Microplastics and nanoplastics in the marine-atmosphere environment

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Abstract

The discovery of atmospheric micro(nano)plastics transport and ocean–atmosphere exchange points to a highly complex marine plastic cycle, with negative implications for human and ecosystem health. Yet observations are currently limited. In this Perspective, we quantify the marine-atmospheric micro(nano)plastics cycle processes and fluxes, with the aim of highlighting the remaining unknowns in atmospheric micro(nano)plastics transport. Between 0.013 and 25 million metric tons per year of micro(nano)plastics are potentially being transported within the marine atmosphere and deposited in the oceans. However, the high uncertainty in these marine-atmospheric fluxes is related to data limitations and a lack of study intercomparability. To address the uncertainties and remaining knowledge gaps in the marine-atmospheric micro(nano)plastics cycle, we propose a future global marine-atmospheric micro(nano)plastics observation strategy, incorporating novel sampling methods and the creation of a comparable, harmonized and global dataset. Together with long-term observations and intensive investigations, this strategy will help to define the trends in marine-atmospheric pollution and any responses to future policy and management actions.

Editor's Summary

Atmospheric transport of microplastics could be a major source of plastic pollution to the ocean, yet observations currently remain limited. This Perspective quantifies the known budgets of the marine-atmospheric micro(nano)plastics cycle and proposes a future global observation strategy.

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Introduction

Over 368 million metric tons (Mt) of single-use plastic were created in 2019 AQ1 AQ2 AQ3 (refs[1,2]) and this is projected to increase AQ4 further owing to rapid and inexpensive plastic production, non-circular economic models and a culture of single-use plastics. Plastic pollution has been found in all environmental compartments, ~~including aquatic,~~from water to soil and air[3,4,5,6]. Projections indicate that plastic pollution will treble by 2040 under a business-as-usual scenario, up to 80 Mt of waste per year (based on 2016 environmental plastic pollution estimates)[7]. Of all the managed and mismanaged plastic waste created, around 12% is projected to enter the aquatic environment and around 22% to enter the terrestrial environment, with an estimated 60 Mt per year lost to just the aquatic and terrestrial environmental compartments by 2030[7,8]. However, there is currently limited assessment of the atmospheric compartment. AQ5 AQ6 AQ7 AQ8

The global oceanic microplastics This "(MP see Box 1 for definitions)" cannot be put here in this first sentence as it makes the sentence illegible and we will loose non specialist readers at this point because of this lack of legibility and flow. Please move it to the next 'microplastic', as illustrated in the edits.

1 delete please ~~(MP; see Box~~ ~~for definitions)~~ cycle[9,10] is currently quantified based on observational and modelled data of ~~MP~~microplastics (MP; see Box 1 for definitions) in marine and fresh water, biota and sediments, as these environments are frequently studied[11,12,13]. Terrestrial runoff, river discharge and marine currents carry micro(nano)plastics (MnP; see Box 1 for definitions) from terrestrial sources to distal areas such as the Arctic, Antarctic and deep-sea locations over months to years[14]. Although it is relatively slow, this mechanism is important in transporting MnP to remote areas, where they can negatively impact marine life[15,16]. Although it is less studied, atmospheric transport research similarly illustrates that wind can transport MnP at trans-continental and trans-oceanic scales[17,18,19,20]. Atmospheric transport is comparably much faster than oceanic transport, 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 could occur through a combination of atmospheric and marine conveyance (Supplementary Note 1), enabling plastic pollutants to infiltrate and influence even the most remote and uninhabited ecosystems of the Earth.

Atmospheric MnP can also affect surface climate, and therefore ecosystem health, via theoretical influences on surface albedo[19], cloud formation[22] and radiative forcing[23] (Supplementary Note 2). ~~Although~~ MnP have diverse colours~~, they~~ and are hypothesized to influence surface albedo and accelerate cryosphere melting when deposited on snow and ice[19,24]. In addition, laboratory-based experiments demonstrate that atmospheric MnP particles are effective ice-nucleation particles, potentially influencing cloud lifetime and albedo[22,25,26]. Similarly, MnP have been modelled to cause positive and negative radiative forcing via direct effects, depending on their size and vertical distribution[23]. For example, greater radiation absorption and resultant atmospheric warming occurs when MnP are present throughout the troposphere[23]. These theories have been hypothesized or modelled (with notable constraints and assumptions), but physical monitoring and observation studies are urgently needed to validate and quantify MnP atmospheric influences. Critically, the only radiative-forcing calculations performed so far were for non-pigmented polymers[23].

Beyond ecosystem health, MnP are also an emergent pollutant of human health concern through ingestion and inhalation[27,28]. Potentially comparable to soot or black carbon, atmospheric MnP transported from proximal or distal sources can result in human exposure through direct inhalation and via the human food web through deposition on agricultural land and water reservoirs, inclusion or contamination during agricultural, food manufacturing and preparation activities. This atmospheric MnP is in addition to other sources of plastic widely used in agriculture, directly added to soils, used in food packaging, or ingested by seafood[9,29,30,31]. As a result, atmospheric 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 current atmospheric MnP data and propose that the atmosphere provides an important but unconstrained flux of marine MnP. Although atmospheric data is still limited, several studies have identified key processes that could

substantially promote global transport of terrestrial MnP to marine environments[18,19]. Furthermore, the incorporation of atmospheric MnP transport processes into the marine MnP cycle highlights the importance of marine MnP export to the atmosphere and potential transportation to terrestrial environments. Therefore, it is important to quantify the atmospheric compartment (emission, transport and deposition) to obtain an accurate estimate of marine MnP fluxes. A collective effort is needed to better quantify and characterize the marine-atmospheric MnP cycle, so that the roles of MnP in the atmosphere, ocean and land can be more fully understood.

Box 1 Key micro(nano)plastic terminology definition and descriptions

Microplastics (MP)

Plastic particles equal to or greater than 1 μm and less than 5 mm in (aerodynamic) diameter[9,10,131,132].

Nanoplastics (NP)

Plastic particles less than 1 μm in (aerodynamic) diameter[9,10,131,132].

Micro(nano)plastics (MnP)

All plastic particles ≤5 mm in diameter (both MP and NP)[131,132,133]. Amounts of MP and NP are measured in the atmosphere as particles or mass per volume of sampled air (in, for example, MP m^{-3}); and their deposition is measured as particles or mass per surface area sampled over a specified duration (for example, MP m^{-2} per day).

Primary MnP

MP manufactured to be 1 μm to 5 mm in diameter (for example, nurdles[134], personal care products[135], textiles[136]).

NP manufactured to be <1 μm in diameter (for example, medical applications[137], printing ink[138], electronics[107,139,140]).

Secondary MnP

MP or NP produced through mechanical, chemical or photodegradation (for example, plastic bottle breakdown to MP and NP on a beach owing to ultraviolet radiation, salt and wave action)[107,141,142,143].

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 (for example, wastewater treatment plant release to a receiving waterway, recycling plant emission caused by mechanical plastic deconstruction, or plastic factory emission due to production activities)[144,145,146].

Diffuse source

MP or NP emission (and re-emission) from activities that have no single emission time and location (for example, road dust or agricultural emissions)[144,145,147,148,149].

Marine plastic cycle processes

MnP 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 a land-use type or activity is limited. This lack of quantification has resulted in numerous assumptions and uncertainties in global modelling and estimation of atmospheric MnP budgets and flux estimates. This section discusses what is known and unknown regarding the sources, transport and deposition of marine-atmospheric MnP.

Sources

Activities that result in atmospheric MnP creation and emission can generally be characterized as terrestrial or marine (Fig. 1). Marine emission of MnP to the atmosphere is an emerging field of research and formative investigation in the field and laboratory point towards MnP ocean–air interface exchange. As such, the coastal zone is thought to serve as a source of MnP through beach sand erosion and entrainment, sea spray and bubble burst ejection along the surf zone caused by wind and waves[34,35,36]. In the coastal and open-ocean environments, MnP particles could be scavenged from the water column by bubbles and ejected into the atmosphere when the bubbles burst[37,38]. 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 edges[39] (Fig. 1). Aquaculture, coastal and offshore fishing have also been identified as a source of marine MnP[40].

Fig. 1

Atmospheric microplastics transport, potential annual flux, burdens and current knowledge gaps.

The atmospheric compartment of the total dynamic microplastics cycle (in million metric tons, Mt, per year) AQ9 can be separated into

burden values are compiled from model analyses[39,79], study flux estimations[38] and reported field studies[37,80,129]. *The coastal zone onshore emission estimate is for localized coastal marine transport at low altitude (<200 m above mean sea level)[37], and does not include long-distance transport microplastics or high-altitude marine (secondary) sourced atmospheric microplastics. Atmospheric micro- and nanoplastics are a key part (potentially up to 25 Mt) of the marine micro(nano)plastics cycle and the calculation of the marine micro(nano)plastics flux.

The emission and (subsequent) atmospheric entrainment (the transition from surface to air followed by atmospheric transport) of agricultural soil MnP have been quantified in the field and estimated in specific soils conditions (well sorted quartz sand, poorly sorted organic soil, semi-arid soils)[41,42]. These studies, which focused on specific processes rather than the complex surface–atmosphere flux, suggest a MnP emission of 0.08–1.48 mg m^{-2} per minute for relatively large MP particles (generally 100–200 µm in size)[41,42] (Box 1). It is acknowledged that there might be local or immediate (re-)deposition, but this is currently unquantified and requires further, focused research. However, if the values are used without localized (re-)deposition considerations,. **AQ10** Aacknowledging that 11% of habitable surface is agricultural (crop) land use (11 million km^2)[43], a global emission of 0.0009 to 0.016 Mt suspended per minute can be estimated when agricultural land is exposed to erosive wind (0.5–22 m per second)[41]. During strong wind events, there is potential for atmospheric emission of agricultural MnP to extend to the region of millions of 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[44,45]. Early estimates suggested potential tyre emissions of aboutpproximately 6 t km^{-1} per year[46]. However, published studies acknowledge the highly variable concentrations of MnP in road dust, owing to spatial, temporal and meteorological characteristics, and road and vehicle or traffic per yearintensity conditions (such as, country, season, vehicle type and road maintenance). **AQ11** Current tyre and brake wear atmospheric emissions are suggested to be up to 40% of total tyre and brake wear emissions, amounting to 0.2–5.5 kg per capita for particles ≤10 µm (refs[19,45]). Alternative emission estimations are based on a constant ratio of tyre wear particles (TWP) to CO_2 (0.49 mg TWP per gram of CO_2) **AQ12** or using the Greenhouse gas–Air pollution Interactions and Synergies (GAINS)[47] model estimations (<0.25 to around **AQ13** 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 Mt per year. It is important to note that many atmospheric MnP findings (MnP m^{-3} or MnP m^{-2}) do not include tyre or brake wear particles, owing to analytical difficulties.

Cities and dense urban living are considered an atmospheric MnP source due to human activities (such as in industry, transport and residential livinghouseholds)[44,48,49], plastic use and waste management (such as landfills, recycling centres and incineration)[49,50,51,52,53]. Although there is a growing dataset of urban atmospheric MnP quantitative characterization, the atmospheric emission rates from specific materials, actions and environments are currently unknown. Within urban environments, atmospheric MnP have been quantified from 0.9 MP m^{-3} (Paris outdoor air[54]) to 5,700 MP m^{-3} (Beijing outdoor air[55]) (Fig. 2, Supplementary Data). However, these estimates were reported without any differentiation to indicate the proportion of MnP transported to each location from a local or distal source, or 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 MP fibre contribution to marine MnP deposition, suggesting a contribution of 7–33 t per year[56]. Owing to the early stage in field observation and MnP source emission research, urban atmospheric MnP emission rates are very uncertain and currently based primarily on theoretical estimates.

Fig. 2

Summary of published micro(nano)plastics atmospheric and marine research.

The marine surface micro(nano)plastics (MP) count **AQ14** (blue-grey base map) **AQ15** is reproduced from the Van Sebille model[130]. The atmospheric MnP sampling sites and values (circles represent air concentrations, triangles represent deposition and squares represent

these atmospheric studies are not directly comparable due to the range of methods and limits of detection of the individual studies; the results are provided here for spatial information. We note that the majority of sampling sites are located in North America, Europe, the Middle East and east Asia, with very little sampling elsewhere around the world. The map shows the spatial limitations of atmospheric MnP research, which highlights the need for global, comparative and standardized sampling.

Microplastics and nanoplastics in the marine-atmosphere environment

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Transport and deposition

There have been numerous quantitative observations of MnP in remote locations where plastic pollution is attributed to atmospheric transport. These include the Ecuadorian Andes[57], the French Pyrenees[17], the Italian Alps[58], US conservation areas[59], snow in the Arctic[39,60], Nunavut (Canadian Arctic)[61], Isle of Heligoland (Germany)[39], the Austrian and Swiss Alps[20,39,62], the Iranian Plateau[63], and the Tibetan Plateau[64]. 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, although there is limited quantitative field observation of atmospheric MnP, the observed atmospheric transport and modelling suggest that the atmosphere contains, transports and deposits MnP throughout the marine environment.

There is a substantial body of literature on MP in the environment. However, most research is focused on the aquatic or terrestrial environments (855 and 366 publications, respectively, in 2020)[65,66]. In total, over 70 published scientific studies (field or laboratory research) are on atmospheric MnP, of which only six focus on the marine environment (Supplementary Data, ~~Google Scholar, Web of Science and Scopus search~~). The concentration of suspended MP particles in urban air range up to 5,700 MP m^{-3} (in Beijing[55]) and studies generally suggest that particle concentrations decrease with distance from city centres[67].

Marine air samples generally present lower atmospheric MP 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, the South China Sea, the Indian Ocean and the Western Pacific Ocean (Fig. 2). However, this marine sampling comprises particles collected predominantly in the range 20 µm to 5 mm (refs[68,69,70]) (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 5 µm (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 MP concentrations of around 2.9 MP m^{-3}, rising to 19 MP m^{-3} during turbulent sea conditions[37]. Bubble and sea spray studies of ocean chemical species suggest that this increase in atmospheric MP could be due to the bubble burst ejection process and spume entrainment[71,72], where the bubble source (horizontally within the water column and spatially such as within a gyre or coastal environment) might be particularly important[18,73].

The deposition of airborne MnP has been measured across a range of terrestrial environments, but publication of marine MnP offshore measurements of air[69] and MnP-deposited snow on ice floes[39] only commenced in 2019 (Supplementary Data). MnP particles collected using passive deposition sampling can present different particle counts and morphology compared to active (pumped) air samples[54,70,74,75,76]. This difference 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.

To quantify the marine-atmospheric MnP flux, both air and depositional field studies must consider the full atmospheric transport process and quantify marine MnP flux. The morphology and quantitative characterization of marine-atmospheric MnP deposition beyond these polar regions are unknown, and thus marine deposition assessments are primarily theoretically modelled estimates owing to the lack of field data. The quantitative assessment of marine aquatic MnP particle ejection to the atmosphere and transport of these particles is also in its infancy, resulting in estimations based on limited field data. Thus, while our current understanding of atmospheric MnP in the marine environment identifies the cyclic nature of MnP movement (ocean–atmosphere flux) the quantification of this flux (deposition, emission and atmospheric concentrations) requires substantial further study.

Marine-atmospheric plastic flux

It is important to understand the atmosphere–ocean interactions to identify what sizes of particle are being transferred and in what amounts, so that the marine-atmospheric limb can be quantitatively characterized. The atmosphere transports predominantly small MnP 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 MnP (Box 1) are also of concern to species and ecosystem health, and so quantifying the marine-atmospheric exchange and transport process is necessary to monitor marine ecosystem health. Conversely, quantifying the marine emission and atmospheric transport of MnP to terrestrial environments is necessary, given that many remote areas, distal from terrestrial MnP sources, could be notably influenced by marine-atmospheric MnP. In this section, the estimates, uncertainties and future improvements in marine-atmospheric fluxes are discussed (Fig. 3).

Fig. 3

Critical known and unknown atmospheric processes.

Shown are some micro(nano)plastics processes that have been (blue) or have yet to be (orange) observed (not modelled), quantified, characterized or parameterized for MnP either in the laboratory or in the field. However, there remain several critical unknown processes in atmospheric MnP dynamics, which include: chemical, mineral and organic material interactions; corona formation and influence; aggregation and colloid action; settling **AQ16** and deposition rates; surface reactivity ratios; diffusivity ratios; in-cloud and below-cloud particle removal; charge change due to chemical interaction with the environment and influence on particle movement; and the influence of ageing, weathering, degradation of density and surface area. These listed 'unknown' processes are indicative, but given **AQ17** that they are untested, this list is not exhaustive or prescriptive. Understanding, quantitative characterization and parameterization of atmospheric MnP processes is vital for accurate modelling of atmospheric MnP transport and accounting for field MnP findings.

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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 that 15% of marine plastic pollution is wind-transported (estimated primary MP marine pollution input of 0.8–2.5 Mt, and therefore 0.12–0.38 Mt ~~of~~by atmospheric deposition)[77]. 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), MP deposition (particles between 1 µm and 5 mm in size) on the marine environment has been estimated to be 10 Mt per year[78]. New NP deposition analysis, considering only the <200 nm particle fraction, suggests that this smaller-sized plastic pollution might result in up to 15 Mt of NP deposition on the ocean surface per year[20]. For context, 10 Mt is equivalent to 3% of current annual global total plastic production (2018, 359 Mt)[78,79], represents 11% of mismanaged plastic waste (2016, 91 Mt per year)[7], is comparable to the MnP entering aquatic ecosystems (11–23 Mt per year)[7,8] and ~~is~~plastic potentially transported to the marine environment (4–13 Mt) (2010)[80] (Fig. 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 MnP (high-density polymers that form a fraction of the total atmospheric and marine plastic pollution) illustrate that >30–34% of these continental

FLEXPART modelling suggests that microplastics and nanoplastics in the marine-atmosphere environment transport and deposition could be around 0.14 Mt per year[19]. This is comparable to the annual quantity of tyre wear reported to enter the oceans via fluvial transport (0.064 Mt per year, tyre wear only)[19]. Gross atmospheric deposition and marine MP flux has also been globally modelled (using the Community Atmospheric Model, CAM)[18]. The CAM estimate incorporates land-based atmospheric MP 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 MP particles, incorporating marine bubble burst ejection and wave action into the marine MP cycle. Gross atmospheric deposition to the ocean is estimated as 0.013 Mt (ref.[18]). It is important to note that the CAM model MP particle size distribution is notably more coarse than the FLEXPART tyre and brake wear modelling, adopting a particle size distribution that is generally above 5 µm and focused on particles 10–50 µm 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 MP ejection to the atmosphere (re)deposits within the marine environment (Fig. 1, Supplementary Note 3).

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The proposed global observation network.

Suggested potential sampling sites (primarily taken from the established WMO and/or GAW networks or European Monitoring and Evaluation Programme stations) illustrated on the map of FLEXPART modelled net deposition of tyre wear and **AQ18** brake wear particles[19] (gross global MP deposition CAM model output is provided in Supplementary Fig. 4). Locations identified with an asterisks (*) 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, 2,373 m); LLN*, Lulin (Taiwan, China, 2,862 m); MHD, Mace Head (Ireland); MLO*, Mauna Loa (USA, 3,397 m); NEU, Neumayer (Antarctica); RPB, Ragged Point (Barbados); RUN*, La Reunion (France, 2,160 m); SMO, American Samoa (USA); SPO*, South Pole (Antarctica, 2,841 m); ZEP, Zeppelin (Norway). The proposed global observation network and suggested sampling sites would provide global, comparative atmospheric MnP observations. Figure adapted from ref.[19], CC BY 4.0.

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Uncertainties

These early marine flux and deposition estimates range from 0.013 to 25 Mt 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 (Fig. 2). Field data is especially scarce 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 characterization of atmospheric MnP primary and secondary source (Box 1) 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 per square metre) 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

source-specific emission). To improve estimates of atmospheric MnP, these emission rates require field observation and validation using advanced field sampling methods (such as 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. Although MnP are often considered hydrophobic, once within the environment it is unknown whether this hydrophobicity changes, for example, as a result of 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 MnP 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 (such as Saharan dust or caesium-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 (Fig. 3) of these generally negatively charged[81,82], 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), uncomparable size fractionation, differing particle characterization (shape, polymer type) and sampling of different processes (for example, snow deposition versus pumped volume of air)[3,83,84]. Atmospheric (terrestrial and marine) MnP studies need to provide comparable results to ensure that the data improve our 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[85,86]. 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[75], Petri dishes with double-sided tape[87], NILU **AQ19** deposition collectors[88], or Brahney Buckets[89] (to name a few) have different blow-by (particles 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[19,45]. However, in practice, these black particles can be difficult to characterize by spectroscopic methods because the signal is limited by absorption of input wavelengths and the strength of vibrational response. Therefore, tyre and brake wear particle chemical characterization is often achieved with destructive thermal degradation methods, without particle morphology characterization[45,90]. 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 disjointt creates uncertainty in total MnP calculations and representation (both atmospheric and marine).

Methods to improve the flux estimate

To improve the accuracy of 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 characterized or parameterized (orange processes highlighted in Fig. 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 MnP both inshore and offshore, ocean ejection of MnP offshore, and coastal and offshore deposition.

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 >50 µm (losing the smaller particle fraction)[21,91]. In addition, pump-sampling devices have been mounted on buoys and ships[38,68,69]. 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[92], but field testing is needed to ensure that 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 <50 µm MnP particles, especially in open-ocean and remote locations, and to provide sample methods close to the water surface.

Although the study of marine MnP emission to the atmosphere via bubble-burst ejection and sea spray processes is in its infancy[35,36,37,73], since the 2000s there has been extensive research on the mechanism of sea-salt aerosol production and other materials involved with ocean–atmosphere exchange[72,93,94]. 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)[95]. The BIMS was originally designed for sea-salt aerosol studies, but its

type device could effectively improve detection of plastics in the marine atmosphere environment. 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[96, 97].

Atmospheric MnP generally fall within the lower range of MP (<500 µm) down to NP, which is a particle size range that is complex to analyse[98, 99] and is within the size 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 11 µm using an µFTIR or 2 µm using a µRaman, but with pixel size limitations and within the LOD of 10 µm for **AQ20** FTIR and 1 µm for Raman under standard analytical setup)[100, 101]. Polymer identification analysis, across the full particle size range, is a vital requirement for MnP analysis and reporting[3, 102, 103]. Analysis of individual particles below 1 µm can be achieved (for example, using equipment such as Raman tweezers, AFM-IR)[98, 104, 105] but it is resource-heavy and difficult to analyse a representative proportion of a field sample. To improve our understanding and flux assessment of atmospheric marine MnP, new techniques and advancements in technology are needed to enable submicrometre particle polymer analysis that provides comparable results to the micrometre particle studies published to date.

There is limited testing or parallel analysis of mass and particle counts to date[84, 85], resulting in mass-based results being mathematically converted to particle counts and vice versa, producing an uncertainty associated with this mathematical estimation. Mass analysis of MnP using destructive methods (thermal degradation) is now possible for very low concentrations of NP in environmental samples[20, 106]. Although 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 characterization analysis is unquantified for MnP studies. To ensure accurate conversion of mass-to-particle count[37, 59] 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 standardized. Although it might not be possible to standardize 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 for MnP: the limits of detection and quantification of studies (LOD and LOQ); a clear description of analytical methods to support interstudy 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; and sample replication and individual replicate results[102, 103, 107, 108]. While visual or graphical representation of MnP findings can be done in coarse particle increments, it is necessary for interstudy 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 data repository or supplementary dataset). 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[109, 110]. Analytical methods have advanced beyond visual identification (effective to around 500 µm)[111, 112, 113] 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 over 30%) of reported particles must be validated using (at least one) of these methods.

Ocean–atmosphere flux estimations using current information have large uncertainties due to data availability, sampling methods and study intercomparability. To improve our ocean–atmosphere flux understanding, a global quantitative characterization of MnP that provides more standardized and comparable data is needed.

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[114]. 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. To quantify these processes, a comprehensive, formalized global programme is needed that follows a harmonized protocol of sampling and analysis. A key objective is to provide comparable datasets that enable detailed characterization 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.

Global long-term observation network

Multi-year measurements at selected long-term observation sites will identify the current state of and trends in atmospheric MnP concentrations. Such long-term observation activities are usually a part of a globally coordinated research or monitoring network(s) to spread the cost and to ensure data uniformity. **AQ21** We propose an organizational approach to address these research needs (Box 2). These activities are broadly divided into measurement studies and modelling studies. The objective of this research organization is to ensure the identified data limitations, interstudy comparability issues and process knowledge gaps are fully addressed with specific objectives in mind. However, there must be cooperation and integration across all activities.

Early modelling of atmospheric MnP gross deposition shows considerable atmospheric deposition into the oceans, especially the Mediterranean Sea, and the North Pacific and North Atlantic oceans (Supplementary Fig. 4)[18]. However, these estimates must be used with caution because much of the deposition theoretically represents both MnP ejected from the ocean surface and transported

in the mid-and high-latitude North Atlantic and North Pacific oceans and the northern Indian Ocean (Fig. 4)[19]. These early findings, although limited to a subset of MP types, provide guidance in establishing location priorities in studies of the global MnP cycle. Place Fig.4 here, within the text that discusses the figure content

To expedite these studies, it is recommend that the existing stations (Fig. 4) in the World Meteorological Organization (WMO) Global Atmosphere Watch (GAW) programme[115,116] be used as the initial long-term monitoring platform network[115,116]. The proposed sites are non-prescriptive but form an effective basis for a long-term observation network for atmospheric MnP. 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[117] and of atmospheric deposition[118]. 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 MP measurements. As such, the WMO/GAW programme presents an ideal and cost-effective global monitoring network with which to commence long-term observation of atmospheric MnP.

The sites (Fig. 4) are suggested on the basis of their capacity to create multi-year time series for extended sets of variables, ranging from atmospheric constituents to atmospheric dynamics, that are 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 efforts and published field data (Supplementary Note 4). A selection of coastal and marine locations would ensure good coverage on a global scale (Fig. 4), 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 should be added to the network (future network expansion) to represent these areas.

Box 2 Proposed global network structure and coordinated international research

Atmospheric transport studies can be divided into measurement studies and modelling studies. Measurement studies are further compartmentalized into exploration, monitoring and process studies, each focusing on advancing atmospheric MnP science at specific points in the plastic cycle. Similarly, modelling studies are divided into long-range transport and deposition or source studies, aimed to quantitatively characterize atmospheric MnP specific to long-range transport, source emissions or other elements of the plastic cycle.

Measurement studies

Monitoring

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

Exploration

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

- Ship-based atmospheric sampling offshore (~~the~~north and southern oceans, **AQ22** the Arctic it is always refered to as "the Arctic" rather than just Arctic and Antarctic)
- Ice cores in Greenland, Antarctica and the Arctic (and other locations)
- High-altitude aircraft measurements, coastal and offshore
- Marine air concentration buoy-type platform measurements

Process

Emission, deposition and transport process studies (potentially including degradation, leaching, Trojan horse and other studies) and quantitative characterization of MnP marine-atmospheric dynamics, including:

- Assessment of the ocean as a source (emission and resuspension of MnP)
- Differentiated wet and dry deposition on ocean and/or 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, and 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 and deposition). Flux trends and responses to policy or practice changes can be derived using these models (long-term data mining and modelled forecasting).

Complementary measurement and modelling studies will provide greater detail of the current state of atmospheric MnP and the potential emission, deposition and transport of MnP within the marine-atmospheric environment.

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[119, 120, 121, 122, 123]. Past and currently active global monitoring networks studying non-plastic atmospheric substances have used a variety of sampling platforms, sampling methods, observations 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. Weekly sampling (to yield monthly mean MnP particle quantitative particle characterization and mass analyses) is recommended, which could initially suffice for the gross characterization of transport quantities (although we acknowledge that for such a novel global study, adjustments will need to 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, hourly or daily sampling, depending on the research focus) undertaken through shorter-term intensive research campaigns using specialized equipment and platforms (such as unmanned aerial vehicles (UAVs) and BIMS). It is important that these exploration and process campaigns create data that are comparable with the global long-term observation dataset, and thus 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 may take several years to develop a full description of the atmospheric MnP burden, flux and trends, owing to the annual and inter-annual variability of conditions that affect entrainment, transport and deposition of atmospheric particles[124]. 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[116], the proposed observational sites will adopt centralized data reporting (similar to the World Meteorology Organisation dataset management).

Proposed sampling platforms

Sampling strategies to achieve long-term observations are initially proposed for fixed stations (Fig. 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 (17–20 m walk-up scaffold sampling towers equipped with elevated atmospheric samplers supported by temporary or permanent field laboratories located on both the continental coast and islands at the terrestrial–marine interface)[119, 120, 121, 122].

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)[123, 125, 126]. Such campaigns are typically 20–40 days in duration and entail frequent location changes, which enable offshore sampling over a wide spatial and temporal range (Supplementary Note 4). Offshore atmospheric MP sampling has been limited to air filter sample collection[38, 68, 69]. Future campaign protocols must be extended to include deposition and NP sampling. Intensive studies to quantitatively characterize the under-studied processes and environmental conditions (Fig. 3) will need to use novel and innovative sampling methods, redesigned and validated specifically for MnP observation. It is expected that 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 BIMS (with advances specific to MnP analysis) is used.

Low-latitude air sampling and vertical and horizontal array sampling over coastal and offshore environments can be achieved through the use of unmanned aerial vehicles. UAVs have limitations on flight duration but can sample over extensive vertical and spatial distances provided sampling payloads are kept minimal[127, 128]. 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, the sea surface and urban areas of potentially high-emission activities) with fewer constraints. This level of control over flight path (and therefore sample precision) could be very useful for intensive air and emission source sampling in the marine environment (Supplementary Note 4). UAVs will enable sampling in locations where access is limited. Use of UAVs could improve measurements of the overall marine-atmospheric MnP burden and help to quantify ocean–atmosphere exchange.

The proposed global observation network and sampling strategy would provide a comprehensive assessment of marine-atmospheric MnP and the ocean–atmosphere MnP flux. Combined with intensive process-, environment- or meteorologically specific focused studies, the global strategy will enable more accurate marine-atmospheric MnP flux estimations, highlight hot spots and key exchange or transport processes that will support improved policy, management and mitigation measures tackling MnP.

Summary and future directions

There is consensus that MP and NP pollution can harm the environment and, potentially, 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.

Reviewing the current state-of-the-art sampling and analysis methods makes it evident that both sampling and analytical methodologies need to be improved 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. The adaptation and advancement of marine and terrestrial sampling methods used in aerosol and atmospheric chemistry research could provide a path towards 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 (such as UAVs, BIMS and deposition collectors). Future sampling campaigns should incorporate a range of open-ocean sampling platforms and sampling methods to help to address the marine-atmospheric MnP research gap.

In conjunction with the complexity of marine-atmospheric MnP sampling, there is a need to improve analytical methods to help to quantify the marine MnP flux. Current analytical methods have advanced to the point where these measurements can be reliably made, but a harmonized approach is fundamental. Despite an increasing particle count with decreasing particle size, until now the majority of analysis has focused on larger MP particles (>10 μm), and there is limited NP analysis and unquantified uncertainties surrounding the comparison of different analytical methods. Analytical advances to enable both mass and particle characterization of marine-atmospheric MnP are necessary, complemented by detailed studies to create an easy comparison between different analytical results. These studies will enable future studies using particle characterization to be directly comparable to mass concentration studies, and will include the NP range.

Early estimates suggest that the atmospheric MnP influx to the oceans are comparable to that from rivers[78]. However, early model estimates show a huge range of uncertainty[18, 19, 78]. An expanded and coordinated global-scale research effort must be undertaken to constrain the uncertainties and provide a clear representation of the marine MnP flux. 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, we recommend the global monitoring effort expands to include research vessels and open-ocean observations, which will complement existing monitoring in inland water bodies and estuary sites.

After several years of network operations, we expect that researchers will be able to identify the key locations, processes and sources of MnP that affect the marine environment. Conversely, this research will also demonstrate the influence and relative importance of emissions from the marine environment that influence the terrestrial atmospheric MnP burden. This improved understanding of MnP flux and the global plastics cycle will be vital for evaluating 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 MnP concentrations.

Supplementary information

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D.A. and S.A. are the lead authors; they undertook the data research, provided substantial contribution to the discussion of content, and undertook the writing and editing of this article. R.A.D. and J.M.P. substantially contributed to the discussion of the content and writing of this article, T.J. and P. Liss substantial contributed to the discussion of the content, and M.B., P. Laj and L.E.R. substantially contributed to the writing. M.K., S.E. and N.E. performed data research and all authors contributed to the review and editing of this article.

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Data availability

The data for Fig. 2 are supplied in the online Supplementary Data file.

Competing interests The authors declare no competing interests.

Supplementary information

Supplementary Information

Supplementary Information

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