Title page

Title: The Atmospheric Cycle of Micro(nano)plastics in the Marine Environment

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Glossary

Microplastic (MP)
Plastic particles <5mm but >1µm (aerodynamic) diameter[1–4]

Nanoplastic (NP)
Plastic particle <1µm (aerodynamic) diameter[1–4]

Micro(nano)plastic (MnP)
All plastic particles ≤5mm (both micro and nano plastic)[1,2,5]. MP and NP can be represented as particles or mass per volume of sampled air, e.g. MP/m^3, and in deposition as particles or mass per surface area sampled over a specified duration, e.g. MP/m^2/day.

Primary micro(nano)plastic
MP manufactured to be 1µm-5mm (e.g. nurdles[6], personal care products[7], textiles[8])
NP manufactured to be <1µm (e.g. medical applications[9], printing ink[10], electronics[11–13])

Secondary micro(nano)plastic
MP or NP produced through mechanical, chemical or photo degradation (e.g. bottle top breakdown to MP and NP on a beach due to UV, salt and wave action)[13–16]

Source
An activity that results in MP or NP emission, described both in location and time and with reference to the plastic particle emission characteristics (primary or secondary).

Point source
MP or NP emission from a defined location at specific times (e.g. waste water treatment plant release to receiving waterway, recycling plant emission due to mechanical plastic deconstruction, plastic factory emission due to production activities)[17–19]

Diffuse source
MP or NP emission (and re-emission) from activities that have no single emission time and location (e.g. road dust or agricultural emissions)[17,18,20–22]

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Title: The atmospheric cycle of micro and nano plastics in the marine environment

Abstract

The discovery of atmospheric transport of micro(nano)plastic (MnP), coupled with ocean-atmosphere exchange of plastics, points to a highly complex cycle of emission, transport, deposition, and resuspension of plastic in the marine environment. In this perspective we consider the quantification of marine atmospheric MnP, MnP sources, transport and marine deposition, and the flux uncertainty (0.013-25 million metric tons per year) due to study inter-comparability and data limitations (~65 published studies). Crucial to advancing the accuracy of this flux is the creation of a comparable, harmonised global dataset representative of the full temporal and spatial extent of our marine atmosphere. We propose a global marine-atmospheric MnP observation strategy, incorporating novel future sampling methods to address key knowledge gaps. Together with long-term observations and intensive investigations, it will help define key source, transport and sinks in the marine-atmospheric MnP cycle, the trends in marine-atmospheric pollution and responses to future policy and management actions.

Key words: atmospheric microplastic, atmospheric nanoplastic marine micro and nanoplastic, atmospheric transport, ocean-atmosphere exchange, plastic cycle, airborne microplastic, aerial transport

1 Introduction

Plastic pollution is a function of production, use and waste, with plastic production increasing annually driven by rapid and inexpensive plastic production, non-circular economic models and a single use plastic culture (368 million metric tons created in 2019)[1,2]. Plastic waste lost to the environment has been evidenced across all environmental compartments (aquatic, soil, air)[3–6] and is projected to rise almost 3 fold to ~80 million metric tons per year by 2040 under a business as usual scenario (from 2016 environmental plastic pollution estimates)[7]. Of the total plastic waste created (managed and mismanaged), ~12% is projected to enter the aquatic environment and ~22% to enter the terrestrial environment, with an estimated ~60 million metric tons per year lost to just these two environmental compartments by 2030 (aquatic, terrestrial)[7,8]. However, there is currently limited assessment of the atmospheric compartment.

Microplastics have been frequently studied in marine and fresh water, biota and sediments[9–11] and these data coupled with models serve as the basis for quantifying global oceanic microplastic[12,13]. The transport of micro(nano)plastic (MnP) in ocean currents is a slow process. Terrestrial runoff, river discharge and marine currents carry MnP from terrestrial sources to distal areas such as the Arctic, Antarctic and deep-sea locations over a period of months to years[14]. Whilst slow, this mechanism is important in transporting MnP to remote areas where they can produce harmful impacts on marine life[15,16]. Atmospheric transport research similarly illustrates that wind can transport MnP at trans-continental and trans-oceanic scales[17–20]. Atmospheric transport is a comparably much faster process, capable of conveying particles from sources to remote locations over a matter of days to weeks[18,20,21]. Long-distance transport to remote and Polar

Regions is also thought to occur through a combination of atmospheric and marine conveyance (Supplementary C1), enabling plastic pollutants to infiltrate and influence even the most remote and uninhabited ecosystems of our planet.

Atmospheric MnP can also affect surface climate, and therefore ecosystem health, via theorised influences on cloud formation[22], surface albedo[19] and radiative forcing[23] (Supplementary C2). Laboratory based studies demonstrate that atmospheric MnP particles are effective ice nucleation particles, and therefore might influence cloud lifetime and albedo[22,24,25]. Similarly, MnP have been modelled to cause positive and negative radiative forcing via direct effects, depending on their size and vertical distribution[23], with greater absorption of radiation (and resultant atmospheric warming) when MnP are present throughout the troposphere[23]. While these theories have been hypothesised or modelled (with notable constraints and assumptions), physical monitoring and observation studies are needed to validate and quantify MnP atmospheric influences. Critically, the only radiative forcing calculations performed to date were for non-pigmented polymers[23]. Since MnPs have diverse colours, they are hypothesised to influence surface albedo and accelerate melting of the cryosphere when deposited on snow and ice[19,26]

Beyond ecosystem health, MnPs are also an emergent pollutant of concern with regards to human health (through ingestion and inhalation)[27,28]. Potentially comparable to soot or black carbon, atmospheric MnP transported from proximal or distal sources can be inhaled or deposited on agricultural land or food preparation areas. Here they can enter the human food web (beyond plastic used in agriculture, directly added to soils, in food packaging, or occurring through marine MnP uptake by seafood)[12,29–31]. As a result this MnP forms part of the threat to global sustainability and the ability of the global community to implement all or most of the United Nations Sustainable Development Goals[32].

In this perspective we synthesize data on atmospheric MnP and propose that the atmosphere provides an important but as yet unconstrained flux of marine MnP. While atmospheric data is still limited, new studies identify several key processes that could substantially promote global transport to the oceans. Modelling suggests that there is considerable atmospheric transport of terrestrial MnP to marine environments[18,19]. Furthermore, incorporation of atmospheric MnP transport processes into marine MnP assessments indicate export of MnP to the atmosphere and potentially to terrestrial environments. To effectively quantify the marine MnP flux, it is therefore important to quantify the atmospheric compartment (emission, transport and deposition). To address this challenge, is it proposed that a collective effort is needed to better quantify and characterise the marine atmospheric MnP cycle and the roles of these processes in the marine MnP fluxes in the atmosphere, ocean and land.

2 Marine plastic cycle processes

Micro and nano plastic that is atmospherically transported to and deposited on the ocean surfaces can originate from a multitude of sources (both marine and terrestrial)[33] and can be conveyed long distances. However, quantitative assessment of atmospheric emission of MnP specific to land use type or activity is limited. This has resulted in numerous

assumptions and uncertainties in early global modelling and estimation of atmospheric MnP budgets and flux estimates.

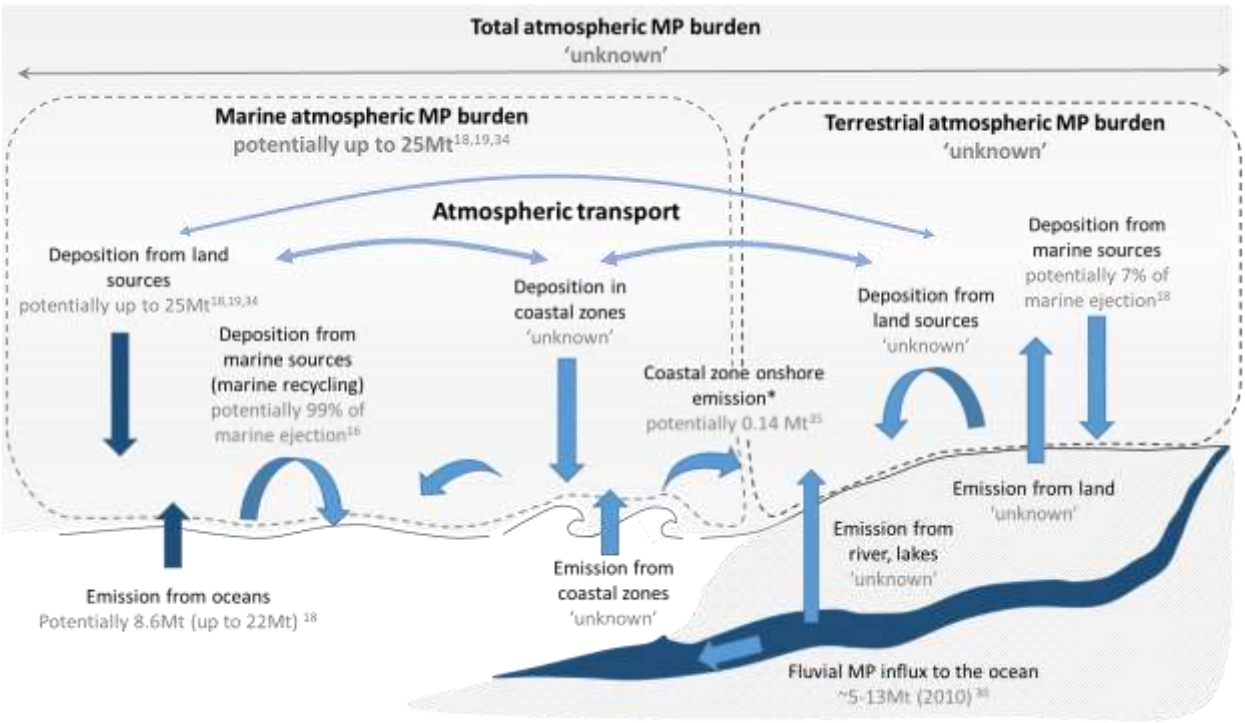

Figure 1. Atmospheric transport, potential annual flux, burdens and current knowledge gaps. The schematic illustrates the atmospheric compartment of the total dynamic microplastic (MP) cycle (in million metric tons, Mt, per year). Values are compiled from model analyses[18,19], early flux estimations[34] and reported field studies[35–37]. *The coastal zone onshore emission estimate is for localised coastal marine transport at low altitude (<200m above mean sea level)[35] and does not include long-distance transport microplastic or high altitude marine (secondary) sourced atmospheric microplastic. Atmospheric micro and nano plastic is a key part of the marine (micro and nano) plastic cycle and the calculation of the marine MnP flux.

2.1 Sources

Activities that result in atmospheric MnP creation and emission can generally be characterised as terrestrial or marine. The coastal zone can serve as a source of MnP through beach sand erosion and entrainment, sea spray and bubble burst ejection along the surf zone due to wind and waves[38]. In the coastal and open-ocean environments, MnP particles can be scavenged from the water column by bubbles and ejected into the atmosphere when the bubbles burst[35,39]. As with coastal zone processes, wind and wave action could increase the rate of ocean emission of MnP, for example along the ever-changing boundary between Arctic and Antarctic sea water and glacial ice or sea ice edge[40]. Aquaculture, coastal and offshore fishing have also been identified as a source of marine MnP[41].

The emission and (subsequent) atmospheric entrainment (the transition from surface to air followed by atmospheric transport) of agricultural soil MnPs have been quantified in the field and estimated in specific soils conditions (well sorted quartz sand, poorly-sorted organic soil, semi-arid soils)[42,43]. These studies, which focused on specific processes rather than the complex surface-atmosphere flux, suggest MnP emission of 0.08-

1.48mg/m^2/minute for relatively large microplastic particles (generally 100-200µm in size)[42,43]. It is acknowledged that there might be local or immediate (re-)deposition, but this is currently unquantified and requires further, focused research. However, if these valued are used without localised (re-)deposition considerations, and acknowledging that 11% of habitable surface is agricultural (crop) land use (11 million km^2)[44], this equates to potential global emission of 0.0009 to 0.016 million metric tons (Mt) suspended per minute when exposed to erosive wind (0.5–22m/s)[42]. During strong wind events, there is potential for atmospheric emission of agricultural MnP to extend to the region of million metric tons per year. The wind erosion and emission rate of smaller MnP still needs to be determined.

Tyre and brake wear become atmospherically emitted and entrained through road use and vehicle movement[45,46]. Early estimates suggested potential tyre emissions of ~6 tons/km/year[47]. However, published studies acknowledge the highly variable concentrations of MnP in road dust due to spatial, temporal and meteorological characteristics, road and vehicle per year conditions (for example country, season, vehicle and road maintenance). Current tyre and brake wear atmospheric emissions are suggested to be up to ≤40% of total tyre and brake wear emissions, amounting to 0.2-5.5kg per capita for particles ≤10µm[19,46]. Alternative emission estimations are based on a constant tyre wear to CO_2 ratio (0.49 mg TWP g^{-1} CO_2) or using the Greenhouse gas–Air pollution Interactions and Synergies (GAINS)[48] model estimations (<0.25-~32 tonnes per year, based on region specific, distance driven and vehicle type emission information). These different estimation techniques result in a global atmospheric flux of tyre and brake wear ranging from <0.15 to 4.3 million metric tons per year. It is important to note that many atmospheric MnP findings (MnP per m^3 or MnP per m^2) do not include tyre or brake wear particles due to analytical difficulties.

Cities and dense urban living are considered an atmospheric MnP source due to human activities (for example commerce, industry, transport, household)[45,49,50], plastic use and waste management (landfills, recycling centres, incineration)[51–55]. While there is a growing dataset of urban atmospheric MnP quantitative characterisation, the atmospheric emission rates from specific materials, actions and environments are currently unknown. Within urban environments, atmospheric MnP has been quantified from 0.9MP/m^3 (Paris outdoor air[56]) to 5700 MP/m^3 (Beijing outdoor air[57]) (Supplementary Figure 1) but without any differentiation within these samples to indicate the proportion transported to this location from a local or distal source, the proportion occurring as local emission or the quantity lost due to atmospheric transport away from the local urban source. One study has used field data extrapolation and simple transport modelling to estimate the indoor microplastic fibre contribution to marine MnP deposition, suggesting a contribution of 7-33 metric tons per year[58]. Due to the early stage in field observation and analysis of MnP source emission research, urban atmospheric MnP emission rates are very uncertain and currently based primarily on theoretical estimates.

2.2 Transport and deposition

There have been numerous quantitative observations of MnPs in remote locations where plastic pollution is attributed to atmospheric transport. These include the Ecuadorian Andes[59], French Pyrenees[17], Italian Alps[60], US conservation areas[61], snow in the Arctic[40,62], Nunavut (Canadian Arctic)[63], Isle of Helgoland (Germany)[40], Austrian and

Swiss Alps[20,40,64], the Iranian Plateau[65], and the Tibetan Plateau[66]. Atmospheric transport of MnP particles is extensive, reaching hundreds to potentially thousands of kilometres from major emission sources (for example, cities, intensive agriculture, industry). Therefore, while there is limited quantitative field observation of atmospheric MnP, the observed atmospheric transport and modelling suggest the atmosphere to contain, transport and deposit MnPs throughout the marine environment.

There is a substantial body of literature on microplastics in the environment. However, the majority of research is focused on the aquatic or terrestrial environments (855 and 366 publications respectively in 2020)[67,68]. In total, over 60 scientific studies (field or laboratory research) have been published on atmospheric MnP, of which only 5 focus on the marine environment (Supplementary Data, Google Scholar, Web of Science and Scopus search). The concentration of suspended microplastic particles in urban air range up to 5700 MP/m^3 (in Beijing[57]) and generally suggest that particle concentrations decrease with distance from city centres[69]. Marine air samples generally present lower atmospheric microplastic concentrations compared to terrestrial levels. Marine atmospheric MnP concentrations of up to 0.06-1.37 MP/m^3 have been reported over the North Atlantic Ocean, South China Sea, Indian Ocean and Western Pacific Ocean (Figure 2). However, this marine sampling comprises particles collected predominantly in the range of 20µm-5mm[70–72] (limited focus or analysis on the smaller particle size range, Supplementary Data) and is thus an underestimation. Comparatively, the Beijing and other terrestrial studies extend down to 5µm (limit of quantification), potentially resulting in relatively elevated particle counts given the increasing particle count with decreasing particle size. However, it has been shown that coastal air samples of wind in an onshore direction (blowing from the sea to the land) can carry elevated microplastic concentrations of ~2.9 MP/m^3, rising to 19 MP/m^3 during turbulent sea conditions[35]. Bubble and sea spray studies of ocean chemical species suggest that this increase in atmospheric microplastic could be due to the bubble burst ejection process and spume entrainment[73,74], where the bubble source (horizontally within the water column and spatially such as within a gyre or coastal environment) might be particularly important[18,75].

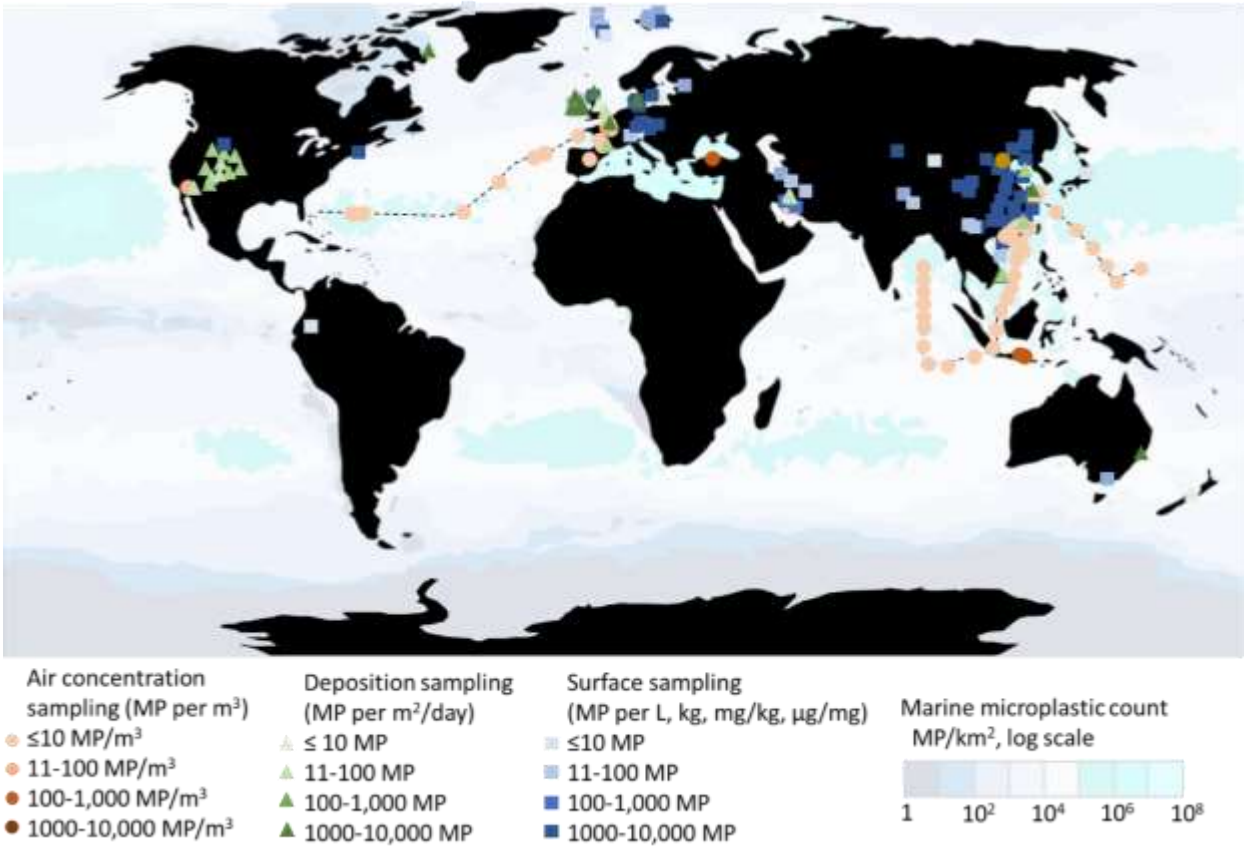

Figure 2. Summary of published micro and nano plastic atmospheric and marine research. The marine surface MnP model results are reproduced from the Van Sebille model of marine MnP (Van Sebille et al. 2015 Figure 3e)[76]. The atmospheric MP values are derived from 65 research studies published to date (Supplementary Data)[17,20,35,40,49,56,57,59–61,63–65,70–72,77–124]. It is noted that these atmospheric studies are not directly comparable due to the range of methodologies and individual studies' limits of detection but are provided here for spatial information. Full details of the atmospheric MnP studies presented are provided in the Supplementary Data. Figure 2 visually illustrates the spatial limitations of atmospheric MnP research and the need for global, comparative and standardised sampling.

The deposition of airborne MnP has been measured across a range of terrestrial environments, but has only recently been measured in the offshore marine environment in the form of deposited snow on ice floes[40]. MnP particles collected using passive deposition sampling can present different particle counts and morphology in comparison to active (pumped) air samples[56,72,81,94,125]. This might be due to the different transport processes in action (for example scavenging, settling, convective or advective transport) or the sampling methodology (active versus passive sampling, deposition versus suspended particle sampling), and is an important area of future investigation. Thus, to quantify the marine atmospheric MnP flux, both air and depositional field studies are necessary to consider the full atmospheric transport process and quantify marine MnP flux. The morphology and quantitative characterisation of marine atmospheric MnP deposition beyond these polar regions is unknown and thus marine deposition assessments are primarily theoretically modelled estimates due to lack of field data. The quantitative assessment of marine aquatic MnP particle ejection to the atmosphere and transport of these particle is also in its infancy, resulting in estimations based on limited field data.

3 Marine-atmosphere plastic flux

It is important to understand the atmosphere-ocean interactions in order to identify what size particles are being transferred and in what quantities-to quantitatively characterise this marine atmospheric limb. The atmosphere transports predominantly small micro and nano plastics compared to fluvial processes, and is a notably faster transport pathway, potentially resulting in substantial marine particle deposition and exchange between the ocean and atmosphere. Smaller micro and nanoplastics are of concern to species and ecosystem health, therefore quantifying the marine atmospheric exchange and transport process is necessary from a marine ecosystem health perspective. Conversely, quantifying the marine emission and atmospheric transport of MnPs to terrestrial environments is necessary as many remote areas, distal from terrestrial micro and nanoplastic sources, could be notably influenced by marine atmospheric MnP.

3.1 Estimates

Early estimates of the atmospheric MnP within the marine environment have been undertaken using simple extrapolation of continental data through to more dynamic atmospheric process modelling. The 2017 IUCN report suggests 15% of marine plastic pollution is wind transported (estimated primary microplastic marine pollution input of 0.8-2.5 million metric tons, therefore 0.12-0.38 million metric tons of atmospheric deposition)[126]. Acknowledging that both primary and secondary MnP particles are atmospherically transported to the marine environment, simplistic extrapolation of atmospheric MnP deposition onto the ocean surface has been carried out. Using the reported remote area atmospheric MnP deposition quantities and the global ocean surface area (3.6×10^8 km^2), microplastic deposition (particles between 1µm and 5mm in size) on the marine environment has been estimated as 10 million metric tons per year[34]. New nanoplastic deposition analysis, considering only the <200nm particle fraction, suggests that this smaller sized plastic pollution might result in up to 15 million metric tons of nanoplastic deposition on the ocean surface per year[20]. For context, 10 million metric tons is equivalent to 3% of current annual global total plastic production (2018, 359 million metric tons)[34,127], represents 11% of mismanaged plastic waste (2016, 91 million metric tons/year)[7], is comparable to the plastic (macro and micro) entering aquatic ecosystems (11-23 million metric tons per year)[7,8] and potentially transported to the marine environment (4-13 million metric tons) (2010)[36] (Figure 1).

Global model estimations have been undertaken using estimated emission rates from terrestrial (and marine) sources and current atmospheric MnP transport dynamics. Lagrangian transport and dispersion modelling (FLEXPART) of tyre and brake wear MnPs (high density polymers that form a fraction of the total atmospheric and marine plastic pollution) illustrate that >30-34% of these continental MnP particles are atmospherically transported and deposited on ocean surfaces (analysis of only MnPs ≤10µm, Figure 4)[19]. FLEXPART modelling suggests that net tyre and brake wear MnP input into the oceans via atmospheric transport and deposition could be ~0.14 million metric tons per year[19]. This is comparable to the annual quantity of tyre wear reported to enter the oceans via fluvial transport (0.064 million metric tons per year, tyres wear only)[19]. Gross atmospheric deposition and marine microplastic flux has also been globally modelled (using the

Community Atmospheric Model, CAM)[18]. The CAM estimate incorporates land based atmospheric microplastic emissions and as such has a high uncertainty due to data availability and associated assumptions. The CAM model includes ocean ejection and recirculation (resuspension) of microplastic particles, incorporating marine bubble burst ejection and wave action into the marine microplastic cycle. Gross atmospheric deposition to the ocean is estimated as 0.013 million metric tons[18]. It is important to note that the CAM model microplastic particle size distribution is notably more coarse than the FLEXPART tyre and brake wear modelling, adopting a particle size distribution generally above 5µm and focused on particles 10-50µm in size. The model suggests that potentially >11% of urban atmospheric deposition comes from sea spray or bubble burst ejection in the marine environment and that up to 99% of the total marine microplastic ejection to the atmosphere (re)deposits within the marine environment (Figure 1, Supplementary C3).

3.2 Uncertainties

These early marine flux and deposition estimates range from 0.013 to 25 million metric tons per year, illustrating the uncertainty resulting from data and research limitations. There is limited global representation of atmospheric MnP concentrations due to the limited number of studies, limited parallel air concentration and deposition studies and the limited global observation extent (Figure 2). Field data is especially scarse in the marine atmospheric environment, a lack that constrains the capacity to accurately calculate and validate estimated and modelled marine environment results of emission, deposition, marine atmospheric burden and flux. As a result, current marine atmospheric MnP understanding and flux estimations are based on available data and assumptions, resulting in large uncertainties around calculated flux and transport results.

A primary knowledge gap is the quantitative assessment of source emissions to the atmosphere, both marine and terrestrial. The quantitative characterisation of atmospheric MnP primary and secondary source emission is needed across the full temporal (all seasons and weather patterns) and spatial range (Arctic to Antarctic, remote to urban areas). Currently, atmospheric emission rates (for example particles or mass released per hour or m^2) are assumed or estimated, both in models and flux calculations due to the complexity of in field study assessment (specifically the disaggregation of background atmospheric MnP presence from the source specific emission). To advance the atmospheric flux accuracy and to understand key sources of atmospheric MnP, these emission rates require field observation and validation using advanced field sampling methods (for example horizontal and vertical array sampling across a prospective source area to define upwind and local atmospheric MnP concentrations relative to emission specific concentrations).

The understanding and experimental validation of wet removal (scavenging) of atmospheric MnP is relatively unknown. While MnPs are often considered hydrophobic, once within the environment it is unknown whether this hydrophobicity changes, for example, due to corona effects, photodegradation and weathering, or leaching of phthalates. Field and laboratory controlled studies are needed to describe changes to the microphysical behaviour of environmental MnPs as a result of environmental exposure and therefore corresponding changes to the emission, transport and deposition behaviour of these particles. Furthermore, entrainment and turbulent mixing dynamics of MnP are

also poorly understood; they are generally modelled using proxies (for example Saharan dust, or Cesium-137) or theoretical particle motions (based on particle mass, shape and density). To improve flux estimates and model outputs, laboratory and field experimentation and data are needed to adequately describe the emission, (re-)entrainment, turbulent mixing and deposition dynamics (Figure 3) of these generally negatively charged[128,129], low density, non-uniform MnP particles.

Comparability between studies is difficult at best. The wide range of sampling methods, analytical techniques and reporting standards has resulted in publication of MnP observations with differing limits of detection (LOD) or quantification (LOQ), incomparable size fractionation, differing particle characterisation (shape, polymer type) and sampling of different processes (for example snow deposition versus pumped volume of air)[3,130,131]. Atmospheric (terrestrial and marine) MnP studies need to provide comparable results to ensure data advances the understanding of source, transport, deposition and flux quantification. To achieve this, inter-method comparison studies are needed to define the method specific limitations and the relative uncertainties of each method, allowing published findings to be directly compared. For example, a sample analysed by µRaman and Nile Red fluorescence microscopy could provide similar MnP counts, but the relative uncertainties for each analytical method have not been quantified to support effective direct comparison. Early comparative studies have started to identify under or over estimations relative to specific analytical methods but without direct comparison and quantification of these uncertainties specific to particle shape, size and polymer type[132,133]. Similarly, there is an assumption that sample collection methods are accurate and effective representations of the environment or medium they sample. However, the respective comparable sampling efficiencies of deposition and air concentration collectors, and the associated uncertainties, are unquantified. For example, deposition sample collectors such as funnels connected to a collection bottle[94], petri dishes with double sided tape[104], NILU deposition collectors[103], or Brahney Buckets[134] (to name a few) have different blow-by (particle not collected due to turbulence at sampler opening resulting from sampler design or wind conditions), entrapment and retention efficiencies, resuspension and sample losses. These comparative analysis and method unknowns result in unquantifiable uncertainties in flux estimates.

Tyre and brake wear can comprise an important fraction of urban MnP pollution and might be an important component of marine atmospheric MnP[19,46]. However, in practice, these black particles can be difficult to characterise by spectroscopic methods because of limited signal due to absorption of input wavelengths and strength of vibrational response. Therefore, tyre and brake wear particle chemical characterisation is often achieved with destructive thermal degradation methods, without particle morphology characterisation[46,135]. As a result, many atmospheric MnP studies either focus on tyre and brake wear or exclude these particle types and quantify classic plastics (for example polyethylene, polypropylene, polyvinyl chloride, polyester, polyethylene terephthalate and others). This has created a disjointed dataset of MnP that does not represent the total (tyre and brake wear plus all other polymer types) MnP concentration, burden, emission or deposition. This disjoin creates uncertainty in total MnP calculations and representation (both atmospheric and marine).

3.3 Methods to advance the flux estimate

To advance the accuracy in the marine atmospheric MnP flux, greater understanding of atmospheric concentrations, deposition, emission and entrainment mechanisms and rates are needed across the global spatial and temporal range. There are numerous atmospheric processes that have not yet been quantitatively characterised or parameterised (orange processes highlighted in Figure 3) which need to be assessed to close the marine air mass balance, advance the particle flux estimation, and limit the uncertainty in flux and transport estimations. These include the vertical distribution of MnPs both on the inshore and offshore, ocean ejection of MnPs offshore, and coastal and offshore deposition.

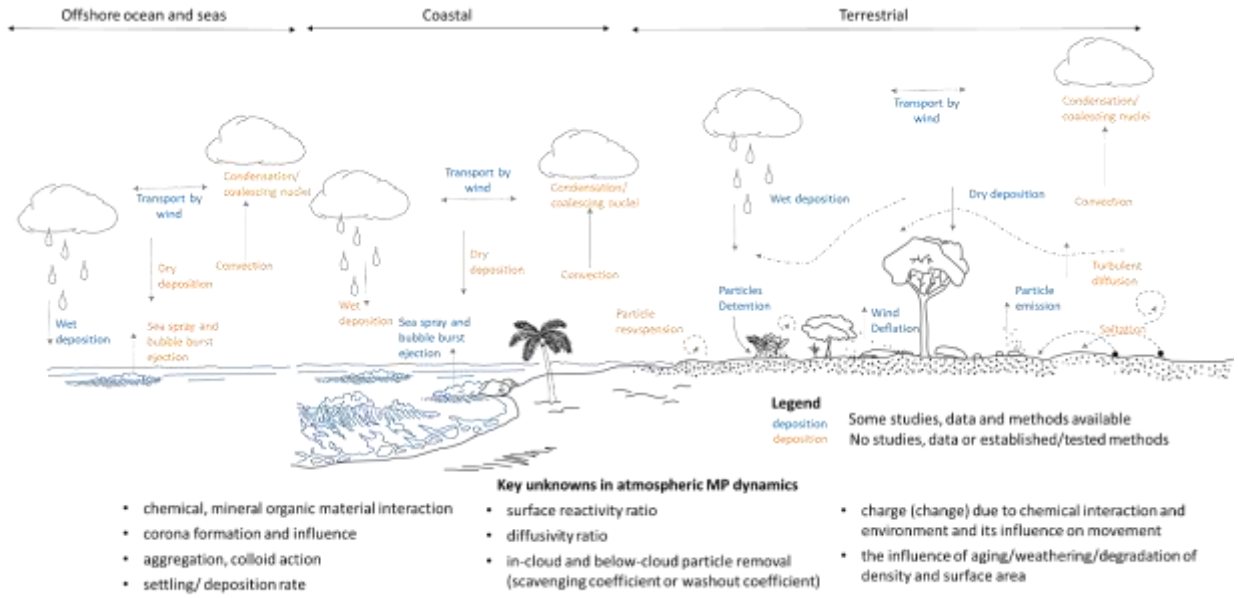

Figure 3. Key atmospheric processes that are known and unknown. Specifically, MnP processes that have been or have yet to be observed (not modelled), quantified, characterised or parameterised for MnP either in the laboratory or in the field. Understanding, quantitative characterisation and parameterisation of these processes is vital for accurate modelling of atmospheric MnP transport and accounting for field MnP findings. The processes listed are indicative, considered to be 'unknowns' in atmospheric transport but given they are untested this list is not exhaustive or prescriptive

It is a challenging task to properly sample atmospheric fluxes of MnP in any environment, but it is particularly difficult in remote marine environments. Marine atmospheric sampling (for dust and particulates, not plastic) has been undertaken using Modified Wilson and Cook samplers (MWAC), which typically collect particles >50µm (losing the smaller particle fraction)[21,136]. In addition, pump sampling devices have been mounted on buoys and ships[39,70,71]. Modified versions of these methods can be included in the array of sampling methods effective for MnP marine atmospheric research on ocean or coastal platforms[137], but field testing is needed to ensure these methods provide appropriate MnP data across the full particle size range and function in the complex marine climate (inclement weather). Method advances and innovation are needed to sample the <50µm MnP particles, especially in open-ocean and remote locations, and to provide sample methods close to the water surface.

While there are limited studies of MnP emissions produced via the bubble-burst ejection and sea spray processes[35,75], since the 2000's there has been extensive research on the mechanism of sea-salt aerosol production and other materials involved with ocean-atmosphere exchange[74,138,139]. These provide a foundation on which to base future research of ocean ejection of MnP to the atmosphere. To quantify ocean MnP emissions via bubble-burst ejection, it might be possible to use sampling methods such as the Bubble Interface Microlayer Sampler (BIMS)[140]. The BIMS was originally designed for sea salt aerosol studies, however its use is limited to calm seas. When used in conjunction with deposition measurements and pumped air sampling campaigns, a BIMS-type device could effectively advance the quantification of ocean-atmosphere MnP exchange in the field. In the laboratory, wave flumes and marine aerosol reference tanks, extensively used in sea-spray aerosol research, could provide a tool to observe and quantify the MnP wave and bubble ejection processes[141,142].

Atmospheric MnPs generally fall within the lower range of microplastics (<500µm) down to nanoplastics, a complex particle size to analyse[143,144] and within the range of concern for environmental and human health. The majority of atmospheric MnP studies are constrained by their particle counts, polymer type and shape, and limit of quantification (published down to 11µm using an FTIR or 2µm using a µRaman, but with pixel size limitations and in LOD of 10µm for FTIR, 1µm for Raman under standard analytical setup)[145,146]. Polymer identification analysis, across the full particle size range, is a vital requirement for MnP analysis and reporting[3,147,148]. Analysis of individual particles below 1µm can be achieved (for example using equipment such as Raman tweezers, AFM-IR)[143,149,150] but is resource heavy and difficult to analyse a representative proportion of a field sample. To advance the understanding and flux assessment of atmospheric marine MnPs, new techniques and advancements in technology are needed to enable submicron particle polymer analysis that provides comparable results to the micron particle studies published to date.

There is limited testing or parallel analysis of mass and particle counts to date[131,132], resulting in mass based results being mathematically converted to particle counts and vice versa, and the uncertainty associated with this mathematical estimation. Mass analysis of MnP using destructive methods (thermal degradation) is now possible for very low concentrations of nanoplastics in environmental samples[20,151]. While thermal degradation methods do not have a theoretical size limit, these methods are constrained by the minimum concentration (total mass) required to achieve detection. However, the uncertainty associated with comparative mass to particle count and particle characterisation analysis is unquantified for nano and micro plastic studies. To ensure accurate conversion of mass-particle count [35,61] and the comparability of analytical results using these different methods, comparative experimental analysis of spectroscopic and thermal degrading methods is necessary for atmospheric MnP samples.

Within the research community, it is acknowledged that reporting must be prescriptive and standardised. While it might not be possible to standardise the collection or analytical methods across individual studies and institutions, future studies need to present the following to ensure a comparable and consistent knowledge base and database of MnPs:

the limits of detection and quantification of studies (LOD and LOQ); a clear description of analytical methods to support inter-study comparison; quality assurance and control (use of field blanks and spiked sample recovery, positive and negative controls); documentation of contamination controls (clean room use, field and laboratory contamination prevention actions); method and calculations for blank correction of sample results; sample replication and individual replicate results[147,148,152,153]. While visual or graphical representation of MnP findings can be done in coarse particle increments, it is necessary for inter-study comparability that findings are presented in the smallest, consistent particle size increments possible (for example a table of 5 µm size increments provided in a study's supplementary information). Similarly, MnP particle sizes need to be presented as physical particle sizes for ecotoxicology assessment and also as aerodynamic diameters for transport modelling and inhalation studies[100,154]. Analytical methods have advanced beyond visual identification (effective to ~500µm)[155–157] and while polymer identification by thermal degradation or spectroscopy (chemical fingerprinting) methods for all particles is not always possible due to resource constraints, a minimum of 10% (ideally 30%+) of reported particles must be validated using (at least one) of these methods.

4 A global strategy

The oceans comprise over 70% of the Earth's surface, highlighting the global importance of understanding the marine atmospheric MnP cycle, transport and exchange processes. Knowledge of these processes is a prerequisite to assessing the risk posed by the atmospheric transport of MnP on species, ecosystems, and human health[158]. Individual MnP studies undertaken suggest that MnP are omnipresent over the oceans and that long-distance transport of atmospheric MnP could be a critical factor in supplying these particles to the oceans. In order to quantify these processes, we need a comprehensive, formalised global program that follows a harmonised protocol of sampling and analysis. A key objective is to provide comparable datasets that enable detailed characterisation of MnP concentrations and properties over the ocean, their temporal and spatial variability, as well as the importance of the atmospheric compartment to marine plastic pollution.

Multi-year measurements at selected long-term observation sites will identify current state and trends in atmospheric MnP concentrations. Such long-term observation activities are usually a part of a globally coordinated research or monitoring network(s) due to cost and to ensure data uniformity. An organizational approach is proposed to addressing these research needs (Box 1). These activities are broadly compartmentalized under Measurement Studies and Modelling Studies. The objective of this research organization is to ensure the identified data limitations, inter-study comparability issues and process knowledge gaps are fully addressed with specific objectives in mind. However, it is important there must be cooperation and integration across all activities.

Box 1| Proposed global network structure and coordinated international research

Measurement Studies

Monitoring Studies

Long-term (multi-year) atmospheric concentration and deposition measurements of MnP at Global Atmosphere Watch (GAW) sites (weekly or monthly composite samples continuously collected using standardised sample collection and analysis methodology, standardised LOD/LOQ)

Exploration Studies

Site specific studies from coast to offshore across a wide range of platforms and analytical methods, including:

- Ship based atmospheric sampling offshore (north and southern oceans, Arctic and Antarctic)
- Ice cores in Greenland, Antarctica, the Arctic (and other locations)
- High altitude aircraft measurements, coastal and offshore
- Marine air concentration buoy-type platform measurements

Process Studies

Emission, deposition and transport process studies (potentially including degradation, leaching, Trojan horse and other studies) and to quantitatively characterise MnP marine atmosphere dynamics, including:

- Assessment of the ocean as a source (emission and resuspension of MnP)
- Differentiated wet and dry deposition on ocean/marine surfaces
- Marine atmospheric MnP source identification
- MnP particle count to mass comparative measurement technique development

Modelling Studies

Transport

Modelling, built from the field study findings, to define the local/national/regional/global transport of atmospheric MnP in the marine (and terrestrial) environment.

Sources

Modelling to identify the potential (key) MnP sources of atmospherically transported particles found in the marine environment, remote and coastal areas.

Process specific models are also needed to quantify and detail ocean-atmosphere exchange (ocean emission or ejection).

Flux

Using global, comparable and uniform datasets that are temporally and spatially representative, global flux modelling will quantify the marine atmospheric MnP burden and flux through quantitative assessment of the full plastic cycle (emission, transport, deposition). Flux trends and responses to policy or practice changes can be derived using these models (long-term data mining and modelled forecasting).

4.1 Global long-term observation network

Early modelling of atmospheric MnP gross deposition shows considerable atmospheric deposition to the oceans, especially the Mediterranean Sea, and the North Pacific and North Atlantic Oceans (Supplementary Figure 4)[18]. However, these estimates must be used with caution since much of the deposition theoretically represents both MnP ejected from the ocean surface and transported from the terrestrial environment[18,35]. Studies looking only at tyre and brake wear show significant net atmospheric MnP deposition in the mid-and high-latitude North Atlantic, North Pacific and the northern Indian Ocean (Figure 4)[19]. These early findings, although limited to a subset of microplastic types, provide guidance in establishing location priorities in studies of the global MnP cycle.

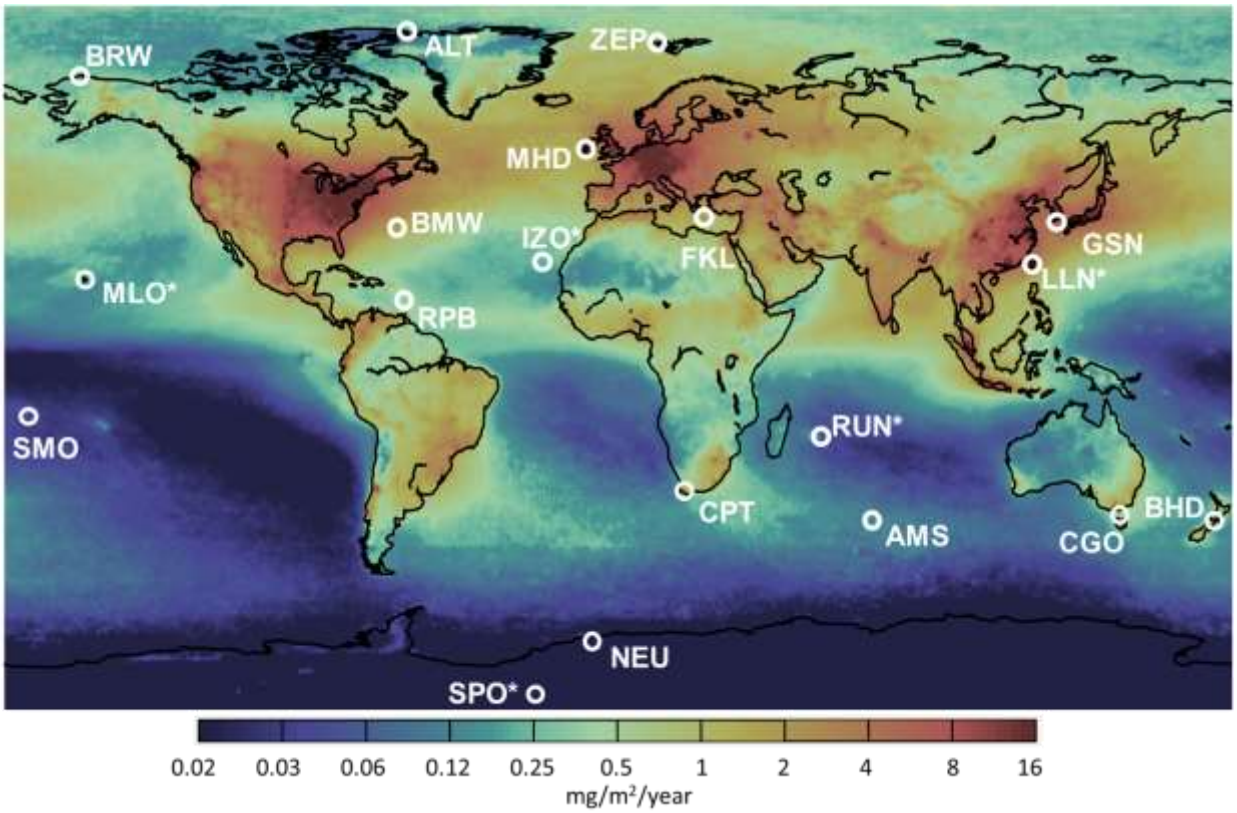

Figure 4. The proposed global observation network. Suggested potential sampling sites (primarily taken from the established WMO/GAW networks or European Monitoring and Evaluation Programme stations) illustrated on the map of FLEXPART modelled net deposition of tyre wear and brake wear particles[19] (gross global MP deposition CAM model output is provided in Supplementary Figure 4). Locations identified with * are high altitude (tropospheric) sites, all other locations are coastal monitoring sites. Potential sites are: ALT Alert (Canada); AMS Amsterdam Island (France); BHD Baring Head (NZ); BMW Tudor Hill (Bermuda); BRW (Barrow, USA); CGO Cape Grim (Australia); CPT Cape Point (South Africa); FKL Finokalia (Greece); GSN Gosan (Korea); IZO Izana (Spain, 2373 m); LLN Lulin (Taiwan, China 2862 m); MHD Mace Head (Ireland); MLO Mauna Loa (USA, 3397 m); NEU Neumayer (Antarctica); RPB Ragged Point (Barbados); RUN La Reunion (France, 2160m); SMO American Samoa (USA); SPO South Pole (Antarctica, 2841 m); ZEP Zeppelin (Norway).

To expedite these studies, it is recommend that the existing stations (Figure 4) in the World Meteorological Organization (WMO) Global Atmosphere Watch (GAW) program[159,160] be used as the initial long-term monitoring platform network[159,160]. The proposed sites are non-prescriptive but form an effective basis for a long-term observation network for atmospheric MnPs. GAW coordinates activities in a global array of fixed platforms and follows a fully developed protocol of high-quality measurements of a wide range of atmospheric composition variables, including aerosol properties[161] and of atmospheric deposition[162]. It is recommended that as part of the international effort all observational sites adopt common measurement and quality assurance protocols and centralized data reporting. At least two GAW stations have tentatively undertaken microplastics measurements. As such, the WMO/GAW program presents an ideal and cost-effective global monitoring network to commence long-term observation of atmospheric MnP.

The sites (Figure 4) are suggested based on their capacity to create multi-year time series for extended sets of variables, ranging from atmospheric constituents to atmospheric dynamics, key to MnP variability analysis. Sites located on isolated coasts or islands are ideal in that they minimize the impact from local and regional sources of MnP. The network configuration includes the most intense deposition areas as identified through early modelling effort and published field data (Supplementary C4). Figure 4 includes a selection of coastal and marine locations to ensure good coverage on a global scale, including regions where transport is potentially weak. Atmospheric MnP modelling suggests transport and deposition "plumes" downwind of North and South America, Africa, Australia and Asia[19]. Long-term observation stations are scarce in these regions and additional stations need to be added to the network (future network expansion) to represent these areas.

4.2 Observation and sampling campaigns

Long-term observations and monitoring activities are designed to provide multi-year to decadal datasets that can illustrate long-term and event specific trends and fluxes[163–167]. Past and currently active global monitoring networks studying non-plastic atmospheric substances have used a variety of sampling platforms, sampling methods, observation and monitoring campaigns. Building on this wealth of marine and atmospheric research experience, the proposed coordinated research strategy incorporates a unified and standardized long-term monitoring campaign. It is recommended weekly sampling (to yield monthly mean MnP particle quantitative particle characterisation and mass analyses), which could initially suffice for the gross characterisation of transport quantities (although it is acknowledged this for such a novel global study, adjustments will be made after initial datasets are created).

In addition to the long-term observations, complementary exploration and process studies would occur within the network. These studies would create high resolution datasets (minute, hour, daily sampling dependent on the research focus) undertaken through shorter-term intensive research campaigns using specialized equipment and platforms (for example, UAVs, BIMS). It is important that these exploration and process campaigns create data comparable with the global long-term observation dataset, therefore following (at an overview level) the basic observation outputs of the long-term dataset. The intensive research campaigns will link detailed process and event specific data and findings to specific source regions, synoptic conditions or transport processes.

The global observation network mayt take several years to develop a full description of the atmospheric MnP burden, flux and trends due to annual and inter-annual variability of conditions that affect entrainment, transport and deposition of atmospheric particles[168]. A fundamental aspect of such a monitoring network is that MnP measurements must be co-located with other observations, in particular aerosol chemical and physical properties and meteorological conditions. In the long run, fixed-point observatories in the ocean should become part of the observation network. As a part of the international efforts[160], the proposed observational sites will adopt centralized data reporting (similar to the World Meteorology Organisation dataset management).

4.3 Proposed sampling platforms

Sampling strategies to achieve long-term observations are initially proposed for fixed stations (Figure 4) using both passive deposition and active (pumped air, such as Tisch HiVol) sampling methods. These sites could include sampling towers similar to those used in the SEAREX and AEROCE networks (17-20m walk-up scaffold sampling towers equipped with elevated atmospheric samplers supported by temporary or permanent field laboratories located on both continental coast and islands at the terrestrial-marine interface)[163–166].

It is proposed that the fixed (coastal and island) long-term observations will be augmented by offshore long-term observations attained from repetitive research vessel campaigns. Research vessels often carry out repeat transits and cruises to the Arctic, Atlantic, Pacific and Antarctic waters (any sea or ocean)[167,169,170]. Such campaigns are typically 20-40 days' duration and entail frequent location changes, which enable offshore sampling over a wide spatial and temporal range (Supplementary C4). Offshore atmospheric microplastic sampling has been limited to air filter sample collection[39,70,71]. Future campaign protocols must be extended to include deposition and nanoplastic sampling. Intensive studies to quantitatively characterise the under-studied processes and environmental conditions (Figure 3) will need to use novel and innovative sampling methods, redesigned and validated specifically for MnP observation. It is expected these will include platforms and methods based on research vessels, aircraft, UAVs, buoys, or temporary sampling towers. Intensive offshore and coastal water interface sampling is novel, and initially it is recommended that methodology such as the Bubble Interface Microlayer Sampler (BIMS) (with advancements specific to MnP analysis) is used.

Low latitude air sampling, vertical and horizontal array sampling over coastal and offshore environments, can be achieved through use of unmanned aerial vehicles. Unmanned aerial vehicles (UAVs) have limitations on flight duration but can sample over extensive vertical and spatial distances provided sampling payloads are kept minimal[171,172]. UAVs are cost-effective, they sample at low airspeed and can maintain a selected altitude and location (for minutes to hours) to allow sampling of specific air masses. Furthermore, UAVs can fly close to high-risk surfaces and locations (for example, sea surface and urban areas, potentially high-emission activities) with fewer constraints. This level of control in flight path and, therefore, sample precision could be very useful for intensive air and emission source sampling in the marine environment (Supplementary C4). UAVs will enable sampling in locations where access is limited. Use of UAV could improve measurements of the overall marine atmospheric MnP burden and help to quantify ocean-atmosphere exchange.

5 Summary and future directions

There is consensus that microplastic and nanoplastic pollution can have an adverse impact on the environment and, potentially, on human health. However, despite the growing body of evidence of the importance of atmospheric MnP, there is limited marine atmospheric MnP information. MnP particles are emitted from primary and secondary sources and transported to the marine atmosphere, but the atmospheric MnP burden is also comprised of resuspended particles. Limited source emission and resuspension

studies, alongside transport and deposition studies, have resulted in high uncertainty in global-scale and marine MnP burden and flux estimations.

On review of the current state-of-the-art sampling and analysis methods it becomes evident that both sampling and analytical methodologies need to be advanced to incorporate the marine atmosphere in the plastic pollution cycle. Terrestrial atmospheric MnP sample collection methods could be implemented to effectively collect coastal and high-altitude samples but have limitations for deployment in the marine environment. Adaption and advancement of marine and terrestrial sampling methods used in aerosol and atmospheric chemistry research could provide an inroad to marine atmospheric MnP collection but require field experimentation and transport process focused studies to test their capabilities and effectiveness. Furthermore, research vessel studies currently provide low altitude air MnP concentrations but have the potential to observe a greater air column sample and ocean-atmosphere exchange if a wider range of sampling methodologies are employed (for example, UAV, BIMS, deposition collectors). To address the marine atmospheric MnP research gap, it is recommended future sampling campaigns incorporate a range of open-ocean sampling platforms and sampling methods.

In conjunction with the complexity of marine atmospheric MnP sampling, there is a need to advance analytical methods to help quantify the marine MnP flux. Current analytical methods have advanced to the point where these measurements can be reliably made but a harmonised approach is fundamental.
Despite an increasing particle count with decreasing particle size, to date the majority of analysis has focused on larger microplastic particles (>10µm), and there is limited nanoplastic analysis and unquantified uncertainties surrounding the comparison of different analytical methods. Analytical advances to enable both mass and particle characterisation of marine atmospheric MnP are necessary, with supporting detail studies to create an easy comparison between different analytical results. This will enable future studies using particle characterisation to be directly comparable to mass concentration studies and include the nano sized particle range.

Early estimates suggest that the atmospheric MnP influx to the oceans are comparable to that from rivers, however early model estimates show a huge range of uncertainty. To constrain the uncertainties and provide clear representation of the marine MnP flux it is recommended an expanded and coordinated global-scale research effort be undertaken. We propose a global observation network built upon existing long-term monitoring platforms to create a baseline and trend analysis dataset, augmented with intensive, short-term monitoring and experimentation research focused on specific processes, events or locations. Looking forward, it is recommended the global monitoring effort expands to include research vessels and open-ocean observation as well as inland waterbodies and estuary sites.

We expect that after a several years of network operations, we will be able to identify the key locations, processes and sources of MnP that impact the marine environment. Conversely, this research will also demonstrate the influence and relative importance of emissions from the marine environment influencing the terrestrial atmospheric MnP

burden. This improved understanding of MnP flux and the global plastic cycle will be vital for evaluations of the success of urgently needed mitigation strategies against plastic pollution. The information is also vital to inform risk assessments for humans and the biosphere, which need to be based on realistic environmental micro- and nanoplastic concentrations.

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Title: The Atmospheric Cycle of Microplastics in the Marine Environment

Supplementary Information

Commentary C1: The importance of atmospheric micro(nano)plastic transport to remote and polar regions

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

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

There is significant evidence of microplastic in the Arctic and Antarctic seas, suggesting that oceanic currents are one key source of Arctic plastic pollution[1,2] in the polar marine environment. However, while polar ice acts as a sponge, collecting marine plastic during its ice formation[3,4], the microplastic found on the surface of ice flows and snow are not directly attributable to marine conveyance. Notable microplastic quantities have been found on Arctic surfaces[5], and early studies have identified microplastic in the Antarctic atmosphere. It is suggested that these particles were either transported long-distance through the atmosphere prior to polar deposition, or were conveyed via marine currents then emitted during ocean turbulence (e.g. through the bubble burst ejection process) and atmospherically transported to the polar surface (land, glaciers or sea ice). Characterising and quantifying the atmospheric transport contribution to remote area and polar micro(nano)plastic is therefore necessary to both understand the remote areas transport processes and to model plastic pollution influx (and future scenarios) to these relatively pristine and remote locations.

Commentary C2: Atmospheric microplastics and climate change

Research on micro(nano)plastic in the context of climate change is extremely limited to date. Life cycle and greenhouse gas (GHG) emission analyses show micro(nano)plastic to contribute to GHG, climate change/global warming potential and photochemical ozone formation/ozone formation[6,7]. Micro(nano)plastic lost to the environment release GHG and interfere with ocean carbon fixation. Early model estimations suggest current GHG from plastic from cradle to grave (incorporating material extraction, production, manufacture, transport and waste management) of 1.34 gigatons CO_2 equivalent/year by 2030, and by 2050 may consume 10-13% of the remaining carbon budget[7].

It has been hypothesised that deposition of brake wear and tyre wear particles on ice and snow may accelerate warming of the cryosphere[8]. Micro(nano)plastic particles, particularly black coloured particles, may function as cryoconites, increasing the snow and ice melt in polar and high elevation locations. Given their hydrophobic nature, microplastic and nanoplastic may act as cloud ice nuclei[9]. Certain regions may therefore be particularly sensitive to the presence of airborne micro(nano)plastic, such

as the Southern Ocean, where cloud albedo is strongly linked to the concentration of ice-nucleating particles[10]. Southern high latitudes have experienced enhanced UV fluxes in recent decades owing to the Antarctic ozone hole; this is thought to enhance the formation of micro(nano)plastics in surface waters and terrestrial environments at southern high latitudes[11].

Airborne particulate matter influences surface climate by absorbing and scattering solar and terrestrial radiation. While total aerosol number concentrations range between 1×10^9 – 1×10^{10} per m^3 over Europe and East Asia[12], the concentrations of airborne micro(nano)plastics in the same regions are much lower; between 0.01 – 5000 MP/m^3.[13,14] Airborne microplastics therefore make only a small contribution to total aerosol abundances in the present-day atmosphere. Electromagnetic scattering and absorption calculations indicate that non-pigmented microplastics are efficient at scattering solar (shortwave) radiation, and at absorbing terrestrial (longwave) radiation[15]. Global climate model simulations show that the balance between shortwave effects (which imply a cooling influence on Earth's surface) and longwave effects (i.e., the greenhouse effect) depends strongly on the assumed vertical distribution of airborne microplastics[15]. The effective radiative forcing (ERF), assuming direct aerosol-radiation interactions only (that is, neglecting aerosol-cloud interactions) is calculated to be on the order of -0.746 – 0.044 mW m^{-2} assuming a surface concentration of 1 MP m^{-3}. Compared with the total aerosol effective radiative forcing of -0.71 to -0.14 W m^{-2} due to aerosol-radiation interactions[16], the microplastic ERF is small in the present-day atmosphere.

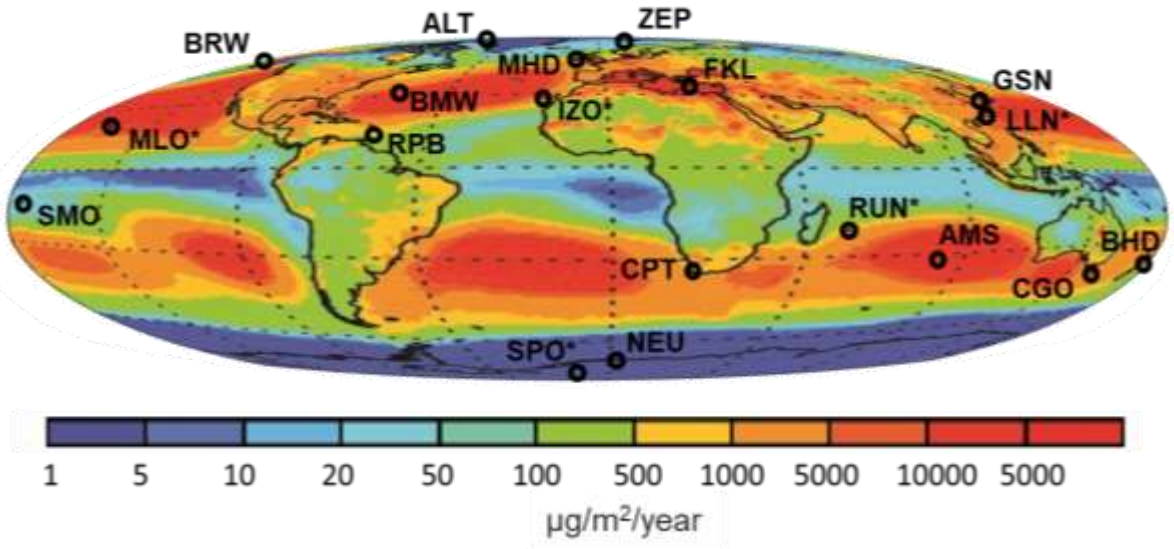
The expected increase in atmospheric micro(nano)plastic abundances due to increasing global plastic use and mismanaged plastic waste will increase microplastic ERF in future, and may influence local and regional climate in regions where airborne micro(nano)plastic concentrations are particularly large. Furthermore, emissions of anthropogenic aerosols are projected to decrease in future, which is linked to expected improvements in air quality[17]. Micro(nano)plastics will therefore make a larger relative contribution to total aerosol ERF in future.

Commentary C3: CAM model global gross microplastic deposition and the global monitoring network

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

The results of the CAM model are notably different from those presented for FLEXPART modelled tyre and brake wear global *net* deposition[8]. This is partly due to the inclusion of marine emission in the CAM model whereas Figure 4**Error! Reference source not found.** FLEXPART models net deposition to the marine environment (no marine emission) therefore providing an insight into the terrestrial net influx of microplastic to the marine environment. The microplastic particle size included in the CAM model also differs from that used in the FLEXPART model, with the CAM model particle size extending from 250µm to 4µm while the FLEXPART model considered particles of either <10µm or <2.5µm (PM10 or PM2.5). As presented Figure C3.1 the quantity of microplastic particles increases significantly with decreasing particle size. Thus the FLEXPART model may incorporate a significantly greater number and mass of smaller, more easily atmospherically transported microplastic particles than the CAM model. It is also important to note the CAM model adopts a global road microplastic emission rate (all polymers in road dust, not just tyre and brake wear) of 96Mt/yr rather than the 284MT/yr used in the FLEXPART model[18]. These highlighted differences illustrate the need for better harmonization and compatible parameterisation and field work to characterise atmospheric micro(nano)plastic particles and transport dynamics to enable advancement of atmospheric micro(nano)plastic modelling.

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



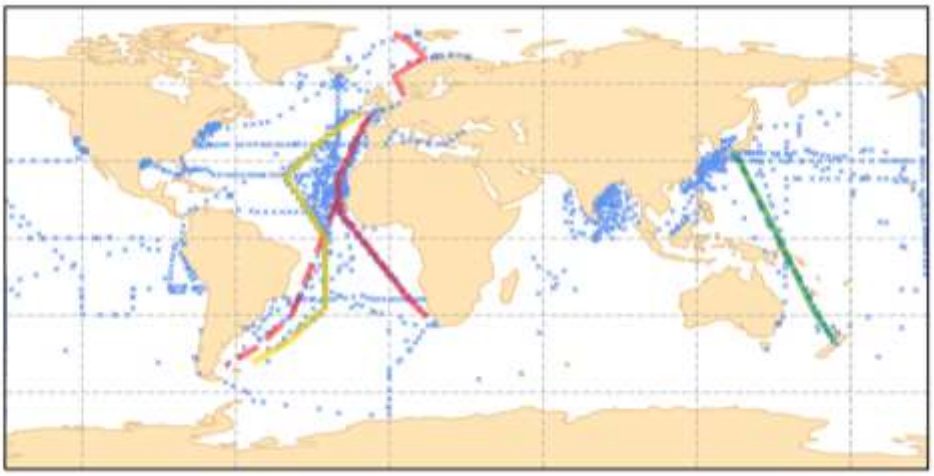
Supplementary Figure 3.1 Possible sampling sites illustrated on maps of CAM modelling of the total atmospheric microplastic deposition (all polymer types)[18]. Locations identified with * are high altitude

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

Commentary C4: Marine atmospheric sampling platforms

Marine Research Vessels

Ships provide access to regions of the ocean that are otherwise inaccessible, functioning with a typical campaign of 20-40 days. While ship-based research (primarily research vessels) can provide sampling from currently unmonitored marine atmospheric locations, the individual voyages result in a dataset that may be spatially and temporally patchy[19] and therefore climatologically less representative. Longer-term monitoring research campaigns such as GEOTRACES[20], the Atlantic Meridional Transect Programme[21] and Alfred Wegener Institute ship-based research program (e.g. RV Polarstern research campaigns)[22] can however provide vital offshore marine atmospheric information and be a valuable monitoring platform when employed multi-annually (potentially also seasonally) over a specific transect. To date only pumped aerosol samplers have been used on vessels, providing data of atmospheric composition (only three marine cruises have sampled and published marine atmospheric microplastic to date (Figure 2, Figure C4.1) across the Atlantic, Pacific Antarctic and Arctic oceans primarily[13,23,24]. In general, rainfall and dry deposition is grossly under-sampled over the oceans considering its importance to the air-to-sea transfer of material to the global ocean, with no precipitation or dry deposition yet undertaken specifically for micro(nano)plastic analysis.



Supplementary Figure 4.1. Locations of aerosol samples collected from UK, French, German, Dutch, New Zealand, Australian and Japanese ships and analysed at UEA from 2000 to 2019 (n = ~2070) (blue)[19]. Atlantic Meridional Transect cruises are shown in yellow, *Polarstern* inter-hemisphere transfers and Arctic campaigns in red and *Transfuture5* voyages in green.

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

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

Fixed coastal or island platforms

Two notable fixed platform networks have been used to analyse the continental aerosol contribution to the marine environment over the past four decades. The SEAREX Ocean Aerosol Sampling Network was designed to characterise marine atmospheric chemistry and the role of continental to marine (ocean) transport within the (western) Pacific Ocean[25]. The AEROCE Network (interlinked with several World Meteorological Organisation observatories and Global Atmosphere Watch Programme(GAW)) undertook similar research extending along the east and west of the central to north Atlantic Ocean. The SEAREX and AEROCE networks used 17-20m walk-up scaffold sampling towers equipped with elevated atmospheric samplers supported by temporary or permanent field laboratories located on both continental coast and islands at the terrestrial-marine interface. Long-term monitoring occurred, including year-round measurements linked to the specific synoptic meteorological conditions, in conjunction with short-term intensive research activities that addressed focused research questions (e.g. sea salt aerosol bubble burst emission quantification).

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

Aircraft, UAV, buoys

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

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

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

Buoys have been successfully used for marine atmospheric sampling, monitoring dust and atmospheric composition in static marine locations at low (near sea surface) elevations[37]. Buoys may provide a complementary long-term static marine sampling network complementary to fixed platforms and shipping vessel sampling, providing

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

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

Platform Type	Advantages	Disadvantages
Ships	• 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)	• Short-term (days or weeks to several months) temporal snapshots • Very dirty environment • May travel at speeds resulting in spatially ranging sample representation
Sailing vessels	• 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	• Short to moderate temporal snapshots • Fewer personnel accommodated than ships • Limited on-board analysis
Island/coastal sites *WMO/GAW permanent sites*	• Synoptic, seasonal and annual variability • Trained personnel • Supporting chemical/ meteorological measurements • Multiple elevations	• Limited geographical locations
Island/coastal sites *Other permanent sites*	• Synoptic, seasonal and annual variability • Possible supporting chemical/ meteorological measurements	• Limited trained personnel • Limited geographical locations
Island/coastal sites *Non-permanent sites*	• Synoptic and possibly seasonal scale variability • Possible supporting chemical/meteorological	• Limited/untrained personnel • Limited geographical locations

	measurements (in some cases)	
Aircraft	• Unlimited marine locations • Trained personnel • Multiple elevations	• Very short-term temporal snapshots • Limited sampling intervals • Very expensive
Drones & UAVs	• Multiple elevations • Relatively Low cost	• Limited marine locations unless launched from ships • Very short-term temporal snapshots • Limited sampling intervals • Limited power availability and payload
Tethered or remotely controlled balloons	• Full range of elevation (surface-PBL-troposphere) • Generally limited to terrestrial release but potentially possible from ships	• 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)
Buoys	• Synoptic, seasonal and annual variability • Possible wide geographical coverage	• Difficult to service • Possible limited power • Excessive sea spray

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