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Chemicals of Emerging Arctic Concern in north-western Spitsbergen snow: Distribution and sources

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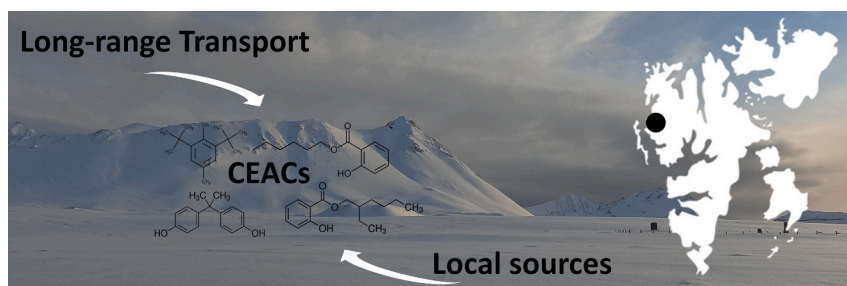
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HIGHLIGHTS

- Personal care product chemicals are present in the snowpack on Spitsbergen.
- Concentrations of some PCPs are different across altitudes.
- UV filters show higher concentrations during winter at most sites.
- Chemicals of concern partition to particulate and dissolved phases differently.

GRAPHICAL ABSTRACT



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ABSTRACT

Personal care products contain chemicals that are considered of emerging concern in the Arctic. In this study, a selected group of personal care products was investigated in the snowpack on north-western Spitsbergen. We report a preliminary study on the spatial and seasonal distribution of 13 ingredients commonly found in personal care products, including fragrance materials, UV filters, BHT and BPA. Possible sources and deposition processes are discussed. Experimental analyses utilizing GC–MS/MS, were complemented with outputs from the HYSPLIT transport and dispersion model. The results reveal the presence of all selected compounds in the snow, both in proximity to and distant from the research village of Ny-Ålesund. For some of these chemicals this is the first time their presence is reported in snow in Svalbard. These chemicals show different partitioning behaviours between the particulate and dissolved phases, affecting their transport and deposition processes. Additionally, concentrations of certain compounds vary across different altitudes. It is observed the relevance of long-range atmospheric transport during winter at most sites, and, regardless of the proximity to human settlements, snow concentrations can be influenced by long-distance sources. This study highlights the need for detailed

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information on CEACs' physical-chemical properties, considering their potential impact on fresh and marine waters during the snowmelt under climate change.

1. Introduction

The implementation of pollution monitoring programs in the Arctic has made it clear that this region is no longer as pristine as it was considered at the beginning of the past century. For a long time, the environmental occurrence of legacy contaminants, such as polycyclic aromatic hydrocarbon (PAHs), polychlorinated biphenyls (PCBs), and polybrominated diphenyl ethers (PBDEs) represented the major concern, and their presence in arctic environmental matrices was widely documented (Balmer et al., 2019; Herbert et al., 2005; Kelly et al., 2008; Muir et al., 1992; Su et al., 2007; Vecchiato et al., 2018, 2015; Xie et al., 2022). Due to international efforts and regulations, the occurrence of these contaminants in the Arctic has decreased (Arctic Monitoring and Assessment Programme (AMAP), 2017). However, there is an increasing interest in another category of chemicals, known as Chemicals of Emerging Arctic Concern (CEACs) (Arctic Monitoring and Assessment Programme (AMAP), 2017). CEACs represent a valid concern due to their rising production and use, the documented effects they have on living organisms (Sonne et al., 2021), and the current lack of knowledge regarding their distribution and behaviour in the environment (Arctic Monitoring and Assessment Programme (AMAP), 2017). In fact, many of these chemicals are not subject to any international regulation (Arctic Monitoring and Assessment Programme (AMAP), 2017). So far, the occurrence of CEACs in the Arctic has been documented in surface waters (Tsui et al., 2014), seawater (Vecchiato et al., 2018), terrestrial and marine biota (Sonne et al., 2021; Vorkamp et al., 2019), wastewater (Gunnarsdóttir et al., 2013), and snow (Lebedev et al., 2018; Vecchiato et al., 2018). The environmental matrix in which these chemicals are more likely to be found may vary depending on the environmental conditions of the deposition area and the specific chemical properties of the compounds (Kallenborn et al., 2018). To comprehend the environmental fate of these chemicals it is important to investigate their distribution, focusing on potential sources of contamination, transport patterns, and deposition processes.

The CEACs include several types of chemicals, including pharmaceuticals, personal care products ingredients (PCPs), currently used pesticides (CUPs), organophosphate esters (OPEs), and per- and poly-fluoroalkyl substances (PFAS) (Arctic Monitoring and Assessment Programme (AMAP), 2017; Kallenborn et al., 2018; Xie et al., 2022). In this study, we focus on ingredients commonly present in PCPs, namely Fragrance Materials (FMs), organic UV filters (UVFs), and other chemicals, such as Butylated hydroxytoluene (BHT) and Bisphenol A (BPA) (Table S1). These chemicals were selected due to their stability, high volume production, and the recent controversy about their environmental impact (Brooke et al., 2008; Gaudin, 2013; Huang et al., 2021; Picone et al., 2021).

Fragrance Materials are commonly used as perfumes in a wide range of PCPs (soaps, shampoos, body lotions, etc.). The selected fragrances are stable enough to be utilized in aggressive commercial products, such as bleach or acid cleaners, and their scent can persist on blotters for weeks to months (Vecchiato et al., 2018). Previous studies detected these analytes in urban (Vecchiato et al., 2021, 2016) and remote (D'Amico et al., 2022; Vecchiato et al., 2020, 2018, 2017) areas deriving from local and distant sources. Moreover, research has shown the potential estrogenic effects of these compounds in humans and rats (Charles and Darbre, 2009; Zhang et al., 2013, 2012). The investigated fragrances also proved to be ecotoxic to copepods, inhibiting the larval development of *A. tonsa* (Picone et al., 2021). In a recent study conducted on *Mytilus galloprovincialis* in the Venice Lagoon adverse effects of Amyl Salicylate were observed, including changes in immune response and cell death (Bernardini et al., 2022).

Organic UVFs are chemicals capable to absorb UV radiations at different absorption bands. They are used in sunscreens, other PCPs, and in industrial products as stabilizers (Huang et al., 2021). Generally, organic UVFs are applied as a blend within different products to increase the UV absorption range (Huang et al., 2021). These substances have been found to be bioaccumulative, phototoxic, and harmful to the endocrine system (Montes-Grajales et al., 2017; Vuckovic et al., 2022). Some of the compounds included in this work are currently under investigation and evaluation. For instance, in 2021, the Scientific Committee for Consumer Safety of the EU Commission concluded that concentrations of Benzophenone-3 and Octocrylene need to be reduced in cosmetic products. Additionally, it is worth noting that Hawaii, the U. S. Virgin Island, and Palau have implemented a ban on Benzophenone-3 (BP3), Octocrylene (OCR), and 2-Ethylhexyl 4-methoxycinnamate (EHMC) due to their adverse effects on coral reefs (Miller et al., 2021). A recent study detected EHMC in the snow of Terra Nova Bay (Antarctica) (D'Amico et al., 2022), partly as a result of human activities in scientific bases and facilities, and possibly due to long-range atmospheric transport.

In addition to the two aforementioned groups of compounds, this study also investigated BHT and BPA since they are present either in PCPs or their packaging. BHT is a widely used synthetic antioxidants in various products, including food, PCPs and plastic manufacturing (Yehye et al., 2015). The European Union (EU) Scientific Committee on Consumer Safety considered its use in cosmetic products to be safe under certain circumstances in 2021, but the environmental aspects were not addressed (EU SCCS, 2021). In vitro experiments conducted on zebrafish have demonstrated that the BHT is an endocrine disruptor (Yang et al., 2018), it may affect the normal organogenesis of zebrafish embryos (Sarmah et al., 2020), and alter their dopamine-related gene expression (Liang et al., 2020). BPA is a plasticizer largely used in the production of plastic packaging and epoxy resins (Vandenberg et al., 2007). It can have carcinogenic effects on a large number of animals, and is also considered an estrogenic and endocrine disruptor (Barroso et al., 2019; Levy et al., 2004; Vandenberg et al., 2007), especially early exposure studies have shown that BPA can affect marine biota (Minier et al., 2008). BPA also poses a risk to human health as it accumulates in tissues and organs, interfering with the normal functioning of the liver and thyroid, among others (Rochester, 2013). Due to its effects on human and animal health, in recent years, the use of BPA has been regulated in the EU and in some states of the United States (Aungst et al., 2014; European Commission, 2018). Its presence in snow was documented in Germany (Prieto et al., 2010), and Minnesota (Ferrey et al., 2018). However, to the best of our knowledge, the presence of BPA in Arctic snow has not been investigated.

All these chemicals can enter the environment in different ways depending on their physical-chemical properties (Table S2), use, and disposal methods. For example, domestic and industrial wastewaters are important sources of BHT (Barbosa et al., 2016), BPA (Liu and Mabury, 2020), and PCPs in general (Anand et al., 2022; Vecchiato et al., 2016). In the aquatic compartment, these compounds can be discharged into both fresh and marine waters through wastewater plants (Emnet et al., 2015; Meng et al., 2016; Rodil et al., 2010) or may directly enter during recreational water activities (O'Malley et al., 2021). Target CEACs can also be emitted directly into the atmosphere. Fragrances in perfumes, and spray sunscreens are released into the air during their use or production (Pegoraro et al., 2020). BHT can enter the atmosphere through evaporation from treated surfaces (Hodgson et al., 1993), while BPA, might be emitted through the burning of plastic or the disposal and burning of electronic waste (Fu and Kawamura, 2010). These chemicals can also enter the atmosphere by re-volatilization from environmental

matrices. Air – sea gas exchange is an important secondary emission source as well as a transport process for fragrance materials (Xie et al., 2007). The UVFs can re-volatilize from the soil, atmospheric particles, water, and even vegetation (Pegoraro et al., 2020). Fugitive air – water emission of BPA has been reported as a mode of emission to the atmosphere near factories and waste treatment plants (Vasiljevic and Harner, 2021).

The presence of PCPs in polar regions demonstrates that their ability to be deposited and accumulate in cold environments, yet little is known

about the primary sources of contamination in remote regions and the main transport processes. However, available studies in the Arctic suggest that their occurrence is linked to inadequate local waste treatment (Gunnarsdóttir et al., 2013; Kallenborn et al., 2018; Tsui et al., 2014), and both long-range and regional oceanic and atmospheric transport (Tsui et al., 2014). In the latter case, rain and snow can scavenge gaseous and particle-bound organic contaminants from the atmosphere, and their efficiency depends on the atmospheric temperature and the chemical characteristics (Lei and Wania, 2004). This process reduces

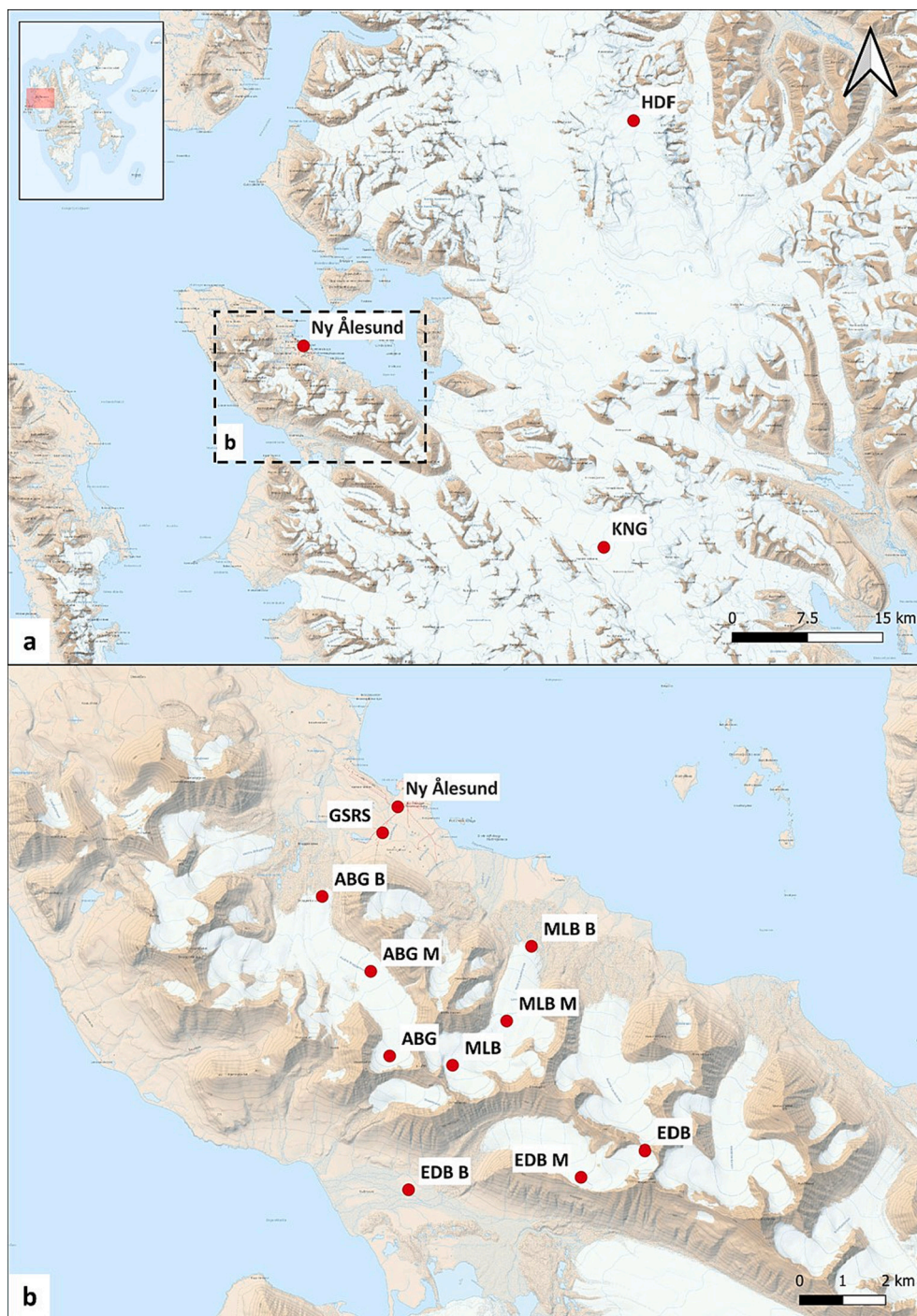


Fig. 1. Ny-Ålesund and the sampling sites. (a) Ny-Ålesund and the sampling points of snow pits on the glaciers Kongsvegen (KNG), and Holtedahlfonna (HDF). (b) Detailed map of the area around Ny-Ålesund. The sampling points on the glaciers: the snow pits the top of the Austre Brøggerbreen (ABG), Midtre Lovénbreen (MLB), Edithbreen (EDB); the surface snow collected at the middle (M) and at the base (B) of the three glaciers; and the sampling point at the Gruvebadet Snow Research Site (GRSR). (Toposvalbard, ©Norwegian Polar Institute).

their presence in the atmosphere by incorporating them into the snowpack, which acts as a temporary reservoir. These compounds are subsequently released again into the environment in a relatively short time after snow melts (Daly and Wania, 2004).

This study focuses on investigating the presence of a selected group of CEACs in snow samples collected from north-western Spitsbergen, specifically at the Gruvebadet Snow Research Site (Ny-Ålesund) and in five glaciers, three of which are situated in the peninsula. To the best of our knowledge, this is the first study investigating the occurrence of BP3, Ethylhexyl Salicylate (EHS), EHMC, OCR, BHT, and BPA in arctic snow.

The objectives of this study are i) to investigate the presence of CEACs in the snow of north-western Spitsbergen and understand their distribution between the particulate and the dissolved phases in a cold environment; ii) to assess the spatial, altitudinal, and seasonal variability of these compounds in the area; and iii) to identify potential sources of contamination, considering the village the main local source in the area.

The aim is to provide an initial overview of the environmental presence of these compounds, which have been overlooked in this region until now. This research contributes to protecting the Arctic ecosystem by providing data on their snow distribution, which can be helpful in defining a future comprehensive monitoring plan for these compounds in the area.

2. Materials and methods

2.1. Study site and sampling methodology

Located in north-western Spitsbergen (Svalbard), Ny-Ålesund was originally built as a mining village. However, since the 1960s, the area has transformed into a permanent international scientific research settlement. The village has its own airport and harbor, hosting research institutes from over 13 countries year-round, with an average of about 40 people during winter and up to 200 people in summer.

In this study, the Gruvebadet Snow Research Site (GSRs, 78.918 N, 11.895 E) is the closest sampling site to Ny-Ålesund. It is situated approximately 1 km south of the village at an elevation of 40 m a.s.l. (Fig. 1). The sampling area lies about 80 m from the Gruvebadet Atmospheric Laboratory, both of which are managed by the Italian National Research Council (CNR).

The selected glaciers are shown in Fig. 1, three are in the Brøggerhalvøya peninsula, near Ny-Ålesund and the coast (Edithbreen (EDB), Midtre Lovénbreen (MLB), Austre Brøggerbreen (ABG)), while the other two (Kongsvegen (KNG) and Holtedahlfonna (HDF)) are inland and more isolated from human settlements. The five glaciers differ in length, altitude, slope, and surrounding topography. The sampling points were chosen along the glaciers' profile to investigate the spatial variability, ranging from areas relatively close to the village to more distant ones. This approach provides a better understanding of the transport processes involved based on distance and altitude. Indeed, the elevation of the glaciers considered in this study ranges from about 400 m and 1120 m, which is significant as the winter boundary layer in the area of Ny-Ålesund also varies within this range (Maturilli and Kayser, 2017). This diverse elevation gives the opportunity to collect sampling points above and below this boundary layer, and to discuss the effects of local and long-distance sources of contamination.

A total of twenty-five snow samples were collected in spring 2021 (Table S3). Considering the purposes of this research, the sampling was planned according to the following criteria: at the GSRs, surface snow samples ($n = 4$; 10 cm of thickness) were collected once per week from the 12th of April to the 3rd of May 2021. At the top of each glacier, instead, one snow pit was dug, and three samples were collected for each pit. The three samples were collected to be representative of the different phases of the 2020/2021 snow season. They consist of a surface snow sample (~20 cm), representing the most recent spring snow deposition, a basal snow sample (the first ~30 cm from the bottom of the

snowpit consisting of bare ice or firn), representative of early fall deposition, and an intermediate snow sample (integrating the snow layers in between the previous two samples), generally representative of late-fall and winter deposition. In addition to the snow samples collected in the summit area of the glaciers, two surface snow samples (about 10 cm deep) were collected at the base and at the middle of the three glaciers closest to Ny-Ålesund (Midtre Lovénbreen, Austre Brøggerbreen, and Edithbreen respectively). The elevation of these sampling points ranges from approximately 57 m and 521 m (Table S3).

The samples were collected in 5 L stainless steel barrels and in aluminium trays using a density wedge cutter made of aluminium.

2.2. Target chemicals for the analyses

In this study we selected 13 ingredients present in PCPs; 7 FM: Amyl and Isoamyl Salicylate (pentyl 2-hydroxybenzoate and 3-methylbutyl 2-hydroxybenzoate), Hexyl Salicylate (hexyl 2-hydroxybenzoate), Benzyl Salicylate (benzyl 2-hydroxybenzoate), Oranger Crystals (1-naphthalen-2-ylethanone), Peonile® (2-cyclohexylidene-2-phenylacetone), and Ambroxif® (Dodecahydro-3a,6,6,9a-tetramethylnaphtho[2,1-b]furan); 4 organic UV filters (UVFs): 2-Ethylhexyl 4-methoxycinnamate (EHMC; 2-ethylhexyl 3-(4-methoxyphenyl)prop-2-enoate), Benzophenone-3 (BP3; (2-hydroxy-4-methoxyphenyl)-phenylmethanone), Ethylhexyl Salicylate (EHS; 2-ethylhexyl 2-hydroxybenzoate), and Octocrylene (OCR; 2-ethylhexyl 2-cyano-3,3-diphenylprop-2-enoate); and other chemicals: Butylated hydroxytoluene (BHT; 2,6-ditert-butyl-4-methylphenol); and Bisphenol A (BPA; 4-[2-(4-hydroxyphenyl)propan-2-yl]phenol) (Table S1).

2.3. Analytical methods

The analytical procedure used for the sample preparation and analysis is described in Vecchiato et al. (2020, 2018). After the collection of snow, the samples were transported to the Italian Station 'Dirigibile Italia', and they were thawed at ambient temperature (around 25 °C). All the samples were filtered with glass microfiber filters (GMF, Sartorius 0.7 µm, 47 mm), and extracted using solid phase extraction (SPE, Waters Oasis® HLB cartridges, 200 mg, 6 cm³, 30 µm particle size, Waters Corp., Milford, MA USA). The SPE cartridges were previously conditioned with pesticide-grade 10 mL of *n*-hexane, 10 mL of dichloromethane (Romil Ltd., Cambridge, UK), followed by 10 mL of ultrapure water (ELGA, High Wycombe, UK). Cartridges and filters were stored at -20 °C and later eluted in the stainless steel clean-room laboratories for organic analyses ISO 7 at the Ca' Foscari University of Venice (Italy). The samples were eluted with pesticide-grade 1 mL of toluene, 15 mL of dichloromethane, and 10 mL of *n*-hexane (Romil Ltd., Cambridge, UK). Eluates were dried with an aliquot of Na₂SO₄ and concentrated to 250 µL under nitrogen flow at 23 °C (Turbovap II®, Caliper Life Science, Hopkinton, MA, USA). Phenanthrene ¹³C and Octocrylene d¹⁰ were used as Internal Standards, and samples were spiked prior to elution (50 µL, 50 ng, and 140 ng abs respectively).

Filters were extracted using 10 mL of *n*-hexane/dichloromethane (1:1 v/v), spiked with Phenanthrene ¹³C and Octocrylene d¹⁰ (50 µL, 50 ng, and 140 ng abs respectively), dried with an aliquot of Na₂SO₄ and sonicated for 15 min. The extracts were collected, and a second round of extraction was done. All combined extracts were concentrated following the same procedure described above for the SPE eluates.

The instrumental analysis was conducted using the GC-MS/MS (Trace 1310 - TSQ 9000 Thermo Fisher). The gas chromatographic separation was executed on a 60-m HP-5MS (0.25 mm I.D., 0.25 µm; Agilent Technologies, Avondale, USA). The initial temperature in the oven is 120 °C (1.0 min) increasing to 180 °C at 25 °C min⁻¹, subsequently increased to 250 °C at 10 °C min⁻¹, and finally to 310 °C at 20 °C min⁻¹ (8.6 min). The SRM transitions were calculated for the selected chemicals, and they are reported in Table S4. All the GC-MS/MS data were analysed using Thermo Scientific Chromeleon 7.3.1

Chromatography Data System (CDS).

2.4. Quality assurance and quality control

To avoid cross-contamination the steel bins and the aluminium trays were rinsed with pesticide-grade acetone and *n*-hexane prior to sampling. All glassware was rinsed with pesticide-grade acetone, dichloromethane, and *n*-hexane prior to use. In the field, the following measures were employed to prevent contamination: the sampling was conducted using nitrile gloves and avoiding any plastic material; and the face of the operator was completely covered by the ski mask. The sampling points were chosen in undisturbed areas, meters away from snowmobile traffic, and avoiding areas where previous activities were conducted from other researchers.

In the clean laboratory, in addition to the needed precautions of the work environment, no creams or other personal care products were used before entering the laboratory.

Concentrations were calculated using the isotopically labelled internal standards. Laboratory blanks ($n = 3$) were prepared filtering and extracting 1 L of ultrapure water (ELGA, High Wycombe, UK) in the clean laboratory of the Italian station 'Dirigibile Italia' under a laminar flow hood, and following the same procedure described for the samples. The results were mean-blank corrected, and the method detection limits (MDL) were calculated as three times the standard deviation of the blank signals and are reported in Table S5. The procedural recoveries of the analytical method were calculated spiking known amounts of the selected analytes in surrogate samples ($n = 5$), range between $92 \pm 3\%$ and $102 \pm 3\%$ in the dissolved fraction and $90 \pm 2\%$ and $100 \pm 2\%$ for the particulates. Slightly lower recoveries were detected for BHT, resulting at $86 \pm 7\%$ and $81 \pm 3\%$, respectively for the dissolved and the particulate fractions.

Among all the samples HDF S stands out as an outlier. This is likely due to sample contamination of Bisphenol A and Benzophenone-3. However, considering the high values of these two chemicals in HDF S compared to the others, the reliability of the other samples should not be affected.

Statistical analyses were performed using the STATISTICA 7 Data Analysis Software System (StatSoft Inc.) and R (RStudio Team, 2023, www.posit.co). All data, including non-detect data, were analysed through a non-parametric Kaplan-Meier test (Helsel, 2005).

2.5. Back trajectory analysis

The origin of air masses was determined using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPPLIT) transport and dispersion model of the National Oceanic and Atmospheric Administration (NOAA) (Stein et al., 2015). The meteorological data were retrieved from the NCEP/NCAR Reanalysis database on the NOAA website. The endpoint of the trajectories is the GSRS, and the trajectories were calculated daily from 10 m above ground level every 6 h starting from one week before the first sampling at the GSRS and, later, during the days separating the samplings.

3. Results and discussion

3.1. Fragrance materials in snow

The concentrations of the seven FMs (\sum_7 FMs) in the 25 samples are shown and summarized in Fig. 2. The results refer to the total concentrations calculated in the dissolved and particulate phases considering the volume of water passing through the filters (Table S8). The total FM concentrations range from 1.4 ng L^{-1} (GSRS 4) to 148 ng L^{-1} (MLB M), where the highest concentration of a single FM compound was also detected for HexS (56 ng L^{-1}) (Table 1). The available literature on the occurrence of FMs in polar regions is limited. The concentrations found in this study are similar to those found in snow samples in the area

around Ny-Ålesund by Vecchiato et al. (2018) (up to 72 ng L^{-1}), and to the ones found in Antarctic seawater during the melting season (Vecchiato et al., 2017) (intermediate range of $18\text{--}64 \text{ ng L}^{-1}$). These concentrations are lower than the maximum concentration (281 ng L^{-1}) found in an ice core from Mt. Elbrus (Caucasus) by Vecchiato et al. (2020).

We observe that the dominant fragrances are the salicylates; Hexyl Salicylate, Amyl, and Isoamyl Salicylate account for 37 %, 25 %, and 20 % of the \sum_7 FMs concentrations, respectively. This is consistent with previous findings in the same area (Vecchiato et al., 2018), and it is understandable considering that salicylates are widely used in personal care products due to their low price, leading to an increase in their consumption over the years (Gaudin, 2013). However, the maximum concentrations of the salicylates found in this study are lower than the potentially toxic levels (Hexyl Salicylate = 57 ng L^{-1} ; Amyl Salicylate = 131 ng L^{-1} ; Benzyl Salicylate = 2795 ng L^{-1}) for brackish and marine invertebrate species (Picone et al., 2021).

3.2. UV filters in snow

The concentration of the \sum_4 UVFs ranges from 0.2 ng L^{-1} (ABG W) to 358.4 ng L^{-1} (HDF S) (Table 1 and Fig. 2). In this study, the dominant UVFs are BP3 and EHS with concentrations that account for 48 % and 40 % respectively of the total concentrations of the UVFs in the 25 snow samples. However, the high percentage of BP3 is linked to the concentration found in HDF S (358.4 ng L^{-1}) which is the same order of magnitude as the maximum value registered in wastewater treatment plant effluents (195 ng L^{-1}) in Antarctica by Emmet et al. (2015). This value represents an outlier within the dataset, and it might be the result of sample contamination during its collection and/or preparation. Although Hermanson et al. (2020) show that high concentrations of organic contaminants at high altitudes in Svalbard glaciers may be related to different types of sources compared to samples collected at lower altitudes, we decided to exclude HDF S from this discussion. If we do not consider this sample, the dominant UVFs concentration becomes EHS (64 %), followed by BP3 (18 %). This is consistent with their high production and manufacturing: in the European Union alone, EHS is manufactured and/or imported between 1000 and 10,000 tonnes per year, while BP3 is between 100 and 1000 tonnes per year. Several studies have been conducted to understand the harmful effects of these two chemicals on human health and the environment. EHS has shown to bioaccumulate and have potentially toxic effects on zebrafish larval metabolism (Zhou et al., 2022). BP3 has recently raised serious concerns due to its phototoxicity (Vuckovic et al., 2022) and its effects on human health as a possible endocrine disruptor (DiNardo and Downs, 2017).

The occurrence of UVFs was detected in several matrices in polar regions. The EHMC, OCR, and BP3 were found in surface seawater in the Arctic Ocean and in the Chukchi Sea with concentrations ranging between 25 and 66 ng L^{-1} for EHMC, $26\text{--}31 \text{ ng L}^{-1}$ for OCR, and $17\text{--}33 \text{ ng L}^{-1}$ for BP3 (Tsui et al., 2014). These concentrations are higher than the ones found in the current study and in other studies conducted on arctic char samples and bird eggs (Lucia et al., 2016; Provencher et al., 2022). In Antarctica, different UVFs were detected in wastewater treatment plant effluents (WWTP) close to scientific stations, in seawater, freshwater, sea ice, and snow with concentrations of the same order of magnitude as the ones found in the current study, except for the maximum values of the Antarctic WWTP which are one order of magnitude higher than ours (D'Amico et al., 2022; Domínguez-Moruco et al., 2021; Emmet et al., 2015).

3.3. BHT and BPA in snow

The BHT is only found in five samples in the particulate phase, and it results below the detection limit in the dissolved phase in all the samples (Tables S6 and S7). The concentrations range between 0.2 ng L^{-1} (ABG M and GSRS 2) and 1.4 ng L^{-1} (EDB B). Literature on the occurrence of

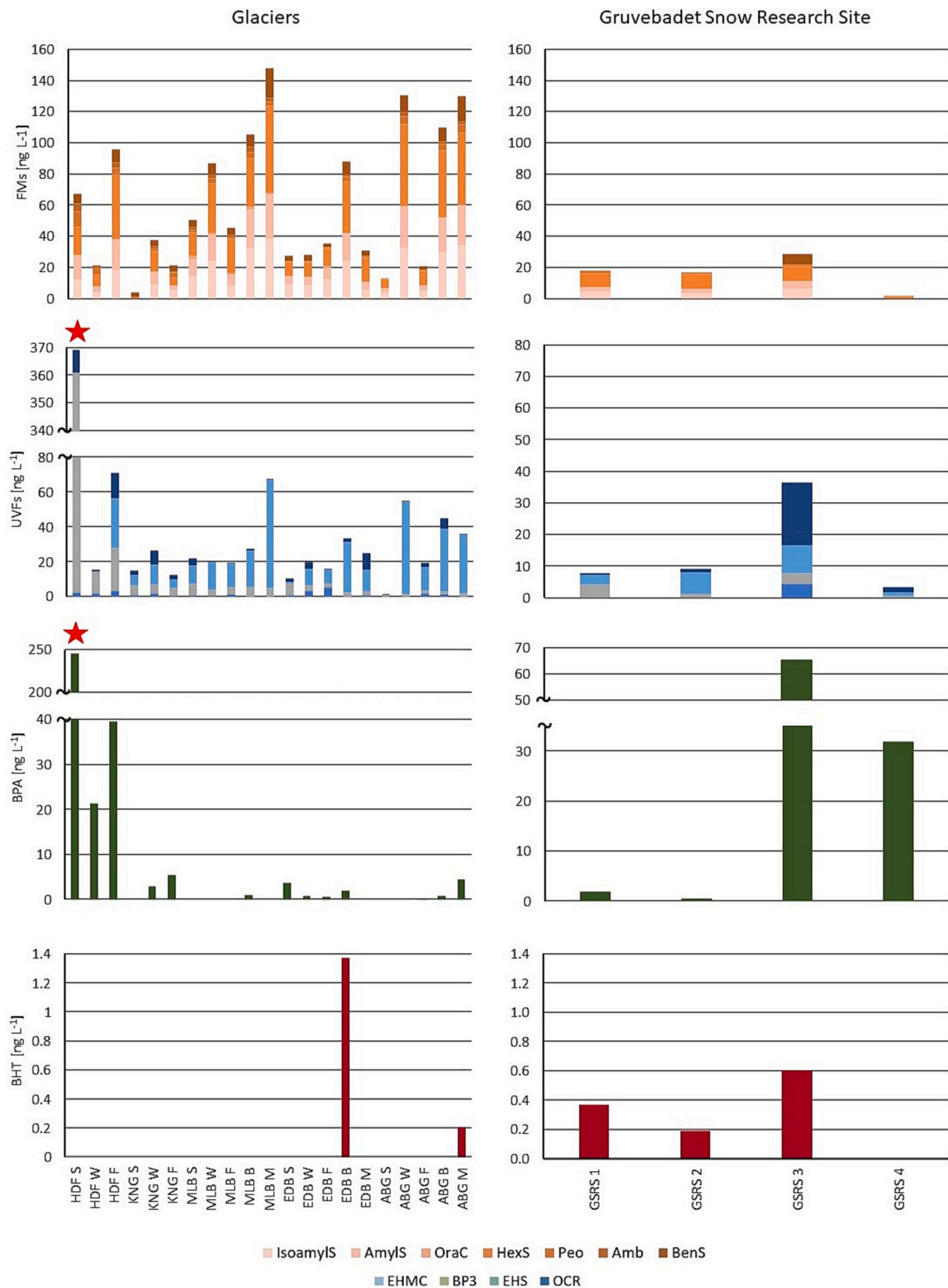


Fig. 2. Fragrance materials (FMs), UV Filters (UVFs), BPA and BHT in snow (ng L^{-1}) at the selected glaciers: S – Spring, W – Winter, F – Fall, B – Base of the glacier, M – Middle of the glacier; and at the Gruevbadet Snow Research Site: GRSR. The red star indicates the analytes and the sample not included in the discussion.

Table 1

The frequency detection in the dissolved and particulate fraction samples (F.D.), percentage distribution in the dissolved (D) and particulate (P) phases of the total concentrations, minimum and maximum detected values, summary statistics were calculated employing a non-parametric Kaplan-Meier test for the 7 FMs, 4 UVFs, BHT, and BPA concentrations (ng L^{-1}) in the twenty-five snow samples. The values refer to the sum of concentrations in the dissolved and particulate phases unless otherwise indicated.

| FMs | IsoamylS | AmylS | OraC | HexS | Peo | Amb | BenS |
|------------------|----------|-------|-------|------|-------|-------|--------|
| F.D. dissolved | 92 % | 92 % | 72 % | 92 % | 100 % | 56 % | 80 % |
| F.D. particulate | 24 % | 24 % | 0 % | 56 % | 0 % | 0 % | 4 % |
| D | 97 % | 97 % | 100 % | 94 % | 100 % | 100 % | 99.5 % |
| P | 3 % | 3 % | 0 % | 6 % | 0 % | 0 % | 0.5 % |
| Min | 3.6 | 2.7 | 0.2 | 1.1 | 0.3 | 1.0 | 2.2 |
| Max | 38.1 | 28.3 | 2.3 | 55.9 | 10.6 | 4.6 | 19.1 |
| Mean | 14.1 | 11.0 | 0.6 | 20.2 | 2.6 | 1.7 | 5.7 |
| Sd | 11.5 | 8.9 | 0.6 | 16.6 | 2.3 | 1.0 | 4.4 |
| Se | 2.3 | 1.8 | 0.1 | 3.3 | 0.5 | 0.2 | 0.9 |

| UVFs | EHMC | BP3 | EHS | OCR |
|------------------|------|-------|------|-------|
| F.D. dissolved | 64 % | 92 % | 76 % | 40 % |
| F.D. particulate | 24 % | 72 % | 60 % | 100 % |
| D | 82 % | 96 % | 86 % | 66 % |
| P | 18 % | 4 % | 14 % | 34 % |
| Min | 0.5 | 0.5 | 0.9 | 0.2 |
| Max | 5.5 | 358.4 | 61.9 | 19.6 |
| Mean | 1.4 | 18.7 | 15.7 | 3.7 |
| Sd | 1.4 | 71.0 | 16.3 | 4.9 |
| Se | 0.3 | 14.2 | 3.3 | 1.0 |

| Other compounds | BHT | BPA |
|------------------|-------|-------|
| F.D. dissolved | 0 % | 44 % |
| F.D. particulate | 20 % | 44 % |
| D | 0 % | 96 % |
| P | 100 % | 4 % |
| Min | 0.2 | 0.2 |
| Max | 1.4 | 245.1 |
| Mean | 0.3 | 17.2 |
| Sd | 0.3 | 50.5 |
| Se | 0.1 | 10.1 |

this compound in polar regions is limited. It was detected in seabird eggs in Canadian Arctic (Provencher et al., 2022), and in Antarctic krill (Wang and Liu, 2017), but to the best of our knowledge its presence was never investigated in snow.

Concentrations of BPA are found in both the dissolved and particulate phases. It is above the detection limit in 17 of the 25 samples, and concentrations range between 0.2 ng L^{-1} (ABG F) and 245 ng L^{-1} (HDF S) (Table S8). The highest concentration found in HDF S represents an outlier, in fact, as observed with BP3, the concentration is the same order of magnitude as the maximum concentration found in a wastewater treatment plant in Antarctica (986 ng L^{-1}) (Emnet et al., 2015). For this reason, as previously discussed for BP3, it was decided to exclude HDF S from the discussion on BPA. This way the range of concentrations becomes $0.2\text{--}65 \text{ ng L}^{-1}$ (GSRS 3). In the Arctic, the occurrence of BPA was documented in marine sediments from the Kongsfjorden (Svalbard) ($0.04\text{--}6.2 \text{ ng g}^{-1}$) (Spataro et al., 2022), in seabirds feathers in the Baltic ($86.8\text{--}809.5 \text{ ng g}^{-1}$ dw maximum values in two species) (Bodziach et al., 2022); and, in Antarctica, it was found in wastewater treatment plants effluents ($4.7\text{--}986 \text{ ng L}^{-1}$), in seawater ($<1.3\text{--}7.7 \text{ ng L}^{-1}$), and in sea ice ($<1.3\text{--}3.8 \text{ ng L}^{-1}$) (Emnet et al., 2015).

3.4. Distribution of CEACs in the dissolved and particulate phases

In Table 1 are listed the distributions of FMs, UVFs, BHT, and BPA in the dissolved and particulate phases. All the target FMs are present in the dissolved phase, and among these only salicylates are present in both phases. The studied UVFs and BPA are present both in the dissolved and particulate phases, while the BHT is solely present in the particulate phase.

Considering the different sampling locations, the percentage of the total concentrations of CEACs present in the dissolved phase range between 49 % (GSRS 1) and nearly 100 % (MLB M) (Fig. S1). In the particulate phase, the sample with the highest percentage is GSRS 1 (51 %), followed by EDB M (24 %) (Fig. S1). Fig. 3 shows the percentage of the distribution for $\sum_7\text{FMs}$, $\sum_4\text{UVFs}$, BHT, and BPA in the two analysed phases in the sampling sites. The most abundant group of chemicals are $\sum_7\text{FMs}$ (4 % - 91 %) and $\sum_4\text{UVFs}$ (9 % - 80 %). The highest percentage of $\sum_7\text{FMs}$ is found during spring in ABG S ($\sum_7\text{FMs} = 91 \%$, 12.8 ng L^{-1}), followed by winter in MLB W ($\sum_7\text{FMs} = 81 \%$, 86.8 ng L^{-1}), and at the base of the glacier MLB B ($\sum_7\text{FMs} = 79 \%$, 105.3 ng L^{-1}). For the UVFs, the sample with the highest percentage is found during spring in KNG S ($\sum_4\text{UVFs} = 80 \%$, 15.1 ng L^{-1}), followed by ABG F ($\sum_4\text{UVFs} = 48 \%$, 19.6 ng L^{-1}) during the fall. The different CEACs are differently distributed among sites: the FMs are abundant at lower elevations and in sites closer to Ny-Ålesund. Conversely, the UVFs are abundant both at high and low elevations. The ABG is the only glacier with a relatively high percentage of both FMs and UVFs, this might be due to its proximity to the village and due to the elevation of the sampling sites (111–451 m) that coincide with the lower oscillation of the winter boundary layer (Maturilli and Kayser, 2017). These two aspects might allow both local and long-range sources to influence the concentration of CEACs measured in the glacier snowpack. Additionally, the glacier is a research site frequently visited by scientists for monitoring and sampling, which could potentially lead to direct contamination.

BPA is detected in all the glaciers and at the GSRS, while BHT is found only in surface snow at Edithbreen, Austre Brøggerbreen, and GSRS.

3.5. Spatial and altitudinal distribution of CEACs in glaciers

The surface snow samples ($n = 10$) collected at the base, the middle, and the top of the glaciers show the presence of most of the target compounds in both phases (Fig. S2). The salicylates are the most frequently detected fragrances (9 out of 10 samples), we also observed the prevalence of the isoamyl, amyl and hexyl salicylates above the benzyl salicylate, in agreement with previous study conducted in the same area (Vecchiato et al., 2018). Among UVFs, the only compound detected in all samples is BP3 (range $1\text{--}25 \text{ ng L}^{-1}$). BPA is detected at low altitudes in the glaciers closer to the village (MLB B, ABG B and ABG M) and at the top and at the base of the Edithbreen on the other side of the peninsula. In the particulate phase, the main detected compounds are the UVFs, BPA, and the salicylates (Table S6). The only ubiquitous chemical in the particulate fraction is OCR ($0.3\text{--}6.0 \text{ ng L}^{-1}$).

The statistical analysis reveals no significant correlation between the concentration of CEACs in surface snow and the proximity to Ny-Ålesund, suggesting that the local influence of the village can be neglected in this case. This could be attributed to the local wind patterns, which are influenced by the Kongsfjorden and the surrounding topography making wind measurements at Ny-Ålesund particularly variable (Platt et al., 2022). The area is also affected by katabatic winds descending from the Kongsvegen glacier and funnelling into the fjord as they move westward (Platt et al., 2022). As all the sampling sites are located eastward of Ny-Ålesund, it is unlikely that they are directly affected by the wind transport of chemicals from the village. On the other hand, the concentrations show a significant negative correlation with the elevation of the sampling sites ($R = -0.73$) (Fig. S3). The samples collected at the top of glaciers are the least contaminated. Concentrations at

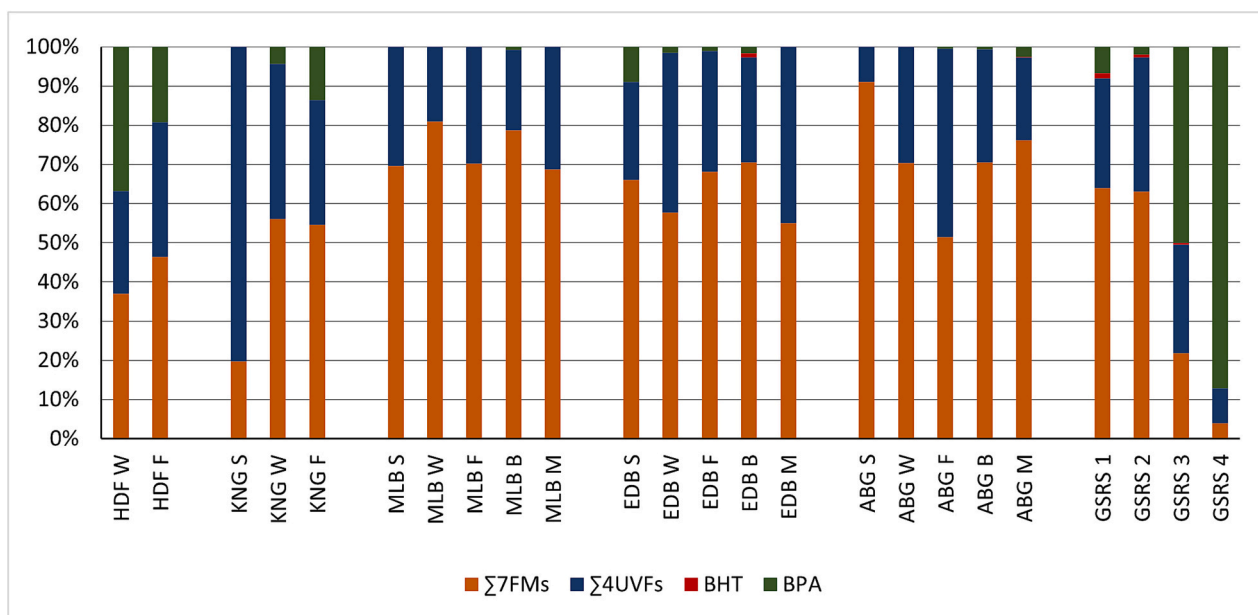


Fig. 3. The percentage of the concentrations (ng L^{-1}) of $\Sigma_7\text{FMs}$, $\Sigma_4\text{UVFs}$, BHT, and BPA in both the dissolved and particulate phases in all the sampling sites. Refer to Fig. 2 for details on single CEACs.

Edithbreen decrease with elevation, while the samples collected at Midtre Lovénbreen and Austre Brøggerbreen show that the highest concentrations are found in the middle of the glaciers, followed by samples collected at the bottom. The position of the slopes of the two glaciers can explain these observations. They face the surrounding area of the village and could be potentially affected by human activities: consequently, the snow at the base could be mixed for example by the passage of the snowmobiles, which would explain why the concentrations there are lower than in the middle of the glacier. This effect is not observed on the Edithbreen, situated on the opposite side of the peninsula and south-exposed, the area is relatively undisturbed compared to the other two sites. Looking at the distribution of the selected compounds in the three glaciers at the three different elevations we found that, even if concentrations are higher at lower altitudes, the salicylates are ubiquitous in the samples regardless of their elevation. It can be inferred, then, that the processes responsible for their deposition are not significantly affected by the vertical gradient. On the contrary, the concentrations of two UVFs, namely BP3 and OCR, are more abundant at the top of the glaciers between ~ 400 m and 630 m. High concentrations of these compounds were also observed in the surface snow sample collected at the top of the KNG (650 m), located at about 20 km from Ny-Ålesund, and where these compounds account for 27 % (BP3) and 15 % (OCR) of the total CEACs concentrations. The range of elevations at which these samples were taken falls within the range of the estimated winter boundary layer above Svalbard (Mayer et al., 2012). It is plausible that these UVFs were transported long-range through the atmosphere and deposited at the top of the glaciers possibly following a preferential scavenging process. It is rather common to observe a different altitudinal gradient between different groups of organic compounds (Daly and Wania, 2005). The altitude and the chemical characteristics play a role in the rate of deposition, in fact, precipitation increases with altitude specially at high latitudes and so does the efficiency of wet deposition of semi-volatile compounds like BP3 and OCR, particularly if associated with particulate matter (Daly and Wania, 2005). The scavenging process through snow deposition can reduce the concentrations of BP3 and OCR in the atmosphere and this would explain why we found different percentages at different altitudes along the profiles of the glaciers. However, further research should be done to

better understand the influence of the vertical gradient on the deposition of these compounds.

3.6. Seasonal distribution of CEACs

The seasonal distribution of CEACs in the snow pit samples ($n = 15$) differs between the two phases. The concentrations in the particulate phase do not show a particular seasonal distribution, while in the dissolved phase the highest cumulative concentration corresponds to the winter season in all glaciers except for Holtedahlfonna (Fig. 4). These results are consistent if we consider that during winter and spring, a phenomenon known as the Arctic Haze affects the atmospheric transport and deposition of contaminants in the Arctic region (Shaw, 1995). The haze reaches its peak between late winter and early spring, and it originates in industrial Eurasia, particularly from Northern Europe and Siberia (Platt et al., 2022). This phenomenon is linked to i) the reduction of solar radiation, and ii) to extremely dry conditions due to low temperatures (Shaw, 1995). During the winter period, the arctic atmosphere is loaded with contaminants and their vertical mixing is limited by the relatively low winter boundary layer. This phenomenon can be responsible for the higher concentration of contaminants found in winter samples at the Austre Brøggerbreen, Edithbreen, Midtre Lovénbreen, and Kongsvegen, suggesting distant sources of contamination. The seasonal values shown in Fig. 4 for Holtedahlfonna do not include BP3 and BPA for the reasons previously discussed. The distinct seasonal distribution, when compared to the other glaciers, can be explained by the elevation of the sampling site (1125 m) which is above the average winter boundary layer and is probably affected by air masses with a different source of origin.

With the exception of Holtedahlfonna, all the snow pits show at least one melting/freezing event during the winter season, plus ice layers at the bottom of the snowpack in the autumn horizon. These post-depositional processes can be related to the more frequent rain on snow events (ROS) and/or melting, and they may have changed the vertical distribution of the target chemicals. However, we do not expect any crossed influence between layers since the migration within the snowpack is confined to the portions between two or more ice crusts. This means, that in this case, any vertical change in distribution stayed

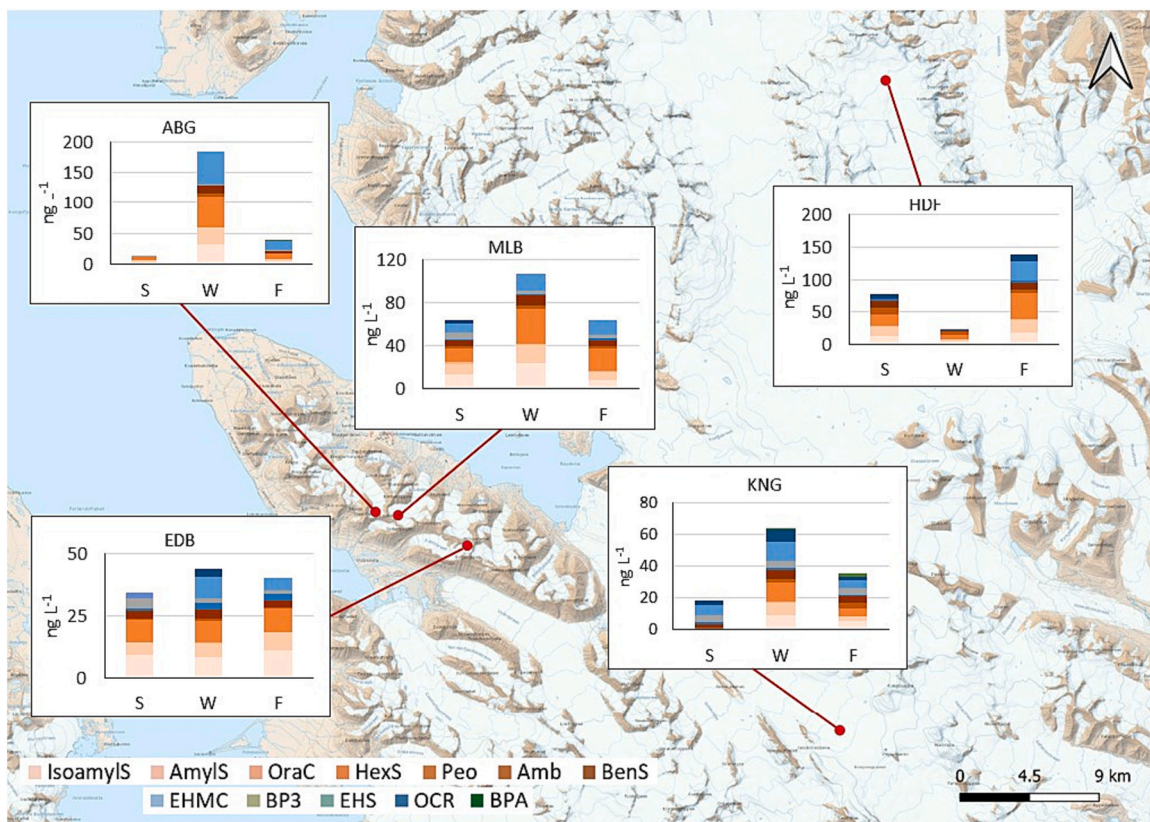


Fig. 4. Seasonal distribution of cumulative concentrations of target contaminants in the dissolved phase of snow collected in snow pits at the top of each glacier. Spring – S; Winter – W; Fall – F. BP3 and BPA not displayed in the HDF S sample.

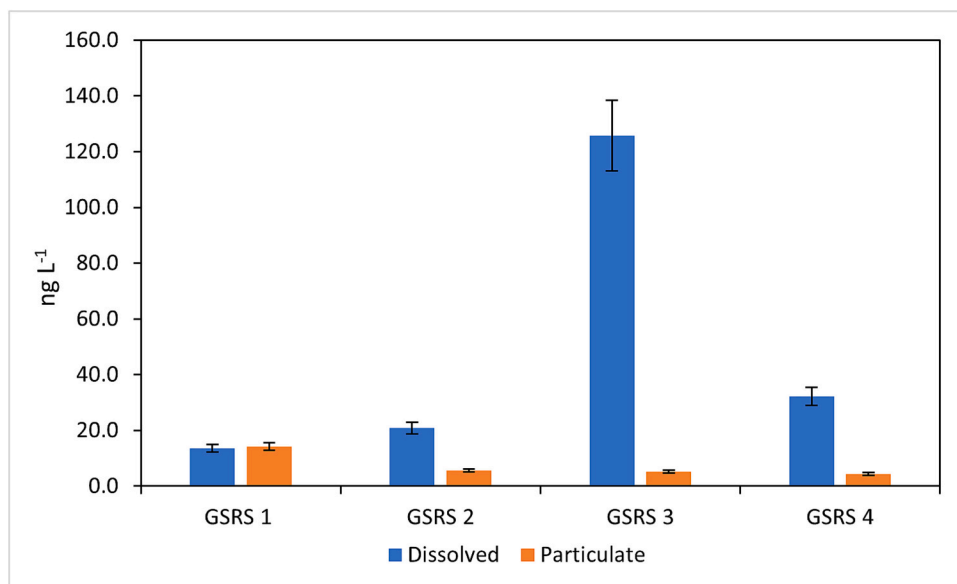


Fig. 5. Concentrations of Σ CEACs in the dissolved and particulate phases in the samples collected at the Grubebadet Snow Research Site (GSRs) between the 12th of April and the 3rd of May 2021.

within the same seasonal horizon without affecting the other two.

3.7. CEACs at the Grubebadet Snow Research Site

The total concentrations (ng L^{-1}) of CEACs (Σ CEACs) collected at the GRS are shown in Fig. 5 separating the two investigated phases. Data on snowpack depth were collected by the automated nivometric

station located next to the sampling site (Scoto et al., 2023) (Fig. S4). Contaminants are mainly found in the dissolved phase rather than in the particulate phase. All the samples are compacted snow except for GRS 2 collected on the 19th of April 2021. During the week between the collection of GRS 1 and GRS 2, no wet depositional event occurred until the same day of the sampling, the increase of concentrations observed in the dissolved phase (GRS 1 = 14 ng L^{-1} ; GRS 2 = 21 ng

L^{-1}) may be due to wet and dry deposition and/or diffusivity. The highest value of \sum CEACs in the dissolved phase was registered on the 27th of April 2021 (GSRS 3 = 126 ng L^{-1}) and it is one order of magnitude higher than those found in the other three samples; on the contrary, the concentrations in the particulate phase remain almost constant in all the samples (Table S6). During the week before the sampling of GSRS 3, the total snow precipitation was 11.5 mm of snow water equivalent with 6.0 mm falling only on the 20th of April (www.yr.no/nb). Concentrations drop one week later (GSRS 4 = 32 ng L^{-1}) concurrently with the decrease of the snowpack thickness, which that week reached the minimum value recorded during the sampling activities at the site. No significant changes in temperature or wind speed were observed during this week so it is reasonable to assume that the thinning of the snowpack is mostly due to post-depositional compaction or erosion by wind.

The salicylates are the compounds with the most abundant concentrations in GSRS 1 (HexS 32 % and IsoamylS 16 %) and GSRS 2 (HexS 36 % and EHS 26 %), but this changes in the two samples collected in the following two weeks. In GSRS 3 and 4 the compound with predominant concentrations is BPA, which reaches 50 % in GSRS 3 and even 87 % in GSRS 4; in both samples the second most abundant chemical is the OCR, followed then by the salicylates. The increase of BPA concentrations in GSRS 3 after a week of important snowfall events suggests that the air masses passing above the area may have been loaded with this compound and that the source may be distant. As mentioned above, in the week between the sampling of GSRS 3 and 4 the snowpack was mainly affected by compaction and the concentrations of all the chemicals decreased possibly due to processes of re-volatilization, degradation, and/or percolation. One possible explanation for the increase in the percentage of BPA concentrations over the other compounds may be related to its low volatility that make BPA more stable within the snowpack, at least within a short time period.

The compounds found at the GSRS are the same found at the glaciers, except for the BHT and the Ambrofix. The first is present in the particulate phase in three of the samples collected at the GSRS (average concentration 0.4 ng L^{-1}), but it is not detected in most of the glaciers except for EDB. These results suggest that the BHT might come from local sources, and it was transported into the atmosphere associated to particles and deposited without reaching the glaciers. Although the proximity of Ny Ålesund, it is unlikely to serve as the primary source of BHT in the area, as we would expect to observe peaks for other compounds. It has been observed that natural BHT production occurs in freshwater phytoplankton (Babu and Wu, 2008). As a result, we can assume that aquatic natural production might be a possible source of BHT, which might be deposited at lower altitudes along the coast. However, the definitive source remains to be determined. Ambrofix, on the other hand, is below the detection limit at the GSRS but it was detected in all the glaciers. Therefore, the village and the air-sea gas exchange do not appear to be major sources of Ambrofix, and it is more plausible that its presence in the glaciers is due to long-range transport.

The salicylates have lower mean concentrations than those in the glacier samples (Table S8). This applies to all salicylates apart from the Benzyl Salicylate which is present only in the GSRS 3 (7 ng L^{-1}) with a concentration equal to the average recorded in the glaciers. The Peonile is the only FM detected in all samples at GSRS (mean concentration 1 ng L^{-1}) and showing concentrations similar to those detected on glaciers (mean concentration 2 ng L^{-1}). Like the Benzyl Salicylate, the EHMC is also present only in GSRS 3 (4 ng L^{-1}), and at a slightly higher concentration than the average detected in the glaciers (2 ng L^{-1}). The situation is different for OCR, which is indeed present in all the samples, but in GSRS 3 (20 ng L^{-1}), the concentration is one order of magnitude higher than in the others (Table S8). For BP3 the concentrations detected at GSRS and on glaciers are similar (mean concentrations 2 and 3 ng L^{-1} respectively). Finally, the BPA was partially discussed above but comparing the GSRS samples with those from glaciers it is observed that the concentrations in GSRS 1 (2 ng L^{-1}) and GSRS 2 (0.5 ng L^{-1}) are

comparable to the mean concentration of BPA on glaciers (2 ng L^{-1}), and they increase of one order of magnitude in GSRS 3 (65 ng L^{-1}) and GSRS 4 (32 ng L^{-1}). However, this is a preliminary study and more sampling points could be useful to have a more in-depth discussion of the differences between GSRS and glaciers in future studies.

3.8. Source analysis

The wind rose illustrating the wind direction, frequency, and mean wind speed for the sampling period from 12th April and 3rd May, is shown in the Supplementary Material (Fig. S5). The analysis was conducted using data from the Norwegian Centre for Climate Services (www.seklima.met.no/). The hourly maximum mean wind speed, and the mean direction in degrees registered over the last 10 min before the observation time were used. The range of the wind speed is between 2 and 8 m/s. It appears that the local winds predominantly moved from SE with a mean speed of 4 m/s, as also described by Vecchiato et al. (2018) in the same area. Winds from N show low frequency, and this suggests that, during the sampling period, GSRS is not affected by wind blowing from the village.

To have a clearer idea on the possible long-range sources of contamination, the NOAA-HYSPLIT model (Stein et al., 2015) was used to study the pathways of the air masses reaching GSRS. The back trajectories show that air masses originated in the Arctic region for most of the sampling dates. However, during the 8 days before the sampling of GSRS 3 the air masses passed above the anthropized areas of western Europe, and so the found chemicals might have been emitted into the atmosphere in these highly populated areas and were transported long-range to the Arctic (Fig. 6). This might explain the higher concentrations found in GSRS 3.

4. Conclusions

These results obtained from this study reveal that CEACs are present in the snowpack near Ny-Ålesund and in glaciers up to 40 km away from the village. For some chemicals investigated, this is the first report of their occurrence in snow samples in the Arctic environment. The implications are crucial if we consider that the annual snow cover is a temporary reservoir for chemicals, and they are re-emitted into the environment in a relatively short time during the melting season.

In line with a previous study (Vecchiato et al., 2018), Ny-Ålesund can represent a source of local contamination, however, we observed that its presence does not predominantly influence the pollution in the specific areas covered by this study. Sources might also have distant origins for most of the compounds found in the selected glaciers and at the GSRS. The empirical results obtained suggest that the target compounds might have the potential to remain in the atmosphere long enough to be transported over long distances and that their physical-chemical properties allow snowfall to remove them and to remain into the snowpack for at least one snow season. This is particularly evident for the UVFs, which are locally used only during the polar day (in late spring and summer), but that show higher concentrations during winter, in most of the cases, except for the Kongsvegen glacier.

It was also observed that the target compounds have a different distribution in the particulate and dissolved phases, this can affect their transport and deposition processes. The salicylates show a similar behaviour among each other suggesting a common source and/or similar environmental transport and deposition pathways. Considering all these aspects, there is a growing need for more detailed information about the physical-chemical properties of CEACs. Further research on this topic is needed to better understand both their behaviour in the atmosphere and in the snowpack, as well as the fate of PCPs in polar environments.

Moreover, the atmospheric transport of chemicals is related to local seasonal conditions (Burkow and Kallenborn, 2000). In the perspective of ongoing climate change, the transport and storage of chemicals will

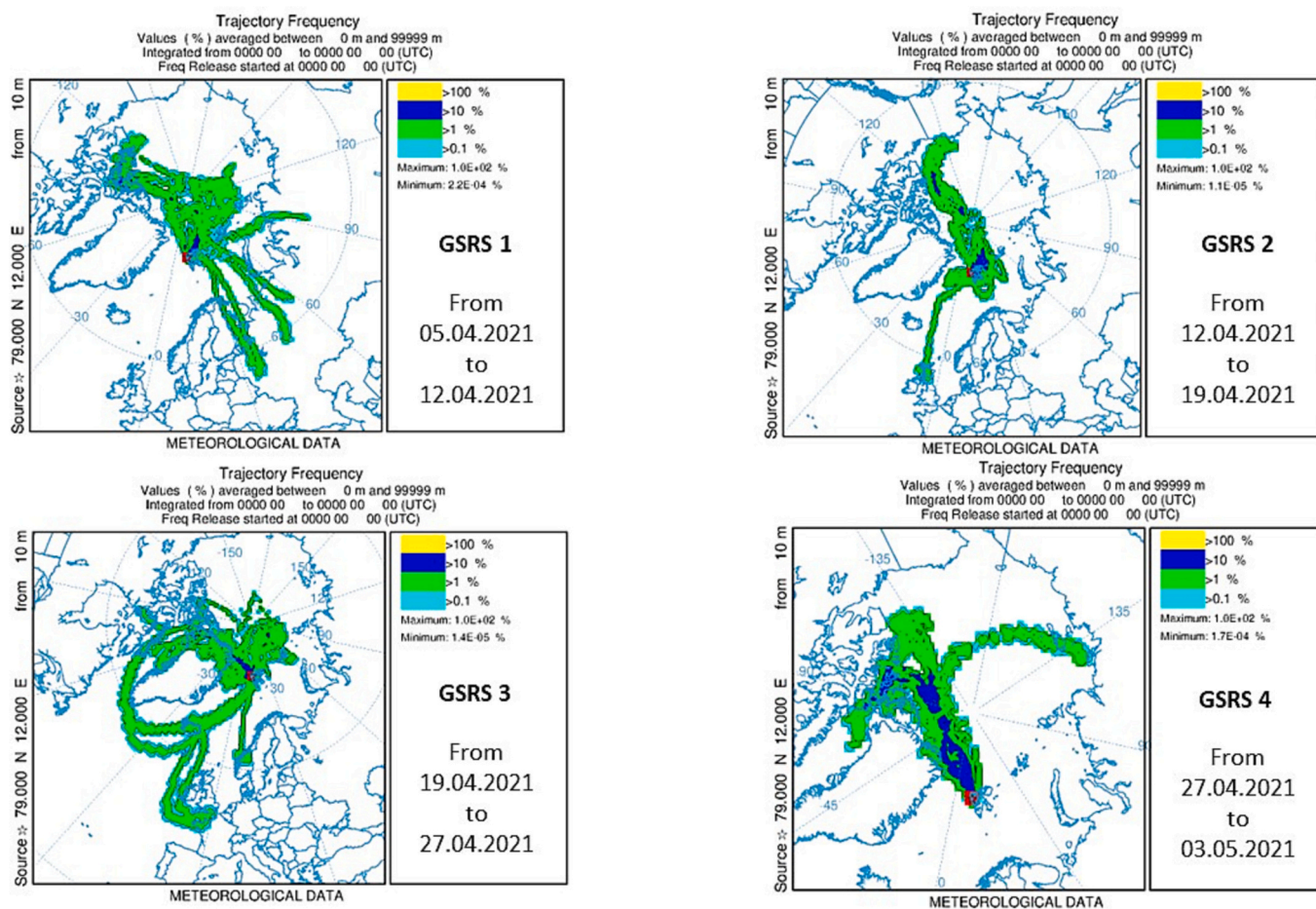


Fig. 6. Back trajectories frequency data simulated with Hysplit 5.0. for each surface snow sample collected at the Gruevbadet Snow Research Site (GSRS).

also change accordingly. Thus, future research should consider these aspects including key processes involved in the release of these chemicals during the melting season.

Even if the concentrations of target chemicals in the seasonal snowpack are below potentially environmentally toxic levels, they could have a significant impact on those recorded in fresh and marine waters as a direct result of the snowmelt process in the area. Monitoring these chemicals in the snow alongside other matrices can hence help to discern and evaluate the impact of the different sources of contamination on the Arctic ecosystem.

CRediT authorship contribution statement

Marianna D'Amico: Investigation, Formal analysis, Writing – original draft, Visualization. **Roland Kallenborn:** Supervision, Project administration, Funding acquisition. **Federico Scoto:** Resources, Writing – review & editing. **Andrea Gambaro:** Resources, Project administration, Funding acquisition. **Jean Charles Gallet:** Resources. **Andrea Spolaor:** Conceptualization, Resources, Supervision, Funding acquisition. **Marco Vecchiato:** Conceptualization, Methodology, Resources, Writing – review & editing, Supervision, Funding acquisition.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2023.168401>.

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