Chemical hazard in glacial melt? The glacial system as a secondary source of POPs (in the Northern Hemisphere) – a systematic review

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Toxicity of compounds belonging to persistent organic pollutants (POPs) is widely known, and their re-emission from glaciers has been conclusively demonstrated. However, the harmful effects associated with such secondary emissions have yet to be thoroughly understood, especially in the spatial and temporal context, as the existing literature has a clear sampling bias with the best recognition of sites in the European Alps. In this review, we elaborated on the hazards associated with the rapid melting of glaciers releasing organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and polycyclic aromatic hydrocarbons (PAHs). To this end, we collated knowledge on: (1) the varying glacier melt rate across the Northern Hemisphere, (2) the content of POPs in the glacial system components, including the less represented areas, (3) the mechanisms of POPs transfer through the glacial system, including the importance of immediate emission from snow melt, (4) risk assessment associated with POPs re-emission. Based on the limited existing information, the health risk of drinking glacial water can be considered negligible, but consuming aquatic organisms from these waters may increase the risk of cancer. Remoteness from emission sources is a leading factor in the presence of such risk, yet the Arctic is likely to be more exposed to it in the future due to large-scale processes shifting atmospheric pollution and the continuous supply of snow. For future risk monitoring, we recommend to explore the synergistic toxic effects of multiple contaminants and fill the gaps in the spatial distribution of data.

1. Introduction

Glacial ice in the Northern Hemisphere covers a significant part of the Arctic and it occurs at high elevations in Europe, Asia, and North America, with patches in South America and Africa. The contemporary climate change results in rapid glacier melt worldwide (Chandler et al., 2016; Moon, 2017; Zemp et al., 2015). It is also among the drivers of regional water and food security crises, potentially leading to a global crisis if appropriate action is not taken (Fraiture et al., 2010; Hanjra and Qureshi, 2010; Ragab and Prudhomme, 2002). Glacial ice constitutes the largest freshwater reservoir on Earth (National Snow & Ice Data Center, 2020) and its melt impacts multiple environmental components (Miner et al., 2017). Glacier shrinking importantly contributes to the climate-driven water supply vulnerability (IPCC, 2014), especially in the mountain and piedmont regions of the temperate and low latitudes. Approximately 10% of the global population depends partly on glacial water sources (IPCC, 2019). Beside the physical aspect of the water volume supplied by glaciers, the water quality is also of concern (Bogdal et al., 2010; Miner et al., 2017; Wang et al., 2008a).

Meltwater may contain chemical substances deposited on the glacier in the past (Bogdal et al., 2011), including bioaccumulating persistent toxic compounds, both legacy and currently emitted. As chemicals representative of this problem, we chose for the review organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) as legacy anthropogenic persistent pollutants, polycyclic aromatic hydrocarbons (PAHs) as persistent toxic substances of mixed origin, with continuous emissions, and polybrominated diphenyl ethers (PBDEs) as a newer class of compounds with similarly concerning properties (Jones and de Voogt, 1999). Hereafter, we will refer to all those pollutant classes in general as POPs (persistent organic

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49   pollutants), despite the PAHs not being listed in the Stockholm Convention (Secretariat of the

50   Stockholm Convention, 2017).

51   The Arctic and the high mountains are far from direct sources of POPs, yet POPs are

52   detected in the snow, surface water, sediments and animal tissue collected in these regions at

53   disproportionally high concentrations. The "cold condensation" hypothesis (Simonich and

54   Hites, 1995) explains that in temperate climates, volatiles evaporate intensely into the

55   atmosphere, and are transported with warm air masses to be trapped by cooling in polar and

56   alpine regions (Blais et al., 1998; Galassi et al., 1997; Ma et al., 2016).

57   Since the early 2000s (Blais et al., 2001), it has been conclusively shown that chemicals

58   can occur in discharge from melting glaciers, reintroducing pollution from old emissions. The

59   problem has been studied extensively since, especially in the Alps (Bogdal et al., 2009;

60   Pavlova et al., 2014; Wang et al., 2008a) and Miner et al. (2017) have recently collected the

61   existing literature pertaining directly to this problem in a tutorial review. Miner et al. (2018)

62   have recently investigated the pollution effect in Alaska, an important site for POPs

63   bioaccumulation monitoring due to the fishing industry supporting local communities. Miner

64   et al. (2019, 2018a) provided a risk assessment for pollutants liberated from melting glacier

65   ice and it is the best attempt to date at quantifying the health hazard connected to this

66   secondary wave of pollutant release.

67   Despite the problem being strongly confirmed in one region of the Northern Hemisphere

68   and present elsewhere, it remains to be determined, in the context of the full pollutant

69   pathway through the glacial system, where such a risk is highest and how it may develop over

70   time. As opposed to the existing tutorial review (Miner et al., 2017) of the problem upon the

71   example of organochlorine legacy POPs, we present here a systematic review on the

72   concentrations of four classes of POPs in the environmental media of the glacial system (for

73   methods of the review see section S1 in Supplementary Information). The following novel

74   insights have been achieved as a result: 1) considering glaciers as secondary sources of both

75   legacy and current emission POPs; 2) collating quantitative data on chemical concentrations

76   of POPs in glacial systems of the Northern Hemisphere, 3) overcoming the spatial sampling

77   bias, which has led to the initial highlighting of 45°N as a maximum pollution zone by Miner

78   et al. (2017), 4) including geographical considerations on the polluted ice distribution and

79   melt timing, 5) renewed review of the possible current and future risk levels connected to the

80   discussed phenomenon.

81   **2. POPs pathways through the glacial system, with an emphasis on the snow cover**

82   Glaciers are complex systems, and the transfer of a POP through them may be simplified

83   in the following way (Fig. 1 & 2). Snowfall, rainfall, and dry deposition are all sources of

84   POPs in glaciers, yet snow is by far the most efficient pathway into the system, both as a

85   scavenger of atmospheric pollutants (Lei and Wania, 2004) and the building material of

86   glaciers. Before the snow is permanently incorporated into the glacier, it may undergo further

87   processes which lead to losses of the pollutant mass stored in them: revolatilisation,

88   decomposition of the pollutant in the snow cover (e.g. through photodegradation; Grannas et

89   al., 2007), and elution with meltwater. It may also capture an extra pollutant load through dry

90   deposition. We focus on processes related to snow cover in this work especially because of

91   their marginal mention in the existing review of the problem (Miner et al., 2017), while other

92   elements of the pollutant pathway are elaborated on in that work.

93   During snowmelt, the concentrations of various contaminants may change considerably,

94   leading to elevated loadings in some environmental media at the expense of others. A few

95   field (Bizzotto et al., 2009; Kozioł et al., 2017; Lafrenière et al., 2006; Schondorf and

96   Herrmann, 1987; Simmleit et al., 1986) and experimental studies (Meyer et al., 2009a, 2009b;

97   Meyer and Wania, 2011, and refs therein) show that the concentrations of various organic

98   pollutants in natural and artificial snowpack are modified during melt, depending both on the

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99   pollutant physicochemical properties and the physical or chemical characteristics of the

100 snowpack. Concentration peaks in meltwater, related to snow and ice melt, have also been

101 studied through modelling (Daly and Wania, 2004; Meyer and Wania, 2011; Morselli et al.,

102 2014). Additionally, Daly and Wania (2004) and Hansen et al. (2006) draw attention to the

103 rapid revolatilisation of the more volatile POPs in the spring, when the snow cover changes

104 properties (grain surface area reduces), resulting in spring emission maxima in the

105 atmosphere, especially for α- and γ-HCH.

106   A pulse release of the substances accumulated in snow is a potential ecotoxicological

107 hazard (Daly and Wania, 2004), and its impact downstream has been confirmed using a

108 dynamic organism–water–sediment model in the proglacial Frodolfo stream, where 98% of

109 the released mass of PCB-70 and PCB-101 was estimated to come from snow melt, and not

110 ice melt (for p,p'-DDE, on the contrary, 92% came from melting ice; Morselli et al., 2014). In

111 broad terms, the more hydrophilic contaminants (e.g., naphthalene among PAHs) become

112 released from melting snow unproportionally fast, while the longest storage in melting snow

113 is connected to particulate matter affinity. For example, benzo[ghi]perylene is typically

114 released in a pulse at the very end of the snowpack melt, as it binds mainly to particulate

115 impurities in snow (Meyer et al., 2009a). However, such effects are relative: a more dilute,

116 "cleaner" Arctic snowpack will release a higher proportion of its pollution load, even of the

117 more hydrophobic types, at the beginning of the melt, since the initial concentrations are so

118 low that they do not exceed the capacity of water to dissolve these contaminants. On the other

119 hand, snowpacks heavily laden with particulate matter may retain much more of all pollutant

120 types until their last portion melts (Meyer & Wania, 2011). The effect of the elution processes

121 has consequences for the incorporation of POPs into glacial ice in the part of the accumulation

122 area which undergoes significant melt, to the effect of making clean snowpacks even cleaner.

123 Conversely, the particulate-heavy snow would become an even more concentrated reservoir

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124 of pollutants. This may be relevant not only to urban snowpacks, but also to glaciers exposed

125 to dust sources (e.g., in Iceland, Kamchatka), although the hypothesis has not been explored

126 yet.

127 Among snowpack properties, the increasing depth of the snow cover enhances Type 1

128 elution behavior *sensu* Meyer et al. (2009a), i.e., flushing of more hydrophilic chemicals from

129 the melting snow early; it does not importantly impact any other elution behavior. Particulate

130 matter content and particle permeability of snow regulate the Type 2 (end of melt period)

131 elution. A high internal snow surface area (snow grain surface area) exacerbates Type 3

132 elution (gradually increasing concentrations in meltwater during the melt season), as opposed

133 to Type 1 which predominates if the grains are coarse and rounded (Meyer & Wania, 2011).

134 Meyer & Wania (2011) admit to the importance of refreezing in the chemical

135 dynamics of natural snowpacks, yet their model does not elaborate on the effects of this

136 process. A field study of Kozioł et al. (2017) shows that superimposed ice, an effect of basal

137 refreezing, stores the chemicals which would be transported in meltwater. Without the effects

138 of refreezing, the Type 2 (hydrophobic, particle-bound) organic chemicals should dominate

139 the storage in glacial ice. However, refreezing may rebalance the ratios, adding more

140 relatively water-soluble compounds to the mix. Unfortunately, the quantitative impacts of

141 refreezing on the preservation of POPs in glacial ice are unknown to date.

142 [Figure 1 here]

143 The snow which has not melted becomes compacted into firn and then ice. The

144 combination of ice flow, glacier shrinkage and upward shift of the boundary between the

145 accumulation and ablation area, i.e. the ELA, leads to the polluted ice being exposed at the

146 glacier front and surface. Meltwater from the polluted layers may be delayed by infiltration

147 towards the glacier bed, yet finally it drains into proglacial rivers. While they typically have a

148 high fine sediment loading, its relatively low organic content may prevent intensive

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149 adsorption of POPs onto these particles (Blais et al., 2001). On the other hand, lateral erosion

150 of rivers may incorporate extra pollution load from eroded proglacial soils (Li et al., 2018b).

151 Finally, polluted sediment is deposited either in proglacial lakes, recording former glacial

152 emissions (Bogdal et al., 2010), or in the sea (Pouch et al., 2017). In proglacial waters, the

153 ecosystems process the pollutants, and the top trophic levels of those are especially impacted

154 due to biomagnification (Mazzoni et al., 2020). Both these waters and living organisms in

155 them are subject to human consumption.

156   [Figure 2 here]

3. Geographical distribution of glacial melt in the context of POPs release

158   Glaciers and ice sheets worldwide cover around 10% of the land area (IPCC, 2019) and

159 store 69% of global freshwater (National Snow & Ice Data Center, 2020), likely of better

160 water quality than other surface waters. Arguably, contamination within it should draw more

161 focused attention than before. Worldwide, multiple ice masses have been experiencing rapid

162 dwindling, especially the Greenland Ice Sheet (GrIS), with an average mass loss rate of $278 \pm$

163 11 Gt yr^{-1}, while all   mountain glaciers and ice caps experienced 220 ± 30 Gt yr^{-1} loss

164 (average data for the years 2006-2015; IPCC, 2019).

165   Glacial mass loss changes in time, which inhibits efficient prediction of POPs reemission

166 in glacial melt. The GrIS provides a striking example, where the loss was exacerbated by $80 \pm$

167 6 Gt/y per decade, on average, in the period 1972-2018, reaching -286 ± 20 Gt/y in 2010–

168 2018, although with short-term fluctuations, e.g. due to the cold summers of 2017 and 2018

169 (Mouginot et al., 2019), or the high melt in the summers of 2012 and 2019 reaching (-600) –

170 (-650) Gt of ice (Velicogna et al., 2020). In GrIS, the majority of mass loss is connected to a

171 small number of outlet glaciers and the coastal area (Forsberg et al., 2017).

172   Glaciers and ice caps, i.e., all glacial ice outside the GrIS and Antarctic Ice Sheet, follow a

173 different response pattern due to their smaller size and some of them being located in the

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174   lower latitudes. In 2006–2016, they have been losing up to 335 ± 144 Gt a^{-1} (Zemp et al.,

175   2019). Future projections (Hock et al., 2019) consistently show at least $22 \pm 8\%$ loss of global

176   glacier coverage and $18 \pm 7\%$ of volume, including small glaciers and ice caps located in

177   Greenland and Antarctica (and $29 \pm 9\%$ and $24 \pm 9\%$, respectively, excluding both), even in

178   the relatively mild RCP2.6 climate scenario. RCP8.5 ("business-as-usual") will lead to total

179   glacier and ice cap area loss of up to $43 \pm 14\%$ and volume loss of up to $36 \pm 11\%$ before

180   2100 (all percentage changes are calculated as means between multiple global glacier mass

181   loss models, and relative to 2015). Temporal distribution of mass losses is also uneven across

182   scenarios: slow to no increase in the absolute ice melt annual rate until 2040 and a decrease

183   afterwards in RCP2.6; as opposed to a constant acceleration of losses (i.e. increasingly

184   negative mass balance) across the 21st century in RCP8.5, with only a plateau towards the

185   2100 due to many glaciers disappearing (Hock et al., 2019).

186   To date, the highest cumulative mass losses have been experienced in Western Canada

187   and USA, Central Europe, Alaska, and Asia Central regions (WGMS, 2020) (Fig. 3). In the

188   future, the highest absolute loss is modelled for the regions of Antarctic periphery, Alaska,

189   Arctic Canada North, Greenland periphery, and the Russian Arctic (regardless of the emission

190   scenario), while the relative reductions between 2015 and 2100 would be highest in Central

191   Europe ($69 \pm 19\%$ for RCP2.6 and $93 \pm 10\%$ for RCP 8.5) and lowest in Arctic Canada North

192   (from $12 \pm 8\%$ to $23 \pm 15\%$ for the respective climate forcings; Hock et al., 2019).

193   Furthermore high percentage losses are projected in Caucasus, Scandinavia, and Low

194   Latitudes; divergent future trajectories could happen in Russian Arctic, Arctic Canada South,

195   Svalbard, and Iceland, especially the latter, which may experience anything between

196   negligible dwindling and an almost total wane of glaciers (Hock et al., 2019).

197   [Figure 3 here]

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Importantly, there are both inherent variability and resulting uncertainty in prediction of the glacier mass balance and its components, different for the summer and the winter balance. Summer balance is typically subject to higher variability and drives the negative total (Medwedeff and Roe, 2017). Furthermore, the same net outcome can be achieved both with high snowfall in winter combined with strong melt in the summer, as with low winter precipitation and slow summer melt. These two situations will have varying impacts on the pollutant storage and release, leading to a fast or slow turnover, respectively. The global climate forcing scenarios RCP2.6, RCP4.5, and RCP8.5 show increasing trends both for temperature and precipitation, with a higher magnitude when the forcing is stronger (Hock et al., 2019; Radić et al., 2014), which could lead to a faster turnover of pollutants and the incorporation of new POPs into the ice. However, the predicted positive temperature trends are stronger in the Arctic (Arctic amplification) (Radić et al., 2014; Shannon et al., 2019), and the forecasted precipitation is much more likely to fall as rain. Under high-end climate scenarios, a substantial drop in snowfall is predicted: by as much as 70% in Central Europe, South Asia east, Svalbard or Iceland. The least affected by such shortages would be the Arctic Canada South, North Asia, and Greenland (-4% – -12%); and only the Arctic Canada North (+22% by 2100) and the Russian Arctic (+7%) would receive extra snowfall then (Shannon et al., 2019).

In the 21st century, models predict a gradual spatial change in glacial melt (maximum meltwater runoff or „peak water"; Huss and Hock, 2018). In the Northern Hemisphere (except most of the Arctic, excluded from the cited study), there are multiple regions with glaciers already past their "peak water" point: most of the European Alps, Canadian Rockies, and the Siberian mountain ranges. Soon (before 2040) peak water will be reached in Norway and the lower parts of the Himalaya. On the other hand, Iceland, Alaska, and the higher parts of the Himalaya are predicted to reach their highest glacial runoff only between 2040 and 2080.

223   Bliss, Hock and Radić (2014) included all Arctic sites in a similar study, effectively placing

224   Svalbard and Arctic Canada in the same category as Iceland and Alaska (however with a

225   steeper increase before the maximum glacial runoff is reached). They also presented the

226   relationships between runoff trends and the glacier area and terminus elevation, for each

227   region. While the elevation has the strongest impact on the trend direction and magnitude, in

228   most regions, also glacier size plays an important role. Notably, in the context of chemical

229   hazards connected with glacier melt, it appears more dangerous if the maximum melt period

230   does not coincide with the most intensive melt of polluted ice layers, preventing dilution of

231   the supplied pollutant concentrations; the non-uniform distribution of pollutants in the glacial

232   ice requires targeted monitoring to determine the impact of asynchronous pollution and melt

233   peaks. Finally, there occur mechanisms within the glacial system which relocate pollution

234   from the initial contaminant storage, which have yet to be fully understood (see Section 2).

235   Glaciological understanding offers insights into the location of pollution maxima within

236   the glacier body, as these correspond to ice of certain age. This age can be estimated with the

237   best accuracy at certain points, were ice cores had been drilled, while its spatial distribution

238   can be found through ice flow modeling (in areas with limited data, the accumulation rate

239   provides indirect information). Ice core drilling campaigns have had worldwide coverage. In

240   the Northern Hemisphere, cores have been collected in the Arctic (Greenland, Svalbard,

241   Kamchatka, Alaska, Canadian Arctic) and mountain ranges in the mid-latitudes on all

242   continents (Himalaya and Tibet, the Rockies, European Alps, Tien Shan, Caucasus, and

243   Altai). A multitude of ice dating methods provide cross-validated robust age estimations

244   (Table S1).

245   The detailed literature review of the investigated ice cores, with respect to the ice

246   thickness corresponding to the maximum emission of the POPs classes of interest in this

247   paper, is provided in Table S1 (Supplementary materials; PAHs are excluded here due to their

continuous deposition). We caution that many core studies did not report the newest

accumulation due to their early publication dates, so especially PBDE are covered for an

uneven number of years. In brief, the measured or calculated layer thickness corresponding to

the maximum deposition of PCBs, OCPs, and PBDEs extended between <0.50 and 124 m

w.e., depending on the location. Globally, such ice typically spanned the thickest in the

European Alps and Alaska, followed closely by Caucasus and Yukon Territory. The thinnest

layers corresponded to the maximum POPs accumulation period in Greenland and Ellesmere

Island, with Svalbard and the Russian Arctic also in the lower part of the spectrum. At all

these locations, the thickness of ice in the period of maximum POPs accumulation rarely

exceeded 30 m water equivalent (w.e.), and frequently was below 10 m. However, multiple

cores in the Alps showed polluted layer thickness values within that range, too, and the

maximum observed in the Alps may encompass higher values than elsewhere due to more

studies being conducted there. Nevertheless, the overall picture gathered from point

observations indicates important reservoirs of polluted ice remaining in the Alps, Alaska,

Yukon, and Caucasus. A full quantitative view would require modelling how far the

information from each of those cores can be extrapolated. Indeed, the ice water equivalent

related to the legacy pollution period can differ significantly (61-68%) between a single-point

measurement and a spatially distributed average, as claimed Pälli et al. (2002) in a study of

Svalbard based on radar measurements. An overall picture can be also derived from

accumulation rates worldwide, while an important perturbation to such an evaluation is glacier

geometry.

The two regions with the best described problem of POPs release from glaciers, i.e., the

European Alps and Canadian Rockies, are also those which have already experienced the

most rapid melt and have passed their "peak water" (Huss and Hock, 2018). While the Alps

may store a further load of these pollutants in their higher parts, other regions with thinner

273   layers of polluted ice are likely to become more important pollutant releasers in the future: at

274   first such locations as Alaska, with a relatively fast ice movement, where the thick layer of

275   polluted ice propagates to the lower parts of the glacier (Hodson, 2014; Miner et al., 2018b).

276   Other regions, where such polluted ice has not been transferred to the ablation zone by ice

277   movement, may join as secondary pollution sources once melting shifts the equilibrium line

278   altitude (ELA) upwards and thus liberates POPs from former accumulation areas. An ELA

279   shifting rapidly and glaciers with a wide tongue (large areas near the current ELA) indicate

280   situations prone to POPs release. Svalbard, Arctic, Canada, Iceland, the Russian Arctic,

281   Greenland periphery, and Himalaya all may yet surprise with locally elevated pollutant

282   release once their thinner layers of POPs storage are exposed. Additionally, Arctic Canada,

283   the Russian Arctic, and Greenland are especially important areas to monitor with respect to

284   emerging contaminants, as they are predicted to receive the most snowfall in the future, which

285   may lead to glaciers in these areas acting as reservoirs for new POPs.

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287   **4. Legacy and current use POPs classes accumulated in glaciers**

288   The POPs contain many groups of chemical compounds, and for the capacity of one

289   review we had to narrow our focus to four selected representative classes of POPs. OCPs and

290   PCBs were chosen because they represent old pollutants that had enough time to spread to the

291   most inaccessible parts of the Earth. Their mass use lasted between the end of the Second

292   World War and the 1970s, when restrictions were imposed by individual countries (Breivik et

293   al., 2007; Koureas et al., 2019; Willett et al., 1998). Subsequently, they were banned by the

294   Stockholm Convention (2008), although DDT is still used in some African countries and

295   India to fight malaria (van den Berg et al., 2012). PBDEs are a newer class of impurity, used

296   commercially since late 1960s and surging in 1970s due the limitations in PCB use. By 1990s,

297   PBDEs were widespread (Śmiełowska and Zabiegała, 2018). In the 1990s, penta-BDE were

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298   withdrawn in Japan (Jinhui et al., 2017), then in 2004 the EU discontinued penta- and octa-

299   BDE, and by the end of the same year the US decided to ban PBDEs completely. 2008

300   brought a ban on deca-BDE in the EU (European Court of Justice, 2008) and on all PBDEs in

301   Canada (Jinhui et al., 2017). The Stockholm International Convention adopted in 2009

302   classified mixtures of penta- and octa-BDE as POPs (Secretariat of the Stockholm

303   Convention, 2011). Deca-BDE restrictions were adopted in 2017, although not as stringent

304   (Secretariat of the Stockholm Convention, 2017). PAHs are formed mainly during incomplete

305   combustion of organic matter, therefore they are of both anthropogenic and natural origin,

306   with continuous production and dispersed sources worldwide (Kozak et al., 2017; Manzetti,

307   2013; Zhang and Tao, 2009).

308   **5. The contents of POPs in elements of the glacial system**

309   The systematic review below refers to the samples of air, snow, water, and bottom

310   sediments, collected in the vicinity of glaciers, thus representing the glacial system as a

311   whole. Three areas of sampling recurred most frequently: the Alps, the Himalayas, and the

312   Arctic. The review was limited to the Northern Hemisphere due to the extensive data as well

313   as the limited POPs transfer across the equator (UNECE, 2010). We excluded tropical glaciers

314   to avoid treating the equator as a sharp physical boundary between similar glaciers on both

315   hemispheres. Tables S2 and S3, containing concentration ranges, have been placed in

316   Supplementary materials due to their size.

317   *5.1. Atmospheric air*

318   Thanks to the Arctic Monitoring and Assessment Programme (AMAP), the air

319   pollution of POPs in the Arctic is constantly monitored since mid-1990s at four stations: Alert

320   (Ellesmere Island, Canada); Stórhöfði (Iceland); Zeppelin (Ny-Ålesund, Svalbard); and Pallas

321   (Finland). Elsewhere in the Arctic, further short-term research was conducted, including the

322   newer monitoring station in northern Greenland (Bossi et al., 2016). Besides seasonal

323 variability, a downward long-term trend was observed for the atmospheric concentrations of

324 most POPs. However, it has recently been approaching an equilibrium with concentrations in

325 other media (AMAP, 2016). On the Tibetan plateau, long-term monitoring has been carried

326 out since 2008, complemented with a few short-term tests, while in the Alps there are three

327 long-term monitoring stations (Weißfluhjoch Switzerland, Sonnblick Austria and

328 Schneefernerhaus Germany) (Tables S2 &S3).

329   The existing studies vary by time frame and elevation, which causes a problem in

330 comparing the data as representative for their respective regions. A distinct relationship

331 observed (Fig. 4) is the proportion of volatile compounds increasing with elevation, in

332 accordance with the cold condensation hypothesis (Finizio et al., 2006; Gong et al., 2014).

333   [Figure 4 here]

334   (Wang et al., 2019) report that the concentrations of hexachlorobenzene (HCB), α-

335 HCH, γ-HCH, and α-endosulfan are similar in the Arctic and the Tibetan plateau. However,

336 the concentrations shown in their graphs differ from those in the data table (the basis for these

337 graphs). From the data in the original articles, we conclude that higher concentrations of α-

338 HCH and HCB were observed in the Arctic. For the remaining relationships, we found no

339 further differences between the data reported by Wang et al. and the articles they were derived

340 from. In comparison with these two areas, the Alps show the lowest concentration values

341 (except γ-HCH, which are higher). In Nuuk, Greenland, a correlation between γ-HCH and

342 temperature suggested their reemission from soil and snow driving the atmospheric

343 concentrations (Bossi et al., 2008).

344 *5.2. Snow cover*

345   The concentration data reviewed here concern snow samples collected either nearby or

346 on the glaciers themselves. Some publications present concentrations of particular PCB

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347 congeners, especially the Dutch 7 (PCB 28, 52, 101, 118, 138, 153, 180) (Table S3), and from

348 these it can be concluded that the highest share in the sum of PCBs (∑PCBs) is brought by the

349 lower chlorine content congeners. These are more volatile and thus prone to travel long

350 distances. Some authors mention also the share of lighter congeners increasing with elevation

351 (Finizio et al., 2006; Kang et al., 2009), which appears in opposition to the claim by Miner et

352 al. (2017) that there are higher transport and deposition rates for organochlorine legacy

353 pollutants with higher molar mass and chlorination (postdepositional processes may produce

354 such a concentration profile instead). Maximum concentrations of ∑PCBs occurred in the

355 Alps and the Arctic, yet due to the large scatter of data, the singular low value from the

356 Himalayas cannot be interpreted as conclusive (Fig. 5a).The highest concentrations of the sum

357 of PAHs (∑PAHs; Fig. 5b), markedly different from the other locations, were observed on

358 Spitsbergen. Such high values may be affected by local emission sources in close proximity to

359 all three sites reported here.

360   [Figure 5 here]

361   While we aimed to review the concentration data on 19 selected OCPs, we found

362 sufficient data for comparisons only for HCHs and DDTs. The largest ΣHCHs contents

363 occurred in the Arctic, especially in the Canadian Arctic (Fig. 5c). Those maximum

364 concentrations represent the 1980-90s, when lindane was still widely used. The concentration

365 distribution of ΣDDTs is more even, perhaps due to the greater durability of DDTs compared

366 to HCHs and the continuous use of DDT to combat malaria (Feng et al., 2007). From the

367 collected data pool, which partially overcomes the overrepresentation of samples from the

368 European Alps, it can be concluded that highlighting the latitude of 45°N as a hotspot of

369 pollution by Miner et al. (2017) was premature.

370 *5.3 Ice and firn*

371   Ice cores carry information about the state of the atmosphere in the past, modified

372   through postdepositional changes in the snow cover. Unfortunately, their collection is costly

373   and thus relatively few studies report POPs content in those. To our knowledge, eighteen

374   cores were collected where the content of compounds reviewed here was determined. These

375   ice cores can be divided into two groups, depending on their temporal extent (and the depth

376   they reached).

377   The first group includes ice cores covering only a few years of accumulation. It

378   consists of cores from Lys Glacier (Villa et al., 2006b), Dasuopu Glacier (Wang et al., 2008c),

379   Mt. Ortles (Kirchgeorg et al., 2016), and Colle Gnifetti (Kirchgeorg et al., 2013). All studies

380   analyzing PCBs and OCPs expose a lack of seasonality in their concentrations. Conversely,

381   for PBDEs, Kirchgeorg et al. (2016) observed a higher concentration in the spring-summer

382   period. There were also differences observed in the case of compounds from the PAHs group,

383   which display a high seasonality associated with the heating period (Wang et al., 2008c).

384   Ice cores from the second group span several years of accumulation. The highest

385   concentrations of OCPs in those were observed in a few ice cores in the ice layers

386   corresponding to the 1960s-1980s (Donald et al., 1999; Hermanson et al., 2005; Wang et al.,

387   2008a). Maximum concentrations depended on the compound and the latitude of the sampling

388   site, which is related both to the proximity to areas with a longer OCP use period and the

389   long-range atmospheric transport effects (Donald et al., 1999). PCBs and PAHs showed some

390   maxima in concentrations which did not coincide across the hemisphere, yet for PAHs there

391   occurred longer-term increasing trends underlying the short-term variability (Gabrieli et al.,

392   2010; Garmash et al., 2013; Gregor et al., 1995; Kawamura et al., 1994; Pavlova et al., 2015,

393   2014; Peters et al., 1995; Vecchiato et al., 2020; Vehviläinen et al., 2002). BDE-209

394   concentrations increased more recently, which matches its emission history (in a core

395 collected prior to PBDE restrictions) (Hermanson et al., 2010). Further temporal

396 considerations on POPs ice core records are discussed in Section 7.2.

397 *5.4 Glacial meltwater*

398   The published studies of the concentrations of OCPs and PCBs in the proglacial

399 streams of the Northern Hemisphere concerned locations in the Alps (Bizzotto et al., 2009;

400 Ferrario et al., 2017; Pavlova et al., 2016; Villa et al., 2006a), Canadian Rockies (Blais et al.,

401 2001; Lafrenière et al., 2006; Miner et al., 2018b), Himalaya (Ali et al., 2018; Sharma et al.,

402 2015) and the Arctic (Kosek et al., 2019, 2018; Mcneely and Gummer, 1984). Although the

403 concentrations of the compounds of interest in stream water were at low levels, their seasonal

404 variability depends on pollutant elution behavior and the availability of its store in glacial ice.

405 Thus, Bizzotto et al. (2009) observed a peak in PCBs concentrations in June, which coincided

406 with the late snowmelt and could be connected to the Type 2 elution behaviour of the higher-

407 chlorinated PCB congeners. The lack of difference between the concentrations in a proglacial

408 and non-glacial stream further confirmed the snowmelt origin of those pollutants in the

409 Frodolfo stream. HCHs, on the other hand, consistently with their more typical elution Type 1

410 or 3, exhibited high concentrations in May already. They continued to show high

411 concentrations throughout the melt season in the proglacial stream (unlike in the non-glacial

412 one), thus indicating an extra release from glacial ice melt. Additionally HCB and p,p'-DDE

413 exhibited differences in favour of proglacial waters, but only for one month, which could be

414 connected to the early melt of glacial ice. (Villa et al., 2006a) also found higher

415 concentrations of pesticides (especially HCHs and DDTs, less so for HCB) in glacial than in

416 non-glacial waters.

417   It was also found that the rate of snow melt affects POPs concentrations in streams.

418 Rapid melting causes an increased release of pollutants into the water, while slow melting

419 allows volatiles to escape into the atmosphere instead (Daly and Wania, 2005; Lafrenière et

420   al., 2006). Another modification on the streamwater concentrations of POPs may be the

421   organic content of the sediment it comes in contact with (Blais et al., 2001).

422   *5.5 Proglacial lake sediments*

423   Lake sediments provide information on historical POP emissions, modified by the

424   processes within the upstream part of the catchment. We have found published lake sediment

425   core analyzes of POPs content from the Alps (Bettinetti et al., 2011; Bogdal et al., 2009,

426   2008; Kohler, M.; Zennegg, M.; Hartmann, P. C.; Sturm, M.; Gujer et al., 2005; Pavlova et

427   al., 2016; Poma et al., 2017; Zennegg et al., 2007), the Himalayas (Guzzella et al., 2011; Sun

428   et al., 2018), and the Arctic (Allen-Gil et al., 1997; Jiao et al., 2009; Mannio et al., 1997; Rose

429   et al., 2004; Sapota et al., 2009; Skotvold and Savinov, 2003). It is impossible to determine an

430   area exposed to the largest amount of pollution, yet there is a common temporal pattern. The

431   maximum content of POPs in sediments occurred during their peak use, then it gradually

432   decreased following the prohibition of use by legal acts. Several publications report a second

433   increase in POP content, signaling their reemission. As this secondary source, melting glaciers

434   are most frequently considered (Bettinetti et al., 2011; Bogdal et al., 2009; Sun et al., 2018),

435   yet lateral erosion of soils by rivers during floods was also proposed (Li et al., 2018a).

436   *5.6 Proglacial soils*

437   Soils are the main place of accumulation of POPs in the terrestrial environment (Kurt-

438   Karakus et al., 2005; Wilcke, 2007). It is believed that the content of clay and natural organic

439   material has a major impact on the ability of soil to accumulate them (Ahn et al., 2006; Boyd

440   et al., 2001; Meijer et al., 2003, 2002), which is consistent with the hydrophobic properties of

441   POPs.

442   A notable feature is at least ten times higher concentrations of PBDEs in soil in the

443   Alps (Tremolada et al., 2008) than in the Arctic (Wang et al., 2015) and the Tibetan Plateau

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444 (P. Wang et al., 2009; Wang et al., 2012; Zheng et al., 2012). PCBs have also been seen at

445 higher concentrations in the Alps (Tremolada et al., 2008) than in the Tibetan plateau (Zheng

446 et al., 2012). However, no notable differences were observed in the content of OCPs between

447 the Alps (Tremolada et al., 2008) and Tibet (Fu et al., 2001; Tao et al., 2011; Wang et al.,

448 2012; Yuan et al., 2014) and in the content of PAHs between the Arctic (Z. Wang et al., 2009)

449 and Tibet (Tao et al., 2011; Wang et al., 2014, 2013; Yuan et al., 2014).

6. Model understanding of POPs concentrations in the glacial system

451 A significant progress in understanding glaciers as secondary sources of POPs has been

452 also achieved through modelling. In this section, according to our approach to treat glaciers as

453 a system connected to other environmental media, we will review models of atmospheric

454 transport of POPs and their entrainment into snow cover and glacial ice. However, as other

455 review articles have reported similar findings, we will refer to them and only report the

456 aspects of the state of the art which are the most relevant to glaciers.

6.1. Atmospheric redistribution of POPs into glaciated areas

458 The atmospheric part of the glacial POPs pathway has been explored through two

459 types of modelling work: 1) the pollution origin interpretation is frequently supported with

460 backward air mass trajectory calculation and 2) the quantitative redistribution of POPs into ice

461 media (including snow) is explored through global or hemispheric POPs fate models. For the

462 first case, HYSPLIT (Rolph et al., 2017; Stein et al., 2015) is frequently used. For example,

463 Hermanson et al. (2020) report that HYSPLIT trajectories show abundant air transport from

464 potential origin areas of organochlorine POPs and Gong et al. (2015) have used HYSPLIT to

465 confirm the long-range transport of OCPs in Tibet. The long-range transport phenomenon

466 highlights the vulnerability of glaciers in remote zones to disproportionally high

467 concentrations of POPs, consistent with the cold condensation hypothesis (Wania and

468   Mackay, 1995) or the newer, "differential removal hypothesis", which places more

469   importance with remoteness from POPs sources than with temperature at the receiving

470   location (Friedman and Selin, 2016).

471   The global redistribution models offer an insight into the future of POPs supply into glaciers

472   hemisphere-wide, especially as their primary emissions diminish. Such models range in

473   complexity (described e.g. by Gusev et al., 2007), and they may also inform about the

474   environmental persistence of various POP types. A thorough review of multiple POPs global

475   fate models for atmospheric applications can be found in the United Nations Hemispheric

476   Transport of Air Pollution 2010 report (UNECE, 2010). In brief, at short time scales, the most

477   important processes governing POPs redistribution are airborne transport, partitioning

478   between gaseous and particulate matter, atmospheric degradation, and deposition. The newer

479   atmospheric models have been refined through the inclusion of more detailed processes (Mu

480   et al., 2017; Whaley et al., 2018). On longer time scales, ocean currents (especially for β-

481   HCH) and reemission from surfaces also contribute importantly (multi-hop mode of transport)

482   (UNECE, 2010). POP fate depends on dry and wet deposition from aerosols and gas, on

483   degradation, volatilization, and physical sequestration (typically into deep sea or soil,

484   although glacial entrainment also belongs in this category). POPs have temperature-dependent

485   physical-chemical properties, which govern their long-range transport.

486   The behaviour of all POP types mentioned in our review has been subject to atmospheric

487   modelling (OCPs were represented by HCHs and DDT). The evaluation of such models

488   against measurement data shows typical agreement within a factor of three to four or less, yet

489   there occur high discrepancies in some cases, revealing gaps in emission inventory or

490   potential improvements to model structure or parameters. Westerly winds and northward air

491   flow towards the Arctic both transfer significant loads of POPs. For example, PCBs are

492   brought to the Arctic mainly from Europe, Russia, and North America (including through the

493   Northern Atlantic as a secondary source – Stemmler & Lammel, 2009), and α-HCH from high

494   emission areas in Asia. Emission changes are influential factors in the modification of POPs

495   deposition in receptor regions (which receive up to 30% of POPs from intercontinental

496   transport). These regions would experience a significant drop in POPs deposition load

497   following emission reduction in the main source areas (e.g., Europe for PCBs, North America

498   for B(a)P, or South Asia for α-HCH). In receptor areas, secondary emission sources become

499   more important, for example, they can contribute up to almost 50% of chlorinated POPs in the

500   Arctic; they are much less important in high emission areas like mainland Europe.

501   Atmospheric levels of POPs are influenced by episodic events and thus they need to be

502   equilibrated over longer time periods to estimate their influence on glacial storage. At the time

503   of the UNECE (2010) review, among the main knowledge gaps was listed the description of

504   the impact exerted by secondary sources of POPs on their global fate and long-term trends in

505   source-receptor relationships.

506   UNECE (2010) reviewed the estimates of climate change impacts on the atmospheric

507   transport of POPs, and these were identified as complex, as they could be exerted through

508   changes in wind fields, atmospheric composition (e.g. aerosol loading), removal efficiency

509   and alterations of the surfaces onto which POPs are deposited, including the shrinking snow

510   and ice cover. The temperature dependence of POPs properties makes them vulnerable to

511   climate change impacts. In a more recent review, focusing especially on climate change

512   impact on the fate of POPs globally, Kallenborn et al. (2012) conclude that the overall effect

513   is obscured by the complex nature of the climate-POP fate relationship and the more

514   important role played by the indirect consequences of climate change (agricultural and

515   industrial). The global shift of activity towards the Arctic combined with the removal of

516   cryospheric barriers and storage (sea ice, snow cover, glaciers) will have a compound effect

517   on POPs contamination. On the other hand, Friedman et al. (2014) showed through their

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518 modelling work that for PAHs in the Arctic, that future emission changes will be much more

519 important than secondary effects caused by climate change. Additional concerns are the

520 increased toxicity of pollutants in higher temperatures and the combined effects of climate

521 change and xenobiotics in already vulnerable ecosystems, therefore it is important to

522 determine the tipping points beyond which such effects accelerate (Noyes et al., 2009; Noyes

523 and Lema, 2015).

524 The issue has been further explored for the Arctic in the ArcRisk project, by using

525 several POPs fate models to estimate both the atmospheric delivery of POPs to the Arctic in

526 the future and their impact on the food web (Carlsson et al., 2018) (with a focus on PCBs, as

527 the "sentinel" compound group). While the general emission decline will govern the future

528 concentrations of PCBs in the atmosphere, some environmental concentrations in the Arctic

529 may increase and their distribution in the Arctic is likely to be impacted (especially through

530 wet deposition, biological uptake and riverine transport). Remobilisation of PCBs from

531 seawater, soils, and ice is predicted. Carlsson et al. (2018) reviewed PCB fate in the Arctic

532 according to the models BETR Research, MPI-MCTM, DEHM-POP, and ECHAM5-

533 MPIOM. Because the models neglect emission reduction, they have predicted a factor of 2

534 increase in PCBs concentrations in the Arctic for the ~+3°C climate change scenario. In

535 general, the mid- and high-chlorinated PCBs showed higher sensitivity to climate change (and

536 higher relative concentrations) in the models than did low-chlorinated PCBs. The ECHAM5-

537 MPIOM model included an ocean biogeochemistry submodel, which highlights the increase

538 in bioavailability of PCB-153 due to its efficient binding to colloidal organic matter in the

539 ocean. According to MPI-MCTM, PCB-153 will become more biodegradable, while the

540 persistence of PCB-28 will increase.

541 The MPI-MCTM was used by Octaviani et al. (2015) also to predict future gateways

542 and transport directions of organochlorine POPs into and out of the Arctic. They concluded

543 that PCB-153 will be net-exported from the Arctic to a higher extent in 2070-2099, while

544 PCB-28 will be net-imported, but at a very low flux level. PCBs will enter the Arctic mainly

545 through Alaska−Northwest Territory, Greenland and Norwegian Sea−Northwest Russia

546 gateways, while exiting the region through the Canadian Arctic, the Denmark Strait and the

547 Russian Far East gateways. DDT will continue to be net-imported into the Arctic, mainly

548 through Alaska−Northwest Territories, Davis Strait−Greenland, Norwegian Sea−Northwestern

549 Russia, and Urals−Siberian gateways. It is noteworthy that an increase in DDT fluxes coming

550 from the Arctic is expected as late as 100 years following their peak emissions. Finally, the

551 authors caution against the effects connected to the newer POPs, giving the example of penta-

552 BDEs, which in the year 2000 have reached a concentration in the Arctic atmosphere which is

553 five times that of PCB153 at its peak primary emission in 1970. Hansen et al. (2015) also

554 predicted that the Arctic will store 38% more HCHs and between 38% less and 17% more

555 PCBs (compared to 1990-2000), due to the redistribution by atmospheric transport. Finally,

556 Friedman and Selin (2016) claim that factors outside the Arctic (whether primary or

557 secondary emissions) will impact the Arctic PCB concentrations stronger in the future than

558 any process happening within the Arctic, thus contradicting the findings of Zhao et al. (2015),

559 who ascribed step changes in Arctic atmospheric PCB concentrations to the effects of a rapid

560 shrinking of sea ice.

561 A recent review adds information on the atmospheric delivery of PAHs to the Arctic, the main

562 glaciated region of the Northern Hemisphere. Balmer et al. (2019) noted that the fraction of

563 the total environmental burden of three PAHs (anthracene, fluoranthene, and benzo[a]pyrene)

564 stored in the Arctic ranged (between various model scenarios) from 0.5% to 12.8%. Winter

565 and spring in the Canadian Arctic experience orders of magnitude higher airborne

566 concentrations of PAHs, an important seasonal difference favouring their accumulation in

567 glacier ice. Models simulating future changes in atmospheric PAHs concentrations reflect

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568 mainly the projected drop in global PAHs emissions, although this effect will be much less

569 pronounced in the Arctic than in the mid-latitudes of the Northern Hemisphere (up to 8%

570 compared to up to 38%).

571 **6.2. Snow and land ice in models**

572 Multi-compartment models at the start of their use did not include separate

573 compartments for the cryosphere. The first to do so were Koziol & Pudykiewicz (2001), who

574 included snow and ice as exchange surfaces in a global distribution study of α- and γ-HCH,

575 yet their model did not allow the analysis of long-term effects, such as storage in glacial ice.

576 A significant improvement in understanding the role of the cryosphere was offered by

577 Hofmann et al. (2012), who included dynamic snow cover and land ice (conceptualized as a

578 stable layer of snow cover over Greenland and Antarctica) in the MPI-MCTM model applied

579 for the global fate of γ-HCH and DDT. In a ten-year simulation, they estimated the recapture

580 in the Northern Hemisphere land ice for 1.83% γ-HCH and 1% DDT. While these numbers

581 may seem small, they corresponded to $20\pm10\%$ of the total burden of these pollutants in the

582 ice-covered areas. The authors have also estimated the residence times in land ice (4.44 years

583 for DDT and 16.14 years for γ-HCH), although the overall residence times of these substances

584 in the environment were not significantly changed by the inclusion of land ice in the model,

585 because both pollutants are incorporated into the ice mostly at the expense of storage in soil.

586 Similarly, Stocker et al. (2007) have modelled the impact of snow and ice cover as a shield for

587 underlying soil and seawater, finding that the barrier was responsible for between 3% (for

588 αHCH) and 73% (for HCB) decrease in soil concentration against the ice-free world. The

589 seawater in the Arctic was even more influenced by the ice cover, containing only between

590 8% (for α-HCH) and 21% (for dacthal) of the hypothetical POPs amounts in the ice-free

591 world. The world with the contemporary ice cover produced higher levels of atmospheric

592 pollution (between 2 and 10 times higher, for HCB and PBDE-209, respectively).

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593   The role of snow cover in POPs fate has been investigated by Hansen et al. (2008) in

594   their model DEHM-POP applied to α-HCH behaviour, by comparing the results of model runs

595   with and without snow cover to 21 monitoring datasets. Eight of these sites experienced an

596   improved model fit to reality through the inclusion of the snowpack module, especially

597   predicting spring maxima in air concentrations due to revolatilisation from the snowpack.

598   Another model by Hansen et al. (2006) predicted that α-HCH and γ-HCH are retained in

599   snowpack longer than fluorene, phenanthrene, PCB-28, or PCB-52, which partition more

600   efficiently into the atmospheric air. Finally, the snow scavenging estimation in models may be

601   subject to bias not only from POPs concentration estimations, but also due to the uncertainties

602   in modelling precipitation totals (Whaley et al. 2018).

603   The specific glacial application of modelling to determine the POPs storage

604   potential has only been applied by one research group. The most extensive models for glaciers

605   as a secondary pollution source have been developed for the Oberaar (Bogdal et al., 2010) and

606   Silvretta glaciers (Steinlin et al., 2016). These studies were able to confirm the role of the

607   glacier as a secondary source of pollution and to determine how much of the deposited

608   pollutant loads has already been released from the Oberaar glacier (half of the PCBs and DDT

609   content being retained in the glacier then) and for the Silvretta glacier. In the latter case, it was

610   estimated that 14 g of PCB-153, 41 g of six indicator PCBs (PCB-28, -52, -101, -138, -153,

611   and -180), and 200 g of ΣPCB was still stored in the glacier. These shares were a small

612   fraction of the PCBs that used to be there. A model of accumulation process in the lake

613   sediment was well adjusted for higher-chlorinated PCB congener content in both studies.

614   Bogdal et al. (2010) pioneered in explaining englacial fate of POPs with a model and

615   achieved a relatively faithful reproduction of the storage and delay in the system, despite the

616   neglection of transport in meltwater. The reverse order (in time) of pollutant incorporation

617   into the glacial ice and its release was consistent with glaciological understanding.   The

25

618   authors have estimated that due to climate warming, 45% of the stored POPs have been

619   released from Oberaar glacier before 2010, as opposed to 21% in the scenario with stable

620   climate. Furthermore, the changing climate has generated peak emission events beyond the

621   scale expected in the stable scenario.

622   Steinlin et al. (2016) combined three models to thoroughly describe the fate of PCBs

623   within the glacial system. The models included PCB incorporation into the glacier surface,

624   transport with the ice flow, and chemical fate in the proglacial lake. They were run for the

625   years 1900−2100 and validated against measured PCB concentrations in an ice core, a lake

626   sediment core, and a glacial stream. All PCB fluxes and their storage in the glacier increased

627   until the 1980s and decreased thereafter. The loss dynamics of the lower-chlorinated

628   congeners followed closely the input dynamics, i.e. a high share of these chemicals was

629   released from the glacier immediately after deposition (unlike higher-chlorinated congeners).

630   After a brief increase in the 2000s, the PCB concentrations in the proglacial stream are

631   predicted to be small, although they are expected to persist throughout the 21st century.

632   Interestingly, ice flowlines explained the high concentrations in the sediment in the

633   1990s, although not the high concentrations in the 2000s. The proposed explanation for this

634   discrepancy was processes in the ablation area unaccounted for in the model. The proposed

635   delay of PCB delivery into the lake (through, for example, storage in the porous ice layer on

636   the glacier surface) was estimated for 10 years (Steinlin et al., 2016), which could be partly

637   explained by the residence time of the particulate organic matter on the glacier surface as

638   cryoconite (Koziol et al., 2019; Takeuchi et al., 2010). Vertical meltwater percolation within

639   the glacier (not included in the model) could also relocate chemicals (Steinlin et al., 2016).

640   The model predicts a constant concentration of approximately 500 pg L^{-1} ΣPCBs in the

641   glacial stream during the next decades, which is three orders of magnitude lower than the

642   concentration considered chronically toxic for fish; the impacts on higher levels of the trophic

643   chain need further exploration. Steinlin et al. (2016) claim also that the underestimated

644   sorption of lower-chlorinated congeners and the dynamic system of glacial lakes (Nellier et

645   al., 2015) necessitate the inclusion of biochemical processes in the model to explain the

646   accumulation of POPs in lake sediments.

7. Chemical hazards in glacial melt?

648   OCPs, PCBs and PAHs were found in all the media discussed here (air, snow, ice,

649   stream water, lake sediment). Their presence was also reported by a small number of

650   publications in other media, such as lake water (Diamond et al., 2005; Guzzella et al., 2011;

651   Z. Wang et al., 2009; Yuan et al., 2014) (Tables S2 & S3, Supplementary materials) or

652   seawater (Ademollo et al., 2021) and sea sediment (Pouch et al., 2017). The modest size of

653   the dataset collected so far on these media prevented the interpretation of those for universal

654   spatial and temporal trends. This warrants further studies, especially of the rarely investigated

655   compound classes, such as PBDEs.

7.1. The quality and uniformity of the published data

657   As the research in this field developed, the methods have been adjusted and as a result

658   the data recorded are not consistent in format across the reviewed works. We describe this

659   here as an important limitation to hemisphere-wide conclusions. A typical problem is

660   differing and non-comparable units, especially of fluxes, e.g., [$\mu g/m^2/y$], and concentrations.

661   In some cases, it was possible to calculate concentrations from fluxes according to standard

662   procedures, including e.g. snow water equivalent, ice density and age, or sediment deposition

663   rate, yet there were instances were unreported raw data hindered our efforts. Some values

664   were available on graphs alone, which decreases the accuracy of reporting. Another difficulty

665   for comparison is the common practice to administer only the sum of the concentrations of

666   compounds for such groups as HCHs or DDTs. In the case of PCBs, the problem is

667   exacerbated because the sum of PCBs may involve a different number of compounds,

668   depending on the study design. For individual PCBs, there are two main ways to present the

669   results: 1) reporting concentrations for 7 indicator PCB compounds (PCB 28, PCB 52, PCB

670   101, PCB 118, PCB 138, PCB 153, PCB 180, also named the Dutch seven); 2) grouping

671   PCBs and providing the sums of compound concentrations with the same number of chlorine

672   atoms in the molecule. PAHs experience similar problems as PCBs in reporting the sums.

673   Finally, the concentrations of individual compounds were sometimes given only in the form

674   of an average or a median, which complicated the interpretation, especially as maximum

675   concentrations are of concern for human health impacts.

676   *7.2. The consequences of the pollution trends in time*

677   The rising temperature and enhanced melt release POPs accumulated in glaciers into

678   the environment. This phenomenon has been observed in two ways: in glacial meltwater

679   (through contrasts in concentrations between snow and ice melt and between glacial and non-

680   glacial streams ; Bizzotto et al., 2009) and in sediment cores from proglacial lakes (as delayed

681   secondary concentration maxima; Bettinetti et al., 2011; Bogdal et al., 2009; Sun et al., 2018).

682   Fig. 6 shows the content of ∑DDTs, ∑HCHs, and ∑PCBs in the sediment cores, representing

683   the data for multiple sites in the European Alps and the Himalaya. The release of pollution

684   began at the end of the 1980s in the Himalayas and at the end of the 1990s in the Alps (except

685   Lake Num Co in the Himalaya, where the pollution was also liberated from the 1990s

686   onwards). Within each group of organochlorine legacy compounds, the maximum

687   concentrations from reemission occurred at a similar time. In both regions, the content of

688   OCPs and PCBs in the layers corresponding to reemissions was equal to or even higher than

689   in the layers from the peak use period. In the Alps, concentrations increased steadily until the

690   period of core collection for all POP groups. It was different in the Himalayas: the POP

691   content decreased there in the early 21st century, although it was not uniform for all sites and

692   compounds. Based on these data, POPs re-emission appears to be ongoing. However, there

693   remain important data gaps, especially profiling a wider scope of contaminant concentrations

694   and including areas such as the Arctic, the Rockies, or the rest of Asia.

695   [Figure 6 here]

696   The past pollution emission trends are reflected in ice cores, and the layers recorded in

697   those are expected to melt out in reverse order. We have systematically plotted the

698   concentrations of POPs in the existing ice core records to gain new insights into the spatial

699   and temporal distribution of POP-enriched layers (Fig. 7). Considering the concentrations of

700   OCPs in ice cores, it can be stated that most of HCHs and DDTs are accumulated in the

701   European Alpine glaciers, while the Himalaya and the Arctic show lower values. For PCBs,

702   however, there were no clear differences (perhaps the Agassiz ice cap had a higher

703   concentration, but there were Alpine glaciers within a similar range). Therefore, the

704   conclusion of Miner et al. (2017), based on three sites in the Alps and Svalbard, of the much

705   lower concentrations in the Arctic, appears invalid in light of the existing hemisphere-wide

706   data. In most cases, OCPs are accumulated in the layer of their period of maximum use, with

707   the exception of Lys Glacier, where DDT content continued increasing over time. The cause

708   of such a phenomenon, as Villa et al. (2003) explain, was the intensified local emission at the

709   time when DDTs were being phased out. The timing of the PCBs accumulation (and potential

710   release) is more evenly distributed in time, and the distributions in ice cores are at least

711   trimodal. Thus, the PCBs released from melting glaciers may be a longer lasting problem,

712   without a clear peak to trace.

713   PAHs ice core records are characterized by a different distribution: an underlying slow

714   increase in time (since 1970s), with overlying high extremes of short duration (some

715   overlapping across multiple locations). The slow increase can be linked to the global

716   consumption of fossil fuels and economic growth, while changes in fossil fuel type (especially

29

717   from coal to petroleum and natural gas) mark drops in these trends (Peters et al., 1995;

718   Vecchiato et al., 2020; Wang et al., 2008b). The short-term maxima may be linked to singular

719   events, such as volcanic eruptions. Especially, there can be found maxima in the hemispheric

720   PAHs ice core record closely coinciding with the eruptions: 1963 Surtsey (Iceland) & 1964

721   Scheveluch (Kamchatka), 1970 Hekla (Iceland), 1978 Westdhal (Alaska) & 1980 (Mt. St.

722   Helens, Washington, USA), and perhaps also the 1989 Redoubt (Alaska) and 1991 Pinatubo

723   (Philippines). The consequences of such a complex temporal pattern are: difficulties in

724   predicting the exact timing of reemission of PAHs from melting glaciers, regional differences

725   in the impacts of such a phenomenon, and its long duration due to the thicker layer being

726   affected by elevated PAHs concentrations.

727   [Figure 7 here]

728   The phenomenon of the reemission of pollutants trapped in a glacier is disturbing, and

729   it shows how legacy pollutants may still cause unexpected problems. It should also be an

730   encouragement to limit the use of any persistent and harmful substances as early as possible,

731   lest they become a similar threat as the DDT. Thus far, the phenomenon has been very well

732   documented in the Alps, observed in the Canadian Rockies, and implied in the Himalayas and

733   Alaska. There have been also concerns raised that due to the rapidly warming climate of the

734   Arctic, the phenomenon will also occur there (Miner et al., 2018b), especially as the cold

735   condensation effect promoted their accumulation in the Arctic climate in the past. As POP

736   sources are more distant in the Arctic and some Arctic glaciers experienced lower past

737   accumulation rates, the amount of pollutants stored in them may be smaller than in mid-

738   latitudes (Hodson, 2014). However, the accumulation phenomenon may be exacerbated in the

739   future due to the transfer of legacy pollution into the Arctic by the global atmospheric

740   circulation (Hansen et al., 2015). While the rest of the Northern Hemisphere is predicted to

741   slowly lose legacy POPs, the Arctic will keep accumulating some of them, especially HCHs.

742   Newer pollutants may also accumulate faster in the Arctic, according to Hansen et al. (2015).

743   Finally, with the increased human activity in the Arctic, local pollution sources may also

744   increase their impact (e.g., Granberg et al., 2017). Thus, fresh snow monitoring on Arctic

745   glaciers presents an important data source for the future occurrence of the discussed pollution

746   release phenomenon. Especially in the case of PAHs, the measured concentrations in Arctic

747   snow exceed decidedly the concentrations in the Arctic ice cores. Since PAHs emissions are

748   linked to fossil fuel consumption, especially coal combustion, and glacier melt intensity

749   depends on the greenhouse gas emission scenario and dark particle deposition, glacial

750   reemission of POPs could be limited through phasing out fossil fuels, a wider use of

751   particulate filters for combustion, and by using more efficient combustion methods.

752   *7.3. Risk assessment strategies concerning the impact of glacial POPs release*

753   As POPs bioaccumulate (Corsolini and Sarà, 2017; Czub and McLachlan, 2004; Van

754   Drooge et al., 2014; Walters et al., 2008), their contents in fauna and flora have been of

755   particular concern (Bard, 1999; Dietz et al., 2019; Pedersen et al., 2015; Routti et al., 2019).

756   Fortunately, publications on long-term monitoring of the organochlorine POPs content in

757   body tissues from the Arctic report a downward trend in OCP concentrations (a general trend

758   – with exceptions; AMAP, 2017). The reemission from glaciers will impact mainly the

759   organisms found in rivers and lakes to which such meltwater drains, and only in the case of an

760   extremely high and long-term release could this also be observed in the marine ecosystem

761   (Geisz et al., 2008).

762   The presence of POPs in living organisms, especially in fish, and in drinking water,

763   raises the question of possible harm to human health. Unfortunately, to our knowledge, very

764   few authors have undertaken an assessment of the risk that glacial meltwater poses for human

765   health. Quadroni & Bettinetti (2017) reported that PCB levels in the fish tested sometimes

766   exceeded European Union (EU) standards regarding potential carcinogenic effects. Risk

767   assessment carried out by K. R. Miner et al. (2019) and K.R. Miner et al. (2018) in the Swiss

768   Alps and Alaska, using the Environmental Protection Agency (EPA) methodology, shows that

769   there is no risk associated with the consumption of proglacial water. However, the

770   concentrations in these waters are high enough that the consumption of fish (which

771   bioaccumulate DDTs, HCHs, and PCBs) leads to a risk of cancer. Such risk is greater for

772   children than for adults because of their lower body weight and the expected longer exposure

773   time. At the same time, in waters from another glacier Lys, in the Italian Alps, a lack of risk

774   was concluded for both fish and water consumption, in terms of their DDTs and HCHs

775   concentrations (Miner, 2018). This highlights that the assessed risk level cannot be

776   generalized at a regional scale at the moment, and warrants future research with a wider

777   spatial coverage.

778   We compared POPs concentrations in glacial streams and rivers with the following

779   water quality guidelines (Table S4): the EPA guidelines, concerning human health (EPA -

780   United States Environmental Protection Agency, 2020), and aquatic life condition (EPA -

781   United States Environmental Protection Agency, 2019) and the Directive of the European

782   Parliament and of the Council on the quality of water intended for human consumption (EU-

783   Groundwater Directive, 2006). The guideline thresholds were only rarely exceeded, among

784   them none of the EU limits. The highest observed concentration in Frodolfo stream (Bizzotto

785   et al., 2009) exceeded the EPA norm more than two times, yet even those concerned a short

786   period of time, while the norm referred to chronic toxicity. Based on these two guidelines, the

787   POPs released from the glacier have not yet caused a known acute or chronic toxicity risk for

788   aquatic organisms or humans.

789   The aforementioned water quality criteria indicate the risk of cancer associated with the

790   consumption of aquatic organisms and water. Using this criterion, which is based on

791   carcinogenicity risk 10^{-6}, we also scanned the collected database for potential threats. These

32

792 recommended concentrations of pollutants were exceeded in many cases. The contents of α-

793 HCH (Bizzotto et al., 2009; Ferrario et al., 2017; Lafrenière et al., 2006; Villa et al., 2006a),

794 HCB (Bizzotto et al., 2009), dieldrin (Lafrenière et al., 2006), and p,p'-DDT (Bizzotto et al.,

795 2009; Ferrario et al., 2017; Villa et al., 2006a) were exceeded at a maximum by one order of

796 magnitude. Moreover, PCB (Bizzotto et al., 2009; Ferrario et al., 2017; Lafrenière et al.,

797 2006; Pavlova et al., 2016) and p,p'-DDE (Bizzotto et al., 2009; Ferrario et al., 2017)

798 guideline concentrations were exceeded by almost three orders of magnitude. This fact

799 encourages the continuation of screening for POPs concentrations in glacial meltwater.

800 However, due to the fact that the highest concentrations of PCBs, HCB, and p,p'-DDE in

801 Bizzotto et al. (2009) occurred in the snow melt period (June) and the highest concentrations

802 of α-HCH, dieldrin, and PCBs were noted by Lafrenière et al. (2006) in lysimeters collecting

803 snow melt water, we vouch for including snow elution patterns in the screening study design.

804 *7.4. Recommendations for the chemical monitoring of glaciers*

805 For exposure monitoring, the most important issue to assess is the timing and intensity

806 of the potential pollution plume release from glaciers, based on the limited existing

807 information. With monitoring being both highly demanding in skilled work and logistics, and

808 thus costly, it is not affordable to fund widespread screening programs for POPs in glacial

809 meltwater streams or in other glacier-related media. However, such information is especially

810 needed where the water and fish supply for large groups of people is connected to melting

811 glaciers, and where there are the most vulnerable ecosystems exposed to such extra stressors

812 in the time of an ongoing ecological crisis (Ripple et al., 2019). Thus we seek here for a

813 monitoring strategy robust enough to protect human and animal health.

814 POPs release from glaciers has been confirmed locally (and regionally in the European

815 Alps), and it is necessary to confirm its occurrence elsewhere, especially since risk levels may

816 differ between glaciers from the same region (Miner, 2018; Miner et al., 2018a). A screening

817   for areas already exposed to POPs release from glaciers (and the chronic toxicity risk) could

818   be based on proglacial lake bottom sediment coring (due to the efficient capture of POPs by

819   sediments). The regions especially vulnerable now and in the future may be located through

820   the interpretation of global chemical fate models, which are increasingly successful at

821   incorporating cryospheric component into the POP cycle, although their spatial distribution

822   could be further refined. Incorporating the functioning of the glacial systems in these models

823   (e.g. ELA shifts, accumulation and ablation rates, glacier geometry) is the next challenge for

824   progress in understanding glaciers as POPs storage.

825   It is also necessary to determine the combined harm posed by multiple substances

826   released in glacial water. Current models focus mainly on individual chemical compounds or

827   groups of similar compounds, so the risk assessment may be underestimated because it omits

828   other (especially newer) substances and possible synergistic effects. A valuable addition to

829   models would be to use toxicity tests based on living organism exposure (such as in

830   Szczepańska et al., 2017). Due to the risk being mainly connected to either chronic toxicity or

831   carcinogenic effects, any planned monitoring of proglacial waters may be performed with

832   passive samplers exposed for the whole melt season, limiting the number of analyses to be

833   performed. These approaches, combined as befits a particular site, would lead to a more

834   comprehensive assessment of human exposure.

835   **8. Conclusions**

836   The threats connected to the melting of glaciers include multiple physical effects, although

837   the chemicals released from glacial melt can also be dangerous. The increased concentrations

838   of POPs were observed both in sediment cores from proglacial lakes and in glacier-fed

839   streams (as compared to non-glacial streams). Such evidence comes mainly from the

840   European Alps and Canadian Rockies. To overcome this sampling bias, we systematically

841 collected published data on four POP class concentrations in glacial environmental media of

842 the Northern Hemisphere.

843 While the Canadian Arctic and European Alpine glaciers appear as the most polluted in

844 the hemisphere, and lower pollution concentrations occur in the Himalayas, the regional

845 concentration ranges frequently overlap, spanning <LOD to several ng/L (for some OCPs and

846 the sum of PBDEs). Only the PAHs content in glaciers exceeds that level, and it is clearly the

847 highest in Canadian Arctic at 35-660 ng/L, with intermediate levels in the Caucasus at 49-166

848 ng/L, the Himalayas at 8.9-100 ng/L or Greenland at 2-103 ng/L, and the lowest in the Alps at

849 0.5-5.2 ng/L (if locations close to high-emission hotspots are excluded). Based on the existing

850 data, the greatest threats associated with the release of POPs will take place in the Canadian

851 Arctic and the Alps, also due to the presence of thicker ice layers in the maximum emission

852 period. However, multiple regions are too poor in data for a conclusive assessment.

853 Especially, the tentative conclusion of maximum legacy POPs concentrations around 45°N

854 (Miner et al., 2017) is not maintained in light of a wider dataset. Therefore, a more

855 comprehensive way to account for the distance from POPs sources, including local hotspots,

856 and the emissions changes in time needs to be used for spatial assessments (cf. Friedman and

857 Selin, 2016; Kallenborn et al., 2012). Finally, the global pollution shifts and the remaining

858 cryosphere extent will increase the importance of monitoring both legacy and new POPs in

859 the Arctic.

860 Although the compounds we discuss are toxic, no immediate toxic effects associated with

861 their release in meltwater are to be expected. They may, however, cause harm through long-

862 term effects related to their ability to bioaccumulate and biomagnify. This is unless serious

863 synergic effects are discovered, on which there should be future research, since the

864 overlapping emission periods of various groups of POPs increase such risk. Furthermore, risk

865 assessments are still missing for the release of PAHs and PBDEs in glacial meltwater,

including remote locations (Himalaya, Arctic). An excess of PAHs may occur especially in volcanic areas of the Arctic (Iceland, Kamchatka). The northern Canadian Arctic and the Russian Arctic may be continuous recipients of current use pollutants, as they are the only areas predicted to receive extra snowfall in high-end warming climate scenarios. Arctic glaciers and the Himalaya are also important prospective study areas since they are yet to experience their maximum melt later on in the 21st century. Hence, estimating the moment when POPs stored there are released and at what concentrations would identify whether an important chemical hazard to humans or ecosystems will occur.

We suggest applying as monitoring tools for the underrepresented areas: 1) drilling sediment cores from proglacial lakes, 2) monitoring bulk concentrations of POPs in meltwater with passive samplers, 3) employing bioassays to estimate the overall toxicity of proglacial waters. It appears that among legacy pollutants, PCBs will require much longer monitoring due to the lack of clear maxima in their accumulation records in glacier ice (similarly PAHs, due to their continuous emissions). The common emission sources of PAHs and greenhouse gases provides extra arguments for phasing out fossil fuels (especially coal) and applying more efficient combustion methods where possible. For the designation of the most exposed study sites, glaciological parameters can be used: 1) high accumulation rate and accumulation area size at the time of maximum pollutant emission, 2) high melt intensity and a rapid upward ELA shift, 3) the width of the glacier at ELA, which determines the area exposed to melt upon such an upward shift; however, on short-term basis, the role of POPs elution from snow should also be included in monitoring design.

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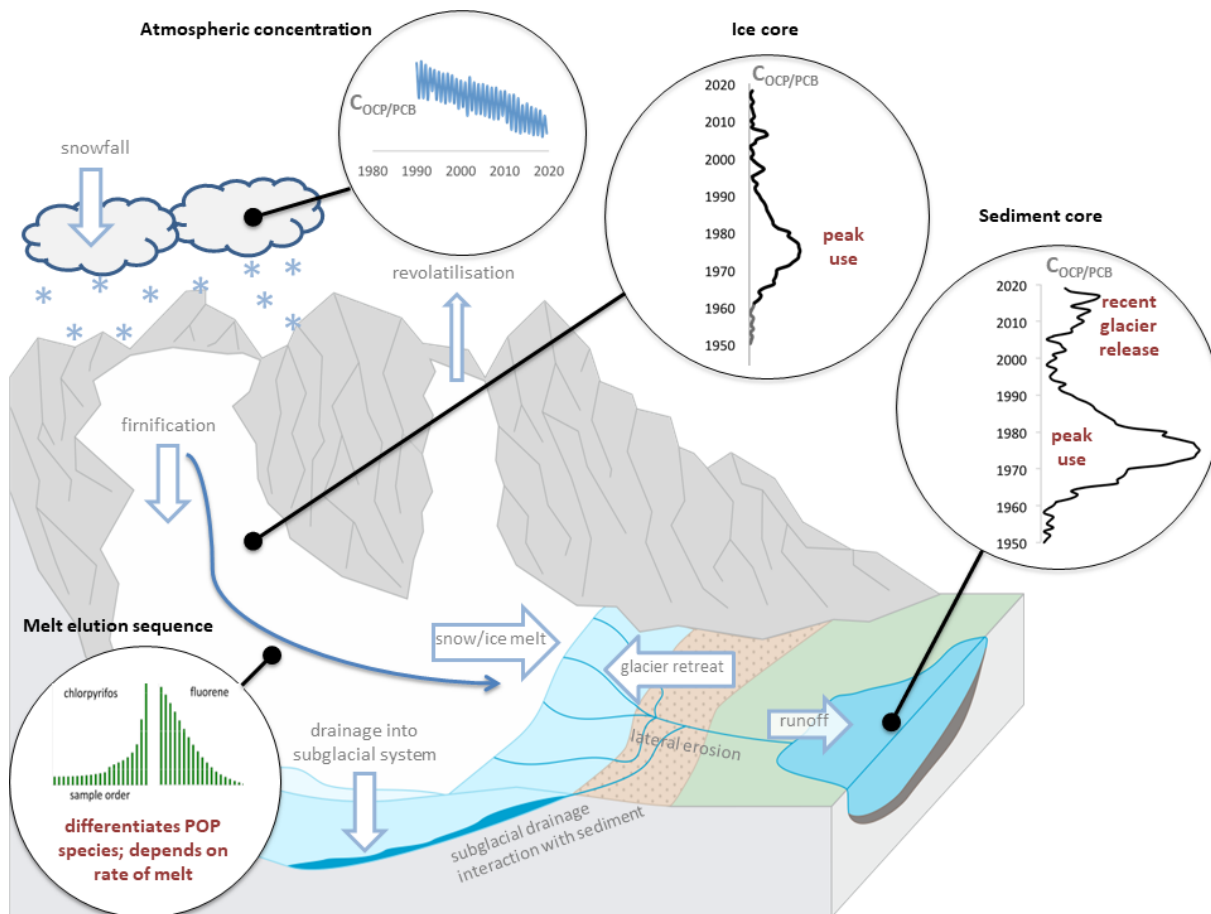1494   https://doi.org/10.1016/j.envpol.2011.09.036

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Figure 1. A diagram of the glacial system with the environmental media potentially storing pollutants (with sample pollutant concentration (C_{POP}, $C_{OCP/PCB}$) graphs).

POPs partitioning between gaseous, dissolved and adsorbed (between phases of water and attachment to impurities)
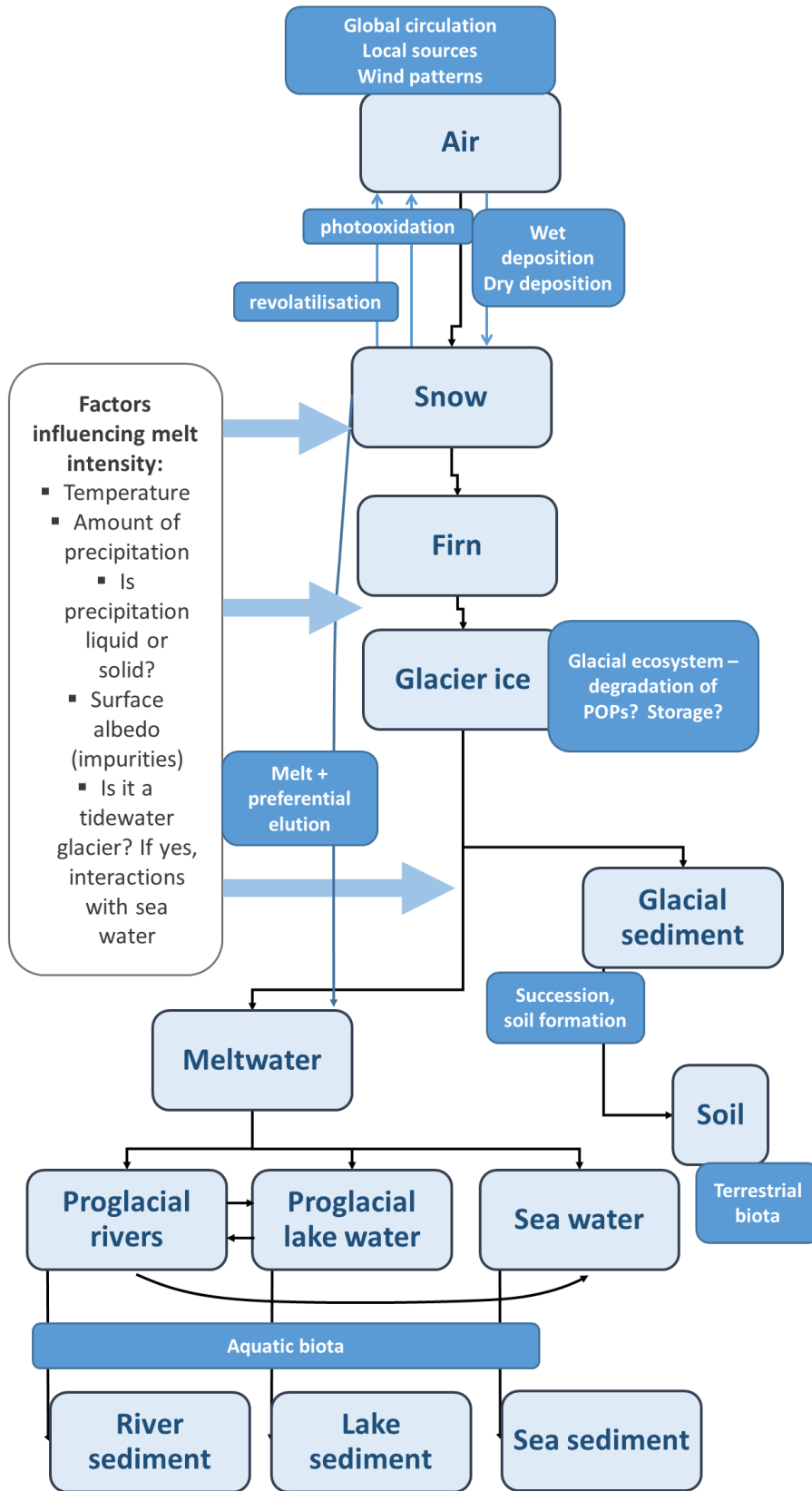
Global circulation
Local sources
Wind patterns

Air

photooxidation

Wet deposition
Dry deposition

revolatilisation

Snow

Factors influencing melt intensity:
- Temperature
  - Amount of precipitation
    - Is precipitation liquid or solid?
    - Surface albedo (impurities)
      - Is it a tidewater glacier? If yes, interactions with sea water

Firn

Glacier ice

Glacial ecosystem – degradation of POPs? Storage?

Melt + preferential elution

Glacial sediment

Succession, soil formation

Meltwater

Soil

Terrestrial biota

Proglacial rivers

Proglacial lake water

Sea water

Aquatic biota

River sediment

Lake sediment

Sea sediment

Figure 2. POPs pathways through the glacial system, including the factors influencing their transport efficiency and storage time.
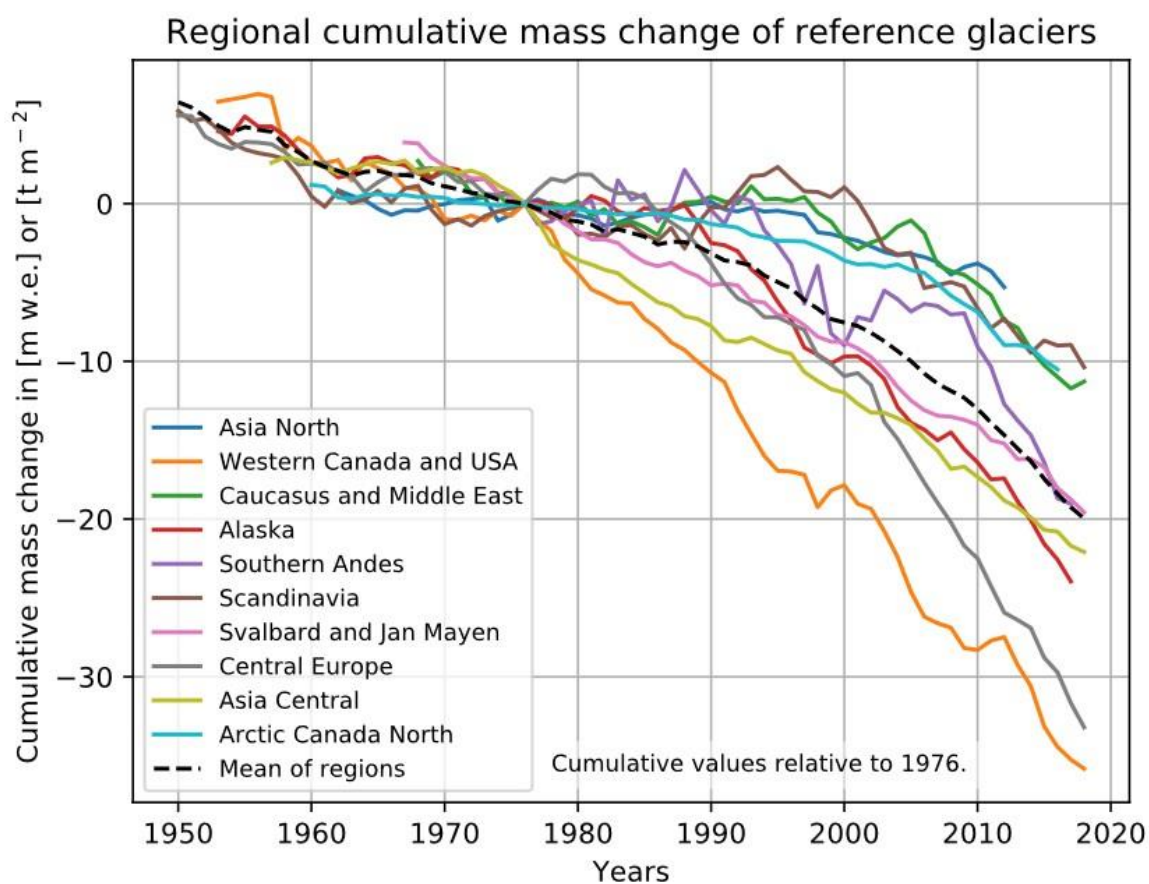
Figure 3. Cumulative mass balance change (since 1976) in glaciers around the world, divided by region. Source: (WGMS, 2020). *Reprinted with permission.*
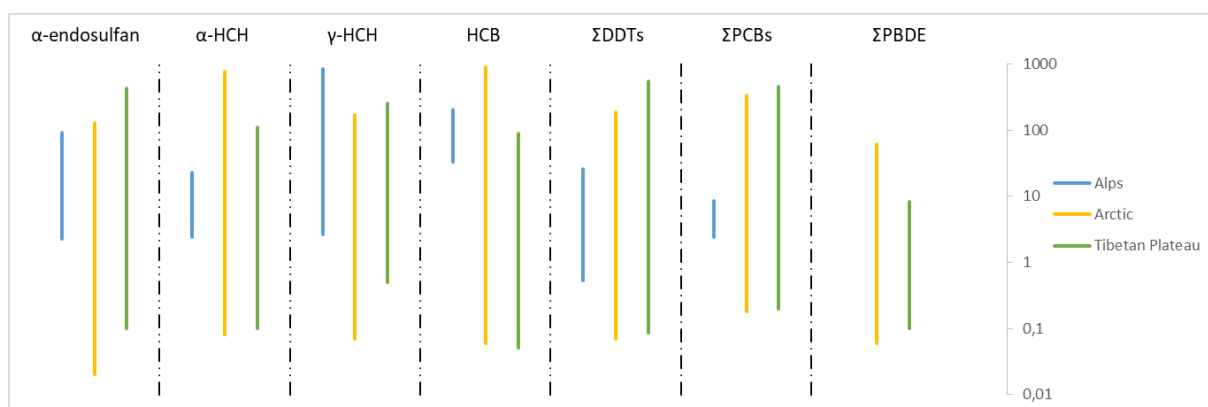
Figure 4. Comparison of the concentration ranges [pg/m³] of atmospheric POPs in the Alps, Arctic and Tibetan Plateau. Data and references used in this figure can be found in Table S2-3.
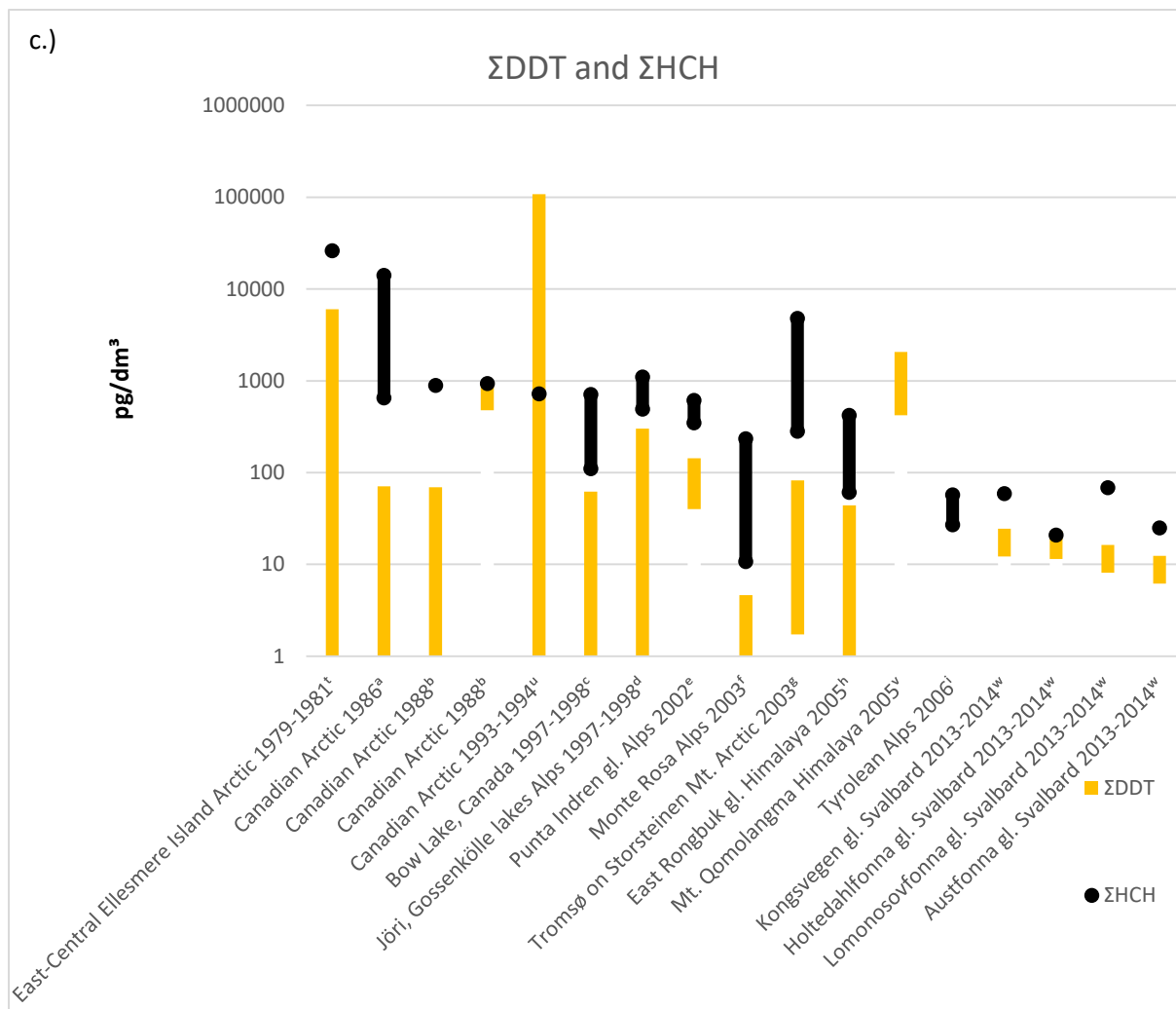
a.) ΣPCB's

b.) ΣPAH's
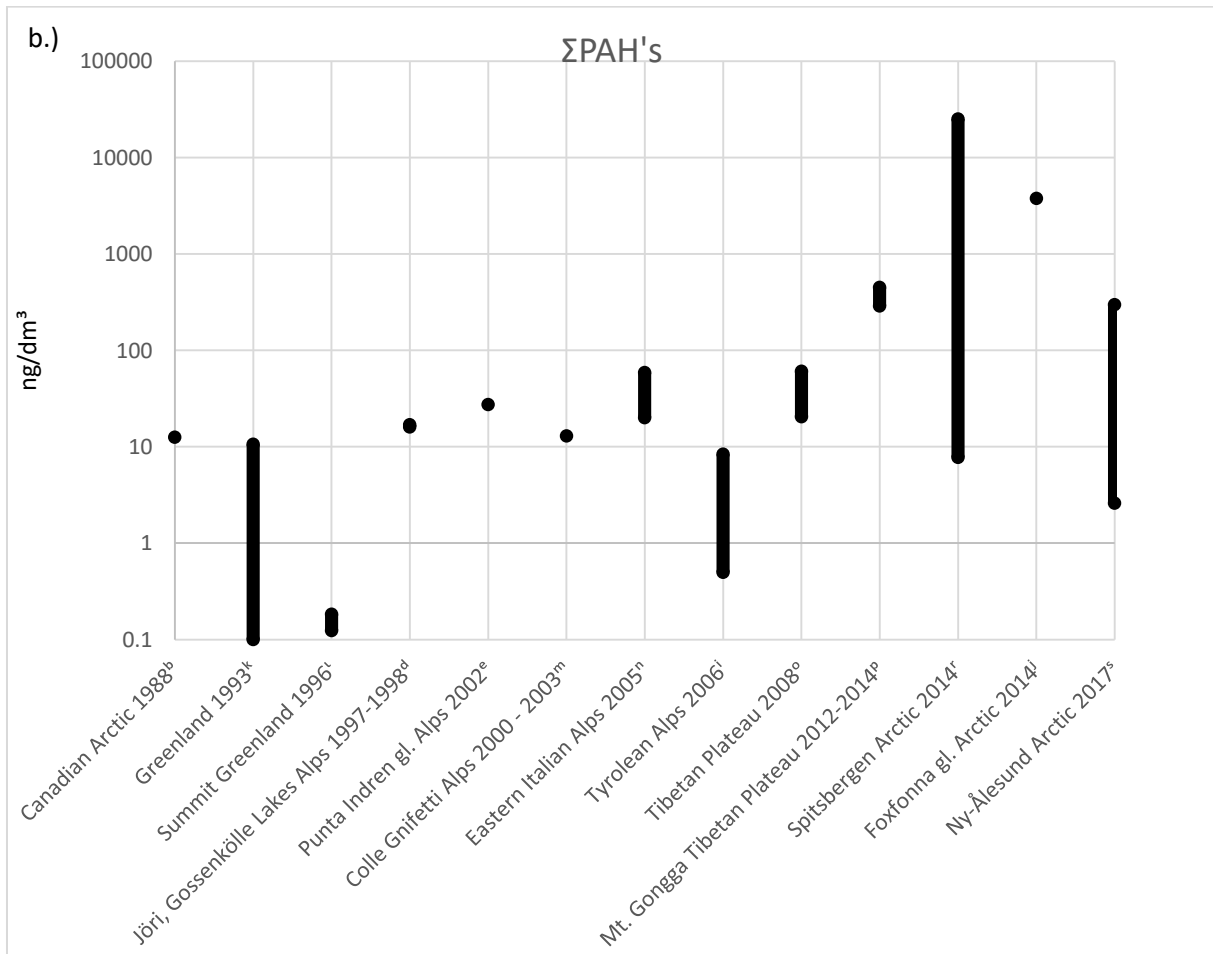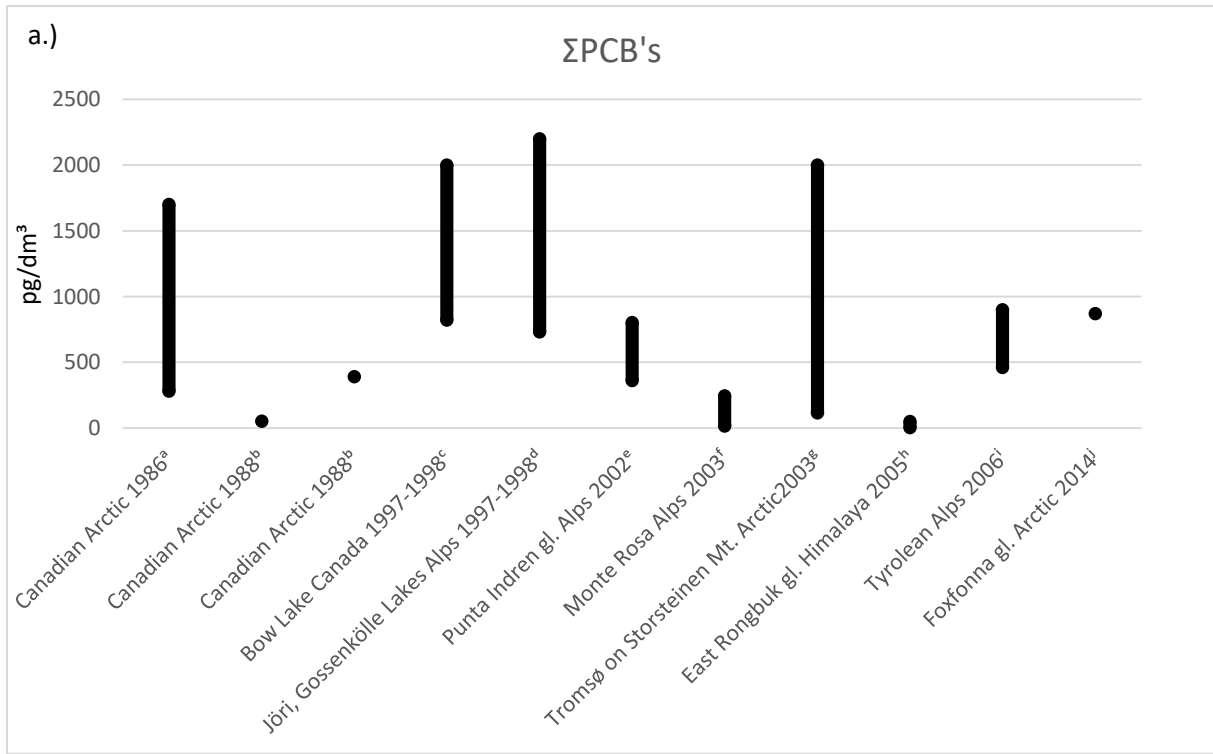
Figure 5. Range of a.) ∑PCBs [pg dm^{-3} of melted snow water], b.) ∑PAHs [ng dm^{-3} of melted snow water] and c.) ∑HCHs and ∑DDTs concentration in [pg dm^{-3} of melted snow water] concentration in snow in various glaciated regions worldwide. a - (Dennis J Gregor & Gummer, 1989), b - (Welch et al., 1991), c - (Lafrenière et al., 2006), d - (Carrera et al., 2001), e - (Herbert et al., 2004), f - (Finizio et al., 2006), g - (Herbert et al., 2005), h - (Kang et al., 2009), i - (Arellano et al., 2014), j - (Kozioł et al., 2017), k – (Masclet et al., 2000), l - (Currie et al., 1998), m - (Gabrieli et al., 2010b)(Gabrieli, Vallelonga, et al., 2010), n - (Gabrieli et al., 2010a)(Gabrieli, Decet, et al., 2010, o - (Li et al., 2011), p - (Yu et al., 2018), r

- (Abramova et al., 2016), s - (Vecchiato et al., 2018), t - (Mcneely and Gummer, 1984), u - (Boyd-Boland et al., 1996), v - (Wang et al., 2007), w - (Hermanson et al., 2020).
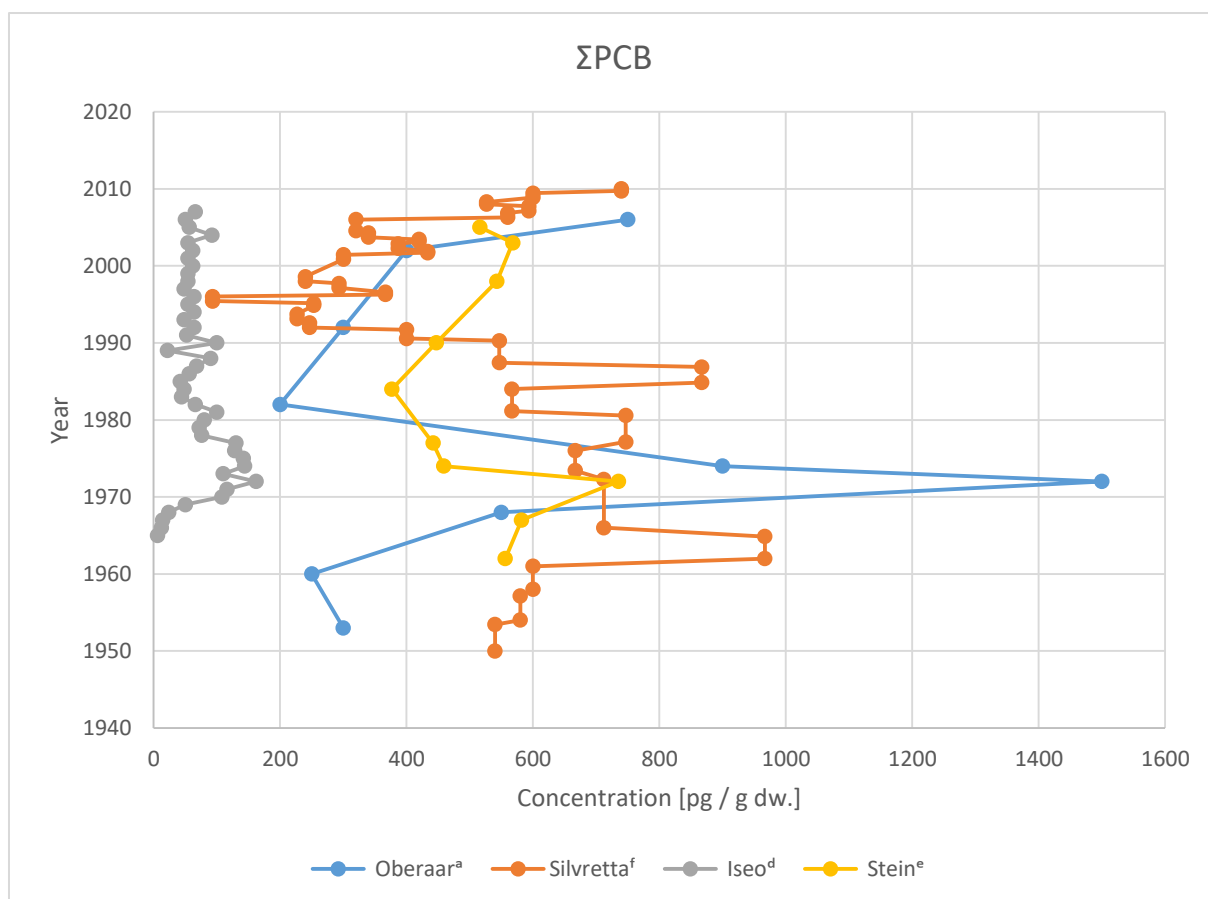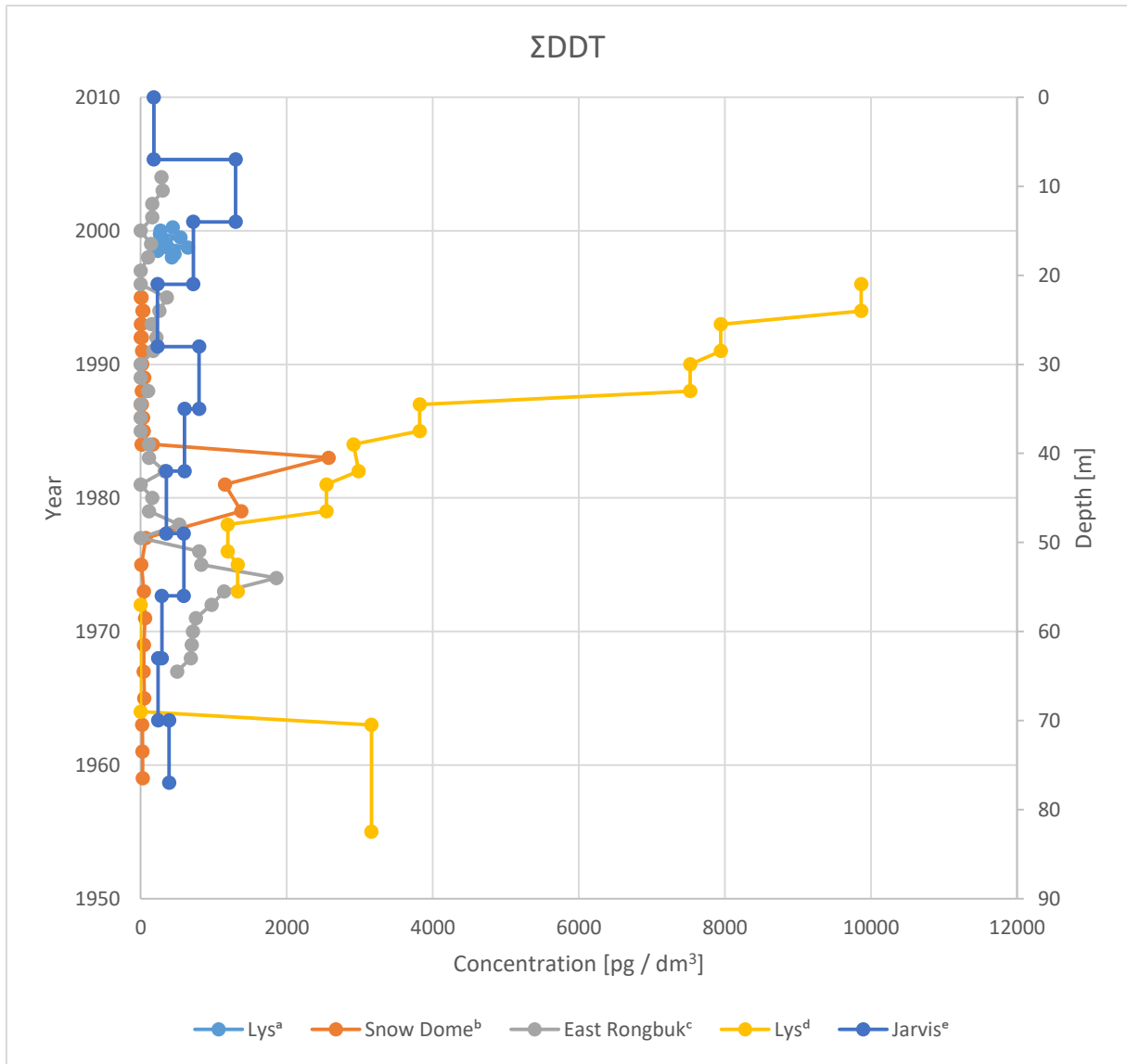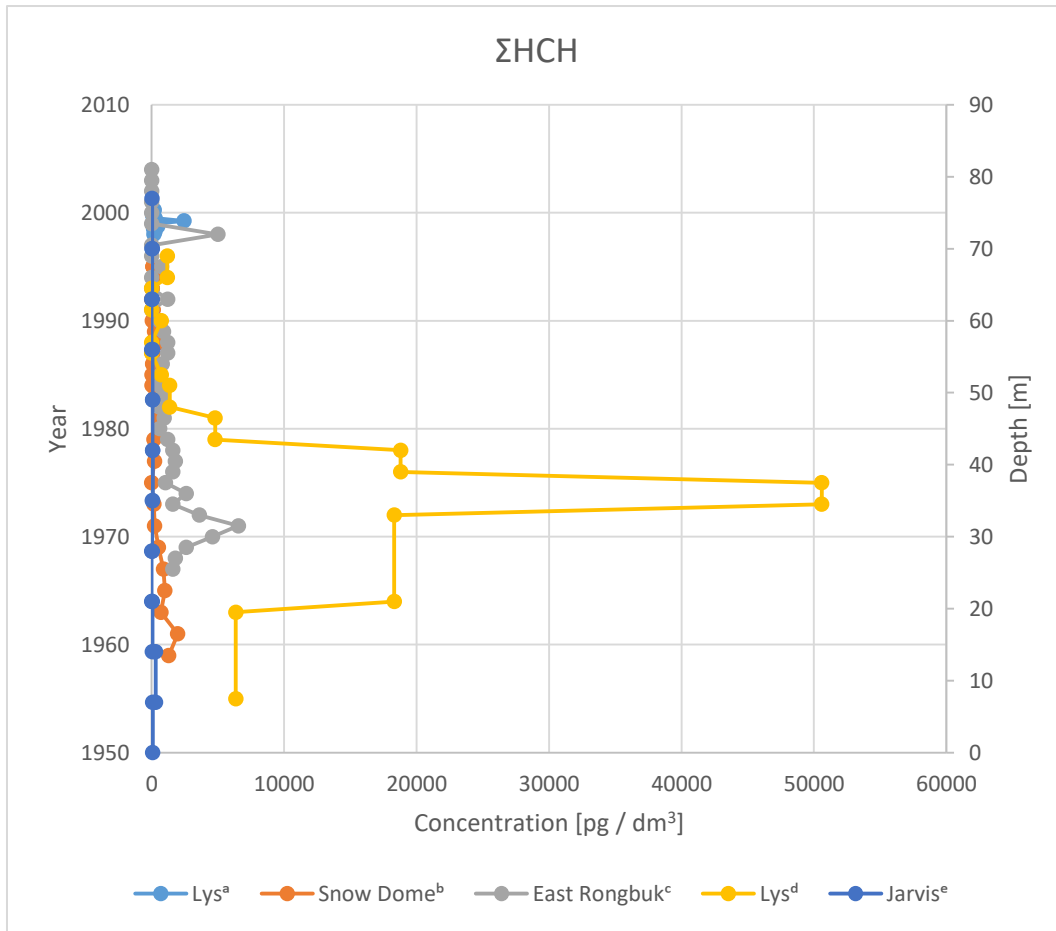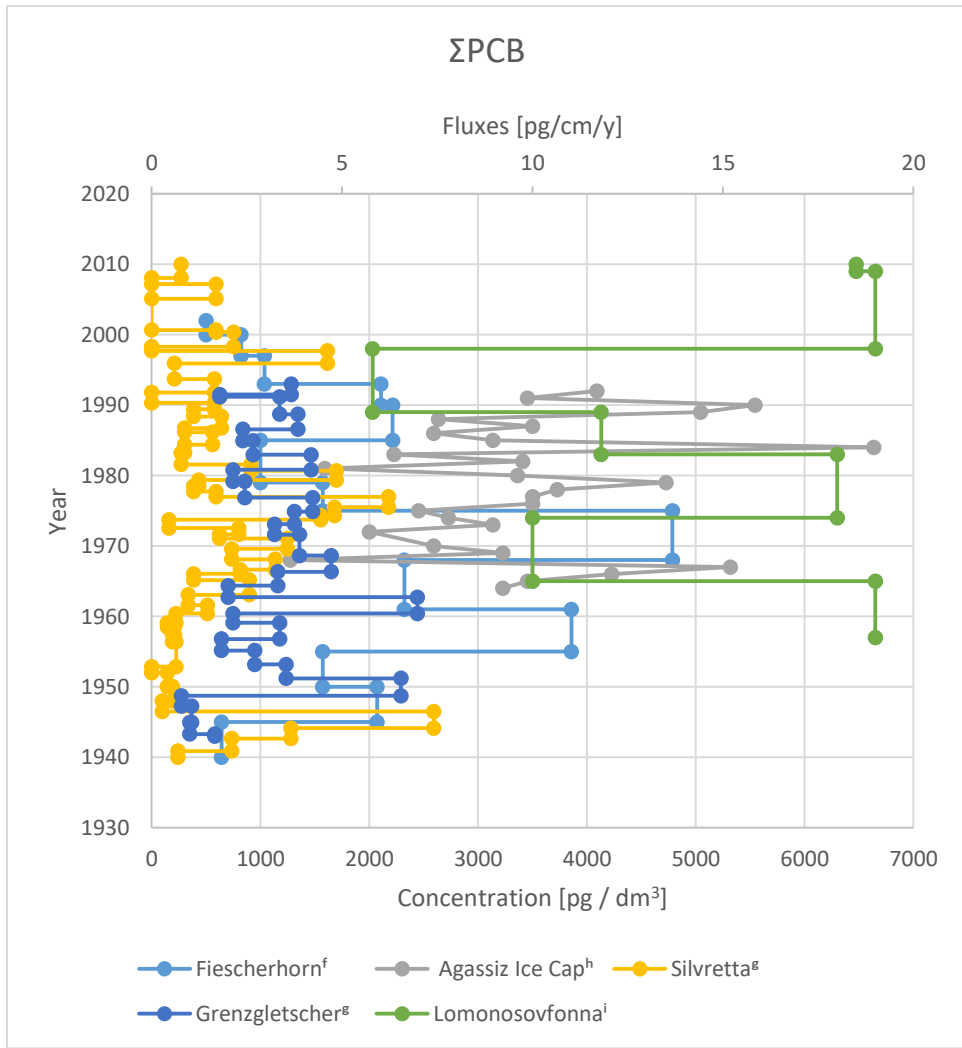

ΣDDT

ΣHCH
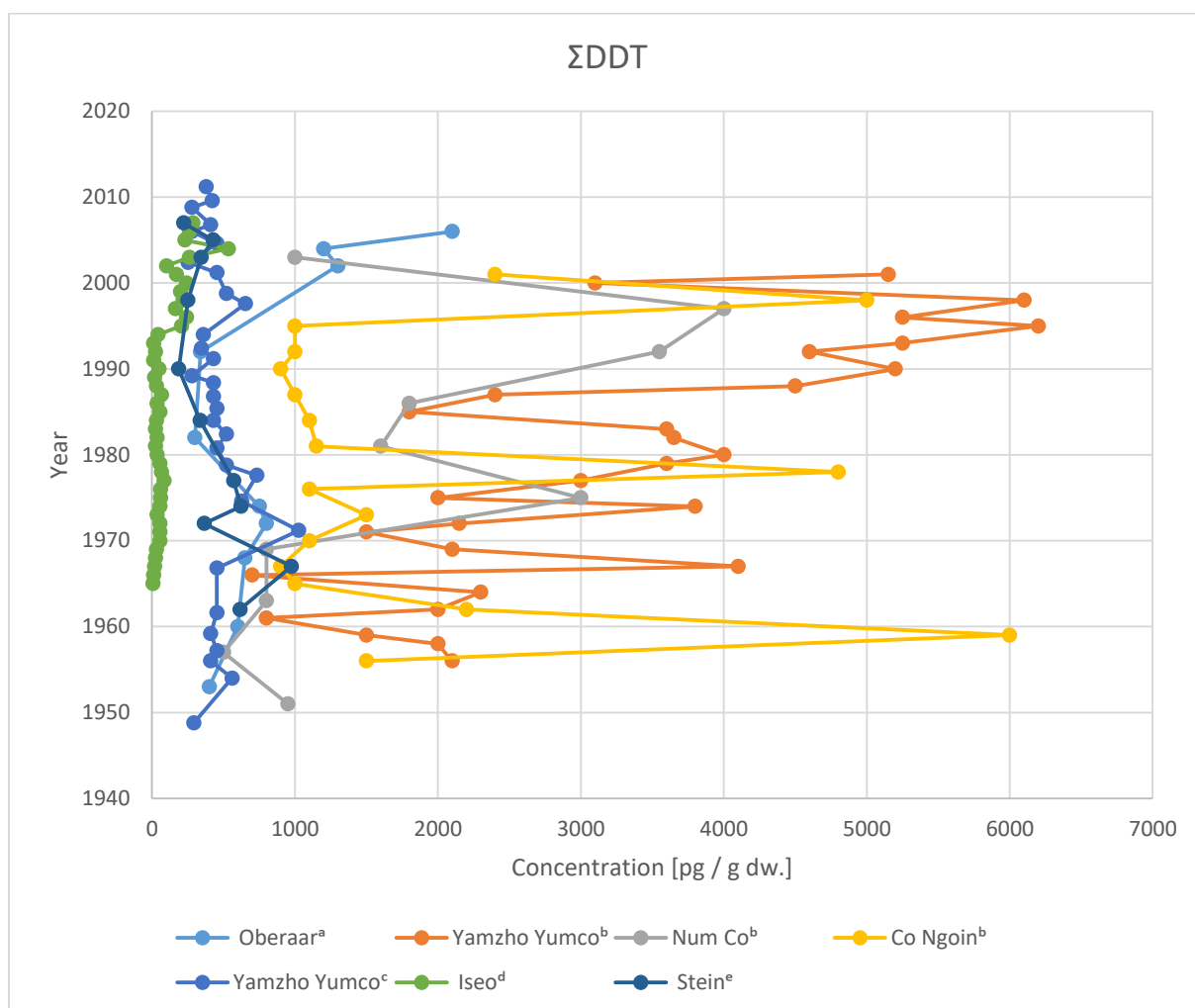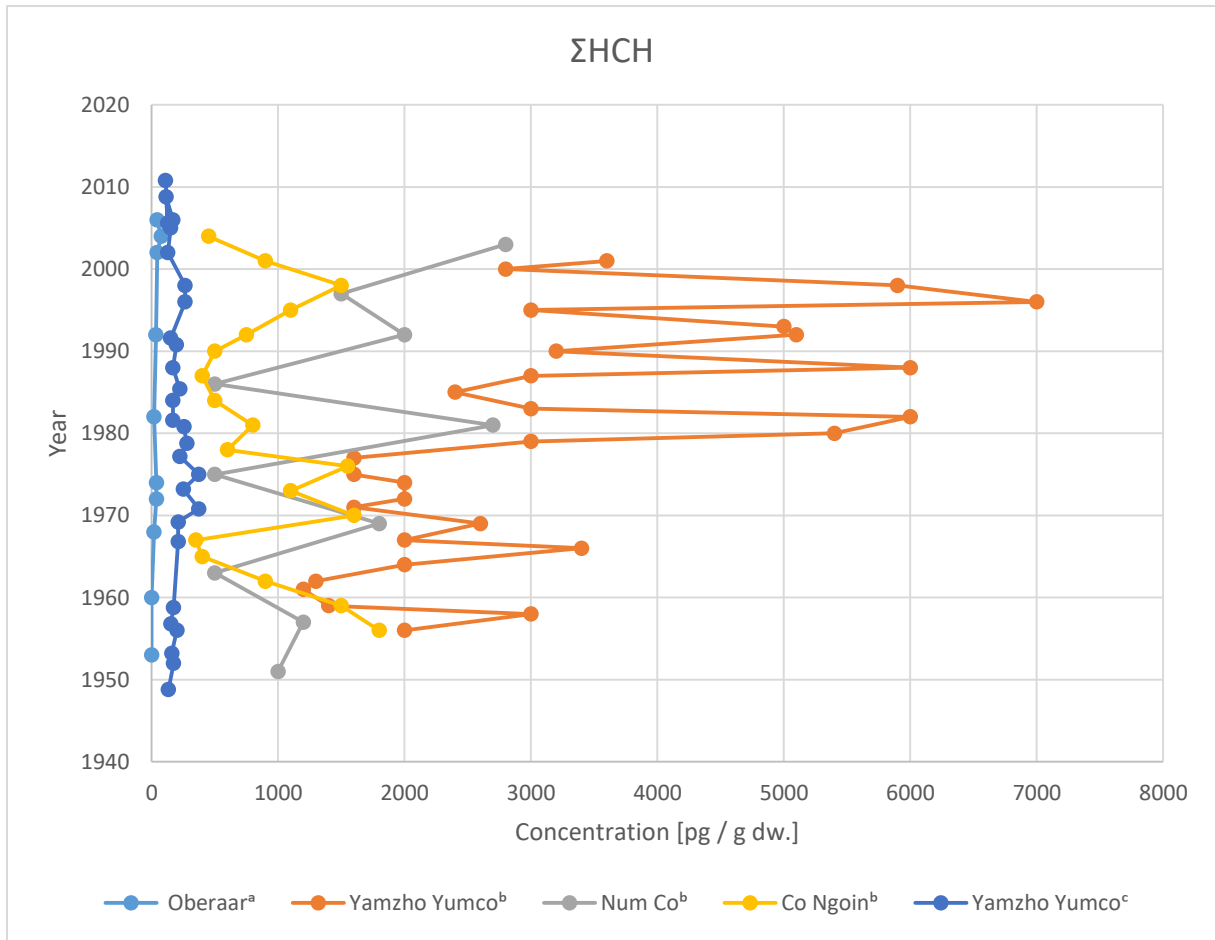
Figure 6. Concentrations of ΣDDT, ΣHCHs, ΣPCBs in the sediment cores from proglacial lakes in the European Alps and Himalaya. a – (Bogdal et al., 2009), b – (Cheng et al., 2014), c – (Sun et al., 2018), d – (Bettinetti et al., 2011), e – (Schmid et al., 2011), f – (Pavlova et al., 2016), e – (Schmid et al., 2011).
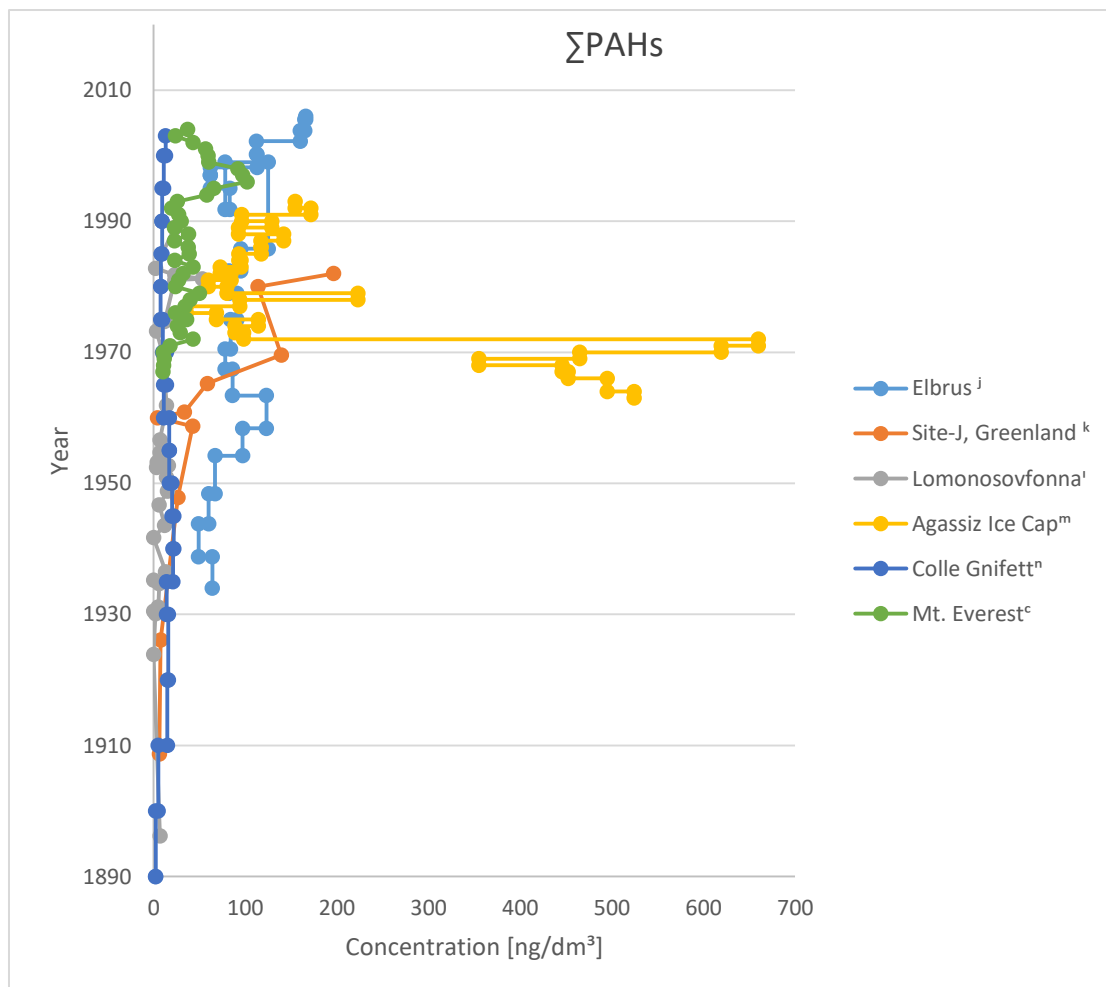
ΣDDT

Concentration [pg / dm³]

Year

Depth [m]

Lysᵃ   Snow Domeᵇ   East Rongbukᶜ   Lysᵈ   Jarvisᵉ

ΣHCH

Legend: Lys[a], Snow Dome[b], East Rongbuk[c], Lys[d], Jarvis[e]

ΣPCB

Figure 7. Concentration of ΣDDTs (please note: the age of layers was not specified for Jarvis glacier, hence the concentrations are plotted using depth), ΣHCHs (Jarvis glacier concentrations are plotted by depth instead of age), ΣPCBs (please note that the contents of the ΣPCBs for Lomonosovfonna should be read from the top, secondary axis (as fluxes)), ΣPAHs in the ice cores from published literature. a – (Villa et al., 2006), b – (Donald et al., 1999), c – (Wang et al., 2008), d – (Villa et al., 2003), e - (Miner et al., 2018), f – (P.A. Pavlova et al., 2014), g – (Pavlina Aneva Pavlova et al., 2015), h – (D.J. Gregor et al., 1995), i – (Garmash et al., 2013), j - (Vecchiato et al., 2020), k - (Kawamura et al., 1994), l - (Vehviläinen et al., 2002), m - (Peters et al., 1995), n - (Gabrieli et al., 2010b).

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