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Distribution of PCNs, PCBs, and other POPs together with soot and other organic matter in the marine environment of the Grenlandsfjords, Norway

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Abstract

The apparently dissolved concentration of polychlorinated naphthalenes (PCNs) and three planar polychlorinated biphenyls (pPCBs) were sampled and analysed in the water column of a marine fjord system. We also measured how much of these persistent organic pollutants (POPs) were associated with suspended particles. The field observations showed that an unexpectedly high portion of the pollutants were particle-associated. The factor of deviation from model predictions had positive linear regression on the soot carbon:particulate organic carbon ratio of the particles, and on estimates of the soot–water distribution coefficient for the PCNs. The spatial distribution of surface sediment concentrations of PCNs and polycyclic aromatic hydrocarbons (PAH) were found to consistently follow the sediment content of soot (f_{SC}) to a larger extent than the bulk organic matter (f_{OC}). There were no systematic differences in the strength of correlation of sediments concentrations of other POPs, i.e. octachlorostyrene and organochlorine pesticides with sediment concentration to f_{SC} and f_{OC}. Mechanisms possible of generating these types of observations, e.g. adsorption to soot carbon in the marine particle, are discussed.

Keywords: POPs; Partitioning models; Soot; Water; Sediment

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1. Introduction

In a previous study the solid–water phase and the geographical distributions of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in sediments were observed to follow the soot carbon (SC) and, to a lesser extent, the organic matter (OM) content of marine particles in the Grenlandsfjords, Norway (Persson et al., 2002). Our current objective was to investigate if these results were valid only for PCDD/Fs, or whether they apply to a broader range of POPs. We therefore extended the analysis to include data for other persistent organic pollutants (POPs), i.e. polychlorinated naphthalenes (PCNs), polychlorinated biphenyls (PCBs), polycyclic

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aromatic hydrocarbons (PAHs), octachlorostyrene (OCS), hexachlorobenzene (HCB), hexachlorocyclohexane (HCH), 1,1,1-trichloro-2,2-bis(4-chlorophenyl)ethane (DDT), and its transformation products DDE and DDD. A primary objective of the current study was to significantly increase the number of available field data on the phase distribution of these POPs, especially PCNs and planar PCBs (pPCBs), against which hypotheses of governing processes can be tested.

Laboratory and field studies on PAHs over the past decade strongly suggest that elevated solid–water phase distribution coefficients (K_d) for PAHs, and their spatiotemporal distribution, stem from efficient interaction with highly aromatic and planar soot particles (Readman et al., 1987; Broman et al., 1990; McGroddy and Farrington, 1995; Gustafsson et al., 1997; Næs et al., 1998; Bucheli and Gustafsson, 2000; Accardi-Dey and Gschwend, 2002; Jonker and Koelmans, 2002). Such pyrogenic soot carbon has been found to make up a significant fraction (2–20%) of the total reduced carbon in marine sediments (Gustafsson and Gschwend, 1998; Middelburg et al., 1999). In this work, we test whether the solid–water and the spatial distribution of other POPs are, analogous to PAHs, influenced by the soot content of the particles. This hypothesis is evaluated by probing the distribution of POPs, SC, and OM in the marine environment of the Grenlandsfjords, Norway. This system is highly contaminated with POPs, and is therefore a suitable study area.

2. Materials and methods

2.1. The study region

The Grenlandsfjords are five joined fjords in the south of Norway (59°5′ N, 9°38′ E). The innermost fjord, Frierfjorden, has since the 1950s been receiving discharges from a magnesium production plant. These discharges contained large amounts of chlorinated organic pollutants, which were formed during the chlorination of magnesium oxide to yield water-free magnesium chloride and during the subsequent electrolysis to the pure metal (Oehme et al., 1989). Emissions of PCDD/Fs from the plant were 1–3 g TEQ/yr (TEQ = 2,3,7,8-TCDD equivalents) from 1992 until it closed down in 2002, but may have been up to three orders of magnitude higher during the earlier years of operation (Knutzen et al., 2003). PCNs from the same plant contribute to a lesser extent to the TEQ concentrations in biota in the Grenlandsfjords. PAHs are also emitted to the Grenlandsfjords, mainly from a ferro-manganese plant (Næs, 1999). Concentrations of POPs are still high in water, sediment and biota (Persson et al., 2002; Knutzen et al., 2003).

2.2. Sample collection and handling

Water samples were taken at various locations in the Grenlandsfjords and the river Skienselva/Farelva during two excursions on 29 June–1 July 1999, and 2–5 May 2000 (Table 1). We collected water column suspended particulates on glass fiber filters (Whatman GF/F, 293 mm diameter), and the apparently dissolved phase was collected in a sorbent plug made of polyurethane foam (PUF, 10 cm inner diameter). Samples of the bottom sediment from the fjords were taken 2–4 May 2000, the top 0–1 cm was collected using a Kajak corer. More details on the sampling of the water column and the bottom sediments are described by Persson et al. (2002). Bottom sediment was also sampled on the 7–8 October 1997, the surface 0–2 cm were collected, but then with a van Veen-grab (Næs, 1999).

2.3. Chemical analysis

PCNs and pPCBs in filter, sorbent and sediment samples were analysed following a methodology reported in Persson et al. (2002). For the quantification of PCNs and planar PCBs (pPCBs) an internal standard containing the $^{13}C_{12}$-labelled PCB congeners 3,3′,4,4′-polychlorobiphenyl (PCB 77), 3,3′,4,4′,5-polychlorobiphenyl (PCB 126) and 3,3′,4,4′,5,5′-polychlorobiphenyl (PCB 169) was added to the sample prior to extraction (Table 1). Laboratory blanks were also analysed, and the amounts detected were subtracted from

Table 1
Sampling year, sampled matrix, extraction method, and chemical analysis

Analyte	Matrix	Year	Extraction	Internal standard	Chemical analysis
PCN, PCDD/Fs, co-planar PCB	Suspended particles on filter, and matter sorbed in PUF	1998–2000	Toluene, Soxhlet > 24 h	$^{13}C_{12}$-labelled PCBs and PCDD/Fs	HRGC-HRMS[a]
α-HCH, γ-HCH, OCS, DDD, DDE, DDT, bulk PCBs	Bottom sediment	1997	Cyclohexane/acetone ultrasonification	PCB 53	GC-ECD[b]
PAHs	Bottom sediment	1997	Pentane	Deuterated PAHs	GC-MSD[b]

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