
Supplementary information

**Microplastics and nanoplastics in the
marine-atmosphere environment**

In the format provided by the
authors and unedited

Title: The Atmospheric Cycle of Microplastics in the Marine Environment

Supplementary Note 1: The importance of atmospheric micro(nano)plastic transport to remote and polar regions

Supplementary Note 2: Atmospheric microplastics and climate change

Supplementary Note 3: CAM model global deposition and the global monitoring network

Supplementary Note 4: Marine atmospheric sampling platforms

Supplementary Table 1: Tabulated summary of sampling platforms and the advantages and disadvantages of their use in atmospheric micro(nano)plastic studies

1 **Supplementary Note 1: The importance of atmospheric micro(nano)plastic**
2 **transport to remote and polar regions**

3
4 The remote and polar regions are natural reserve areas relatively untouched by human
5 activity. They provide the last bastions of natural ecosystems and biodiversity. If
6 human activities and resource exploitation results in catastrophic ecosystem collapse
7 and biodiversity loss, these natural reserve areas provide the potential for future
8 natural regeneration.

9
10 It is possible the microplastic and nanoplastic are not transported solely by either
11 atmospheric transport or aquatic (ocean) transport to these remote areas and poles.
12 Plastic particles may undergo re-entrainment multiple times in terrestrial and marine
13 environments, resulting in long-distance transport via a cyclic entrainment-deposition-
14 re-entrainment process. This may allow for plastic pollution to reach areas that do not
15 have air or ocean currents feeding directly into them from polluted areas.

16
17 There is significant evidence of microplastic in the Arctic and Antarctic seas,
18 suggesting that oceanic currents are one key source of Arctic plastic pollution^{1,2} in the
19 polar marine environment. However, while polar ice acts as a sponge, collecting
20 marine plastic during its ice formation^{3,4}, the microplastic found on the surface of ice
21 flows and snow are not directly attributable to marine conveyance. Notable
22 microplastic quantities have been found on Arctic surfaces⁵, and early studies have
23 identified microplastic in the Antarctic atmosphere. It is suggested that these particles
24 were either transported long-distance through the atmosphere prior to polar
25 deposition, or were conveyed via marine currents then emitted during ocean
26 turbulence (e.g. through the bubble burst ejection process) and atmospherically
27 transported to the polar surface (land, glaciers or sea ice). Characterising and
28 quantifying the atmospheric transport contribution to remote area and polar
29 micro(nano)plastic is therefore necessary to both understand the remote areas
30 transport processes and to model plastic pollution influx (and future scenarios) to these
31 relatively pristine and remote locations.

32
33 **Supplementary Note 2: Atmospheric microplastics and climate change**

34
35 Research on micro(nano)plastic in the context of climate change is extremely limited
36 to date. Life cycle and greenhouse gas (GHG) emission analyses show
37 micro(nano)plastic to contribute to GHG, climate change/global warming potential and
38 photochemical ozone formation/ozone formation^{6,7}. Micro(nano)plastic lost to the
39 environment release GHG and interfere with ocean carbon fixation. Early model
40 estimations suggest current GHG from plastic from cradle to grave (incorporating
41 material extraction, production, manufacture, transport and waste management) of
42 1.34 gigatons CO₂ equivalent year⁻¹ by 2030, and by 2050 may consume 10-13% of
43 the remaining carbon budget⁷.

44
45 It has been hypothesised that deposition of brake wear and tyre wear particles on ice
46 and snow may accelerate warming of the cryosphere⁸. Micro(nano)plastic particles,
47 particularly black coloured particles, may function as cryoconites, increasing the snow
48 and ice melt in polar and high elevation locations. Given their hydrophobic nature,
49 microplastic and nanoplastic may act as cloud ice nuclei⁹. Certain regions may
50 therefore be particularly sensitive to the presence of airborne micro(nano)plastic, such

51 as the Southern Ocean, where cloud albedo is strongly linked to the concentration of
52 ice-nucleating particles¹⁰. Southern high latitudes have experienced enhanced UV
53 fluxes in recent decades owing to the Antarctic ozone hole; this is thought to enhance
54 the formation of micro(nano)plastics in surface waters and terrestrial environments at
55 southern high latitudes¹¹.

56
57 Airborne particulate matter influences surface climate by absorbing and scattering
58 solar and terrestrial radiation. While total aerosol number concentrations range
59 between $1 \times 10^9 - 1 \times 10^{10}$ per m^3 over Europe and East Asia¹², the concentrations of
60 airborne micro(nano)plastics in the same regions are much lower; between 0.01 –
61 5000 $MP\ m^{-3}$.^{13,14} Airborne microplastics therefore make only a small contribution to
62 total aerosol abundances in the present-day atmosphere. Electromagnetic scattering
63 and absorption calculations indicate that non-pigmented microplastics are efficient at
64 scattering solar (shortwave) radiation, and at absorbing terrestrial (longwave)
65 radiation¹⁵. Global climate model simulations show that the balance between
66 shortwave effects (which imply a cooling influence on Earth's surface) and longwave
67 effects (i.e., the greenhouse effect) depends strongly on the assumed vertical
68 distribution of airborne microplastics¹⁵. The effective radiative forcing (ERF), assuming
69 direct aerosol-radiation interactions only (that is, neglecting aerosol-cloud interactions)
70 is calculated to be on the order of $-0.746 - 0.044\ mW\ m^{-2}$ assuming a surface
71 concentration of $1\ MP\ m^{-3}$. Compared with the total aerosol effective radiative forcing
72 of -0.71 to $-0.14\ W\ m^{-2}$ due to aerosol-radiation interactions¹⁶, the microplastic ERF is
73 small in the present-day atmosphere.

74
75 The expected increase in atmospheric micro(nano)plastic abundances due to
76 increasing global plastic use and mismanaged plastic waste will increase microplastic
77 ERF in future, and may influence local and regional climate in regions where airborne
78 micro(nano)plastic concentrations are particularly large. Furthermore, emissions of
79 anthropogenic aerosols are projected to decrease in future, which is linked to expected
80 improvements in air quality¹⁷. Micro(nano)plastics will therefore make a larger relative
81 contribution to total aerosol ERF in future.

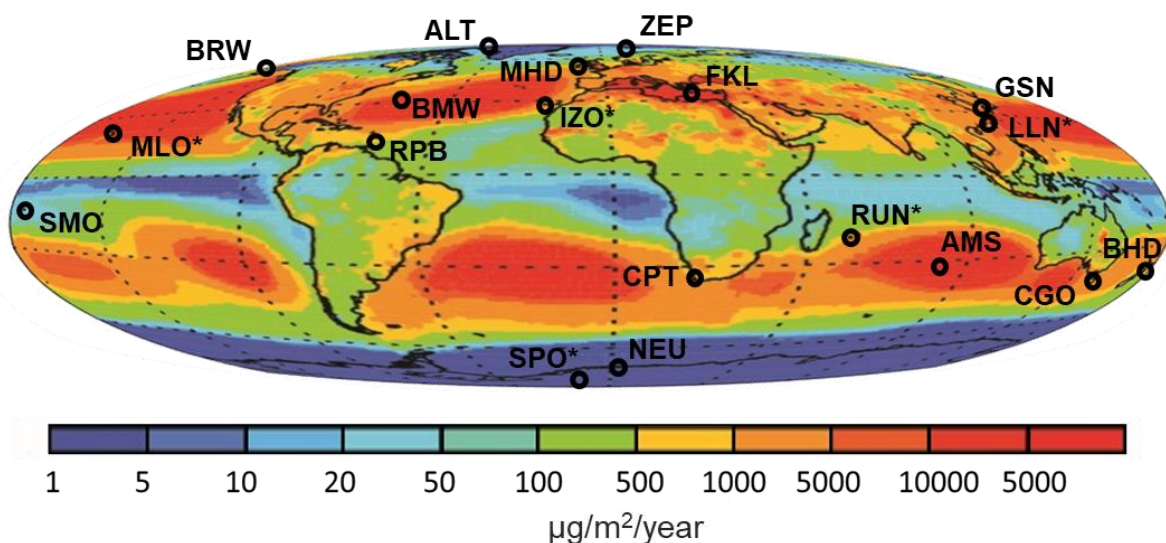
82 83 **Supplementary Note 3: CAM model global gross microplastic deposition and the** 84 **global monitoring network**

85
86 The Community Atmospheric Model (CAM) model has been used to estimate gross
87 microplastic deposition across the globe¹⁸. The results presented in Figure C3.1
88 highlight the high deposition of atmospheric microplastic across both northern and
89 southern hemisphere oceans (especially over the Pacific and Mediterranean
90 Oceans)¹⁸ as well as key terrestrial deposition areas in Europe, China, India, the
91 middle east, central and northern Africa eastern south America and the USA. The CAM
92 model results provided an interesting backdrop to the marine microplastic flux
93 question, providing an early insight into the gross deposition over the marine
94 environment (microplastic atmospherically transport from the terrestrial environment
95 as well as marine (secondary) sourced microplastic emission and (re)deposition.
96 These early global atmospheric microplastic model findings are important as it is the
97 first tentative assessment of total atmospheric microplastic transport, considering all
98 plastic polymer types and incorporating marine microplastic atmospheric emission.

99

100 The results of the CAM model are notably different from those presented for
 101 FLEXPART modelled tyre and brake wear global *net* deposition⁸. This is partly due to
 102 the inclusion of marine emission in the CAM model whereas Figure 4 **Error! Reference**
 103 **source not found**. FLEXPART models net deposition to the marine environment (no
 104 marine emission) therefore providing an insight into the terrestrial net influx of
 105 microplastic to the marine environment. The microplastic particle size included in the
 106 CAM model also differs from that used in the FLEXPART model, with the CAM model
 107 particle size extending from 250µm to 4µm while the FLEXPART model considered
 108 particles of either <10µm or <2.5µm (PM10 or PM2.5). As presented Figure C3.1 the
 109 quantity of microplastic particles increases significantly with decreasing particle size.
 110 Thus the FLEXPART model may incorporate a significantly greater number and mass
 111 of smaller, more easily atmospherically transported microplastic particles than the
 112 CAM model. It is also important to note the CAM model adopts a global road
 113 microplastic emission rate (all polymers in road dust, not just tyre and brake wear) of
 114 96Mt/yr rather than the 284MT yr⁻¹ used in the FLEXPART model¹⁸. These highlighted
 115 differences illustrate the need for better harmonization and compatible
 116 parameterisation and field work to characterise atmospheric micro(nano)plastic
 117 particles and transport dynamics to enable advancement of atmospheric
 118 micro(nano)plastic modelling.

119
 120 The proposed GAW/WMO long-term monitoring stations have been overlaid onto the
 121 global *gross* atmospheric microplastic deposition results from the CAM model analysis
 122 to illustrate that the proposed stations incorporate locations appropriate to analyse
 123 marine microplastic (re)emission and recycling as well as net marine microplastic
 124 deposition. The proposed GAW/WMO sampling sites extend onto high marine
 125 atmospheric microplastic gross deposition areas of the Atlantic, Mediterranean and
 126 Pacific as well as the low deposition zones of the Arctic and Antarctic. The proposed
 127 fixed platform network would therefore be effective in providing both an understanding
 128 of net marine micro(nano)plastic deposition and gross marine micro(nano)plastic
 129 cycling, advancing the early estimation of the terrestrially sourced atmospheric
 130 micro(nano)plastic flux to the marine environment and quantification of the marine
 131 atmospheric micro(nano)plastic cycle.



132
 133 Supplementary Figure 3 Possible sampling sites illustrated on maps of CAM modelling of the total
 134 atmospheric microplastic deposition (all polymer types)¹⁸. Locations identified with * are high altitude

135 (tropospheric) sites, all other locations are coastal monitoring sites. Sites are as follows: ALT Alert
136 (Canada); AMS Amsterdam Island (France); BHD Baring Head (NZ); BMW Tudor Hill (Bermuda); BRW
137 (Barrow, USA); CGO Cape Grim (Australia); CPT Cape Point (South Africa); FKL Finokalia (Greece);
138 GSN Gosan (Korea); IZO Izana (Spain, 2373 m); LLN Lulin (Taiwan, China 2862 m); MHD Mace Head
139 (Ireland); MLO Mauna Loa (USA, 3397 m); NEU Neumayer (Antarctica); RPB Ragged Point (Barbados);
140 RUN La Reunion (France, 2160m); SMO American Samoa (USA); SPO South Pole (Antarctica, 2841
141 m); ZEP Zeppelin (Norway).

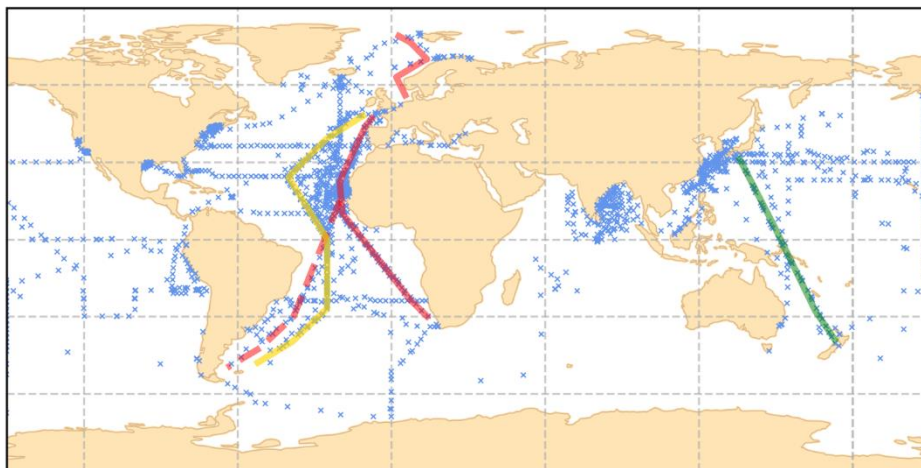
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143 **Supplementary Note 4: Marine atmospheric sampling platforms**

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145 Marine Research Vessels

146 Ships provide access to regions of the ocean that are otherwise inaccessible,
147 functioning with a typical campaign of 20-40 days. While ship-based research
148 (primarily research vessels) can provide sampling from currently unmonitored marine
149 atmospheric locations, the individual voyages result in a dataset that may be spatially
150 and temporally patchy¹⁹ and therefore climatologically less representative. Longer-
151 term monitoring research campaigns such as GEOTRACES²⁰, the Atlantic Meridional
152 Transect Programme²¹ and Alfred Wegener Institute ship-based research program
153 (e.g. RV *Polarstern* research campaigns)²² can however provide vital offshore marine
154 atmospheric information and be a valuable monitoring platform when employed multi-
155 annually (potentially also seasonally) over a specific transect. To date only pumped
156 aerosol samplers have been used on vessels, providing data of atmospheric
157 composition (only three marine cruises have sampled and published marine
158 atmospheric microplastic to date (Figure 2, Figure C4.1) across the Atlantic, Pacific
159 Antarctic and Arctic oceans primarily^{13,23,24}. In general, rainfall and dry deposition is
160 grossly under-sampled over the oceans considering its importance to the air-to-sea
161 transfer of material to the global ocean, with no precipitation or dry deposition yet
162 undertaken specifically for micro(nano)plastic analysis.



163

164 Supplementary Figure 4. Locations of aerosol samples collected from UK, French, German, Dutch, New
165 Zealand, Australian and Japanese ships and analysed at UEA from 2000 to 2019 (n = ~2070) (blue)¹⁹.
166 Atlantic Meridional Transect cruises are shown in yellow, *Polarstern* inter-hemisphere transfers and
167 Arctic campaigns in red and *Transfuture5* voyages in green.

168

169 To advance marine shipping research programmes, it is recommended both
170 atmospheric deposition and air concentration (pumped air sampling) be undertaken
171 on research cruises, and that these cruises occur across the range of seasonal
172 conditions and over multiple years (long-term monitoring). This would result in
173 illustration of seasonal and annual trends in marine atmospheric micro(nano)plastic

174 composition and provide information of the deposition rate of atmospheric
175 micro(nano)plastic to the ocean. With quantitatively characterised marine atmospheric
176 micro(nano)plastic deposition information, back trajectory location specific models can
177 identify the potential marine or terrestrial source of these particles, improving the early
178 estimations of marine atmospheric micro(nano)plastic flux. Once a substantive marine
179 atmospheric MP database is available, global flux modelling (such as undertaken by
180 Brahney et al. and Evangeliou et al.^{8,18}) to quantify the overall marine atmospheric
181 micro(nano)plastic flux can be executed with greater accuracy (both terrestrial
182 provisioning of micro(nano)plastic to the oceans and marine micro(nano)plastic
183 contribution to the terrestrial atmospheric micro(nano)plastic burden). Furthermore,
184 vessels transects can be designed to follow major continental outflows to examine the
185 trend in micro(nano)plastic atmospheric transport along established meteorological
186 conveyance pathways. Such micro(nano)plastic specific vessel based sampling help
187 document gradients in continental-sourced micro(nano)plastic concentrations and
188 potentially opposing gradients in sea surface microlayer sourced micro(nano)plastic,
189 demonstrating the spatial atmospheric micro(nano)plastic burden and influence of
190 terrestrial or offshore marine conditions.

191

192 Fixed coastal or island platforms

193 Two notable fixed platform networks have been used to analyse the continental
194 aerosol contribution to the marine environment over the past four decades. The
195 SEAREX Ocean Aerosol Sampling Network was designed to characterise marine
196 atmospheric chemistry and the role of continental to marine (ocean) transport within
197 the (western) Pacific Ocean²⁵. The AEROCE Network (interlinked with several World
198 Meteorological Organisation observatories and Global Atmosphere Watch
199 Programme(GAW)) undertook similar research extending along the east and west of
200 the central to north Atlantic Ocean. The SEAREX and AEROCE networks used 17-
201 20m walk-up scaffold sampling towers equipped with elevated atmospheric samplers
202 supported by temporary or permanent field laboratories located on both continental
203 coast and islands at the terrestrial-marine interface. Long-term monitoring occurred,
204 including year-round measurements linked to the specific synoptic meteorological
205 conditions, in conjunction with short-term intensive research activities that addressed
206 focused research questions (e.g. sea salt aerosol bubble burst emission
207 quantification).

208

209 The SEAREX and AEROCE networks long-term monitoring has paved the way for
210 marine atmospheric science breakthroughs over the past four decades^{26–29}. The
211 sampling strategy and network design was shown to be effective in capturing the
212 continental atmospheric contribution and influence to the marine environment and
213 helped to establish protocols and field campaign design for global marine atmospheric
214 monitoring strategies. During the SEAREX and AEROCE long-term monitoring
215 campaigns key results illustrated the needs for meteorological consideration in the
216 efforts to minimise sample contamination, the island effect (especially on condensation
217 nuclei concentrations) and the importance of sampling times steps of <24 hours to
218 ensure synoptic situations to be attributed to individual samples and back-trajectory
219 analysis can be undertaken with greater certainty. The SEAREX and AEROCE
220 networks provide a proven global network strategy that could be effectively utilised to
221 collect representative samples and undertake analyse consistent and comparable
222 analysis of global marine atmospheric micro(nano)plastic.

223

224

225 Aircraft, UAV, buoys

226 Manned aircraft have been used to collect atmospheric samples for short term or snap
227 shot monitoring of atmospheric composition^{30–32}. Recently, one campaign has used
228 manned aircraft to sample for micro(nano)plastic (over a populated terrestrial area)³³.
229 Manned aircraft can provide access to spatial locations and elevations which are
230 difficult to reach, operating at a variety of altitudes (PBL, troposphere and
231 stratosphere) over terrestrial or marine environments. Similar to constraints identified
232 with ship sampling, manned aircraft sampling occurs over a spatial extent rather than
233 single location and has campaign duration constraints (limited by flight times).
234 Furthermore, micro(nano)plastic (specifically environmentally weathered and
235 degraded particles) are very fragile and shatter under notable impact (such as
236 sampling at high wind speeds or at significant velocity). Therefore, significant further
237 research is necessary to quantify the efficiency of traditional research aircraft for
238 atmospheric micro(nano)plastic sampling. Studies are needed to ensure particle
239 integrity is retained and to quantify the micro(nano)plastic loss due to filter inlet
240 pressure (preventing particles from entering the sampling chamber due to aircraft
241 speed and inlet design) and by/through pass (particles by-passing or being sucked
242 through the filter due to pressure differential or excessive vacuum).

243

244 Unmanned aircraft systems (UAVs) have recently emerged as an effective low altitude
245 (PBL, troposphere) sampling platforms^{34–36}. While UAVs have not yet been used to
246 sample atmospheric MP, their low airspeed coupled with elevation range and access
247 to remote or difficult to sample locations suggests they may be an effective platform
248 for atmospheric micro(nano)plastic sampling. The low operational cost and easy
249 availability of UAV make them very attractive. Significant recent advances allow for a
250 complete automatic flight from launch to landing following a predetermined flight path.
251 However, these UAV have payload and/or duration limitations, requiring sampling
252 equipment to be minimalist (and battery powered active sampling is required). There
253 are also spatial and elevation limitations as under air law in most countries, the UAV
254 must stay within visual line of sight (VLOS) and have an altitude restriction of 400 feet
255 above ground level (~120m). These restrictions can be overcome under special
256 permits requiring substantial investment in both equipment and training. Larger long
257 range UAV such as the Global Hawk UAV, one of the largest available, has a 680kg
258 payload, an 8500 nautical-mile range and a 24-hour endurance. Unfortunately, this
259 aircrafts cruising speed is of a similar nature to the standard research aircraft.

260

261 Fixed wing and vertical take-off and landing (VTOL) fixed wing, a more commonly
262 available UAV, can have a 5kg payload, 100km flight range (within VLOS) and 5-hour
263 endurance. These may create opportunities for micro(nano)plastic research due to
264 their ability to fly very close to potential sources in complete safety whilst staying within
265 aircraft regulations. Many heavy lift multirotor UAV are available with up to 20kg
266 payload, however very few have a longer than 20 min flight duration severely limiting
267 their applicability to micro(nano)plastic research due to small sample size. For coastal
268 operations, low elevations and remote locations (marine launched UAVs) UAVs could
269 provide an effective sampling platform for marine and terrestrial atmospheric MP.

270

271 Buoys have been successfully used for marine atmospheric sampling, monitoring dust
272 and atmospheric composition in static marine locations at low (near sea surface)
273 elevations³⁷. Buoys may provide a complementary long-term static marine sampling

274 network complementary to fixed platforms and shipping vessel sampling, providing
 275 near surface air concentration or depositional data. Power supply on buoys can be
 276 limited, constraining sampling methodology to passive or low power consumption
 277 activities. Sample collection via buoys includes complexity due to their isolation
 278 (accessing buoy samples by ship may be difficult at regular intervals) resulting in
 279 potentially irregular sampling times and longer sample durations. Despite these
 280 limitations, use of buoys to undertake specific research intensive field assessment
 281 may be effective where low elevation sampling is required, and buoys may form part
 282 of a long-term monitoring network when combined with other global initiatives.

283

284 **Supplementary Table 1. Tabulated summary of sampling platforms and the**
 285 **advantages and disadvantages of their use in atmospheric micro(nano)plastic**
 286 **studies**

287

Platform Type	Advantages	Disadvantages
<u>Ships</u>	<ul style="list-style-type: none"> • Unlimited marine locations • Can have trained personnel and appropriate technical capacity (i.e. clean labs) • Potential of ocean-atmosphere exchange sampling • Long-term monitoring through repeated visitation of selected offshore stations over multiple years (e.g. GEOTRACES program) 	<ul style="list-style-type: none"> • Short-term (days or weeks to several months) temporal snapshots • Very dirty environment • May travel at speeds resulting in spatially ranging sample representation
<u>Sailing vessels</u>	<ul style="list-style-type: none"> • Unlimited marine locations • Can accommodate trained personnel • Functional in mild to moderate weather conditions • Slow passage speed supporting effective spatial sampling • Potential for ocean-atmosphere exchange sampling • Relatively low cost 	<ul style="list-style-type: none"> • Short to moderate temporal snapshots • Fewer personnel accommodated than ships • Limited on-board analysis
<u>Island/coastal sites</u> <i>WMO/GAW permanent sites</i>	<ul style="list-style-type: none"> • Synoptic, seasonal and annual variability • Trained personnel • Supporting chemical/meteorological measurements • Multiple elevations 	<ul style="list-style-type: none"> • Limited geographical locations
<u>Island/coastal sites</u> <i>Other permanent sites</i>	<ul style="list-style-type: none"> • Synoptic, seasonal and annual variability • Possible supporting chemical/meteorological measurements 	<ul style="list-style-type: none"> • Limited trained personnel • Limited geographical locations
<u>Island/coastal sites</u> <i>Non-permanent sites</i>	<ul style="list-style-type: none"> • Synoptic and possibly seasonal scale variability • Possible supporting chemical/meteorological 	<ul style="list-style-type: none"> • Limited/untrained personnel • Limited geographical locations

	measurements (in some cases)	
<u>Aircraft</u>	<ul style="list-style-type: none"> • Unlimited marine locations • Trained personnel • Multiple elevations 	<ul style="list-style-type: none"> • Very short-term temporal snapshots • Limited sampling intervals • Very expensive
<u>Drones & UAVs</u>	<ul style="list-style-type: none"> • Multiple elevations • Relatively Low cost 	<ul style="list-style-type: none"> • Limited marine locations unless launched from ships • Very short-term temporal snapshots • Limited sampling intervals • Limited power availability and payload
<u>Tethered or remotely controlled balloons</u>	<ul style="list-style-type: none"> • Full range of elevation (surface-PBL-troposphere) • Generally limited to terrestrial release but potentially possible from ships 	<ul style="list-style-type: none"> • Limited access due to expense, licensing • Snapshot sampling rather than continuous • Potential for constraints in spatial control • Limited sampling equipment payload (when considering multiple elevation sampling)
<u>Buoys</u>	<ul style="list-style-type: none"> • Synoptic, seasonal and annual variability • Possible wide geographical coverage 	<ul style="list-style-type: none"> • Difficult to service • Possible limited power • Excessive sea spray

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289
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