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21 22 Monitoring microplastics in the atmosphere and cryosphere in the circumpolar North: A case for multi-compartment monitoring

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23 Abstract

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The atmosphere and cryosphere have recently garnered considerable attention due to their role in 24 25 transporting microplastics to and within the Arctic, and between freshwater, marine, and terrestrial environments. While investigating either in isolation provides valuable insight on the 26 fate of microplastics in the Arctic, monitoring both provides a more holistic view. Nonetheless, 28 despite the recent scientific interest, fundamental knowledge on microplastic abundance, and 29 consistent monitoring efforts, are lacking for these compartments. Here, we build upon the work of the Arctic Monitoring and Assessment Programme's Monitoring Guidelines for Litter and 30 Microplastic to provide a roadmap for multi-compartment monitoring of the atmosphere and 31 cryosphere to support our understanding of the sources, pathways, and sinks of plastic pollution 32 across the Arctic. Overall, we recommend the use of existing standard techniques for ice and 33 atmospheric sampling and to build upon existing monitoring efforts in the Arctic to obtain a 34 more comprehensive pan-Arctic view of microplastic pollution in these two compartments. 35

Keywords: air, Arctic, atmospheric deposition, sea ice, ice cores, atmospheric transport 36

37 1. Introduction

Plastic pollution including larger plastic litter and microplastics (≤ 5 mm) has been identified as 38 39 an emerging concern in the Arctic (AMAP 2017; PAME 2019), especially given its inherent 40 complexity of morphology (e.g., color, shape, size), chemical composition (i.e., polymer type, additives), and associated chemicals (Rochman et al. 2019). Further, microplastics are ubiquitous, 41 42 and have been detected in numerous biotic and abiotic samples across the circumpolar North, 43 including mammals (e.g., Moore et al. 2020; Carlsson et al. 2021), seabirds (e.g., Baak et al. 2020; 44 Trevail et al. 2015), fish (e.g., Morgana et al. 2018; Kühn et al. 2018), invertebrates (e.g., Fang et

al. 2018; Iannilli et al. 2019; Granberg et al. 2020), seawater (e.g., Ross et al. 2021; Tekman et al. 45 2020), wastewater (e.g., Herzke et al. 2021), sediment (e.g., Bergmann et al. 2017; Kanhai et al. 46 47 2019; Mu et al. 2019), sea ice (e.g., Obbard et al. 2014; Peeken et al. 2018), lake water (e.g., 48 González-Pleiter et al. 2021), and atmospheric deposition (i.e., wet deposition (e.g., Bergmann et 49 al. 2019); dry-deposition; (Hamilton et al. 2021)). Despite the widespread presence of 50 microplastics in the Arctic, their sources remain poorly understood, including the relative importance of local and distant sources of microplastics (Hallanger and Gabrielsen 2018; Herzke 51 et al. 2021; PAME, 2019). 52

53 Sources and pathways of microplastics have been reviewed by Brown (2015) and Li et al. (2020). We consider sources of plastic as their "origin of anthropogenic input into the environment". With 54 55 regard to the Arctic, sources can thus be within or outside the Arctic, i.e., microplastics in the Arctic can be from local sources or be locally introduced via long-range transport. We consider 56 pathways of microplastics as the physical transport process, e.g., with ocean currents (van Sebille 57 58 et al. 2020) or via atmospheric transport (e.g., Allen et al. 2019), that move microplastic particles in the environment. The majority of studies on the transport of microplastics have focused on ocean 59 pathways (e.g., Lusher et al. 2015; Tekman et al. 2020).). Ocean currents originating in the south 60 have been proposed to function as conveyor belts, carrying microplastics from the more densely 61 62 populated southern areas in Europe to the Arctic (Cózar et al. 2017; Tekman et al. 2020). Further, 63 local sources, such as untreated wastewater, can cause considerable microplastic pollution, which may be regionally distributed within the aquatic environment (Herzke et al. 2021). In addition, he 64 2019 report of the Arctic Council Working Group on the Protection of the Marine Environment 65 66 (PAME) identified atmospheric circulation as a potentially important transport pathway (PAME, 67 2019). However, given the limited empirical data and lack of harmonised methodologies for Arctic Science Downloaded from conscience pub.com by ALFRED-WEGENER-INSTITUT on 06/02/22 For personal use only. This Just-IN manuscript is the accepted manuscript prior to copy editing and page composition. It may differ from the final official version of record.

68 sample collection, it is not vet possible to estimate the magnitude of atmospheric transport of 69 microplastics to the Arctic. Similarly, little is known about microplastic abundance within the 70 Arctic cryosphere, including land-fast ice, pack ice, and land-based ice (e.g., ice caps, ice fields, seasonal ice in freshwater lakes and rivers, glaciers). These ice types are of different origin and 71 72 therefore likely to have different sources of microplastic contamination. Therefore, it is essential 73 that we monitor the atmosphere and cryosphere to fully understand the fate and transport of microplastics into and within the Arctic, including the role of air and ice as a transport medium 74 and, with regard to the cryosphere, as a reservoir for microplastics (Figure 1). 75

The Arctic Monitoring and Assessment Program (AMAP) has outlined a multi-compartment approach, which has the potential to improve our overall understanding of microplastic movement within the Arctic environment (AMAP, 2021a). Here, we expand upon AMAP's Litter and Monitoring Guidelines for air, ice, and snow (AMAP, 2021b) and discuss the strengths and limitations of monitoring microplastics in the atmosphere and cryosphere. Further, we highlight research gaps that should be prioritized for future monitoring efforts across the circumpolar North.

83 2. State of the science

2.1 Microplastic in the atmosphere and long-range transport

Although microplastics (e.g., microfibres, fragments, films, and foams) have been identified in
both polar regions (Isobe et al. 2017, Waller et al. 2017; Peeken et al. 2018b; PAME 2019, Materić
et al. 2022), the majority of studies on microplastics in the atmosphere, ice, and snow have focused
on Arctic environments (e.g., Obbard et al. 2014; Peeken et al. 2018a; Bergmann et al. 2019;
Kanhai et al. 2020; Von Friesen et al. 2020; Brahney et al. 2021; Kim et al. 2021, Materić et al.

2022). Like other atmospheric particles, microplastics are expected to undergo long-range 90 91 transport via air currents followed by wet and dry deposition onto water and land (Allen et al. 92 2019). Compared to ocean currents, air masses can widely distribute microplastics, within a matter 93 of hours or days (Stohl, 2006). Liss (2020) suggests that the atmosphere may contribute as much as 10 million tonnes of microplastic per year to the oceans worldwide. This is comparable to 94 95 estimates of riverine inputs of 5–13 million tonnes per year (Jambeck et al. 2015). Based on simulations of atmospheric transport of road wear particles, Evangelious et al. (2021) estimated 96 that 5-10% of all tire and brake wear particles in the size fraction $<10 \ \mu m$ (particulate matter 10 97 $[PM_{10}]$) emitted globally are transported to the Arctic. However, imperial measurements to 98 99 confirm these measurements are lacking. Furthermore, nanoplastic particles from tire wear were recently detected in a 14 m deep Greenland firn core (Materić et al. 2022). Microplastic particles 100 can undergo physical changes during atmospheric transport, including fragmentation, UV 101 102 degradation, and chemical weathering. Cai et al. (2017) recorded signs of degradation such as 103 grooves, pits, fractures, and flakes on microplastic particles collected in atmospheric deposition 104 and suggested they were caused by collision and friction, as well as chemical weathering due to 105 the high irradiation levels in the atmosphere. Fragmentation during transport likely increases the 106 potential for long-range transport (Biber et al. 2019).

107 The strong seasonal changes in the Arctic may also impact the transport of airborne microplastic, 108 e.g., changes in air mass transport, the presence or absence of UV light, as well as its intensity, 109 and the impact of sea spray, on the levels of microplastics in both air and water (Allen et al. 2020). 110 The polar sunrise and Arctic haze season are known to create reactive environments that could 111 both enhance the deposition of microplastics or cause fragmentation, which may result in longrange transport of smaller particles. Thus, monitoring of airborne microplastic should ideally takeplace throughout the year, similar to other contaminants (Wong et al. 2021).

114 Few studies have addressed the trajectory or transport pathways of microplastics in the 115 atmosphere. Nonetheless, they generally note that microfibres are the most common shape 116 identified in atmospheric deposition samples (e.g., Dris et al. 2016; Cai et al. 2017; Bullard et al. 117 2021). In addition, Wright et al. (2020) showed a predominance of microfibres in microplastics bulk deposition in London (UK) and estimated travel distances of 12 and 60 km for non-fibrous 118 119 and fibrous material, respectively, with an influence area of fibrous microplastics from 640 to 8700 120 km². Using air mass trajectory analysis, Allen et al. (2019) estimated a travel distance of 95 km 121 for microplastics observed in the Pyrenees. Notwithstanding these few studies, the atmospheric 122 transport of microplastics has been widely noted as a gap in knowledge (e.g., Allen et al. 2019; 123 Wright et al. 2020; Zhang et al. 2020; Bullard et al. 2021).

124 **2.2** Microplastics in the cryosphere

125 Cryosphere matrices (e.g., sea ice, land-fast ice, ice caps, ice fields, glaciers, etc.) tend to sequester 126 microplastics, and act as temporary storage and regional transport vector (Obbard et al. 2014; Peeken et al. 2018a; von Friesen et al. 2020; Kanhai et al. 2020; Ásmundsdóttir et al. 2020; Kim 127 128 et al. 2021). The mechanism of microplastic sequestration is likely dependent upon the origin of 129 the ice (e.g., seasonal sea ice versus ice fields created by snowpack). Atmospheric deposition (e.g., 130 wet and/or dry deposition), as a pathway for microplastics into Arctic sea ice, was suggested by Geilfus et al. (2019), who found high microplastic concentrations in the surface layer of an open 131 132 sea ice tank experiment. However, when measuring in-situ sea ice cores from the Baltic Sea they 133 could not corroborate these experimental results (Geilfus et al. 2019). The Baltic findings are in

line with observations of Arctic sea ice cores, which generally lack high concentrations of microplastics in the surface (Peeken et al. 2018a, Kanhai et al. 2020). This is further supported by Kim et al. (2021), who showed that less than 1% of observed microplastic entrapped in sea ice could be related to snowfall in the western Arctic Ocean, while the remaining proportion was a result of microplastics sequestered from seawater. In contrast, Bergmann et al. (2019) recorded comparably higher concentrations of microplastic in Eurasian Arctic snow, which might be explained by more polluted air masses or analytical differences.

141 Microplastics identified in land-based snowpack and ice (e.g., ice caps, ice fields) are a direct result 142 of both wet and dry atmospheric deposition (Kim et al. 2021; Ambrosini et al. 2019; Bergmann et 143 al. 2019; Geilfus et al. 2019; Cabrera et al. 2020; Materić et al. 2020; Stefánsson et al. 2021). There 144 is evidence of microplastics in glacial debris from the Forni Glacier, Italian Alps, by Ambrosini et 145 al. (2019) at concentrations comparable to those found in European marine and coastal sediments 146 (Gomiero et al. 2019; Haave et al. 2019). Microplastics recently observed in snow covering the 147 Vatnajökull ice cap in Iceland also suggest their presence in compacted deeper glacial layers (Stefánsson et al. 2021). Furthermore, Materić et al. (2022) identified nanoplastic in the Greenland 148 149 ice sheet and attributed these findings to long-range transport as the source (Materić et al. 2022). 150 In concert, organic contaminants, transported to polar regions in the gaseous phase or associated with particles, have been found in multi-year high-altitude ice caps and ice fields, where 151 152 atmospheric deposition is the main source of contaminant transport (e.g., Hermanson et al. 2010; Na et al. 2020; Gao et al. 2020; Xie et al. 2020; Hermanson et al. 2021). These sites have yet to be 153 154 investigated for microplastics.

Another important feature of Arctic sea ice is its seasonal cycle of growth and melt. For example,the European Arctic margin is influenced by drift ice formed on the Siberian shelves and carried

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by ocean currents to the Fram Strait via the Transpolar Drift (Serreze et al. 1989). Studying various 157 158 sea ice cores along the Transpolar Drift, Peeken et al. (2018a) could show that ocean currents had 159 a unique microplastic fingerprint, which was reflected in their sea ice. In addition, similar polymer 160 compositions and plastic shapes between the western Arctic Ocean and the Arctic Central Basin 161 suggest a strong connectivity between these two basins and a considerable input of microplastics 162 through the Pacific inflow in the Bering Street into the Arctic basin (Kim et al. 2021). Upon entering the major outflow gateways of the Arctic, microplastics are likely released from the 163 164 marginal ice zone (Obbard et al. 2014; Peeken et al. 2018a; Von Friesen et al. 2020; Kim et al. 2021). Displacement of microplastics from the marginal ice zone into deep-sea sediments at the 165 166 HAUSGARTEN observatory in the Fram Strait was proposed by Bergmann et al. (2017) and further corroborated by modelling of microplastic pathways in Fram Strait sediments and water 167 (Tekman et al. 2020). Furthermore, Fang et al. (2018) observed high microplastic concentrations 168 169 in benthic organisms caught below the ice covered Pacific inflow gateway (Fang et al. 2018). 170 Given the marked reduction in age, thickness, and extent of Arctic sea ice cover in recent decades 171 (Polyakov et al. 2012; Stroeve et al. 2012), it is likely that sequestered microplastic will be 172 increasingly released by the major outflow gateways into Arctic and sub-Arctic pelagic water 173 systems. In a warming Arctic, the occurrence, movement, and freeze thaw cycles of ice can be anticipated to play an even stronger role in the link between the atmospheric, aquatic, and 174 175 terrestrial environments with regard to microplastic accumulation and transport.

These studies, although limited in number, already indicate the presence of microplastics both in the atmosphere and cryosphere, with implications for transport to and distribution within the Arctic. Considering the rapid changes in the Arctic cryosphere (Ásmundsdóttir and Scholz 2020), ice may play a dynamic role in the storage, transport, and release of microplastics. However,

published knowledge on the connectivity between the role of ocean currents and atmospheric input of microplastic in the Arctic is lacking. Future monitoring studies should include multicompartment monitoring to enhance our understanding of the linkages and governing factors controlling the exchange of microplastic between compartments and to obtain a better understanding of sources and pathways of microplastic in the Arctic.

186 **3. Sampling methods and challenges**

187 **3.1 Sampling the atmosphere**

188 Although the monitoring of air and ice is important for a holistic understanding of microplastic occurrence in the Arctic, sample collection faces practical challenges. The routine collection of air 189 samples for microplastics in the Arctic is limited because of the remoteness, harsh climatic 190 191 conditions (e.g., wind, frigid temperatures), and limited access to power (AMAP, 2021b). 192 However, there is a growing knowledge base on the atmospheric sampling of microplastics that 193 can provide examples of appropriate sampling strategies. In general, atmospheric studies on 194 microplastics have used traditional air and precipitation monitoring methods, such as active air samplers, bulk deposition samplers (Dris et al. 2016; Allen et al. 2020; Roblin et al. 2020), wet-195 196 only deposition samplers (Brahney et al. 2020; Roblin et al. 2020), dry dust collectors (Brahney et 197 al. 2020), and snow samplers (Figure 2). Nonetheless, the strong wind conditions in the Arctic are 198 a challenge compared to less exposed regions.

Sampling methods that allow continuous measurements throughout the year are beneficial foratmospheric microplastic research; however, the lack of electrical infrastructure can make

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continuous active air sampling a challenge. One solution is to use existing Arctic research stations 201 202 for atmospheric monitoring. The stations used for contaminant monitoring were recently described 203 by Wong et al. (2021) and include the Zeppelin Observatory on Svalbard, Alert and Little Fox 204 Lake in Canada, Villum Research Station in Greenland, Stórhöfði in Iceland, Pallas in Finland and 205 Andøya in Northern Norway. The study also included the stations Amderma and Tiksi in Northern Russia (Wong et al. 2021). However, extending current sampling programs to microplastics will 206 207 require adjustments to equipment and procedures, as well as dedicated quality assurance/quality 208 control (QA/QC) protocols for microplastics. Sampling sites co-located with meteorological 209 measurements will provide valuable supporting information of high relevance for data 210 interpretation, such as wind speed, wind direction, precipitation, and temperature These data can 211 provide insights into seasonal variability of microplastic concentrations due to changes in wind 212 patterns or short-term transport events. Alternatively, passive sampling methods can be employed 213 as a screening method to determine microplastics in an area at a given time. Passive sampling 214 methods for plastic particles have been explored and developed as a way to increase spatial 215 coverage, provide a relative comparison between different areas, and evaluate relative atmospheric 216 deposition at a particular time (Pienaar et al. 2015). As they are usually more easily operated than 217 active samplers, passive sampling methods (e.g., moss bags, petri dishes) can engage local 218 communities and further enhance capacity building in the field of microplastic monitoring. In this 219 context, moss and lichen biomonitoring appear to be a cost-effective tool to study airborne 220 contamination including microplastic deposition (Roblin and Aherne, 2020; Loppi et al. 2020) and 221 through the use of moss or lichen bags (Temple et al. 1981) they may be particularly suitable 222 during winter conditions. When compared with snow samples, moss bags are considered to provide 223 a more homogeneous and better controlled sampling method (Salo et al. 2016).

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3.2 Sampling the cryosphere

While various glacial coring programs are ongoing in the Arctic, primarily targeting climate 225 226 reconstruction (e.g. Weißbach et al. 2016), there are currently no land-based cryosphere coring 227 campaigns for microplastic (i.e., glaciers, ice caps, ice fields) in the circumpolar North, although 228 legacy samples from such campaigns have been analysed (Materić et al. 2022). However, sea ice 229 sampling has been described for the Arctic, and several studies evaluating plastics have used traditional coring techniques (e.g., Kovac corers; Obbard et al. 2014; Peeken at al. 2018a), which 230 231 can be applied to sea ice sampling. Monitoring programs that have a particular interest in mass-232 based abundance of microplastics in sea ice or potential impacts of microplastic to ice-based 233 organisms, are encouraged to collect several replicate cores from the same ice floe. Furthermore, 234 additional sea ice cores can provide valuable ancillary data for temperature, salinity, black carbon 235 content, and biological parameters (e.g., chlorophyll, cell counts) to provide a more holistic view 236 of the sampled sea ice and thus evaluate how microplastics might affect ecosystem services. 237 Specific markers, such as rare Earth elements, are helpful for elucidating the history of the sampled sea ice (e.g., riverine input; Laukert et al. 2017). Sampling ice caps, ice fields, and glaciers also 238 239 requires drilling tools (e.g., US Ice Drilling and Design Operations hand auger (76 mm) and further 240 handling is similar to that of sea ice cores (e.g., Materić et al 2022). When evaluating ice from glaciers, ice fields, ice caps, etc., it is important to take replicate cores for high-resolution age-241 depth data. Moreover, replicates are highly recommended for more robust data to compensate for 242 243 heterogeneous distribution within both land and sea-based ice samples.

244 3.3. QA/QC practices

In general, field sampling carries the risk of contamination, which should be reflected in sampling 245 246 protocols, i.e., field techniques should be employed that prevent procedural contamination during 247 the collection of cryosphere and atmospheric samples. For example, samples should be taken 248 against the prevailing wind direction. Field technicians in warmer weather should not use gloves 249 and in colder weather should wear natural fibers (i.e., wool, leather or cotton) for hands and head. 250 Field sheets should record the material types and colors being worn including footwear while 251 sampling, possible, clothing of field technicians should be analysed as a means of QA/QC. 252 Likewise, laboratory facilities with controlled, particle free environments and techniques must be 253 ensured for the processing of the samples. Laboratory technicians should wear cotton laboratory 254 coats and work within a clean room and laminar air flow hood when available.

255 Procedural laboratory and field blanks are of the utmost importance in order to evaluate method 256 quality and provide accurate data, especially given that plastic particle counts are often quite low 257 in Arctic regions. During field sampling, procedural blanks (e.g., one for every 10 samples, or at 258 least one per sampling site) should undergo the exact same processing as a field sample. For 259 example, when taking active air samples, an additional sampling head (Figure 2) should be taken 260 to the field, loaded in the air sampling apparatus, attached to the pump, and allowed to draw air 261 for <30 seconds. Similarly, for passive samplers, blanks should be brought to the field, deployed, and immediately retrieved. Blanks should be covered, stored, transported, processed, and analysed 262 in the same way as the environmental samples. This way, procedural contamination throughout 263 264 the entire sampling and analysis process can be evaluated and results can be corrected or flagged 265 accordingly (Figure 3). For ice sampling, entire cores or individual sections should be cut with a 266 stainless steel, non-coated blade (e.g., bone saw). The outer part of the core (i.e., firn) should be cleaned with a non-plastic, non-coated grater (e.g., stainless steel, ceramic) to ensure the removal 267

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of any surface contamination. Ice core or snow melting should occur in a pre-cleaned, sealed stainless steel or glass jar to further prevent procedural contamination. Plastic airborne contamination in the sample preparation area should be monitored and reported alongside the results of the environmental samples.

Furthermore, it is imperative that particle specification methods are included for all compartments 272 when reporting results (i.e., polymer type, colour, shape, length, and diameter). Sample analyses 273 274 should have multiple lines of evidence, such as microscopy (stereo or fluorescence) and chemical 275 identification techniques to determine polymer type (e.g., Raman spectroscopy, Fourier Transform 276 Infrared (FTIR), Laser Direct Infrared (LDIR) imaging, pyrolysis/gas chromatography-mass 277 spectrometry (GC-MS)). Further, external quality control schemes are being developed for 278 microplastics and should be utilized, e.g., Quality Assurance of Information for Marine 279 Environmental Monitoring in Europe (QUASIMEME; van Mourik et al. 2021).

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4. Recommendations for future monitoring and research priorities

282 Atmosphere and cryosphere microplastic research is still in its infancy, which poses challenges for 283 standardised monitoring (Figure 4). Furthermore, the resulting data gaps hamper our 284 understanding of transport processes and the role of local and distant sources. Since the various components of the cryosphere are quite different, research and monitoring strategies need to be 285 286 adapted to each, while still allowing connections between the compartments. While ice caps and 287 Arctic lakes are strongly impacted by atmospheric deposition (Figure 1; e.g., Louto et al. 2019), 288 the marine and riverine cryosphere might be more influenced by the plastic particle load of the 289 underlying water currents (Figure 1; e.g., Peeken et al. 2018). While atmospheric deposition is shown to be a contributing factor of microplastics in various water bodies, it remains a challenge
to quantify its importance; thus, active air monitoring at dedicated locations is necessary to provide
insight into the role of atmospheric deposition.

293 The relative contributions of different pathways to the marine environment, including ocean 294 transport, riverine inflows, atmospheric deposition, and biological transport, might differ between 295 locations, seasons, and for different types of plastics. This needs further research to be properly 296 quantified, however, reliable and comparable methods are essential and should be a primary area 297 of development. Experiences and lessons learned from better-developed research on marine 298 microplastics can be used and adapted to address questions relating to microplastics in other 299 environmental compartments (e.g., QA/QC, quantification, and identification techniques). This 300 involves building upon already existing monitoring infrastructure and co-creating monitoring 301 programs with Northern partners that address local interests towards Northern led research.

Plastic pollution of the Arctic environment directly affects Arctic communities, as microplastics 302 303 have the potential to accumulate in Arctic food chains (Moore et al. 2022). In addition, 304 microplastics in the air could also be inhaled by local Arctic community members, especially in 305 areas prone to sea spray (Allen et al. 2020). The monitoring needs for plastic pollution across the 306 Arctic provide opportunities for Indigenous and community-based produced and co-produced 307 research and long-term monitoring programs, including sampling campaigns with appropriate 308 QA/QC schemes. For example, Hamilton et al. (2021) used simple passive air sampling methods 309 (i.e., petri dishes lined with double sided sticky tape) deployed by local partners in Nunavut, 310 Canada. The deployment of these samplers was used in part to determine atmospheric deposition 311 (i.e., dry dust deposition), but they were also used as a pilot project to determine feasibility and 312 usability in collaboration with local partners. Working together to produce manageable and Page 15 of 32

replicable monitoring methods that are guided and led by Indigenous researchers is crucial as we 313 314 work toward a strategic monitoring effort across the circumpolar North. Opportunities of aligning 315 monitoring priorities in the field of litter and microplastics with interests of northern and Indigenous communities and co-developing monitoring strategies have been discussed by 316 Provencher et al. ****. The National (Canada) Inuit Strategy for Research produced by the Inuit 317 318 Tapiriit Kanatami (ITK) organization, representing about 65,000 Inuit in the Canadian Arctic, has 319 presented a National Inuit Strategy for Research (ITK 2018). While each Indigenous group and 320 local communities across the Arctic will be different with varying research priorities and interests, 321 these principles could also be applied outside Canada, across the circumpolar North, with an 322 emphasis on community collaboration and co-production of monitoring efforts moving forward.

323 Contaminant monitoring infrastructure exists across the Arctic (e.g., Provencher et al. ****; Hamilton et al. ****; Bergmann et al. 2017; Parga Martinez et al. 2020), which could be built upon 324 325 in an effort to create a similar circumpolar monitoring program for plastic pollution in the 326 atmosphere and cryosphere. The Arctic air monitoring stations are equipped with active air samplers that collect a variety of organic contaminants (e.g., flame retardants, pesticides, 327 328 polychlorinated biphenyls), which could be expanded to include plastic particles. At Villum 329 Research Station in Greenland, a pilot project has been initiated on microplastic determinations in snow samples, with a strong focus on QA/QC protocols There is also a network of air quality 330 stations, close to or within the Arctic. For example, in Nunavut there are stations in Arviat, Igaluit, 331 332 and Kugluktuk. At these stations, gasses and particles (e.g., ozone, nitrogen dioxide, and PM_{2.5}) 333 are routinely monitored and provide potential sites that could be expanded for microplastics 334 research. Further, the European Monitoring and Evaluation Program (EMEP) includes monitoring sites across the European Arctic that could be expanded upon to include microplastic sampling. 335

Despite the growing interest regarding microplastic pollution in sea ice (e.g., Obbard et al. 2014; 336 337 Peeken et al. 2018a; von Friesen et al. 2020, Kim et al. 2021), there are currently no established 338 research or monitoring sites for sea ice (PAME, 2019). Monitoring could be implemented at 339 existing research stations by collecting extra cores for microplastic. For example, current regular 340 sea ice sampling occurs in the Hudson Bay, Cambridge Bay, and in Northern Baffin Bay, Canada. 341 Another targeted area could be Northeast Greenland in the outflow of sea ice from the Arctic Ocean as well as Young Sound (e.g., Daneborg/Zackenberg stations 74° N), where it is possible to 342 343 collect drifting sea ice during the summer months. Additionally, regular sampling campaigns like the ones occurring in Fram Strait (FRAM Pollution Observatory as part of HAUSGARTEN 344 345 Observatory) could monitor the outflow of Arctic sea ice and study the processes at the interface 346 between the ocean and the atmosphere by ship-based sampling. However, it is imperative to include extensive QA/QC protocols during ship-based sampling due to the high potential for ship-347 348 based contamination (Leistenschneider et al. 2021). Selected fjords near Svalbard or reoccurring 349 Central Arctic research vessel expeditions could include additional sea ice core sampling and air sampling programs for microplastics. Furthermore, collaborations with existing research programs 350 351 could be fostered to acquire additional (legacy) ice cores for plastic contamination from established 352 ice monitoring programs (e.g., US National Science Ice Core Facility, Canadian Ice Core 353 Laboratory, EGrip and NGrip on Greenland).

Estimates for the contribution of long-range atmospheric transport of microplastics versus local sources are lacking for both the marine and the terrestrial cryosphere. In contrast to previous assumptions, there are now indications that local sources play a role in the overall microplastic pollution in the Arctic ocean. For example, recent studies by Ross et al. (2021) Von Friesen et al. (2020) and Herzke et al. (2021) showed higher concentrations of anthropogenic microparticles

close to wastewater outlets and in the marginal sea ice zone. Currently over four million people 359 360 live in the Arctic (Heleniak and Bogoyavlensky, 2015) and most have no access to proper waste 361 management or wastewater treatment. Thus, plastic debris from openly exposed waste disposal 362 sites (e.g., open-pit landfills, open-pit burning) and microplastic from treated and untreated 363 wastewater enters the marine environment continuously (Magnusson et al. 2016; Granberg et al. 364 2019; Gomiero 2019; Herzke et al. 2021) and could be a local source for ice contamination and atmospheric deposition. Other local microplastic pollution sources in the Arctic are related to 365 366 shipping, fisheries, and tourism (PAME 2019). Typical polymers of these activities like varnish, polyamide, and polyethylene were traced to very small microplastic particles in Arctic sea ice 367 368 (Peeken et al. 2018a). Thus, the estimate of local sources should be an integral part of future 369 monitoring activities, which could include community-based assessments of plastic pollution (e.g., monitoring ice caps close to local communities or ice samples in a gradient along wastewater 370 371 effluent outlets).

372 River systems are another critical pathway that connects the freshwater, marine and terrestrial compartments, and should be monitored for plastic inputs in the Arctic (Frank et al. 2021; 373 374 Yakushev et al. 2021). Understanding the role of riverine transport can be important in 375 understanding the fate of microplastics, particularly in the cryosphere. For example, since a large 376 fraction of Arctic sea ice is created on shallow shelves (e.g., Laukert et al. 2017) or as anchor ice 377 on the actual seafloor in shallow areas (Reimnitz et al. 1987), microplastic with riverine origin or 378 resident in sediment can easily be transported as far from its sources as Fram Strait (Peeken et al. 379 2018a, Tekman et al. 2020). Given that 11% of the global riverine discharge enters the Arctic 380 Ocean (Fichot et al. 2013), Russian and Canadian rivers likely constitute important pathways for microplastic to the Arctic Ocean (Yakushev et al. 2021). Recent estimates suggest that previous 381

studies overestimated the worldwide input of plastic from rivers, implying much longer residence 382 383 time of plastics particles in the surface ocean (Weiss et al. 2021). Nonetheless, river systems should 384 be included in future monitoring activities, especially given the fact that most of the Arctic rivers 385 have a freezing cycle, which might further enhance the fragmentation of plastic litter and lead to 386 fast speeds of river currents in the melting season, which could promote particle transport to river 387 deltas. This information can be used to fuel two- and three-dimensional simulations of particle 388 transport trajectories, which have previously been used to identify the pathways of various polymer 389 types in the Arctic (Tekman et al. 2020). This will also improve 1-D thermodynamic models, which 390 together with the backtracking of sea-ice floes are a good tool to track the incorporation of various 391 polymer types during sea-ice growth (Peeken et al. 2018). Furthermore, robust models can improve our ability to evaluate any increasing accumulation of microplastic in the Arctic over time on the 392 scale of several decades, as well as studying the role of winter convection for downwelling 393 394 processes of microplastic to the seafloor and thus interconnecting with this compartment (Lusher et al. ****; Bergmann et al. 2017). 395

396

397 5. Conclusion

In addition to the proposed reporting methods highlighted in the AMAP Litter and Monitoring Guidelines (AMAP, 2021b), a multi-compartment monitoring approach can provide a more comprehensive understanding of microplastics in the pan-Arctic, including their transport to and distribution within the Arctic. Monitoring efforts should include multi-compartment sampling when appropriate, combining sampling of glaciers and atmospheric deposition, or sea ice and surface water, supplemented with relevant ancillary data for each compartment. To propel this area 404 of research out of its exploratory phase, and to create and sustain monitoring research efforts, 405 opportunistic sampling alongside existing monitoring programs is recommended. Furthermore, 406 knowledge sharing and collaboration with local communities, with an emphasis on community 407 research priorities, is crucial in creating successful and robust long-term monitoring programs 408 across the circumpolar North. Ultimately, a holistic monitoring approach that includes multiple 409 knowledge streams will increase our understanding of the inputs and outputs of microplastics in 409 various environmental compartments across the Arctic.

411

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429 writing, editing and writing.

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Figure 1: Graphic depicting the atmosphere and cryosphere compartments and transport pathways ofmicroplastics into and within the Arctic.



Figure 2: Sampling equipment for atmospheric microplastics, photographs showing (from left to right)
active air sampling (with sampling head), wet deposition only sampling, NILU bulk deposition collector,
and passive air sampling (including moss bags).

444

<u>Atmosphere</u>

Active sampling / Bulk deposition / Wet or dry deposition only / Passive sampler

- Prepare all collection vessels (e.g., bucket, Nipher gauge, petri-dish, etc.,) at the same time (including blanks)
- Deploy collection vessel/sampler to the air at collection site and recover immediately. Record exposure time
- Cover and store in the same manner as other samples
- Process alongside samples to account for procedural contamination throughout the entire process.

<u>Cryosphere</u>

Land and sea-based ice samples

- Prepare a moist collection vessel (e.g., stainless steel jar, glass bucket, etc., w/filtered reverse osmosis water)
- Expose collection vessel to the sampling environment for the same duration as ice core sampling
- The opening of the container should be big enough to collect/store an ice core (e.g., 9 cm wide) in order to be representative for field contamination
- Process alongside samples to account for procedural contamination throughout the entire process.

General considerations

Regardless of matrix

- One blank for every 10 samples, and/or 1 blank for every sampling site
- Blanks should be prepared, treated, and analyzed alongside samples to account for procedural contamination from the start of the process through the analysis phase
- Blank data should either be reported along side the sample data, or blank subtracted.

446 Figure 3: Preparation of procedural blanks and general considerations for proper quality
 447 assurance/quality control methods for atmosphere and cryosphere sampling.

448



451 Figure 4: Flow chart highlighting recommendations for monitoring, reporting, and future research452 priorities for microplastic sampling in the Arctic atmosphere and cryosphere.

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