

PERSPECTIVE

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# Addressing the importance of microplastic particles as vectors for long-range transport of chemical contaminants: perspective in relation to prioritizing research and regulatory actions

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## Abstract

Over the last several years there has been increasing concern regarding the environmental fate and potential global transport of plastic debris, particularly in the form of microplastic particles (MPs). The global transport of MPs has also triggered concerns regarding the potential role that its mobility may represent towards influencing the long-range environmental transport (LRET) of particle-bound chemicals, particularly the large number of chemicals known to be added to plastic. This perspective considers the various lines-of-evidence that might be used towards understanding the LRET of persistent organic pollutants (POPs). For instance, it has been proposed that the LRET of POPs is facilitated by global fractionation processes that facilitate the mobility of chemicals from source regions towards remote locations, such as the polar regions, where they have the potential to accumulate. These processes are influenced by the physicochemical properties of POPs and can result in various transport mechanisms influencing environmental fate and transport. Here I suggest that there are similarities that can be drawn, whereby knowledge of how differences in the physicochemical properties of MPs relative to different emission scenarios, can influence the relative importance of sequestration processes that may result in global fractionation of MPs. Several challenges are identified throughout the perspective, with an urgent need towards the development and application of standard sampling and analytical methods being identified as critical for enabling datasets to be reliably compared for use in better understanding potential source-receptor relationships, as well as advancing the characterization and quantification of various environmental fate processes. In many instances, it is suggested that advances in our understanding can be facilitated based on knowledge obtained in other areas of research, such as in relation to studies developing tools to evaluate the mobility of particulate organic matter in aqueous environments or from studies investigating the fate and mobility of atmospheric particulates. Recognizing that not all MPs are equal, with respect to environmental fate and toxicological effects, knowledge regarding which types of MPs are likely to be subject to LRET can only strengthen our ability to evaluate their role as vectors of transport for plastic associated chemicals and the associated risks that their LRET may represent. Nevertheless, several outstanding issues remain that would benefit from constructive discussions between all stakeholders. It is anticipated that this perspective can play a role in initiating those discussions.

**Keywords:** Long-range environmental transport, Plastic additives, Exposure assessment

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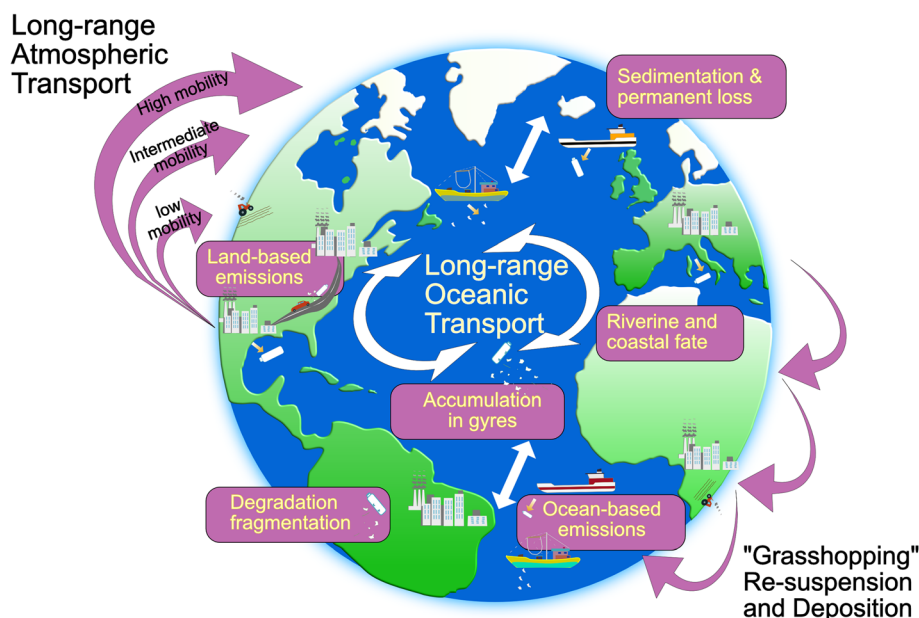
## Introduction

Characterizing and quantifying the relationship between an exposure to a physical or chemical stressor and a toxicological adverse effect represents the underlying principle of risk assessment. Assessing exposure is the process of estimating or measuring the magnitude, frequency and duration of exposure to a stressor, which ideally includes characterization of the sources, pathways, routes and the associated uncertainties [1, 2]. Recognizing that all physical and chemical stressors have the potential to cause harm to human and ecological health – actions aimed at reducing the magnitude, frequency and duration of their exposure represents an aspirational goal within any global sustainability agenda [3].

Assessing the exposure and risks of chemical substances used in commerce represents an important element of chemical risk assessment conducted by national, regional and international regulatory agencies. Organic chemicals, for instance, are routinely screened and prioritized for exposure mitigation based on their potential to be persistent, bioaccumulative and toxic (PBT) [4–9]. In addition to identifying substances as PBT, the Stockholm Convention on persistent organic pollutants (POPs) includes the evaluation of an additional property that specifically assesses the potential for an organic chemical to be subject to long-range environmental transport (LRET) [4, 6]. For a comprehensive summary of the regulatory instruments and the evolution of PBT and POP screening criteria please see Matthies et al. [4]. Because the combined properties of persistence and LRET can result in an organic chemical

to become globally distributed and bioavailable over relatively long-time scales, exposure and the potential risk of an adverse effect to organisms and human populations in remote areas can represent a significant cause for concern.

Nearly 30 years ago, Wania and Mackay [10] proposed a mechanism of LRET for POPs that involved a process referred to as global fractionation, whereby chemicals are characterized as being latitudinally fractionated as a result of varying volatility encountered due to changes in environmental temperatures. The mechanism proposed raised potential concerns regarding chemicals with vapour pressures within a certain low range, in that they may preferentially accumulate in polar regions [10]. Figure 1 is a schematic illustration that includes the key elements of the global fractionation and ‘grass-hopping’ concepts. Briefly, the concept, which has been widely used towards helping to understand the relationship between the physicochemical properties of POPs and LRET [11–16], aims at characterizing how chemicals might cycle between air, water and soils at varying temperatures encountered in the global environment. Warm temperatures at low latitudes, for instance, would favour evaporation from water and soil, causing chemicals to become mobile and subject to long-range atmospheric transport (LRAT), until they encounter lower temperatures at higher latitudes, which would favour deposition. The lower temperatures encountered in the polar regions would result in ‘condensation’ and thus an accumulation of chemicals in those regions that may eventually exceed toxicological



**Fig. 1** Schematic illustration of the concepts of global fractionation and “grasshopping” as proposed by Wania and Mackay [10]. Superimposed are various processes known to influence the environmental fate and transport of microplastic particles

thresholds of concern, if not appropriately managed. Semi-volatile organic chemicals, on the other hand, may be subject to LRET due to a process of “grasshopping”, whereby organic chemicals might be subject to seasonal, and even diurnal, cycling between air, water and soil, with their mobility occurring through a series of ‘hops’, commonly referred to as the ‘grasshopper effect’ [17]. Following the observations of Wania and Mackay [10], screening and prioritizing the potential of PBT and/or POPs to undergo LRET has since been greatly facilitated through the development and application of a wide range of modelling tools [6, 18–20]. Environmental monitoring data are also widely used as complementary information to results obtained from the models, which support a weight-of-evidence approach aimed at strengthening confidence in identifying new candidate PBT and POPs.

Over the last several years, concern regarding the environmental fate and potential global transport of plastic debris, particularly in the form of microplastic particles (MPs) has increased, including the potential role that MPs may play towards influencing the LRET of particle-bound chemicals, including POPs and plastic additive chemicals (PACs). A relatively simplistic model, which assumes that because most plastic polymers are buoyant in water, is that the release of plastic debris into the aquatic environment will result in its LRET causing the global distribution of these materials and contamination of remote regions to potentially occur [21]. Monitoring data supporting the global distribution of plastic debris and MPs are substantive, with a recent exponential increase in the number of publications aligned to this specific issue [22]. While there is no disagreement regarding the global distribution of plastic debris and MPs, the understanding of the sources, environmental fate and transport of these materials remains uncertain [23].

This perspective sets out to consider various lines-of-evidence that might be used in reflecting what we do and don’t know with respect to the LRET of plastic debris and MPs, with particular emphasis on their potential role as transport vectors for PACs. Questions considered in this perspective include:

- What do we know about the LRET of plastic debris and MPs?
- Are there similarities between the LRET of MPs and POPs?
- What do we know about the role that MPs play as vectors of LRET for PACs?

It should be noted that I focus primarily on the issue of MPs as potential vectors of PACs, primarily because these are chemicals we know are added to plastic polymers and therefore their potential to be transported by MPs generated from plastic products may represent a source of concern. The issue related to the role of MPs

as vectors for POPs has been addressed in various publications and although there continues to be an ongoing debate, from my perspective I perceive that the weight-of-evidence supports exposure to POPs via the LRET of MPs to be minimal relative to other transport pathways, see for instance [24–27].

### **Are plastic debris and microplastic subject to LRET?**

Researchers have estimated between 19 to 23 million tonnes of plastic debris entered aquatic ecosystems in 2016 [28]. Direct inputs due to riverine transport are projected as representing 2.8–18.6% of coastal emissions, with the top 20 polluting rivers largely located in south-east Asia, accounting for 67% of the global annual input [29]. Estimates for the global mass of plastic debris present in the open ocean surface layer, however, represent only a small percentage of the estimated annual emissions into the marine environment [30] – causing many to consider ‘where is the missing plastic?’

Several processes have been suggested in an effort to address the question of the ‘missing plastic’, including its potential uptake and accumulation in living organisms, rapid losses with respect to buoyancy and subsequent sedimentation to the deep ocean, as well as the potential for stranding on beaches and the role of degradation via microbes and photochemical processes towards mineralizing the plastic [31–39]. However, there continues to be a debate regarding the relative importance of the various processes with respect to the environmental fate and transport of MPs, which results in varying interpretations of data presented throughout the literature. Most recently, Wiess et al. [40] provide a statistical re-analysis based on updated data on MPs, the output of which suggests that previous estimates of river fluxes potentially represent overestimates, of up to three-orders of magnitude. Their reanalysis results in the average residence time of MPs at the ocean surface increasing from days to years, which they suggests strongly reduces “the theoretical need for a missing sink” [40], an observation that is also consistent with Pabortsava and Lampitt [36]. Nevertheless, Wiess et al. [40] acknowledge that their estimates are based on underlying data that contain significant uncertainties, but that their observations are perceived to be better aligned with an understanding that surface oceanic MP stocks, particularly within oceanic gyres, result from slow accumulation processes, an observation that is inconsistent with suggestions that there exists relatively rapid environmental fate processes that might help to sequester surface oceanic MPs. An additional notable observation from Wiess et al. [40] relates to differences in the properties of MPs between those observed in rivers and those in accumulation areas, whereby the observed differences are suggested to

potentially represent a selective sequestration of smaller size MPs. An implication from Weiss et al. [40], could therefore relate to the potential role of a global fractionation process that may cause some types of MPs to be more susceptible to LRET relative to others, analogous to processes illustrated in Fig. 1 in relation to the LRET of POPs.

Given the various interpretations presented throughout the literature in relation to environmental fate and transport processes, however, it may be premature and potentially naïve to suggest we do not need to address possible sinks for MPs in the environment. While there is a role for science to communicate advocacy messages aimed at encouraging efforts to quickly adopt measures aimed at reducing the unnecessary release of plastic and MPs to the environment, there remains a need to conduct fundamental science that can produce insight and knowledge for strengthening the characterization and quantification of environmental fate and transport of plastic and MPs – insight that would greatly strengthen our ability to assess environmental and human health risks. Ideally the research community can support both without negatively impacting the other. The publication of Weiss et al. [40] does indeed provide a good example illustrating how these messages can be balanced. For instance, they raise awareness regarding the need for action aimed at reducing plastic entering the environment, while also drawing attention to the underlying uncertainties in the data they use in their estimates and are specific with respect to identifying research urgently needed to help reduce those uncertainties. For instance, by advocating the need for the development and application of standard sampling methods and reliable extrapolation models that can help to avoid errors when performing mass balance estimates of MPs in the environment.

In addition to the development and application of standard sampling and analytical methods, however, I would include a continued need towards improved characterization and quantification of environmental fate processes that might influence the mobility and potential sequestration processes that Weiss et al. [40] draw attention to, since these processes represent important factors that strongly influence environmental fate and transport. An example of a potentially important environmental fate process, which may have previously been underestimated and overlooked, relates to research that has drawn attention to the relative importance that coastal systems may play in acting as a barrier to both transporting plastic to the open ocean and for open ocean plastic to reach coastal beaches. In 2017, Hardesty et al. [23], for instance, suggested the need for research aimed at characterizing and quantifying a loss term for coastal regions, which would represent an improvement towards

better understanding the Coast-Ocean-Coast suspension and resuspension mechanisms that influence the mobility of plastic debris and MPs in coastal zone environments. Building on the suggestion of Hardesty et al. [23] is the development and application of a model based on five model parameters by Lebreton et al. [30], who, in 2019, estimated the influence of including a mass fraction term of 97% to describe the annual stranding and settling of plastic debris along coastal shorelines. Model results demonstrate a significant time lag between the release of plastic debris to the coastal environment relative to debris present in the open ocean, with predictions that land-based plastic debris currently present in the open ocean originates from the 1990s. The stranding parameter in the model thus implies that the fate of plastic debris released to the environment is strongly influenced by the different mechanisms that cause marine litter to be captured by coastal landmasses—by beaching and sedimentation, for example—which represents a feasible explanation for the likely location of the missing plastic [41], but which is also consistent with a slow accumulation process reported by Weiss et al. [40]. While the observations of Lebreton et al. [30] are based on a simplified model system, they do represent an incremental step in our efforts as a scientific community to strengthen our understanding of environmental fate and transport processes, which from my perspective demonstrate a level of humility that reminds us that we are attempting to understand highly complex systems that are influenced by a variety of competing processes, each with their own intrinsic variability. The observation that losses of MPs due to beaching and sedimentation along coastal systems, however, should not represent a distraction towards taking actions aimed at significantly reducing the release of plastic debris to the environment. On the contrary, the insight could offer potential guidance towards finding effective solutions for reducing releases, which in this instance should include targeted solid waste management actions for regions identified as major sources, particularly in coastal areas [42–45].

With respect to direct sources of oceanic plastic debris and MPs, several studies report a predominance of fishing and shipping related materials in both surface waters and sediment. Dowarah and Devipriya [46], for instance, observed a strong correlation between fishing activity and MPs abundance at beaches along the Indian Puducherry coast and a weak correlation with recreational/tourist activities. In their study reporting the presence of both MPs and larger plastic debris items in the Southern Ocean, Suraria et al. [47] observed only 5 MPs south of the subtropical front and 17 macrolitter items, with estimated mean concentrations being  $188 \pm 589$  MPs/km<sup>2</sup>. The concentrations south of the subtropical front were notably an order of magnitude lower than those north of



this area, suggesting the importance of the subtropical front as a barrier for floating debris into the Southern Ocean. While it is difficult to attribute the plastic debris sources to the marine environment, approximately 10% of macrolitter could be identified as being associated with fishing related activity, such as in the form of fishing buoys [47]. Observations of Suraria et al. [47] are also notably consistent with earlier research investigating the presence of macrolitter in the sea around the Antarctic [48–50]. The presence of derelict and lost fishing gear represents a significant source of concern in the marine environment, where observations of marine organisms becoming entangled, suffocating or being victims of ghost fishing activity have been reported [51–53].

In Arctic waters, MP detection has been estimated at 28,000 MPs/km<sup>2</sup> [54]. These concentrations are consistent with levels reported for other ocean surface waters, but are approximately two orders of magnitude greater than those in the Antarctic, on a particle count basis. The dominant MP types observed are fibres, consisting of Rayon (30%), polyester (15%) and polyamide (15%) [54]. Although, Lusher et al. [54] note that the ability to attribute the sources of MPs observed in the open ocean cannot be accurately characterized, the shapes and compositions imply that the sources likely relate to the degradation and fragmentation of larger plastic items, such as fibres from shipping activity or fishing equipment, recreation and offshore industries (i.e. ocean-based activities). They also suggest that the relative importance of emissions from local sources (such as sewage discharges from communities within the Arctic) should not be underestimated. These observations are generally consistent with Ross et al. [55], who report a predominance of polyester fibres, with Pacific-influenced western Arctic fibres to have properties of more weathered fibres and at lower concentrations, while those from the eastern Atlantic-influenced Arctic are less weathered, most likely because they originate from newer sources. Large North American and Eurasian rivers that flow into the eastern Arctic Ocean may represent regional sources of polyester fibres, with possible origination from textiles discharged via wastewater treatment effluent. Most recently, Dibke et al. [56] report on the potential sources of MPs in the German Bight, where their data suggests a dominance of MPs generated from shipping activity in coastal waters and that previous estimates of MPs to the marine environment have underestimated the importance of ships. Most importantly, Dibke et al. [56] propose that the widely cited 80% terrestrial- to 20% marine-based debris ratio for MPs is potentially inverted, implying that the major sources of MPs to the marine environment are ocean-based. The observations reported above are presented to illustrate the various challenges that the scientific community encounter when

attempting to define source-receptor relationships. A major challenge, in this instance, relates to how well we can confidently differentiate between local and distant sources, whereby the presence of human activity in the open ocean and in the Arctic, cannot be neglected as potential factors that may influence the observations of MPs reported in those remote locations.

Complicating the overall ability to evaluate source-receptor relationships is the role of degradation and fragmentation of plastic debris and MPs in the open ocean, which is widely understood to represent an important source of MPs. Lebreton et al. [30] estimate that two-thirds of MPs generated from the degradation and fragmentation of plastic debris emitted from 2000 and earlier have disappeared from the ocean surface layer, most likely from settling, ingestion, aggregation, stranding or additional degradation into smaller particles. Research has also emerged demonstrating that buoyant MPs can become rapidly subject to biofouling and hetero-aggregation, which tends to increase the relative MP density, enhancing settling rates [57–60]. These various processes combine to cause researchers to speculate that sediments, both coastal and deep-ocean, likely represent ultimate sinks for MPs [38, 41, 61, 62], an observation that is consistent with existing knowledge regarding the mass flux of organic carbon from riverine sources into the marine environment. Peng et al. [62], for instance, report concentrations of MPs in hadal sediments of the Mariana Trench, representative of the deepest region of the oceans. The largest proportion of polymers detected in sediment samples is reported as polyester (19%), followed by polyurethane (14%), polyamide (12%), polyvinyl chloride (PVC) (10%) and rayon (10%), which they attribute as most likely originating from textiles, ropes, fishing gear, and plastic packaging materials [62]. While the MP sources are uncertain, they speculate that the accumulation of plastic debris and its degradation and fragmentation in the North Pacific Subtropical Gyre into MPs, followed by sedimentation to the deep sea environment, may represent the most likely source [62]. Their observations therefore support that transfer to the deep ocean likely represents a sink for MPs emitted to the open ocean [62, 63], although the ultimate fate of MPs in deep sea sediments remains largely unknown. Other notable factors that may influence the transport of MPs into deep sea sediments, such as those in the Mariana Trench, are attributed to the downslope horizontal transport of sediment, triggered by episodic earthquakes and the cycling of sediment via resuspension/deposition processes [61, 63, 64].

While research implies a variety of competing processes that influence the environmental fate and transport of MPs, from which it may initially appear are challenges that would be near-impossible to characterize

and quantify, there are potentially opportunities to leverage learnings from other areas of research. Research aimed at characterizing and quantifying the vertical and horizontal transport of particulate organic matter (POM), for instance, may represent insight that could be readily applied towards understanding the mass flux of MPs from rivers to the marine environment [34, 61, 65–67]. As noted by Harris [61], however, few studies reporting on MPs in the aquatic environment actually report the sedimentation rate associated with the system being monitored for MPs. Harris [61] suggests that proxies, such as total organic carbon and sediment grain size, could represent important sources of information for better understanding which environments are most likely to support sequestration processes for MPs [68]. Observations that continental slope sediment drift deposits contain higher concentrations of MPs than deep sea environments suggests the important role that coastal zone environments represent towards the sequestration of MPs [61, 69–73]. This is particularly relevant to MPs that may have properties, such as density and particle size, consistent with the POM being discharged from a riverine system [61]. The hydrological factors that influence the fate of POM in these systems, has the potential to influence the fate of the MPs in a similar manner. An important observation has been that low energy systems, such as shallow areas, fjords, estuaries and bays that receive significant inputs from land-based sources, have much higher sediment concentrations of MPs than those observed in deeper environments, which also tend to be more remote relative to direct emission sources [61, 63]. A challenge to the MP research community, therefore, is to embrace cross-discipline research opportunities and actively consider how existing knowledge may potentially be useful in helping to strengthen our understanding of environmental fate and transport [45, 61].

It should be acknowledged that the discussion presented above has focused primarily on the fate and transport of MPs in the aquatic environment, primarily because this is where much of the research has taken place over the last several years. However, there is a growing awareness of the presence of MPs in the atmosphere, with several studies drawing attention to the potential role that LRAT may play in facilitating the mobility of MPs from source to remote locations [74–80]. In the interest of brevity, however, it is not my intention to comprehensively review studies reporting MPs in various environmental media as part of this perspective. Nevertheless, I do think it appropriate to draw attention to underlying aspects that are common to all MP environmental monitoring data. For instance, a confounding factor that accompanies the reporting of MPs in both air and water are concerns that have been raised regarding the lack of standardized methods for measuring MPs, which results in accompanying concerns

with respect to the relative quality of the data presented and associated challenges when attempting to extrapolate and compare data between studies that use different sampling and analytical methods and which report their data using inconsistent metrics [40, 81–84]. Drawing attention to the concerns raised in relation to the reporting of MPs in environmental media, however, is not meant to undermine the added value of the studies reporting MPs in any particular environmental compartment, but rather to take the opportunity to draw parallels between all environmental data. With specific attention to the environmental fate and transport of MPs in air, I would draw attention to the considerable amount of research that has been conducted in relation to characterizing and quantifying the environmental fate and transport of atmospheric particulates. Is it possible that the MP research community might actively work towards identifying constructive opportunities to apply those learnings for use in better understanding the mobility of MPs in air, similar to suggestions to consider utilizing knowledge that has been gained through the monitoring and modelling of POM in aquatic systems?

In Fig. 1, I have superimposed various key processes understood to influence the environmental fate and transport of MPs, including illustrative examples of the various sources that may complicate our ability to differentiate local versus distant sources of MPs detected in the environment, onto the illustrative schematic commonly used to represent the processes that influence the global mobility and fractionation of POPs. Below I reflect on the feasibility of building upon the fundamental principles used in developing models that have been used to evaluate the LRET of POPs for use in assessing the LRET of MPs. For instance, a theme I wish to consider builds upon recognizing that MPs represent a complex heterogeneous mixture of particles of varying polymeric composition, shape, size and density. Therefore, it seems reasonable to anticipate that the mobility of MPs in the global environment is subject to fractionation processes that are likely analogous to the heterogeneity of the physicochemical properties of organic chemicals, and which may result in MPs of a given set of properties to be subject to mobility consistent with the concept of ‘fliers’, ‘swimmers’, ‘single-hoppers’, and ‘multi-hoppers’, terminology introduced by Wania (13) when relating the properties of POPs to varying transport mechanisms.

### The role of models

Models have been developed and applied to estimate MP mobility following their release into source regions, with their properties potentially influencing fractionation as they move away from emission sources [85]. Most recently, Schernewski et al. [67] applied a model to estimate the aqueous-phase residence time of polyethylene

and polypropylene (PE/PP) particles of sizes between 20 and 500  $\mu\text{m}$  discharged from waste water treatment systems within the watershed of the Baltic Sea region. Their results suggest that PE/PP entering the Baltic Sea has an average residence time in the water column of 14 days before they are washed ashore, whereas polyethylene terephthalate (PET) demonstrated seasonal variability with respect to its residence time, being about 14 days in the winter and 3–7 days in the summer [67]. The seasonal variation observed for PET is due to seasonal differences in hydrology in relation to the properties of PET. Because both PE/PP and PET have short residence times, Schernewski et al. [67] observe strong accumulation zones for MPs in the vicinity of emission sources, an observation that is also consistent with Turrell [86] who report results of a model describing the sources of marine plastic litter and MPs to the Scottish Atlantic and North Sea Coast, which are dominated from sources related to littering along the Scottish coast. There is thus a complex interaction between the properties of MPs, the influence of weathering, biofouling and aggregation, as well as hydrodynamic processes that influence their relative mobility in the environment that needs to be appreciated [41].

Modelling the environmental dispersion and deposition of organic chemicals in the environment to evaluate whether the chemicals have a local, regional or global scale impact has represented an important element towards evaluating the LRET of POPs [6]. Scheringer [87], for instance, demonstrated that the spatial scale needed to characterize the dispersion and deposition of organic chemicals is strongly influenced by their physicochemical properties. Defining the concept of a spatial range,  $R$ , which is a property that quantifies the spatial dispersion of a chemical as a probability distribution relative to its point of release, Scheringer [87] introduce a mass-independent metric for screening and prioritizing moderately and persistent organic chemicals. An important observation is that persistence is a necessary condition for LRET, since in the absence of persistence a chemical would have limited spatial dispersion. However, not all persistent chemicals are necessarily subject to LRET. For environmental dispersal to occur from its point of release, the chemical must have a sufficiently long residence time or persistence in a mobile media, such as air or water capable of supporting global scale transport [17]. For volatile and semi-volatile persistent organic chemicals, a combination of physicochemical properties combine to support their potential for LRAT, whereas water soluble persistent organic chemicals may be subject to LRET in water. Very hydrophobic persistent organic chemicals with log octanol-water partition coefficient ( $K_{OW}$ )  $> 6$ , on the other hand, and which strongly sorb to POM in the ocean, are observed to have

reduced LRET, particularly when transfer of POM to the deep ocean is included when modelling the spatial range of these types of chemicals [88]. As noted by Scheringer et al. [88], evaluating the LRET of POPs requires an appreciation that environmental fate and transport are multimedia problems, and that processes, such as export to the deep sea should be considered when evaluating their global distribution, especially for those with  $\log K_{OW} > 6$ . I think the observation of Scheringer et al. [88] in this instance is applicable to the LRET of MPs, i.e. the interactions of MPs in the environment require an appreciation of the multimedia processes that influence their environmental fate and transport.

Complementary to the concept of spatial range, as a metric for evaluating LRET, has been the development of models used to estimate a characteristic travel distance (CTD). According to van Pul et al. [89], for instance, the atmospheric residence time can be considered as the most indicative parameter for evaluating the potential of atmospheric pollutants to contribute to LRET, which can be determined from their physicochemical properties. Van Pul et al. [89] describe the atmospheric residence time as the time period in which the mass of the pollutant in the atmospheric boundary layer is reduced by 50%, and which is derived based on the total removal rate resulting from all removal processes from the atmospheric boundary layer, including wet and dry deposition and photochemical degradation of the chemical. Building on the concept of an atmospheric 'half-distance', Bennett et al. [20] introduce the concept of the CTD, which is the distance from the source region at which the concentration of the pollutant in air is reduced by 63%. Consistent with deriving  $R$ , the CTD is obtained from a multimedia environmental fate model, whereby the CTD can be shown to be strongly influenced by the intrinsic physicochemical properties of the chemical, not by the mass of chemical emitted to the environment [19, 90].

Considering the important insight that has been gained from the development and application of models and the metrics of LRET that have been proposed for organic chemicals, there may be similar opportunities to consider the development and application of metrics to assess the LRET of MPs. Much of the discussion related to the mobility of MPs has tended to be speculative, correlating detection of MPs in remote locations with their potential for LRET. As discussed above, there exists significant uncertainties, however, which inhibits the quantification of source-receptor relationships for MPs detected in the environment, whereby the relative impact of local sources cannot be excluded [41]. Having tools that enable the evaluation of the distance that MPs may travel until a reduction within a Lagrangian cell of 63% in concentration is observed, analogous to the CTD

for instance, may represent a tool that could be used to help strengthen our understanding between the properties of the particles relative to their environmental fate and behaviour within complex multimedia environments. The sensitivity of factors such as polymer density, size, and shape as well as environmental processes that may influence weathering, dispersion and deposition, should all be assessed in an effort to determine the sensitivity of both the physicochemical properties of MPs relative to varying environmental fate processes, in both air and in water [41]. Intuitively, the more dense polymers, such as polyvinyl chloride (PVC) are likely to undergo limited LRET in either air or water as compared to less dense polymers, such as polystyrene. However, as particle size decreases the relative influence of density may become less sensitive towards influencing mobility, and this may differ depending on whether the particle is emitted to air or water, thus insight regarding which particle sizes and/or shapes may be subject to LRET can be better communicated. Consequently, a more mechanistic approach based on the properties of the particles using models, would greatly strengthen informed discussions and help the decision-making process by providing guidance with respect to communicating which types of MPs may be subject to LRET. The objective should be towards screening and prioritizing which combination of properties of MPs relative to mode of entry to the environment and environmental fate processes most strongly influence their potential mobility [91]. This insight would also benefit technological innovation in the area of material sciences, by providing knowledge that can be used in the development of plastic products in the future that would not only minimize the release of MPs during all life-cycle stages of the product, but which would also ensure that the use of materials would not include properties that might result in the LRET of MPs, in instances where MPs may unintentionally be generated.

### **The role of monitoring data**

When considering the availability of monitoring data, combined with the ubiquitous use of plastic articles, particularly PE/PP and PET, as well as the use of polyester and polyamide textiles, such as in relation to fishing and shipping activity, the presence of plastic debris and MPs are likely to be strongly correlated with any human activity. Therefore, a strong correlation between human activity and the emissions of MPs represents a reasonable assumption that can greatly complicate our ability to interpret the LRET potential of MPs solely based on monitoring data. For instance, even in remote locations human activity can occur, whereby the improper and careless handling of plastic waste can result in relatively large releases of plastic, either intentionally or

unintentionally [92]. This statement, however, includes a certain element of subjectivity, since it should be noted that I am not aware of any agreed upon definition regarding the actual distance MPs and/or plastic debris need to travel for the materials to be subject to a LRET designation. My perception, for instance, would be that source-receptor distances of about 300 km are representative of local/regional relationships, which would not be sufficient to meet a designation of LRET. Thus, shipping and/or fishing activity off the coast of remote islands that result in the release and subsequent beaching of plastic debris on their beaches, would, in my opinion, reflect a local source. Of course this suggestion is purely speculative on my part, and is intended to be controversial in an effort that will hopefully result in stimulating a constructive dialogue regarding how we might characterize and quantify the various lines-of-evidence of LRET for MPs.

Nevertheless, a direct correlation between the magnitude of human activity, such as defined by population density, and the release of MPs remains a data gap that would benefit from improved characterization and quantification. It may be, for instance, that even a small number of individuals can generate a significant amount of plastic debris and MPs. Directly correlating human activity with exposure to MPs represents a significant challenge towards characterizing which fraction of MPs detected in remote locations is due to local sources relative to that of LRET [41]. There are both similarities and differences when comparing to the evaluation of monitoring data regarding the LRET of POPs. POPs detected in remote locations that are dominated by a strong industrial or agricultural use scenario are representative of an important line-of-evidence towards supporting LRET, in that their concentrations tend to be elevated in highly industrialized regions. POPs more widely used in commerce and detected in remote regions, on the other hand, require additional lines-of-evidence aimed at evaluating the relative importance of local versus distant sources. Quantifying the magnitude, frequency, and duration of concentrations along urban-rural-remote gradients can therefore potentially represent useful insight regarding the mobility of pollutants. It is anticipated that with the development and application of standard sampling and analytical methods, aimed at producing reliable and consistent datasets for MPs, that an improved appreciation regarding environmental fate processes that may sequester particles based on their physicochemical properties can help towards a more accurate understanding of LRET, consistent with how our understanding related to the LRET of POPs has evolved over the last several decades.

Factors that may confound the interpretation of MPs concentration data, however, must therefore include a



comprehensive understanding of the relative quality of the data presented. A number of issues have been raised regarding the quality of environmental data, with a key limitation identified as a lack of availability of standard methods with respect to the collection, processing and analysis of samples collected from all environmental media, including air, water, soil, sediment and biota [63, 82, 93–97]. Lack of standardized methods has resulted in many concerns being raised, including the potential that data reported on MPs in the environment may either over- or underestimate environmental concentrations. Factors such as background contamination can result in higher concentrations whereas losses during sample handling and processing can result in lower concentrations. Often sampling methods are limited based on the lower size range that can be quantified, with additional challenges encountered when attempting to compare data between studies due to variability regarding how concentrations are reported as well as poor confidence in relation to inter-laboratory comparison [98]. While these issues are generally well understood within the MP research community, and there exists a sense of optimism that they can be resolved within a reasonable timeframe, it would intuitively be beneficial to consider how to apply improved analytical tools towards improved characterization and quantification of source-receptor relationships. Specifically, quantification of rates that influence environmental fate, such as rates of fragmentation and degradation, biofouling, sedimentation and atmospheric deposition represent some of the key factors highlighted in @@@Fig. 1, and which influence environmental fate and transport. Additionally, I would consider efforts aimed at deriving emission factors, for instance, that might be correlated with varying types of human activity as also representing an important source of information that could be used towards improved source-receptor characterization. Characterization and quantification of emission factors, for instance, could represent opportunities to perform a priori exposure assessments, which may prove more practical than attempting to deduce emission sources from monitoring data a posteriori.

As is common for studies reporting on MPs in the environment, the above represents a ‘grocery-list’ of research needs and the accompanying challenge regarding how to best prioritize what to advance first. Given the important source of information that monitoring data represent, my preference would be towards advocating resource towards the development and application of standard sampling and analytical methods. We urgently require reliable and consistent data, in the absence of which creates a myriad of issues that are difficult to resolve due to underlying uncertainties that have not been addressed, primarily

because the data have been generated using non-standard approaches.

#### **Are MPs subject to LRET – yes and no**

Concerns regarding the presence of plastic debris in the environment represent an important challenge for society to urgently address [99]. I do not think there is any disagreement from anyone regarding that statement. The development and application of mechanistically-based models and the acquisition of reliable and consistent monitoring data are urgently needed to better identify and characterize the sources, sinks and environmental fate processes of MPs. This information is required to more effectively communicate the most efficient and tangible actions that can be taken to help reduce environmental exposure [100]. Releases of plastic to the environment occur locally in all regions of the planet, with no immediate ability to discriminate between remote and highly urbanized areas with respect to magnitude, frequency and duration of the emission [41]. Due to the ubiquitous release of plastic debris and MPs to the environment, and a certain element of subjectivity regarding how different individuals potentially perceive local versus distant sources, the ability to quantify LRET will continue to represent a significant source of uncertainty and debate. Generalizations, such as implying that all plastic debris and MPs have the potential for LRET fail to appreciate how the properties of the particles influence their environmental fate and mobility. A concern raised, therefore, relates to how such generalizations potentially inhibit the development and application of holistic tools that could strengthen our overall understanding of environmental fate and transport of MPs, as well as in relation to their role as vectors of transport for PACs, as I will consider in the next section. Screening and prioritising which combinations of properties and environmental conditions are most problematic, with respect to LRET of MPs represents a critically important process in helping to identify tangible actions that could be taken towards reducing societal concerns as well as helping to enable better input in relation to assessing both environmental and human health risks. A hypothesis to consider is that not all MPs are equal with respect to their potential to cause an adverse effect, consequently, I think it is prudent to identify the properties of MPs that we are most concerned, particularly when considering their potential to negatively impact remote ecosystems. An element to consider relates to the relative concern of the MPs themselves towards eliciting an adverse effect versus their potential to transfer a sufficient mass of PACs, for which the intrinsic hazard may represent a greater cause for concern than the particle itself. Having knowledge in relation to which types of MPs are potentially subject to LRET is important, since the use of PACs in plastic varies depending on polymeric

type and its commercial use. The objective of this section has been aimed at considering why improved understanding of which properties of MPs are most likely subject to LRET would be helpful, insight which when coupled with improved understanding of the use of PACs in plastic, should ideally result in holistic approaches for evaluating LRET as well as enabling directed strategies for assessing and mitigating risk. In other words, knowledge of the properties of MPs that may be subject to LRET and the PACs associated with those particles can be used to:

- Support effects testing that targets the testing of materials of greatest environmental concern
- Support technological innovation, particularly in the space of materials science, to develop plastic products that both minimize the potential release of MPs, particularly those with properties that are most likely subject to LRET, and the use of PACs that may be subject to LRET as a consequence of particle-bound transport.

In this section I thus propose that the LRET of MPs is indeed probable, but that there most likely exists various sequestration processes that strongly influence their relative LRET potential, and that, analogous to the LRET of POPs, mobility occurs as a global fractionation process. Identifying the properties of MPs and environmental release scenarios that most likely support LRET

potential should therefore represent an important goal for the research community to address.

### Does plastic represent a source of exposure to plastic-additive chemicals?

To address environmental and human health exposure to PACs requires an appreciation of their use in plastic articles throughout society. Uses of PACs include their application in a broad range of globally distributed consumer products, such as food packaging, electronics, textiles, furniture and appliances. The use of PACs is also associated with plastic materials used in various industries, such as in the construction, automotive and aerospace sectors [99]. Table 1 provides a summary of several widely used plastic polymers in relation to the different classes of PACs and the range of inclusion levels that may be associated with them.

PACs use is predicted to increase to about 9.75 million tonnes in 2024, with the greatest usage associated with flexible PVC film [105]. Since PACs are not chemically bound to the plastic polymer, but rather added to the plastic during manufacture they are subject to diffusion through the plastic along thermodynamic concentration gradients. The migration of a PAC from the plastic to the gas phase is a two-step process, which includes diffusion from the bulk polymer to the surface followed by evaporation, and which can be described by Fick's second law of diffusion. Diffusivity in the polymer typically

**Table 1** Summary of average densities of commonly used polymers and the application of representative additives and estimates of typical percentages added (weight/weight) in commonly used polymers [101–104]

Polymer	Density (g/cm <sup>3</sup> )	Anti-oxidant	Flame retardant	Plasticizer	UV Stabilizer
Typical amount range (% wt/wt)		0.05–3	0.7–25	10–70	0.05–3
<i>Thermoplastic</i>					
Acrylonitrile butadiene styrene (ABS)	0.98	✓			✓
Polyamide 66 (Nylon 66)	1.24	✓	✓	✓	✓
Polycarbonate (PC)			✓		✓
Polyethylene (PE) (amorphous)	0.85				
• Low-density PE (LDPE)	0.89	✓	✓		✓
• High-density PE (HDPE)	0.96	✓	✓		✓
Polyethylene (PE) (crystalline)	1.00				
Polypropylene (PP)	0.99	✓			✓
Polystyrene (PS)	1.04	✓	✓	✓	✓
Poly(vinyl chloride) (PVC)	1.39	✓	✓	✓	✓
<i>Thermosetting</i>					
Epoxide resin	1.2	✓	✓	✓	✓
Phenol-formaldehyde resin	1.36		✓	✓	
Unsaturated polyester resin	1.23–2.3	✓	✓	✓	✓
Polyurethanes	1.2	✓	✓	✓	✓

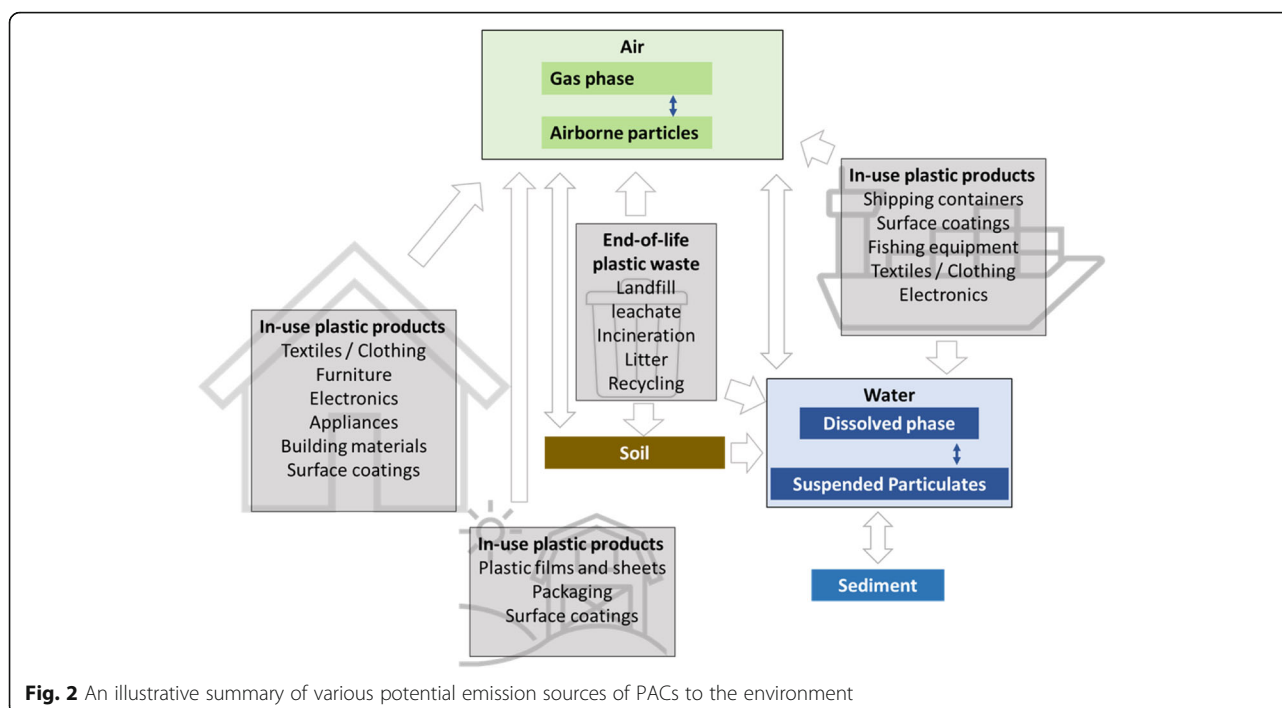
increases with the concentration of the PAC in the plastic, which can be explained by the increase of the free volume and mobility of the polymer molecules in the presence of the plasticizer [106]. Since both diffusion and evaporation influence the total loss of the PAC from the plastic, the overall rate of loss is determined by the slower process, which for low volatility chemicals will be the rate of evaporation. For PACs with high boiling points, such as di(2-ethylhexyl phthalate) (DEHP) it has been observed that a thin film can form at the surface of the plastic [106], which can facilitate the migration of the chemical from the plastic into dust particles that may have settled on the plastic product. Thus, the diffusion of PACs can result in their potential to leach from the plastic into the surrounding environment either directly through evaporative losses or in relation to the fate of thin films containing PACs forming at the surface of the plastic.

Chemical leaching rates from different plastic products can be measured and/or estimated based on knowledge of their physicochemical properties, such as their vapour pressure [107–110]. PACs, however, typically have relatively low vapour pressures, inhibiting the rate of loss due to evaporation. For instance, the emission rates of phthalate ester plasticizers have been reported to range from 0.3–4.9  $\mu\text{g}/\text{m}^2/\text{h}$  from a 0.5 mm thick PVC plate, whereas the emission rates of the more hydrophobic polybrominated diphenyl ether flame retardants are  $<0.5 \mu\text{g}/\text{m}^2/\text{h}$ , measured at a temperature of 60 °C from a 3 mm thick polycarbonate/ABS plate [107]. Emission rates, are also observed to vary, depending on the polymer type and thickness, as well as with changes in environmental temperature or in relation to the aging of the plastic product (107, 108). A general observation is that as hydrophobicity increases and vapour pressure decreases, emission rates also decrease. Flame retardants and UV stabilizers tend to have physicochemical properties consistent with low emission rates.

The expected lifetime of in-use plastic products, such as textiles, furniture, appliances, electronics, etc. can be on the order of several decades [99], which can result in concerns regarding the continuous leaching of PACs from widely used consumer products. Over the last several years considerable research aimed at reporting both estimated and measured emission rates for characterizing and quantifying human exposure due to oral ingestion, inhalation and dermal uptake have improved our understanding of environmental emissions [111–117]. Emissions of PACs from consumer products in indoor environments, industrial activities such as shipping and agriculture, and other human-related activities all represent potentially important sources to the environment (Fig. 2) [112].

When considering the summary illustrated in Fig. 2, with respect to the variable emission sources of PACs to the environment, it is important to appreciate that estimates of environmental emissions require the development and application of mechanistically consistent source emission categories, which can be used towards strengthening the reliability of models aimed at evaluating the environmental fate and transport of PACs via different emission pathways and exposure scenarios [112]. It is well understood that the emission of PACs from a specific in-use plastic product will be strongly influenced by a chemical's physicochemical properties, the properties of the plastic article and the environmental properties, such as temperature. Depending on the environmental properties where the in-use plastic product is used, such as air flow rates, the composition and volume fraction of other particles and dust, as well as the properties of other infrastructure, such as indoor surfaces, the surrounding environment can play an important role influencing the fate of PACs released from the product. Because the direction of flux associated with the emission of PACs from products is dependent on the properties of the chemical and the direction of the chemical fugacity gradient, the relative importance of a specific product as a source or sink for PACs can be determined [112]. Estimates of indoor concentrations of PACs, for instance, have been derived through the application of multimedia environmental fate models [112]. When coupled with assumptions of indoor-outdoor air-exchange fluxes, the far-field exposure of PACs, such as phthalates and flame retardants [3, 118–120], can be estimated when coupled with models of emissions for indoor environments.

It is generally assumed that because of their ubiquitous use throughout society, PACs, will be widely distributed throughout the environment, an assumption that is supported based on the detection and widespread reporting of PACs in all environmental compartments [120–128], which has fuelled concerns with respect to ecological exposure and potential environmental risks. Figure 2 implies that emissions to air may represent an important pathway facilitating environmental transport and fate, however, emissions associated with down-the-drain release scenarios can also result in significant emissions to waste water treatment plants, where chemicals can be subject to removal via biodegradation, with subsequent releases to the environment possible either via effluent discharges or onto agricultural soils in the form of biosolids for the undegraded fraction [128]. Furthermore, storm water runoff can contribute to additional emissions to surface waters [126, 129]. Given the ubiquitous use of plastic products throughout society, many of which contain PACs, the need to derive robust source emission categories are needed towards better



**Fig. 2** An illustrative summary of various potential emission sources of PACs to the environment

characterization and quantification of environmental emissions [112]. Although the emphasis here largely targets the use of PACs in plastic products, it should be further appreciated that chemicals typically used in various plastic products are also observed in other materials and products, such as cotton and wool and surface coatings. For instance, Avagyan et al. [130] report the presence of the benzotriazole photostabilizer 2-(2H-benzotriazol-2-yl)-4,6-di-tert-pentylphenol (UV-328) in cotton at a mean concentration of 106 ng/g; Sørensen et al. [131] report the highest levels of PACs associated with the degradation of wool fibres with the lowest levels of additives observed in PET fibres; whereas Luongo et al. [132] report that benzothiazoles and quinolines were present in > 80% of textiles (both cotton and synthetic materials) analysed, with significant reductions in concentrations following repeated laundering.

Despite the overwhelming data related to the emissions of PACs from consumer products and their ubiquitous detection in all environmental media, arguments have recently been made that plastic debris and MPs in the environment may provide a mechanism that can result in their transport to remote regions, such as the Arctic [133]. As discussed in the previous section, it is not possible to either prove or disprove that MPs are subject to LRET. It is thus problematic to demonstrate that the LRET of MPs results in the particle-bound transport of PACs to remote locations. Recognizing that the use of PACs and their weight fraction inclusion levels vary between different types of polymers (Table 1),

knowledge regarding which types of MPs are subject to LRET would represent an important source of information when attempting to estimate more accurately the mass of PACs that may be transported via their inclusion in MPs. A general observation, for instance, is that flexible PVC contains the greatest mass of chemical plasticizers, consequently, the LRET of MPs consisting of PVC would potentially warrant greater cause for concern than in the instance of PE, for which the use of chemical plasticizers is uncommon. If we consider a simplistic model, whereby the relative differences in density between PVC and PE are used as parameters that influence mobility, with the density of PVC > PE, then the relative importance of PVC to act as a LRET vector of PACs will be minimal, assuming that the greater density of PVC limits mobility and potential exposure to locations near emission sources. For PACs that may be associated with PE, on the other hand, and for which greater mobility can be assumed based on the simplistic model applied here, we may wish to further consider the influence of kinetic processes, such as leaching rates, which are likely to increase as particle size decreases or, alternatively, as surface area increases. Thus, knowledge regarding the size and surface area of MPs subject to LRET may be useful in better understanding the potential rate of losses of the PACs in those polymers to the surrounding environment while they are in transit from source to receptor, and which would enable an estimate of the mass of PAC that may potentially become bioavailable in remote ecosystems. The illustrative example given here thus



draws attention to data and environmental fate processes that would benefit our ability to derive estimates of environmental and human health risks, in that they potentially enable the derivation of the mass of chemical transported. Two important questions can thus be addressed through the development and application of models used to evaluate the LRET of MPs, particularly when coupled with models that already exist for organic chemicals. 1. What is the mass of PAC transported via MPs, and is it sufficient to exceed toxicological thresholds of concern? 2. How does the mass of PAC transported via MPs compare to other transport pathways that might influence their LRET? The discussion thus draws attention to a potential need towards refining existing tools that estimate the LRET of chemicals to introduce an additional transport pathway, i.e. the inclusion of MPs as a transport and exposure pathway for PACs, which should be considered relative to all other transport and exposure pathways. In this instance, the characterization and quantification with respect to the relative contribution between transport and exposure pathways is suggested to represent a valuable source of information for providing context and strengthening confidence by providing a holistic exposure assessment.

Recognizing the implications described above, Andrade et al. [133] estimate that 13,500 t of PACs are transported within buoyant plastic matrices globally. Their estimate assumes that the mass of PACs associated with plastic debris is equivalent to 5% of the 0.27 million tonnes of plastic estimated to be floating at the surface of the ocean. When the assumption that PACs are representative of 5% of the total amount of plastic debris is combined with estimates of plastic transported to the Arctic by Zarfl and Matthies (i.e. between 0.062–0.105 million tonnes), the total fraction of PACs in plastic debris transported to the Arctic is estimated as representing nearly 40% of the total amount. In their assumptions, however, Andrade et al. assume an average weight fraction of PACs across all plastic products, an assumption that is unlikely consistent with weight-fraction inclusion levels found in the different types of polymers that dominate the fraction of plastic debris found in the environment. Data reported by De Frond et al. [134], for instance, report weight fractions of between 0.05–2.5% for plastic polymers most routinely observed in environmental samples, or less than half the weight fraction assumed by Andrade et al. [133]. The assumption adopted by Andrade et al. [133] is thus likely skewed due to the higher inclusion levels associated with PVC, which tends to have a higher density and inclusion level of PACs. Caution should thus be used not to over-interpret the significance of the estimates reported by Andrade et al. [133]. Furthermore, Andrade et al. do not differentiate differences between the properties of the

plastic, such as with respect to size and shape, or take into consideration the influence of various weathering processes on the properties of plastic, which are all understood to influence the kinetic rates of desorption.

Nevertheless, the estimate derived by Andrade et al. [133] represents one approach for considering the mass of PACs that may be associated with plastic debris relative to the mass that might be found associated with in-use plastic products. Geyer et al. [99], for example, estimate that in 2015 there were 2500 Mt of plastic currently in-use and that 4900 Mt of plastic had been discarded to landfills or released to the environment between 1950 and 2015. Using the assumption of Andrade et al. [133], i.e. that the inclusion of PACs in plastic is 5%, would imply that there is approximately 125 Mt of PACs contained within in-use products and 245 Mt of PACs that have been discarded. Of the discarded fraction, Jambeck et al. [135] estimate country-specific fractions of mismanaged plastic waste that would enter the marine environment for 20 countries, with releases ranging from between 0.9–27.7%. Assuming that the average fraction of mismanaged plastic waste can be applied to the global situation (i.e. 4% of 4900 Mt or 196 Mt entered the marine environment between 1950 and 2015), would imply that approximately 9.8 Mt of PACs would be in plastic debris, or 3% of the total amount used up to 2015. Thus, while better understanding the fate of PACs associated with plastic debris would help improve overall understanding of environmental fate and transport, the estimates illustrate a critical need towards ensuring our understanding of the environmental fate, transport and exposure of PACs associated with the remaining 97% is appropriately addressed. The development and application of tools capable of evaluating the fate and exposure of PACs during both in-use and at end-of-life would thus represent a more holistic approach.

The illustrative estimates above imply that a considerable reservoir of PACs can be found associated with in-use products throughout society [101]. Various recent advances in exposure science imply that risk assessment frameworks are evolving to address the different use scenarios and exposure pathways, which should include end-of-life fate, such as in landfills, captured within recycling processes and as plastic debris and MPs as part of a larger evaluation strategy (Fig. 2). While the fraction of PACs found in MPs has raised concerns and has drawn important attention to a potentially novel exposure pathway for humans and the environment, the relative extent of the frequency, duration, and magnitude that the exposure scenario represents should be considered within a holistic framework which includes an evaluation of the relevance of all emission sources.

Previous efforts to evaluate human exposure to PACs via ingestion of seafood and drinking water that may be contaminated with MPs containing chemical additives have been conducted [103, 136]. The approaches taken can be perceived as conservative, combining data reporting maximum concentrations of MPs in seafood and drinking water with maximum concentrations of PACs reported in plastic debris and MPs in the environment, and assuming that 100% of chemicals associated with the plastic can be systemically available. Based on these assumptions, the mass of PACs associated with MPs contaminating seafood and drinking water suggests minimal human health risks [103, 136]. Similarly, environmental studies have also attempted to evaluate the relative importance that ingestion of MPs contaminated with PACs may represent relative to other exposure pathways, such as through the natural diet. Koelmans et al. [137], for instance, report that exposure and ingestion of MPs containing bisphenol A and nonylphenol by lugworms resulted in tissue concentrations that were observed to be in the lower global concentration ranges for these chemicals, implying that ingestion of MPs did not represent a significant source of exposure relative to other exposure pathways. The various attempts to characterize and quantify the potential risks associated with the role that MPs may play as a vector of transport, however, are in contradiction to several other studies that report a relatively important contribution to PACs due to their leaching after ingestion [138–140].

Within the context of what may appear to be an ongoing debate or controversy regarding the question as to whether or not plastic debris and MPs are vectors of transport of PACs, it is important to recognize that there is no disagreement that the process can occur [141]. The main issue relates to the relative importance of the exposure scenario and a desire to develop an improved mechanistic appreciation related to the various factors that might influence exposure to PACs within a holistic framework. Numerous studies have been conducted trying to characterize and quantify the leaching of PACs from MPs. However, it has been demonstrated that these studies also fail to address a number of quality assurance and quality control criteria related to the design and execution of the study, and which have been identified as important for reliably demonstrating the statistical significance of MPs to act as chemicals vectors. Specifically, Koelmans et al. [141] recently evaluated data produced by 61 studies reporting on the role that MPs may play as a vector of transport for chemicals and recommend the adoption of criteria identified as being critical for characterizing and quantifying the extent to which the leaching of chemicals from plastic might be important as an exposure pathway. Their analysis found significant variability in relation to the quality of studies, with

limitations identified for studies both demonstrating a vector effect and those not finding the process to be of significance [141]. There is thus perhaps an argument to be made to prioritize research aimed at an improved mechanistic understanding that can characterize and quantify key parameters needed in developing a holistic assessment framework, regarding the relative importance of MPs as a vector for exposure to PACs, and a reduction in studies attempting to prove or disprove that plastic can or cannot act as a vector of exposure.

Some suggest that screening and prioritization tools be developed to identify physicochemical properties of PACs and use scenarios that may represent greatest concern, either from their release related to in-use products or via leaching from plastic debris and MPs [101, 134]. Persistent PACs chemicals with  $\log K_{OW} < 3$ , for instance, will likely be subject to significant leaching from products over a relatively short time frame. Is it possible to identify the products and use scenarios that these types of chemicals are associated with – what are the environmental and human health implications? Intuitively, as  $\log K_{OW}$  increases a higher fraction of the PAC will likely remain in the plastic, can we characterize products and use scenarios in helping to quantify releases and potential exposure? In order to support a holistic assessment framework will be the need to estimate emissions due to leaching, for instance, as a function of product life expectancy, which can be useful in helping to evaluate potential exposures from consumer products, such as electronics, appliances, furniture and textiles, which can be anticipated to have average lifetimes > 10 years [99, 107]. Complementary to an improved understanding of the losses of PACs from products is a need for characterization and quantification of typical weight fraction inclusion levels, where it may be possible to couple various sources of information when investigating plastic debris in the environment as a tool towards characterizing a ‘fingerprint’ that might be useful in evaluating the age of the debris [142] – information that can potentially be useful in understanding environmental persistence.

## Conclusion

Given the numerous challenges and uncertainties articulated in this perspective, differentiating local versus global sources of plastic debris and MPs in the environment and their potential to act as vectors of LRET for PACs represents an important and non-trivial research need. This perspective attempts to imply an urgent need towards a coordinated strategy for the development of mechanistic models and analytical tools that can strengthen our overall understanding of the environmental fate, transport, and exposure of MPs. These tools are needed to help screen and prioritize the combination

of physicochemical properties and environmental factors that may facilitate the LRET of MPs. Building on the observations of Wania and Mackay [10, 15], are there analogies, for instance, with respect to transport mechanisms that influence POPs and MPs and which can be used to facilitate a mechanistically-based assessment of the LRET of MPs? Knowledge of the polymer composition, density, size and shape of MPs and their potential mobility under different environmental conditions can be used to a) identify potential geographic hot-spots and b) evaluate the types of PACs that may be associated with those materials and which can be used to help inform the application of models for characterizing and quantifying their leachability and potential exposure. When combined with source emission categories of PACs from in-use products and subsequent environmental release scenarios, a holistic exposure assessment may potentially be realized that strengthens and improves our overall understanding of environmental human health implications.

Tackling the significant uncertainties that accompany this issue will intuitively require coordinated and collaborative research that builds on the expertise of researchers from a wide range of disciplines. The current scenario that divides the LRET vector issue between those wanting to demonstrate that the process is of significant importance and those arguing that it is potentially not of significance only creates barriers that prevent an important constructive dialogue and which, in my opinion, only negatively impacts advances in scientific understanding and reducing uncertainties. Moving forward it is thus critically important to identify research opportunities that bring groups and expertise together to address data gaps necessary to inform effective and tangible actions that decision makers and risk managers can apply with a high level of confidence [143].

Over the last several years there have been a number of reviews collating and interpreting environmental monitoring data of MPs in all environmental media. A common feature of those reviews is a 'grocery-list' of research needs, which may result in various funding agencies encountering a challenge regarding which research need to support. From my perspective, there is an urgent need to support the development and consistent application of standard sampling and analytical methods. These are needed for strengthening our reliability and confidence in data reporting MPs in air, water, soil, sediment and biota. The acquisition and reporting of high-quality consistent data will result in better understanding of spatial and temporal trends, as well as enable better characterization and quantification of environmental fate processes, such as sedimentation, atmospheric deposition, resuspension, and potentially helping to inform the derivation of emission factors in relation to different

human activities. In parallel to these research needs are studies that can characterize and quantify rates of degradation and fragmentation of polymers of varying composition, size and shape, as well as biofouling and weathering processes.

Lastly, a key element of this perspective has been directed on the question of whether or not MPs are subject to LRET. While it is clear that it is impossible to demonstrate that MPs are not subject to LRET, I do think it is possible for us to develop modelling tools that can identify the properties of MPs that may be subject to LRET. However, in order to enable the modelling tools to prove meaningful within a regulatory decision-making context, we will need to develop an understanding and consensus in relation to distances that do and don't represent LRET. I think that the topics that I touch upon in this perspective will indeed trigger a range of responses, which given the importance of this topic is intentional. The objective of this perspective therefore has not been to necessarily argue for a definitive and prescriptive way for moving forward, but to present one possible way regarding how we might proceed and some of the underlying rationale. I am hopeful that we will at some point be able to bring together expertise from various areas and varying perspectives to engage in a constructive dialogue that helps us create a thorough and mechanistically-sound framework for assessing the LRET of MPs and the chemicals that may be associated with them.

#### Abbreviations

CTD: Characteristic travel distance; DEHP: di(2-ethylhexyl phthalate); LRAT: Long-range atmospheric transport; LRET: Long-range environmental transport;  $K_{OW}$ : Octanol-water partition coefficient; MPs: Microplastic particles; PACs: Plastic additive chemicals; PBT: Persistent bioaccumulative and toxic; PE: Polyethylene; PET: Polyethylene terephthalate; POM: Particulate organic matter; POPs: Persistent organic pollutants; PP: Polypropylene; PVC: Polyvinyl chloride; R: Spatial range

#### Acknowledgements

The authors acknowledge feedback and comments from various individuals, including Anastasia Swearingen and Brett Howard (American Chemical Council), Russ LaMotte (Beveridge & Diamond PC), Jens Christopher Otte (BASF).

#### Disclaimer

The authors alone are responsible for the views expressed in this publication and they do not necessarily represent the views, decisions or policies of the World Health Organization.

#### Author's contributions

The conception and design of the perspective entirely initiated, written, read and approved by TG.

#### Funding

TG acknowledges funding support from the International Council of Chemical Associations.

#### Availability of data and materials

Not applicable.

## Declarations

### Competing interests

TG has received financial support from the International Council of Chemical Associations in relation to providing consultancy support and comments on the Stockholm Convention on Persistent Organic Pollutants intersessional working group on long-range environmental transport and the intersessional working group on the draft risk profile for UV-328.

Received: 17 August 2021 Accepted: 18 August 2021

Published online: 30 August 2021

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