



Terrestrial inputs govern spatial distribution of polychlorinated biphenyls (PCBs) and hexachlorobenzene (HCB) in an Arctic fjord system (Isfjorden, Svalbard)[☆]



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ABSTRACT

Considerable amounts of previously deposited persistent organic pollutants (POPs) are stored in the Arctic cryosphere. Transport of freshwater and terrestrial material to the Arctic Ocean is increasing due to ongoing climate change and the impact this has on POPs in marine receiving systems is unknown. This study has investigated how secondary sources of POPs from land influence the occurrence and fate of POPs in an Arctic coastal marine system.

Passive sampling of water and sampling of riverine suspended particulate matter (SPM) and marine sediments for analysis of polychlorinated biphenyls (PCBs) and hexachlorobenzene (HCB) was carried out in rivers and their receiving fjords in Isfjorden system in Svalbard. Riverine SPM had low contaminant concentrations (<level of detection–28 pg/g dw ΣPCB₁₄, 16–100 pg/g dw HCB) compared to outer marine sediments (630–880 pg/g dw ΣPCB₁₄, 530–770 pg/g dw HCB). There was a strong spatial gradient in sediment PCB and HCB concentrations with lowest concentrations in river estuaries and in front of marine-terminating glaciers and increasing concentrations toward the outer fjord. This suggests that rather than leading to increased concentrations, inputs of SPM from land lead to a dilution of contaminant concentrations in nearshore sediments. Preliminary estimates of SPM:water activity ratios suggest that terrestrial particles (with low contaminant concentrations) may have the potential to act as sorbents of dissolved contaminants in the coastal water column, with implications for bioavailability of POPs to the marine food web. There is concern that ongoing increases in fluxes of freshwater, sediments and associated terrestrial material (including contaminants) from land to the Arctic Ocean will lead to increased mobilization and transport of POPs to coastal ecosystems. However, the results of this study indicate that on Svalbard, inputs from land may in fact have the opposite effect, leading to reduced concentrations in coastal sediments and waters.

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1. Introduction

Persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs) and hexachlorobenzene (HCB), are highly resistant towards degradation and can undergo long-range transport (LRT) to remote areas, where they bioaccumulate and biomagnify

and can potentially cause adverse effects (AMAP, 2004; Rigét et al., 2019). Despite the Stockholm Convention ban, these compounds are still present in the environment, due to slow degradation, but also because they are continuously released into the environment from diffuse secondary sources or as unintended industrial by-products, e.g. PCB-11 and HCB (AMAP, 2016; Barber et al., 2005;

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Carlsson et al., 2018). The occurrence of PCBs and HCB in remote as well as more populated areas on Svalbard is well-documented through several studies and monitoring programmes and their environmental behaviour is fairly well-known, which make them well-suited for studies on the impact of environmental processes on contaminant cycling. Furthermore, these persistent and bio-accumulative compounds are known to pose a risk for long-lived Arctic top predators, where concentrations are often above thresholds for potential toxic effects (Carlsson et al., 2018; Hung et al., 2016; Jartun et al., 2009; Pedersen et al., 2011). These and other studies have shown how POPs are stored in terrestrial compartments such as glaciers, rivers (especially suspended particulate matter; SPM) and their catchment areas, where they can subsequently be released as secondary emissions to the receiving marine environment (Carlsson et al., 2018; Garmash et al., 2013; Kallenborn et al., 2012). More attention has recently been directed to the potential re-mobilization of previously deposited contaminants due to increased temperatures, change in precipitation patterns and accelerated glacier ablation potentially leading to enhanced secondary release of legacy POPs that have been stored in the cryosphere (Bogdal et al., 2009; Carlsson et al., 2012; Kallenborn et al., 2012). Permafrost thaw is expected to lead to increased mobilization and downstream transport of organic matter (OM) and OM-associated contaminants, and may also drive increased erosion and sediment transport to Arctic coastal systems (AMAP, 2017; Hanssen-Bauer et al., 2019). This may change concentrations of contaminants available for uptake at the base of the food-web and consequently also food web accumulation in affected ecosystems (McGovern et al., 2019). The glacial influence on the total POP burden to the receiving coastal environments in the Arctic is not fully understood, although some recent studies have investigated differences in contaminant concentrations in sediments between lakes and the receiving fjords (Sun et al., 2020). However, transport and fluxes of organic contaminants from meltwater to the receiving marine environment and their fate in coastal waters have gained less attention (Ma et al., 2015; Pouch et al., 2017, 2018; Sapota et al., 2009).

Freshwater delivery to the Arctic Ocean is highly seasonal, with most discharge taking place during an intensive summer melting season (ca. June–September on Svalbard). In this short period, large volumes of freshwater, and associated terrestrial material, including dissolved and particle-bound contaminants, are transported from the terrestrial environment to the coastal zone. However, little is known about the magnitude, seasonality and impact of these fluxes on the marine environment, and how these changing inputs from land could affect contaminant concentrations in coastal biota. Contaminants bound to heavier particles may settle out quickly in river estuaries, while those bound to finer suspended sediments may be transported further out into the marine environment. High inputs of SPM from land may also lead to changes in partitioning of contaminants between the dissolved and particulate phase, with important implications for bioavailability and uptake into the marine food web (Eggleton and Thomas, 2004).

In previous studies regarding POP distribution within Arctic fjords, sample collection has only included water or sediments within the fjord areas (Carlsson et al., 2012; Ma et al., 2015; Pouch et al., 2017, 2018; Sapota et al., 2009) without investigation of the associated meltwater rivers. This only allows a comparison of e.g. inner- and outer fjord locations, with the assumption that the inner stations to a greater extent reflect terrestrial sources of contaminants.

The overall aim of this study is to investigate how terrestrial secondary sources may affect occurrence and fate of PCBs and HCB in the receiving marine fjords by measuring dissolved and particle bound contaminant concentrations along the freshwater-marine

continuum (i.e. in rivers, estuaries and fjords). We hypothesize that inputs of PCBs and HCB from land will cause increased contaminant concentrations in nearshore coastal waters, but high loads of suspended sediments will lead to a reduced bioavailability of these contaminants due to 'scavenging' of dissolved contaminants.

2. Material and methods

2.1. Study area

The present study was conducted in Isfjorden area on the west coast of Spitsbergen, Svalbard (78°20'N 15°20'E). The fjord system includes several inner fjord arms, including Adventfjorden, Billefjorden and Tempelfjorden (Fig. 1), which receive freshwater mainly from marine terminating glaciers and sea ice (in Billefjorden and Tempelfjorden) and river runoff from glaciers, snow melt and precipitation on land. These fjords have polar features with local polar water in Billefjorden and sea ice cover (Billefjorden and Tempelfjorden) during wintertime, but also receive warm North-Atlantic water (Fraser, 2018). All the fjords are ice free during summer and autumn (Nilsen et al., 2008). The area around Isfjorden (and Svalbard in general) is characterized by glacial landscapes, permafrost and sparse vegetation. Consequently, the area is subject to seasonal erosion processes and transport of large amounts of sediment into the receiving coastal areas during the summer season of June to September (Bogen and Bønsnes, 2003; Forwick et al., 2010). In general, the west coast of Spitsbergen receives little precipitation (200–400 mm/year), but precipitation rates have increased by 20–30% for some stations on Spitsbergen since 1971 and other measurement stations show more frequent heavy rainfall events today compared to the 1970s (Hanssen-Bauer et al., 2019). There are three small settlements in the Isfjorden complex. Long-yearbyen, a small Norwegian settlement with ca. 2400 inhabitants, is located in the inner part of Adventfjorden. Pyramiden, an abandoned Russian mining settlement, is located in Billefjorden, and Barentsburg, an active Russian mining settlement with ca. 500 inhabitants, is located in Grønfjorden.

2.2. Sampling

Sampling was carried out between June and September 2018. Passive sampling of water and sampling of river suspended particulate matter (SPM) and marine sediments for analysis of POPs were carried out in five rivers (Sassenelva, Gipsdalselva, Adventelva, DeGeerelva and Ebbaelva) and their receiving fjord arms (Billefjorden, Adventfjorden, and Tempelfjorden (including Sassenfjorden)), and outer Isfjorden; Fig. 1, Tables S1–S2). The study rivers differ in their catchment characteristics (e.g. degree of glacial cover, size) and geochemistry (McGovern et al., 2020). Sampling sites were selected in order to provide a gradient from inner to outer fjord, and are categorized as follows: 1) rivers, 2) "inner": inner fjord sites (including glacier fronts and river estuaries), 3) "middle": mid-fjord sites in the three fjord arms studied, and 4) "outer": sites in the outer part of Isfjorden (Fig. 1).

2.3. Passive sampling of POPs in water

Passive sampling was carried out to investigate the concentration of target compounds in the aqueous phase. Passive samplers (PAS), made of silicone rubber (SR) and spiked with Performance Reference Compounds (PRCs), were deployed at 10 different river (n = 6) and marine (n = 4) locations for 12 weeks, from mid-June to mid-September (Fig. 1 and Table S1). Marine samplers were attached to a metal mesh and deployed 5 m below the surface.

River samplers were attached to a metal rod that was attached in the riverbed. Two replicates were deployed at each sampling location and field blanks were exposed to air during the deployment as well as during retrieval. Upon recovery, samplers were washed on-site with *in situ* water to remove particles and biofouling and stored in metal tins at $-20\text{ }^{\circ}\text{C}$ until further analyses at the Norwegian Institute for Water Research (NIVA), Oslo. Unfortunately, many of the PAS were lost due to currents, weather and polar bears; only Ebbaelva, Tempelfjorden and Adventfjorden samplers were retrieved (Table S1). Aqueous concentrations were calculated based on the amount of target compounds accumulated in the sampler and dissipation of PRCs, based on Smedes and Booij (2012) as described in the supplemental information.

2.4. Sampling of riverine suspended particulate matter (SPM) and marine sediments

SPM samples ($n = 5$) were collected in the same rivers as the

passive samplers were deployed in (Fig. 1, Table S2). A stainless-steel sedimentation box (Fig. S1), based on a design described in Schulze et al. (2007) was used. These boxes collect suspended sediments by reducing the incoming water flow velocity which causes particles to deposit in the container, thus providing a time-integrative collection of SPM for the exposure period. The boxes were mounted about 30 cm below the river surface and were deployed for approximately 24 h. SPM was collected from the box using a pre-cleaned metal spoon, transferred to clean glass jars and stored at $-20\text{ }^{\circ}\text{C}$. The collection capacity of the sedimentation box varies with SPM concentration, water velocity and particle size, and thus the amount of sediments collected in each river varied from ca. 10 g in Ebbaelva to 2 kg in Adventelva.

Marine sediment samples ($n = 19$) were collected with a Van Veen grab. The uppermost 0–2 cm of undisturbed sediments were sampled using a stainless-steel spoon. Sediment was transferred to pre-cleaned glass jars and kept dark and frozen ($-20\text{ }^{\circ}\text{C}$) until analysis. Sampling locations were chosen to cover a gradient from

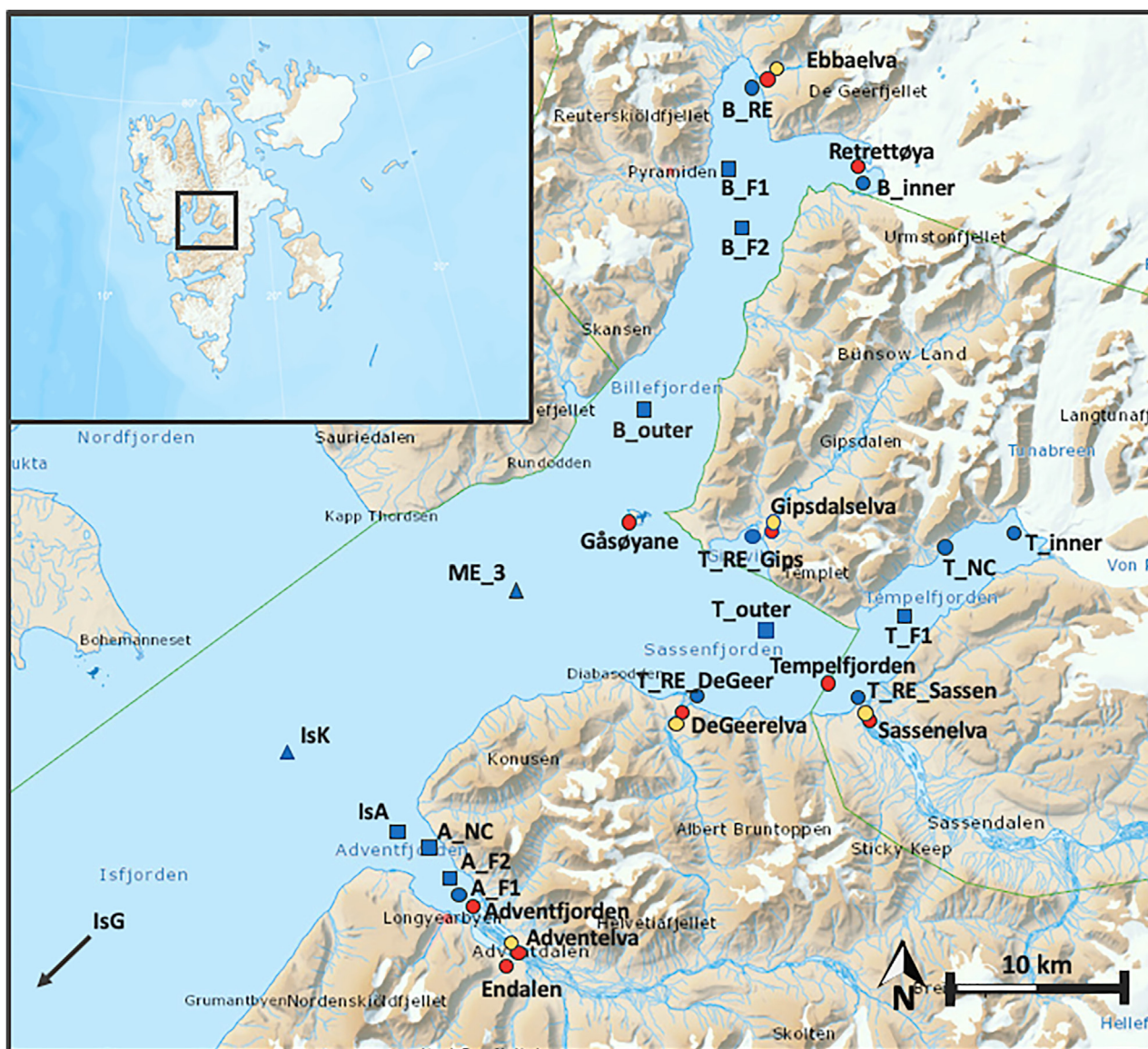


Fig. 1. Map of Svalbard and Isfjorden with Adventfjorden (south), Sassen- and Tempelfjorden (East) and Billefjorden (North). Note that the IsG sediment station is outside the map, close to the mouth of Isfjorden. Red = passive sampling stations, blue = marine sediment sampling and yellow = riverine SPM sampling stations. The map is modified from <https://toposvalbard.npolar.no> (Norwegian Polar Institute). Table S2 shows classifications (illustrated by shapes here in Fig. 1) of stations into river (circles), inner (circles), middle (square) and outer stations (triangles). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

shore (i.e. estuaries outside the studied river and glacier front sites) to outer fjord. Sampling stations were divided into categories of “Inner” (including the sub-categories near shore, glacier front and estuary), “Middle” and “Outer” stations based on their location in the fjord (Fig. 1, Table S2).

2.5. Chemical analyses

Analyses of riverine SPM, marine sediments and aqueous concentrations (PAS) are based on methods described in Pintado-Herrera et al. (2016) and are briefly described in the supporting material. All samples were analyzed for 14 target PCBs, selected to provide a range in K_{OW} and to target PCBs that are often present at detectable levels in the Arctic environment (PCB-28, -31, -44, -52, -101, -105, -118, -138, -149, -153, -156, -170, -180, -194) and HCB. The listed PCBs are hereafter referred to as \sum_{14} . Congeners included in \sum_{7} PCBs are PCB-28, -52, -101, -118, -136, -153 and -180. The passive samplers were in addition analyzed for PRCs (CB-1, -2, -3, -10, -14, -21, -50, -78, -104, -145). Separation and quantification were carried out by a gas chromatograph connected to a triple quadrupole mass spectrometer (GC-MS/MS). Instrumental parameters are provided in SI.

2.6. Supporting parameters

From each sampling station, a sub-sample was analyzed for total organic carbon (TOC) and particle size distribution (Ugelstad, 2019). Particle size distribution was determined at the University of Tromsø (Norway) using laser diffraction (Ugelstad, 2019). Sediment TOC concentrations were determined at the University of California Davis Stable Isotope Facility after removal of inorganic carbon by acidification. Data on TOC content and fine sediment fraction (<63 μm) are included in Table S2, together with other information about the sediment/SPM-sampling stations.

2.7. Quality assurance and control

Due to poor chromatographic separation of CB-28 and -31 in the sediment samples, these two congeners were quantified together and thus reported as CB-28/31. For the PRC-spiked passive samplers, interference (most likely from the PRC-compound CB-50) made it difficult to quantify CB-28 and -31 and they are thus not reported for passive samplers.

For each analytical batch, one or two procedural blanks were included. For passive samplers, field blanks and transportation blanks were analyzed; these were spiked samplers that were also used to determine initial PRC concentrations in unexposed samplers. Level of detection (LOD) was based on background levels in blank samples and signal to noise (S/N) ratios. LOD was set 3 times the mean blank concentration. If no compounds were detected in blank samples, LOD was set to $S/N = 3$.

2.8. Calculations of sediment-water activity ratios

When the riverine SPM is released into the coastal water column, the direction of transfer of hydrophobic organic contaminants in the system will be governed by the gradient or difference in activity of the chemicals in the water (A_w) and that of those sorbed to the river SPM (A_{SPM}):

$$\frac{A_{SPM}}{A_w} \quad (1)$$

A ratio <1 indicates that the SPM acts as a sink for chemicals present in the water column, while a ratio >1 predicts a desorption of chemicals from SPM into the water column. The freely dissolved concentrations (C_w) of PCBs and HCB in water are proportional to their activity. For SPM, the contaminant activity can be assumed to be proportional to C_w , which the river SPM would be able to sustain under equilibrium conditions (C_{w-SPM}). When a measurement of C_{w-SPM} is not possible, it can be modelled with the following equation, assuming that the contaminants sorb mainly to the organic carbon fraction of the sediments:

$$C_{w-SPM} = \frac{C_{SPM}}{f_{OC} \cdot K_{OC}} \quad (2)$$

where C_{SPM} is total contaminant concentration in SPM (ng/kg dw), f_{OC} is the fraction of organic carbon in SPM and K_{OC} is the organic carbon (TOC)-normalized partition coefficient for our chemicals of interest (L/kg). Combining equations (1) and (2) gives:

$$\frac{A_{SPM}}{A_w} = \frac{C_{w-SPM}}{C_w} = \frac{C_{SPM}}{f_{OC} K_{OC} C_w} \quad (3)$$

The activity ratio can therefore be estimated from the contaminant concentrations measured in the SPM and estimated for the C_w in the water column, K_{OC} can be obtained from the literature since these are not available specifically for this particular SPM (4). Assuming equilibrium partitioning between water and organic carbon, K_{OC} can be calculated from the following relationship (Chiou et al., 1983; Karickhoff et al., 1979; Xia, 1998).

$$\log K_{OC} = 0.97 \log K_{OW} - 0.50 \quad (4)$$

as a function of the $\log K_{OW}$ of the PCBs (K_{OW} values from Hawker and Connell (1988)). For HCB, $\log K_{OW} = \log K_{OC} = 5.5$ was used, and with K_{OW} as given in Smedes et al. (2009).

2.9. Statistical analyses

R (R Core Team, 2020) were used for statistical analysis. All values below LOD were replaced with a random value between $\frac{1}{2}$ -LOD and LOD. Values below LOD were not included in any sum (e.g. \sum_{14} PCBs) calculations. For statistical analysis, a p-value of ≤ 0.05 was chosen as threshold for significance. Multivariate statistical analyses were carried out using the ‘vegan’ package (Oksanen et al., 2017) in R. Briefly, principal components analysis (PCA) was used to explore variation in contaminant concentrations in riverine SPM and marine surface sediments, while redundancy analysis (RDA) was used to assess the variance in the dataset explained by fjord and sampling location within the fjord (as defined in section “Sampling”; including rivers, river estuaries, glacier fronts, as well as “Middle” and “Outer” stations).

3. Results and discussion

To the authors best knowledge, this is the first study measuring riverine SPM- and aqueous concentrations of PCBs and HCB in rivers, their estuaries and their receiving high Arctic fjords simultaneously. Both dissolved (PAS) and particle associated (marine surface sediments and riverine SPM) PCBs and HCB were detected in the analyzed samples (Tables S3 and S4), supporting previous indications of transport from secondary POP sources to marine Arctic environments (Carlsson et al., 2012). Concentrations of all analyzed compounds in all samples can be found in SI (Tables S3 and S4).

3.1. Water and sediment concentrations

Among the analyzed compounds, HCB dominated in the aqueous phase, ranging from 14 to 66 pg/L with highest concentrations in Ebbaelva (Table S4). To our knowledge, no previous studies have reported PCB or HCB concentrations in river water from Svalbard. Slightly lower aqueous concentrations (2–8 pg/L) of HCB have been measured in seawater from Kongsfjorden and Liefdefjorden (Hallanger et al., 2011), and in surface seawater outside Western Spitsbergen (Gioia et al., 2008). However, in both these previous studies, water sampling has been carried out with high volume water samplers (i.e. active sampling). Allan et al. (2011) reported dissolved concentrations of HCB between 48 and 117 pg/L in seawater around Bjørnøya using passive sampling.

$\sum\text{PCB}_{12}$ (CB-28 and -31 not quantified) ranged from 0.7 to 4.3 pg/L in seawater from Adventfjorden, Tempelfjorden and river Ebbaelva (Table S4). $\sum\text{PCB}_7$ (except PCB-28) ranged between 0.4 and 3.8 pg/L in these samples and are comparable to concentrations measured in surface water using active sampling on the west coast of Spitsbergen (range $\sum\text{PCB}_7$: 3.11–5.99 pg/L in Kongsfjorden) and in the Arctic Ocean (0.3–2.8 pg/L for $\sum\text{PCB}_{12/13}$) (Carrizo and Gustafsson, 2011; Gioia et al., 2008; Hallanger et al., 2011). Although the PCB congeners analyzed in these studies differed, and although there is high analytical uncertainty associated with these low concentrations, the concentrations observed in the current study do appear to fall within the reported range from the Barents Sea region. However, since only three passive samplers were retrieved, spatial patterns in aqueous concentrations of PCBs and HCB cannot be determined. While we do use these PAS data to provide preliminary coarse estimates of riverine transport and to estimate sediment/water activity ratios, we recognize that these estimates carry a great deal of uncertainty.

$\sum\text{PCB}_{14}$ and HCB concentrations were low in riverine SPM, ranging from n. d.-28 pg/g dw and 16–100 pg/g dw respectively. However, it is important to note that SPM concentrations in the investigated rivers are highly seasonal and can also change strongly

in response to high flow events, highlighting the “snapshot” nature of these samples. Concentrations were similarly low in surface sediments from inner fjord stations (ranging from 12–170 pg/g dw and 10–110 pg/g dw for PCBs and HCB, respectively) and increased toward the outer fjord (630–880 pg/g dw of $\sum\text{PCB}_{14}$ and 530–770 pg/g dw of HCB) as shown in Fig. 2 and Table S3.

The highest sediment concentration of $\sum\text{PCB}_{14}$ (1600 pg/g dw) was found in marine sediment collected outside Pyramiden (sample B_F2), which is likely attributable to local pollution from the abandoned Pyramiden mine settlement (Evenset et al., 2009; Jartun et al., 2009; Pedersen et al., 2011). Concentrations of $\sum\text{PCB}_7$ (and HCB (Table S1) at the outer stations were comparable with levels previously reported for surface sediments around Svalbard, which typically range between 500 and 1500 pg/g dw for $\sum\text{PCB}_7$ and 400–800 pg/g dw for HCB (Green et al., 2010; Zaborska et al., 2011).

Riverine SPM samples were collected with sedimentation boxes (Fig. S1). These boxes are likely to systematically under-sample the finest particles since they are less likely to settle. This particle fraction is often the most organic rich, meaning that they have higher sorption capacity for hydrophobic compounds compared to larger, less organic rich particles and hence, a different sampling set-up might have provided slightly higher concentrations of the studied compounds compared to this set-up.

3.2. Contaminant gradients

As outlined above, we observed a strong spatial gradient in HCB and PCB concentrations, with low concentrations in riverine SPM and estuarine/glacially-impacted sediments and increasing concentrations toward the outer fjord (Fig. 2, Table S3). This increase from rivers to outer fjord suggests that, while rivers do deliver PCBs and HCB to the marine environment, the low concentrations in riverine SPM compared to marine sediments further out from land results in a ‘dilution’ of sediment contaminant concentrations in nearshore coastal waters that receive large inputs of relatively

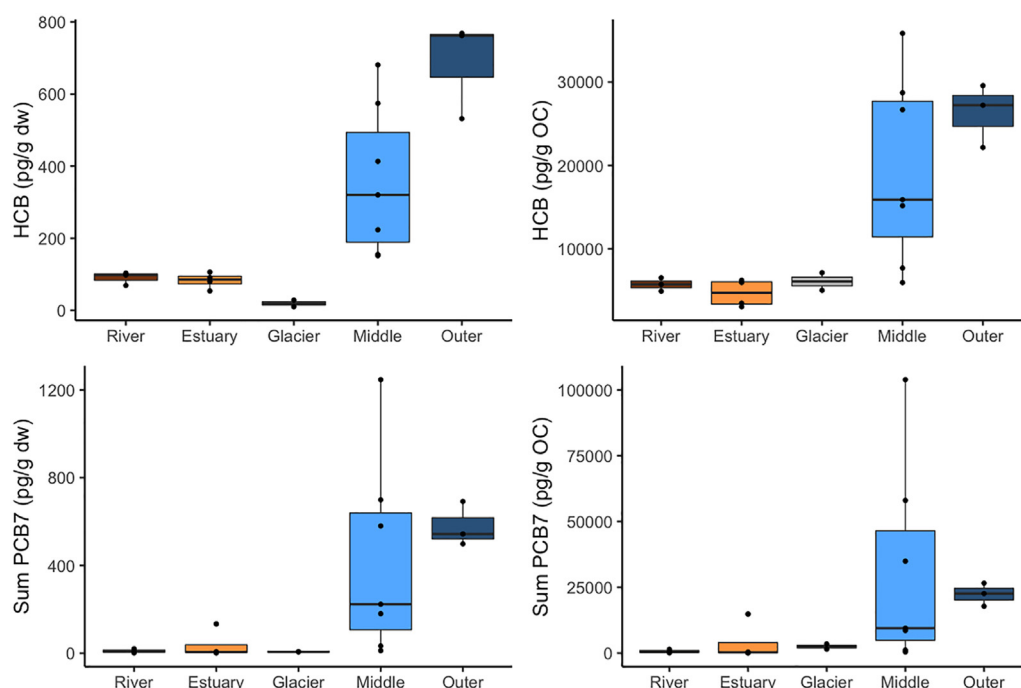


Fig. 2. Concentrations of HCB (top left) and PCB₇ (bottom left) increased from riverine SPM, estuaries and glacier fronts to outer marine sediments both before and after OC normalization. High concentrations observed for some ‘middle’ fjord stations reflect historical contamination in Billefjorden.

uncontaminated terrestrial particulate matter. This dilution effect appears to play a key role in structuring surface sediment contaminant concentrations in the Isfjorden system.

A similar increase from rivers to the outer fjord was also observed after OC-normalization of PCB and HCB concentrations (Fig. 2). The higher absolute and OC-normalized concentrations observed for outer fjord stations (ME_3, IsK and IsG; Table S2) than for strongly terrestrially-influenced inner fjord sites (and in riverine SPM) suggest that long-range marine and atmospheric transport are the main sources of these contaminants to Isfjorden, rather than secondary emissions from land, and that this gradient cannot only be explained by higher inorganic sediment loads in rivers and inner fjords. Prior to OC-normalization, mean concentrations of $\sum\text{PCB}_7$ in outer fjord sediments were approximately 58-fold higher than in riverine SPM, with an 8-fold difference observed for HCB. After OC-normalization, $\sum\text{PCB}_7$ concentrations in outer fjord sediments were 5-fold higher than in riverine SPM and inner fjord sediments, HCB concentrations were 33-fold higher. The much larger contrast in absolute and OC-normalized $\sum\text{PCB}_7$ than for HCB between riverine SPM and outer fjord sediments suggests that inputs from land may play a more important role in coastal cycling of HCB than for PCBs. This is also consistent with our preliminary observations of relatively high HCB concentrations in river water (Table S4).

The strong gradients in contaminant concentrations observed from the inner to outer fjord (Fig. 2) are further illustrated in the results from the PCA analysis (Fig. 3), which shows that differences in overall contaminant concentrations in sediments are driving most of the variability between sites rather than differences in congener distribution. Inner fjord stations (including river estuaries and glacier fronts) separated from sites that are further offshore (the mid and outer fjord stations) along PC1, which accounted for 90.5% of the total variation in the dataset (Fig. 3) and had strong positive loadings for all contaminants. Billefjorden's history of local contamination (Pyramiden settlement; (Evenset et al., 2009; Jartun et al., 2009), drives a separation along PC2, although only 7.2% of the total variation in the dataset is explained by this PC (Fig. 3). We also found that many congeners present in Russian PCB mixtures

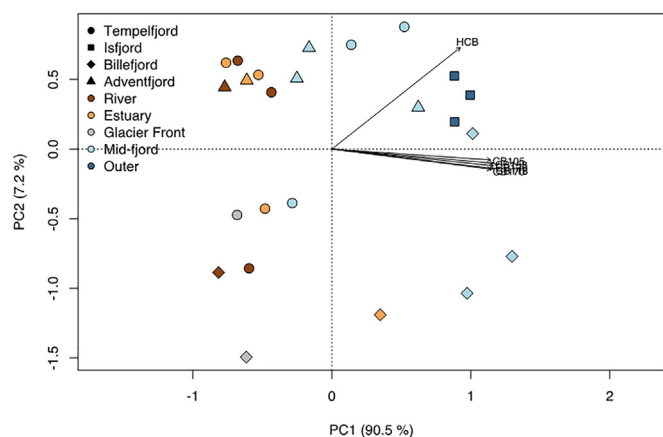


Fig. 3. Principal components analysis (PCA) of log-transformed contaminant concentrations (only including compounds where at least 20% of the samples were above the LOD: 105, 118, 138, 153, 170 and HCB). PC1 explains 90.5% of the variation in the dataset, while PC2 explains 7.2% of the variance in the dataset. Sampling fjord is indicated by shape, while sampling location ("site type") within the fjord is indicated by colour. Based on RDA variance partitioning, together "site type" and "fjord" explain 73.2% of the variance in the dataset ($P = 0.001$), with partial effects of 36.9% for "site type" ($P = 0.001$), and 17.5% for fjord ($P = 0.005$). Estuary and glacier front represents samples from the inner stations. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

were represented in samples from Billefjorden (Fig. 4). However, exclusion of Billefjorden from the analysis does not change these results, indicating that differences in local contamination are not driving these results. Furthermore, RDA variance partitioning (Fig. S2) revealed that location within the fjord (e.g. site type) was a better predictor for contaminant concentrations than fjord (explaining 36.9% vs. 17.5% of the variance), despite the known between-fjord differences in contamination history (Fig. S2). This further highlights the key role played by large inputs of terrestrial particulate matter (with low contaminant concentrations) in driving spatial patterns in contaminant concentrations through dilution.

These findings contradict our hypothesis of terrestrial inputs as an important source of POPs to Svalbard's coastal waters, as well as the results of previous studies of spatial distribution of PCBs and HCB in Svalbard fjords (e.g. Wijdefjorden and Woodfjorden; NE Spitsbergen and Hornsund, Kongsfjorden and Adventfjorden; W Spitsbergen), where highest concentrations of $\sum\text{PCB}_7$ and HCB were found in surface sediments from the inner parts of the fjords, which the authors related to influence of terrestrial/meltwater runoff (Pouch et al., 2017, 2018; Sapota et al., 2009). However, rivers were not included in those studies, and the inner fjord sites were further away from land outflows such as rivers and glaciers (~2–7 km) compared to the present study, where we were able to sample directly in river mouths and shallow waters (down to 7 m depth) since we were working from small boats.

Several factors make it difficult to assess the actual transport and fate of POPs that are stored in- and released from ice: Firstly, the more volatile compounds (i.e., HCB and low chlorinated PCBs) are subject to re-evaporation from the glacial surface into the atmosphere (Hung et al., 2016). Secondly, meltwater might consist of a mixture of melted ice of post- and pre-industrial origin. Direct (and seasonally-resolved) measurements of meltwaters will help to better understand the importance of snow and glaciers as secondary sources of contaminants.

Our results suggest that input of terrestrial/riverine sediments with low contaminant concentrations is the dominant factor governing the spatial distribution of PCBs and HCB within Isfjorden. Reasons for the relatively low levels of SPM-associated contaminants observed in the present study as well as in a previous study in Kongsfjorden (Ma et al., 2015) could be that riverine particles consist of a mixture of eroded surface soil, permafrost- and particulate matter from glacial abrasion processes, which has had limited exposure to the ambient environment and therefore limited possibility to accumulate local- and LRT contaminants. Seen in the light of climate change occurring in the Arctic, it cannot be excluded that increased erosion and glacial meltwater could lead to lower contaminant concentrations in runoff from terrestrial catchments, while elevated inputs of terrestrial particles may also result in dilution of PCBs and HCB in nearshore marine sediments, which could in turn lead to lower exposure to POPs for coastal biota (Hanssen-Bauer et al., 2019; McGovern et al., 2019).

The impact of terrestrial run-off on concentrations of organic contaminants in coastal sediments is also apparent in contaminant profiles between sites. In particular, PCB-52, which in recent studies has been found to be a major congener in surface soils, vegetation and surface snow in Svalbard (Aslam et al., 2019; Bartlett et al., 2019), was more prominent in terrestrial runoff (riverine SPM) and in sediments in river estuaries and at glacier fronts than in outer fjord sediments (Fig. 4). Relative concentrations of HCB and PCBs also differed along these gradients, with HCB concentrations typically higher than PCB concentrations in riverine SPM and estuarine sediments, and similar to or lower than PCB concentrations in outer fjord sediments (Fig. 4, Table S3). This is also consistent with our observations that, although still high, HCB

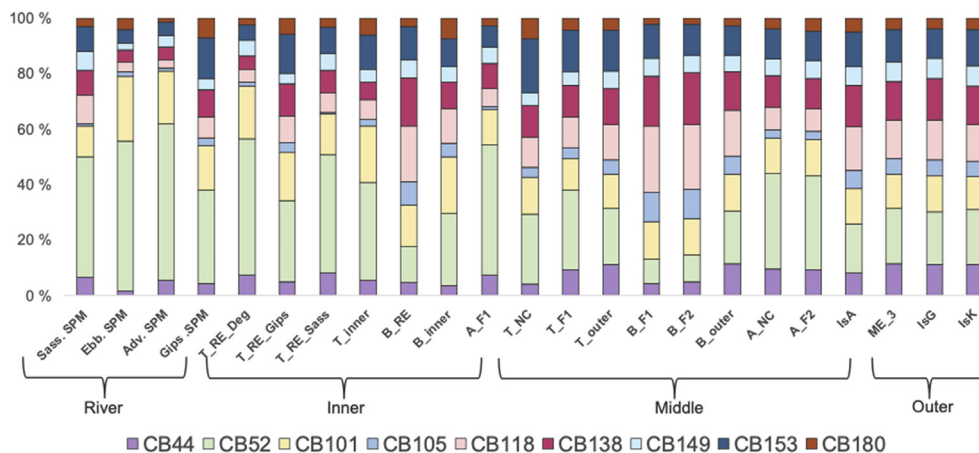


Fig. 4. Relative distribution of the most abundant of the analyzed PCB-congeners in riverine SPM and sediment samples.

concentrations in outer fjord sediments were less elevated relative to riverine SPM and inner fjord sediments than was observed for PCBs, suggesting that terrestrial runoff may play a more important role for cycling of HCB compared to PCBs in the Isfjorden system.

3.3. Riverine contribution of contaminants to fjords

Atmospheric deposition (dry deposition) to the ocean around Svalbard has been estimated to be about 0.0023 kg/km² year⁻¹ for ∑PCB₇ and 0.010 kg/km² year⁻¹ for HCB (Green et al., 2010). Due to local differences in wet deposition (precipitation), the total deposition in fjords around Svalbard may differ from those in more open sea areas, but the calculations still provide an estimate of the size order of LRT. If the numbers from Green et al. (2010) are applied to Isfjorden (3084 km²), this corresponds to estimated inputs of 7093 g/year of ∑PCB₇ and 30 800 g/year of HCB through dry deposition. These estimates are based on limited data, and represent gross rather than net deposition, since they do not account for volatilization. However, volatilization is likely low for this cold Arctic system, particularly for higher molecular weight congeners. From local sources (Pyramiden and Barentsburg), the Norwegian Water Resources and Energy Directorate (NVE) has estimated a yearly flux of 2800 g ∑PCB₇/year (Pedersen et al., 2011).

Due to few results for contaminant concentrations and a lack of seasonal river discharge data it was impossible to calculate reliable data for riverine fluxes of PCBs and HCB. However, in order to assess the contribution from rivers in relation to other sources, a rough estimate was made based on previously reported annual sediment yields for catchments on Svalbard (as summarized in Hanssen-Bauer et al., 2019) and the range of PCB and HCB measured in

riverine SPM (Tables 1 and S3). Although these estimates are highly uncertain, relying on extrapolation of PCB and HCB concentrations from a very small number of riverine SPM samples, and on extrapolation of sediment yields from a limited number of Svalbard rivers, they provide a first estimate of riverine transport vs other sources of PCB to the marine environment on Svalbard. Our estimates of inputs of SPM-associated POPs to Isfjorden in the present study are <1–28 g/y of PCB₁₄ and 16–100 g/y of HCB (Table 1).

This suggests that contribution from land would be at least two orders of magnitude lower than other sources of inputs to Isfjorden, such as direct atmospheric deposition and local sources. The transport to Isfjorden with oceanic currents is not considered here, since the magnitude of the Atlantic water influence to the fjord is considered to be minor relative to atmospheric contribution of legacy POPs (Carlsson et al., 2018). Although our estimated fluxes from land represent conservative estimates, as they do not include dissolved HCB or PCB, and use low estimates of sediment yields, using highest reported sediment yields for non-glaciated and glaciated areas on Svalbard, would only lead to an 8–9 fold increase in estimated SPM-associated POPs fluxes to Isfjorden, suggesting that terrestrial inputs can certainly act as a source of PCB and HCB to Svalbard’s coastal waters, but to a lesser degree that direct atmospheric deposition. However, once again, these estimates are coarse and should be treated carefully in comparison with other studies. Nevertheless, they provide preliminary estimates of contaminant transport under given conditions, and with the purpose to provide a semi-quantitative comparison of the magnitude of input of contaminants from different sources, and the relative importance of the terrestrial environment as a source of contaminants to Arctic coastal waters.

Table 1
PCB and HCB inputs from different sources to Isfjorden.

Sources	Compound(s)	Input (mass/year)	Sources
Atmospheric deposition ^a	∑PCB ₇	7093 g/y	Calculations based on Green et al. (2010)
	HCB	30 800 g/y	
Local sources ^b	∑PCB ₇	2800 g/y	Jartun et al. (2009); Pedersen et al. (2011). This study.
Terrestrial SPM ^c	∑PCB ₁₄	<1–28 g/y	
	HCB	16–100 g/y	

^a Based on atmospheric dry-deposition of 0.0023 kg/km²/year PCB₇ and 0.010 kg/km²/year HCB and Isfjorden’s surface area (3084 km²; Nilsen et al., 2008).
^b From the local settlements Pyramiden and Barentsburg.
^c Based on previously published sediment yields for Svalbard catchments for non-glaciated and glaciated areas, and the total area (7309 km²) and degree of glaciation (~40%) of the Isfjorden catchment (Forwick et al., 2010). Since reported sediment yields are highly variable, we took a conservative approach, using lowest reported sediment yields for both non-glaciated (28 t/km²/y) and glaciated (303 t/km²/y) areas, resulting in estimated SPM fluxes to Isfjorden of 1009 kt/y. We then paired estimated SPM flux with min-max ∑PCB₁₄ and HCB concentrations measured in riverine SPM to estimate annual sediment-associated fluxes of PCB and HCB from land.

Higher amount of dissolved HCB (>95% of total) compared to particulate HCB in Ebbaelva suggests that terrestrial contribution of HCB may be mostly linked to the dissolved fraction for this river. For the sum of PCBs, about 50% of the total was measured in the dissolved fraction. A rough estimate on dissolved PCB and HCB input from rivers based on PAS results in the present study can be found in Table 1. Despite high levels of SPM in the studied rivers and the sorptive nature of POPs towards particles - and organic matter in particular, this study also demonstrates the importance of taking dissolved contaminants into consideration when transport and fluxes are quantified in riverine runoff. It is also likely to be a high degree of seasonal variability in dissolved contaminant transport (as is also the case for particulate organic contaminants). In particular, during spring freshet, there is a high potential for remobilization and transport of dissolved contaminants previously deposited on soils and snow, when snow is melting and water flows through the uppermost organic soils at the snow-soil interface. This highlights the importance of future studies focusing on seasonality in contaminant concentrations and fluxes from land to Arctic coastal ecosystems.

3.4. Particle -water distribution

Due to low PCB and HCB concentrations in riverine SPM compared to sediment in the outer parts of the fjords, we hypothesized that terrestrial particles might have the potential to sorb freely dissolved contaminants from the water column as they enter the marine environment (i.e. have a “scavenging” effect. When the riverine SPM is released into the coastal water column, the direction of transfer of hydrophobic organic contaminants in this system will be governed by the gradient and differences in chemical activity (which is proportional to their concentration) in the water (A_w) and that of those sorbed to the river SPM (A_{SPM}). The SPM acts as a potential sorbent of these compounds in cases where $A_{SPM}/A_w < 1$ (equation (3)). Due to uncertainties in K_{oc} , a ratio between 0.2 and 5 is approximately where the SPM is predicted to be close to equilibrium with the aqueous phase (Prokeš et al., 2012). A_{SPM}/A_w was estimated for SPM samples from rivers with PAS data available from the near-shore estuarine fjord area (Adventelva and Sassenelva, Fig. 5). Values of K_{oc} had to be obtained from the literature since they are not available specifically for the SPM investigated in the present study. It is important to keep in mind that these estimates are based on single snapshot samples of riverine SPM for each river, and on a single deployment of passive samplers in receiving marine waters ($n = 2$ replicates retrieved for Adventfjorden and only one replicate retrieved for Tempelfjorden). Given the limited data available and the fact that PCB concentrations in river and fjord waters and SPM are likely to exhibit high seasonal variability, these results should be treated as a preliminary exploration of the potential for terrestrial particles to have a “scavenging” effect.

The SPM acts as a potential sorbent of freely dissolved hydrophobic contaminants in cases where $A_{SPM}/A_w < 1$.

As shown in Fig. 5, the SPM sorption capacities show a slight “scavenging” effect of PCBs and HCB from the water column and onto the riverine particles for mid- and high chlorinated PCBs. Overall, our predictions rather show conditions close to equilibrium (0.2–5, e.g. Prokeš et al. (2012)), due to the low TOC fraction associated with the SPM for these rivers (1.4–1.8%, Table S2). Equation (3) shows that the activity ratio is dependent on the value of K_{oc} , and that a higher value would decrease the ratio further towards a situation where SPM acts as a sink for PCBs/HCB present in the water column ($A_{SPM}/A_w < 1$). The K_{oc} values used can be

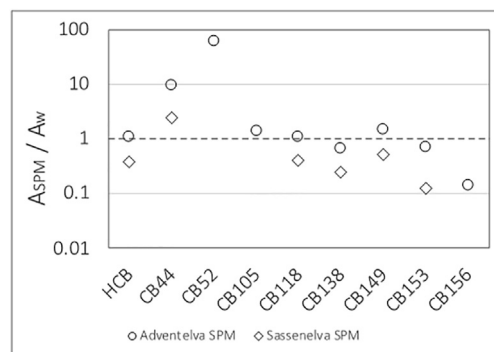


Fig. 5. Suspended particulate matter (SPM)/water-activity ratios for the PCBs. Only congeners with concentrations > LOD in both aqueous phase and SPM is shown in the figure, hence no ratios were calculated for PCB-101, -170, 180 and 194. PCB-52 was <LOD in the water sample from Tempelfjorden (where Sassenelva drains), while PCB-105 was <LOD in Sassenelva SPM.

considered as conservative, as real values can be one or two orders of magnitude higher depending on the type of particulate organic carbon (Aslam et al., 2019). Furthermore, as previously mentioned, the sedimentation boxes used in the current study are also likely to have led to systematic under-sampling of finer and more organic-rich SPM, which would also lead to an underestimate of the sorption capacity of the SPM. Much of the area in Isfjorden contains coal which is also present in marine and riverine sediments and coal seems to have a higher affinity for PCBs and HCB compared to other organic matter (Evenset et al., 2009; Lohmann et al., 2005). Differentiation between coal and other organic matter was not part of this study, but we suggest that future studies should investigate this in more detail, since a potential sorption of freely dissolved contaminants could lead to reduced bioavailability and food web uptake of POPs in coastal waters. Future studies should also consider potential between-catchment differences as well as seasonality in sorptive capacity of terrestrial particles and A_{SPM}/A_w as well as how scavenging effects could differ between a broader range of congeners.

4. Conclusions

This study has shown that inputs from land play an important role in governing concentrations of PCB and HCB in the receiving marine environment, but not in the way we had expected. While we expected that inputs from land would lead to elevated PCB and HCB concentrations in Svalbard's coastal environment, our observations suggest that contribution from terrestrial sources is low. Instead, high fluxes of SPM with relatively low PCB and HCB concentrations from Svalbard's glaciated catchments to the marine environment result in strong dilution of POPs in coastal sediments, leading to strong spatial gradients in sediment concentrations from inner to outer fjord stations. By combining riverine SPM measurements with passive sampling of dissolved POPs, we also explore the potential for terrestrial particles to scavenge freely dissolved POPs from coastal waters, possibly leading to reduced availability for uptake into coastal food webs. These observations suggest that climate-change driven remobilization of organic contaminants from land to coast may be mitigated by concurrent increases in particle delivery, particularly in glaciated regions, with the potential to lead to reduced POPs concentrations in impacted coastal waters, sediments and food webs. These results also point to a need for an increased research focus on the role of inputs from land on contaminant cycling and effect in Svalbard's coastal ecosystems.

CRedit author statement

Sverre Johansen: Conceptualisation, Methodology, Investigation, analysis, writing. Amanda Poste: Conceptualisation, Formal analysis, writing, Funding acquisition. Ian Allan: Validation, Methodology, Supervision, writing. Anita Evenset: Conceptualisation, writing. Pernilla Carlsson: Methodology, Investigation, Supervision, writing.

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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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envpol.2021.116963>.

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