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ScienceDirect
Chemosphere 67 (2007) S16–S21
CHEMOSPHERE
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Persistent organic pollutants, brominated flame retardants and synthetic musks in fish from remote alpine lakes in Switzerland

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Accepted 26 May 2006
Available online 17 January 2007

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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Keywords: Fish; Alpine lakes; POPs; Synthetic musks; PBDE; PCDD/F

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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0045-6535/$ - see front matter © 2006 Elsevier Ltd. All rights reserved.
doi:10.1016/j.chemosphere.2006.05.080

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.

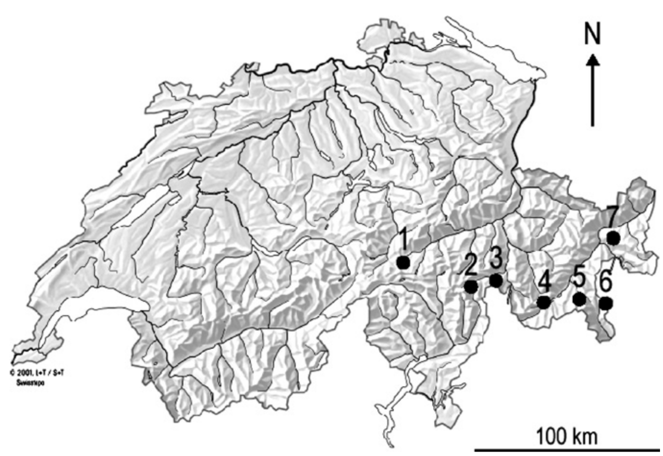

Fig. 1. Locations of the alpine lakes south eastern Switzerland (1: Lai da Tuma, 2: Laghetto Moesola, 3: Surettasee, 4: Lägh dal Lunghin, 5: Lei da Diavolezza, 6: Lagh dal Teo, 7: Lai Grond).

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,

Table 1
Parameters of alpine lakes included in the present study

Lake	Longitude (E)	Latitude (N)	Altitude (m)	Surface area (km^2)	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

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