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Seabird-mediated transport of organohalogen compounds to remote sites (North West Greenland polynya)

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HIGHLIGHTS

- The lake with bird droppings was acidic (pH = 3.4), the others close to neutral (pH = 8).
- Seabirds efficiently transport organohalogen compounds to Arctic sites.
- The bird-mediated deposition preserves the less stable and more volatile compounds.
- Organohalogen deposition was highest in the upper sediments of the polynya lake.
- Birds are introducing important organohalogen amounts in Arctic sites, except DDTs.

GRAPHICAL ABSTRACT

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ABSTRACT

The role of sea birds as carriers of pollutants over long distances was evaluated by analyzing organochlorine and organobromine compounds in lake sediment cores from three remote sites around the North Water polynya (North West Greenland). One lake, NOW5, was in the vicinity of a little auk (*Alle alle* L.) bird colony, whereas the other two lakes, NOW14 and Q5, were undisturbed by seabirds. The former was strongly acidic (pH = 3.4) but the latter had a pH close to 8. Due to the guano loading, NOW5 exhibited higher chlorophyll concentrations (74 μg/L) than the other two lakes (1.6–3.4 μg/L), higher content of total phosphorous (0.34 mg/L vs. 0.007–0.01 mg/L) and total nitrogen (3.75 mg/L vs. 0.21–0.75 mg/L).

The concentrations of all organohalogen compounds were substantially greater in NOW5 than in the other lakes, indicating the strong influence of these seabirds in the transport and deposition of these compounds to remote sites. However, not all compounds showed the same increases. Hexachlorocyclohexanes and endosulfans were more than 18 times higher in NOW5, the drin pesticides and hexachlorobenzene (HCB), between 9.5 and 18 times and DDTs, polybromodiphenyl ethers (PBDEs), polychlorobiphenyls (PCBs) and chlordanes about 2.7–6 times. These differences demonstrated that the bird-mediated deposition has preservation effects of the less stable and more volatile compounds, e.g. those with log Kaw < −2.4, log Koa < 9 and/or log Kow < 6.8.

The sedimentary fluxes of PCBs, HCHs, drins, chlordanes, PBDEs, HCB and endosulfans were highest in the upper sediment layer of the polynya lake (year 2014). In contrast, the highest DDT fluxes were found in 1980. These trends

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indicate that despite restrictions and regulations, bird transport continues to introduce considerable amounts of organohalogen pollutants to the Arctic regions with the exception of DDTs, which show successful decline, even when mediated by bird metabolism.

1. Introduction

Organic molecules with high degrees of halogen substitution, e.g. organochlorine and organobromine compounds, were produced and used due to their high chemical stability, fire and thermal resistance, and, in some cases, pesticide activity. Unfortunately, these properties caused environmental pollution problems and human health deleterious effects. First, their high stability to environmental degradation and semi-volatility boosted their spread over long distances through the atmosphere. Second, their hydrophopic properties favoured their accumulation in flora and fauna, which led to biomagnification along the aquatic and continental trophic chains.

A substantial number of compounds with these properties have been phased out of use and production. Most organochlorine compounds such as polychlorobiphenyls (PCBs), aldrin, dieldrin, endrin, heptachlor, and other legacy pollutants listed in the Stockholm Convention's "dirty dozen" (Korosi et al., 2017) have been banned worldwide in the last decades. For instance, among the banned compounds, aldrin, dieldrin and endrin were used as insecticides from the 1950s to the early 1970s (Blais and Muir, 2005). Aldrin is readily metabolised to dieldrin in plants and animals which is more resistant to biodegradation making it more toxic (Vorkamp et al., 2004). Giving another example, among hexachlorocyclohexanes (HCHs), γ-HCH was widely used around the globe but the overall usage significantly decreased in the 1980s and 1990s (Hung et al., 2010) despite the fact that it is still in use in some countries. Endosulfan and methoxychlor belong to a new group of less persistent chlorinated pesticides and are still used in Europe, USA and, most likely, in other world areas.

Despite of these restrictions, many of these compounds called persistent organic pollutants (POPs) are still known to linger in the various environmental compartments from close and distant sites of human activity, reaching the most remote places of the planet, such as high mountains (Grimalt et al., 2001, 2004a; Grimalt et al., 2004b) and Arctic areas, where they were never produced or used (Ma et al., 2011, 2016; Rigét et al., 2010b). In the absence of significant local anthropogenic inputs, the regional and global distributions of these compounds is increasingly reflecting phase partitioning between the environmental reservoirs such as air, water, soil, vegetation and ice, where POPs get accumulated. In general, the concentrations of phased-out or heavily restricted contaminants like PCBs, DDTs, hexachlorobenzene (HCB), chlordanes, dieldrin and their metabolites are now stabilizing or decreasing (Muir and de Wit, 2010). In contrast, many brominated flame retardants, for example, the polybromodiphenyl ethers (PBDEs), which have the same bioaccumulative properties such as PCBs, are still in production and use and their concentrations are in fact, increasing in the Arctic (de Wit et al., 2010).

Knowledge on contamination trends and origins is critical for determining the extent of POP transport and accumulation to the Arctic (Rigét et al., 2020; Rigét et al., 2016; Rigét et al., 2019). In addition to atmospheric transport, some studies indicate that migratory species may also play a critical role in translocating POPs between ecosystems, increasing productivity in otherwise unproductive systems (Blais et al., 2007; Evenset et al., 2004; Michelutti et al., 2009a, 2010). This is especially true for sea birds which feed at sea and come to land to breed, bringing nutrients as well as contaminants into freshwater systems (Blais, 2005; Davidson et al., 2018; Evenset et al., 2007a; González-Bergonzoni et al., 2017; Polis et al., 1997). Many avian species are migratory, travelling thousands of kilometers from breeding sites to wintering grounds, spending majority of their life in both areas (Wang et al., 2019). In recent decades, a number of surveys have indicated large accumulations of various POPs in birds all across the world, suggesting that pollutant levels in these organisms are related to their surrounding environments (Blais et al., 2007; Bustnes et al., 2012; Mehlum and Daelemans, 1995; Roosens et al., 2007; Huertas et al., 2016; Wang et al., 2019). Seabirds may be the most globally relevant bio-vectors since they are keystone species that represent the dominant form of wildlife along coastlines worldwide and form dense nesting colonies that can number in the millions of individuals (Michelutti et al., 2009a).

Local nutrient enrichment from guano has been documented, but the possibility of POP contamination in areas near seabird nesting sites has only been probed by a handful of studies (Blais, 2005; Choy et al., 2010; Evenset et al., 2007a, 2007b; Michelutti et al., 2009a, 2009b). Most of these studies have been conducted at Cape Vera on Devon Island (Nunavut, Canada; 76°15′N, 89°15′W) where a colony of northern fulmars (*Fulmarus glacialis*, L.) was observed to release POPs and metals to the environment through their guano (Blais, 2005; Choy et al., 2010; Michelutti et al., 2009a, 2009b). Another study was conducted in Lake Ellasjøen (Bjørnøya; Barents Sea; 74°30′N, 19°00′E) where colonies of little auk (*Alle alle*), kittiwake (*Rissa tridactyla*, L.) and glaucous gull (*Larus hyperboreus*, Gunnerus) are present (Evenset et al., 2007a, 2007b). These studies have been focussed on the transport of PCBs and DDTs, and, in the case of Lake Ellasjøen, also hexabromocyclododecanes.

In the present study, we are exploring the efficiency of transport of POPs by guano deposits in one Greenland site, at the southern end of the North Water Polynya, Northern Baffin Bay, which lies between Greenland and Canada (Fig. 1). This site allocates the largest polynya in the Arctic which covers around 85,000 km^2 with little auk. Our study focusses not only with the most hydrophobic POPs, e.g. PCBs, and DDTs, but also on more hydrophilic compounds such as aldrin, dieldrin, endrin, heptachlor, HCHs, chlordanes, endosulfans and HCB. PBDEs have also been considered. North Water Polynya (NOW) is one of the most productive marine environments in the Arctic owing to the combination of year-round nutrient rich, open waters and constant light in the summer, making it an ideal study location (González-Bergonzoni et al., 2017; Ribeiro et al., 2021). Three freshwater lakes located along the coast of NOW were selected based on their ecological conditions: one with a large migratory colony of little auk in the catchment (North Water; NOW5), one with no birds and historical human presence (Hunter gatherer Inuit-Thule people) from around 1400 CE (NOW14) and one without any interference from birds or humans (Q5).

Sediment cores were collected in each of these lakes to provide time-trend information on whether increased restrictions in the use of some products at lower latitudes may have resulted in decreasing levels in the Arctic and/or whether banned compounds are still transported to this remote environment due to possible time lags (Malmquist et al., 2003). In this regard, paleolimnological studies in remote sites provide useful information on the fate and transport processes of these pollutants (Fernandez et al., 2000; Korosi et al., 2017). By using lake sediments, past atmospheric deposition rates can indirectly be estimated and compared with current contamination levels. Unfortunately, there is a dearth of information on depositional chronology of concentrations or fluxes of contaminants in freshwater lake sediments from the Arctic compared to the high altitude freshwater lakes (Grimalt et al., 2001; Grimalt et al., 2004a, 2004b). Only a few studies have provided information on concentrations and fluxes of contaminants in freshwater lakes in the European Arctic (Evenset et al., 2007a) and Canadian Arctic (Muir et al., 1995, 1996; Stern et al., 2005), making the present study an important contribution to the available literature.

Here, we seek to understand what has been the transfer of POPs to these reference Arctic freshwater lakes in Greenland at present and in the past, and how the presence of bird colonies may have affected their deposition pattern. Specifically, a chronological comparison of the POPs accumulation

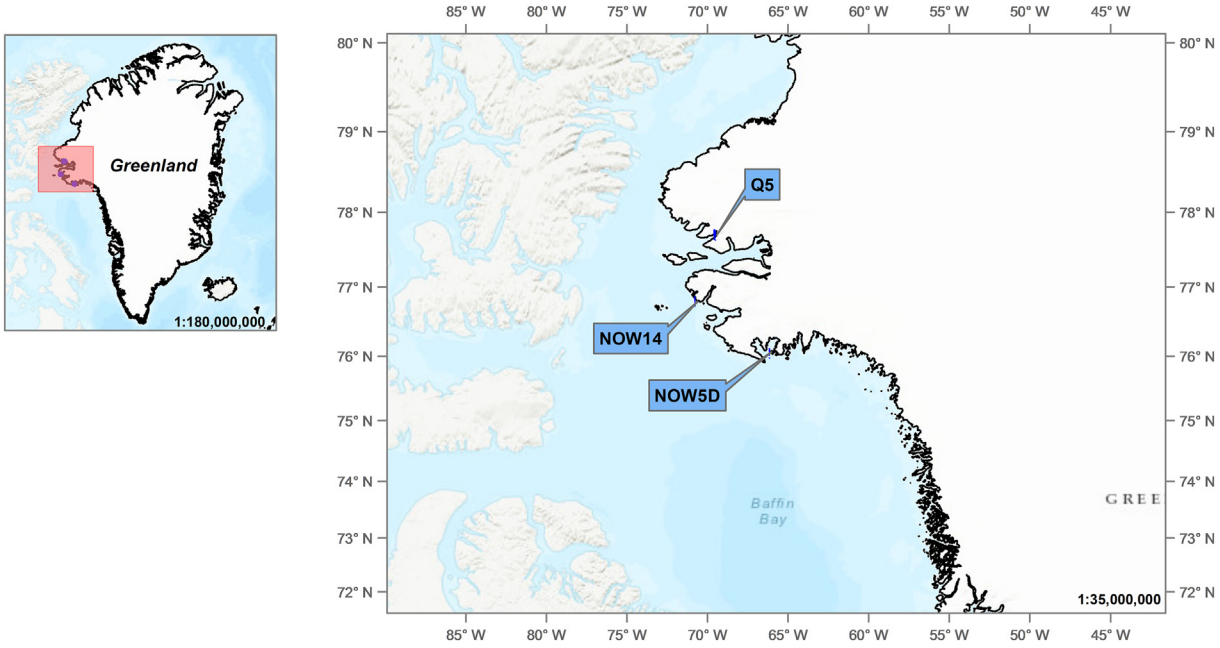

Fig. 1. Maps showing the location of the studied lakes. NOW5 is the lake hosting a polynya.

in sediment cores from the three selected lakes, starting from the industrialisation period to the present, is reported.

2. Materials and methods

2.1. Sampling

Samples were collected from the three freshwater lakes on the coast of NOW polynya in the summers of 2014 (NOW5), 2015 (NOW14), and 2016 (Q5; Fig. 1). Lake coring was carried out using an HTH gravity corer with an internal diameter of 8.5 cm. The cores were sliced and stored frozen until analysis. Further details of sampling and dating of the sediment cores have been described elsewhere (Davidson et al., 2018).

2.2. Chemicals and reagents

Acetone, ethyl acetate, cyclohexane, isooctane and copper (particle size <63 μm) were from Merck (Darmstadt, Germany). The Florisil cartridges (20CC; 5 g) were acquired from Waters (Milford, MA, USA). Standards of 1,2,4,5-tetrabromobenzene (TBB), PCB 209 and the solution mixture of OCs were purchased from Dr. Ehrenstoffer (Augsburg, Germany). Solution mixtures of BDEs, BDE 118 and [3-^{13}C] BDE-209 were acquired from Cambridge Isotope Laboratories (Andover, MA, USA).

2.3. Analytical method

The frozen sediment core slices were freeze-dried overnight. Sediments (0.1–1.0 g) were placed in a glass centrifuge tube (10 mL), spiked with surrogate standards (TBB and PCB 209) and kept in contact overnight. After that period, the samples were extracted with 10 mL ethyl acetate/cyclohexane (5:2, v/v) by vortex mixing for 1 min and ultrasonic stirring (10 min). The extract was centrifuged (10 min; 3000 rpm) and transferred to 40 mL flasks. This procedure was repeated twice. The total extracts were then concentrated to 1 mL by vacuum rotary evaporation. About 200 mg of activated copper was added to the extracts and left overnight to eliminate sulphur. Clean-up was performed using 5 g florisil cartridges after conditioning that were eluted with 20 ml of ethyl acetate/cyclohexane (5:2, v/v). This extract was finally concentrated to 0.5 mL.

Copper was activated by ultrasound stirring in a suspension of hydrochloric acid (25%; v/v) for 15 min. This procedure was followed by several Milli-Q washes for acid elimination until pH 7. Two rinses with acetone removed the water, and lastly, the activated copper was stored in hexane at −20 °C.

2.3.1. Organochlorine compound analysis

Before chromatographic analysis, an internal standard of PCB-142 was added to each sample. The OCs were identified and quantified by gas chromatography (GC; Agilent Technologies 7890 N) coupled to mass spectrometry (MS, Agilent Technologies 5975C) operating in negative chemical ionisation mode (GC-NICI-MS). This instrument was provided with a HP-5MS capillary column (60 m length, 0.25 mm internal diameter, 0.25 μm film thickness; JW Scientific) protected with a retention gap. The injection was performed in splitless mode. Injector and detector temperatures were 280 °C and 325 °C, respectively. The oven temperature was held at 90 °C for 2 min, increased to 130 °C at 15 °C/min and to 310 °C at 4 °C/min with a final holding time of 10 min. Ultrapure helium and ammonia were used as carrier and reagent gasses, respectively.

2.3.2. Polybromodiphenyl ether analysis

Before chromatographic analysis, internal standards of BDE-118 and [3-^{13}C]BDE-209 were added to all the samples. A GC (Agilent Technologies 7890N) coupled to a MS (Agilent Technologies 5975C) operating in negative chemical ionisation mode (GC-NICI-MS) was used for identification and quantification of the PBDE congeners (17, 28, 47, 66, 71, 85, 99, 100, 138, 153, 154, 183, 190 and 209). The instrument was equipped with a low bleed fused silica capillary column (15 m length, 0.25 mm ID, 0.10 μm film thicknesses; DB-5MS) protected with a retention gap. The oven temperature was programmed from an initial temperature of 90 °C which was kept for 1.5 min followed by heating to 200 °C at 40 °C/min, a second increase up to 275 °C at 5 °C/min and a third increase to 300 °C at 40 °C/min. This temperature was held for 10 min and then increased to 310 °C at 10 °C/min with a final holding time of 2 min. Ammonia was used as reagent gas. Identification and quantification were performed by injection of PBDEs standard solutions (Vizcaino et al., 2009; Bravo et al., 2017).

2.4. Quality assurance

Three procedural blanks were analysed with each batch of samples. Identification and quantification of OCs and PBDEs were performed by

injection of external standards at different concentrations, and all the samples were blank corrected. Method detection limits were calculated from the average signals of the procedural blank levels plus three times the standard deviation. They ranged between 0 and 0.0020 ng/g for the individual OCs and 0.001 and 0.0030 ng/g for the brominated compounds. The limits of quantification were calculated from the averages of the procedural blanks plus five times the standard deviation. They ranged between 0 and 0.0050 ng/g for the individual OCs and between 0.001 and 0.10 ng/g for PBDEs. One-half of the limits of detection and limits of quantification were assigned to non-detected and non-quantified values, respectively. Pentachlorobenzene was below the LOD in most sediment sections and therefore not included in further analysis.

3. Results and discussion

The lakes had a strong difference in terms of depositional setting. The waters of NOW5 were highly acidic with a pH of 3.4, whereas the pH values of NOW14 and Q5 were close to 8 (Table 1). This difference can be attributed to the influence of *A. ale* guano depositions (Davidson et al., 2018). In Spitzbergen, guano depositions of these birds also resulted in acidic lake water (Zwolicki et al., 2013). These highly acidic values have not been reported in other lakes receiving high amounts of guano depositions from other bird species.

These guano depositions are also responsible for the other observed differences between the lake waters. Thus, NOW5 has higher total phosphorous and nitrogen, 0.34 mg/L and 3.75 mg/L, respectively, than the other two lakes, 0.007–0.01 mg/L and 0.21–0.75 mg/L, respectively. Obviously, these differences led to strong contrasts in lake productivity, e.g. NOW5 show high chlorophyll concentrations (74 μg/L vs. 1.6–3.4 μg/L, respectively).

3.1. Concentrations

The concentrations of all organochlorine and organobromine compounds studied were higher in NOW5 lake than in the other two reference lakes (NOW14 and Q5; Table 2). The median concentrations of DDTs, PBDEs, PCBs, and chlordanes in the NOW5 core, 1.9 ng/g, 1.3 ng/g, 1.2 ng/g, and 0.040 ng/g, respectively, are about 2.7–6 times higher than the medians in NOW14 and Q5 lake cores, 0. 43–0. 64 ng/g, 0.22–0.37 ng/g, 0.26–0.44 ng/g, and 0–0.010 ng/g, respectively. In the case of the drin pesticides and HCB, the NOW5 medians of 0.74 ng/g and 0.36 ng/g, respectively, are about 9.5–18 times higher than in NOW14-Q5, 0.050–0.078 ng/g and 0.020–0.023 ng/g, respectively. HCHs' and endosulfans' medians in NOW5, 0.18 ng/g and 0.020 ng/g, respectively, are more than 18 times higher than in NOW14-Q5, 0–0.010 ng/g and below detection limit, respectively.

Table 1
Characteristics of the studied lakes.

	NOW5 (Salve Ø)	NOW14 (Nuuliit)	Q5 (Qaanaaq)
Latitude	76.04386	76.800518	77.70713
Longitude	−65.99149	−70.60262	−69.41987
Core length to 1900 (cm)	0–12.5	0–5.5	0–12.5
Year of sampling	2014	2015	2016
Depth (m)	18	1	17
pH	3.4	8.3	7.98
Chla (μg/cm^2) or μg/L	74	3.4	1.6
T (°C)	2.4	11.90	8.1
TP (mgP/L)	0.37	0.01	0.007
TN (mg/L)	3.75	0.75	0.212
Living organisms in the catchment	Birds (little auk, thick-billed murre, common eider)	Inconsistent presence of Humans (Thule people)	None

The concentrations of all POPs tested in lakes with no bird influence (NOW14 and Q5; Table 2) are lower than those reported in the lacustrine cores from high mountain European lakes (Grimalt et al., 2004a), Yukon lakes (Rawn et al., 2001) and Lake Ellasjøen (Bear Island; Norway; Evenset et al., 2007b) (Table 3). The concentrations of HCB, HCHs, endrins, and endosulfans in NOW14 and Q5 (Table 2) are also lower than those reported in cores from other Arctic lakes, including the Canadian Arctic, Alaska and Bear Island (Table 3).

However, the concentrations of DDTs in these lakes with no bird influence (max. 0.8–1.7 ng/g; Table 2) are higher than those observed in lake cores from Alaska and Devon Island (max 0.15–0.20 ng/g; Stern et al., 2005; Allen-Gil et al., 1997) and similar to those from some Canadian Arctic lakes (Muir et al., 1995, 1996). The concentrations of PCBs (max. 0.48–0.90 ng/g; NOW14 and Q5; Table 2) are also lower than those reported in lacustrine cores from Svalbard and the Canadian Arctic (max 2.5–59 ng/g; Muir et al., 1995, 1996; Rose et al., 2004), similar to those from Kangerlussuaq (Greenland; max 0.60–1.3 ng/g; Malmquist et al., 2003) and higher than in lake cores from Alaska (0.12 ng/g; Allen-Gil et al., 1997). The concentrations of PBDEs (max. 0.27–0.37 ng/g; Table 2) are comparable to those reported in lake cores from Devon Island (Nunavut, Canada; max. 0.25 ng/g; Stern et al., 2005) and higher than those reported in Kangerlussuaq (Greenland; max. 0.001–0.02 ng/g; Malmquist et al., 2003).

Except for DDT in Lake Redon, which has comparable values (max. 15–16 ng/g; Tables 2–3), the concentrations of all POPs from the core of the lake of strong bird influence, NOW5, are lower than those reported in the lake cores from remote high mountains of Europe (Grimalt et al., 2004a). In contrast, the higher concentrations caused by guano deposition mean that these POP concentrations in the core from this lake (NOW5) which is situated very close to NOW14 and Q5 has now higher values than in many of the Arctic sites discussed above.

Similarly, the maximum concentrations of DDTs, drins, PBDEs, PCBs, HCHs, HCB, chlordanes and endosulfans, in NOW5, which are 16 ng/g, 7.9 ng/g, 6.2 ng/g, 3.8 ng/g, 1.2 ng/g, 0.65 ng/g, 0.47 ng/g, and 0.25 ng/g, respectively, are higher than those in Kangerlussuaq lakes (Greenland), where PCBs, PBDEs, chlordanes and HCB concentrations, were 0.60–1.3 ng/g, 0.001–0.02 ng/g, 0.03–0.6 ng/g, and 0.01–1.5 ng/g, respectively (Malmquist et al., 2003; Table 3). They are also higher than those in Devon Island lake cores (Canadian Arctic) for DDTs, max. 0.20 ng/g, HCHs, 0.33, ng/g, PCBs, 2.7, ng/g, PBDEs, 0.25, ng/g, chlordanes, 0.08, ng/g, dieldrin, 0.43, ng/g, and endosulfans, 0.60 ng/g (Stern et al., 2005). A comparison of the mean core concentrations in NOW5 and the mean top 2 cm from lake sediment in Alaska (Elusive, Schrader, Feniak and Desperation Lakes from the Brooks Range) shows that the concentrations of HCB, 0.17 ng/g, HCHs, 0.090 ng/g, PCBs, 0.12 ng/g, DDTs, 0.15 ng/g, endosulfans, below detection limit, and chlordanes, 0.030 ng/g (Allen-Gil et al., 1997) are higher in the seabird influenced Greenland polynya lake (Tables 2 and 3). The organohalogen concentrations in NOW5 are similar to those found in other remote Canadian Lakes such as Yukon Lakes (Rawn et al., 2001) concerning DDTs, 0.86–21 ng/g, HCB, 1.1–1.9 ng/g, and HCHs, 0.46–1.6 ng/g, or lakes located between 49°N and 82°N (Muir et al., 1996) for HCB, 0.3–1.8 ng/g, HCHs, 0.05–2.9 ng/g, chlordanes, 0.08–3.3 ng/g, and dieldrin, 0.05–3.2 ng/g. However, these Canadian lakes exhibit higher PCB concentrations, 2.5–59 ng/g (Tables 2 and 3). PCBs are also found in higher concentrations than NOW5 lake in cores from Svalbard, 2.6–14 ng/g (Rose et al., 2004) and Ellasjøen, 72 ng/g (Bear Island; Evenset et al., 2007b). In this last site, the concentrations of DDTs, 4.0 ng/g, and PBDEs, 0.73 ng/g, are lower than in NOW5.

3.2. Depositional fluxes

The sedimentation fluxes of all organochlorine and organobromine compounds considered for study were higher in the core of the NOW5 lake than in the cores of the other two reference lakes (NOW14 and Q5; Table 4). The medians of the fluxes of DDTs, PBDEs, PCBs, and chlordanes in NOW5 core, 10 pg cm^{-1} y^{-1}, 4.2 pg cm^{-1} y^{-1}, 5.9 pg cm^{-1} y^{-1}, and 0.15 pg cm^{-1} y^{-1}, respectively, are about 1.9–4.2

Table 2
Statistical parameters of the concentrations of the organohalogen compounds found in the studied cores (ng/g dry weight).

	NOW5						NOW14						Q5					
	Min	Max	Top[i]	Mean	Median	SD[j]	Min	Max	Top[i]	Mean	Median	SD[j]	Min	Max	Top[i]	Mean	Median	SD[j]
HCB[a]	0	0.65	0.35	0.30	0.36	0.23	0.010	0.030	0.020	0.018	0.020	0.0075	0.010	0.060	0.011	0.023	0.020	0.014
PCBs[b]	0.14	3.8	1.7	1.5	1.2	0.96	0.10	0.48	0.26	0.28	0.26	0.10	0.070	0.90	0.48	0.43	0.44	0.22
HCHs[c]	0	1.2	0.71	0.29	0.18	0.30	0	0.020	0	0.011	0.010	0.0094	0	0.010	0	0.0029	0	0.0047
Drins[d]	0.020	7.9	0.16	1.6	0.74	2.2	0.040	0.19	0.050	0.078	0.050	0.052	0.020	0.16	0.020	0.078	0.050	0.052
DDTs[e]	0.12	16	0.12	2.7	1.9	3.4	0.35	0.80	0.35	0.60	0.64	0.14	0.19	1.7	1.1	0.56	0.43	0.38
Chlordanes[f]	0	0.47	0.19	0.080	0.040	0.12	0	0.040	0.010	0.014	0.010	0.010	0	0.020	0.0060	0.048	0	0.0070
PBDEs[g]	0.22	6.2	2.5	1.6	1.3	1.5	0.27	0.48	0.37	0.35	0.37	0.061	0.11	1.1	0.27	0.29	0.22	0.26
Endosulfans[h]	0	0.25	0.25	0.062	0.020	0.083	0	0	0	0	0	–	0	0.010	0.0010	0.00071	0	0.0024

[a] Hexachlorobenzene.
[b] Polychlorobiphenyls (28 + 52 + 101 + 118 + 138 + 153 + 180 congeners).
[c] Hexachlorocyclohexanes (α + β + γ isomers).
[d] Aldrin + dieldrin + endrin.
[e] 2,4′-DDE + 4,4′-DDE + 2,4′-DDD + 4,4′-DDD + 2,4′-DDT + 4,4′-DDT.
[f] cis + trans chlordanes.
[g] 17 + 28 + 47 + 66 + 71 + 85 + 99 + 100 + 138 + 153 + 154 + 183 + 190 + 209 congeners.
[h] α- + β-endosulfans + endosulfan sulphate.
[i] Value at the top of the core.
[j] Standard deviation.

times higher than the medians in NOW14 and Q5 lake cores, 2.4–4.2 pg cm^{-1} y^{-1}, 1.3–2.2 pg cm^{-1} y^{-1}, 1.7–2.8 pg cm^{-1} y^{-1}, and 0–0.080 pg cm^{-1} y^{-1}, respectively. In the case of the drin pesticides and HCB, the NOW5 core medians of 5.2 pg cm^{-1} y^{-1} and 1.4 pg cm^{-1} y^{-1}, respectively, are about 9.5–13 times higher than in NOW14-Q5 core, 0.48–0.55 pg cm^{-1} y^{-1} and 0.11–0.12 pg cm^{-1} y^{-1}, respectively. The medians of HCHs and endosulfans in NOW5 core, 0.84 pg cm^{-1} y^{-1} and 0.070 pg cm^{-1} y^{-1}, respectively, are more than 21 times

Table 3
Sedimentary concentrations and fluxes of organohalogen compounds from other remote lakes.

Sites	Location	Compound	Concentrations (ng/g)	Flux (pg cm^{-2} y^{-1})
Pyrenees (Lake Redon)	42°38′34″N 0°46′13″E	DDTs	Max[a]: 15; Top[b]: 15	
(Grimalt et al., 2004a)		PCBs	Max: 7.3; Top: 7.3	
(1942–1992)		HCB	Max: 1.6; Top: 1.6	
Tatras	49°10′0″-49°13′36″N 20°0′39″-20°10′0″E	DDTs	Max: 30–190; Top: 1–50	
(Grimalt et al., 2004a)		HCHs	Max:1–10; Top: 1	
(1942–2001)		PCBs	Max: 36–86; Top: 5.6–45	
		HCB	Max: 0.5–3.2; Top: 1	
Canadian Arctic	75°34.420′N 89°18.545′W	DDTs	Max: 0.20; Top: 0.05	Max: 1.5; Top 0.71
Devon Island		HCHs	Max: 0.33; Top: 0.22	Max: 3.8; Top: 3.1
(Stern et al., 2005)		PCBs	Max: 2.7; Top: 2.6	Max: 37; Top 37
(1908–1997)		PBDEs	Max:0.25; Top: 0.17	Max: 2.8; Top: 2.8
		Chlordanes	Max: 0.08; Top: 0.05	Max: 1.3; Top: 1.3
		Dieldrin	Max: 0.43; Top: 0.28	Max: 4.6; Top: 4.0
		Endosulfan	Max: 0.60; Top: 0.60	Max: 0.040; Top: 0.040
Ellasjøen (Bear Island)	74°30′N 19°00′E	PCBs	Max: 72; Top: 48	
(Evenset et al., 2007b)		DDTs	Max: 4.0; Top: 1.6	
(1881–1994)		PBDEs	Max: 0.73; Top: 0.73	
Svalbard	77°33′-79°48′N	PCBs	Max: 2.6–14; Top: 2.6–14	
(Rose et al., 2004)	9°30′-15°00′E			
(1980–1998)				
Kangerlussaq (Greenland)	67°16′-67°02′N	PCBs	Max: 0.60–1.3; Top: 0.11–1.2	Mean: 3.0
(Malmquist et al., 2003)	51°46′-51°07′W	PBDEs	Max: 0.001–0.02; Top: 0–0.02	
(1940–2000)		Chlordanes	Max: 0.03–0.6; Top: 0–0.6	1.0–4.0
		HCB	Max: 0.01–1.5; Top: 0–0.2	0.10–2.0
Yukon Lakes (Canada)	59°47′-64°00′N	PCBs	Max: 33.5–11; Top: 33.5–2.2	135–1150
(Rawn et al., 2001)	128°46′-136°10′W	DDTs	Max: 0.86–21; Top: 0.86–3.5	6.6–1300
		HCHs	Max: 0.46–1.6; Top: 0.13–1.4	7.5–36
		HCB	Max: 1.1–1. 9; Top: 0.06–1.1	4.2–87
Canadian Arctic	49°42′-81°45′N	PCBs	Max: 2.5–59; Top: 2.5–40	11–425
(Muir et al., 1995, 1996)	71°30′-126°16′W	HCB	Max: 0.3–1.8; Top: 0.1–1.8	3.2–3.9
(1895–1990)		DDTs	Max: 0.1–12; Top: 0.1–9.5	11–69
		Chlordanes	Max: 0.08–3.3; Top: 0–1	3.8–4.2
		HCHs	Max: 0.05–2.9; Top: 0–1	3–5.3
		Dieldrin	Max: 0.05–3.2; Top: 0–1	4.3–4.7
Alaska	68°16′-68°50′N	HCB	Mean top 2 cm: 0.17	
(Allen-Gil et al., 1997)	144°60′-158°45′W	HCHs	Mean top 2 cm: 0.090	
		PCBs	Mean top 2 cm: 0.12	
		DDTs	Mean top 2 cm: 0.15	
		Endosulfans	Mean top 2 cm: 0	
		Chlordanes	Mean top 2 cm: 0.030	

[a] Maximum value of the core.
[b] Value at the top of the core.

Table 4
Statistical parameters of the sedimentary deposition fluxes of the organohalogen compounds found in the studied cores (pg cm^{-2} y^{-1}).

	NOW5						NOW14						Q5					
	Min	Max	Top[i]	Mean	Median	SD[j]	Min	Max	Top[i]	Mean	Median	SD[j]	Min	Max	Top[i]	Mean	Median	SD[j]
HCB[a]	0	4.9	4.9	1.5	1.4	1.3	0.030	0.30	0.28	0.14	0.12	0.11	0.045	0.67	0.067	0.18	0.11	0.17
PCBs[b]	0.77	25	24	7.7	5.9	6.3	0.30	3.6	3.6	1.9	1.7	1.2	0.44	5.5	2.9	2.7	2.8	1.3
HCHs[c]	0	9.9	9.9	1.6	0.84	2.1	0	0.20	0	0.058	0.040	0.071	0	0.073	0	0.017	0	0.028
Drins[d]	0.080	34	2.2	7.7	5.2	8.9	0.17	0.70	0.70	0.43	0.48	0.18	0.12	0.98	0.12	0.50	0.55	0.28
DDTs[e]	1.7	47	1.7	12	10	10	1.9	7.2	7.2	3.8	4.2	1.8	1.0	10	6.4	3.6	2.4	2.3
Chlordanes[f]	0	3.6	2.7	0.53	0.15	0.94	0	0.14	0.14	0.10	0.080	0.11	0	0.44	0.036	0.048	0	0.11
PBDEs[g]	1.5	35	35	8.4	4.2	8.2	0.80	5.1	5.1	2.5	2.2	1.7	0.57	7.1	1.7	2.0	1.3	1.9
Endosulfans[h]	0	3.5	3.5	0.41	0.070	0.78	0	0	0	0	0	–	0	0.061	0.0061	0.0043	0	0.015

[a] Hexachlorobenzene.
[b] Polychlorobiphenyls (28 + 52 + 101 + 118 + 138 + 153 + 180 congeners).
[c] Hexachlorocyclohexanes (α + β + γ isomers).
[d] Aldrin + dieldrin + endrin.
[e] 2,4′-DDE + 4,4′-DDE + 2,4′-DDD + 4,4′-DDD + 2,4′-DDT + 4,4′-DDT.
[f] cis + trans chlordanes.
[g] 17 + 28 + 47 + 66 + 71 + 85 + 99 + 100 + 138 + 153 + 154 + 183 + 190 + 209 congeners.
[h] α- + β-endosulfans + endosulfan sulphate.
[i] Value at the top of the core.
[j] Standard deviation.

higher than in NOW14-Q5 core, 0–0.040 pg cm^{-1} y^{-1} and below detection limit, respectively.

Comparison of the data from Tables 3 and 4 shows that these sedimentation fluxes of all POPs in the lakes of the present study are lower than those described in lacustrine cores from Kangerlussuaq (Greenland; Malmquist et al., 2003), the Canadian Arctic (Muir et al., 1995, 1996) and Yukon Lakes (Rawn et al., 2001). However, there are two exceptions, the drins in the Canadian Arctic cores which have similar mean fluxes than NOW5 core, 4.3–4.7 pg cm^{-1} y^{-1} and 5.2 pg cm^{-1} y^{-1}, respectively (Tables 3 and 4), and the mean flux of PCBs in the Kangerlussuaq (3.0 pg cm^{-1} y^{-1}) which are lower than those of NOW5 core, 7.7 pg cm^{-1} y^{-1}. Comparison of the results of the present study with the POP composition in the cores from North West Greenland and Devon Island (Stern et al., 2005) shows that the latter had higher sedimentation fluxes than cores NOW14 and Q5 for of all POPs except in the case of DDTs whose maximum sedimentation rates were 1.5 pg cm^{-1} y^{-1}, 7.2 pg cm^{-1} y^{-1} and 10 pg cm^{-1} y^{-1}, respectively, and therefore higher in North West Greenland (Tables 3 and 4). Conversely, for all POPs except PCBs, sedimentation fluxes are higher in the NOW5 core, which is influenced by the bird polynya, which values are 27 pg cm^{-1} y^{-1} and 37 pg cm^{-1} y^{-1}, respectively.

3.3. Influence of birds on lacustrine POP accumulation

The specific conditions of sedimentation in NOW5 are strongly influenced by the bird population, as shown by the sedimentary concentration and flux differences between the three lakes chosen for study (Tables 2 and 4, respectively), resulting in the observed significant increase in organochlorine and organobromine compounds compared to the seabird free lakes.

Because POPs tend to partition from gas and liquid phases to organic phases, the accumulation of atmospheric POPs is also driven in part by gradients in organic carbon content of soils and sediments (Nizzetto et al., 2010). The fertilizing effects of guano deposits are the main sources of organic carbon in the lakes. Furthermore, eutrophication and the accumulation of organic matter can be related with chlorophyll *a* concentration. Phytoplankton in the water column is a transport driver of organohalogen compounds to the sediments both in lacustrine (Meijer et al., 2006) and marine environments (Dachs et al., 2002). In lacustrine environments, the sedimentation fluxes of the organohalogen compounds driven by phytoplankton sinking in the water column have been related to the water column chlorophyll *a* concentrations by Eq. (1) (Meijer et al., 2006),

$$F_{sed} = 1.8 \cdot C_p \cdot 10^{(1.82+0.62\log(Chla))}/(69Chla + 146.8) \quad (1)$$

where F_{sed} is the sedimentation flux of one organohalogen compound (ng m^{-2} d^{-1}), Chla is the concentration of chlorophyll *a* (kg m^{-3}) and C_p is the concentration of the organochlorine compound in phytoplankton (per volume of water; ng m^{-3}).

The estimated coefficients for the C_p values in lakes NOW5, NOW14 and Q5, 0.16 m d^{-1}, 0.024 m d^{-1} and 0.015 m d^{-1}, respectively, are obtained by calculating the chlorophyll *a* values defined by Eq. (1) using the chlorophyll concentrations of Table 1. These result in ratios of 6.5 and 10 for the relative sinking rates for NOW5/NOW14 and NOW5/Q5, respectively.

These values are intermediate between the ratios of the measured concentrations or deposition rates, in the range of 1.9–21 for NOW5/NOW14 and 2.1–13 for NOW5/Q5 when calculated using the median values (Table 5). These ratios are consistent with the enhanced effect of the bird deposition. As shown in a previous study (Meijer et al., 2009), significant transport from air to water occurs in the gas phase at air to water constants, log Kaw, higher than −2.4, and in the particulate phase at octanol to air constants, log Kow, higher than 9. Accumulation from the water column into sediments is observed at octanol to water constants, log Kow, higher than 6.8 (Meijer et al., 2009). These conditions are fulfilled by most congeners of the ΣPCBs and ΣPBDEs and DDT metabolites (Table 5). The chlordanes also have constants close to these threshold values (Table 5). Accordingly, these compounds are those having lower NOW5/NOW14 and NOW5/Q5 ratios (1.9–4.2) because their deposition and accumulation in the sediments already occurs irrespectively of bird mediation. The other compounds do not have physical chemical constants favouring transport from air to water (HCB, some HCHs, α- and β- endosulfans) or accumulation in the sediments (HCB, HCHs, drins, endosulfans) (Table 5). In these cases, the difference in the accumulation is more relevant if enhanced by bird deposition and the NOW5/NOW14 and NOW5/Q5 ratios are much higher (≥ 9.5).

Furthermore, the rates of Table 5 indicate that bird-mediated deposition has a preservation effect of the less stable compounds, e.g. endosulfans (Guerin and Kennedy, 1992), whose concentrations and fluxes are much higher in NOW5 than in the other lakes.

The effect of the birds in transporting POPs to the high arctic lakes can reflect two processes: 1) from lower to higher latitudes when they migrate and 2) from the marine environment to inland where they feed and transport meal for their chicks. This transport is in the same direction as that described in the global distillation model and the tendency for semi-volatile POP chemicals to become concentrated in cold environments (Wania and Mackay, 1993, 1996; Grimalt et al., 2001; Grimalt et al., 2004a, 2004b) but through specific biogenic mechanisms. In any case, the polynya effect leads to higher rates of POPs in

Table 5
Ranges of the physical-chemical constants of the organohalogen compounds found in the lakes.

	Log Kaw[a]	Log Koa[b]	Log Kow[c]	NOW5/NOW14[d]	NOW5/Q5[d]
HCB[e]	−1.158	7.38	5.73	12	13
PCBs[f]	(−2.087, −3.388)[g]	(7.71, 11.66)	(5.62, 8.24)	3.4	2.1
HCHs[h]	−3.677	(7.82, 8.88)	(3.93, 4.14)	21	>20
Drins[i]	(−1.801, −3.388)	(7.86, 8.59)	(5.20, 6.06)	11	9.5
DDTs[j]	(−2.769, −3.468)	(9.12, 10.38)	(5.87, 6.91)	2.4	4.2
Chlordanes[k]	−2.702	8.92	6.22	1.9	–
PBDEs[l]	(−3.516, −6.131)	(9.40, 18.42)	(5.88, 12.11)	1.9	3.2
Endosulfans[m]	(−2.576, −4.877)	(6.41, 8.54)	(3.66, 3.83)	>20	>20

[a] Air-water constants.
[b] Octanol-air constants.
[c] Octanol-water constants.
[d] Calculated from the median values of the calculated sedimentation rates.
[e] Hexachlorobenzene.
[f] Polychlorobiphenyls (28 + 52 + 101 + 118 + 138 + 153 + 180 congeners).
[g] Range of values for the compounds included in the group.
[h] α + β + γ Hexachlorocyclohexanes.
[i] Aldrin + dieldrin + endrin.
[j] 2,4′-DDE + 4,4′-DDE + 2,4′-DDD + 4,4′-DDD + 2,4′-DDT + 4,4′-DDT.
[k] cis + trans chlordanes.
[l] Polybromodiphenyl ethers (17 + 28 + 47 + 66 + 71 + 85 + 99 + 100 + 138 + 153 + 154 + 183 + 190 + 209 congeners).
[m] α- + β-endosulfans + endosulfan sulphate. Constant data from US EPA (2022) and Beyer et al. (2002).

the lake sediments which are not diluted by the organic matter increases due to the higher productivity associated to the increase of nutrients from the guano deposits.

3.4. Temporal trends

With the major threat of climate change looming over the Arctic, favouring snowmelt re-emission of POPs that were deposited long ago in the various environmental compartments in the cold climate (Arellano et al., 2011, 2015, 2018; Grimalt et al., 2009; Gustavsson et al., 2019; AMAP, 2021), multiple new sources of POPs may emerge, leading to changes in transport rates and additional deposition in the lakes. Consequently, monitoring lakes such as NOW5 with high sedimentation rates and supplies of contaminants from multiple sources is extremely beneficial since changes in contaminant input are reflected in the lake sediments.

Examination of the temporal trends of the sedimentary deposition fluxes of the measured pollutants (Fig. 2) show much higher values for NOW5 than the other lakes. This is consistent with the overall results of Table 4 and highlights the biogenic POP transport effect linked to guano deposits once more. The most distinct feature of the majority of time profiles from this core is the increase to highest values in the most recent core sections, e.g. 2014, for many of the compounds examined, PCBs, HCHs, drins, chlordanes, PBDEs, HCB and endosulfans. Total DDTs are the lone exception showing a maximum peak in 1980. The values in the upper sections (most recent years) in the other two cores are significantly lower, with no increase. These temporal trends in core NOW5 show that despite the banning of many of the POPs, the pollutants released into the environment at lower latitudes are still transported to Arctic areas and have an impact on their ecosystems and that migratory bird activity is a main agent of long-range transport in the polynya.

In addition to this main feature, the cores from NOW5 show other patterns such as a maximum of the sedimentation fluxes of drins, PCBs, HCHs, DDTs, PBDEs, and endosulfans at 1980–1985 (Fig. 2). This maximum likely reflects an episode of highest pollution by all these organohalogen pollutants and is coincident with the largest flux of DDTs recorded in this core. It is also coincident, with dating uncertainties, with a maximum of 4,4′-DDT and chlordanes in a lacustrine core from Devon Island (75°34.420′N 89°18.545′W; Stern et al.,

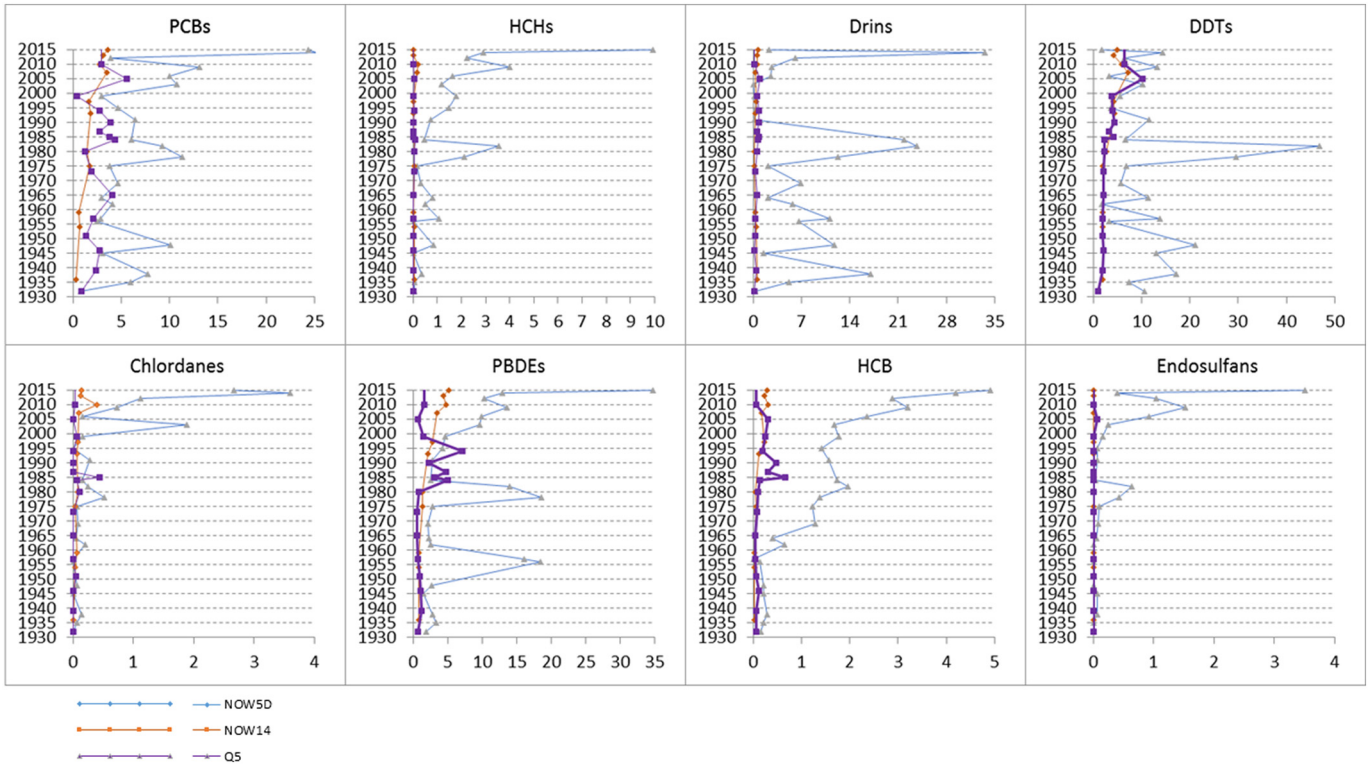

Fig. 2. Downcore sedimentary deposition fluxes of the organohalogen compounds found in the studied cores (pg cm^{-2} y^{-1}).

2005). Other lower maxima reflected higher inputs from specific comments, e.g. PBDEs in 1955, PCBs, drins, and DDTs before 1955.

The fluxes of HCB followed a different pattern showing a continuous increase from 1960 to 2015 and a maximum value in this last year (Fig. 2). HCB has been shown to display quite constant concentrations in the atmosphere in different remote sites, 35–85 pg m^{-3} in Europe (Van Drooge et al., 2004), 39–50 pg m^{-3} in North America (Daly et al., 2007), 56 pg m^{-3} in the Tibetan Plateau (Zhu et al., 2014) and 60 pg m^{-3} in Brazil (Guida et al., 2018) which is close to background pollution levels, while the other organohalogen compounds identified in the lakes may reflect episodic inputs.

In general, the temporal changes of POPs in Arctic samples reported in the literature were showing decreasing trends for most organochlorine compounds and increases for the PBDEs. However, recent reports are describing that the global warming is leading to re-emission of POPs already deposited in cold areas which effects in the Arctic regions (AMAP, 2021). Concerning PBDEs, a study of sediment cores from Søndre Strømfjord, near Kangerlussuaq (West Greenland), performed in 2000 showed maximum concentrations in the top sediment sections of four of the seven cores studied (Malmquist et al., 2003). de Wit et al. (2010) also showed increasing trends for PBDEs in the freshwater sediments from the Canadian Arctic and from Greenland. In the sediment core of Lake Ellasjøen (Bear Island; Barents Sea, with a seabird colony), the PBDEs concentration of the congeners (BDE-28, 47, 99, 100, 153), 0.73 ng/g, also peaked in the surface sediments (Evenset et al., 2007b).

PCBs concentration trends in Arctic biota have been reported to be decreasing (Rigét et al., 2010a, 2010b). Decreasing trends were also identified in cores from Kangerlussuaq, West Greenland, where the concentration maximum of these pollutants peaked in 1970 (Malmquist et al., 2003). Similar decreasing trends were recorded in Lake Ellasjøen where the concentration maximum was observed in the core section of 1966 (Evenset et al., 2007b). In other studies, i.e. Alaska (Gubala et al., 1995) and Finland (Kjeller and Rappe, 1995), PCBs were primarily found in layers starting 1950s with maxima in the 1970–1980s. A study from the Baltic, on the other hand, found PCBs appearing early in the 20th century (Muir et al., 1996). Sanders et al. (1992) also reported low levels of PCBs (mostly low molecular weight PCBs) in early 1900s. PCBs were commercially produced between the 1930s and 1980s (Bigus et al., 2014). However, on Devon Island, the concentrations of these pollutants in the sediment cores of lakes with strong bird influence (Cape Vera) show highest concentrations in the most recent core sections (Michelutti et al., 2009a, 2009b). This study likewise showed a strong contrast between the PCB concentrations in the core with bird influence (3.8 ng/g at the top) and no bird inputs (almost below limit of detection) which is consistent with the observations of the present study (Fig. 2). The PCB profiles from Devon Island lake sediment cores located at 75°34.420′N 89°18.545′W also showed maximum concentrations in the top sections (19 μg m^{-2} y^{-1}; Stern et al., 2005).

Lindane (γ-HCH) was used until recently when it was banned globally under the Stockholm Convention in 2009 (Kirk and Gleason, 2015). Arctic biota, especially seabird population, have showed decreasing trends for α- and γ-HCH (lindane) in Arctic Canada and Greenland but increasing trends of β-HCH in East Greenland black guillemot eggs (Rigét et al., 2016). Since β-HCH has stronger affinity for organic matter than α-HCH and γ-HCH, is found in higher concentrations in biota (Li and Macdonald, 2005). However, in NOW5, the concentrations of all three isomers of HCHs were similar and showed highest concentrations in the top sections, indicating a distinct temporal trend than in the cores lacking bird influence, which showed much lower concentrations and fluxes (Fig. 2). The HCH profiles in cores from Devon Island located at 75°34.420′N 89°18.545′W also showed maximum concentrations in the top sections and in the section dated in 1975 (1.95 and 2.2 μg m^{-2} y^{-1}, respectively; Stern et al., 2005).

Dieldrin, aldrin, endrin and chlordanes were banned or heavily restricted prior to the year 2000 (Kirk and Gleason, 2015). Although aldrin was widely used, it is readily degraded to dieldrin in the environment, which is the form of major concern in the Arctic. Most of the studies have focused solely on dieldrin, but in the present study all three drins were considered. Dieldrin and endrin were found in equally high concentrations in the core sections of highest sedimentation in NOW5.

HCB was first used in the 1950s with peak usage between 1970s and 1980. Despite the fact that it is currently banned it is commonly found in remote environments. For instance, in one study of freshwater lakes from Alaska, it was detected at concentrations of 0.17 ng/g dw in the top 2 cm of the sediment cores (Allen-Gil et al., 1997). In the current study, this compound is again found in highest concentration in the lake core of strong guano deposition with highest fluxes in the most recent core sections whereas the cores from the lakes free from bird influence show much lower concentrations and no discernable temporal trend.

As stated previously, the temporal profile of DDTs in the core from lake NOW5 was the only contaminant that did not show maximum values in the top sections of the core but in the section that corresponded to the year 1980. This temporal trend is in agreement with the banning policies implemented in European and north American countries. This profile is equivalent, within model age uncertainty, with the profile of 4,4′-DDT in a core from Devon Island which showed maximum concentrations in the section dated in 1988 (DV09; 1.6 μg m^{-2} y^{-1}; Stern et al., 2005). However, the lake core temporal trend of Lake Ellasjøen (Bear Island; Barents Sea) which also has a seabird colony shows the highest DDT concentrations at 1966 (4 ng/g; Evenset et al., 2007b). These comparisons of the ages from different core records are tentative because the resolution of the time models is not the same. In any case, the NOW5 core witnesses that the DDT restrictions successfully decreased the transport of this insecticide and metabolites to the Arctic regions which is in contrast with the observations of the other organohalogenated POPs.

4. Conclusions

According to the findings of this study, the levels of contamination in the lake NOW5 are positively correlated to the presence of birds in the lake catchment. The other two lakes are also polluted, albeit to a much lesser extent and mainly as a result of atmospheric deposition. The median concentrations of DDTs, PBDEs, PCBs, and chlordanes in the polynya lake are about 2.7–6 times higher than the median concentrations in the lakes with no bird influence. In the case of the drin pesticides and HCB, the former have a median that is about 9.5–18 times higher than the latter, and these ratios are more than 18 times higher for HCHs and endosulfans. When the changes in sedimentation fluxes between these lakes are considered, similar ratios are found, indicating that bird-mediated deposition has a preservation effect of the less stable and more volatile compounds.

The results have also shown that the highest sedimentary deposition fluxes of PCBs, HCHs, drins, chlordanes, PBDEs, HCB and endosulfans of the polynya lake were found in the upper sediment layer, corresponding to 2014. Thus, despite the fact that many of the organochlorine and organobromine compounds have been banned, those that have already been released to the environment at lower latitudes are still being transported to Arctic areas and impacting the ecosystems there.

In contrast, the maximum sedimentation fluxes of DDTs were found at the layer corresponding to 1980 which, within age model uncertainties, is consistent with the maximum sedimentation flux of this pesticide found in Devon Island (Stern et al., 2005) and could reflect that the restrictions in the use of this compound successfully decreased the transport of this insecticide and metabolite to the Arctic regions, even when mediated by bird metabolism.

CRediT authorship contribution statement

NN analysis, interpretation of results, writing. MB conceptualization, supervision, interpretation of results, review and editing. SB conceptualization, supervision, interpretation of results, TAD sampling, conceptualization, interpretation of results, review and editing. EJ interpretation of results, review and editing. JOG supervision, interpretation of results, writing, review and editing, funding acquisition.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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