Atmospheric microplastics: A review on current status and perspectives

Yulan Zhang[a,b,*], Shichang Kang[a,b,f], Steve Allen[c,**], Deonie Allen[c], Tanguang Gao[d], Mika Sillanpää[e]

[a] State Key Laboratory of Cryospheric Sciences, Northwest Institute of Eco-Environment and Resources, Chinese Academy of Sciences, Lanzhou 730000, China
[b] CAS Center for Excellence in Tibetan Plateau Earth Sciences, Chinese Academy of Sciences, Beijing 100101, China
[c] Department of Civil and Environmental Engineering, University of Strathclyde, Glasgow, Scotland, UK
[d] Key Laboratory of Western China's Environmental Systems (Ministry of Education), College of Earth and Environmental Sciences, Lanzhou University, Lanzhou 730000, China
[e] Department of Civil and Environmental Engineering Florida International University Miami, FL, USA
[f] University of Chinese Academy of Sciences, Beijing 100049, China

ARTICLE INFO

ABSTRACT

Microplastics have recently been detected in the atmosphere of urban, suburban, and even remote areas far away from source regions of microplastics, suggesting the potential long-distance atmospheric transport for microplastics. There still exist questions regarding the occurrence, fate, transport, and effect of atmospheric microplastics. These questions arise due to limited physical analysis and understanding of atmospheric microplastic pollution in conjunction with a lack of standardized sampling and identification methods. This paper reviews the current status of knowledge on atmospheric microplastics, the methods for sample collection, analysis and detection. We review and compare the methods used in the previous studies and provide recommendations for atmospheric microplastic sampling and measurement. Furthermore, we summarize the findings related to atmospheric microplastic characteristics, including abundance, size, shapes, colours, and polymer types. Microplastics occur in the atmosphere from urban to remote areas, with an abundance/deposition spanning 1–3 orders of magnitude across different sites. Fibres and fragments are the most frequently reported shapes and the types of plastic which generally aligns with world plastic demand. We conclude that atmospheric microplastics require further research and greater understanding to identify its global distributions and potential exposure to human health through further field sampling and implementation of standardized analytical protocols.

1. Introduction

Microplastics are an emerging concern worldwide (PlasticsEurope, 2018; Rochman et al., 2019; Zeng, 2018). The common definition of microplastics is a plastic particle 5 mm to 100 nm in size (GESAMP, 2016; Masura et al., 2015; Thompson et al., 2004). A more recent definition of microplastics follows the logical differentiation along standard international unit nomenclature (SI units) of microplastics = 5 mm − 1 μm (Hartmann et al., 2019). Due to the evolving research on plastic particles, nanoplastics are also of particular concern because it is expected to be as ubiquitous as its bulk counterparts (Alimi et al., 2018; Bergman et al., 2015; Hartmann et al., 2019). Nanoplastics is usually categorized as plastic particles smaller than 1 μm, which is also an important priority with regard to seafood safety as well as

enhancement of contaminant transport in the environment and potential risks to human health (Alimi et al., 2018; Bank and Hansson, 2019; Hartmann et al., 2019; Zeng, 2018). However, environmental nanoplastics are yet to be quantified widely. Therefore, microplastics are the targeted plastic particles to review in this study.

Microplastics can be categorized as primary or secondary plastics. The primary microplastics are intentionally manufactured microplastic particles for particular applications (for example microbeads); secondary microplastics are created by fragmentation and degradation of macroplastics, including fibres from synthetic textiles (GESAMP, 2016; Zeng, 2018). Such a distinction is of possible importance to the study of atmospheric transport due to the difference in shape that may affect its aerodynamics and therefore atmospheric transport. There is strong evidence that microplastics are entering into the environment at all

steps in the life cycle of a plastic product - from producers to waste management, with the potential for trophic transfer and human health exposure (Bank and Hansson, 2019; GESAMP, 2016).

Microplastics have been found in quite diverse media, from soils to aquatic systems (e.g., oceans, rivers, shorelines, and swamps), and digestive tracts of both vertebrates and invertebrates (Auta et al., 2017; Li et al., 2018; Prata et al., 2019; Ribeiro-Claro et al., 2017; Rochman, 2015). The majority of research to date has focused on the marine environment; however, attention is increasingly being paid to other environmental compartments (Bank and Hansson, 2019; Horton and Dixon, 2018). The atmosphere is an important pathway by which many suspended materials are transported regionally or globally (Camarero et al., 2017; GESAMP, 2016). Recent studies have illustrated that atmospheric microplastic particles can be transported to ocean surface air and even remote areas (Allen et al., 2019a; Ambrosini et al., 2019; Klein and Fischer, 2019; Liu et al., 2019a, 2019b; Zhang et al., 2019). The atmosphere includes kinds of processes, e.g. the wind speed and directions, up/down drafts, convection lift and turbulence. Therefore, they are considered as important vectors to affect microplastic transport, and which further influence the flux mechanism and source-sink dynamics of plastic pollution in both marine and terrestrial environments (Bank and Hansson, 2019; Liu et al., 2019b; Zhang et al., 2019). Currently, due to their inhalation and combination with other pollutants (mercury or PAHs), microplastics are thought to be an emergent component of air pollution (Barboza et al., 2018; Gasperi et al., 2018; Liu et al., 2019; Rochman et al., 2019; Tourinho et al., 2019; Wright and Kelly, 2017).

Compared to the plethora of microplastics studies in marine environment and growing number of studies in terrestrial environments (Alimba and Faggio, 2019; Auta et al., 2017; Prata et al., 2019), research on atmospheric microplastics has only recently gained attention. To date, very few studies have been conducted on atmospheric microplastic. The majority of studies so far published focus on atmospheric deposition, a passive collection of deposited material at a selected location. Several studies have been longitudinal (extending over multiple seasons up to 12 months) (Dris, 2016; Klein and Fischer, 2019) but extended or long-term monitoring and a global perspective of atmospheric microplastic pollution has yet to be undertaken.

The characteristics, including abundance, size, shapes, and components haven been studied and reported for urban, suburban, and remote areas (Allen et al., 2019b; Dris et al., 2015; Klein and Fischer, 2019). Dris et al. (2017) and Liu et al. (2019) investigated fibres in indoor and outdoor air, identifying that indoor dust is a non-negligible source of human exposure to microplastics. To better understand current status of atmospheric microplastics, it is necessary to collate and compare current research findings, to determine the current state of knowledge and to compare atmospheric microplastic characteristics with microplastics from other environments. Furthermore, the potential impact of atmospheric microplastics on transport and deposition to remote areas and humans via food webs is as an emerging global concern. This review presents the state of knowledge in atmospheric microplastic pollution research with a focus on its current progress, knowledge gaps and recommendations to support standardized and comparable future research.

2. Microplastic analysis methodology

2.1. Sample collection

The majority of the published atmospheric microplastics research to date has been undertaken using a passive collector (total deposition), described in the methodologies published by Allen et al. (2019a), Cai et al. (2017), Dris et al. (2017) and Klein and Fischer (2019). Early studies used non-standardized collection equipment, collecting a range of wet and/or dry deposition for varying periods and precipitation quantities. However, recent advances in passive sampling of atmospheric deposition have resulted in a metallic/glass standardized system designed by NILU (Norwegian Institute for Air Research). This system provides a plastic-free standardized method for passive atmospheric deposition, which is ideal for microplastic research. The benefits of these total or bulk deposition samplers is ease of use, methodology standardization and no requirement for power to the study site. Implementation of this standardized sampling method allows studies to be performed in remote locations with minimal infrastructure at a very low cost but to a standard protocol for collection. Another reason for using a standardized sampler is that the volume of blowby (wind lifting particles out of the collection funnel before entrapment) is a known volume which allows for comparison to other deposited material in addition to other plastic studies.

Road and indoor dust have been sampled using different sampling methods, e.g., sweeping, vacuum, and active pumped sampling, which may cause difficulty in data comparison. Liu et al. (2019) collected indoor dust deposition using hog bristle brushes and transferred to sample bags as completely as possible (unknown amount of material retained in the brush). While this method is easily replicable, it is difficult to determine the relative quantity of air sampled or whether the collected microplastics were solely atmospheric deposition. Abbasi et al. (2017) investigated road dust for heavy metals, microplastics and mineralogical characteristics, collecting sampled using a dustpan and brush. Similarly, Dehghani et al. (2017) collected road dust for microplastic analysis using an anti-static wooden brush. This study was careful to note the meteorological conditions prior to and during the sampling, selecting sampling times with specific dry periods preceding the sample time periods to try and provide an indication of the duration of dry deposition. This is useful in further comparative analysis of microplastic deposition (MP m^{-2} d^{-1}); however, it is difficult to directly compare these findings to true atmospheric deposition collectors (such as the NILU collector) as the quantity of residual microplastic left on the sample surface is unknown.

Active pumped samplers, effective atmospheric microplastic sampling method, are successful used in sampling known volumes of air over defined periods at selected locations (Hayward et al., 2010). This is a highly effective sample collection method that follows a standard protocol for collection, can be correlated to site specific meteorological conditions and known terrestrial/ocean surface conditions. Active pumped air sampling is an established method for atmospheric pollution monitoring (microplastic and other established atmospheric pollutants), used over the past decade and more to monitor atmospheric chemistry such as mercury, lead, carbon and microbes (Dommergue et al., 2019). Dris et al. (2017) used active air pumped sampling methodology to enable a known volume of indoor air to be sampled (filtered). This provides an advancement in standardization of sampling protocol and, while being more intensive in sample resources (electricity and equipment requirements) is highly replicable. In conjunction with the Dris et al. (2017) study with use of active air pumped sampling method, Liu et al. (2019a) used active samplers placed on rooftops, pumping 100 ± 0.1 L/min, to sample Shanghai city air mass microplastic content and in a further study to sample ocean air microplastic in a marine voyage across the China Sea (Shanghai-Mariana Islands study) (Liu et al., 2019b).

Passive atmospheric deposition samplers provide a location and time specific indication of the quantity of microplastic falling onto the surface (e.g. urban road surface, rural field or remote mountain top). Active samplers sample pumped air and therefore provide a sample of microplastics in the air mass rather than deposited microplastic pollution. Active pumped air samples provide an indication of the quantity of microplastics in the air mass that may not deposit. As a result, the use of passive samplers to collect atmospheric deposition (wet and/or dry) is recommended in conjunction with active pumped air sampling to gain a full picture of air MP content. To ensure the validity, rigor and future comparative capacity of all microplastic research published, it is vital to clearly state the following in all field and laboratory studies: the type of

equipment used to sample microplastics; the duration and dates of all sampling (representing the time period); the spatial location of the samples (location and elevation). This information, in conjunction with the equipment analytical limitations (e.g. the limitation on particle size, particle type) will ensure the research findings can be compared to other, international, microplastic studies. Furthermore, use of multiple sampling methods at one location (i.e. air pump + dry/wet deposition sampler) will provide microplastic samples representative of both air mass and deposition, and will enable future scavenging (i.e. by rainfall) to be quantified.

2.2. Criteria for visual identification of microplastics

A great majority of plastics produced globally are based on non-renewable fossil fuel resources (GESAMP, 2016). In general, plastic particles >500 μm are visually identified by their shape and colour under a stereomicroscope, with subsequent confirmation using a chemical analytical method (Hidalgo-Ruz et al., 2012; Nguyen et al., 2019). The technique used for identification of atmospheric microplastics is not entirely the same due to the weathering and size of the particles. However, during identification of atmospheric microplastics, the following guidelines are usually used:

✓ Plastics must have no biogenic (cellular or organic) structures (Dris et al., 2015).
✓ Biofilms and other organic or inorganic adherents have to be removed from the microplastic particles to avoid artefacts that impede clear and accurate identification (Löder and Gerdts, 2015).
✓ Fibres are expected to have a relatively even or consistent thickness along their entire length and illustrate three dimensional bending (Dris et al., 2015).
✓ Fragments and films are expected to have relatively homogeneous colouring and illustrate a level of transparency or clarity (Löder and Gerdts, 2015). However, extremely weathered particles may show strong internal colouring 'spots' with a loss or bleaching of colour at the particle edges and surface.
✓ Aged plastic, such as expected in environmental samples present embrittled and weathered surfaces, and to have irregular shapes with broken and sharp edges (Hidalgo-Ruz et al., 2012). Weathered plastics may also show pitting, gouging and scratched/torn surfaces (Zhou et al., 2017).
✓ Colour can be a plastic identifier and ranges from transparent and variations of white to bright orange, blues, greens and purples through to black (Hidalgo-Ruz et al., 2012; Löder and Gerdts, 2015; Nguyen et al., 2019). Transparent, red and green fibres should be examined with high magnification to confirm their nature (Dris et al., 2015). It is noted that biogenic and plastic material becomes bleached during the sample preparation process (H_2O_2 digestion) that makes coloured plastic particulates less visible and more difficult to differentiate from residual (post digestion) biogenic material (Allen et al., 2019a).

2.3. Sample preparation

2.3.1. Organic matrix removal

There is an ongoing evolution and advancement in sample preparation for microplastic analysis. Early research identified microplastics through visual techniques without sample preparation beyond simple filtration (placing the material onto a filter platform), using colour, shape, size and reaction to heat (hot needle test) as methods to indicate plastic composition (Hidalgo-Ruz et al., 2012; Marine and Environmental Research Institute, 2015; Silva et al., 2018). As microplastic research has extended beyond simple sample matrices and the analytical methods have advanced to allow smaller particle analysis, it has become necessary to separate small microplastics, <500 μm, from the remaining sample material. This is particularly important when

samples include significant organic material as the organic matter creates interference in spectrographic analysis; causing increased noise in the spectra, screening and bio-coating of plastic particles (Löder and Gerdts, 2015). Organic removal has been undertaken through a variety of methods, including KOH, NaOH, HNO_3, HCl, H_2O_2, H_2O_2 + H_2SO_4, H_2O_2 + Fe, and enzymatic methods (Hanvey et al., 2017; Löder et al., 2017; Renner et al., 2018). Atmospheric deposition studies have used sodium hypochlorite (NaClO) or hydrogen peroxide (H_2O_2) as digestion methods for organic removal to date (Allen et al., 2019a; Dris, 2016; Klein and Fischer, 2019; Stanton et al., 2019). A level of consensus regarding effective methodology is growing in microplastics research in general, using a form of H_2O_2 digestion in controlled temperature environment for a selected period (relative to the quantity of organic material). Recent research has identified Fenton's reagent as an effective advancement in sample preparation methodology (Hurley et al., 2018; Prata et al., 2019; Tagg et al., 2017).

2.3.2. Density separation

The methods of sample preparation are not yet standardized, and a variety of organic removal and density separation methods have been used. Density separation is relatively simple, requiring material to be suspended or settled in liquid of various densities. Density separation has been undertaken using freshwater (1.0 g mL^{-1}), sea water (1.03 g mL^{-1}), sodium chloride (>1.2 g mL^{-1}), calcium chloride (>1.35 g mL^{-1}), sodium polytungstate solution (>1.5 g mL^{-1}), sodium bromide (>1.6 g mL^{-1}), zinc bromide(>1.7 g mL^{-1}), zinc chloride (>1.7 g mL^{-1}) and sodium iodide (>1.8 g mL^{-1}) (Li et al., 2018; Quinn et al., 2017). To date, density separation for atmospheric samples has been completed zinc chloride (S. Allen et al., 2019; Dris et al., 2017). It is noted that when settling fine dust particles in atmospheric deposition (Saharan dust and similar material) it was necessary to lightly agitate the settling tubes (60 rpm) to prevent collation of fine dust on/around microplastic particles and subsequent loss of microplastic sample material through deposition.

2.4. Analytical measurements

2.4.1. Visual methods

Early atmospheric microplastic research involved a simple visual microscopic reporting of plastic presence and quantification (Dris et al., 2015). While effective for large obvious microplastic particles, it can be difficult to accurately determine if particulates are plastics when considering particles <500 μm (Käppler et al., 2016; Silva et al., 2018). This severely limits the size fractions which can be examined using only visual reporting, because the abundance of microplastic particles appear to increase almost exponentially with decreasing particle size (Araujo et al., 2018). More recent studies have used visual identification coupled with confirmation of plastics presence (as opposed to organic or inorganic material) by various hot-needle techniques (Hendrickson et al., 2018; Hidalgo-Ruz et al., 2012; Marine and Environmental Research Institute, 2015; Silva et al., 2018). However, visual identification and sorting of microplastic is strongly affected by human bias, microscopy quality, sample matrix and size limitation due to microscope resolution (Hidalgo-Ruz et al., 2012; Li et al., 2018). For particles <500 μm, it is recommended that non-visual, spectroscopy methods be used to determine if particles are plastic (overview of microplastic analysis methods is provided in Table 1).

2.4.2. Thermochemical methods

The use of pyrolysis coupled with mass spectrometry is one of the non-visual methods to determine microplastic in a sample. To date, pyrolysis mass spectrometry (e.g. Py/GC/MS) analysis has not been undertaken on atmospheric samples. Py/GC/MS can identify the type of plastic in a sample (e.g. PET, PVC, PE) and the concentration of this plastic type (ppb) through thermoanalytical methods. It is not possible to define the number of particles or the shapes using this method, and

Table 1
A summary of sampling methods for atmospheric microplastics from previous studies.

Sampling method	Related equipment	Types	Study area	References
Active sampling	1. Palmex Rain Sampler with a sampling area of 0.014 m2 (diameter of 135 mm) constructed of ultraviolet-resistant polyvinyl chloride (PVC) and stainless steel	Dry and wet atmospheric fallout	the Pyrenees mountains, remote area	Allen et al., 2019
	2. NILU Particulate Fallout Collector (p.no. 9721) with a sampling area of 0.03 m² (diameter 200 mm) constructed of high-density PE and stainless steel			
	A pump (Stand-alone sampling pump GH300, Deltanova, France) allowed to sample 8 L/min of indoor air on quartz fibre GF/A Whatman filters (1.6 mm, 47 mm)	Dry deposition	Paris, France	Dris et al., 2018, 2017
	Suspended atmospheric microplastics were collected using a KB-120F type intelligent middle flow total suspended particulate sampler (Jinshida, Qingdao) with an intake flow rate of 100 ± 0.1 L/min.	Dry deposition	Shanghai, China & West Pacific Ocean	Liu K et al., 2019a, 2019b
	The dust collection system comprised a low-volume sampler unit and a filter changer with an intake tube and sampling head (inlet) to collect PM from air that is drawn through a size-selective inlet and through the filter media.	Air	Asaluyeh County, Iran	Abbasi et al., 2019
Passive sampling	A sampling device equipped with a glass bottle	Dry and wet deposition	Yantai, China	Zhou et al., 2017
	A sampling device was equipped with a glass bottle (30 cm × Φ15 cm, i.e., opening area is 0.0177 m2, volume is 5.31 L) and a fixed support	Dry and wet deposition	Dongguan, China	Cai et al., 2017
	A funnel in a 20 L glass bottle on the rooftop of Paris-Est Creteil University	Wet and dry deposition	Paris, France	Dris et al., 2015; Dris et al., 2016; Dris et al., 2017
	Total particulate samplers consists of a 150 cm long PVC-pipe, a PE-funnel and a 2 l PE-bottle. 1. The indoor dust was collected from 4m² of floor in each bedroom and 4m² in the living room using hog bristle brushes. 2. The outdoor dust was collected simultaneously from the windowsills and open-air balconies connected to the apartments following the same protocol.	wet and dry deposition indoor and outdoor dust	Hamburg, Germany 39 major cities of China	Klein and Fischer, 2019 Liu C et al., 2019

thermoanalytical methods are by nature destructive. The quantity or size of plastic particles necessary within the sample to obtain a clear results has been suggested as 100 μm (Fries et al., 2013; Gillibert et al., 2019; Käppler et al., 2016; Löder and Gerdts, 2015). However, there have been significant recent advancements in pyrolysis methods coupled with spectrometry techniques have been used to identify smaller quantities of particles in environmental and laboratory experiment samples (Fischer and Scholz-Böttcher, 2017; Käppler et al., 2018; Materić et al., 2019; Materić et al., 2017). These advancements, including the use of thermal desorption (TDS-GC/MS) coupled with thermogrametric analysis (TGA) and solid phase extraction may provide enhanced analysis. TDS-GC/MS may enable identification of sample composition in environmental samples with very small particles and plastic quantities and are potentially usefully atmospheric microplastic analysis methods in the future (David et al., 2018; Dümichen et al., 2017; Dümichen et al., 2015; Renner et al., 2018).

2.4.3. FTIR spectroscopy

Both FTIR and Raman spectroscopy measure the reaction of the various chemical bonds in materials to an energy (light) source (Centrone, 2015). The use of vibrational spectroscopy for atmospheric microplastic started with Dris et al. (2016) where Attenuated Total Reflectance Fourier Transform Infra Red (ATR-FTIR) and micro spectroscopy was used to study fibres (with a minimum size limit of 50 μm). FTIR has been used extensively as a tool for characterization and more recently mapping for particle counting and size distribution (Bergmann et al., 2019; Primpke et al., 2017). FTIR determines a particles composition (it's molecular structure) through examination of the sample using an IR wavelength range of 400–4000 cm^{-1}. A proportion of the wavelengths are absorbed by the particle being analysed. By determining which wavelengths were absorbed and transforming the absorption using the Fourier Transform function a spectrum describing the particles composition is created. This spectrum is cross referenced against reference libraries and/or analysed for its individual chemical structure to define the particle composition (Everall et al., 2007; Ribeiro-Claro et al., 2017).

The early popularity of FTIR for all microplastics research potentially stemmed from the easy to use libraries and ease of analytical and equipment operation. The advantage of FTIR is a higher throughput (compared to hot-needle and visual analysis), the ability to analyse a smaller particle size (below 500 μm), characterize it and automation of particle spectral analysis through polymer spectral libraries. FTIR's long history of use for polymer industry quality control offers substantial library spectra of virgin polymer types though environmentally aged plastics spectra are often not as clear. Some early studies used Attenuated Total Reflectance (ATR) to gain spectra from a particle. This system requires placing the individual particles (particles large enough to be manipulated using tweezers) between two points before running the analysis. The minimum size that can be physically manipulated for ATR limits its practical use for atmospheric microplastics (Dris, 2016, 50 μm).

FTIR equipped to include a confocal microscope (known as μFTIR) and focal Plane Array (FPA) with Mercury Cadmium Telluride (MCT) liquid nitrogen cooled detectors has reduced the practical particle size down toward the diffraction limit. For infrared (IR) this limit is theoretically 10 μm (particle diameter) as the whole wavelength must pass through the material, however given the normally weathered surface of environmental microplastic samples it is difficult to get reliable signals below ~20 μm, especially when automated (Gillibert et al., 2019). The application of FPA μFTIR for atmospheric microplastic is limited by this diffraction limit and has been shown to illicit 35% underestimation of particles below 20 μm (Käppler et al., 2016). MCT detectors must be cooled using liquid nitrogen to minimize the noise created by dark energy passing through the detector. This means the liquid nitrogen dewar must be maintained at least every 8 h for most machines.

2.4.4. Raman spectroscopy

Though both Raman and FTIR are considered vibrational spectroscopy, Raman is different to FTIR in which it uses a higher frequency (normally 532 nm) laser to excite the surface of a material until it emits photons. The photons are normally emitted in line with the laser (Rayleigh scatter) but 1 in 10–7 photons are emitted at right angles and are known as Raman scattering. Raman is relatively new to microplastics research and does not have the history in industrial polymer research, as such the libraries are not yet well developed. The theoretical limitations of Raman are sub-micron however 10 μm is the current smallest published due to surface weathering and the energy imparted to the particle can be destructive; however it is expected to reduce to 2 μm with improved techniques (Allen et al., 2019a; Araujo et al., 2018). The smallest atmospheric plastic particle using μFTIR is 11 μm (Vianello et al., 2019).

The main issue with short wavelength Raman lasers is fluorescence. The wavelengths traditionally used for Raman are very close to many maximum excitation wavelengths which causes the particle to fluoresce. This fluorescence obfuscates the signal denying analysis of the chemical bonds. To get past this, studies are now using a near IR laser of 785 nm (Allen et al., 2019a). This has an impact on particle sizes and more power is needed to elicit the same Raman response which may increase the possibility of particle destruction. Though initial purchase costs of μRaman is higher than the equivalent μFTIR, the Raman generally uses Thermo Electrically Cooled (TEC) detectors which obviates the necessity for liquid nitrogen cooling. This simplifies operation, reduces costs and makes it possible to operate overnight without supervision increasing runtime per day. It is thought that with further library and technique development that μRaman will be the preferred tool for atmospheric microplastics due to the smaller particle size in this emerging field. Recent advances in nano-FTIR, Raman Enhanced Atomic Force Microscopy (RE-AFM), and Raman tweezers (RT) may change this with particle characterization as small as 20 nm (Gillibert et al., 2019; Huth et al., 2012; Meyns et al., 2019). Raman tweezers use the same principle as optical tweezers (OT) which traps particles in liquid with the force of interactions with light between the tips and particle. RT adds the ability to gain a spectra from the particles as well as size and shape making it a promising technique. It is noted that the material must be suspended in liquid which may make it unsuitable for all atmospheric samples types as suspension in an added liquid may contaminate. It is noted by Meyns et al. (2019) that Nano-FTIR has trouble identifying polystyrene which will limit the applicability for environmental samples. Though it may be possible to characterize nanoplastic on these machines, current technology means a very slow throughput and significantly higher costs.

μFTIR and μRaman spectroscopy (FTIR and Raman advanced to allow microparticle analysis rather than *meso* or macro particle analysis) enables the analysis of small microplastics directly on filters without any visual pre-sorting and open the possibility for automatization. The current preferred filter is Anodisc (aluminium oxide) with pore size of 0.2 μm (Allen et al., 2019a; Bergmann et al., 2019). However, it is noted that Anodisc is active in the near IR wavelength (785 nm) making sub 10 μm analysis difficult. The use of plastic filter types (such as polycarbonate, PTFE, etc) are discouraged for FTIR as in transmittance the light must pass through the filter. Similarly, when approaching the detection limits stray light from the Raman laser may gain signal from the filter and obscure the signal from the target particle. Cellulosic type filters tend to warp when drying which makes automation of focus difficult, so they are not recommended for either technique. Both μFTIR and μRaman are effective in identifying >20 μm microplastic particles, with advances enabling particle mapping and automated counts. Compared with μFTIR spectroscopy, μRaman techniques can theoretically analyse particles down to sub-μm (diffraction limit of 250 nm compared to μFTIR at ~10 μm), providing a higher resolution analysis for these increasingly important small microplastic particles (Araujo et al., 2018; Renner et al., 2018). The practical limit of

μFTIR on aged microplastics appears to be around 20 μm without substantial extra effort and expertise provided by the analyst. Käppler et al. (2016) reports that a significant amount (35%) of small microplastics (<20 μm) are lost (underestimation of MP) during μFTIR analysis compared to μRaman imaging. The μFTIR analysis has the benefit of extensive polymer library resources for identification and comparative analysis. However, as μRaman use increases it is expected a similar library resource will evolve, providing an identification and comparison functionality equivalent to the current μFTIR detail and availability. As a result, μRaman provides a slight advantage at present with regards to the lower limitation of particle size analysed and in the cost to perform the analysis.

It is recommended that atmospheric microplastic analysis can be undertaken using a spectroscopic analysis methodology due to visual methods being ineffective for small particle sizes such as those found in atmospheric deposition samples. It is recommended that spectroscopy, μRaman or μFTIR, be used to characterize and quantify atmospheric microplastics to aid in understanding sources and fates of this material. Analysis of microplastic <1 μm in environmental samples is still in the early stages of technological advancement. Further research to support sub-micron microplastic analysis techniques is essential. Development of thermochemical (Py-GCMS, TD) analysis methods may provide an avenue to identifying concentration and plastic type in samples containing microplastics <1 μm, acknowledging information on particle counts and shapes are not possible following these destructive methodologies.

3. Current knowledge of atmospheric microplastics

3.1. Occurrence and abundance

The spatial distribution of atmospheric microplastic studies, presented in Fig. 1, illustrates the new and developing nature of this research focus. Using a number of the different sampling and analytical methods discussed, the published atmospheric MP studied (Fig. 1) have identified, quantified and characterized atmospheric microplastics at these remote to urban or industrial locations.

The overall abundance for atmospheric microplastics are presented in Table 2. The average abundance of atmospheric microplastics varied greatly among different studied areas. In the European cities, the mean microplastic abundance from dry and wet deposition has been found between 118 (Paris) and 275 (Hamburg) particles $m^{-2} d^{-1}$ (Dris et al., 2016; Dris et al., 2015; Klein and Fischer, 2019). While in Dongguan city of China, the abundance of non-fibrous microplastics and fibres ranged from 175 to 313 particles $m^{-2} d^{-1}$ in the atmospheric deposition (Cai et al., 2017). Deposition flux of atmospheric microplastics in Yantai (a coastal city) of China attained a maximum of 602 particles $m^{-2} d^{-1}$ (Zhou et al., 2017). In the remote area of the Pyrenees Mountains, the result illustrates an average microplastic particles deposition of 365 particles $m^{-2} d^{-1}$ (Allen et al., 2019a). Atmospheric microplastics observed from urban cities to remote pristine areas further indicates that microplastic pollution has become a global issue (Bank and Hansson, 2019).

The deposition rate of fibres in indoor environments of Paris is between 1586 and 11,130 fibres $m^{-2} d^{-1}$ with an abundance of fibres between 0.3 and 1.5 particle m^{-3} (Dris et al., 2017). Abundance of suspended atmospheric microplastics in Shanghai from filtered air ranges from 0 to 4.18 particles m^{-3}, with an average level of 1.42 ± 1.42 particles m^{-3} (Liu et al., 2019a). Higher abundance was observed in Shanghai compared to Paris, possibly due to more anthropogenic activities, population densities, and industrialization levels.

Precipitation (wet deposition, including rainfall and snowfall) events may be a positive drivers in atmospheric microplastics deposition (Allen et al., 2019a; Dris et al., 2016). Micro/nano plastic was recently shown by Ganguly and Ariya (2019) (laboratory study) to be
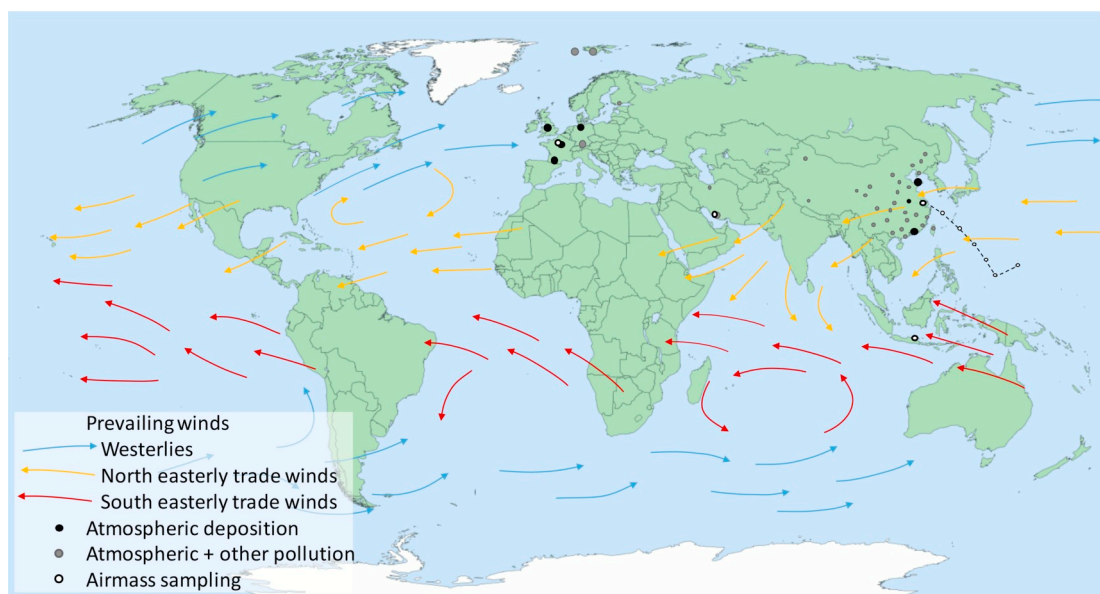
Fig. 1. A mapped representation of the atmospheric microplastic studies published to date. For further details on plastic quantities and characterization in these studies, refer to Fig. 2 and Table 1.

an efficient cloud ice nuclei which may explain the slight correlations with MP counts to snow events from Allen et al. (2019b). Snow is considered as a positive method of microplastic deposition (atmospheric particle scrounging) depositing atmospheric particulates in urban areas and on the sea or Arctic regions. The recent study by Bergmann et al. (2019) indicates that deposition of microplastics ranged from 190 to 154×10^3 particles L^{-1} and $0–14.4 \times 10^3$ particles L^{-1} in melted snow sampled from Europe and the Arctic, respectively. This abundance was suggested by the authors (Bergmann et al., 2019) to be 4–7 orders of magnitude higher than concentrations previous reports from Dongguan and Paris (Cai et al., 2017; Dris et al., 2017). A large proportion of this discrepancy is expected to result from significant differences in methodology, most specifically the limit of particle size analysed (11 μm in the Arctic study, ~50 μm in Paris and ~200 μm in Dongguan). Rain and snow are thought to be effective scavenging mechanisms for aerosol particles and the findings in these studies emphasize the need for event specific sampling enabling individual rain/snowfall event atmospheric deposition to be determined, and for short time period dry deposition sampling to occur to support meteorological correlation to atmospheric deposition. Spatially, the Artic and Pyrenees studies illustrate the encroachment of atmospheric microplastic pollution in remote areas. These studies provide a new perspective on transport of atmospheric microplastics.

Outdoor abundance of microplastics is significantly lower than that of indoor environments. Dris et al. (2017) found indoor concentrations ranged between 1.0 and 60.0 fibres m^{-3}, while outdoor concentrations ranged between 0.3 and 1.5 fibres m^{-3}. In China, outdoor atmospheric microplastic abundance has been reported as up to 4.18 particles m^{-3} (Shanghai) and in Surabaya, Indonesia, up to 174 particles m^{-3}. In the indoor dust samples from 39 major cities of China, microplastics (detected PET) abundance ranges from 1550 to 120,000 mg kg^{-1} with a median abundance of 26,800 mg kg^{-1} (Liu et al., 2019). These findings show that indoor microplastics may be an important source of atmospheric microplastics and contribute to atmospheric deposition (Dris et al., 2017).

At present, one of the major limitations of current perspectives on microplastic pollution research is the lack of harmonization or standardization of data and methodologies that are widely used within the research community. However, this is improving as scientists have now developed formal definitions to ensure transferability and reproducibility of research results and greater clarity in the reporting structure

(detailing sampling and analytical methods and their limitations) (Hartmann et al., 2019). Correlation to meteorological conditions and sample period representation are further important study data that need to be considered in the design and provided in the reporting of future atmospheric microplastic research.

3.2. Physical characterization: shapes, size, and colours

3.2.1. Shapes

Microplastics in the environment appear in a wide diversity of shapes and size (Rocha-Santos, 2017). Frequent description of microplastic shapes includes spheres, beads, pellets, foam, fibres, fragments, films, and flake (Hidalgo-Ruz et al., 2012). These shapes depend on the original form of primary microplastics, the degradation and erosion processes of plastic particle surface, and residence time at the environment. It has been suggested that degraded microplastics with sharp edges illustrate a recent introduction into the environment while smooth edges are associated with a large residence time (Hidalgo-Ruz et al., 2012; Rocha-Santos, 2017). Diverse shapes including fibre, foam, fragment, and film have been detected in the atmospheric microplastics proved by previous studies. In Dongguan, Shanghai, Yantai, and Paris (urban centres), fibres were the dominant shape (>60%) for the atmospheric microplastics (Table 2). However, in Hamburg, the dominant shape of atmospheric microplastics detected were fragments, contributing to 95% of the total particle numbers and only 5% comprised fibres (Klein et al., 2019). Atmospheric deposition studies in remote areas also suggested dominant fragment shape (Allen et al., 2019a; Bergmann et al., 2019). In Iran, fibrous (33.5%) and granule (65.9%) microplastics were the most abundant shapes in street dust (Abbasi et al., 2019; Dehghani et al., 2017).

The shape of microplastics has been often used to infer their origin and pathway because certain shapes may be more prolifically shed from particular products (Helm, 2017; Rochman et al., 2019). Fibres, for example, are the dominant shape found in the urban atmospheric deposition of Shanghai, are likely closely connected to the increasing production of synthetic fibre (clothing, upholstery, or carpet); while fragmented microplastics could possibly result from the exposure of larger plastic items to strain, fatigue, or UV light (K. Liu et al., 2019a). Surface texture (e.g., adhering particles, grooves pits, fractures and flakes) of fibres indicate that mechanical abrasion and chemical weathering might play a key role on the degradation of microplastics in

Table 2
A summary on characteristics of microplastics in possible atmospheric depositions in the previous studies. Studies that are a mixture of both atmospheric and other sources of microplastics (e.g. road pollution) are indicated with an asterix *.

Study area	Sample types	MP abundance	MP size	Shapes	Colours	Composition	Method	References
Paris, France	Total atmospheric fallout (dry&wet deposition)	29–280 particles m^{-2} d^{-1}; Ave: 118 particles m^{-2} d^{-1} (110 particles m^{-2} d^{-1} in urban, 53 particles m^{-2} d^{-1} in suburban)	100–500 μm; 500–1000 μm; 1–5 mm (50% fibres >1000 μm)	>90% fibres; ~10% fragments	Blue; Red	N/A	Stereomicroscope; μFT-IR	Dris et al., 2015
Paris, France	Atmospheric fallout	2.1–355.4 fibres m^{-2} d^{-1}	50–200 μm:3%; 200–600 μm: 42%; 600-1400 μm: ~40% (50–4850 μm)	Fibres	N/A	29% synthetic	Stereomicroscope; μFT-IR	Dris et al., 2016
Paris, France	Indoor and outdoor air	Outdoor: 0.3–1.5 fibres m^{-3} (1586–11,130 fibre m^{-2} d^{-1}); Indoor: 1–60 fibres m^{-3}	50–450 μm: >80%	Fibres	N/A	PP	Stereomicroscope; μFT-IR	Dris et al., 2017
Hamburg, Germany	Atmospheric deposition	275 particles m^{-2} day^{-1} (Range: 136–512 particles m^{-2} day^{-1})	<63 μm: ~60%; 63–300 μm:~30%; >300 μm: ~20%; Fibres: 300–5000 μm: 68%; 63–300 μm: 25%; <63 μm: 7%	Fragment: >90%; Fibres: <10%	N/A	PE: 48.8%; EVAC: 22%; PTFE; PAV	μ-Raman	Klein and Fischer, 2019
* Tehran metropolis, Iran	Urban dust	88–605 items per 30 g dry dust	Predominant: 250–500 μm (100–1000 μm)	Granule dominant60%; Fibres:35%; Sphere:5%	Black, grey, orange, yellow, white, transparent, green, blue, red, pink	N/A	Fluorescence microscopy; SEM/EDS	Dehghani et al., 2017
* Asaluyeh County, Iran	Suspended dust, urban dust	0.3–1.1 item m^{-3}	100–1000 μm:	Fibres; Granules	White-transparent: >70%; blue-green:; Yellow-orange; Black	N/A	Fluorescence microscopy; SEM/EDS	Abbasi et al., 2019
Dongguan, China	Atmospheric fallout (dry&wet deposition)	175–313 particles m^{-2} d^{-1} (natural fibres 73%; PP 9%, PE 14%)	majority of fibres to be 200–700 μm in length (200–4200 μm)	Fibres (80%); foams; Films	Blue; Red; Transparent; Grey	PE, PP, PS	Stereomicroscope; μ-FTIR	Cai et al., 2017
Shanghai, China	Suspended atmospheric microplastics	1.42 ± 1.42 items m^{-3} (maximum 4.18 items m^{-3})	23-500 μm: >50% (23–5000 μm)	Fibres: 67%; Fragment: 30%; Granules: 3%	black, blue, red, transparent, brown, green, yellow, and grey	PET, PE, PES, PAN, PAA, and Rayon	Stereomicroscope; μ-FT-IR analysis	Liu K et al., 2019a
Yantai, China	Atmospheric deposition	Fibres: 115–602 items m^{-2} d^{-1}; Others: 40 items m^{-2} d^{-1}	<500 μm: 50% (100–300 μm dominant) followed by 0.5–1 mm	Fibres:95%; Film; Foam	N/A	N/A	Stereomicroscope; μ-FT-IR analysis	Zhou et al., 2017
* 39 major cities in China	Indoor and outdoor dust	PET: 1550–120,000 mg kg^{-1} (indoor); 212–9020 mg kg^{-1} (outdoor); PC: 4.6 mg kg^{-1} (indoor); 2.0 mg kg^{-1} (outdoor)	N/A	Fibres dominant: 75% outdoor; 85% indoor; Granule; Cellulose and Rayon: natural sources	N/A	PET: >60%; PC, Nylon, PE, PP, PMMA, PU, PEI, Alkyd	μ-FT-IR	Liu C et al., 2019
Pyrenees mountains, Europe	Atmospheric dry & wet deposition	249 fragments, 73 films and 44 fibres m^{-2} d^{-1}	predominant fibre lengths of 100–200 μm and 200–300 μm (10–5000 μm)	Fragments; 68% fibres; films	N/A	PS, PE, PP, PVC, PET	Stereomicroscope; μ-Raman	Allen et al., 2019

Table 2 (*continued*)

| Study area	Sample types	MP abundance	MP size	Shapes	Colours	Composition	Method	References
* Europe and Arctic	European snow, Arctic snow (wet deposition)	190–154 × 10³ items L⁻¹ European snow; 0–14.4 × 10³ items L⁻¹ Arctic snow	11–150 μm; 11–250 μm; more than 80% ≤ 25 μm (11–475 μm)	Fibres	N/A	Varnish, Nitrile rubber, PE, Polyamide rubber	μ-Raman, FTIR imaging	Bergmann et al., 2019
* Helsinki, Finland	Urban snow (wet deposition)	700 items m⁻² of melted snow (market place); 1400 (road side); 16,600 (residential area)	0.3–4 mm	N/A	N/A	PE, PP	FTIR	Pikkarainen et al., 2019
* Italian Alps	Supraglacial debris	74.4 items kg⁻¹ of sediments (dry weight)	N/A	N/A	Black 31%, Blue 22%, Red 17%, Transparent 17%, Light blue 9%, Violet 4%	Polyester 39%, PA 9%, PE 9%, PP 4%, Unknown 39%	μ-FTIR	Ambrosini et al., 2019
Surabaya, Indonesia	Suspended atmospheric microplastics	132.75–174.97 items m⁻³	<500 μm: 5%, 500–1000 μm: ~13%, 1000–1500 μm: ~30%, 1500–2000 μm: ~14%, 2000–2500 μm: ~20%, 2500–3000 μm: ~7%, 3000–3500 μm: ~2%, 3500–4000 μm: ~2%, 4000–4500 μm: ~3%, 4500–5000 μm: ~4%	Fibres dominant, fragments, films, pellets	NA	PE, PET, cellophane	FTIR	Asrin and Dipareza, 2019
* Nottingham, UK	Atmospheric wet and dry deposition, urban dust	0–31 fibres m⁻² d⁻¹	38 μm–5 mm	fibres	NA	Acrylic, polyamide, polyester, polupropolene	FTIR	Stantin et al. 2019
West Pacific Ocean (Open Ocean)	Suspended atmospheric microplastics	0–1.37 items m⁻³; Ave.: 0.06 items m⁻³	More than 50% <500 μm (20 μm - 2 mm)	Fibres: 60%, Fragment: 31%, Granule: 8%, Microbead	Black, blue, brown, green, grey, orange, pink, purple, red, transparent, white, yellow	PET 57%, PE: 10%, PE-PP: 6%, PES, AlK, EP, PA, PAN, Phe, PMA, PP, PS, PVA, PVC	Stereomicroscope, μ-FTIR	Liu K et al., 2019b
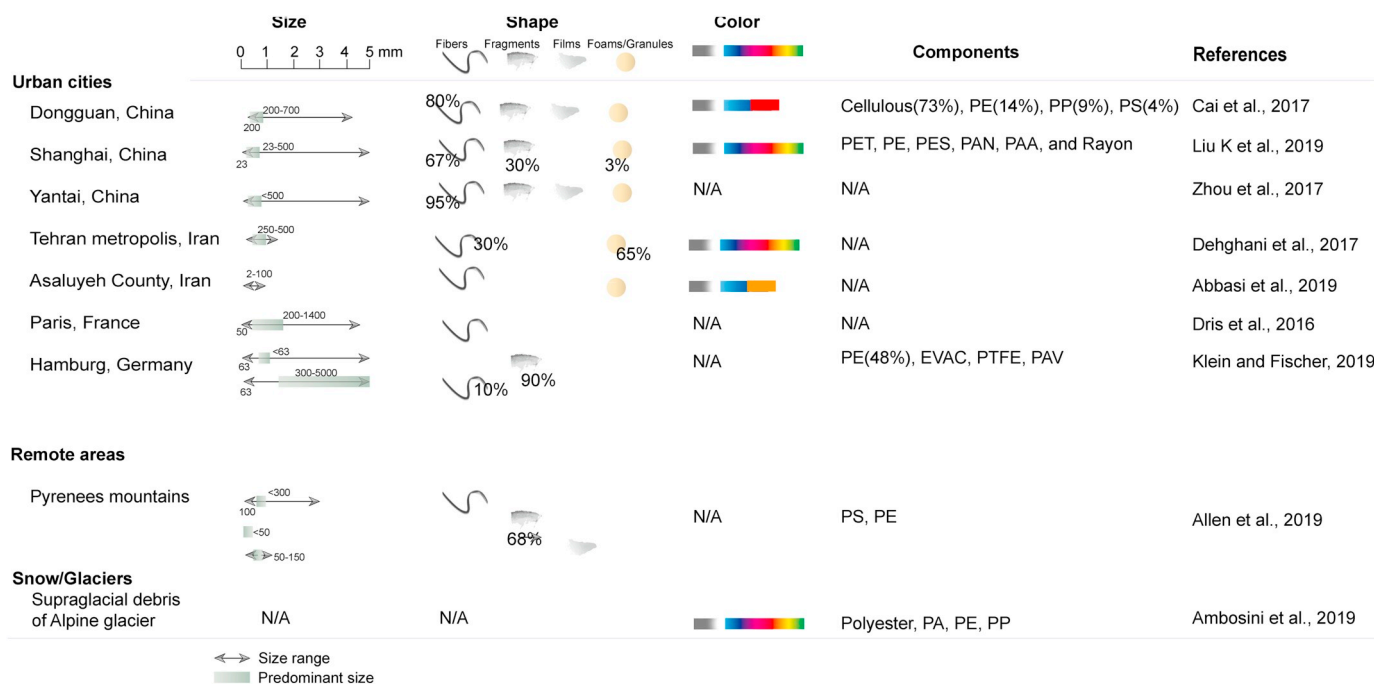
Fig. 2. A summary of characteristics of atmospheric microplastics from the literatures.
* It is acknowledged that not all sampled collected in the research represent atmospheric deposition or atmospheric microplastic pollution.

the atmospheric environment (Cai et al., 2017; Zhou et al., 2017). Microplastic fibres can range in thickness and/or width from 1 to ~500 μm (Cole, 2016; Jemec et al., 2016; Napper and Thompson, 2016). It is noted that for small microplastics it can be difficult to identify if the particle is a fibre due to the mechanical and/or chemical degradation of the material resulting in reduction of the fibre length such that the width and length of the fibre are similar. The differentiation between fibre and fragment for smaller microplastics may therefore be ineffective.

The different shapes of microplastics may affect the transport of this pollutant through the environment. For instance, films can be very thin and flat, and therefore provide a greater surface area for atmospheric conveyance relative to fragments of the same mass (Allen et al., 2019a). The influence of shape on atmospheric transport is currently unknown and required further research. Specific shapes or sizes of microplastics may have greater potential to cause physical harm to organisms, with smaller angular particles passing membrane barriers more easily than particles presenting regular surfaces or longer edges (Hidalgo-Ruz et al., 2012; Rochman et al., 2019).

3.2.2. Size

Plastic particle size is a major factor determining the item's interaction with biota and its environment fate (Besseling et al., 2017; Hüffer et al., 2017). Generally, microplastic size limits are operationally defined by the sampling and analysis method (Hartmann et al., 2019). Microplastics encompass a broad range of sizes, which are typically considered to be 1 μm to 5 mm in length (GESAMP, 2016; Hartmann et al., 2019). Compared to microplastics from aquatic and sediment environments (Auta et al., 2017; Hanvey et al., 2017; Prata et al., 2019), the predominant size of atmospheric microplastics is much smaller (Table 2). For example, in the Pyrenees Mountains, the predominant length of plastic fibres was less than 300 μm (~50%) with a greater proportion of fragments (fragment sizes <50 μm, 70% of microplastic particles) within the samples (Allen et al., 2019a). In a European urban city of Hamburg, the majority of fragments were < 63 μm (~60%), followed by 63–300 μm (~30%); while fibres were predominantly between 300 and 5000 μm in length (Klein and Fischer, 2019). Dris et al. (2016) primarily found fibres of 200–600 μm (~40%),

whereas Cai et al. (2017) report predominant fibre lengths of 200–700 μm (~30%). In Yantai and Shanghai, the predominant particle size is <500 μm (~50%) (Liu et al., 2019a). Among the previous studies, the longest atmospheric microfibre identified is ~5000 μm (Cai et al., 2017; Dris et al., 2016). Film and foam size have not been specifically evaluated in the majority of previous atmospheric microplastic research. Only the study in Pyrenees Mountains shows the predominant film diameter is 50–200 μm, larger than the predominant fragment size (Allen et al., 2019a). For snow in European and Arctic regions, 80% of the detected microplastics were ≤ 25 μm, and 98% of all particles were < 100 μm (Bergmann et al., 2019). Such results indicates that amount of microplastic particles decreased with increasing size (Bergmann et al., 2019; Isobe et al., 2015) and that particle size is a highly important aspect of atmospheric microplastic analysis and research.

3.2.3. Colour

Microplastics have been reported in a range colours, including red, orange, yellow, brown, tan, off white, white, grey, blue, green, and so on (Bergmann et al., 2019; Rochman et al., 2019). The most commonly reported are blue and red fibres (Hidalgo-Ruz et al., 2012). Dark, white, transparent, or translucent particles may be underrepresented during visual inspection (Hartmann et al., 2019). Colour is useful to identify potential sources of plastic debris as well as potential contaminations during sample preparation (Hartmann et al., 2019; Rocha-Santos, 2017). Clear and transparent items have been ascribed to polypropylene, white to polyethylene and opaque colours to LDPE (Rocha-Santos, 2017). However, the colour of a plastic particle cannot easily be used to deduce the type or origin. Importantly, colour information can be biased as brighter colours are spotted more easily during visual inspection (Rochman et al., 2019).

In the study of Paris atmospheric microplastics, Dris et al. (2015) pointed out that there was a tendency to overestimate brightly coloured fibres (blue, red) in comparison with other particles because they are more easily recognized. In Shanghai city, atmospheric microplastics were variously coloured including black, blue, red, transparent, brown, green, yellow, and grey particles (Liu et al., 2019a) (Fig. 2). Among them, blue and black microfibres comprised the majority of the

atmospheric microplastics, accounting for 25% and 28% of the total microplastics, respectively. For microplastic from supraglacial debris, both fragment and fibres were of diverse colours, with black and blue dominating (31% and 22%, respectively) (Ambrosini et al., 2019). Optical microscope images of selected polymers from Dongguan city showed atmospheric microplastics have colour of blue, red, grey, and transparent (Cai et al., 2017). Discoloration of microplastics can take place during weathering as well as sample preparation (particularly with oxidative digestion such as H_2O_2), which should be considered in data reporting and interpretation (Allen et al., 2019a; Rochman et al., 2019). It is considered that colour can be helpful in the initial visual assessment of microplastics in atmospheric samples, but due to the predominantly small particle size and often significant weathering of these particles colour analysis is less important than spectral or chemical identification of these microplastics.

3.3. Components

The chemical composition is the most fundamental criterion for defining plastic pollution (Hartmann et al., 2019). Microplastics are also composed of a diverse suite of polymer types (Rochman et al., 2019). A variety of polymers are synthesized and used for domestic and industrial purposes. The structure (backbone) of plastic polymers can define a plastic's physical and chemical properties (categorized to be thermoplastics and thermosets) (PlasticsEurope, 2018). The greatest plastic demand and most highly produced polymer types are polypropylene (PP, 19.3%), low-density polyethylene (LDPE, 17.5%), high-density polyethylene (HDPE, 12.3%), polyvinyl chloride (PVC, 10.2%), polyurethane (PUR, 7.7%), polyethylene terephthalate (PET, also known as polyester, 7.4%), and polystyrene (PS, 6.6%) (PlasticsEurope, 2018). Polymer composition in seawater reported from published literature indicates that PE is the dominant polymer, followed by PP and PS; while PE, PP, PS, and PES are major polymer types on beaches and subtidal sediments (Zeng, 2018).

For the atmospheric microplastics, chemical composition varies over different regions (Fig. 3). The main polymers in the coastal city of Yantai were PET in the case of most of the fibres, PVC in the case of some fibres and films, PE for the fragments, and PS for the foams (Zhou et al., 2017). In Shanghai, synthetic compounds comprised 54% of the observed particles, of which PET, PE, PES, PAN, PAA, and rayon comprised 91% of the microplastics (Liu et al., 2019a). In Dongguan city, microplastics of three different polymers (e.g., PE, PP, PS) were identified (Cai et al., 2017). Microplastics in dust deposition from Chinese major cities were mainly determined as PET and PC (Liu et al., 2019). In the Hamburg city of Germany, PE and ethylvinyl acetate (EVA) copolymers dominated in the atmospheric microplastics samples (48.8% and 22.0%, respectively) (Klein and Fischer, 2019). The predominant plastic found in the samples from remote area of Mountains is PS (as fragments), closely followed by PE (S. Allen et al., 2019). For the snow fallen out in Arctic, polymer types were found to vary extensively; varnish (acrylates), plasticized rubber and polyamides were among the most high identified microplastics. In contrast, in the European snow microplastics composition was primarily (67%) by polyimide, varnish, rubber, EVA, and PE (Bergmann et al., 2019). In the supraglacial debris of an Alpine glacier, most microplastic items were made of polyesters, followed by PA, PE, and PP (Ambrosini et al., 2019). To date there is no clear correlation or explanation for the variability or composition of polymer types in atmospheric samples. Further research is needed to establish if there is a predominant group of polymers occurring in atmospheric microplastic pollution and whether this polymer composition changes due to sample location and particle distance travelled.

3.4. Comparison with microplastics from marine and terrestrial environments

From the above sections, we notice the characteristics of atmospheric microplastics vary widely among studies from the dry and/or wet depositions and sampling of airmasses. This is also true for microplastics in marine environment with a range from undetected to more than 100,000 items m^{-3} (Zeng, 2018). Microplastic abundance
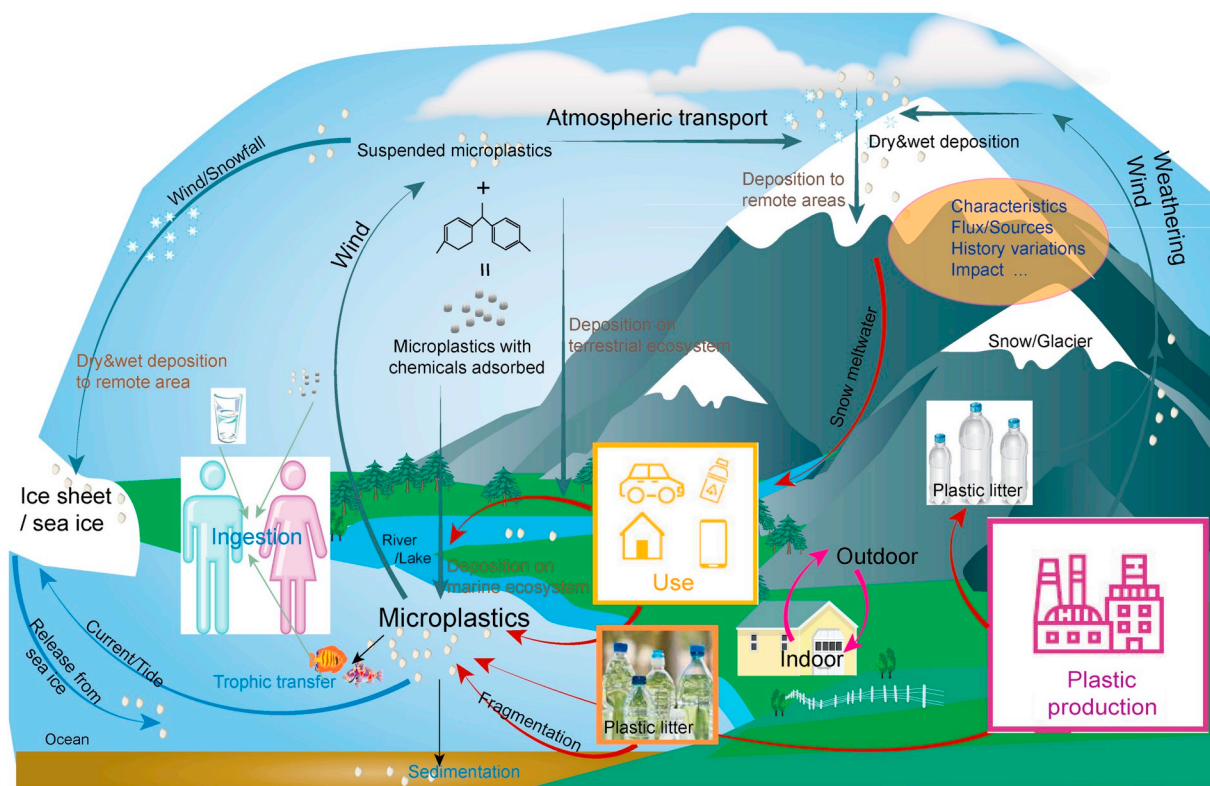

Fig. 3. Conceptual model of atmospheric microplastics in the environment.

tends to increase significantly with decreasing size (Isobe et al., 2015). Meanwhile, the mean microplastic size reported in individual studies depends on the size range of the microplastic sampled and analysed. For example, in aquatic environments, mean size of microplastics ranges from one to a few millimetre for samples collected using nets with mesh size of 200–1000 μm; however, mean size of microplastics collected by using smaller net mesh size (50–63 μm) have shown a mean size of <700 μm (Isobe et al., 2015; Zeng, 2018). For atmospheric microplastics, particle size tends to be much smaller. Fibres observed in indoor and outdoor air are mainly in the lower size range (50–80% between 100 and 500 μm) with only a small proportion being larger than 500 μm (10–30% between 500 and 1000 μm or between 1000 and 5000 μm) (Dris et al., 2017). Fragments observed in the atmospheric deposition concentrated on the range of <100 μm (Allen et al., 2019a; Klein and Fischer, 2019).

Previous studies indicate that fibres and fragment are dominant shapes of microplastics in seawater, beach sediments, and freshwater (Fu and Wang, 2019; Isobe et al., 2015; Zeng, 2018). Such findings imply that secondary microplastics contribute to microplastic abundance more than primary plastics in the marine and freshwater environment. In the atmosphere, fibres and fragment are also the dominant shapes (Fig. 2). Fibres originate from fabric, for example, clothes and textiles (Allen et al., 2019a; Browne et al., 2011; Cai et al., 2017). Fragments are thought to originate from disposable plastics via fragmentation (Allen et al., 2019a; Zeng, 2018). Textile fibres are an important source of indoor dust (Dris et al., 2017; Liu et al., 2019). PET is commonly used to produce polyester fibre, fabric, and cording for textiles (Kuczenski and Geyer, 2010), and this widespread use can help explain the high levels of PET MPs in indoor dust. As shown in Fig. 2 and Table 2, the higher concentration of fibres in indoor air compared to those measured outdoors suggest that a large fraction of the fibres may be transferred to outdoors through the air exchange. This could contribute to atmospheric fallout and indoor atmospheric fibres and particles could also enter the aquatic systems through runoff.

Different polymers have different densities (Hidalgo-Ruz et al., 2012), which can affect the pathway of microplastics into the atmosphere. Less dense polymers, such as PE, PP, and expanded PS, are widespread in the water and atmospheric fallout. The variety of polymer types found in atmospheric samples published to date does not indicate a clear or obvious delineation between less and denser polymer types. Conversely, other polymers with heavier densities (e.g. PS, PVC, PES) have also been observed in atmospheric deposition/air mass sampling. PS and PE are used in many single-use plastic items and in packaging material as indicated by a European Strategy for Plastic in a Circular Economy (Allen et al., 2019a). The link or correlation between marine, terrestrial and atmospheric microplastic composition has not yet been considered in detail, and there is not yet sufficient terrestrial, freshwater or atmospheric microplastic research to provide indications on these interlinkages and source-pathways. While some studies appear to have similar atmospheric microplastic deposition composition to previously published aquatic studies (e.g. Dongguan (Cai et al., 2017)) other studies show significantly different and variable composition (Allen et al., 2019a; Bergmann et al., 2019; Klein and Fischer, 2019).

Shapes and polymers of microplastics show different sorption of hydrophobic contaminants and can facilitate the transport of contaminants (Alimi et al., 2018; Tourinho et al., 2019). In the aquatic or sediment environment, microplastics are always found as part of a mixture or diverse suite of chemicals (Rochman et al., 2019).This indicates that microplastics can adsorb organic chemical and trace metals from the surrounding environment (Hermabessiere et al., 2017; Rochman, 2015). Overall, hydrophobic compounds are attracted to the neutral areas on the microplastic surface, while hydrophilic or charged compounds are attracted to the negative areas on the microplastic surface with electrostatic interactions and media characteristics being most important (Tourinho et al., 2019). In European seabass, microplastics were found to influence the bioaccumulation of mercury

(Barboza et al., 2018). Currently, studies on aggregation, toxicity, sorption of contaminants for microplastics in the atmosphere are sparse. Substantial further research is needed to understand the scope of this issue.

4. Perspectives

4.1. Atmospheric transport of microplastics

Microplastic pollution appears ubiquitous in marine, freshwater, terrestrial and now atmospheric environmental compartments (Allen et al., 2019a; Bergmann et al., 2019; Horton and Dixon, 2018). These environments are interlinked, with a diverse network of source-pathway-sink connections which can influence the flux and retention of microplastics among such environmental matrices. In recent years, atmospheric transport of microplastics has been considered an important vector and that could lead to deposition of microplastics to land or aquatic environments (Allen et al., 2019a; Bergmann et al., 2019; Zhang et al., 2019). Such transportation strongly impacts the source-sink dynamics of plastic pollution in different ecosystems including transfer between terrestrial and marine environment (Bank and Hansson, 2019; Windsor et al., 2019). In Shanghai city and the west Pacific Ocean, a study based on characteristics of atmospheric microplastics suggests that marine microplastics may ultimately derive from terrestrial environments (Liu et al., 2019a). Suspended atmospheric microplastics may be an importance source of microplastics pollution in the ocean, including the pollution caused by textile microfibres (Liu et al., 2019b). Atmospheric transport plays a significant role on the transport and potential environmental sinks for microplastics. The density and shape of microplastic particles will have important effects on their transport (Horton and Dixon, 2018). However, little is known about the processes governing transport of microplastics within air (Allen et al., 2019a; Dris et al., 2017). Specifically, it is not known to what extent atmospheric fallout contributes to aquatic and terrestrial contamination. More researches are needed in this area, spatially, with regards to source-pathway-sink processes, transport parameters and relative to meteorological conditions.

Latest research show that atmospheric transport of microplastics can reach remote areas without any local source of plastics (Allen et al., 2019a). Evidence of microplastics on an Alps and Tibetan glaciers has been observed (Ambrosini et al., 2019; Zhang et al., 2019). Microplastics transported by wind to high latitudes may be the cause of microplastics deposition on glaciers (Fig. 3). Microplastics in snowfall (and rainfall) may be another important way for microplastics occurrence in surface ocean and Arctic environments (Bergmann et al., 2019).

Measurement of atmospheric microplastic flux could help quantify the contributions of atmospheric microplastics to the marine or terrestrial ecosystems. However, microplastics flux from atmospheric deposition has not been widely studied at present. Glaciers in the cryosphere regions are ideal environments to accumulate pollutants from the atmosphere through dry deposition or snowfall. Due to its low temperature and remoteness from human activities, pollutant records in snow have been effectively used to calculate the flux from atmospheric deposition. Similar processes may also be effective in analysis of microplastics, one of the most ubiquitous pollutants released by anthropogenic activities. More importantly, accumulation of microplastic particles in ice cores will provide temporal variations, in a similar way to lacustrine archive microplastics (lake sediments) (Turner et al., 2019).

To date, only two atmospheric microplastic studies have attempted to examine the transport pathway or trajectory of these particles. The first attempt at analyzing atmospheric microplastic transport was presented by Allen et al. (2019b) where the particle transport was evidenced to be greater than 100 km. This study used simplistic meteorological and particle settling velocity calculation and well-known

atmospheric Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) to examine dynamic atmospheric transport. A further study by Liu et al. (2019b) has used HYSPLIT to consider the possible sources of atmospheric microplastic (air mass sampling) creating back trajectories relative to the sample period. Lagrangian atmospheric models such as HYSPLIT, LAGRANTO and FLEXPART are useful tools to consider where atmospheric particles and pollutants may have travelled from Allen et al. (2019b). They can be used to identify the potential source of an atmospheric pollutant and the atmospheric trajectory along which it may have travelled (distance, elevation, atmospheric mixing etc.). These models are well established and used of atmospheric modelling of pollutants, particles and gasses such as mercury, caesium and dust and have great potential for more in depth and detailed analysis of atmospheric microplastic transport. However, at present the parameters necessary to adequately describe and characterize atmospheric microplastics are unknown. The density, shape and size of atmospheric particles can be attained from field samples, but global, regional or land use specific generalizations are difficult. The entrainment potential, deposition and detention processes and potentials, atmospheric settling velocities with/without collation or cohesion of homo/heterogeneous atmospheric particles are all unknown. The efficiency of precipitation scavenging, influence of atmospheric microplastics in atmospheric ice nucleation and electrical charge of the particles on atmospheric transport is unknown and so un-parameterized (Allen et al., 2019b; Ganguly and Ariya, 2019; van der Does et al., 2018). Atmospheric particle transport modelling is an important future focus of atmospheric microplastic transport research, with significant further research needs and challenges in definition and description of atmospheric microplastic transport dynamics.

It is widely considered that the oceans represent a sink for a large proportion of microplastics, with terrestrial and freshwater environments acting as important sources and pathways for microplastics to the sea (Jambeck et al., 2015). Atmospheric microplastics link the processes influencing flux and retention of microplastics in environment (Horton and Dixon, 2018; Liu et al., 2019b). The atmospheric transport for microplastics to remote areas and its potential global impact on contributions to microplastics in marine and terrestrial ecosystems is a challenge facing the development of the plastic source-pathway-sink model (Bank and Hansson, 2019).

4.2. Risk estimation for human exposure

Microplastics present in the environment can be ingested by different types of organisms, including species widely used in the human diet (Li et al., 2018; Prata, 2018; Rochman et al., 2015). The recent findings of atmospheric microplastics highlight the broad spatio-temporal scales of the processes that influence the sources, fate, transport and effects of microplastics on the environment and its inhabitants, including humans (Bank and Hansson, 2019). Epidemiologic studies indicated ambient atmospheric particles air pollution is linked to adverse respiratory and cardiovascular effects (Churg and Brauer, 2000). Although the visually observed microplastic fibres are supposedly too large to be inhaled; there is an exposure may occur through dust ingestion, particularly for young children (Dris et al., 2017; Wright and Kelly, 2017). Previous studies identified cellulosic and plastic fibres in human lungs (excised lung cancers and lung biopsies) (Pauly et al., 1998; Wright and Kelly, 2017) and for workers in plastic processing factories to demonstrate breathing and health problems (coughing, dyspnea, wheezing, occupational asthma (Kremer et al., 1994). Microplastic particles (>100 μm) have also been demonstrated as bio-persistent and to pass the gastrointestinal tract epithelium (Wright and Kelly, 2017). Human exposure of microplastics especially via dust ingestion can potentially be estimated based on the atmospheric microplastic concentration.

Simplistic modelling has estimated that approximately 7665 particles of microplastics are inhaled annually by people in Shanghai (East China) from outdoor environments (K. Liu et al., 2019a). Meanwhile, indoor dust is a non-negligible source of human exposure to MPs, accounting for a geomean daily intake of 17,300 ng/kg-bw (average body weight) of PET microplastics in children of Chinese major cities (Liu et al., 2019). In Iran, it is estimated that a mean of 3223 and 1063 MP yr^{-1} is ingested by children and adults, respectively (Dehghani et al., 2017). For context, the flocking area of a polyester microfibre plant may have airborne particles of 7 mg m^{-3}, up 1000,000 fibres m^{-3} (Wright and Kelly, 2017). Most of the inhaled microplastic fibres are potentially subjected to mucociliary clearance; however, some may persist in the lung causing localized biological responses, including inflammation, especially in individuals with compromised clearance mechanisms (Gasperi et al., 2018; Wright and Kelly, 2017).

Microplastic is also a pollutant transport medium for other toxic elements such as DDT and hexachlorobenzene (Laskar and Kumar, 2019). The sorption of chemicals (e.g., PAHs, mercury) to microplastics may become a threat to biota, when ingestion occurs or through leaching and/or desorption of adsorbed and plastic composite chemicals. Associated contaminants such as PAHs desorb and lead to genotoxicity while the plastic itself and its additives (dyes, plasticizers, PFAs, phthalates) lead to health effects including reproductive toxicity, carcinogenicity and mutagenicity (Gasperi et al., 2018; Latini et al., 2003; Wirth et al., 2008; Wright and Kelly, 2017). Microplastic bioaccumulation in the environment is in the early stages of research, with very little known in the marine, freshwater or terrestrial environments and no examination with relation to the atmospheric environment yet (Drummond et al., 2019). It is known that phthalates and other plastic components can cause detrimental impacts on human health, as illustrated by past BPA studies (endocrine disruption) and DEHP research (modified gene expression, shortened gestation periods, lower birth weights) (Bhat et al., 2020; Latini et al., 2003; Nardelli et al., 2015; Peretz et al., 2014). There is also evidence of phthalates such as BPA in the atmosphere as aerosols in notable quantities (up to 17,4000 pg m^{-3}) (Fu and Kawamura, 2010). The effect of atmospheric microplastics, their chemical components and their adsorbed pollutants on human and ecosystem health is unknown, but the potential of micro and nano plastic to influence this is of concern (Lehner et al., 2019; Wright and Kelly, 2017). However, the interactions between microplastics with other organic pollutants and metals in the atmosphere, their impacts on and interaction with the environment, humans and ecosystem health are virtually unstudied and need to be better understood.

5. Conclusions

Microplastics are now acknowledged as atmospheric pollutants and particulates. Recent studies have demonstrated the existence of microplastics in the area of urban, rural and remote atmosphere and atmospheric deposition. As an atmospheric pollutant, there is significant potential for long-range transport and therefore influence on locations far from microplastic pollution sources.

Among the published studies, relative abundance of atmospheric microplastics reflects a wide range of characteristics and quantities across different regions. Fibres and fragments are the most frequently identified microplastic shapes in atmosphere. Conclusion on size distribution in these studies are difficult to draw due to the differences in targeted particle size. Because of its light-weight, durability, and other intrinsic features, atmospheric microplastics can be transported to remote areas and deposited through dry or wet deposition. Wind, snow-fall, and weathering play an important role on atmospheric microplastics from sources to ocean or land surfaces.

Current atmospheric microplastic research is in the early stages, and therefore suffers from insufficient comparable data on abundance and characterization. This is especially the case in remote areas and concerning microplastic composition due to the non-standardized operation protocols for microplastic sampling and detection used to date.

Standardized methods for sampling and measurement of atmospheric microplastics will allow reproducibility and comparability of results and will lead to the quality data to necessary conduct risk assessments. Worldwide research on spatial and temporal variations of atmospheric microplastics depositions needs to be further enhanced. Studies are also needed to better understand the interaction between atmospheric microplastics and other chemicals, ecosystems and human exposure.

Declaration of Competing Interest

None

Acknowledgments

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.earscirev.2020.103118.

References

Abbasi, S., Keshavarzi, B., Moore, F., Delshab, H., Soltani, N., Sorooshian, A., 2017. Investigation of microrubbers, microplastics and heavy metals in street dust: a study in Bushehr city. Iran. Environ. Earth Sci. 76. https://doi.org/10.1007/s12665-017-7137-0.

Abbasi, S., Keshavarzi, B., Moore, F., Turner, A., Kelly, F.J., Dominguez, A.O., Jaafarzadeh, N., 2019. Distribution and potential health impacts of microplastics and microrubbers in air and street dusts from Asaluyeh County. Iran. Environ. Pollut. 244, 153–164. https://doi.org/10.1016/j.envpol.2018.10.039.

Alimba, C.G., Faggio, C., 2019. Microplastics in the marine environment: current trends in environmental pollution and mechanisms of toxicological profile. Environ. Toxicol. Pharmacol. 68, 61–74. https://doi.org/10.1016/j.etap.2019.03.001.

Alimi, O.S., Farner Budarz, J., Hernandez, L.M., Tufenkji, N., 2018. Microplastics and nanoplastics in aquatic environments: aggregation, deposition, and enhanced contaminant transport. Environ. Sci. Technol. 52, 1704–1724. https://doi.org/10.1021/acs.est.7b05559.

Allen, S., Allen, D., Phoenix, V.R., Le Roux, G., Duranteza, P., Simonneau, A., Stéphane, B., Galop, D., 2019a. Atmospheric transport and deposition of microplastics in a remote mountain catchment. Nat. Geosci. 12, 339–344. https://doi.org/10.1038/s41561-019-0335-5.

Allen, D., Allen, S., Sonke, J., Phoenix, V., 2019b. Atmospheric transport and our planetary boundary layer as an environmental compartment of microplastics pollution. In: Cocca, M., Di Pace, E., Errico, M., Gentile, G., Montarsolo, A., Mossotti, R. (Eds.), International Conference on Microplastic Pollution Inthe Mediterranean Sea. Springer Water. Springer, Cham, Capri, pp. 1–8.

Ambrosini, R., Azzoni, R.S., Pittino, F., Diolaiuti, G., Franzetti, A., Parolini, M., 2019. First evidence of microplastic contamination in the supraglacial debris of an alpine glacier. Environ. Pollut. 253, 297–301. https://doi.org/10.1016/j.envpol.2019.07.005.

Araujo, C., Nolasco, M.M., Ribeiro, A.M.P., Ribeiro-Claro, P.J.A., 2018. Identification of microplastics using Raman spectroscopy: latest developments and future prospects. Water Res. 142, 426–440. https://doi.org/10.1016/j.watres.2018.05.060.

Auta, H.S., Emenike, C.U., Fauziah, S.H., 2017. Distribution and importance of microplastics in the marine environmentA review of the sources, fate, effects, and potential solutions. Environ. Int. 102, 165–176. https://doi.org/10.1016/j.envint.2017.02.013.

Bank, M.S., Hansson, S.V., 2019. The plastic cycle: a novel and holistic paradigm for the anthropocene. Environ. Sci. Technol. https://doi.org/10.1021/acs.est.9b02942. acs.est.9b02942.

Barboza, L.G.A., Vieira, L.R., Branco, V., Figueiredo, N., Carvalho, F., Carvalho, C., Guilhermino, L., 2018. Microplastics cause neurotoxicity, oxidative damage and energy-related changes and interact with the bioaccumulation of mercury in the European seabass, Dicentrarchus labrax (Linnaeus, 1758). Aquat. Toxicol. 195, 49–57. https://doi.org/10.1016/j.aquatox.2017.12.008.

Bargmann, M., Gutow, L., Klages, M., 2015. Marine anthropogenic litter. Springer, Berlin, pp. 456.

Bergmann, M., Mützel, S., Primpke, S., Tekman, M.B., Trachsel, J., Gerdts, G., 2019. White and wonderful? Microplastics prevail in snow from the Alps to the Arctic. Sci.

Adv. 5, eaax1157. https://doi.org/10.1126/sciadv.aax1157.

Besseling, E., Quik, J.T.K., Sun, M., Koelmans, A.A., 2017. Fate of nano- and microplastic in freshwater systems: a modeling study. Environ. Pollut. 220, 540–548. https://doi.org/10.1016/j.envpol.2016.10.001.

Bhat, R.A., Kumar, D., Bhat, S.M., Sofi, I.R., 2020. Historical perspective of bisphenol A and phthalates in the environment and their health effects. In: Handbook of Research on Environmental and Human Health Impacts of Plastic Pollution. IGI Global, pp. 246–262. https://doi.org/10.4018/978-1-5225-9452-9.ch013.

Browne, M.A., Crump, P., Niven, S.J., Teuten, E., Tonkin, A., Galloway, T., Thompson, R.C., 2011. Accumulation of Microplastic on Shorelines Worldwide : Sources and Sinks. pp. 9175–9179.

Cai, L., Wang, J., Peng, J., Tan, Z., Zhan, Z., Tan, X., Chen, Q., 2017. Characteristic of microplastics in the atmospheric fallout from Dongguan city, China: preliminary research and first evidence. Environ. Sci. Pollut. Res. 24, 24928–24935. https://doi.org/10.1007/s11356-017-0116-x.

Camarero, L., Bacardit, M., de Diego, A., Arana, G., 2017. Decadal trends in atmospheric deposition in a high elevation station: Effects of climate and pollution on the long-range flux of metals and trace elements over SW Europe. Atmos. Environ. 167, 542–552. https://doi.org/10.1016/j.atmosenv.2017.08.049.

Centrone, A., 2015. Infrared imaging and spectroscopy beyond the diffraction limit. Annu. Rev. Anal. Chem. 8, 101–126. https://doi.org/10.1146/annurev-anchem-071114-040435.

Churg, A., Brauer, M., 2000. Ambient atmospheric particles in the airways of human lungs. Ultrastruct. Pathol. 24, 353–361. https://doi.org/10.1080/019131200750060014.

Cole, M., 2016. A novel method for preparing microplastic fibers. Sci. Rep. 6, 1–7. https://doi.org/10.1038/srep34519.

David, J., Steinmetz, Z., Kučeřík, J., Schaumann, G.E., 2018. Quantitative analysis of poly (ethylene terephthalate) microplastics in soil via thermogravimetry-mass spectrometry. Anal. Chem. 90, 8793–8799. https://doi.org/10.1021/acs.analchem.8b00355.

Dehghani, S., Moore, F., Akhbarizadeh, R., 2017. Microplastic pollution in deposited urban dust, Tehran metropolis. Iran. Environ. Sci. Pollut. Res. 24, 20360–20371. https://doi.org/10.1007/s11356-017-9674-1.

Dommergue, A., Amato, P., Tignat-perrier, R., Magand, O., Thollot, A., Joly, M., Bouvier, L., Sellegri, K., Vogel, T., Sonke, J., Jaffrezo, J., Andrade, M., Moreno, I., Labuschagne, C., Martin, L., Zhang, Q., Larose, C., Pearce, D.A., 2019. Methods to investigate the global atmospheric microbiome. Front. Microbiol. 10, 1–12. https://doi.org/10.3389/fmicb.2019.00243.

Dris, R., 2016. First Assessment of Sources and Fate of Macro- and Micro- Plastics in Urban Hydrosystems: Case of Paris Megacity. Université Paris-Est.

Dris, R., Gasperi, C.J., Rocher, A.V., Saad, B.M., Renault, N., Tassin, B., 2015. Microplastic contamination in an urban area: a case study in Greater Paris. Environ. Chem. 12, 592–599.

Dris, R., Gasperi, J., Saad, M., Mirande, C., Tassin, B., 2016. Synthetic fibers in atmospheric fallout: a source of microplastics in the environment? Mar. Pollut. Bull. 104, 290–293. https://doi.org/10.1016/j.marpolbul.2016.01.006.

Dris, R., Gasperi, J., Mirande, C., Mandin, C., Guerrouache, M., Langlois, V., Tassin, B., 2017. A first overview of textile fibers, including microplastics, in indoor and outdoor environments. Environ. Pollut. 221, 453–458. https://doi.org/10.1016/j.envpol.2016.12.013.

Drummond, J., Krause, S., Nel, L., Allen, S., Allen, D., Simon, L., Doaudy, C., 2019. Gathering at the Top? Environmental Controls of Microplastic Uptake and Biomagnification in Aquatic Food Webs.

Dümichen, E., Barthel, A.K., Braun, U., Bannick, C.G., Brand, K., Jekel, M., Senz, R., 2015. Analysis of polyethylene microplastics in environmental samples, using a thermal decomposition method. Water Res. 85, 451–457. https://doi.org/10.1016/j.watres.2015.09.002.

Dümichen, E., Eisentraut, P., Bannick, C.G., Barthel, A.K., Senz, R., Braun, U., 2017. Fast identification of microplastics in complex environmental samples by a thermal degradation method. Chemosphere 174, 572–584. https://doi.org/10.1016/j.chemosphere.2017.02.010.

Everall, N., Griffiths, P., Chamlers, J., 2007. Vibrational Spectroscopy of Polymers: Principles and Practice. Wiley.

Fischer, M., Scholz-Böttcher, B.M., 2017. Simultaneous trace identification and quantification of common types of microplastics in environmental samples by pyrolysis-gas chromatography-mass spectrometry. Environ. Sci. Technol. 51, 5052–5060. https://doi.org/10.1021/acs.est.6b06362.

Fries, E., Dekiff, J.H., Willmeyer, J., Nuelle, M.-T., Ebert, M., Remy, D., 2013. Identification of polymer types and additives in marine microplastic particles using pyrolysis-GC/MS and scanning electron microscopy. Environ Sci Process Impacts 15, 1949. https://doi.org/10.1039/c3em00214d.

Fu, P., Kawamura, K., 2010. Ubiquity of bisphenol A in the atmosphere. Environ. Pollut. 158, 3138–3143. https://doi.org/10.1016/j.envpol.2010.06.040.

Fu, Z., Wang, J., 2019. Current practices and future perspectives of microplastic pollution in freshwater ecosystems in China. Sci. Total Environ. 691, 697–712. https://doi.org/10.1016/j.scitotenv.2019.07.167.

Ganguly, M., Ariya, P.A., 2019. Ice nucleation of model nano-micro plastics: a novel synthetic protocol and the influence of particle capping at diverse atmospheric environments. ACS Earth Sp. Chem. https://doi.org/10.1021/acsearthspacechem.9b00132.

Gasperi, J., Wright, S.L., Dris, R., Collard, F., Mandin, C., Guerrouache, M., Langlois, V., Kelly, F.J., Tassin, B., 2018. Microplastics in air: are we breathing it in? Curr. Opin. Environ. Sci. Heal. 1, 1–5. https://doi.org/10.1016/j.coesh.2017.10.002.

GESAMP, 2016. Sources, fate and effects of microplastics in the marine environment: part 2 of a global assessment. In: IMO/FAO/UNESCO-IOC/UNIDO/WMO/IAEA/UN/

UNEP/UNDP Joint Group of Experts on the Scientific Aspects of Marine Environmental Protection.

Gillibert, R., Balakrishnan, G., Deshoules, Q., Tardivel, M., Magazzù, A., Donato, M.G., Maragò, O.M., Lamy de La Chapelle, M., Colas, F., Lagarde, F., Gucciardi, P.G., 2019. Raman tweezers for small microplastics and nanoplastics identification in seawater. Environ. Sci. Technol. 53, 9003–9013. https://doi.org/10.1021/acs.est.9b03105.

Hanvey, J.S., Lewis, P.J., Lavers, J.L., Crosbie, N.D., Pozo, K., Clarke, B.O., 2017. A review of analytical techniques for quantifying microplastics in sediments. Anal. Methods 9, 1369–1383. https://doi.org/10.1039/C6AY02707E.

Hartmann, N.B., Hüffer, T., Thompson, R.C., Hassellöv, M., Verschoor, A., Daugaard, A.E., Rist, S., Karlsson, T., Brennholt, N., Cole, M., Herrling, M.P., Hess, M.C., Ivleva, N.P., Lusher, A.L., Wagner, M., 2019. Are we speaking the same language? recommendations for a definition and categorization framework for plastic debris. Environ. Sci. Technol. 53, 1039–1047. https://doi.org/10.1021/acs.est.8b05297.

Hayward, S.J., Gouin, T., Wania, F., 2010. Comparison of four active and passive sampling techniques for pesticides in air. Environ. Sci. Technol. 44, 3410–3416. https://doi.org/10.1021/es902512h.

Helm, P.A., 2017. Improving microplastics source apportionment: a role for microplastic morphology and taxonomy? Anal Methods 9, 1328–1331. https://doi.org/10.1039/c7ay90016c.

Hendrickson, E., Minor, E.C., Schreiner, K., 2018. Microplastic abundance and composition in western lake superior as determined via microscopy, Pyr-GC/MS, and FTIR. Environ. Sci. Technol. 52, 1787–1796. https://doi.org/10.1021/acs.est.7b05829.

Hermabessiere, L., Dehaut, A., Paul-Pont, I., Lacroix, C., Jezequel, R., Soudant, P., Duflos, G., 2017. Occurrence and effects of plastic additives on marine environments and organisms: a review. Chemosphere. https://doi.org/10.1016/j.chemosphere.2017.05.096.

Hidalgo-Ruz, V., Gutow, L., Thompson, R.C., Thiel, M., 2012. Microplastics in the marine environment: a review of the methods used for identification and quantification. Environ. Sci. Technol. 46, 3060–3075. https://doi.org/10.1021/es2031505.

Horton, A.A., Dixon, S.J., 2018. Microplastics: an introduction to environmental transport processes. Wiley Interdiscip. Rev. Water 5, e1268. https://doi.org/10.1002/wat2.1268.

Hüffer, T., Praetorius, A., Wagner, S., Von Der Kammer, F., Hofmann, T., 2017. Microplastic exposure assessment in aquatic environments: learning from similarities and differences to engineered nanoparticles. Environ. Sci. Technol. 51, 2499–2507. https://doi.org/10.1021/acs.est.6b04054.

Hurley, R., Lusher, A.L., Olsen, M., Nizzetto, L., 2018. Validation of a method for extracting microplastics from complex, organic-rich, environmental matrices. Environ. Sci. Technol. 52, 7409–7417. https://doi.org/10.1021/acs.est.8b01517.

Huth, F., Govyadinov, A., Amarie, S., Nuansing, W., Keilmann, F., Hillenbrand, R., 2012. Nano-FTIR absorption spectroscopy of molecular fingerprints at 20 nm spatial resolution. Nano Lett. 12, 3973–3978. https://doi.org/10.1021/nl301159v.

Isobe, A., Uchida, K., Tokai, T., Iwasaki, S., 2015. East Asian seas: a hot spot of pelagic microplastics. Mar. Pollut. Bull. 101, 618–623. https://doi.org/10.1016/j.marpolbul.2015.10.042.

Jambeck, J.R., Geyer, R., Wilcox, C., Siegler, T.R., Perryman, M., Andrady, A., Narayan, R., Law, K.L., 2015. Plastic waste inputs from land into the Ocean. Science (80-.) 347, 768–771. https://doi.org/10.1126/science.1260879.

Jemec, A., Horvat, P., Kunej, U., Bele, M., Kržan, A., 2016. Uptake and effects of microplastic textile fibers on freshwater crustacean Daphnia magna. Environ. Pollut. 219, 201–209. https://doi.org/10.1016/j.envpol.2016.10.037.

Käppler, A., Fischer, D., Oberbeckmann, S., Schernewski, G., Labrenz, M., Eichhorn, K.-J., Voit, B., 2016. Analysis of environmental microplastics by vibrational microspectroscopy: FTIR, Raman or both? Anal. Bioanal. Chem. 408, 8377–8391. https://doi.org/10.1007/s00216-016-9956-3.

Käppler, A., Fischer, M., Scholz-Böttcher, B.M., Oberbeckmann, S., Labrenz, M., Fischer, D., Eichhorn, K.J., Voit, B., 2018. Comparison of μ-ATR-FTIR spectroscopy and py-GCMS as identification tools for microplastic particles and fibers isolated from river sediments. Anal. Bioanal. Chem. 410, 5313–5327. https://doi.org/10.1007/s00216-018-1185-5.

Klein, M., Fischer, E.K., 2019. Microplastic abundance in atmospheric deposition within the Metropolitan area of Hamburg, Germany. Sci. Total Environ. 685, 96–103. https://doi.org/10.1016/j.scitotenv.2019.05.405.

Klein, A., Ravetta, F., Thomas, J.L., Ancellet, G., Augustin, P., Wilson, R., Dieudonné, E., Fourmentin, M., Delbarre, H., Pelon, J., 2019. Influence of vertical mixing and nighttime transport on surface ozone variability in the morning in Paris and the surrounding region. Atmos. Environ. 197, 92–102. https://doi.org/10.1016/j.atmosenv.2018.10.009.

Kremer, A.M., Pal, T.M., Boleij, J.S.M., Schouten, J.P., Rijcken, B., 1994. Airway hyperresponsiveness, prevalence of chronic respiratory symptoms, and lung function in workers exposed to irritants. Occup. Environ. Med. 51, 3–13. https://doi.org/10.1136/oem.51.1.3.

Kuczenski, B., Geyer, R., 2010. Material flow analysis of polyethylene terephthalate in the US, 1996-2007. Resour. Conserv. Recycl. 54, 1161–1169. https://doi.org/10.1016/j.resconrec.2010.03.013.

Laskar, N., Kumar, U., 2019. Plastics and microplastics: a threat to environment. Environ. Technol. Innov. 14, 100352. https://doi.org/10.1016/j.eti.2019.100352.

Latini, G., De Felice, C., Presta, G., Del Vecchio, A., Paris, I., Ruggieri, F., Mzzeo, P., 2003. In utero exposure to di-(2-ethylhexyl)phthalate and duration of human pregnancy. Environ. Health Perspect. 111, 1783–1785. https://doi.org/10.1289/ehp.6202.

Lehner, R., Weder, C., Petri-Fink, A., Rothen-Rutishauser, B., 2019. Emergence of nanoplastic in the environment and possible impact on human health. Environ. Sci. Technol. https://doi.org/10.1021/acs.est.8b05512.

Li, J., Liu, H., Paul Chen, J., 2018. Microplastics in freshwater systems: a review on occurrence, environmental effects, and methods for microplastics detection. Water Res. 137, 362–374. https://doi.org/10.1016/j.watres.2017.12.056.

Liu, C., Li, J., Zhang, Y., Wang, L., Deng, J., Gao, Y., Yu, L., Zhang, J., Sun, H., 2019. Widespread distribution of PET and PC microplastics in dust in urban China and their estimated human exposure. Environ. Int. 128, 116–124. https://doi.org/10.1016/j.envint.2019.04.024.

Liu, K., Wang, X., Fang, T., Xu, P., Zhu, L., Li, D., 2019a. Source and potential risk assessment of suspended atmospheric microplastics in Shanghai. Sci. Total Environ. 675, 462–471. https://doi.org/10.1016/j.scitotenv.2019.04.110.

Liu, K., Wu, T., Wang, X., Song, Z., Zong, C., Wei, N., Li, D., 2019b. Consistent transport of terrestrial microplastics to the ocean through atmosphere. Environ. Sci. Technol. 1–12. https://doi.org/10.1021/acs.est.9b03427.

Löder, M.G.J., Gerdts, G., 2015. Methodology used for the detection and identification of microplastics—a critical appraisal. In: Bergmann, M., Gutow, L., Klages, M. (Eds.), Marine Anthropogenic Litter, pp. 201–227. https://doi.org/10.1007/978-3-319-16510-3.

Löder, M.G.J., Imhof, H.K., Ladehoff, M., Löschel, L.A., Lorenz, C., Mintenig, S., Piehl, S., Primpke, S., Schrank, I., Laforsch, C., Gerdts, G., 2017. Enzymatic purification of microplastics in environmental samples. Environ. Sci. Technol. 51, 14283–14292. https://doi.org/10.1021/acs.est.7b03055.

Marine & Environmental Research Institute, 2015. Guide to Microplastic Identification. Marine & Environmental Research Instatite.

Masura, Julie, Baker, Joel, Foster, Gregory, Arthur, C., 2015. Laboratory Methods for the Analysis of Microplastics in the Marine Environment.

Materić, D., Peacock, M., Kent, M., Cook, S., Gauci, V., Röckmann, T., Holzinger, R., 2017. Characterisation of the semi-volatile component of dissolved organic matter by thermal desorption - proton transfer reaction - mass spectrometry. Sci. Rep. 7, 1–8. https://doi.org/10.1038/s41598-017-16256-x.

Materić, D., Ludewig, E., Xu, K., Röckmann, T., Holzinger, R., 2019. Brief communication: analysis of organic matter in surface snow by PTR-MS - implications for dry deposition dynamics in the Alps. Cryosphere 13, 297–307. https://doi.org/10.5194/tc-13-297-2019.

Meyns, M., Primpke, S., Gerdts, G., 2019. Library based identification and characterisation of polymers with nano-FTIR and IR-sSNOM imaging. Analyt. Methods 11, 5195–5202. https://doi.org/10.1039/C9AY01193E. arXiv:1906.10243.

Napper, I.E., Thompson, R.C., 2016. Release of synthetic microplastic plastic fibres from domestic washing machines: Effects of fabric type and washing conditions. Mar. Pollut. Bull. 112, 39–45. https://doi.org/10.1016/j.marpolbul.2016.09.025.

Nardelli, T.C., Erythropel, H.C., Robaire, B., 2015. Toxicogenomic screening of replacements for Di(2-Ethylhexyl) phthalate (DEHP) using the immortalized TM4 sertoli cell line. PLoS One 10, 1–17. https://doi.org/10.1371/journal.pone.0138421.

Nguyen, B., Claveau-Mallet, D., Hernandez, L.M., Xu, E.G., Farner, J.M., Tufenkji, N., 2019. Separation and analysis of microplastics and nanoplastics in complex environmental samples. Acc. Chem. Res. 52, 858–866. https://doi.org/10.1021/acs.accounts.8b00602.

Pauly, J.L., Stegmeier, S.J., Allaart, H.A., Cheney, R.T., Zhang, P.J., Mayer, A.G., Streck, R.J., 1998. Inhaled cellulosic and plastic fibers found in human lung tissue. Cancer Epidemiol. Biomark. Prev. 7, 419–428.

Peretz, J., Vrooman, L., Ricke, W.A., Hunt, P.A., Ehrlich, S., Hauser, R., Padmanabhan, V., Taylor, H.S., Swan, S.H., Vandevoort, C.A., Flaws, J.A., 2014. Bisphenol A and reproductive health: Update of experimental and human evidence, 2007-2013. Environ. Health Perspect. 122, 775–786. https://doi.org/10.1289/ehp.1307728.

PlasticsEurope, 2018. Plastics – The Facts 2018: An Analysis of European Plastics Production, Demand and Waste Data.

Prata, J.C., 2018. Airborne microplastics: Consequences to human health? Environ. Pollut. 234, 115–126. https://doi.org/10.1016/j.envpol.2017.11.043.

Prata, J.C., da Costa, J.P., Girão, A.V., Lopes, I., Duarte, A.C., Rocha-Santos, T., 2019. Identifying a quick and efficient method of removing organic matter without damaging microplastic samples. Sci. Total Environ. 686, 131–139. https://doi.org/10.1016/j.scitotenv.2019.05.456.

Primpke, S., Lorenz, C., Rascher-Friesenhausen, R., Gerdts, G., 2017. An automated approach for microplastics analysis using focal plane array (FPA) FTIR microscopy and image analysis. Anal. Methods 9, 1499–1511. https://doi.org/10.1039/C6AY02476A.

Quinn, B., Murphy, F., Ewins, C., 2017. Validation of density separation for the rapid recovery of microplastics from sediment. Anal. Methods 9, 1491–1498. https://doi.org/10.1039/C6AY02542K.

Renner, G., Schmidt, T.C., Schram, J., 2018. Analytical methodologies for monitoring micro(nano)plastics: which are fit for purpose? Curr. Opin. Environ. Sci. Heal. 1, 55–61. https://doi.org/10.1016/j.coesh.2017.11.001.

Ribeiro-Claro, P., Nolasco, M.M., Araújo, C., 2017. Characterization of microplastics by Raman spectroscopy. Compr. Anal. Chem. 75, 119–151. https://doi.org/10.1016/bs.coac.2016.10.001.

Rocha-Santos, Teresa Duarte, 2017. Characterization and Analysis of Microplastics. Elsevier.

Rochman, C.M., 2015. The complex mixture, fate and toxicity of chemicals associated with plastic debris in the marine environment. In: Bergmann, M., Gutow, L., Klages, M. (Eds.), Marine Anthropogenic Litter. Springer, Cham, pp. 117–140. https://doi.org/10.1007/978-3-319-16510-3.

Rochman, C.M., Tahir, A., Williams, S.L., Baxa, D.V., Lam, R., Miller, J.T., Teh, F.C., Werorilangi, S., Teh, S.J., 2015. Anthropogenic debris in seafood: plastic debris and fibers from textiles in fish and bivalves sold for human consumption. Sci. Rep. 5, 1–10. https://doi.org/10.1038/srep14340.

Rochman, C.M., Brookson, C., Bikker, J., Djuric, N., Earn, A., Bucci, K., Athey, S., Huntington, A., McIlwraith, H., Munno, K., De Frond, H., Kolomijeca, A., Erdle, L., Grbic, J., Bayoumi, M., Borrelle, S.B., Wu, T., Santoro, S., Werbowski, L.M., Zhu, X., Giles, R.K., Hamilton, B.M., Thaysen, C., Kaura, A., Klasios, N., Ead, L., Kim, J.,

Sherlock, C., Ho, A., Hung, C., 2019. Rethinking microplastics as a diverse contaminant suite. Environ. Toxicol. Chem. 38, 703–711. https://doi.org/10.1002/etc.4371.

Silva, A.B., Bastos, A.S., Justino, C.I.L., da Costa, J.P., Duarte, A.C., Rocha-Santos, T.A.P., 2018. Microplastics in the environment: challenges in analytical chemistry - a review. Anal. Chim. Acta 1017, 1–19. https://doi.org/10.1016/j.aca.2018.02.043.

Stanton, T., Johnson, M., Nathanail, P., MacNaughtan, W., Gomes, R.L., 2019. Freshwater and airborne textile fibre populations are dominated by 'natural', not microplastic, fibres. Sci. Total Environ. 666, 377–389. https://doi.org/10.1016/j.scitotenv.2019.02.278.

Tagg, A.S., Harrison, J.P., Ju-Nam, Y., Sapp, M., Bradley, E.L., Sinclair, C.J., Ojeda, J.J., 2017. Fenton's reagent for the rapid and efficient isolation of microplastics from wastewater. Chem. Commun. 53, 372–375. https://doi.org/10.1039/c6cc08798a.

Thompson, R.C., Olsen, Y., Mitchell, R.P., Davis, A., Rowland, S.J., John, A.W.G., McGonigle, D., Russell, A.E., 2004. Lost at sea: where does all the plastic go? Science (80-.) 304, 838.

Tourinho, P.S., Kočí, V., Loureiro, S., van Gestel, C.A.M., 2019. Partitioning of chemical contaminants to microplastics: Sorption mechanisms, environmental distribution and effects on toxicity and bioaccumulation. Environ. Pollut. 252, 1246–1256. https://doi.org/10.1016/j.envpol.2019.06.030.

Turner, S., Horton, A.A., Rose, N.L., Hall, C., 2019. A temporal sediment record of microplastics in an urban. J. Paleolimnol. 61, 449–462. https://doi.org/10.1007/s10933-019-00071-7.

van der Does, M., Knippertz, P., Zschenderlein, P., Giles Harrison, R., Stuut, J.-B.W., 2018. The mysterious long-range transport of giant mineral dust particles. Sci. Adv. 4. https://doi.org/10.1126/sciadv.aau2768.

Vianello, A., Jensen, R.L., Liu, L., Vollertsen, J., 2019. Simulating human exposure to indoor airborne microplastics using a Breathing Thermal Manikin. Sci. Rep. 9, 1–11. https://doi.org/10.1038/s41598-019-45054-w.

Windsor, F.M., Durance, I., Horton, A.A., Thompson, R.C., Tyler, C.R., Ormerod, S.J., 2019. A catchment-scale perspective of plastic pollution. Glob. Chang. Biol. 25, 1207–1221. https://doi.org/10.1111/gcb.14572.

Wirth, J.J., Rossano, M., Potter, R., Puscheck, E., Daly, D., Paneth, N., Krawetz, S., Protas, B., Diamond, M., 2008. A pilot study associating urinary concentrations of phthalate metabolites and semen quality. Syst Biol Reprod Med 54, 143–154. https://doi.org/10.1080/19396360802055921.

Wright, S.L., Kelly, F.J., 2017. Plastic and human health: a micro issue? Environ. Sci. Technol. 51, 6634–6647. https://doi.org/10.1021/acs.est.7b00423.

Zeng, E.Y. (Ed.), 2018. Microplastic Contamination in Aquatic Environments - An Emerging Matter of Environmental Urgency. Elsevier. https://doi.org/10.1016/C2016-0-04784-8.

Zhang, Y., Gao, T., Kang, S., Sillanpaa, M., 2019. Importance of atmospheric transport for microplastics deposited in remote areas. Environ. Pollut. 24, 112953. https://doi.org/10.1016/j.envpol.2019.07.121. (in review).

Zhou, Q., Tian, C., Luo, Y., 2017. Various forms and deposition fluxes of microplastics identified in the coastal urban atmosphere. Chin. Sci. Bull. 62, 3902–3909. https://doi.org/10.1360/n972017-00956.