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Are ingredients of personal care products likely to undergo long-range transport to remote regions?†

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Personal care products (PCPs) contain contaminants of emerging concern. Despite increasing reports of their presence in polar regions, the behavior of PCP ingredients under cold environmental conditions remains poorly understood. Snow collected around Villum Research Station at Station Nord, Greenland, between December 2018 and June 2019 was extracted in a stainless steel clean-room and analyzed for seven fragrance materials, four organic UV-filters and an antioxidant using gas chromatography-tandem mass spectrometry. All twelve target PCPs were detected, with elevated concentrations during two sampling events potentially tied to air mass transport from northern Europe and the northern coasts of Russia. To contextualize the presence of these PCP chemicals in high Arctic snow, we estimated their (i) partitioning properties as a function of temperature, (ii) equilibrium phase distribution and dominant deposition processes in the atmosphere at temperatures above and below freezing, and (iii) potential for long-range environmental transport (LRET). Even though most PCPs are deemed to be gas phase chemicals predominantly deposited as vapors, rapid atmospheric degradation is expected to limit their LRET. On the other hand, the less volatile octocrylene is expected to be sorbed to atmospheric particles, removed *via* wet and dry particle deposition, and possibly exhibit a higher potential for LRET by being protected from attack by photooxidants. The contrast between consistent detection of PCP chemicals in high Arctic snow and relatively low estimated LRET potential emphasizes the need for further research on their real-world atmospheric behavior under cold conditions.

Environmental significance

With improved analytical techniques and a more diverse list of target substances, the number of organic contaminants detected in samples taken from remote polar regions is steadily increasing. In some instances, the detection of these substances stands in marked contrast to theoretical expectations, manifested in predictions of a limited potential for long-range environmental transport (LRET) made with simple assessment models. This raises challenging questions related to (i) the interpretation of the detection in remote regions as evidence of the potential for LRET and (ii) the reliability of current LRET assessment models. While the weight-of-evidence compiled here indicates that the presence of PCP in snow collected in Northern Greenland is due to atmospheric transport from lower latitudes, it is questionable whether that can be interpreted as invalidating current LRET assessment models. High production volumes and environmental release rates, when combined with efficient air mass transport and low analytical detection limits, could explain detection, even if the fraction of the released amount undergoing LRET is very small.

1. Introduction

In recent decades, more and more organic contaminants have been detected in remote regions worldwide,[1–7] highlighting that the impact of human activities extends beyond mid-latitudes, affecting even seemingly pristine polar areas. Contaminants are often found in remote environments either because they were released locally or transported there *via* air or water.[8–10] Determining whether their presence in these regions results from long-range environmental transport (LRET) holds regulatory significance. This is particularly relevant to the Stockholm Convention on Persistent Organic Pollutants (POPs), adopted in 2001 and in force since 2004, which stipulates that global action

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is warranted if a chemical is "likely, as a result of its long-range environmental transport, to lead to significant adverse human health and/or environmental effects."[11] LRET, occurring *via* air, water, or migratory species, is a key screening criterion for determining whether an organic compound is eligible for nomination for inclusion in the convention.

To have the potential for adverse effects as a result of LRET, a chemical must (i) be released in significant quantities into air or water, (ii) be transported over large distances, and (iii) accumulate in exposure-relevant media in remote regions.[10] LRET is quantified using different metrics: transport-oriented LRET metrics only consider process (ii), whereas target-oriented metrics take process (iii) into account.[12] Deposition from the atmosphere occurs *via* wet or dry processes. In cold remote regions, snowfall can scavenge organic compounds either in the vapor phase or while sorbed to particles thereby transforming the snowpack and glacier ice into temporary reservoirs.[13,14] The efficiency of snow deposition depends on a compound's physicochemical properties, and, for the particle bound compound, on the particle size as well as snow characteristics.[13,15]

In recent years, in addition to legacy contaminants, the attention of environmental chemists has increasingly turned to chemicals of emerging concern, including those found in personal care products (PCPs).[16–18] This has been prompted by the risk these compounds may pose to both human health and the environment.[19–24] While for the most part PCPs are unregulated internationally, in a few jurisdictions, such as Hawaii, the U.S. Virgin Island and Palau, the use of some organic UV-filters (*e.g.*, benzophenone-3 (BP3), octocrylene (OCR), ethylhexyl methoxycinnamate (EHMC)), has been banned. They were found to be phototoxic and harmful to local marine fauna and ecosystems.[24,25]

Most studies on the occurrence of PCPs in the environment have focused on temperate zones in mid-latitudes,[16,26,27] where PCPs ingredients are frequently found near densely populated areas, consistent with such areas being the primary location of their environmental release.[16,28–31] Specifically, PCPs can enter the environment during their production, use, or disposal.[27,30,32] However, PCPs have also been detected in different biotic and abiotic environmental matrices in polar regions.[33–38] Specifically, the presence and distribution of PCPs in Antarctic and Arctic snow[39–41] suggests the possibility for LRET of PCPs. The presence of PCPs in remote, cold regions prompts several key questions: what are the mechanisms by which these compounds reach such areas? How do they behave under cold temperature conditions? What are the primary processes governing their transport and deposition? In response, the Arctic Monitoring and Assessment Programme (AMAP), and the Scientific Committee on Antarctic Research (SCAR) have encouraged further research on the topic of PCPs in polar regions and the implementation of monitoring plans.[42,43] However, investigations into the physicochemical properties of these compounds at different temperatures and on their potential for LRET have been limited.

This study employed a multidisciplinary approach, combing field measurements and modelling to increase the understanding of environmental transport and deposition of twelve PCPs (7 fragrance chemicals, 4 UV-filters, 1 antioxidant) in cold remote environments. The fieldwork focused on investigating the temporal variation of these PCPs in surface snow at Villum Research Station at Station Nord in Northeastern Greenland. The modelling study involved estimating key chemical properties of the PCPs and their temperature dependence, determining their equilibrium phase distribution in the atmosphere, identifying dominant deposition processes at temperatures above and below freezing, and assessing their LRET potential employing a modified version of the OECD tool.[44,45]

2. Material and methods

2.1 Sampling location

Surface snow was collected at Villum Research Station (VRS) in Greenland. Operating all year round, VRS accommodates up to 14 researchers. It is situated at the military outpost Station Nord, staffed by 5 soldiers, on Princess Ingeborgs Peninsula in north-east Greenland (81.6014°N, −16.6620°W; 24 m a.s.l.), approximately 800 km from the nearest human settlement (Fig. 1). The area is particularly dry and cold, with an average annual temperature of −15.1 °C, an annual average precipitation of ~328.7 mm, and year-round presence of sea ice.[46] Prevailing winds originate from the SSW, maintaining an average speed of 4 m s^{-1}. Contaminants emitted from Eurasia and North America reach Greenland *via* atmospheric LRET, affecting the northern and eastern coastlines.[47] Anthropogenic contaminants at Station Nord have been shown to originate primarily in Eurasia.[4,6,47–52]

2.2 Sampling procedure

Surface snow samples of the top 20 cm of the snowpack were collected every two weeks at a location about 2 km upwind (*i.e.* with respect to the main wind direction of SSW) from VRS[54] between 17th December 2018 and 6th June 2019, for a total of eleven sampling events. The reproducibility of the sampling procedures and of the analyses was evaluated by collecting and analyzing duplicate samples (with one exception). The collection procedure involved metal buckets for snow sampling, subsequent melting under ambient air conditions at VRS. For each sample, the resulting meltwater was collected in two duplicates in separate glass jars. Each jar was covered with a double layer of aluminum foil and closed with a screw cap. Buckets and jars had been pre-cleaned with MilliQ water, acetone, methanol, and *n*-hexane. All solvents used were pesticide-grade (Romil Ltd., Cambridge, UK). The percentage difference of the PCP concentrations between duplicated surface snow samples (Table S6†) was on averaged 12% and therefore generally comparable to the analytical uncertainty of the analytical method.[34]

Daily average temperature and snow depth were calculated using five-minute average data measured *in situ* at VRS from December 2018 and June 2019 (Fig. S1†). Temperatures never went above zero degrees during the entire sampling period; average, minimum and maximum temperatures are shown in Fig. S1.†

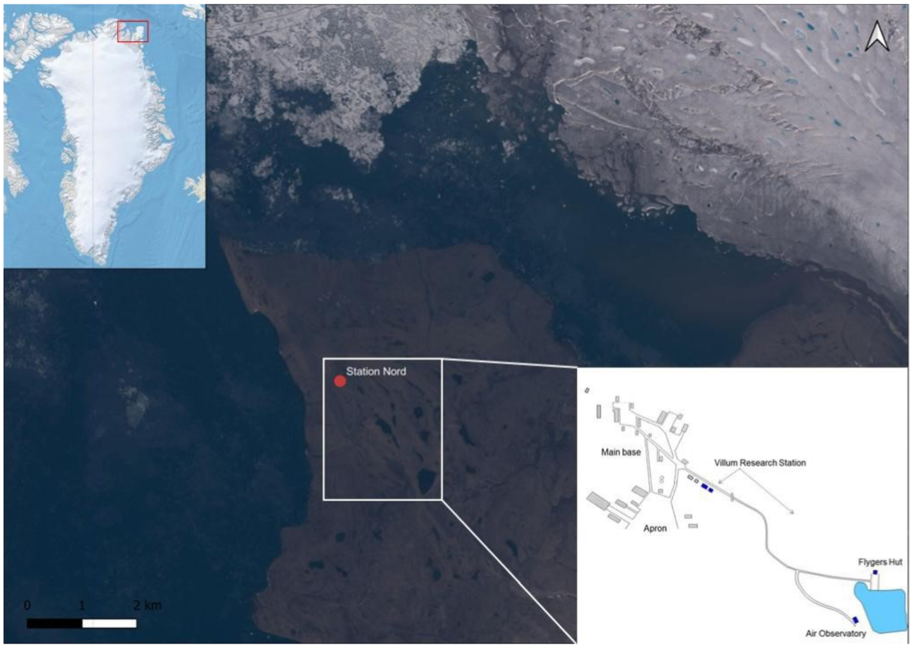

Fig. 1 Map of Villum Research Station with its buildings (blue) relative to Station Nord military outpost. Flygers hut and air observatory are located about 2 km outside the main base of Station Nord. The snow samples were taken about 300 meters south of the air observatory (figure created with QGreenland (v2) [software], National Snow and Ice Data Center. Fisher *et al.* (2022) https://doi.org/10.5281/zenodo.6368651 and using the map from Skov *et al.* (2020)).[53]

2.3 Sample extraction

Samples in glass jars were stored and kept frozen at −20 °C until and during transport to Europe and remained frozen until their preparation in the stainless steel clean-room laboratories for organic analyses (ISO 7) at Ca' Foscari University in Venice. In contrast to previous work[34] the meltwater in this study was not filtered to separate dissolved and particulate fractions. Samples were left thawing at ambient temperature inside the clean-room and extracted immediately after thawing. Solid phase extraction (SPE, Waters Oasis® HLB cartridges, 200 mg, 6 cc, 30 μm particle size, Waters Corp., Milford, MA USA) of melt water samples used cartridges conditioned with 10 mL of *n*-hexane, 10 mL of dichloromethane, and 10 mL of ultrapure water (ELGA, High Wycombe, UK). After spiking 0.5 ng phenanthrene-^{13}C and 140 ng octocrylene-d^{10} onto the cartridges as internal standards, they were eluted using separately 1 mL of toluene, 15 mL of dichloromethane, and 10 mL of *n*-hexane. Laboratory personnel did not use PCPs before entering the laboratory. Eluates were dried with Na_2SO_4 and concentrated to 250 μL under nitrogen flow at 23 °C (Turbovap II®, Caliper Life Science, Hopkinton, MA, USA). Laboratory blanks ($n = 3$) consisted of 1 L of ultrapure water (ELGA, High Wycombe, UK) that was extracted in the clean-room laboratories using the same procedure as was used for meltwater 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 (Table S3†). The recoveries of the analytical method for the selected analytes are above 90%, as reported in D'Amico *et al.*[34]

2.4 Instrumental analysis

Twelve PCPs (Table S1†) including seven fragrance materials (FMs; amyl salicylate (AmylS) and isoamyl salicylate (IsoamylS), hexyl salicylate (HexS), benzyl salicylate (BenS), oranger crystals (OraC), peonile (Peo), and ambrofix (Amb)), four organic UV-filters (UVFs; ethylhexyl methoxycinnamate (EHMC), benzophenone-3 (BP3), ethylhexyl salicylate (EHS), octocrylene (OCR)) and the antioxidant butylated hydroxytoluene (BHT) were quantified in the extracts using gas chromatography tandem mass spectrometry (GC-MS/MS, Trace 1310 – TSQ 9000 Thermo Fisher). The GC separation was performed on a 60 m HP-5MS (0.25 mm I.D., 0.25 μm; Agilent Technologies, Avondale, USA). Starting at 120 °C (maintained for 1.0 min), the GC oven temperature increased to 180 °C at 25 °C min^{-1}, then to 250 °C at 10 °C min^{-1}, and finally at 20 °C min^{-1} to 310 °C, which was held for 8.6 min. Details on the performance of the analytical method are reported by D'Amico *et al.*[34]

2.5 Back trajectory analysis

To determine the origin and pathways of air masses arriving at the sampling location near VRS, we calculated five-days back trajectories using the HYSPLIT transport and dispersion model of NOAA[55] and meteorological data from the NCEP/NCAR reanalysis. The mid-boundary layer height was selected for the calculations, setting the trajectory's starting height to the middle of the dynamically calculated atmospheric boundary layer. For each sampling date, back trajectories were calculated every six hours over the preceding five days, yielding a total of 220 individual back trajectories.

2.6 Estimating the properties of PCP ingredients

Experimental data on the equilibrium partitioning ratios between air, water and n-octanol (K_{OW}, $K_{AW,}$ and K_{OA}) for the twelve PCPs are scarce, especially as a function of temperature. To address this, we estimated consensus values by averaging the predictions obtained from different methods:[56] QSARs implemented in EPI Suite,[57] OPERA,[58] SPARC,[59] COSMOtherm,[60] and poly-parameter linear free energy relationships (ppLFERs). For the ppLFERs we used solute descriptors predicted with the IFSQSARs in EAS-E suite[61] unless experimental solute descriptors were available in the UFZ LSER database.[62] Partitioning ratios at different temperatures (Table S4†) were either directly estimated at 25 °C, 0 °C, and −6.8 °C using COSMOtherm, or the predicted values at 25 °C were adjusted using the van't Hoff equation along with internal energies of phase transfer, that were themselves predicted using different methods. Further details are provided in the ESI Section 1.† Degradation half-lives in water and soil were estimated with OPERA.[58] The gas phase reaction rate with OH radicals was obtained as the average of predictions made with OPERA version 2.9.1 (ref. 58) and AOPWIN version 1.92.[57]

2.7 Estimating atmospheric phase distribution of PCP ingredients

The atmospheric fate of PCPs, based on their equilibrium distribution between the gas phase, organic particles, and liquid or frozen hydrometeors at 25 °C and −25 °C, was predicted using chemical partitioning space plots. These plots use the particle-air distribution ratio ($K_{particle/air}$) on the x-axis and either the rain-air ($K_{rain/air}$) or snow-air distribution ratio ($K_{snow/air}$) on the y-axis.[13] $K_{particle/air}$, $K_{rain/air}$ and $K_{snow/air}$ were estimated from K_{OW}, $K_{AW,}$ and K_{OA} using equations and default parameters given in Table S5.† These plots facilitate the identification of a compound's predominant atmospheric deposition processes both above and below the freezing point: chemicals predominantly partitioning to atmospheric particles are subject to dry and wet particle deposition, whereas those in the gas phase undergo dry gaseous deposition or react with OH radicals. Lastly, wet gaseous deposition is linked to either dissolution in liquid cloud water or adsorption to the surface of snow and ice crystals.[13] Chemicals that are partitioning to more than one phase can be subject to more than one deposition process and their atmospheric fate becomes more sensitive to variability in parameters such as temperature, particle concentration and composition and cloud liquid water content.

2.8 Estimating the potential of PCP ingredients for LRET

The potential for LRET of the selected PCPs, upon their release into air, water, and soil, was estimated using the emissions fractions approach (EFA) as implemented in a modified version of the OECD P_{ov} & LRTP screening tool (referred to as "the tool").[44,63] The tool incorporates a steady-state, fugacity-based multimedia mass balance model explicitly developed to facilitate screening of organic chemicals with respect to their potential for LRET and persistence, and to therefore support decision-making in the context of preparing nominations to the Stockholm convention.[45] The EFA version of the tool calculates three LRET metrics: the fractions of the total amount of a chemical emitted into the environment of a source region that is dispersed (φ_1), transferred (φ_2), or accumulated (φ_3) in a remote region.[44] Each metric represents a fraction of the preceding one and can be calculated using three different emission scenarios: 100% emissions to air, water, or soil. Movement between source and remote regions can occur *via* water and air. Whereas the parameterization of atmospheric deposition processes in the model traces its origin to the original version of the tool,[45] the modified version as described in Breivik *et al.*[44] includes intermittent precipitation. By representing precipitation as occurring episodically and not as a constant drizzle, we avoid underestimating atmospheric residence time and travel distance of particle-bound chemicals and chemicals with low Henry's law constant during dry periods.[44,64,65] More details about the EFA metrics can be found in Breivik *et al.*[44,63] The version of the tool we used allows the remote region to have either the same temperature (25 °C) as, or a lower temperature (0 °C) than, the source region.

The input parameters required by the tool are: molar mass; log K_{AW} and log K_{OW} (Table S8†) along with their temperature dependence as expressed through the internal energies ΔU_{OA} and ΔU_{OW}; half-lives in water and soil; the reference rate constant for reaction in air (k_{RAref}); and the activation energies for degradation in air, water and soil (E_{aAir}, E_{aWater}, E_{aSoil}). The values used for the 12 PCPs are listed in Table S10.† Further details about the specific equations employed by the model are available in ESI Section 2.†

3. Results and discussion

3.1 Personal care products in snow at Villum Research Station

The twelve PCPs were detected in at least one sample during the sampling period (Fig. 2 and Table S7†), with the salicylates and OCR being the most abundant in terms of concentrations (Fig. 3). The concentrations (in the text hereafter reported as the arithmetic mean concentration of PCP chemicals in duplicate snow samples in ng L^{-1}) fluctuate across the eleven sampling events, with snow collected on April 22nd and May 21st exhibiting levels one or even two orders of magnitude higher than observed at other times (Fig. 2). Specifically, IsoamylS and AmylS (184 ng L^{-1} and 120 ng L^{-1}, respectively), as well as HexS (188 ng L^{-1}) reached their peak concentration on May 21st, and OCR peaked on April 22nd (167 ng L^{-1}). These compounds are the only ones exceeding 100 ng L^{-1} during the sampling period (Fig. 3).

The occurrence of some of the detected PCPs in polar regions has been previously reported. For instance, FMs and UVFs have been detected in snow samples from Svalbard,[34] with concentration ranges (FMs 1.5–147 ng L^{-1}; UVFs 1.2–369 ng L^{-1}) comparable to those reported here (Table S7†). An exception is BHT, which exhibited a concentration range (0.2–1.4 ng L^{-1}) one order of magnitude lower than the peak concentration observed at VRS (12.4 ng L^{-1}). In seawater collected near Ny-Ålesund, Svalbard, FMs concentrations (below detection limit to 5.8 ng L^{-1}) were two orders of magnitude lower than the

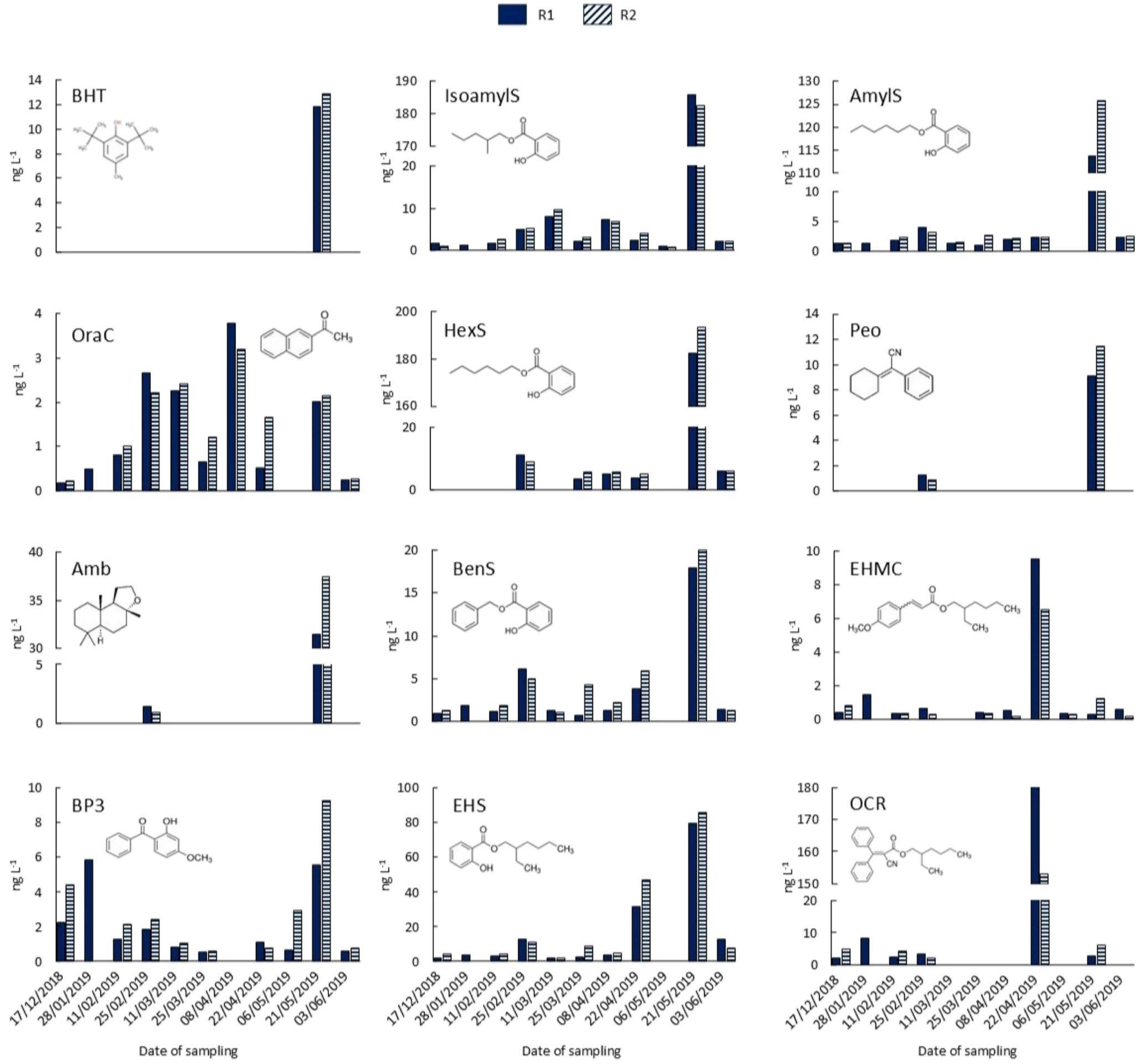

Fig. 2 The concentrations of selected PCP chemicals (ng L^{-1}) in the duplicated snow samples (R1 and R2) collected at VRS between mid-December 2018 and the beginning of June 2019.

maximum levels found here.[40] Similarly, FMs concentrations in Antarctic seawater[37] were one order of magnitude lower than those reported here. Furthermore, the UVF EHMC has been identified in snow from Terra Nova Bay at concentrations ranging from 0.4 ng L^{-1} to 3.1 ng L^{-1} (ref. 39) and in wastewater treatment plant effluents from the Mario Zucchelli Station, where concentrations reached the μg L^{-1} range.[66]

3.2 Atmospheric transport and local contributions to PCPs in polar regions

The origin of PCP chemicals in polar environments remains not fully understood. Previous studies suggested that they reach polar regions through atmospheric LRET.[37,39,40] To test this hypothesis, it is fundamental to understand the atmospheric behavior of these compounds and identify the mechanisms governing their deposition.

Research conducted at mid-latitudes indicates that UVFs emissions vary seasonally, with peak concentrations occurring in the summer. For example, elevated air concentrations of EHS, BP3, EHMC, and OCR were reported in Toronto, Canada,[30] while seawater concentrations of EHS and EHMC along the Japanese coastline also peaked during summer.[67] This seasonal increase is likely driven by greater sunscreen use and recreational activities at coastal areas.[67]

In the Arctic, aerosol transport and deposition are influenced by seasonal variations in source regions, meteorological conditions, and pathways. During winter and spring, LRET from sources in mid-latitudes contribute significantly, while

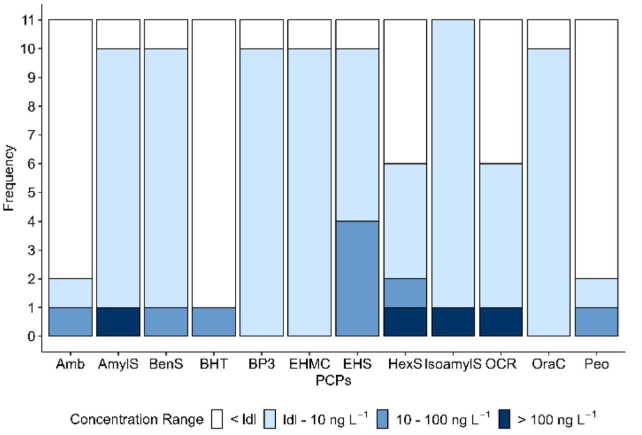

Fig. 3 Frequency distribution of concentration ranges for each PCP chemical: below the detection limit (<ldl); ldl–10 ng L^{-1}, 10–100 ng L^{-1}, and >100 ng L^{-1}. Concentrations refer to the average of duplicates samples. The bar heights indicate the total number of samples, while the gradient of blue shades represents increasing concentration levels from low to high.

local sources, including sea ice and open ocean regions, tend to contribute more in summer.[52,68,69] The transport of particles from lower latitudes, combined with limited removal through wet deposition, suggests that distant sources and dry deposition may influence the concentrations of chemicals at VRS.[70–72] At VRS, aerosol removal *via* wet deposition is lowest during mid-winter but increases in spring, coinciding with the onset of Arctic haze.[51,73]

The five-day back trajectories calculated for the sampling dates (Fig. S2†) show that during most of the sampling period, air masses over Northern Greenland originated within the Arctic (Eurasia and North America). However, between April 17th and 22nd, 2019, air masses originated from regions south of the Arctic circle, consistent with reports that the Arctic region experienced an intense episode of warm and humid air intrusion from mid-latitudes in April 2019.[74] Snow sampled on April 22nd had elevated concentrations of OCR (167 ng L^{-1}), EHS (39 ng L^{-1}), and EHMC (8 ng L^{-1}). For OCR and EHMC these are the highest concentrations of the whole sampling period, suggesting that LRET from mid-latitudes likely contributed to these elevated levels. A recent study also attributed the occurrence of EHMC in Antarctic snow to LRET from distant sources.[39] Back trajectories calculated for the time prior to the May 21st sampling date indicate that air masses originated from the Arctic Ocean, Greenland and the northwestern coast of Russia (Fig. S2†). Snow sampled on that day had high concentrations of salicylates (HexS = 188 ng L^{-1}, IsoamylS = 184 ng L^{-1}, AmylS = 120 ng L^{-1}).

Local wind patterns provide additional insights into potential sources. Throughout the study period (December to June), prevailing winds at the sampling location were primarily from the south-west (SW), suggesting that the research station, located north-west of the sampling site, is unlikely to be the main source of contamination. Wind rose analyses focused on the 2 weeks before the two most contaminated sampling dates further support this conclusion (Fig. S3†). Prior to April 22nd, winds blew mostly from SW, with some contributions from the W, aligning with the hypothesis of LRET. In contrast, prior to May 21st, winds mainly blew from E and SW.

No precipitation data are available for VRS between April and May 2019. To indirectly identify precipitation events, snow depth data (Fig. S1†) collected at five-minute intervals from December 2018 to June 2019 were analyzed. A significant threshold indicative of a possible precipitation event was defined as an increase in snow depth of more than 5 cm within 24 hours. In the night from May 14th to 15th, 2019, one week before the contaminated snow was sampled on May 21st, a substantial increase of about 6 cm was recorded. Meteorological data (*e.g.*, wind speed, temperature) were analyzed to rule out alternative explanations for the change in snow depth, such as wind redistribution or sublimation masking precipitation signals. Since temperatures never exceeded 0 °C during the study period (Fig. S1†), sublimation and melting effects are negligible. Increased snow depth during conditions with strong winds >7.5 m s^{-1} might indicate drifting snow.[75] As the maximum wind speed recorded during the 24 hours preceding the observed snow depth increase on May 14th, 2019, was 6.4 m s^{-1} (Fig. S4†), we conclude that this 6 cm increase can be attributed to a precipitation event. Back trajectories calculated for the date of this precipitation event (Fig. S5†) show air mass origin within the Arctic.

The combined analyses of back trajectories, wind roses, and snow depth suggest that LRET from mid-latitudes is a possible contributor to the elevated PCP levels observed in spring. Although local sources cannot be entirely excluded, particularly for certain salicylates, the results indicate that distant sources and atmospheric LRET play a predominant role in explaining the presence of PCP chemicals at VRS.

3.3 The partitioning properties of PCPs

The partitioning properties estimated for the 12 PCP chemicals are shown in Tables S8 and S9.† OPERA predictions were excluded from the final averages as they represented outliers. This exclusion resulted in a more consistent adherence of the three partitioning ratios to thermodynamic constraints ($K_{OW} = K_{OA} \times K_{AW}$), showing an average misclosure error of only 0.04 log units (Table S8†).

Several chemicals, namely BHT, IsoamylS, AmylS, HexS, Peo, and EHS, exhibit log K_{AW} values around −3 and log K_{OA} values between 7.5 and 9, indicating similar partitioning properties (Table S8†). In aquatic environments, most of these compounds, with log K_{OW} values averaging ∼4.5 and ranging from *ca.* 3 to 6, could either be truly dissolved or be bound to particles, depending on the concentration of organic matter in the water. Among the fragrance materials, Amb displays significant hydrophobicity (log $K_{OW} = 5.2$) and volatility (log $K_{OA} = 7.46$). Its residence time in the atmosphere will primarily be affected by degradation reactions. However, due to its intended purpose as a fragrance compound (Table S2†), it needs to be at least somewhat resistant to atmospheric degradation suggesting a potential for atmospheric LRET. OraC and BenS have a log $K_{AW} < -4$. EHMC and BP3 are less volatile compared to the other

PCPs. The $\log K_{OA}$ value of 10.2 estimated for EHMC here is in reasonable agreement with values for the two EHMC isomers ($\log K_{OA} = 9.64$ for E-EHMC, and $\log K_{OA} = 9.02$ for Z-EHMC) obtained with a gas chromatographic retention time methods.[76] OCR exhibits low volatility ($\log K_{OA} > 13$), and with a $\log K_{OW} > 7$ it is expected to be particle-bound in water.

3.4 The phase distribution of PCPs in a cloud and dominant atmospheric deposition processes

To gain deeper insights into the atmospheric behavior of the 12 PCP chemicals at different temperatures, their partition ratios were superimposed onto atmospheric equilibrium partitioning spaces constructed based on the framework by Lei and Wania.[13] Fig. 4A illustrates the phase distribution in a warm cloud as a function of $K_{rain/air}$ and $K_{particle/air}$, while Fig. 4B shows the phase distribution in a cold cloud, using $K_{snow/air}$ and $K_{particle/air}$ as coordinates. The phase distributions of the 12 PCP chemicals are represented by lines, which illustrate their behavior across temperature ranges of 0 to 25 °C in warm clouds and between −25 and 0 °C in cold clouds. In warm clouds, most PCP chemicals, particularly the FMs, are expected to primarily partition into the atmospheric vapor phase (Fig. 4A). OCR is the only compound that tends to be sorbed to particles, especially at temperatures close to the freezing point, while at warmer temperatures even OCR starts to partition notably into the vapor phase. In cold clouds (Fig. 4B), FMs, BHT and EHS remain in the vapor phase, while UVFs other than EHS start to partition to particles. At temperatures close to −25 °C, as much as half of

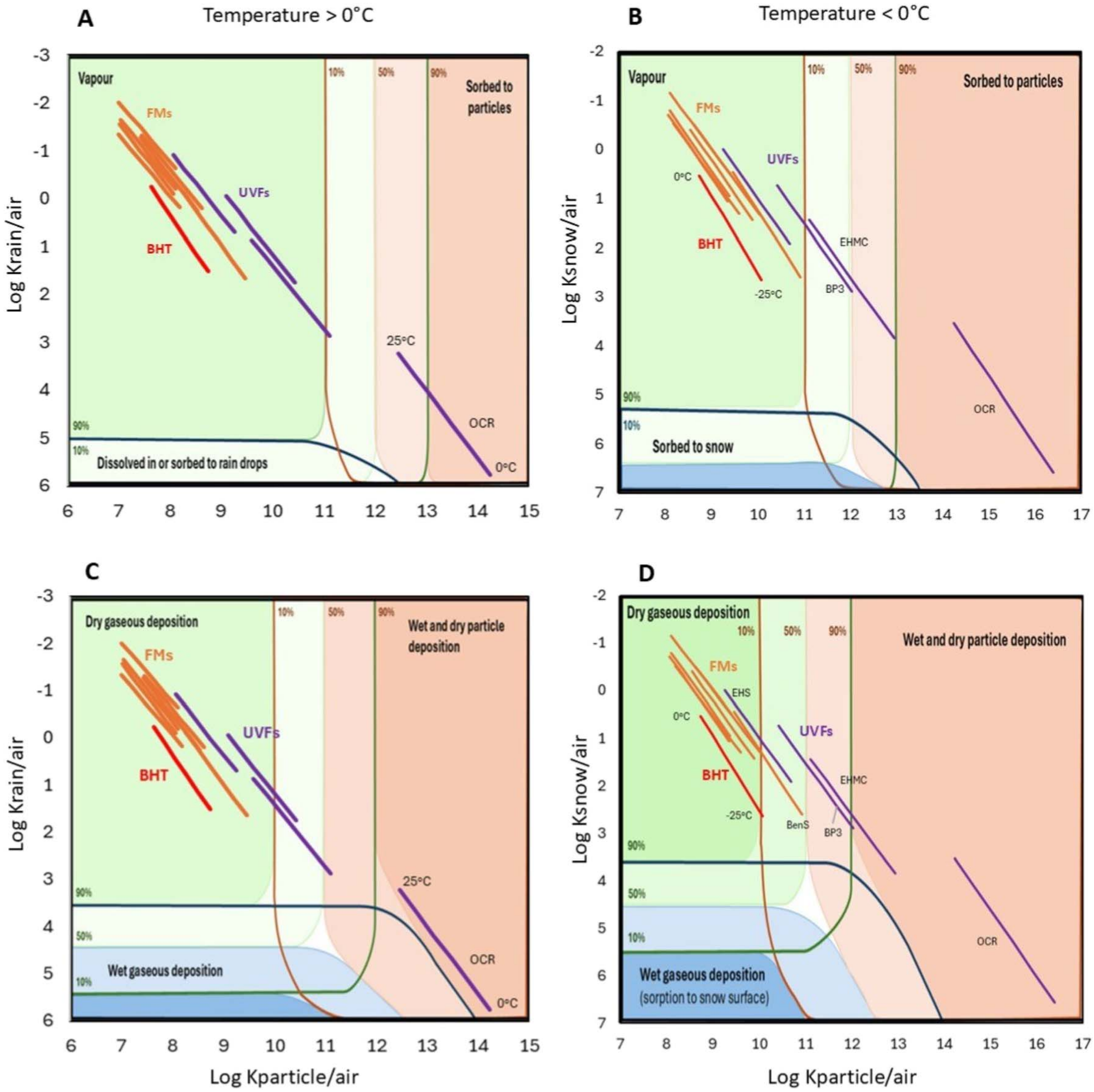

Fig. 4 Equilibrium phase distribution of the 12 PCPs in a warm (temperature range 0 to 25 °C) (A) and a cold (temperature range 0 to −25 °C) cloud (B), and the dominant atmospheric deposition processes at temperatures above (C) and below (D) 0 °C. They are a function of the particle-air, rain-air, and snow-air distribution ratios. The green areas represent dominant gas phase distribution (A and B), and dry gaseous deposition (C and D). Red areas designate particle sorption (A and B), and wet and dry deposition (C and D). Finally, blue areas show liquid water (A) or snow (B) distribution, and wet gaseous deposition (C and D).

BP3 is expected to sorb to particles. The gas/particle partitioning of EHMC, located in the transition region, is particularly sensitive to temperature, with ~10% and ~90% sorbed onto particles at 0 and −25 °C, respectively. OCR is always sorbed to particles at sub-zero temperatures.

The phase distribution of PCP in the atmosphere determines their dominant deposition mechanism. Chemicals in the vapor phase are removed by dry gaseous deposition, those sorbed to particles are subject to both wet and dry particle deposition, and chemicals dissolved in raindrops or sorbed to the snow surface undergo wet gaseous deposition.[13] Fig. 4 illustrates the dominant deposition processes for temperatures above (Fig. 4C) and below the freezing point (Fig. 4D). The parameters used in the construction of the boundaries in Fig. 4C and D are the same as used by Lei and Wania.[13] The deposition of BHT and of the FMs other than BenS is dominated by dry gaseous deposition at any temperature between −25 and 25 °C. In contrast, OCR is solely subject to wet and dry particle deposition across the entire simulated temperature range. For the UVFs EHMC and BP3 particle-associated deposition processes only become relevant when temperatures drop below 10 to 15 °C and are dominant at subzero temperatures. At higher temperatures they are removed only by dry gaseous deposition. Temperatures have to drop below −10 °C for particle-associated deposition processes to become relevant for the larger salicylates, *i.e.* the FM BenS and the UVF EHS.

While FMs exhibit similar behavior as a category, UVFs show more diverse behaviors at different temperatures depending on the specific type of compound. Moreover, many UVFs fall in the transition region being subject to more than one deposition mechanism depending on specific snow area, particle content in the atmosphere, and kinetic parameters such as deposition velocities and precipitation rate.[13] These results imply that each UVFs must be studied individually to fully understand their environmental behavior and impact. The FMs appear to be less sensitive to variations in temperature and as a result, their distribution is more uniform and predictable. For the twelve selected PCP chemicals, there are only two main deposition mechanisms: dry gaseous and wet and dry particle deposition. This implies that, at any temperature, they can deposit even without wet precipitation. While data on the occurrence of the 12 PCPs in precipitation is lacking, our theoretical analysis suggests that if these compounds were to exist within the atmosphere of cold environments, their inherent chemical and physical characteristics should facilitate their deposition and their incorporation in the snowpack. This implies that investigating PCP chemicals within the snowpack of cold regions could indeed serve to understand their presence in the atmosphere and potentially interpret their transport processes.

3.5 Potential for LRET of personal care product chemicals

The EFA approach to LRET assessment, when based on the simple multimedia fate and transport model within the OECD LRT and P_{ov} assessment tool, distinguishes between a source and remote environment. The default assumption is that both these environments are at 25 °C.[44,63] Here we additionally perform calculations with a remote region at a temperature of 0 °C. A temperature gradient between source and remote region could enhance target-oriented LRET by increasing rates of deposition and reducing rates of degradation and re-evaporation in the remote region, a phenomenon described as "polar cold-trapping".[77] While polar temperatures, especially in our study location in Northern Greenland, are well below the freezing point, a remote temperature of 0 °C is used because the model in the tool is not set up to describe snow and ice.

The maximum φ values for the 12 PCP chemicals across different emission modes are shown in Fig. 5 and in Table S11.† Dashed horizontal lines in Fig. 5 represent φ thresholds for potential POP-like behavior. These lines correspond to the minimum φ values calculated for the initial 'dirty dozen' POPs and α-HCH, with thresholds varying for remote environments at 25 and 0 °C (Table S12†). The thresholds are reported for comparison, because these POPs can be assumed to meet the LRET criterion stipulated by the Stockholm convention; as such, these thresholds are not purely scientific but reflect regulatory decisions that might change over time.

The potential for LRET is estimated to be relatively low and similar for most PCP chemicals, irrespective of whether the remote region is warm or cold (Fig. 5). The emission fractions estimated for the PCP chemicals are consistently below the thresholds for POP-like LRET. The one notable exception is the UV-filter OCR, which always has φ values above the thresholds when emitted into air. Its low volatility leads to sorption onto

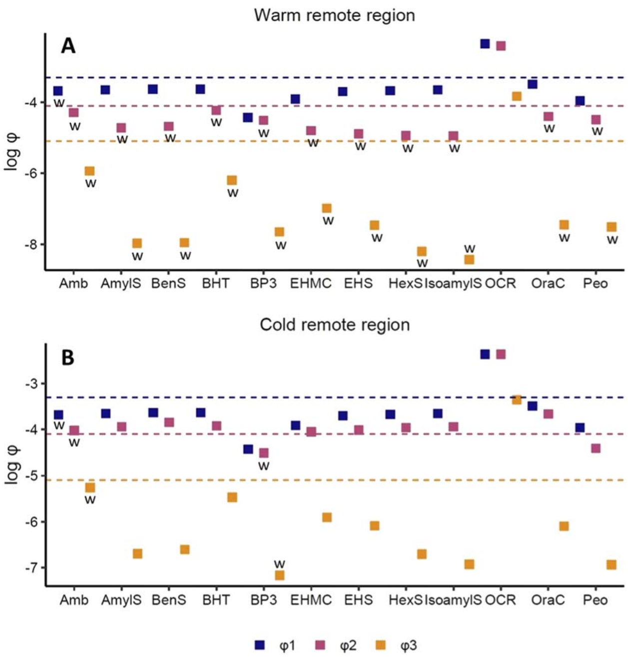

Fig. 5 The fraction of the emitted amount of the selected 12 PCP chemicals estimated to be dispersed (φ_1, blue), transferred to a remote region's surface media (φ_2, purple) and accumulated in those surface media (φ_3, orange), both in a warm (A) (25 °C) and cold (B) remote environment (0 °C). Lines refer to the thresholds derived from established POPs at the same environmental temperatures. A "w" next to a bar indicates that the maximum φ value was obtained when the chemical is emitted to water, as opposed to air.

particles, which is assumed to protect it from reactions with photo-oxidants like the OH radical. Overall, the φ values mostly increase when the temperature of the remote region is lowered (Fig. 5B), consistent with slower degradation rates and increased precipitation scavenging at lower temperatures. However, the thresholds for POP-like behavior similarly increase, so that the position of the PCP relative to these thresholds remains relatively unchanged.

Highest dispersion (φ_1) always occurs when the PCPs are released into air, except for Amb which has the highest φ_1 when emitted to the water. In the warm remote region scenario, the transfer (φ_2) and accumulation (φ_3) of PCP chemicals in surface media are more pronounced when emitted into water (Fig. 5A). In contrast, when the remote region is cold, φ_2 and φ_3 are larger if the PCPs are released into the atmosphere, except for Amb and BP3, whose φ_S are higher when emitted to water. Displaying the φ_S for the emission mode that results in the highest value in Fig. 5 represents a worst-case scenario. While the typical use of PCPs involves their release *via* wastewater, it is important to note that direct air emissions are also possible when the chemicals are present in spray products. Nonetheless, in real-world conditions, the most likely mode of emission is water.

Although the φ_1 and φ_2 values for the PCPs are lower than the thresholds, they do not deviate significantly from them, with a mean difference of 0.62 for φ_2. Conversely, the φ_3 values for the PCPs are notably lower than the threshold, with a mean difference of 2.39. This suggests that, even if the selected PCPs do reach a remote cold region, their accumulation there is likely to be less significant compared to established POPs.

3.6 Possible explanations for PCPs in remote regions

The theoretical assessment indicates that the studied PCPs have limited potential for LRET. This suggests that, after their release into the environment at mid-latitudes, their likelihood of reaching and accumulating in remote regions is relatively low compared to POPs. Nonetheless, their presence at VRS has been confirmed, and their occurrence in other remote regions has also been documented.[34,37,39–41] This apparent discrepancy may originate from various contributing factors. In this section, we aim to propose and examine multiple hypotheses to identify alternative possible sources and explanations.

The persistent presence of FMs at VRS throughout the study period suggests that these compounds are transported through atmospheric processes, with dry gaseous deposition playing a significant role in their deposition at VRS. The vapor-phase presence of these compounds across a wide temperature range (from −25 to +25 °C) suggests that they might remain in the atmosphere enough to be transported and deposited even in the absence of precipitation. The contamination event on May 21st, characterized by high salicylate concentrations, coincides with a precipitation event one week earlier, supporting the possibility of transport from Eurasia, considering air masses originating from areas within the polar circle. OCR, particularly, stands out as the only compound with a LRET potential comparable to that of POPs, likely due to its predicted sorption to particles at all temperatures. The elevated concentration of OCR, observed in the sample taken on April 22nd supports the hypothesis that it was transported from northern Europe through atmospheric LRET, with dry deposition potentially being the primary removal mechanism, given the absence of precipitation during the week prior to sampling.

One plausible alternative explanation is that the presence of selected PCPs predominantly originates from local sources. In recent years, efforts have been expended on identifying the primary sources of local contamination in polar regions.[35,38,41,78–80] Wastewater treatment plants (WWTP) are often identified as significant contributors to contamination in polar regions,[35,41,79] alongside ships navigating polar waters.[8,80] WWTPs in the Arctic are less advanced and relatively outdated compared to those in mid-latitudes.[35] However, local emissions from ships are unlikely to be a significant source at VRS, primarily due to the presence of year-round ice along the coast and in the fjord system, which remains ice-free for only about one month during the summer. Furthermore, the presence of PCP chemicals is closely tied to human personal care practices, with local contributions of PCP ingredients influenced by factors such as local population density, which is extremely low at Station Nord, seasonal habits and requirements (*e.g.*, the utilization of UV-filters varying with the seasons), as well as prevailing environmental conditions such as dark and sunlight period. Interestingly, the concentration range of the same UVFs analyzed in this study were found to be higher in northwestern Svalbard during the dark winter months (15–55 ng L^{-1}), despite the fact that these compounds are not used locally during that time.[34]

Another hypothesis is that the global scale of production, use and emissions of these PCPs is so substantial that it ensures their detection in remote regions, even if their potential for LRET is relatively low. Since Eurasia and North America are the source regions most likely to impact VRS, information about the average annual production of the analyzed PCP chemicals (tonnes per year) was collected for the European Union[81] and the United States of America,[82] while no official data could be found for Russia. Salicylates, for instance, are classified as high production volume chemicals.[83] Given their significant production volumes, ranging from 10 to 10 000 tonnes per year in the European Union alone, and their price range of less than 10 to 30 euros for 100 g (Table S2†), it is likely that they are largely emitted at lower latitudes, remain in the gas phase, and reach the Arctic. This makes it plausible to detect them at VRS and other polar sites (Table 1), even if their potential for LRET should be relatively low.

Lastly, the EFA model may overlook certain critical processes. For instance, the quantitative structure–property relationships used in this study might underestimate the half-lives of the compounds. Additionally, these half-lives could be considerably longer if the very low concentrations of photo-oxidants during the polar night were considered. However, the highest concentrations of PCP chemicals were found in snow sampled in April and May, when sunlight has returned even to high Arctic latitudes. While the model provides a preliminary indication of a contaminant's behavior in the environment, actual environmental dynamics may deviate. This might explain

Table 1 Literature data of salicylates in various matrices in polar and mountain regions

Location	Environmental matrix	Chemicals	Range (ng L^{-1})	Reference
Ny Ålesund (Svalbard)	Snow	Isoamyl, AmylS, HexS, BenS, EHS	ldl[a]–8	40
			1–62	34
Terra Nova Bay (Antarctica)	Sea water	Isoamyl, AmylS, HexS, BenS	ldl–29	37
	Wastewater		2–29	
Mount Elbrus (Russia)	Ice	Isoamyl, AmylS, HexS, BenS	7–102	84

[a] ldl – below the detection limit.

the episodes on April 22nd and May 21st, which account for the highest concentrations at VRS. The explicit consideration of the intermittency of precipitation in the EFA model makes sure that the potential for atmospheric LRET of PCPs ingredients with low Henry's law constant or strong particle sorption, such as the UVFs, is not underestimated. However, as the analysis of Fig. 4 showed, the PCP chemicals do generally not have a sufficient affinity for the aqueous phase for vapor scavenging to be an important deposition process.

Although the tool does not account for riverine transport,[85] and water transport in the EFA model is parameterized to reflect oceanic advection, contaminant transport *via* river water is not relevant for Northeastern Greenland. However, it could play a role in other Arctic regions with large North-bound rivers such as Siberia and the Canadian Arctic.

Potentially all the aspects proposed here play a role and may affect the presence of PCP chemicals in remote regions to a different extent. This highlights the need for further research on this topic, adopting an interdisciplinary approach that integrates field data, PCPs production and import data, and insights into human habits related to the use of PCPs, especially in remote regions. Moreover, the integration of atmospheric concentrations data for the selected PCPs would enhance our understanding of their dynamic processes.

4. Conclusions

This study highlights the presence of PCPs in snow samples collected at VRS in Northern Greenland, with most targeted PCPs detected throughout the sampling period, with highest concentrations observed during two events in spring. Back trajectory analyses suggest that medium and long-range atmospheric transport, with air masses originating from northern Europe and the northwestern coasts of Russia, contributes to the presence of these compounds. The partitioning behavior of the PCPs indicates that dry gaseous deposition is the dominant deposition mechanism. Whereas the Tool predicts relatively low potential for LRET for the measured PCP chemicals, high production and release volumes might help explain their occurrence in polar remote regions. Notably, OCR stands out from the investigated PCP chemicals by its low volatility, suggesting consistent sorption to atmospheric particles and removal from the atmosphere through wet and dry particle deposition. Whereas a relatively high estimated potential for LRET distinguishes OCR from other PCP chemicals, this high estimate is a result of (i) a high potential LRET assumed for atmospheric particles in the modified version of the OECD tool and (ii) the assumption that particle-sorbed substances are not subject to reactions with photooxidants.

These findings highlight the need for further research on atmospheric quantification of these compounds to improve understanding of their environmental behavior and transport mechanisms. Such research is important for resolving the discrepancies between observed concentrations at VRS and model-based LRET predictions, ultimately advancing our knowledge of PCPs' impact in polar regions.

Data availability

The data supporting this article have been included as part of the ESI.†

Conflicts of interest

There are no conflicts to declare.

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