface snow samples gathered in Victoria Land at several sampling sites located at different altitudes (from sea level to 3,000 m) and at varying distances from the sea. The total PCB concentration showed no significant spatial variations (range 0.28–0.73 pg g^{-1}; mean value 0.52 pg g^{-1}). Moreover, samples from a 2.5-m deep pit at the Hercules Névé collected in summer 1993–1994 and 1994–1995 showed slight higher total PCB concentration (1 pg g^{-1}) in the deepest samples (presumably deposited around 1986-1988) than in surface snow (0.65 pg g^{-1}). This result seems to corroborate previous findings and agrees with the general decreasing trend in POP concentration in the atmosphere of Antarctica and the sub-Antarctic islands during the 1980s and 1990s. These values were about 4 times lower than the average 4.1 pg g^{-1} reported for Canadian Arctic snow.

Lichens and mosses are the principal component of terrestrial flora in many ecosystems of Antarctica whose nutrient supply depends largely on atmospheric deposition. Thus, they can play a very important role as bio-monitors and long-term integrators of persistent contaminant deposition.

POPs were measured in several samples collected in a large area of Victoria Land. Total PCBconcentration in mosses ranges between <5 and 34 ng g^{-1} dry wt. These values are of the same order of magnitude as those reported for the moss *Hylocomium splendens* in Norway, e.g. 6.7–52 ng g^{-1} dry wt. The concentration ranges of HCB, 4,4'-DDE and 4,4'-DDT were 0.85–1.9, 1.1–7.9 and 0.54–0.91 ng g^{-1} dry wt., respectively. 4,4'-DDE showed higher concentration than 4,4'-DDT in all samples, which is consistent with a long-range transport as responsible for the transformation of 4,4'-DDT to 4,4'-DDE after release into the environment. α-HCH and γ-HCH concentration ranges were 0.43–4.0 and 0.18–1.6 ng g^{-1} dry wt., respectively. In almost all cases α-HCH was found in higher concentration than the γ-isomer. The high proportion of the α-isomer is consistent with previous observations in other remote sites, as these areas currently reflect past usage of α -HCH enriched technical mixtures. These low values of the Antarctic mosses confirm the absence of local pollution sources and the lower use of organo-chlorine (OC) compounds in the southern hemisphere.

The overall tendency of lower molecular weight PAH compounds, less chlorinated PCB congeners as well as other more volatile POPs as HCB to be prevalent indicates that long-range atmospheric transport is the most important source of contamination in Antarctica, although high POP levels in proximity of scientific stations need to be continuously monitored.

The reduced frequency of occurrence and concentration of pesticides in Antarctic pelagic plankton can be attributed to decreases in use and to the subsequent decrease of atmospheric concentration over the past few decades. The decline in α- + γ-HCH in Antarctic plankton over time yields an estimated environmental half-life of 2 yr for HCHs in coastal Antarctic surface waters. Phytoplankton (mainly diatoms) and mixed zooplankton (copepods, amphipods and krill) samples from Ross Sea showed a total PCB concentration of 1 and 4.2 ng g^{-1} wet wt. respectively.

Krill (Euphausia superba): in 2008 samples from the eastern Antarctic sector HCB was the single most abundant compound quantified : 4.37 ng g^{-1} lipid wt or 0.2 ng g^{-1} wet wt. HCB concentrations were comparable to those detected at this trophic level in both the Arctic and temperate northwest Atlantic, lending support for the hypothesis that HCB will approach global equilibrium at faster rate than other chlorinated pesticides- p,p'-DDE was detected at notable concentrations: 2.6 ng g^{-1} lipid wt 0.13 ng g^{-1} wet wt; PCB content was very low (1.2 ng g^{-1} lipid wt and 0.5 ng g^{-1} wet wt) in relation to the Arctic and also to previous data (about three orders of magnitude lower than values reported for the Ross Sea).
Pelagic marine mammals, samples in 2008 of Antarctic Type C killer whales: PCBs, DDTs, CHLs, HCB and HCHs were determined in blubber samples and was found that Type C killer whales have the lowest levels of POPs (except HCB) of any killer whale population studied to date.

Coastal benthic organisms The Antarctic shelf is dominated by a single suborder of fish (notothenioids), and most benthic and epibenthic species are notothenioids belonging to the genus *Trematomus* (e.g. *T. bernacchii*). *Trematomus bernacchii* is an ideal bioindicator of local contamination because it has restricted home ranges and is ubiquitous.

Penguins About 90% of the avian biomass in Antarctica consists of penguins. In 2008 – samples of Adelie penguin eggs and fat: p,p'-DDE levels have not declined in the Palmer population of Adelie penguins in more than 30 years. In contrast, ΣDDT decreased significantly from 1975 to 2003 in Arctic seabird eggs. ΣDDT in the fat of Adelie penguins from Cape Crozier measured in 2006 was significantly higher than that measured in 1964. p,p'-DDT/p,p'- DDE ratios <1.0 for several Antarctic organisms, including Adelie penguin eggs, suggest contamination by old DDT. But two independent measurements of ΣDDT indicate that 1-4 kg y^{-1} ΣDDT is currently being released into the Antarctic marine environment due to glacier ablation.

In 2003 - samples of Weddell seals blubber from King George Island: DDT (11-19 ng g-1) and PCB (1-2.5 ng g^{-1}) concentrations are the lowest value so far detected in comparable marine mammals from all over the world and one order of magnitude lower than in samples of the same species from other sites in the Antarctica. This suggests a wide variability of organohalogen levels in the Antarctica, depending on the geographic site;

World regional comparison Average DDT, PCB, HCB and HCH concentrations in samples of Weddell seal blubber from various Antarctic locations are from one to three orders of magnitude lower than average values in the blubber of Arctic seals. Finally, the γHCB/αHCH ratio was >1 while in Arctic samples it was always <1;

In samples of 2002 – eggs of Adélie penguins and South Polar skuas: skua eggs had the highest concentrations of total PCDD/DFs (181 pg g–1), and an estimated concentrations of 2,3,7,8-tetrachlorodibenzo-p-dioxin equivalents (TEQs) (PCDDs, PCDFs and dioxin-

like PCBs) in skua eggs of 344 pg g^{-1} higher than in the liver of polar bears (mean = 120 pg g^{-1}). These concentrations were close to values that may cause adverse health effects. The mean value in skua eggs, for instance, was only two- fold less than the toxicity threshold value reported for American kestrel eggs.

Most of the studies reveal that higher concentration of PCB, PAH and PBDE are measured in the vicinity of research station and their sewage outfall.

Krill . It seems that there is no bio-magnification from plankton to krill. Between contaminants the organochlorine pesticides dominate with HCB being the most abundant.

Type C killer whales have the lowest POPs concentrations (except HCB) but still several times higher (up to 90 fold) than the Antarctic minke whale. This is likely due to diet type, which for the first one consists of fish and other higher trophic level species, while for the second one it consists mainly of krill.

The ECA REPORT 2009 includes the following recomendations

1. An internationally coordinated Antarctic Monitoring and Assessment Programme (AnMAP) should be established.

2. All published data should be collected in a global database and archived in a way in which they can be used effectively for global assessment data. The possibility to point out gaps will make it possible to involve research units of various national research programmes in Antarctica in a coordinated network aimed at covering such knowledge gaps.

3. A database should be created, with limited access if required, containing all data collected within the national monitoring programmes on the local environmental impacts of research stations, according to the Madrid protocol.

The following table presents results of PFOS in biota of Antarctica from five research studies (Bengtson Nash et al., 2010; Giesy and Kannan, 2001; Llorca et al., 2012; Schiavone et al., 2009; Tao et al., 2006) that are displayed in figure 5.2.4g of the report.

Table 1 *Levels of PFOs and PFASs in biota and superficial soil in five research studies.*

	PFCs (ng/mL or g wet weight)					
	Year	**Substance**	**Median (range)**	**Location**	**Population/ Nr of samples**	**Reference**
ALGAE	2010	PFPeA PFHxA PFHpA PFOA PFNA PFBS PFOS FOSA	(1.0 – 1.69) nd nd (0.30 – 1.80) nd nd (66.3 – 111) Nd	Ardley Island, Antarctica	3	Llorca, M. et al., 2012
ANTARCTIC KRILL (Euphausia superba) whole body	2006	PFCs	Nd	60-70º S; 30-80º E East-Antarctic sector, Antarctic	16 (pooled adult samples)	Bengtson Nash et al., 2010
Southern Ocean Albatrosses (eight species: Black-browed, Gray-headed, Laysan, Light-manted Sooty, Royal, Shy, Wandering, Yellow-nosed) livers	2004-2005	PFOs PFOA PFCAs	2.2 (<0.5 – 20.7) (<0.6 – 7.84) Nd	Southern Ocean	102	Tao et al., 2006
WHITE CHINNED PETREL (Procellaria aequinoctialis) muscle	1999-2005	PFOS	1.2 (one bird)	51-52º S; 73-75º E Migratory	5	Bengtson Nash et al., 2010
ANTARCTIC PETREL (Thalassoica antarctica) muscle	1999-2006	PFCs	Nd	65-66º S; 52-87º E Antarctic	5	Bengtson Nash et al., 2010
SOUTH POLAR SKUA (Stecorarius maccormicki) plasma, eggs and whole blood	1990s	PFOs	1.2 (<1 - 1.4)	Ross Sea region, Terra Nova Bay, Antarctica	2	Giesy and Kannan, 2001
	1998/1999	PFOs	2.51 (2.08 - 3.12) 0.88 (0.24 - 1.36)	Antarctica	3 eggs 3 blood	Tao et al., 2006
SUPERFICIAL	2010	PFPeA	Nd	Aitcho Islands,	3	Llorca M., 2012

SOIL		PFHxA PFHpA PFOA PFNA PFBS PFOS FOSA	(0.16 – 0.83) nd nd nd nd (0.32 – 0.36) Nd	Antarctica		
GENTOO PINGUIN (Pygoscelis papua) dung	2010	PFPeA PFHxA PFHpA PFOA PFNA PFBS PFOS FOSA	Nd (19.9 – 237) nd (0.63 – 3.98) (0.78 – 4.33) (10.9 – 45.9) (95.2 – 603) Nd	Ardley Island, Neko Bay, Winter Island, Antarctica	6	Llorca M., 2012
GENTOO PINGUIN (Pygoscelis papua) tissues	2010	PFPeA PFHxA PFHpA PFOA PFNA PFBS PFOS FOSA	(0.11 – 2.27) (0.26 – 0.61) nd nd (0.28 – 0.31) nd (0.063 – 0.103) Nd	Ardley Island, Antarctica	6	Llorca M., 2012
GENTOO PINGUIN (Pygoscelis papua) **and ADÉLIE PINGUIN** (Pygoscelis adeliae) eggs	2004-2005	PFOS	0.28 (0.13 - 0.49) 0.38 (0.18 - 0.89)	King George Island, South Shetland, Antarctic Peninsula	13 Gentoo 13 Adélie	Schiavone et al., 2009
ADÉLIE PINGUIN (Pygoscelis adeliae) muscle	1998-2003	PFCs	Nd	67º S; 62º E Ross Sea region and east-Antarctic sector, Antarctic	4 adults 3 chicks 1 adult	Bengtson Nash et al., 2010
	2001	PFOs	Nd	Admiral Bay, South Shetlands, Antarctica	8 blood	Tao et al., 2006
	1995-1996	PFOs	Nd	Edmonson point, Antarctica	6 eggs	Tao et al., 2006
ANTARCTIC FUR SEAL (Arctocephallus gazella) muscle and liver	2003-2004	PFOS	1.29 (0.42–3.59) 9.01 (1.85-17.25)	Livingston Island, South Shetland, Antarctic Peninsula	20 muscle 17 liver	Schiavone et al., 2009
ANTARCTIC FUR SEAL (WESTERN SECTOR) liver and muscle	2004	PFOS	2.0 (one adult)	54º S; 38º W Antarctic, Sub-Antarctic	6	Bengtson Nash et al., 2010

ANTARCTIC FUR SEAL (Arctocephallus gazella) **(EASTERN SECTOR)** muscle	2002-2003	PFCs	Nd	53º S; 73º E Heard and McDonald Island, Antarctic, Sub-Antarctic	5	Bengtson Nash et al., 2010
WEDDELL SEAL (Leptonychotes weddelii) muscle and liver	1970-1995	PFCs	Nd	68º S; 78º E East-Antarctic sector, Antarctic	5	Bengtson Nash et al., 2010
	1995-2005	PFOs	<35	Terra Nova Bay, Antarctic	1	Giesy and Kannan, 2001
SOUTHERN ELEPHANT SEALS (Mirounga leonina) blood	2004-2005	PFOs	0.53 (<0.08 – 3.52)	Elephant Island, South Shetlands, Antarctic Peninsula	59	Tao et al., 2006
HUMPBACK WHALE (Megaptera novaeangliae) muscle	2006	PFCs	Nd	27º S; 153º E East-Antarctic sector, Antarctic	1	Bengtson Nash et al., 2010

References

Abad E, F Pérez, J.J Llerena, J. Caixach, J. Rivera (2003) Evidence of a specific pattern of poly- chlorinated dibenzo-p-dioxins and dibenzofurans in bivalves. Environ. Sci. Technol., 37, 5090– 5096.

Abd-Allah, A.M., (1992). Determination of DDTs and PCBs residues in Abu- Quir and El-Max bayes, Alexandria Egypt. Toxicological and Environmental Chemistry 36, 89-97.

Addison, R.F., D.C. Muir, M.G. Ikonomou, L. Harwood, T.G. Smith and J. Alikamik (2014).
Temporal trends in "legacy" organochlorine contaminants in blubber of ringed seals (*Phoca hispida*) from Ulukahaktok, NT, Canada between 1972 and 2010.Science of the Total Environment 466-467 (2014) 564-576.

Addison R.F., D.C.G. Muir , M.G. Ikonomou, L. Harwood, T.G. Smith (2009). Hexachloro-cyclohexanes (HCH) in ringed seal (*Phoca hispida*) fromUlukhaktok (Holman), NT: trends from1978 to 2006. Sci Total Environ 2009;407:5139–46

Andral B., J.Y. Stanisiere, D. Sauzade, E. Damier, H. Thebault, F. Galgani, P. Boissery (2004) Monitoring chemical contamination levels in the Mediterranean based on the use of mussel caging. Mar. Pollut. Bull., Vol. 49, 9-10, 704-712.

Andral B., F. Galgani, C. Tomasino, M. Bouchoucha, C. Blottiere, A. Scarpato, J. Benedicto, S. Deudero, M. Calvo, A. Cento, S. Benbrahim, M. Boulahdid, C. Sammari (2011): Chemical Contamination Baseline in the Western Basin of the Mediterranean Sea Based on Transplanted Mussels . Arch Environ Contam Toxicol (2011) 61:261–271.

AMAP, 1998. AMAP Assessment Report: Arctic Pollution Issues. AMAP, Oslo Norway. 859 PP. (see De March 1998)

AMAP, 2003. AMAP Assessment 2002: Human Health in the Arctic AMAP, Oslo Norway xiv + 137pp

AMAP, 2004. AMAP Assessment 2002. Persistent Organic Pollutants in the Arctic, AMAP, Oslo, Norway xvi+310pp. (see de Wit et al 2004).

AMAP, 2009. AMAP Assessment 2009: Human Health in the Arctic. AMAP, Oslo, Norway xiv+259pp.

Bartnicki, J., A. Gusev, W. Aas, H. Fagerli & S. Valiyaveetil (2008): Atmospheric supply of nitrogen, lead, cadmium, mercury and dioxins/furans to the Baltic Sea in 2006. EMEP report to HELCOM. Available at: http://www.helcom.fi /environment2/ hazsubs/EMEP/en_GB/emep2006/

Bidleman, T., R. Macdonald, J. Stow 2003. Sources, occurrence, trends and pathways in

the physical environment. Canadian Arctic contaminants assessment report II. Ottawa; Ministry of Indian Affairs and Northern Development. 2003, 361pp.

Bengston Nash, S. (2011) Persistent organic pollutants in Antarctica : current and future research priorities. J.Environ. Monit. 13, 497- 504.

Bengston Nash, S.M., C.A. Waugh and M. Schalbach (2013). Metabolic Concentration of Lipid Soluble Organochlorine Burdens in the Blubber of Southern Hemisphere Humpback Whales Through Migration and Fasting. Environmental Sci.Technol. 47, 9404-9413.

Bengtson Nash, S., Rintoul, S. R., Kawaguchi, S., Staniland, I., Hoff, J. v. d., Tierney, M. and Bossi, R. (2010). "Perfluorinated compounds in the Antarctic region: Ocean circulation provides prolonged protection from distant sources." Environmental Pollution 158(9): 2985-2991.

Bidleman, T.F., K. Kylin, L.M. Jantunen, P.A. Helm and R.W. Macdonald. (2007). Hexacholrocyclohexane (HCHs) in the Canadian Archipielago. 1. Spatial distribution and pathways of α-, β-, and γ-HCHs in surface water. Environ Sci Technol 2007; 41, 2688-2695.

Bignert, A., S. Danielsson, E. Nyberg, L. Asplund, U. Eriksson, U. Berger, & P. Haglund (2009): Comments concerning the national Swedish contaminant monitoring programme in marine biota. Swedish Museum of Natural History. 153 pp.

Bignert, A., S. Danielsson, A. Strandmark, E. Nyberg, L. Asplund, U. Eriksson, U. Berger, A. Wilander & P. Haglund (2008): Com-ments concerning the national Swedish contaminant monitoring programme in marine biota, 2008. Swedish Museum of Natural History. 142 pp.

Bignert, A., E. Nyberg, K. Sundqvist & K. Wiberg (2007): Spatial variation in concentrations and patterns of the PCDD/F and dioxin-like PCB content in herring from the northern Baltic Sea. Journal of Environmental Moni- toring, 9: 550-556.

Borell A., A. Aguilar (2007) Organochlorine concentrations declined during 1987-2002 in western Mediterranen bottlenose dolphins, a coastal top predator. Chemosphere, 66, 347-352.

Braune B, Muir D, de March B, Gamberg M, Poole K, Currie R, Dodd M, Duschenko W, Eamer J,
Elkin B, Evans M, Grundy S, Hebert C, Johnstone R, Kidd K, Koenig B, Lockhart L, Marshall H,
Reimer K, Sanderson J, Shutt L. 1999. Spatial and temporal trends of contaminants in Canadian Arctic
freshwater and terrestrial ecosystems: a review. Sci Total Environ. 230: 145-207.

Butt, C.M, U. Berger, R. Bossi and G.T. Tomy (2010). Levels and trends of poly-and perfluorinated compounds in the arctic environment . Science of the Total Environment 408 (2010) 2936-2965.

Caixach J, M Calvo, A Bartolomé, O Palacios, M Guerra, E Abad, J Rivera (2007) Analysis of PBDEs, DL-PCBs and PCDD/Fs in caged mussels in the Western Mediterranean Sea. Mytilos Project, Organohalogen Compd, 69, 243–246.

Cantillo, A.Y., 1998. Comparison of results of Mussel Watch Programmes of the United States and France with worldwide mussel watch studies. Marine Pollution Bulletin 36, 712-717.

Castro-Jiménez J., Deviller G., Mariani G., Skejo H., Umlauf G, Wollgast J., Laugier T., Héas-Moisan K., Léauté F., Munschy C., Tixier C., Tronczyński J., 2008. PCDD/F and PCB ambient concentrations, congener patterns and occurrence in a Mediterranean coastal lagoon (Etang de Thau, France). *Environmental Pollution*, 156 : 123-135.

Cohen MD, Draxler RR, Artz R, B. C, Bartlett P, Cooney P, Couchot K, Dickar A, Eisl H, Hill
C, Quigley J, Rosenthal JE, Niemi D, Ratte D, Deslauriers M, Laurin R, Mathewson-Brake L,
McDonald J. 2002. Modeling the atmospheric transport and deposition of PCDD/F to the Great
Lakes. Environ. Sci. Technol. 36: 4831-4845.

Cousins, A.P., M. Remberger, J. Andersson, L. Kaj, K. Strömberg, Y, Ekheden, B. Dusan, E. Brorström-Lundén & I. Cato (2005): Results from the Swedish National Screening Programme 2004 – Sub-report 5: Mirex and Endosulfan. IVL report B1641. 40 pp. IVL Swedish Environmental Research Institute Ltd.

De March, B.G.E., C. De Wit, D.C.G. Muir, B.M. Baume, D.J. Gregor, R.J. Nostrom et al. (1998) Ch 6 Persistent Organic Pollutants. AMAP Assessment Report AMAP 1998, p 183-372.

de Wit, C.A. , A.T. Fisk, K.E. Hobbs, D.C.G. Muir, G.W. Gabrielsen, R. Kallenborn et al. (2004) AMAP Assessment 2002. Persistent Organic Pollutants in the Arctic, AMAP, Oslo, 2004 Norway xvi+310pp.

ECA report (2009). Persistent Organic Pollutants (POPs) in the Antarctic environment A Review of Findings by The SCAR Action Group on Environmental Contamination in Antarctica. R. Fuocoa, G. Capodagliob, B Muscatelloa and M. Radaellib a Department of Chemistry and Industrial Chemistry, University of Pisa (Italy), Department of Environmental Sciences, University of Venice (Italy) February 2009 .

Environment Canada and the U.S. Environemntal Protection Agency. 2014. State of the Great Lakes 2011. Cat No. En161-3/1-2011E-PDF. EPA 950-R-13-002. Available at http://binational.net

Evans MS, Muir D, Lockhart L, Stern G, Ryan M, Roach P. 2005. Persistent organic pollutants and
metals in the freshwater biota of the Canadian Subarctic and Arctic: An overview. Sci. Total
Environ. 351-352: 94-147.

Fiedler H. 2007. National PCDD/PCDF release inventories under the Stockholm Convention
on Persistent Organic Pollutants. Chemosphere 67: S96-S108.

Genualdi, S.A., K.J. Hageman, L.K. Ackerman, S. Usenko and S.L.M.Simonich (2011). Sources and Fate of Chiral Organochlorine Pesticides in Western U.S. National Park Ecosystems. Environ Toxicol Chem 30, 1533-1538

Gessy, J.P. And K. Kannan. (2001). Global distribution of perfluorooctanate in wildlife. Environ. Sci Technology 2001, 35, 1339-1342.

Gomara B, L Bordajandi, M Fernandez, L Herrero, E Abad, M Abalos, J Rivera (2005) Levels and trends of polychlorinated dibenzo-p-dioxins/ furans (PCDD/ Fs) and dioxin-like polychlorinated biphenyls (PCBs) in Spanish commercial fish and shellfish products, 1995–2003, J. Agric. Food Chem., 53, 8406–8413.

Gómez-Gutiérrez, A.I., E. Jover, L. Bodineau, J. Albaigés, J.M. Bayona (2006). Organic contaminant loads into the Western Mediterranean Sea: estimate of Ebro river inputs. Chemosphere 65, 224-236.

Gómez-Gutiérrez, A.I., Garnacho, E., Bayona, J.M., Albaigés, J. (2007). Assessment of the mediterranean sediments contamination by persistent organic pollutants. Environmental Pollution, 148, 396-408.

Gusev, A. 2009. Atmospheric deposition of PCDD/F on the Baltic Sea. HELCOM Indicator Fact Sheets (2009). Available at: http:// www.helcom.fi /environment2/ifs/ifs2009/ en_GB/pcddfdeposition/
(January 2014)

HELCOM (1990). Baltic Sea Declaration. Available at http://www.helcom.fi /stc/fi les/Ministeri- alDeclarations/RonnebyDecl1990.pdf (January 2014)

HELCOM (1993). The Baltic Sea Joint Comprehensive Environmental Action Programme. Balt. Sea Environ. Proc. No. 48.

HELCOM (2007). Baltic Sea Action Plan. Helsinki Commission. Available at: www.helcom.fi

HELCOM (2009). Hazardous substances of specific concern to the Baltic Sea – Final

report of the HAZARDOUS project. Balt. Sea Environ. Proc. No. 119.

HELCOM (2010)a Hazardous substances in the Baltic Sea – An integrated thematic assessment of hazardous substances in the Baltic Sea. Balt. Sea Environ. Proc. No. 120B.

HELCOM, (2010)b Ecosystem Health of the Baltic Sea 2003–2007: HELCOM Initial Holistic Assessment. Balt. Sea Environ. Proc. No. 122.

HELCOM (2012) Development of a set of core indicators: Interim report of the HELCOM CORESET project. PART A: Description of the selection process. Balt. Sea Envin. Proc. No 129 A.

HELCOM (2013). HELCOM core indicators: Final report of the HELCOM CORESET project. Balt. Sea Environ. Proc. No. 136

Hoferkamp, L., M.H. Hermanson, D.C.G. Muir (2010). Current use pesticides in Arctic media; 2000-2007. Science of the Total Environment 408 (2010) 2985-2994.

Holmström, K.E., U. Jarnberg & A. Bignert (2005): Temporal trends of PFOS and PFOA in guillemot eggs from the Baltic Sea, 1968- 2003. Environmental Science & Technology 39:80-84.

Holmström, K.E. & U. Berger (2008). Tissue distribution of perfluorinated surfactants in common guillemot (*Uria aalge*) from the Baltic Sea, Environ. Sci. Technol. 42:5879-5884.

Hung H, R. Kallenborn, K. Breivik, Y. Su, E. Brorstrom-Lunden, K.Olafsdottir et al. (2010) Atmo- spheric monitoring of organic pollutants in the Arctic under the Arctic Monitoring and Assessment Programme (AMAP): 1993–2006. Sci Total Environ 2010;408: 2854–73

ICES 2008: ICES Advice, Book 1: Development of proposals for environmental assessment criteria. Available at: http://www.ices.dk/ committe/acom/comwork/report/2008/ Special%20Requests/OSPAR%20Develop-ment%20of%20proposals%20for%20 environmental%20assessment%20criteria. Pdf (January 2014)

Isosaari, P., H. Kankaanpää, J. Mattila, H. Kiviranta, M. Verta, S. Salo & T. Vartiainen (2002): Spatial Distribution and Temporal Accu- mulation of Polychlorinated Dibenzo-p-dioxins, Dibenzofurans, and Biphenyls in the Gulf of Finland. Environ. Sci. Technol. 36:2560-2565.

Kannan, K., S. Corsolini, J. Falandysz, G. Oehme, S. Focardi, J.P. Giesy (2002): Perfl uorooc- tanesulfonate and related fl uorinated hydrocarbons in Marine Mammals, Fishes, and Birds from Coasts of the Baltic and the Mediterranean Seas, Environ. Sci. Technol. 36: 3210-3216."

Karl, H. & U. Ruoff (2007). Dioxins, dioxin-like PCBs and chloroorganic contaminants in herring, Clupea harengus, from different fishing grounds of the Baltic Sea. Chemosphere 67:S90-S95."

Landers, D.H., Simonich, S.L., Jaffe, D.A., Geiser, L.H., Campbell, D.H., Schwindt, A.R., Schreck, C.B., Kent, M.L., Hafner, W.D., Taylor, H.E., Hageman, K.J., Usenko, S., Ackerman, L.K., Schrlau, J.E., Rose, N.L., Blett, T.F. and Erway, M.M. (2008). The fate, transport, and ecological impacts of airborne contaminants in western national parks (USA). Western Airborne Contaminants Assessment Project final report: U. S. Environmental Protection Agency, Office of Research and volume 1. Development, NHEERL, Western Ecology Division. Corvalis, OR.

Law,R.,Hanke,G.,Angelidis,M.Batty,J.,Bignert,A.,Dachs,J.,Davies,I.,Denga,A.,Duffek,B., Herut,H.,
Hylland,K.,Lepom,P.,Leonards,P.,Mehtonen,J.,Piha,M.,Roose,P.,Tronczynski,J.,Velikova, V.
and 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, 2010. 161pp. Scientific
and Technical Research series, ISSN 978-92- 79- 15648- 9.DOI10.2788/85887

Letcher, R.J. , J.O. Bustnes, R. Dietz, B.M. Jenssen, E.H. Jørgensen, C. Sonne, J. Verreault, M.M. Vijayan, G.W. Gabrielsen (2010). Exposure and effects assessment of persistent organohalogen contaminants in arctic wildlife and fish. Science of the Total Environment 408 (2010) 2995-3043

Li YF and R.W. Macdonald (2005). Sources and pathways of selected organochlorine pesticides to the Arctic and the effect of pathway divergence on HCH trends in biota: a review. Sci Total Environ 2005;342:87-106.

Lizak, R., J. Piskorska-Pliszczyńska, B. Kowalski, J. Rachubik, M. Warenik & T. Wijaska (2007): PCDD, PCDF, and dl-PCB in Baltic herring, salmon and sprat from Western Gotland Basin of the Baltic Sea. Bull. Vet. Ins. Pulawy 51:661-666.

Llorca,M., Farre, M., Tavano, M.S., Alonso, B., Koremblit, G.,Barcelo, D., 2012. Fate of a broad spectrum of perfluorinated compounds in soils and biota from Tierra del Fuego and Antarctica. Environmental Pollution163,158-166.

Munschy C, N Guiot, K.Héas-Moisan, C Tixier, J Tronczyjski (2008) Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) in marine mussels from French coasts: Levels, patterns and temporal trends from 1981 to 2005. Chemosphere, 73, 945–953

NCP 2013. Canadian Arctic Contaminants Assessment Report On Persistent Organic

Pollutants – 2013.
Muir D, Kurt-Karakus P, Stow J. (Eds). Northern Contaminants Program, Aboriginal Affairs and Northern Development Canada, Ottawa ON. xxiii + 487 pp + Annex

OSPAR (2005). 2005 Assessment of data collected under the Coordinated Environmental Monitoring Programme (CEMP). OSPAR Publication 235/2005. ISBN 1-904426-77-8.

OSPAR (2006). 2005/2006 CEMP Assessment: Trends and concentrations of selected hazardous substances in the marine environment. OSPAR Publication 288/2006. ISBN 1-905859-26-0.

OSPAR (2007). 2006/2007 CEMP Assessment: Trends and concentrations of selected hazardous substances in the marine environment. OSPAR Publication 330/2007. ISBN 978-1-905859-69-6.

OSPAR (2008). 2007/2008 CEMP Assessment: Trends and concentrations of selected hazardous substances in sediments and trends in TBT-specific biological effects. OSPAR Publication 378/2008. ISBN 978-1-906840-19-8.

OSPAR (2009)a. CEMP assessment report: 2008/2009 Assessment of trends and concentrations of selected hazardous substances in sediments and biota, OSPAR Commission, 2009. OSPAR Publication 390/2009. ISBN 978-1-906840-30-3.

OSPAR (2009)b. Background Document on Assessment Criteria used for assessing CEMP Monitoring Data for the Concentrations of Hazardous Substances in Marine Sediments and Biota in the Context of
QSR 2010. OSPAR Publication 461/2009. ISBN 978-1-907390-08-1.

OSPAR (2009)c. Trends in atmospheric concentrations and deposition of nitrogen and selected hazardous substances to the OSPAR maritime area. OSPAR publication : 447/2009 ISBN 978-1-906840-87-7

OSPAR (2012)a. Finding common ground Towards regional coherence in implementing the Marine Strategy Framework Directive in the North-East Atlantic region through the work of the OSPAR Commission. OSPAR publication 578/2012 ISBN 978-1-909159-12-9"

OSPAR (2012)b. MSFD Advice document on Good environmental status - Descriptor 8:Contaminants.Version of 2 March 2012 Prepared by the OSPAR Committee of Hazardous Substances and Eutrophication (HASEC). OSPAR publication 584/2012. ISBN 978-1-909159-17-4

OSPAR (2013)a.. Comprehensive Atmospheric Monitoring Programme in 2011, OSPAR Publication 597/2013. ISBN 978-1-909159-30-3

OSPAR (2012)c. Comprehensive Atmospheric Monitoring Programme. Deposition of air

pollutants around the North Sea and the North-East Atlantic in 2010. OSPAR publication 564/2012. ISBN 978-1-907390-79-1

OSPAR (2013)b. Riverine Inputs and Direct Discharges to Convention Waters - OSPAR Contracting Parties' RID 2011 Data Report. OSPAR. Publication 598/2013, ISBN 978-1-909159-31-0.

OSPAR (2013)c. Levels and trends in marine contaminants and their biological effects – CEMP Assessment report 2012. OSPAR. Publication 596/2013, ISBN 978-1-909159-29-7.

Picer, M., N. Picer, (1991). Long-term trends of DDTs and PCBs in sediment samples collected from the Eastern Adriatic Coastal Waters. Bulletin of Environmental Contamination and Toxicology 47, 864-873.

Rigét, F., A. Bignert, B. Braune, J. Stow and S. Wilson (2010). Temporal trends of legacy POPs in Arctic biota, an update. Science of the Total Environment 408 (2010) 2874-2884.

Schiavone, A., Corsolini, S., Kannan, K., Tao, L., Trivelpiece, W., Torres, D., Focardi, S., 2009. Perfluorinated contaminants in fur seal pups and penguin eggs from South Shetland, Antarctica. Science of the Total Environment 407, 3899-3904.

Selliah S. Reiner E, Toner D, Dawood R, Farag R, Ali B. 2008. Northern Contaminants Quality
Assurance Program: Synopsis of Research Conducted under the 2007-2008 Northern Contaminants Program. Indian and Northern Affairs Canada, Ottawa ON. p. 176-181.

Sole M, C Porte, J Albaiges (2001) Hydrocarbons, PCBs and DDT in the NW Mediterranean deep- sea Deep-Sea Research I, 48, 495-513.

Sonne C. 2010. Health effects from long-range transported contaminants in arctic top predators: An
integrated review based on studies of polar bears and relevant model species. Environ. Int. 36:
461-491.

Sturludottir E., H. Gunnlaugsdottir, H.O. Jorundsdottir, E.V. Magnusdottir, K. Olafsdottir, G. Stefansson (2013). Spatial and temporal trends of contaminants in mussel sampled around the Icelandic coastline. Science of the Total Environment 454-455 (2013) 500-509.

Tao, L., Kannan, K., Kajiwara, N., Costa, M.A., Fillman, G., Takahashi, S., Tanabe, S., 2006. Perfluorooctanesulfonate and related fluorochemicals in albatrosses, elephant seals, penguins and polar skuas from the Southern Ocean. Environmental Science and Technology 40, 7642-7648.

Taleb Z, I Benali, H Gherras, A Ykhlef-Allal, B Bachir-Bouiadjra, JC Amiard, Z Boutiba (2009) Biomonitoring of environmental pollution on the Algerian west coast using caged mussels Mytilus galloprovincialis, Oceanologia, 51, 1, 63-84.

Tolosa, I., J.M Bayona, J. Albaigés (1995). Spatial and temporal distribution, fluxes, and budgets of organochlorinated compound in Northwest Mediterranean sediments. Environmental Science and Technology 29, 2519-2527.

Tolosa, I., J.W. Readman, S.W. Fowler, J.P Villeneuve, J. Dachs, J.M Bayona, J. Albaigés (1997). PCBs in the western Mediterranean. Temporal trends and mass balance assessment. Deep-Sea Research II 44, 907-928.

UNEP (1990). Assessment of the State of Pollution of the Mediterranean Sea by Organohalogen Compounds. MAP Technical Report No. 39. Athens.

UNEP/FAO/WHO (1996) Assessment of the state of eutrophication in the Mediterranean Sea. MAP Technical Reports Series No. 106. UNEP, Athens, 211 pp.

UNEP (2002). Regionally Based Assessment of Persistent Toxic Substances. Mediterranean Regional Report. UNEP, Geneva, Switzerland.

UNEP (2007). MED POL Database. UNEP(2006) Biological effects monitoring program, MAP technical report, series 166, 244 pages

UNEP/MAP-MED POL (2009). Hazardous substances in the Mediterranean an assessment of the MEDPOL Database (J.Pon , C. Murciano, J. Albaigés) final report 91 p.

UNEP/MAP- Plan Bleu (2009) State of the Environment and Development in the Mediterranean, UNEP/MAP-Plan Bleu, Athens, 2009.

UNEP/MAP RAC/SPA (2010)a Synthèse subrégionale « Méditerranée Occidentale » des documents nationaux d'identification des propriétés majeures des écosystèmes et d'évaluation de l'état écologique et des pressions sur la biodiversité marine et côtière. Par Thierry Perez & Arthur Antonioli, Ed. CAR/ASP, Tunis; 54 pp.

UNEP/MAP RAC/SPA (2010)b Identification of important ecosystem properties and assessment of ecological status and pressures to the Mediterranean marine and coastal biodiversity in the Ionian Sea and the Central Mediterranean. Edited by Ben Haj, S. Ed. RAC/SPA, Tunis; 50 pages.

UNEP/MAP RAC/SPA (2010)c Identification of important ecosystem properties and assessment of ecological status and pressures to the Mediterranean marine and coastal biodiversity in the Adriatic Sea. Öztürk, B., Ed. RAC/SPA, Tunis; 50 pages.

UNEP/MAP RAC/SPA (2010)d Identification of important ecosystem properties and

assessment of ecological status and pressures to the Mediterranean marine and coastal biodiversity in the Aegean Sea-Levant Sea. Edited by Boero, F. Ed. RAC/SPA, Tunis; 80 pages.

UNEP/MAP (2011) Reviewing MED POL marine monitoring activities and planning for the new integrated MAP monitoring system UNEP/MAP Athens 2011

UNEP/MAP (2012) Initial Integrated Assessment of the Mediterranean Sea : fulfilling step 3 of the ecosystem approach process. UNEP/MAP Athens 2012.