Persistent organic pollutants, brominated flame retardants and synthetic musks in fish from remote alpine lakes in Switzerland

Peter Schmid [a,*], Martin Kohler [a], Erika Gujer [a], Markus Zennegg [a], Marco Lanfranchi [b]

[a] *Empa, Swiss Federal Laboratories for Materials Testing and Research, Laboratory for Analytical Chemistry, Überlandstrasse 129, CH-8600 Dübendorf, Switzerland*
[b] *Agency for Nature and Environment of the Canton of Grisons, Gürtelstrasse 89, CH-7001 Chur, Switzerland*

Abstract

Remote alpine lakes do not receive any direct aquatic inputs from anthropogenic activities. Therefore, these ecosystems may receive persistent organic compounds (POPs) by direct atmospheric deposition, only. Consequently, fish dwelling in these ecosystems represent an excellent indicator for the long-term atmospheric input of bioaccumulating and persistent contaminants. In the present study, fish from seven remote alpine lakes, located between 2062 and 2637 m above sea level in south eastern Switzerland (Grisons), were investigated. Lipid-based fish tissue concentrations of pesticides including dichlorodiphenyltrichloroethane (DDT) and its transformation products (2,4′-DDT, 4,4′-DDT, 2,4′-dichlorodiphenyldichloroethane (DDD), 4,4′-DDD, 2,4′-dichlorodiphenyldichloroethene (DDE), 4,4′-DDE), as well as dieldrin, heptachlor *exo*-epoxide (HPEX), hexachlorobenzene (HCB), hexachlorocyclohexanes (HCH), polychlorinated biphenyls (PCB), polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/F), and polybrominated diphenyl ethers (PBDE) were measured. In addition, seven synthetic musk compounds (Crysolide (ADBI), Phantolide (AHMI), Fixolide (AHTN), Traseolide (ATII), Galaxolide (HHCB), musk ketone (MK), and musk xylene (MX)) were determined. Concentrations of PCB, PCDD/F, and PBDE were in the same range as in fish from the major lakes situated in the Swiss plateau, indicating mainly atmospheric input of these persistent compounds. In contrast, concentrations of synthetic musks which are used as fragrances in laundry detergents and cosmetic products were distinctly lower than concentrations in fish from Swiss plateau lakes which receive inputs from waste water treatment plants.
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1. Introduction

Global transfer of persistent semivolatile organic compounds (SVOCs), such as persistent organic pollutants (POPs), is based on long-range atmospheric transport in combination with condensation and volatilization processes. Within the global atmosphere, evaporation in warmer areas and subsequent deposition in colder places leads to a transfer of contaminants towards the poles.

The phenomenon of atmospheric migration of chemically stable, semivolatile chemicals, including polychlorinated biphenyls (PCB) and dichlorodiphenyltrichloroethane (DDT), has been predicted and associated with the term "global distillation" by Goldberg (1975). Later on, this phenomenon has been extensively reviewed by Mackay and Wania (1995). For alpine regions with low average temperatures, Grimalt et al. (2001) have shown that accumulation of organochlorine compounds, such as pesticides and PCB, is enhanced by temperature-controlled condensation, as well. For less volatile components, however, such as polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/F) and polybrominated diphenyl ethers (PBDE),

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* Corresponding author. Tel.: +41 44 823 46 51; fax: +41 44 823 40 41.
 E-mail address: peter.schmid@empa.ch (P. Schmid).

particle-bound deposition was shown to be the dominating transfer mechanism.

The hydrology of remote alpine lakes is determined by direct input through precipitation and inputs from the surrounding catchment area. In contrast to lakes situated in the Swiss midlands there are no direct inputs from waste water treatment plants (WWTP) and other anthropogenic sources. Therefore, the input of persistent organic compounds into these lakes is controlled by atmospheric deposition and condensation. Following these input processes, contaminants are bioaccumulated by the creatures dwelling in and feeding on these ecosystems. Situated to the uppermost trophic level and providing maximum enrichment of persistent contaminants through bioaccumulation, fish from remote alpine lakes represent an excellent indicator for atmospheric long-term input of persistent organic compounds, including POPs.

In the present study, fish from seven alpine lakes from south eastern Switzerland (Grisons) situated between 2062 and 2631 m above sea level were investigated (see Fig. 1). With the exception of Laghetto Moesola, significant input from local anthropogenic emissions can be

excluded. In pooled muscle tissue representing 2–19 individual fish of each lake, concentrations of pesticides as well as their transformation and side products (DDT, dichlorodiphenyldichloroethene (DDE), dichlorodiphenyldichloroethane (DDD), dieldrin, heptachlor *exo*-epoxide (HEPX), hexachlorobenzene (HCB), α-, β-, and γ-hexachlorocyclohexane (α-, β-, and γ-HCH)) will be reported. Next to organochlorine pesticides and their metabolites, persistent organic compounds including PCB, PCDD/F, and PBDE, as well as synthetic musk compounds such as Crysolide (ADBI), Phantolide (AHMI), Fixolide (AHTN), Traseolide (ATII), Galaxolide (HHCB), musk ketone (MK), and musk xylene (MX) will be reported and discussed.

2. Experimental section

2.1. Sample material

Brown trout (*Salmo trutta fario*), brook trout (*Salvelinus fontinalis*), alpine char (*Salvelinus alpinus*), and lake trout (*Salvelinus namaycush*) were caught in August and September 2003 in seven remote alpine lakes (see Fig. 1 and Table 1). Whole fish were wrapped in aluminium foil and stored at −20 °C until analysis.

2.2. Chemicals

Authentic reference materials, including pesticides, PCB, PCDD/F, PBDE, and synthetic musks were obtained from the following sources: AccuStandard Inc. (New Haven, CT, USA), Cambridge Isotope Laboratories (Andover, MA, USA), Dr. Ehrenstorfer GmbH (Augsburg, Germany), Givaudan Roure (Dübendorf, Switzerland), LGC Promochem GmbH (Wesel, Germany), and Riedel-de Haën (distributed by Fluka, Buchs, Switzerland). The following isotope labelled standards were purchased from Cambridge Isotope Laboratories: mixture of the $^{13}C_{12}$-PCB 28, 52, 101, 138, 153, and 180 (EC-4058), mixture of the $^{13}C_{12}$-PCB 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, and 189 (EC-4937), $^{13}C_{12}$-1,2,7,8-tetra-
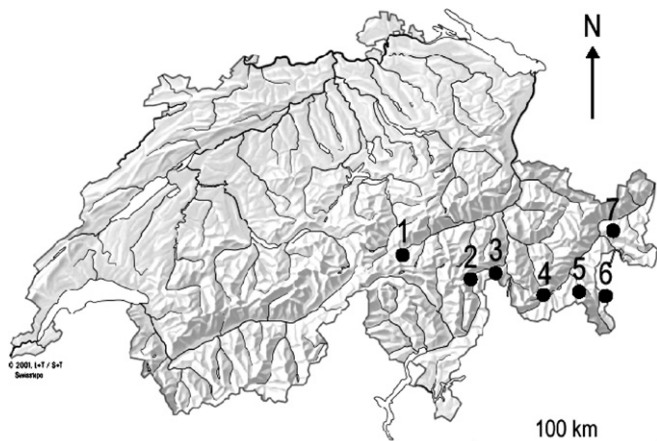

Fig. 1. Locations of the alpine lakes south eastern Switzerland (1: Lai da Tuma, 2: Laghetto Moesola, 3: Surettasee, 4: Lägh dal Lunghin, 5: Lei da Diavolezza, 6: Lagh dal Teo, 7: Lai Grond).

Table 1
Parameters of alpine lakes included in the present study

Lake	Longitude (E)	Latitude (N)	Altitude (m)	Surface area (km²)	Average depth (m)	Ice coverage	Characteristics
Lai da Tuma (L. Tuma)	8.6735	46.6339	2345	0.0255	6.8	Nov–June	Remote
Laghetto Moesola (L. Moesola)	9.1718	46.4955	2062	0.0597	5.0	Nov–June	Nearby pass road
Surettasee	9.3486	46.5385	2195	0.0367	5.3	Nov–July	Slight tourism
Lägh dal Lunghin (L. Lunghin)	9.6756	46.4188	2484	0.0513	9.2	Nov–July	Remote
Lei da Diavolezza (L. Diavolezza)	9.9740	46.4218	2573	0.0349	5.8	Oct–June	Water used for preparation of slopes for downhill skiing
Lagh dal Teo (L. Teo)	10.1132	46.3655	2353	0.0363	12.8	Sept–June	Remote
Lai Grond (L. Grond)	10.1266	46.7305	2631	0.0163	4.8	Nov–July	Remote

CDF (EF-1438), mixture of the $^{13}C_{12}$-2,3,7,8-chlorosubstituted PCDD and PCDF (EDF-4067), $^{13}C_{12}$-4,4'-DDE (CLM-1627-1.2), $^{13}C_{12}$-dieldrin (CLM-4726-1.2), $^{13}C_6$-γ-HCH (CLM-1282), $^{13}C_{12}$-BDE 28 (EO-5002), $^{13}C_{12}$-BDE 47 (EO-4982), $^{13}C_{12}$-BDE 99 (EO-4983), $^{13}C_{12}$-BDE 100 (EO-4993), $^{13}C_{12}$-BDE 153 (EO-4984), $^{13}C_{12}$-BDE 183 (EO-4985). $^{13}C_{12}$-4,4'-DDT, $^{13}C_6$-HCB, and $^2H_3$-AHTN were from Dr. Ehrenstorfer GmbH. $^{15}N_3$-MX (1-*tert*-butyl-3,5-dimethyl-2,4,6-trinitrobenzene) was synthesized in our laboratories.

2.3. Sample preparation

As the mountain lake fish were too small for individual analyses of trace contaminants such as PCDD/F, the available sample material had to be pooled lake-wise, irrespective of different fish species (L. Tuma and Surettasee, see Table 2). Muscle tissue (fillet) was cut in pieces and sample pools of approximately 300 g were prepared and homogenized using a hand blender. Lipids were extracted according to Fürst et al. (1989). Following lipid extraction, lipid contents were determined gravimetrically. For the analysis of PCDD/F and dioxin-like PCB, samples were processed as reported previously (Zennegg et al., 2002). Sample preparation for all other analytes was performed by gel permeation chromatography (Fürst et al., 1989). These extracts were used for gas chromatography/high resolution mass spectrometry (GC/MS) analysis without further purification.

2.4. Quantitative analysis

Quantitative determination of the contaminants was based on GC/MS with single ion monitoring (SIM) using isotope-labelled internal standards. GC/MS analyses were carried out on a MAT 95 mass spectrometer (Thermo Finnigan MAT, Bremen, Germany) coupled to a gas chromatograph HRGC Mega 2 series (Fisons Instruments, Rodano, Italy) equipped with an A200S auto sampler (CTC Analytics, Zwingen, Switzerland). Samples were injected in splitless mode (splitless time 20 s) at an injector temperature of 260 °C. For separation of PCDD/F and dioxin-like PCB, a J&W DB-Dioxin column 60 m ×

0.25 mm (film thickness 0.15 μm) was used. The initial temperature was 180 °C. After 1.5 min, the temperature was ramped at 20 °C/min up to 220 °C and at 2 °C/min up to 260 °C. GC separation of pesticides and synthetic musks was carried out on a 20 m × 0.30 mm glass capillary column coated with a DB-5 analogue stationary phase (PS 086, Fluka, Buchs, Switzerland) with a film thickness of 0.15 μm using hydrogen at 40 kPa as carrier gas. The following temperature program was used: 110 °C (1 min), 20 °C/min up to 150 °C, 4 °C/min up to 220 °C, 20 °C/min up to 260 °C (5 min). PBDE were separated on a 10 m × 0.28 mm glass capillary (stationary phase polydimethylsiloxane PS 347.5 from Fluka, film thickness 0.10 μm) with hydrogen at 20 kPa as carrier gas. The temperature program was 110 °C (1 min), 12 °C/min up to 320 °C (5 min). The ion source was operated at 180 °C (PBDE: 220 °C), the electron energy was 56 eV, and the mass spectrometer was tuned to a mass resolution of $m/\Delta m = 10\,000$. The two most abundant signals of the molecular ion clusters (DDE, HCB, HCH, PCB and PCDD/F) or of typical fragment ions (DDD, DDT, dieldrin, HPEX, synthetic musks, PBDE) were recorded in SIM mode. Calculation of concentrations was based on comparison with reference mixtures, and detection limits were based on a signal-to-noise ratio of 3:1.

2.5. Analytical quality control

Blank samples were prepared by submitting pure solvents spiked with internal standards to the complete clean-up procedures. Blank levels of HCH, MK, MX, ADBI, AHMI, and ATII were below the lowest concentration detected in any of the samples. For the remaining analytes, blank levels were less than 10% of the lowest concentration detected in any of the samples.

2.6. Data processing

Calculation of toxic equivalents (WHO-TEQ) was based on the Toxicity Equivalency Factors of the World Health Organization (WHO-TEF (van den Berg et al., 1998)). For congeners below the detection limit, WHO-TEF were calculated assuming a concentration representing 50% of

Table 2
Parameters of fish investigated in the present study

Lake	Stocking	Fish species	No. of pooled individuals	Total weight (g)	Total fillet weight (g)	Average fat content (%)
L. Tuma	Annual (500–700 fingerlings of brown trout)	Brown trout, alpine char	16	1332	555	2.03
L. Moesola	Annual (100–150 fingerlings of brown trout)	Brown trout	10	2096	712	2.78
Surettasee	Stocking unknown	Brook trout, alpine char	19	1034	420	1.89
L. Lunghin	Last stocking in 1992 (lake trout)	Lake trout	2	593	289	2.93
L. Diavolezza	Last stocking in 1977 (lake trout)	Lake trout	12	2074	723	3.78
L. Teo	Annual (150 fingerlings of brown trout)	Brown trout	10	884	300	1.33
L. Grond	Last stocking in 1978	Brown trout	10	1894	710	1.26

the detection limits of the particular congener. The detection limits of these 17 congeners contributed less than 5% to the total WHO-TEQ in any sample.

3. Results and discussion

An overview of the contaminant concentrations in the pooled fish samples from the seven lakes is shown in Table 3. Within the organochlorine pesticides, DDT (given as sum of 2,4′-DDT, and 4,4′-DDT, 2,4′-DDD, 4,4′-DDD, 2,4′-DDE, 4,4′-DDE) is the most prominent compound, reaching concentrations between 130 and 1100 ng/g lipid weight (lw). Concentrations of dieldrin, HCB, γ-HCH and HPEX, pesticides now banned by the Stockholm Convention, reach levels between 1.1 and 100 ng/g lw. Within this concentration range, all other persistent organic contaminants were encountered. This includes the polybrominated diphenyl ethers (sum of BDE 28–183 was 13–110 ng/g lw) and various synthetic musks (0.79–230 ng/g lw). BDE 47 was the most prominent congener (6.6–39 ng/g lw), followed by BDE 99, 100, and 153. Within the group of synthetic musks, the highest tissue concentrations were detected for AHTN (20–54 ng/g lw) and HHCB (42–230 ng/g lw).

Since the concentrations of the individual contaminants span a rather wide range for the seven lakes, the questions raises how accurately the observed concentrations in fish reflect the exposure of the respective lakes to these contaminants. Next to geographical aspects (proximity to nearby mountain pass roads, tourism), aspects of fish population management need to be considered. As landlocked salmo-

nid fish are known to exhibit occasionally cannibalistic behaviour, stocking of the fish population by introduction of fingerlings can be a pathway of contaminant input to the resident fish population and therefore be a substantial pathway of contaminant input into these lakes. As the fish populations of some of the lakes (L. Tuma, L. Moesola, and L. Teo) are supported by regular stocking (see Table 2) it can indeed not be excluded that the contaminant levels in these fish are influenced. As a matter of fact, the average DDT levels in fish from these three lakes (764 ± 270 ng/g lw) are clearly higher than those from the four lakes (380 ± 200 ng/g lw) that did no receive fish by stocking. However, there is no clear difference between the levels and contaminant patterns in fish from lakes with and without recent stocking.

With regard to the PCB (see Table 4), the highest concentrations were detected for PCB 153 reaching values up to 560 ng/g lw in the composite sample of L. Tuma. The PCB congener patterns of these samples are dominated by hexa- and hepta bromo biphenyl congeners (PCB 138, PCB 153 and PCB 180). Based on the concentrations of the dioxin-like coplanar PCB congeners (with PCB 118 being the most prominent representative), WHO-TEQ between 0.030 and 0.12 ng/g lw were calculated. Levels of dioxins (PCDD/F), expressed as WHO-TEQ reached levels between 0.0022 and 0.019 ng/g lw (Table 4). Hence, coplanar PCB represent the major source of dioxin toxicity in this fish.

The same finding was reported in an investigation on the exposure of fish from 10 lakes situated in the plateau of Switzerland to PCB and PCDD/F by Zennegg et al.

Table 3
Compilation of total concentrations of persistent organic pollutants in fish from alpine lakes in the Grisons, Switzerland (ng/g, lipid weight (lw) based)

	L. Tuma	L. Lunghin	L. Moesola	Surettasee	L. Diavolezza	L. Teo	L. Grond
Pesticides							
Total DDT[a]	1100	390	680	370	130	550	630
dieldrin	57	16	22	43	100	12	19
HCB	22	15	12	22	14	11	12
γ-HCH	1.8	4.9	2.6	1.6	1.8	1.1	1.4
HPEX	8.5	4.0	4.3	6.2	14	3.0	4.2
PBDE							
BDE 28	0.57	0.80	0.42	1.1	0.26	0.22	0.38
BDE 47	24	39	12	14	6.6	9.5	12
BDE 99	14	37	6.0	5.3	3.5	9.4	7.0
BDE 100	8.5	12	4.2	5.9	1.6	5.0	3.8
BDE 153	3.2	10	0.30	2.1	0.61	2.6	1.5
BDE 154	0.52	1.2	<0.22	<0.089	<0.12	<0.11	<0.26
BDE 183	1.1	6.9	0.86	0.91	0.40	0.84	0.67
Total PBDE	52	110	24	30	13	28	25s
Synthetic musks							
MX (musk xylene)	1.9	12	1.8	1.9	1.3	2.5	2.2
MK (musk ketone)	2.2	2.6	2.5	2.2	2.0	2.9	2.1
ADBI (Crysolide)	33	28	29	8.7	29	35	27
AHMI (Phantolide)	1.7	2.5	1.2	0.79	6.3	2.0	3.3
ATII (Traseolide)	1.7	2.1	1.9	1.4	1.3	2.1	1.7
AHTN (Fixolide)	30	38	54	20	27	27	20
HHCB (Galaxolide)	50	78	230	42	46	53	44

[a] Sum of 2,4′-DDD, 4,4′-DDD, 2,4′-DDE, 4,4′-DDE, 2,4′-DDT, 4,4′-DDT.

Table 4
Concentrations of PCB, dioxin-like PCB, and PCDD/F in fish from alpine lakes in the Grisons, Switzerland (ng/g, lipid weight (lw) based)

	L. Tuma	L. Lunghin	L. Moesola	Surettasee	L. Diavolezza	L. Teo	L. Grond
Indicator PCB							
PCB 28	3.2	3.5	4.7	3.2	1.6	2.9	3.3
PCB 52	9.0	6.0	12	4.4	4.0	2.7	4.6
PCB 101	78	25	83	26	18	19	44
PCB 138	340	120	340	130	50	120	220
PCB 153	570	210	550	190	73	210	460
PCB 180	310	120	310	120	29	170	210
Dioxin-like PCB							
PCB 77	0.90	1.3	1.0	0.75	0.58	0.56	0.71
PCB 81	0.065	0.066	0.082	0.031	0.022	0.024	0.047
PCB 105	30	15	36	11	4.5	11	22
PCB 114	1.8	0.85	2.1	0.54	0.19	0.57	1.2
PCB 118	110	62	140	38	14	37	80
PCB 123	13	7.2	14	5.1	2.1	4.2	13
PCB 126	0.84	0.83	0.78	0.45	0.26	0.43	0.64
PCB 156	26	11	31	11	3.0	12	17
PCB 157	6.4	3.9	6.5	2.7	0.85	2.8	3.9
PCB 167	18	9.5	20	7.8	2.7	8.9	14
PCB 169	0.12	0.13	0.088	0.059	0.027	0.060	0.089
PCB 189	5.2	2.9	5.5	2.5	0.50	3.1	3.3
WHO-TEQ PCB	0.12	0.10	0.12	0.059	0.030	0.057	0.088
PCDD/F							
2,3,7,8-TCDD	0.0015	0.00058	0.0017	0.00029	0.00014	0.00042	0.00055
1,2,3,7,8-PeCDD	0.0039	0.0014	0.0043	0.0011	0.00034	0.00049	0.0012
1,2,3,4,7,8-HxCDD	0.00067	0.00039	0.00076	0.00033	0.00010	0.00042	0.00055
1,2,3,6,7,8-HxCDD	0.0025	0.0012	0.0042	0.00086	0.00025	0.00079	0.0014
1,2,3,7,8,9-HxCDD	0.00067	0.00031	0.00056	0.00032	0.00018	<0.00058	0.00065
1,2,3,4,6,7,8-HpCDD	0.0019	0.00068	0.0026	0.00087	0.00029	0.0018	0.0016
OCDD	0.0035	0.0017	0.0035	0.0034	0.0010	0.0067	0.0030
2,3,7,8-TCDF	0.034	0.022	0.043	0.012	0.0086	0.0098	0.012
1,2,3,7,8-PeCDF	0.0052	0.0021	0.0060	0.0016	0.00067	0.0014	0.0016
2,3,4,7,8-PeCDF	0.013	0.0037	0.0140	0.0029	0.0015	0.0022	0.0038
1,2,3,4,7,8-HxCDF	0.0016	0.0005	0.0022	0.00057	0.00018	0.00032	0.00080
1,2,3,6,7,8-HxCDF	0.0033	0.0013	0.0043	0.0010	0.00029	0.00091	0.0017
1,2,3,7,8,9-HxCDF	0.00020	<0.00013	0.00011	<0.00023	0.00006	<0.00030	0.00043
2,3,4,6,7,8-HxCDF	0.0014	0.00043	0.0018	0.00049	0.00020	0.00054	0.00080
1,2,3,4,6,7,8-HpCDF	0.00075	0.00033	0.0012	0.00055	<0.00086	0.00056	0.0012
1,2,3,4,7,8,9-HpCDF	<0.00058	<0.00019	0.00028	<0.00050	<0.00013	<0.00060	0.00078
OCDF	<0.00081	<0.00050	0.00076	<0.00086	<0.00024	<0.0011	0.0010
WHO-TEQ PCDD/F	0.017	0.0066	0.019	0.0045	0.0022	0.0034	0.0056
Total WHO-TEQ	0.13	0.11	0.14	0.063	0.033	0.060	0.094

(2002). If these data are compared to the WHO-TEQ levels of the present study, no distinct differences can be observed, neither in the absolute concentrations nor in the congener patterns of the PCB and PCDD/F. It can therefore be concluded that major input of PCB and PCDD/F originates from atmospheric deposition – which is supposed to be similar for both lake types – rather than from direct aquatic input. As all lakes situated in the plateau receive inputs of effluents from waste water treatment plants, a significant input of PCB and PCDD/F through that pathway would be indicated by increased corresponding levels in fish from these lakes. Furthermore, cold trapping of SVOCs in the alpine lakes under investigation (mean annual temperature in the range of 0 °C) would be indicated by a relative increase of the lower chlorinated PCBs in the congener patterns compared to plateau lakes

(mean annual temperature 8 °C). However, the PCB congener patterns in fish from the different lakes are not homogenous enough to allow such a comparison which is further impeded by a series of confounding factors such as species differences and various age of individual fish.

Some of the same fish samples (*Coregonus* sp., whitefish) from Swiss plateau lakes mentioned above have also been investigated for PBDE (Zennegg et al., 2003). Based on a comparison of the concentrations with various lake parameters, the authors concluded that PBDE input into these lakes is likely to be mainly due to atmospheric deposition. This hypothesis is supported by the data of the present study: the PBDE concentrations reported in the present work are slightly below or similar to the levels reported by Zennegg et al., and concurrent PBDE congener patterns were observed.

Literature data on the occurrence of persistent organic contaminants in fish from alpine lakes is sparse. In a study on the cold trapping of SVOCs in mountain lakes, Grimalt et al. have gathered fish data from another alpine lake (Jörisee, 2519 m) located in the same region as the lakes of the present study (Grimalt et al., 2001): concentrations of HCB as well as of PCB 153 and 180 in brown trout were within the range of concentrations of the present study; the level of PCB 52 was above that range. Reference data for pesticide residues in fish from other Swiss lakes are sparse. Concentrations of DDT, HCB, γ-HCH and PCB in several fish species (mainly *Alosa fallax lacustris*, pumpkinseed) from Lago di Lugano located in the southern part of Switzerland are similar or slightly higher than the levels of the present study (Ceschi et al., 1996). Vives et al. have determined organochlorine compounds (Vives et al., 2004a) as well as PBDE (Vives et al., 2004b) in fish from alpine lakes in Europe and from a lake in Greenland. Again, corresponding concentrations and similar congener patterns (PCB, PBDE) have been observed. In contrast to organochlorine pesticides and PCB, a relatively minor temperature effect has been observed for PBDE (Vives et al., 2004b). Due to their lower vapour pressure, deposition of these compounds is expected to be preferentially particle-mediated.

Synthetic musks are used as fragrances in laundry detergents and in cosmetic products. Due to their usage pattern, they enter aquatic systems mainly via waste water or via effluents from WWTP. In contrast, the alpine lakes under investigation are exclusively fed by direct and indirect atmospheric precipitation. Though synthetic musks in fish from alpine lakes are still detectable, the concentrations are far below levels observed in fish from lakes and rivers receiving inputs of effluents of WWTP (Geyer et al., 1994; Eschke et al., 1995; Ceschi et al., 1996; Gatermann et al., 1999; Fromme et al., 2001). As the range of vapour pressures of synthetic musks is similar to the range covered by the PCB, it can be supposed that they are transferred by evaporation from water systems fed by direct input followed by wet and gaseous deposition (condensation) in alpine lakes. According to the global distillation model, accumulation is expected to be less pronounced for the more volatile polycyclic musks than for nitro musks (Grimalt et al., 2001). Vapour pressures of AHTN and HHCB (0.073 and 0.068 Pa) are indeed similar to the vapour pressure of HCB (0.063 Pa), whereas the vapour pressures of MK and MX (0.00004 and 0.00003 Pa (Peck and Hornbuckle, 2004)) are below that of PCB 180 (0.00013 Pa). Until now, no reference data have been available for levels of these compounds in fish from remote aquatic ecosystems where input of persistent organic contaminants is exclusively based on atmospheric deposition. Therefore, atmospheric distribution processes should not be overlooked when the environmental fate of synthetic musks is investigated.

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