

Historical trends in persistent organic pollutants and metals recorded in sediment from Lake Ellasjøen, Bjørnøya, Norwegian Arctic

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Levels of Polybrominated diphenyl ethers are increasing in sediment from a Norwegian, Arctic lake.

Abstract

Ellasjøen receives contaminants from both atmospheric and biological transport (seabird guano). Historical trends of selected persistent organic pollutants and metals were determined by interpretation of down-core contaminant profiles in conjunction with sediment layer ages, determined by the ²¹⁰Pb method. Polychlorinated biphenyls (PCBs) and dichloro-diphenyl-trichloroethane and metabolites (DDTs) increased from about 1930 until 1970 (max. sum PCBs 71.8 ng/g dry weight (dw), max sum DDTs 4.0 ng/g dw), declining afterwards to 46.4 ng/g dw and 1.6 ng/g dw, respectively. Polybrominated diphenylethers (PBDEs) concentrations are increasing in Ellasjøen, reaching 0.73 ng/g dw in 2001. Hexabromocyclododekane (HBCD) was measured only in sediment from 1–2 cm depth (0.43 and 3.88 ng/g dw of α - and γ -HBCD, respectively). Most metals (As, Cd, Co, Ni and Zn) showed a similar pattern as PCBs. No clear trends were observed for Cr, Pb or V. Mercury (Hg) has increased gradually since the beginning of the industrial era.

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1. Introduction

During recent years, multidisciplinary studies have been carried out on Bjørnøya (Bear Island) in the central Barents Sea (74°30'N, 19°00'E) elucidating the fate and presence of persistent organic pollutants (POPs) in this pristine Arctic environment (Bustnes et al., 2000; Evenset et al., 2004, 2005; Henriksen et al., 2000). The studies have revealed that Ellasjøen, a lake located in the southern mountainous part of Bjørnøya (Fig. 1) is a “hot-spot” for contamination. Even though there are no industrial or agricultural activities on the island, sediment

and biota from Ellasjøen contain high concentrations of POPs (Evenset et al., 2004, 2005). Seabirds (mainly kittiwake (*Rissa tridactyla*), little auk (*Alle alle*) and glaucous gull (*Larus hyperboreus*)), feeding in the marine environment, deposit large amounts of guano (excrement) directly into the lake or in the catchment area of the lake. Guano has recently been identified as an important transport medium for contaminants from the marine to the limnic environment (Blais et al., 2005; Evenset et al., 2004, 2005). Additionally, Ellasjøen is located at the base of a mountainous terrain, and receives 3–4 times more precipitation than the flatter parts of Bjørnøya (Evenset et al., submitted for publication). The high precipitation rate probably leads to a relatively high deposition of air transported contaminants. These two mechanisms have caused some of the highest POP-concentrations ever recorded in remote areas (AMAP, 2004).

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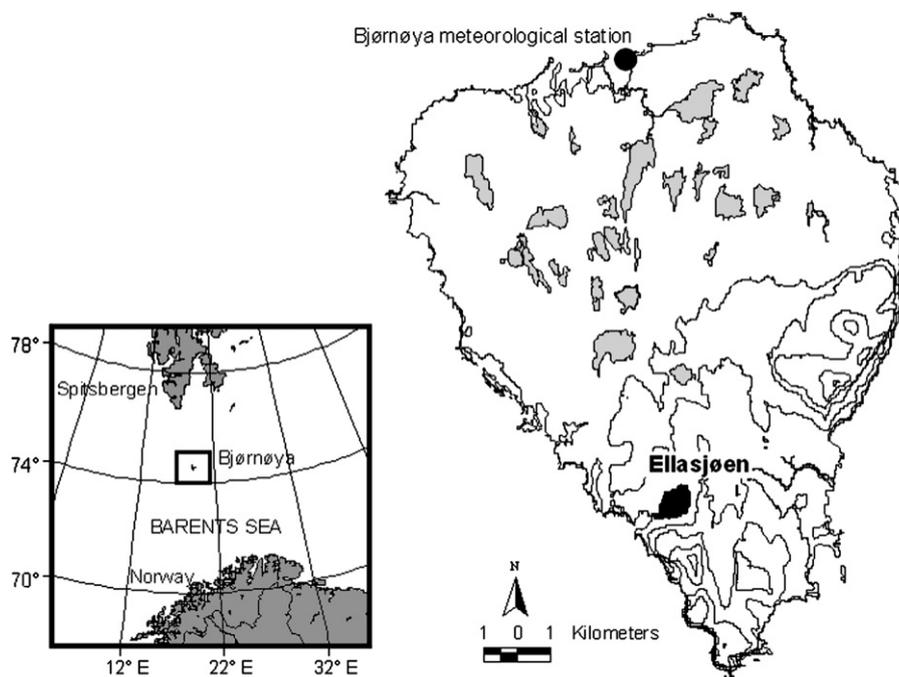


Fig. 1. Bjørnøya (74°30'N, 19°00'E) in the central Barents Sea, Norway. Lake Ellasjøen is located on the southern, mountainous part of the island.

The aim of the present investigation was to evaluate the historical development of contaminant supply to Ellasjøen over the entire industrial era. One method that has often been used to determine historical changes in contaminant deposition is the analyses of sediment cores. However, in most Arctic lakes the sedimentation rate is low compared to lakes from temperate regions. Due to low inputs of organic matter from the terrestrial environment (little vegetation in the catchment area) and low primary production, also the input of organic carbon to the sediment is usually low in Arctic lakes. This makes them less significant reservoirs for hydrophobic compounds than temperate lakes (Diamond et al., 2005; Macdonald et al., 2000). However, due to nutrient input from seabirds, Ellasjøen is a relatively productive Arctic lake. Sediment cores from Ellasjøen were therefore considered to be well suited for studies of historical changes in contaminant input.

There is little published information on concentrations or fluxes of contaminants in lake sediments from the European Arctic, but dated sediment cores have been used to infer the depositional history of contaminants in Canadian Arctic lakes (Muir et al., 1995, 1996) and in European alpine lakes (Fernandez et al., 1999, 2000; Grimalt et al., 2001). The results from these studies indicate that concentrations of phased-out or heavily restricted contaminants, like polychlorinated biphenyls (PCBs) and dichloro-diphenyl-trichloroethane and metabolites (DDTs), now have stabilized or are decreasing. In contrast to e.g. PCBs, many brominated flame retardants (BFRs), like some polybrominated diphenyl ether (PBDE) formulations and hexabromocyclododecane (HBCD), are still in production and use. PBDEs belong to a group of additive BFRs widely used in electronic equipment, insulation material and furniture, while HBCD is extensively used in Europe in expanded and extruded polystyrene

for thermal insulation foams, building and construction, and furniture. However, data on the environmental concentrations of these chemicals, especially HBCD, are still scarce. The occurrence of PBDEs and HBCD in remote air samples (Alaee et al., 1999; Remberger et al., 2004) implies that these compounds have the potential for long-range atmospheric transport. Historical reconstructions of BFR levels in lake sediments from temperate areas (Hoh et al., 2005; Song et al., 2004, 2005; Zegers et al., 2003; Zhu and Hites, 2005) generally indicate increasing concentrations towards surface sediments. The same trend seems to occur in Arctic lakes (Malmquist et al., 2003), but few data series are available to confirm this finding.

Some Arctic areas have also been contaminated with metals generated in other regions of the Northern Hemisphere (Dietz et al., 1998). Different trends have been reported for different metals, depending on usage pattern, discharges, transport pathway and metal properties (i.e. sediment affinities). In general, metal concentrations in Arctic lakes have increased since the onset of the industrial revolution (Dietz et al., 1998). In some studies a slight reduction in the concentration of some metals has been recorded in recent years (Dietz et al., 1998; Hermanson, 1993). Most information is available for lead (Pb), cadmium (Cd) and mercury (Hg), due to concerns for their high toxicity, relatively high volatility and large volume discharges to the atmosphere from industrial processes (Barrie et al., 1992). An enrichment of metals in soil samples from areas affected by seabird guano has also been documented (Headley, 1996; Godzik, 1991).

In the present study we examine the sedimentary profile and historical trend in supply of selected phased-out and present-used POPs and metals to Ellasjøen. Historical changes in contaminant input were determined by mapping the down-core

depth distribution of selected compounds with down-core sediment ages determined by ^{210}Pb sediment geochronology (Robbins, 1978).

2. Materials and methods

2.1. Sampling

Four replicate sediment cores (10 cm diameter), with lengths from 24.5–33.0 cm were collected in April 2001 from the deepest part of Ellasjøen (34 m depth), using a gravity coring device deployed from the ice. Cores having an undisturbed sediment-water interface were extruded and sectioned into 1 cm intervals on site. The sediment was transferred to glass-jars for POPs-analyses and plastic-containers for sediment dating, analyses of TOC and metals. All samples were frozen to $-20\text{ }^{\circ}\text{C}$ immediately after sampling and kept frozen until analysis. One core was used for each of the following determinations: sediment ages, POPs, metals and total organic carbon (TOC).

2.2. Sediment geochronology

Down-core profiles of ^{210}Pb and ^{137}Cs (Robbins, 1978) were used to assign a constant sediment accumulation rate to the Ellasjøen sediment core. Radio-nuclide determinations were performed on the upper 10 cm (^{210}Pb and ^{137}Cs) and selected deeper slices (^{210}Pb only) of the core by the Institute of Oceanology, Poland. 0.2 grams of sediment from each depth interval, spiked with ^{209}Po as a tracer, was digested with hydrofluoric acids (Flynn, 1968). Polonium isotopes were extracted using ion exchange chromatography. Purified samples were subsequently deposited on silver discs for quantification of ^{210}Po (daughter of ^{210}Pb) and ^{209}Po by alpha spectrometry. Subsequently, individual sediment slices were prepared for non-destructive ^{137}Cs counting using a HPGe detector calibrated with IAEA certified reference material. Total organic carbon (TOC) was also determined on selected depth intervals using a Leco IR 212 carbon analyzer.

2.3. POP analyses

18 PCB congeners (CB 28, 52, 99, 101, 105, 118, 126, 128, 138, 149, 153, 156, 169, 170, 180, 183, 187 and 194), DDT and metabolites (*o,p'*-DDD, *o,p'*-DDE, *p,p'*-DDE, *o,p'*-DDT and *p,p'*-DDT), 10 PBDE-congeners (BDE 28, 47, 71, 77, 99, 100, 138, 153, 154 and 183) and HBCD (α -, β -, and γ -isomers) were quantified by the Norwegian Institute for Air Research. All standard materials were purchased from Promochem, Wesel, Germany.

2.3.1. Extraction and clean-up

Approximately 10 g of freeze-dried sample was spiked with 20 μl of internal standard solution containing between 77 and 500 $\text{pg}/\mu\text{l}$ of $^{13}\text{C}_{12}$ -*p,p'*-DDE, $^{13}\text{C}_{12}$ -*p,p'*-DDT, $^{13}\text{C}_{12}$ -labeled PCBs no. 28, 52, 101, 118, 153 and 180, $^{13}\text{C}_{12}$ -labeled PBDEs no. 28, 47, 99, 153 and 183, as well as 1.3 $\text{ng}/\mu\text{l}$ D_{18} -labeled HBCD.

Soxhlet extraction of the sediment samples was performed with 150 mL toluene at reflux for 8 h. The toluene extract was concentrated to 0.5 mL in a Turbovap evaporator (ZyMark, Hopkinton, MA, USA) while changing the solvent to dichloromethane. Sulphur was removed by gel permeation chromatography, using a dual pre-packed Envirogel system (Bio-Beads S-X3 resins, Waters, Milford, MA, USA) at a flow rate of 5 mL/min dichloromethane. The fraction between 75 and 106 mL was collected and concentrated to 0.5 mL (Turbovap) while changing the solvent to cyclohexane. The extract was additionally cleaned-up using a florisil column packed with 1.5 g pre-treated (heated 12 h at $450\text{ }^{\circ}\text{C}$) and deactivated (0.5% (w/w) water) florisil (particle size: 0.15–0.25 mm) and 2 g of anhydrous granulated sodium sulphate on top. The column was pre-washed with 10 mL *n*-hexane/dichloromethane (3:1; v/v) before the sample was applied. All analytes were eluted with 8 mL *n*-hexane/dichloromethane (3:1; v/v). The extract was concentrated to approximately 0.1 mL (Turbovap and nitrogen stream) and the recovery standard octachloronaphthalene (20 μl of a 0.6 $\text{ng}/\mu\text{l}$ solution in iso-octane) was added.

2.3.2. Analysis and quantification of PCBs, PBDEs and DDT

Gas chromatography coupled to mass spectrometry (GC/MS) was employed for separation and quantification of all POP-analytes, with the exception of HBCD. Samples were analysed on a Mega II 8065 gas chromatograph (Fisons, Milan, Italy) coupled to an MD800 quadrupole mass spectrometer (Finnigan MAT, San Jose, CA, USA) operated in the electron impact mode. Samples were injected onto a DB5MS capillary column (30 m length, 0.25 mm i.d., 0.25 μm film thickness, J&W Scientific, Folsom, CA, USA). Separation was performed using helium at a constant column head pressure of 80 kPa, applying the following temperature program: $70\text{ }^{\circ}\text{C}$ (hold time 2 min), then at $15\text{ }^{\circ}\text{C}/\text{min}$ to $180\text{ }^{\circ}\text{C}$ and at $5\text{ }^{\circ}\text{C}/\text{min}$ to the final temperature of $280\text{ }^{\circ}\text{C}$ (hold time 15 min). The GC/MS interface temperature was held at $250\text{ }^{\circ}\text{C}$ and the ion source temperature at $220\text{ }^{\circ}\text{C}$. The mass spectrometer was operated in selected ion monitoring mode (SIM).

2.3.3. Analysis and quantification of HBCD

A sample volume of 10 μL was injected into the separation system using a 1100 Series HPLC system (Agilent Technologies, Palo Alto, CA, USA). HPLC separation was carried out on a C_{18} column (X-Terra MSC18: 3.5 μm particles, 150 mm length, 2.1 mm i.d., Waters, Milford, MA, USA) applying a flow rate of 250 $\mu\text{L}/\text{min}$ of methanol (A); acetonitrile (B); and water (C). The following gradient was used: 0 min, 10% A/2.5% B/87.5% C; 0–12 min, linear change to 88% A/12%B; followed by 12 min isocratic elution with 88% A/12%B. A time-of-flight (TOF) mass spectrometer (LCT, Micromass, Manchester, England) was used employing electrospray ionization in the negative ion mode (ESI(-)) and the following parameters; cone voltage, -13 V ; desolvation temperature, $250\text{ }^{\circ}\text{C}$; source temperature, $110\text{ }^{\circ}\text{C}$; nitrogen cone gas flow, 15 L/h. Mass spectra were registered in full scan mode (mass range m/z 600 to 700).

2.3.4. Quality control

For each compound a quantification mass and a control mass relating the area proportions to the measurements of standard compounds were acquired. Quality assurance of the analytical method was carried out by measurements of laboratory blanks and standard reference material. Because of the lack of commercial standard materials for BFR in sediments a sediment origin from the EU project BROCC "Biological Reference Materials for Organic Contaminants" (G6RD-CT-2001-00518) was used to control the performance of the analytical method. The limit of detection (LOD) was calculated as 3 times signal/noise in the respective solvent blank analyses. The detection limits are therefore a function of the mass extracted and varied between samples (0.01–0.3 $\text{ng}/\text{g dw}$). All values below LOD were marked as non-detects. The Limit of quantification (LOQ) was calculated as 10 times noise in the respective sample chromatograms, when no lab-blank contamination occurred. In case of lab-blank contamination (only the case for BDE 47 and CB153) 10 times signal area in the lab blank was used as LOQ. Samples with recovery below 50% were not accepted for quantification. The average recovery for the ^{13}C labeled compounds lay between 68 and 72% for the PBDEs and 65 and 85 for the PCBs.

2.4. Metal analyses

The metals arsenic (As), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), mercury (Hg), nickel (Ni), lead (Pb), vanadium (V) and zinc (Zn), were quantified by Analytica, Sweden. Dried sediment samples ($50\text{ }^{\circ}\text{C}$ for 48 h) were mixed with 7 M HNO_3 and sealed in teflon containers for microwave digestion (10 min at 375 W and 55 min at 275 W). Cooled samples were transferred to test tubes and diluted to 10 mL. Metals were quantified with Plasma emission spectrometry (ICP-AES) and plasma mass spectrometry (ICP-QMS). Analytical quality was confirmed through analyses of certified reference sediment (LGC-Promochem). Detection limits varied between 0.01 (Cd and Co) and 2 (V) $\text{mg}/\text{kg dw}$, and the analytical uncertainty was between 16 (Pb) and 50% (Hg).

3. Results

3.1. Sediment age determination

The ^{210}Pb geochronology method assumes that ^{210}Pb , in excess of its parent isotope ^{226}Ra , varies exponentially with

sediment depth normalized for sediment porosity changes. An average sediment accumulation rate for the entire core is then determined from the slope of the least-squares fit to an exponential plot of excess ^{210}Pb against sediment depth. The constant sediment accumulation rate determined for the Lake Ellasjøen core was 0.7 mm/yr ($R^2 = 0.98$) which corresponds to a flux of 620 g/m²/year (Carroll and Lerche, 2002). This result was verified by data from the independent radionuclide proxy, ^{137}Cs (Table 1). ^{137}Cs was not detected below the 4–5 cm sediment depth interval in accordance with the first appearance of ^{137}Cs in the 1940's at the beginning of nuclear weapons testing in the European Arctic. A surficial peak in ^{137}Cs content occurred in 1986 (Chernobyl accident) while a sub-surface peak corresponds to the peak in nuclear weapons testing activities in the early 1960's. The correspondence between ^{137}Cs and ^{210}Pb profiles indicates that in Ellasjøen, the rate of sediment accumulation and/or rate of ^{210}Pb supply to the lake bottom varied little with time. The organic carbon concentration was highest in the surface sediment, constituting 6% of the dry weight. Due to natural decomposition processes, the TOC-concentration decreased downwards in the core (Table 1).

3.2. Historical trends in POP-concentrations

3.2.1. PCBs

Relatively high concentrations of PCBs were measured in the upper sediment layers from Ellasjøen (Fig. 2A). The concentration of PCBs increased until approximately 1965–1970 (max. sum PCB = 71.8 ng/g dw, 1422 ng/g OC). At the beginning of the 1970's the concentration started to decrease, reaching 46.4 ng/g dw (779 ng/g OC) in surface sediments. PCBs were detected back to the period 1886–1897, i.e. before PCBs were taken into use. However, the concentrations below 5–6 cm (median age 1928) were low (<2 ng/g dw) compared to concentrations in the upper sediment layers.

The dominating PCB congener in all sediment layers was CB 153 (hexa-chlorinated) followed by CB 138 (hexa-chlorinated) and CB 180 (hepta-chlorinated). The congener pattern was relatively consistent throughout the length of the core, although

Table 1
 ^{210}Pb derived sediment ages corresponding to the mid-point of each depth interval

Depth interval (cm)	TOC (%)	Median age	Range	^{137}Cs content (Bq/kg)
Surface		2001		
0.5	6.0	1994	1987–2001	137 ± 7
1.5	5.9	1980	1973–1987	8.8 ± 3.2
2.5	5.1	1966	1959–1973	23.9 ± 4.5
3.5	4.4	1953	1946–1959	7.2 ± 3.5
4.5	3.8	1940	1934–1946	4.1 ± 1.6
5.5	3.5	1928	1921–1934	<lod
6.5	3.5	1915	1909–1921	<lod
7.5	3.3	1903	1897–1909	<lod
8.5	3.3	1892	1886–1897	<lod
9.5	3.3	1881	1875–1886	<lod

The results compare well with the observed sub-surface peak in ^{137}Cs associated with the peak time period of nuclear weapons testing (shaded and bold). <lod = at or below the detection limit (2 Bq/kg).

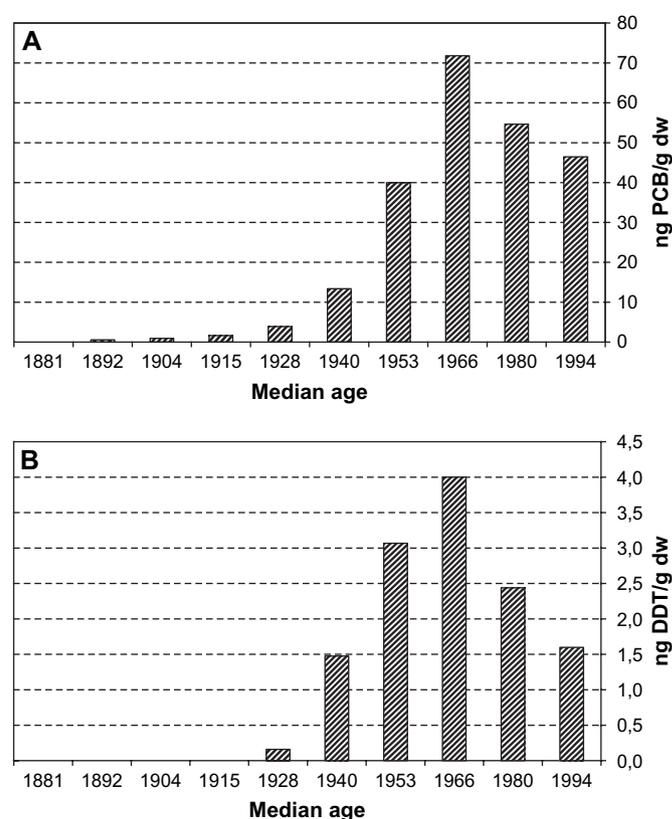


Fig. 2. Concentrations of PCBs (sum of 18 congeners, A) and DDTs (B) in a sediment core from Ellasjøen, Bjørnøya, April 2001.

there appeared to be a slight reduction in the share of higher chlorinated congeners down through the core (Fig. 3).

3.2.2. DDTs

The same trends as for PCBs was revealed for DDTs, with a rapid increase in concentrations (to 4 ng/g dw, 78.5 ng/g OC) until about 1965–1970, and thereafter a decrease to 1.6 ng/g dw (26.8 ng/g OC) in surface sediments. DDTs were detected as far back as approximately 1930 (Fig. 2B). The stable metabolites, *o,p'*-DDE and *p,p'*-DDE, constituted about 90% of sum DDT in all samples.

3.2.3. Brominated flame retardants

PBDEs could only be detected in the upper 4 cm of the sediment core, corresponding to about the last 50 years. The sumPBDE-concentration in the surface sediment was 0.73 ng/g dw (12.2 ng/g OC, Fig. 4). PBDE-concentrations were several times lower than the PCB-concentrations in the same sediment slices, but contrary to PCB-concentrations the concentrations of PBDEs have continued to increase towards present time. More precisely, there has been a more than five fold increase in PBDE-concentrations in sediments from Ellasjøen over the last 50 years. In the slices with median age 1966 and 1980, PCB-concentrations were about 300 times higher than PBDE-concentrations (although fewer PBDE- than PCB-congeners were included), while the ratio between the two compound groups decreased to about 140 in the slice with median age 1980 and to about 65 in the surface sediment.

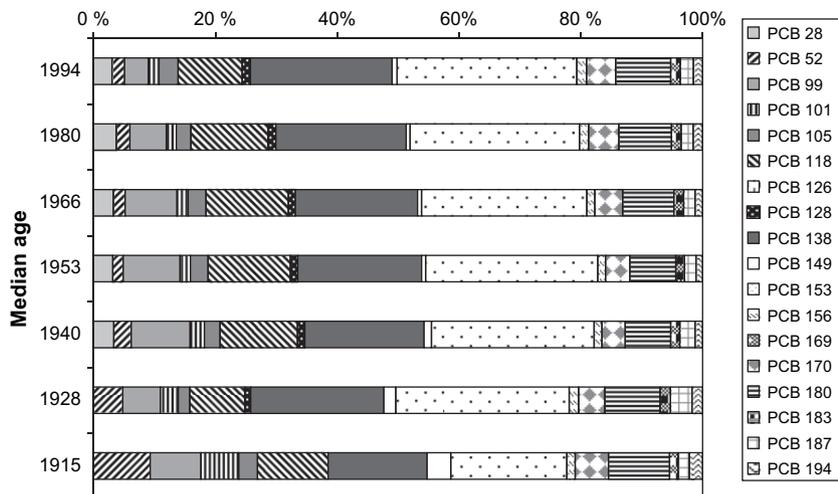


Fig. 3. Relative share of PCB-congeners in a sediment core from Ellasjøen, Bjørnøya, April 2001. PCBs were detected as far back as in sediment with an estimated median age of 1892, but due to very low concentrations (sum PCB <1 ng/g) the profiles for the lowest layers are not included in the figure.

Analyses of 10 BDE-congeners resulted in the detection of only BDE 28, 47, 99, 100 and 153. The dominant congener was BDE 47, followed by BDE 99, 28 and 100 (Fig. 4). The relatively heavy congener BDE 153 was only found in the surface sediment.

HBCD was only detected in the depth interval from 1–2 cm (median age 1980). Only the α - and γ -HBCD isomers (0.43 and 3.88 ng/g dw or 7.3 and 65.8 ng/g OC, respectively) were measured in this depth interval. The concentration was considerably higher (about 11 times higher) than the PBDE concentration in the same slice. β -HBCD concentration was lower than the detection limit (<0.06 ng/g dw) in all samples.

3.3. Historical trends in metal concentrations

The deepest layer from the core (26–28 cm) was defined as a “reference” layer, since this layer was deposited in pre-industrial times (>300 years ago). Concentrations of most metals (As, Cd, Co, Cr, Cu, Ni and Zn) increased from pre-industrial times to approximately the 1970’s (2–3 cm) declining thereafter. No clear temporal trends were observed for Pb,

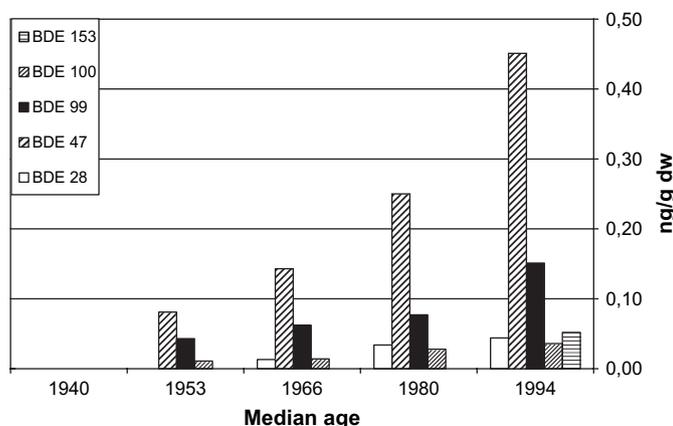


Fig. 4. PBDEs (ng/g dw) in the upper 5 cm of a sediment core from Ellasjøen, Bjørnøya, April 2001.

Cr and V, while the Hg concentration increased slowly from pre-industrial times until present (Table 2).

Enrichment factors (i.e. the ratios between levels in the sediment section with the highest metal concentrations (2–3 cm) and the reference sediment (ratio: 2–3 cm/reference), as well as the ratio between surface sediment and reference sediment (ratio: surface/reference)) were used to investigate temporal trends in metal concentrations. The highest enrichment factors were found for Co (4.85 and 3.92) and Ni (3.26 and 2.76). These metals also showed a marked increase in concentrations around 1920 (Table 2). Relatively high enrichment factors were also found for Zn (2.21 and 1.90) and Cd (1.96 and 1.67).

4. Discussion

The patterns and trends in POPs and metals observed in the sediment record from Ellasjøen are the result of three main components, one relating to source regions (emissions), one relating to a biological vector, namely seabirds (Evenset et al., 2004, 2005) and the third relating to fate processes that the contaminants undergo after entering the lake. Anaerobic degradation has been suggested to be an important removal process for some POPs in Arctic lake sediments (Muir et al., 1995), and may explain lower concentrations observed in deeper sediment layers. However, this can not explain the decreasing concentrations of PCBs and DDTs toward the sediment-water interface.

In addition to acting as a vector transferring contaminants between marine and limnic systems, seabirds add nutrients in the form of seabird guano to Ellasjøen. This nutrient addition has resulted in a relatively high production in the lake, leading to high organic carbon contents (6%) in the lake sediments in Ellasjøen compared to other Arctic lakes (1.2–6%, mean 3%) (Muir et al., 1995; Skotvold et al., 1997). A high production also result in the relatively high sediment accumulation rate (0.7 mm/year or 620 g/m²/year) for Ellasjøen as compared to other Arctic lakes (120–500 g/m²/year) (Muir et al., 1995; Appleby, 2004). Organic-rich sediments in turn, act as a contaminant sink through scavenging of contaminants

Table 2
Metal-concentrations (mg/kg dw) in a sediment core from Ellasjøen, Bjørnøya, April 2001

Median age	1994	1980	1966	1953	1940	1928	1915	1904	1892	1881	Pre-ind. (ref.) (>300 y)	Ratio: surface/ref.	Ratio: 2–3 cm/ref.
As	6.98	7.78	8.64	8.40	7.28	6.61	5.58	5.58	5.79	5.71	5.29	1.32	1.63
Cd	2.20	2.35	2.57	2.50	2.12	1.46	1.32	1.21	1.24	1.26	1.31	1.67	1.96
Co	42.3	46.9	52.4	52.1	32.8	20.1	12.5	10.7	10.4	9.95	10.8	3.92	4.85
Cr	14.4	16.3	17.0	14.9	15.1	17.9	14.2	16.9	15.5	12.3	12.6	1.14	1.35
Cu	34.4	35.1	36.3	36.8	37.6	30.1	30.1	28.1	27.7	26.9	26.6	1.29	1.36
Hg	0.155	0.134	0.124	0.117	0.107	0.102	0.090	0.124	0.104	0.097	0.083	1.87	1.49
Ni	52.5	57.9	62.0	59.7	47.1	30.4	22.3	21.1	20.7	19.2	19.0	2.76	3.26
Pb	86.6	90.7	89.2	87.2	86.0	81.4	82.7	83.5	89.6	95.1	77.8	1.11	1.15
V	18.1	20.3	20.8	18.5	18.2	21.2	17.3	19.9	18.7	15.8	15.9	1.14	1.31
Zn	373	391	434	411	314	236	201	193	205	213	196	1.90	2.21

Enrichment factors for surface sediment and sediment with median age 1966 (2–3 cm) are also given.

from the dissolved to the particulate phase, and due to this process sediment from Ellasjøen is an area of enhanced POPs and metal deposition. The data generated through this investigation allow: i) reconstruction of contaminant introductions to the lake; ii) the assessment of how the observed patterns reflect available information on known sources and emissions to the region; and iii) a comparison of contaminant fluxes from this area with available information on other areas in the Arctic.

4.1. Historical changes in POP-concentrations

4.1.1. PCBs

The levels of PCBs in sediment from Ellasjøen are high compared to levels measured in other Arctic lakes (Muir et al., 1996; de March et al., 1998), and even compared to levels in sediment from lakes closer to industrialised areas. For instance, an average PCB-concentration (sum of 19 congeners) of 3.6 ng/g dw (compared to 46.4 ng/g dw in Ellasjøen) was measured in surficial sediment from Lake Superior in Canada collected in 2001/2002 (Song et al., 2004). The data from Ellasjøen show that the concentrations of PCBs started to decrease after they were banned or strongly restricted in a number of countries, i.e. around 1970–1980 (de March et al., 1998). A decreasing trend is confirmed through a comparison of levels in surface sediment collected from Ellasjøen in 1996 with levels in surface sediment collected in 2001. Evenset et al. (2004) reported a PCB-concentration (sum of 7 congeners) of 60 ng/g dw in surface sediment collected from Ellasjøen in 1996. The sum of the same 7 congeners from the present study amounts to 36.6 ng/g dw.

Low concentrations of PCBs were measured down to 8–9 cm from the sediment-water interface. According to the sediment dating, this sediment is from before PCBs were used. The same picture was also apparent for the other measured contaminants. The occurrence of contaminants in pre-industrial core segments is a problem that has been encountered also in other studies (Muir et al., 1995; Sanders et al., 1992; Rose et al., 2004). Several reasons have been suggested for this: 1) molecular diffusion; 2) sediment coring artifacts; 3) bioturbation; or 4) analytical artifacts. In the present study the first three explanations are most likely, since no POPs were found in the lowest sections of the core

(30–32 cm), and no contamination of blank samples occurred in the laboratory. Molecular diffusion is probably also of minor importance since only a small fraction of the contaminants will be in the pore water, compared to the fraction bound to sediment particles. However, the surface sediment layer was soft and unconsolidated, and it was therefore difficult to prevent small amounts from leaking to deeper layers during the extrusion of the core. Bioturbation may also have caused some mixing of the sediment. There are few benthic species living in Ellasjøen (mainly chironomids) (Klemetsen et al., 1985) and most of the benthic life is limited to the upper 2 cm of the sediment. Still, bioturbation may cause and have caused some sediment mixing in the past, when the deep core layers were closer to the sediment-water interface.

The dominating PCB congeners in the sediment from Ellasjøen were CB 153, 138 and 180. Generally, abiotic samples from the Arctic environment have a low proportion of higher chlorinated congeners and a high share of the lower chlorinated, more volatile congeners. According to the “global fractionation” hypothesis (Wania and Mackay, 1993) POPs with relatively high mobility (e.g. PCBs with 1–4 chlorine atoms) will be more predominant in polar regions than POPs with lower mobility (e.g. PCBs with 4–8 Cl-atoms or DDTs). This has been verified through a number of studies in polar and mid-latitude regions (AMAP, 2002). For example, Muir et al. (1996) found that di-, tri- and tetra-chloro congeners predominated in sediment samples from lakes in the Canadian Arctic, especially in samples from Amituk lake (75°03'N, 93°46'W) which is located close to the latitude of Bjørnøya (74°30'N, 19°00'E). In Muir's and coworkers study di/tri-CBs represented from 32–57% of sum PCBs in the high Arctic sediments. No di-CBs were analyzed in sediment from Ellasjøen, but the tri-CBs measured constituted no more than 2.3% of sum PCB. Hexa-CBs dominated in the sediment from Ellasjøen, with a relative share of 47%. The differences between Bjørnøya and the Canadian Arctic may partly be explained by differences in sources or distance from source areas, since Bjørnøya is located closer to some of the important source areas in Europe than Amituk Lake, which is also believed to be primarily affected by sources from Europe following a transpolar trajectory. However, input from seabirds may also explain the congener pattern in

sediment from Ellasjøen. Contaminants transported by seabirds or excreted in seabird guano, have passed through parts of the marine food chain and have gone through one or more bioaccumulation cycles. These bioaccumulation processes have altered the contaminant pattern leading to an enrichment of the most persistent contaminants, like the higher chlorinated PCB-congeners. Analyses of guano from seabirds caught around Ellasjøen have verified that the PCB-pattern in guano is very similar to the pattern found in the sediment samples (Evenset et al., submitted for publication; Herzke et al., 2003). This indicates that input of guano is affecting both sediment concentration and congener pattern of PCBs in Ellasjøen.

4.1.2. DDTs

The DDT-concentrations were relatively high in the upper sediment layers compared to levels measured in other Arctic lakes (de March et al., 1998; Muir et al., 1995). The DDT-concentration showed the same temporal trend as the PCB-concentration, with the highest concentration appearing in the layer with median age 1966. The commercial production of DDT started in the 1940s and in Europe and North-America the use was heavily restricted during the 1960s and phased out during the mid-1970s. Thus, the trend in DDT-concentrations found in the core from Ellasjøen agrees well with the usage pattern. Further confirmation of this trend is seen by comparing the surface sediment concentration of DDTs in the present study (1.6 ng/g dw) with the concentration in surface sediment collected in 1996 (6.9 ng/g dw) (Evenset et al., 2004).

The persistent metabolite of DDT, *p,p*-DDE was dominant in all samples, indicating input of heavily weathered DDT. *p,p*-DDE is also the dominant DDT-related compound in guano from the seabirds that occur in high numbers around Ellasjøen (Evenset et al., submitted for publication; Herzke et al., 2003).

4.1.3. BFRs

In contrast to PCBs and DDTs that are exhibiting a decreasing trend in recent time, the concentration of PBDEs is increasing in Ellasjøen. The results indicate that the PBDE-concentration in sediment from Ellasjøen has doubled every 15 years for the last 30 years, but the concentration of PBDE is still very much lower than the concentration of PCB and about half the DDT-concentration. An increasing trend in PBDE-concentrations has also been reported from the Great Lakes (Song et al., 2004, 2005; Zhu and Hites, 2005) and Western Europe (Zegers et al., 2003). Thus, the PBDE-concentrations in Ellasjøen follow the same trend that has been reported from lakes located closer to heavily populated and industrialized areas, and in fact the PBDE-concentration in surface sediment from Ellasjøen (0.73 ng/g dw) was in the lower range of what has been reported from the Great Lakes (sum of 9 PBDE congeners from 0.5–4 ng/g dw) (Song et al., 2004, 2005).

The dominant congener in all samples was BDE 47, followed by BDE 99, 28 and 100. The same congeners are also

most prevalent in the commercial penta-BDE formulation (Boon et al., 2002; Sjödin et al., 1998), which is now banned by the European Union and North-America together with octa-mixtures. Also the congener pattern for PBDEs closely resembled the pattern in seabird guano (Akvaplan-niva, unpublished data).

HBCD exhibited an unusual pattern which we currently cannot fully explain. No HBCD was detected in surface sediments, a surprising result given that HBCD is still in use (de Wit, 2002). In fact, HBCD was only measured in one depth interval corresponding to a median age of 1980 (sum HBCD 4.31 ng/g dw). The concentration of HBCD in this segment was about 11 times higher than the PBDE-concentration in the same segment. Although the γ -HBCD isomer was dominant, the α -isomer was present in quantifiable amounts; a pattern that is consistent with results from the limited number of studies reporting levels of individual HBCD-isomers in sediment (Birnbaum and Staskal, 2004).

4.2. Historical trends in metal concentrations

Metals are naturally occurring in the environment, and therefore the local geology will affect metal concentrations in soil and sediments. The bedrock on Bjørnøya is mainly composed of Devonian, Carboniferous and Permian sandstones, shales, coal seams, conglomerates and fossiliferous limestones (Horn and Orvin, 1928; Worsley and Edwards, 1976). Differences in geology complicate comparisons of metal levels between different areas. A common method to interpret results from metal analyses is therefore to compare levels in pre-industrial sediment (here sediment from 26–28 cm was used) with levels in sediment closer to surface.

The concentrations of As, Cd and Zn increased steadily from the start of the 20th century and to approximately the 1970s. Thereafter the concentrations decreased. The non-ferrous metal industry is one the largest anthropogenic source for atmospheric As, Cd and Zn. Generally, the European emissions of these metals have decreased during recent years (Dietz et al., 1998). Thus the flux of As, Cd and Zn in Ellasjøen reflect the discharge pattern for these metals quite well.

Co and Ni exhibited a somewhat unique pattern compared to the other metals, since their concentration increased dramatically around 1920 (Co 4.2 times from 1915 to 1966 and Ni 2.8 times from 1915 to 1966). During the same period also the Cu-concentration increased (1.2 times). The reason for this is not known, but the increases coincide or slightly precede (possibly an artifact of downwards contamination during core extrusion) the initiation of production at the large copper- and nickel- smelters in Nickel and Zapolyarny in Northwest Russia. High levels of Co, Cu and Ni in Eastern Finnmark, Northern-Norway, have been connected to discharges from this industry (Rognerud and Fjeld, 2001; Steinnes et al., 2005). During recent years the concentrations of these metals have decreased in sediment from Ellasjøen. The peak production at the large smelter on the Kola Peninsula was in 1980 (Wright and Traaen, 1992). Thus the decreasing sediment concentrations of Co, Ni and Cu in recent sediment from

Ellasjøen may reflect this associated decrease in emissions. However, previous investigations have concluded that discharges of these metals mainly lead to local contamination (Rognerud and Fjeld, 2001; Steinnes et al., 2005). The results from the present study suggest that also areas further away from the source receive Co and Ni-, and to a smaller degree Cu-contamination.

No clear temporal trends were obvious for Cr, Pb or V. In some studies from Arctic areas an increasing Pb-concentrations was measured after the introduction of leaded gasoline on the northern hemisphere (Hermanson, 1993), with a concomitant reduction after the phasing out of leaded gasoline (AMAP, 2002). In other studies no clear temporal trends for Pb have been identified (Outridge et al., 2002; Rognerud et al., 1993). The natural background levels of Pb are high on Bjørnøya (Skotvold et al., 1997), and the natural levels may be masking any changes superimposed by anthropogenic inputs. Another explanation for the low Pb-input may be that Bjørnøya has mainly received atmospheric inputs from areas with low Pb-emissions. It has been documented that high emissions of Pb have occurred in Western-Europe, but the results for the other metals suggest that Bjørnøya to a large degree has been affected by aerosols from Northwest Russia, i.e. areas with lower Pb-emissions (Dietz et al., 1998).

V and Cr are also metals that are subject to long-range atmospheric transport and an increase in the concentration from pre-industrial times would therefore be expected. However, no clear increases in concentrations from pre-industrial times have been identified for these metals in other studies (Rognerud and Fjeld, 2001; Rognerud et al., 1999). It is therefore likely that the anthropogenic impact is so small that it is camouflaged by the natural background levels.

The concentration of Hg increased steadily from pre-industrial to present time. The enrichment factors for Hg were in the same range as factors previously reported for lakes in northern parts of Finland, Norway and Sweden (Dietz et al., 1998; Rognerud et al., 1993). The results suggest a continuing input of Hg to Arctic sediments.

Previous studies have shown that heavy metals are enriched in soil samples collected in or close to seabird colonies (Godzik, 1991; Headley, 1996). Seabird transport may to some degree have affected the concentration of metals in Ellasjøen, but the effect on metal concentration is not as dramatic as the effect of this biological transport mechanism on POP-concentrations. We suggest that the reason for this is that the accumulation of POPs to a large degree is related to biomagnification processes, which do not apply to most metals (except Hg).

4.3. Overall assessment of contaminant fluxes

Fluxes of phased-out and present used POPs, as well as most metals, in sediment cores from Ellasjøen on Bjørnøya accurately reflect usage and discharge patterns for the most of the measured contaminants; a fact that supports the conclusion that historical trends in contaminant accumulation within Ellasjøen can be confidently interpreted based on the ^{210}Pb

determined sediment ages. In other studies, delays have been reported in the initiation of major shifts in emissions (e.g. increase, decrease, stabilization) relative to the timing of their appearance in the Arctic sedimentary record (de March et al., 1998; Muir et al., 1995, 1996). However, this does not seem to be the case in Ellasjøen. Although Bjørnøya is a remote island, it is relatively close to several contaminant source areas in central Europe (<2500 km) and Northwest Russia (about 700 km to Nickel).

In addition to changes in long-range atmospheric input, fluctuations in seabird populations may have had an influence on the flux of contaminants to Ellasjøen. However, long-term data series on seabird population trends around Ellasjøen for the species of interest (little auk, kittiwake and glaucous gull) are limited (Anker-Nilssen et al., 2000) preventing any assessments on how guano inputs have varied through the 20th century. The fact that the historical development in contaminant concentrations reflects usage patterns of contaminants fairly well suggests that the contaminant supplies from seabirds have remained relatively constant over time. Both the relatively slow down-core decrease in organic carbon content in the sediment and the stability of congener patterns recorded throughout the cores serves as further corroboration of this hypothesis.

5. Conclusions

The levels of PCBs (18 congeners) and DDT and metabolites increased in Ellasjøen from about 1930 until 1970 (max. sum PCBs 71.8 ng/g dry weight (dw), max sum DDTs 4.0 ng/g dw), declining afterwards to 46.4 ng/g dw and 1.6 ng/g dw, respectively. PBDE concentrations have been increasing in Ellasjøen, reaching 0.73 ng/g dw in 2001. Most metals (As, Cd, Co, Ni and Zn) showed a similar pattern as PCBs. However, no clear trends were observed for Cr, Pb or V. Mercury (Hg) has increased gradually since the beginning of the industrial era. Due to a relatively high sedimentation rate and supplies of contaminants both from long-range atmospheric transport and seabird guano, changes in contaminant input from source areas are rapidly reflected in lake sediment from Ellasjøen. This lake is therefore a useful site for monitoring changes of contaminant input to the European Arctic.

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