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Arctic fjord: The influence of ocean currents vs. glacial runoff**

The Spatial Distribution of Organochlorine Pesticides and Halogenated Flame Retardants in the Surface Sediments of an Arctic Fjord: the Influence of Ocean Currents *vs*. Glacial Runoff

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Abstract

Selected organochlorine pesticides (OCs) and halogenated flame retardants (HFRs) were analyzed in surficial fjord sediments collected down the length of Kongsfjorden, Svalbard in the Norwegian high Arctic. Hexachlorocyclohexane (α-HCHs) was found to be the most abundant OC in the sediment, followed by BDE-209 > chlordane > α-endosulfan > Dechlorane Plus (*anti*-DP)> trifluralin Concentration ranges were high over the relatively small study area of the fjord (e.g. ΣHCH: 7.2-100 pg g^{-1} dry weight (dw)), with concentrations broadly similar to, or lower than, measurements conducted in other parts of the Arctic. Concentrations of legacy OCs, including both HCH isomers and chlordane showed a decreasing trend from the outer, seaward end of the fjord to the inner, glacier end of the fjord. Conversely, sediment concentrations of α- and β-endosulfan (0.1-12.5 pg g^{-1} dw) increased from the outer fjord to the inner fjord. This contrasting pattern may be attributed to the influence of historical *vs.* contemporary sources of these chemicals to the fjord area, whereby the North Atlantic/West Spitzbergen oceanic current dominates the transport and input of the legacy OCs, whereas atmospheric deposition and meltwater runoff from the glaciers influence the inner fjord sediments for endosulfan. Interestingly, BDE-209 and Dechlorane Plus did not reveal any clear spatial trend. It is plausible that both glacial runoff and oceanic current end members are playing a role in introducing these chemicals to the fjord sediments. The relatively low fractional abundance of the *syn*-DP isomer (f_{syn}), however, indicates the long-range transport of this chemical to this Arctic site.

Keywords: Organochlorine pesticides, Halogenated flame retardants, Surface sediment, West Spitsbergen Current, Glaciers, Arctic, Kongsfjorden

Introduction

Persistent organic pollutants (POPs) are globally distributed chemicals that are present in the Polar Regions and remote ocean environments (MacDonald et al., 2000; Muir and de Wit, 2010). They have been observed in a variety of environmental compartments (air, water, soil, sediments) and can accumulate in marine and terrestrial foodwebs due to their physical-chemical properties (Riget et al., 2010). For legacy chemicals like organochlorine pesticides (OCs) and polychlorinated biphenyls (PCBs) their fate in the Arctic and transfer between key environmental compartments is still the focus of interest, particularly in light of climatic perturbations affecting chemical transport pathways (Ma et al., 2011). For newer chemical substances such as certain pesticides, deca-BDE and alternative halogenated flame retardants (HFRs), such as the highly chlorinated flame retardant Dechlorane Plus (DP), their sources to the Arctic and subsequent fate are not well understood.

A key aspect to the fate of persistent chemicals is their transfer to the ocean environment and burial in benthic sediments, the latter considered an important long term global sink for these chemicals (Dachs et al., 2002). It is estimated that ~90% of sedimentary burial of organic matter occurs in the continental margin sediments (Hedges and Keil, 1995), with shelf sediments considered a significant reservoir on a global basis and possible semi-permanent sink of POPs (Jonsson et al., 2003). In cold environments climate change is affecting the rates of glacier ablation and melt, with evidence from Alpine lake sediment profiles showing remobilization of glacier-associated POPs following ice retreat/melt (Bogdal et al., 2009). This effect has not been observed in the Arctic although enhanced melting of coastal glaciers will influence sedimentation rates in coastal waters due to changing melt water runoff (Darby et al., 2009) and this may also influence contaminant occurrence in coastal waters and sediments.

To investigate the influence of glacial runoff on contaminant fate, Kongsfjorden, a glacial fjord located on the west coast of Svalbard at 79° N, 12° E, was selected as a study location. This fjord is influenced by both Atlantic and Arctic water masses with incursions of the relatively warm North Atlantic current making this fjord system sub-Arctic rather than Arctic. Inputs from large tidal glaciers create steep gradients in sedimentation rates and salinity along the length of the fjord (Figure 1). Thus, the partially closed water body of Kongsfjorden is viewed as an ideal site for monitoring environmental changes associated with a warming Arctic (Hop et al., 2002; Svendsen et al., 2002). In this study, levels of selected OCs (HCHs, chlordane, endosulfan and trifluralin) as well as HFRs (BDE-209 and DP) were measured in surface sediment samples, with the aim of examining spatial variations in sediment-bound concentrations, and relating these to the influence of glacial runoff and oceanic transport, as well as other factors, on chemical fate. The measurement of HFRs is particularly interesting as these chemicals have high-production volumes, have widespread use and, in some cases, are sufficiently persistent to undergo long-range transport (de Wit et al., 2010).

Materials and Methods

Sediment sampling

Surficial sediment samples (0-2cm in depth) were collected from 27 locations during the period of July 21-24, 2009 along the length of Kongsfjorden. The sediment samples were collected with a stainless steel box corer and scooped using a pre-cleaned stainless steel scoop and placed into baked and solvent-rinsed aluminum containers. All samples were stored at -20°C until further analysis. Sediments were then carefully oven dried and sieved (100 mesh size sieve) before gentle grinding and homogenization. The sampling information is summarized in Table S1.

Chemicals

88   All solvents (methanol, acetone, dichloromethane and *n*-hexane) were purchased from LGC

89 Standards, Germany. They were residue grade and additionally distilled in a full glass unit prior to use.

90 The samples were analyzed for the following OCs, including: α/γ-HCHs, chlordane, α/β-endosulfan

91 and trifluralin, which were purchased from Dr. Ehrenstorfer GmbH (Germany) as well as BDE-209

92 and DP, which were purchased from Wellington Laboratories. Trifluralin-d_{14}, ^{13}C–BDE–209,

93 ^{13}C–synDP and ^{13}C-HCB were purchased from Cambridge Isotope Laboratories. PCB-207 were also

94 obtained from Dr. Ehrenstorfer GmbH. Both ^{13}C-HCB and PCB-207 were used as injection standards

95 (see details below).

96 *Extraction, clean-up and analysis*

97   Sediment extraction, clean-up and analysis of the samples were based on our previously published

98 method (Zhong et al., 2012). Briefly, 10 g of dried sediment were spiked with the surrogate standard

99 trifluralin-d_{14}, HCH-d_6, and ^{13}C–BDE–77 prior to extraction, then Soxhlet extracted with

100 dichloromethane (DCM) and purified on 10% water-deactivated silica column. The extracts were

101 purified by eluting with 20 mL of *n*-hexane (fraction 1), followed by 30 mL of DCM/acetone (1:1)

102 (fraction 2), and evaporated to a final volume of 50 μL. ^{13}C-HCB and PCB-207 were added to fraction

103 1 and 2 as injection standard, respectively. Analysis of OCs and DPs was performed via GC/MS (6890

104 GC/5975 MSD) equipped with a 30 m HP-5MS capillary column (Diameter: 0.25 mm; Film thickness:

105 0.25 µm, Agilent Technologies, US) operating in electron capture negative chemical ionization mode

106 (ECNCI). BDE-209 was analyzed using GC-MS-ECNI equipped with a 15 m DB-5MS capillary

107 column (Diameter: 0.25 mm; Film thickness: 0.10µm, Agilent Technologies, US). The fragment

108 masses (m/z) monitored for quantification and quality control are summarized in Table S2 in the

109 supporting information.

110 *QA/QC*

111 A method blank was run for each batch of samples and six method blanks were obtained in total.

112 Mean absolute blank values of chemicals ranged from 0.27-14 pg per sample. Method detection limits

113 (MDLs) were derived from mean field blank plus three times the standard deviation (σ). For analytes

114 not present in the field blanks then instrumental detection limits (at signal-to-noise ratio of

115 three-to-one) were used to derive MDLs instead. Taking the average mass of sediment extracted as 10

116 g, then MDLs ranged from 0.067 to 2.6 pg g^{-1}. The surrogate recoveries ranged from 60% to 114%

117 (mean: 86 ± 21%) for ^{13}C–BDE–77, from 72% to 121% (mean: 101 ± 14%) for HCH-d_6 and from

118 57% to 137% (mean: 96 ± 19%) for trifluralin-d_{14}, respectively. The concentrations of target

119 compounds reported were corrected with the corresponding surrogates (e.g. HCH-d_6 for α/γ-HCHs

120 and *cis-/trans-* chlordane; trifluralin-d_{14} for trifluralin; ^{13}C–BDE–77 for BDE-209 and DPs).

121

122 **Results and Discussion**

123 *Concentrations of OCs in surface sediment of Kongsfjorden*

124 *Hexachlorocyclohexanes and chlordane*

125 Both α- and γ-HCH were detected in the surface sediment samples and their concentrations are

126 shown in Table S3. Generally, compared with other OCs, α-HCH was the most abundant compound in

127 the sediment with concentrations ranging from 7.0 to 99.2 pg g^{-1} dry weight (dw) with an arithmetic

128 average (SD) of 29.9 ± 25.8 pg g^{-1} dw. However, concentrations of γ-HCH (lindane) was significantly

129 lower ranging from <MDL to 2.06 pg g^{-1} dw, and average (SD) of 0.45 ± 0.47 pg g^{-1} dw. The sum of

130 α- and γ-HCH in all surface sediment samples ranged from 7 to 100 pg g^{-1} dw. Our results are

131 comparable with earlier measurements of HCHs in marine sediments of the Bering Strait and Chukchi

132 Sea, which ranged from 40 to 60 pg g^{-1} dw (Iwata et al., 1994). Generally, HCHs concentrations in

133 Kongsfjorden were lower than those reported from recent studies conducted on coastal and marine

134 sediments elsewhere in the Arctic. Specifically, our results were lower than HCHs observed in surface

135 sediments of Tromsø harbor, Norwegian Sea (40-790 pg g^{-1} dw) (Dahle et al., 2000) and also lower

136 than those reported towards the Russian side of the Barents Sea (e.g. Guba Pechenga, 50-680 pg g^{-1}

137 dw; Pechora Sea (located in th esouth-eastern part of Barents Sea), 200-1000 pg g^{-1} dw; Kola Bay,

138 100-400 pg g^{-1} dw with α-HCH only), (Loring et al., 1995; Savinov et al., 2003). Moreover, α-HCH

139 concentrations were comparable to concentrations observed in fresh water lake sediments in the Arctic,

140 although levels of γ-HCH were significantly lower. Examples include lakes Ellasjøen and Øyangen

141 located on Bjørnøya Island, Norway (α-HCH < 200 pg g^{-1} dw; γ-HCH ~ 400 pg g^{-1} dw) as well as

142 Lake DV09 on Devon Island, Canada (α-HCH 120 pg g^{-1} dw; γ-HCH 80 pg g^{-1} dw) (Evenset et al.,

143 2004; Stern et al., 2005). Very high HCH concentrations were observed in lakes located in the

144 Ny-Ålesund region of northern Svalbard (~1000s pg g^{-1} dw) (Jiao et al., 2009). The reasons for these

145 high concentrations are not entirely clear, but coupled to the low isomer ratios (α/γ ≤ 1) they indicate

146 localized input, or at least rapid and efficient atmospheric transport from ongoing sources from afar.

147 Concentrations of HCHs in surface sediments from nearby fjord areas from the same study were lower

148 and comparable to the concentrations in this study (Jiao et al., 2009; Sapota et al., 2009).

149   Concentrations of chlordane isomers in Kongsfjorden sediments are shown in Table S3.

150 *Cis*-chlordane (CC) concentrations, with an average (SD) of 2.6 ± 2.4 pg g^{-1} dw, were slightly lower

151 than *trans*-chlordane (TC), with an average concentration of 4.0 ± 3.3 pg g^{-1} dw. Our results are

152 comparable with earlier measurements of chlordane in marine sediments of the Bering Strait and

153 Chukchi Sea, which ranged from 7.5 -9.3 pg g^{-1} dw (sum of CC and TC) (Iwata et al., 1994), but lower

154 than the fresh water lake sediments in the Arctic, such as lakes Ellasjøen (220 pg g^{-1} dw) and Øyangen

155 (80 pg g^{-1} dw) located on Bjørnøya Island, Norway (Evenset et al., 2004).

156 *Comment on the spatial distribution of legacy HCHs and chlordane*

157 The spatial distribution of HCHs in the surface sediment of Kongsfjorden is shown in Figure 2.

158 Generally, an increasing trend was exhibited from the inner fjord to the outer fjord for α-HCH.

159 Concentrations of this isomer averaged 61.4 pg g^{-1} dw for the outer fjord (including Station KS 16, 18,

160 19, 20, 21, 22), while the average value for the inner fjord (the remaining station) was only 17.3 pg g^{-1}

161 dw. The lowest concentration was found at station KS 11, closest to the glacier snout in the inner fiord,

162 while the highest concentration was observed at KS 22, the outermost station located in the fiord

163 mouth towards the open sea. Similarly, concentrations of γ-HCH were highest in the outer fjord

164 (average value 0.97 pg g^{-1} dw) compared to the inner fjord (average value 0.24 pg g^{-1} dw) and this

165 was also the case for chlordane, with the highest concentrations observed in the outer fiord sites of KS

166 20 to 22. Significant correlations (Pearson correlation) were evident between the concentrations of

167 α-HCH, γ-HCH, CC and TC, indicating similar processes to be affecting the distribution of these

168 legacy OCs. Given that the atmosphere has been the major source of these chemicals to the remote

169 marine environment, including the Arctic, over many decades, then surface ocean waters are likely to

170 be saturated with respect to the contemporary atmosphere (Li and Macdonald, 2005). Indeed, in the

171 case of α-HCH previous loading into surface Arctic waters has resulted in marine waters re-supplying

172 α-HCH to the overlying atmosphere (Jantunen and Bidleman, 1995, 2006), especially as

173 concentrations in arctic air have declined in recent decades (Hung et al., 2010). Levels of α-HCH are

174   widely reported in both North Atlantic and Arctic marine surface waters (Lohmann et al., 2009).

175   As Kongsfjorden is located on the northwest coast of Svalbard, it is affected by both Atlantic and

176   Arctic water masses (Hop et al., 2002), with the Western Svalbard coastal waters influenced by the

177   northernmost extension of the warm North Atlantic Current, which feeds the West Spitsbergen current

178   (WSC, Figure 1). The WSC significantly influences the ecosystem of the outer fjord (Svendsen et al.,

179   2002), and it is plausible that the influx of Atlantic water via the WSC is likely to account for the

180   relatively high levels of these legacy contaminants to the sediments in the outer reaches of the fjord,

181   resulting in the observed concentration gradient for legacy OCs from outer to inner fjord.

182   *Endosulfan and trifluralin*

183   Endosulfan and trifluralin can be considered as current use pesticides, although endosulfan has been

184   recently included in the Stockholm Convention on POPs (http://chm.pops.int/). After HCH and HCB,

185   endosulfan is one of most abundant OCs in Arctic air (Weber et al., 2010) and there is no evidence of a

186   declining trend in air concentrations from the long-term air monitoring data at Alert in the Canadian

187   High Arctic compared to other legacy OCs (Hung et al., 2002; Hung et al., 2005). In the surface

188   sediment of Kongsfjorden, α-endosulfan ranged from 0.1-9.5 pg g^{-1} dw, with an average (SD) of 2.7 $\pm$

189   2.5 pg g^{-1} dw, while β-endosulfan was in the range of <MDL-3.0 pg g^{-1} dw and averaged at 0.73 $\pm$

190   0.83 pg g^{-1} dw. α-endosulfan was detected in all samples, although a lower rate of detection was

191   observed for β-endosulfan (67% of samples). Only limited information is available for endosulfan in

192   the sediments across the Arctic. For example, the concentrations measured here are one magnitude

193   lower than endosulfan concentrations measured in the sediments of fresh water Arctic lakes, where

194   only α-endosulfan has been observed e.g. average concentrations of 40 and 30 pg g^{-1} dw were

195   observed in lake DV09 in Devon Island, Canadian Arctic and lake Ellasjøenin in Bjørnøya island,

Norwegian Arctic, respectively (Evenset et al., 2004; Stern et al., 2005).

Trifluralin was also present in the sediments of Kongsfjorden (Figure 3), although concentrations (with an average of 0.14 ± 0.42 pg g^{-1} dw) were low compared to the other pesticides. Apart from station KS22 where the concentration was 1.97 pg g^{-1} dw, the concentrations measured at the other stations were all <0.15 pg g^{-1} dw (Table S3). Trifluralin is a current-use herbicide although due to its persistence and ability to undergo long-range environmental transport it has been proposed as a candidate chemical to the UN-ECE Convention on Long-range Transboundary Air Pollution (CLTAP) (UNECE, 2007). Data on trifluralin concentrations in different arctic compartments are sparse, although trifluralin residues in surface sediments of the fresh water lake, Ellasjøenin, were estimated as 40 pg g^{-1} dw (Evenset et al., 2004). The Western Airborne Contaminants Assessment Program (WACAP) report included the analysis of sediment cores taken from lakes located in Alaska although trifluralin was reported as below detection limits (Landers et al., 2008). Observations of trifluralin in Canadian Arctic air as well as Arctic rivers are generally low or below detection limit (Hung et al., 2005; Muir et al., 2007; Su et al., 2008) indicating that trifluralin input to the Canadian/North American Arctic is low relative to other POPs. While the levels of trifluralin are low in the sediment samples collected in this study the quantifiable presence of this chemical in western Spitsbergen coupled to the earlier sediment data from Bjørnøya would indicate that agricultural sources are affecting the European Arctic and Svalbard in particular. To support this, Trifluralin has also been reported from dated snow and ice-cores in the Canadian and European Arctic (Hoferkamp et al., 2010; Ruggirello et al., 2010). For example, the trifuralin accumulation flux observed in an ice core taken from the Holtedahlfonna icecap on northwest Spitsbergen (Svalbard) was 2.32 pg cm^{-2} yr^{-1} (for the 1995-2005 dated firn layer) and markedly higher than that observed on Devon Island ice-cap in the

218   Canadian Arctic of 0.016-0.022 pg cm^{-2} yr^{-1}.

219   *Comment on the spatial distribution of Endosulfan and Trifluralin*

220   The spatial distribution of both endosulfan isomers in the Kongsfjord sediments is shown in Figure

221   3. In contrast to legacy OCs, a decreasing trend was observed from the inner to the outer fjord. The

222   highest concentrations for both isomers were found at station KS1 (9.5 and 3.0 pg g^{-1} dw for

223   α-endosulfan and β-endosulfan respectively), while in the outer fjord, the average value for

224   α-endosulfan was only 1.1 pg g^{-1} dw with β-endosulfan below detection limit for most of these

225   stations. The inner fjord of Kongsfjorden is strongly influenced by large tidal glaciers (e.g. the

226   Kongsbreen). In summer, the discharge of freshwater and associated sediment from adjacent glaciers is

227   most pronounced in the inner fjord with a gradual reduction in the magnitude of glacial effects with

228   distance from the glaciers (Hop et al., 2002). The accumulation of endosulfan in surface glacial snow

229   has already been detected in Svalbard (Herbert et al., 2006) and the subsequent release and transport of

230   endosulfan in melt water during periods of seasonal thaw has been observed (Wania et al., 1999; Helm

231   et al., 2002). Specifically, endosulfan was one of the most abundant OCs in the ice core taken from the

232   Holtedahlfonna icecap with the highest accumulation flux of 6.8 and 2.8 pg cm^{-2} yr^{-1} for α- and

233   β-endosulfan, respectively, observed in the upper most firn layer (1995-2005) (Ruggirello et al., 2010).

234   Therefore, glacial melt water runoff with associated particulate matter is likely to account for the high

235   concentration of endosulfan observed in the sediments of the inner fjord. A clear spatial pattern in

236   trifluralin concentrations was not apparent, although the highest concentration was found at station

237   KS22 located towards the outer fjord. The low levels of this chemical, in most cases close to the

238   method detection limit, hamper our ability to observe a spatial trend.

239   *Concentration and distribution of BDE-209 in surface sediment of Kongsfjorden*

240 BDE-209 was observed at relatively high concentrations in the sediment samples, with an average

241 (SD) concentration of 79.7 $\pm$ 53.2 pg g^{-1} dw (Table S4). The concentrations for BDE-209 are

242 comparable to other remote regions like the Canadian Basin (1.6-101.6 pg g^{-1} dw), Canadian lakes

243 from southern Ontario to Ellesmere Island (42-100 pg g^{-1} dw), but lower than those observed in the

244 sub-arctic regions like Bering Strait (9.0-804.9 pg g^{-1} dw) and Chukchi Sea (n.d.-707.3 pg g^{-1} dw) as

245 well as Tromsø harbor in Norway (420 pg g^{-1} dw) (de Wit et al., 2006; Cai et al., 2012).

246 The spatial distribution of BDE-209 in the surface sediment of Kongsfjorden is shown in Figure 4.

247 Concentrations in the outer fjord (average 104.2 pg g^{-1} dw) were slightly higher than the inner fjord

248 (average 67.4 pg g^{-1} dw). The highest concentration was found at Station KS18 (233.8 pg g^{-1} dw),

249 followed by Station KS21 (137.7 pg g^{-1} dw) in the outer fjord. Such distribution indicates the

250 influence of Atlantic water via the WSC. Moreover, despite its high MW and prevalence in the

251 atmospheric particle-phase, BDE-209 can undergo LRAT to remote regions including the Arctic

252 (Wania and Dugani, 2003; Breivik et al., 2006). BDE-209 has been shown to be the dominating PBDE

253 congener in the European coastal atmosphere (North Sea) and has also been detected in the

254 atmosphere of the Greenland Sea (relatively close to western Svalbard) (Moller et al., 2011b; Moller et

255 al., 2012).Furthermore, a scientific cruise from the North Pacific to the high Arctic (Canadian Basin)

256 observed significant particle-bound dry deposition flux of BDE-209, higher than other PBDEs,

257 although it was rarely detected in surface seawater (Moller et al., 2011a). We suspect this chemical is

258 efficiently exported from surface waters to deeper waters by particle mediated scavenging in the water

259 column. Therefore, both marine and glacier inflow could be seem as the potential sources of BDE-209.

260 However, its uniform distribution and relatively high levels in the sediment, especially the notable

261 concentration observed in the sediment at Station KS18, indicate the influence of local sources. This

262 might include the nearby Arctic scientific stations in Ny-Ålesund as well as the influence of shipping,

263 including large cruise vessels which operate in the fjord during ice-free periods

264 *Concentration and distribution of Dechloranes in surface sediment of Kongsfjorden*

265   Consisting of the *syn-* and *anti-*stereoisomer, DP was detected in most of the surface sediment

266 samples, with 78% and 94% detection frequency for *syn*-DP and *anti*-DP, respectively. Sediments

267 concentration of *syn*-DP ranged from n.d. to 5.4 pg g^{-1} dw and averaged 1.4 ± 1.5 pg g^{-1}, while *anti*-DP

268 was in the range of n.d.-15.9 pg g^{-1} dw, and averaged 4.5 ± 4.3 pg g^{-1} (Table S4). DP has been

269 produced and released into the environment for more than forty years (Sverko et al., 2008) and was

270 first detected in the North American Great Lakes region (Hoh et al., 2006), and has been recently

271 observed in the remote parts of the Arctic and Antarctica (Moller et al., 2010; Moller et al., 2011a).

272   The *syn*-DP fraction (f_{syn}) expressed as the ratio of *syn*-DP to the sum of isomers was calculated

273 for the surface sediment. The mean f_{syn} in the surface sediment was 0.28 ± 0.26, while the f_{syn} value in

274 the commercial mixture is estimated to be 0.32 (Wang et al., 2010). In the nearby marine region of the

275 East Greenland Sea, the mean f_{syn} values in both the atmosphere and seawater were > 0.5. This

276 increasing f_{syn} trend with increasing distance from sources was observed in both the North American

277 Great Lakes region and in the Atlantic (Hoh et al., 2006; Moeller et al., 2010), which indicated that the

278 *syn*-isomer is the more stable isomer leading to stereo-selection or enrichment during atmospheric

279 transport. The higher stability of *syn*-DP toward photodegradation has also been demonstrated in a

280 laboratory study (Sverko et al., 2008). However, subsequent settling in the water column as well as

281 further degradation (e.g. biodegradation) may affect the stereoisomer fractionation of DP present in

282 sediments. Our results revealed that the *anti*-isomer might be more stable and persistent in sediment,

283 which is consistent with observations from the North American Great Lakes and a river located in

284 northeastern China (Qiu et al., 2007; Tomy et al., 2007; Qi et al., 2010).

285 The spatial distribution of DP in surface sediments of Kongsfjorden is shown in Figure 5. Both DP

286 isomers showed higher concentrations in the outer fjord (average at 12.1 pg g^{-1} dw) than the inner

287 fjord (average at 2.9 pg g^{-1} dw), with the highest concentration displayed at Site KS21 (*syn*-DP-5.4 pg

288 g^{-1} dw and *anti*-DP-15.9 pg g^{-1} dw), while *syn*-DP was close to or below detectable concentrations at

289 the sample stations in the inner fjord. This distribution would indicate the influence of marine currents

290 affecting the levels of DP in the sediment. High DP levels have already been observed in the seawater

291 of the Fram Strait passage, located between the northeastern coast of Greenland and Spitsbergen

292 (Moeller et al., 2010). As a highly chlorinated and hydrophobic HFR, DP was found predominantly in

293 the particle phase in both air and seawater, akin to BDE-209, and this will influence both its LRAT

294 (Wania and Dugani, 2003) and subsequent behavior in the water column following deposition (Dachs

295 et al., 2002).

Conclusion

297 The present study provides concentrations of OCs, including HCHs, chlordane, endosulfan and

298 trifluralin, as well as HFRs, including BDE-209 and DPs in the surface sediment of Kongsfjorden,

299 Svalbard, Norwegian Arctic. Concentrations of legacy OCs showed a decreasing trend from outer to

300 the inner fjord, indicating a marine signal, possibly the North Atlantic Current to be influencing the

301 distribution of these contaminants to the fjord. In contrast the concentrations of endosulfan increased

302 from outer to inner fjord, reflecting the contribution of melt water and sediment input containing

303 endosulfan from adjacent glaciers to the inner fjord. For BDE-209, there isn't a clear spatial pattern in

304 the fjord. Such a uniform profile suggesta both marine and glacier inflow influences as well as the

305 existence of local sources. Relative lower levels of the fractional abundance of *syn*-DP (f$_{syn}$) indicate

306 greater stability and hence persistence for the *anti*-DP isomer in the sediment. The role of

307 particle-mediated transport for current-use pesticides and HFRs, both in the atmosphere and in the

308 water column, and the fate of particles during seasonal thaw events requires additional research.

309

314

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453   Figure Captions

454   Figure 1. Location of Kongsfjorden, Svalbard, Norwegian Arctic, and the currents influencing the

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456   Figure 2. Distribution of HCHs and HEPT in surface sediment of Kongsfjorden

457   Figure 3. Distribution of trifluralin and endosulfan in surface sediment of Kongsfjorden

458   Figure 4. Distribution of BFRs in surface sediment of Kongsfjorden
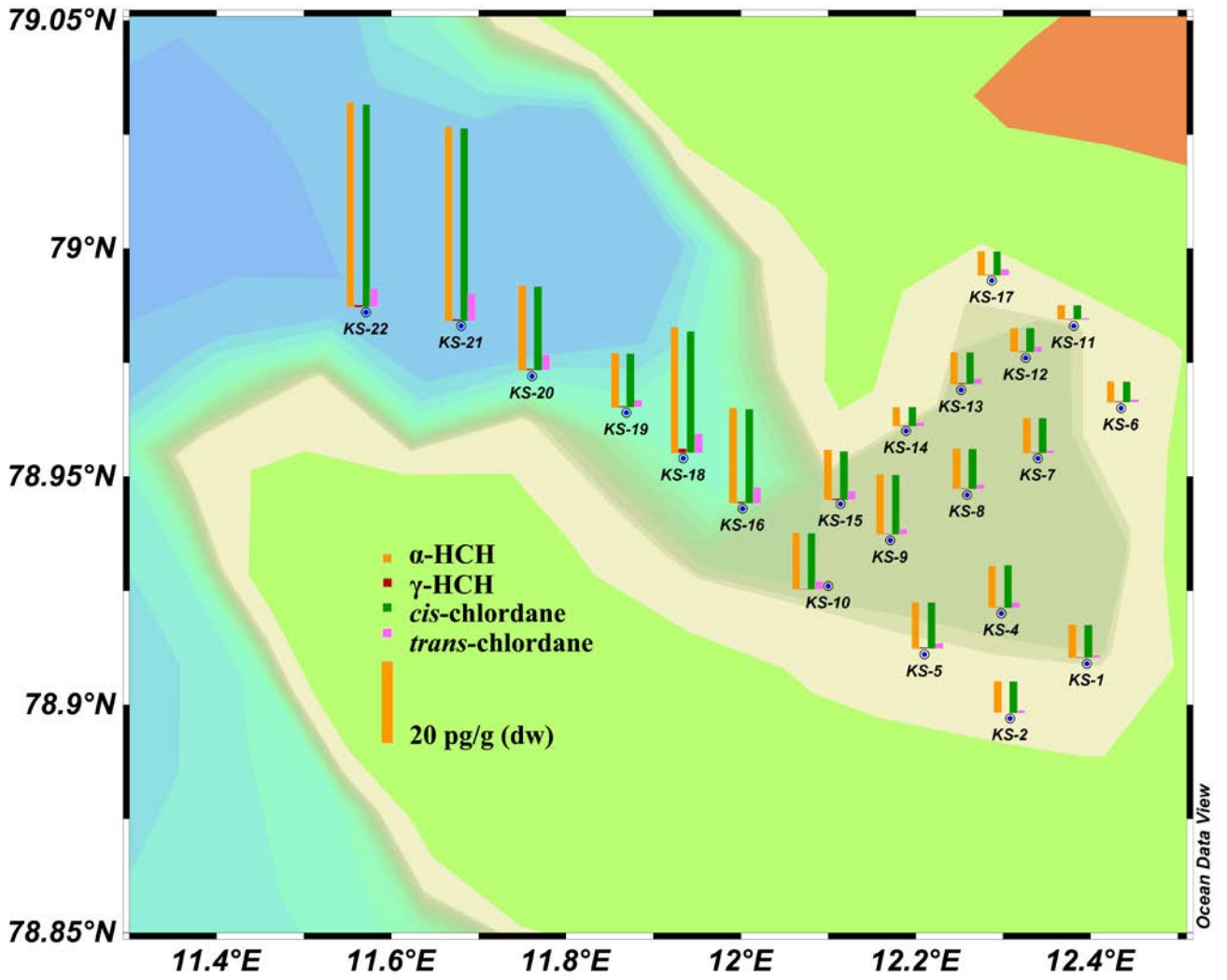
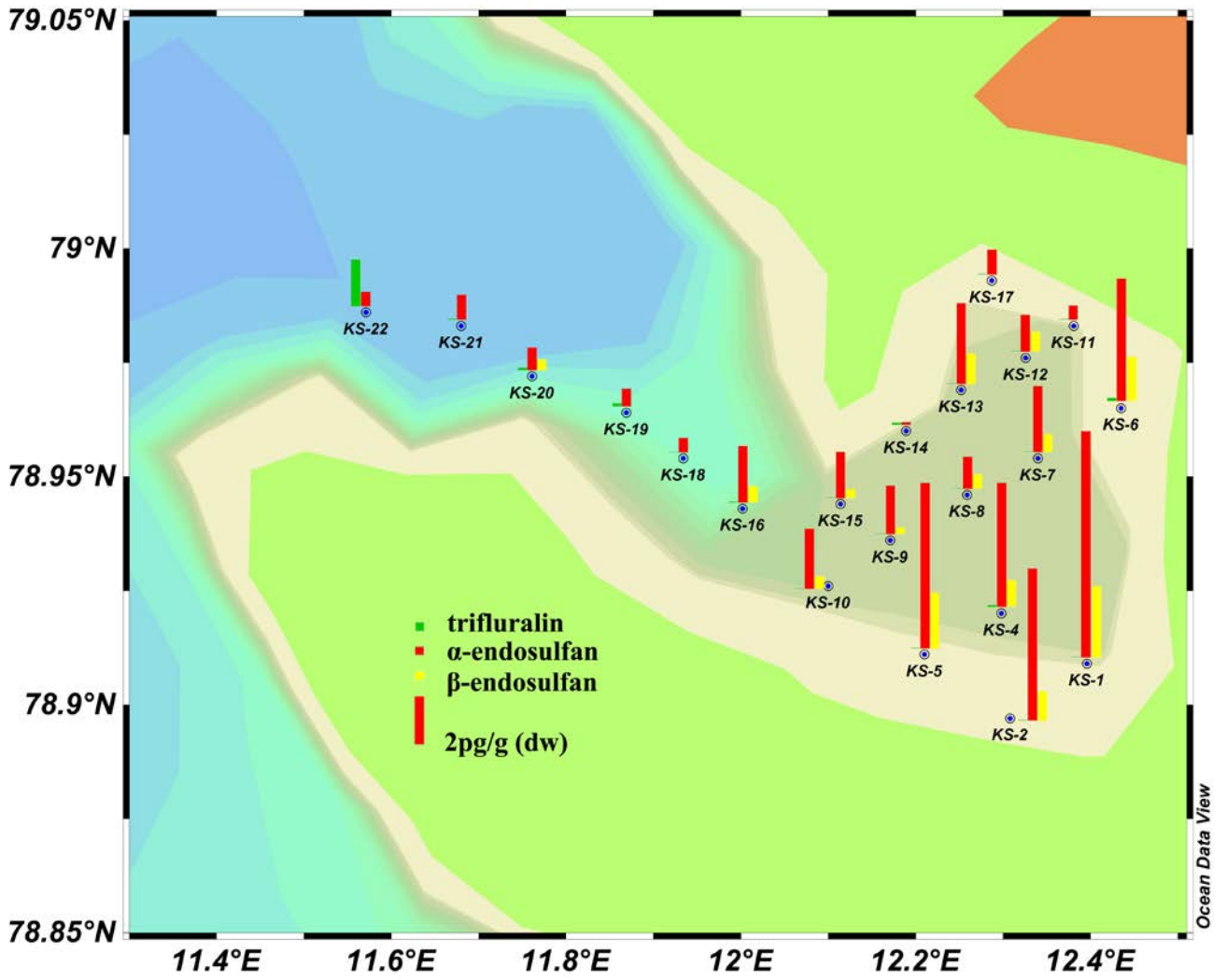459   Figure 5. Distribution of DP in surface sediment of Kongsfjorden
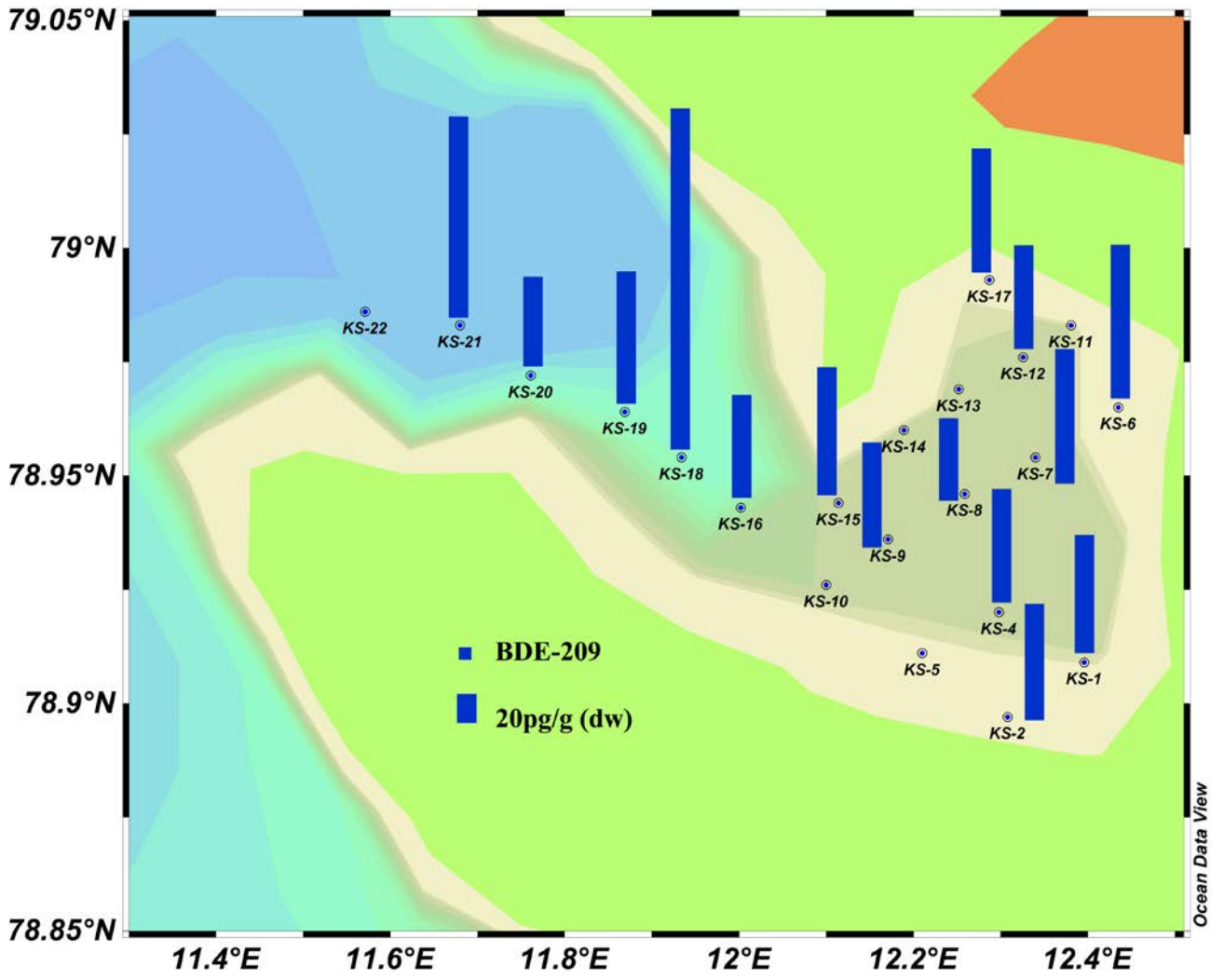
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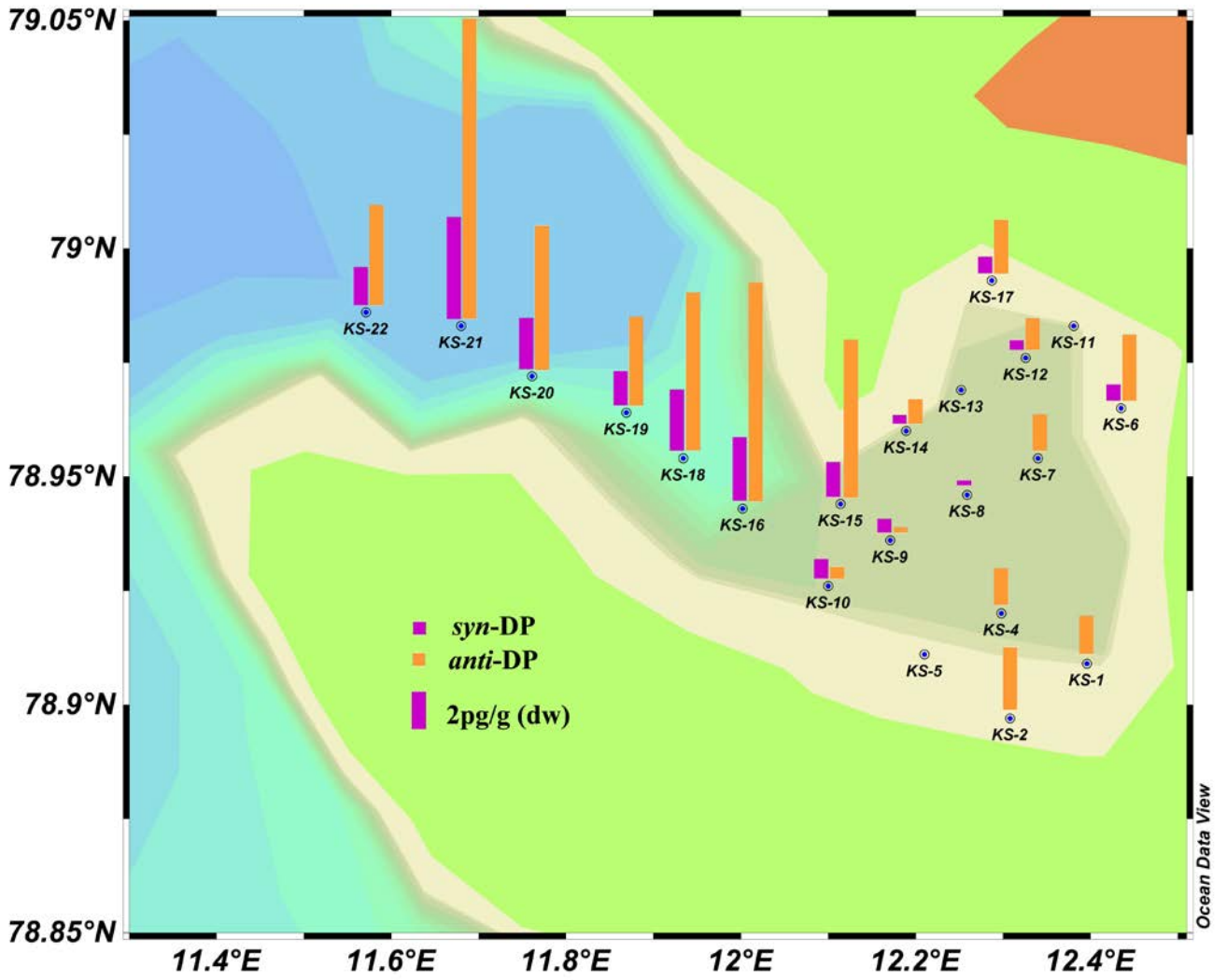462 **Figure 1.**

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Figure 4.

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