Faculty of Science and Engineering

School of Geography, Earth and Environmental Sciences

2023-12

Suspended and deposited microplastics in the coastal atmosphere of southwest England

Kyriakoudes, G

https://pearl.plymouth.ac.uk/handle/10026.1/21694

10.1016/j.chemosphere.2023.140258 Chemosphere Elsevier BV

All content in PEARL is protected by copyright law. Author manuscripts are made available in accordance with publisher policies. Please cite only the published version using the details provided on the item record or document. In the absence of an open licence (e.g. Creative Commons), permissions for further reuse of content should be sought from the publisher or author.

ELSEVIER

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere





Suspended and deposited microplastics in the coastal atmosphere of southwest England

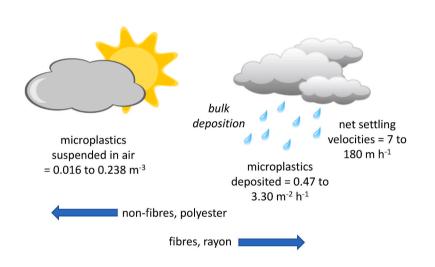
Giannis Kyriakoudes, Andrew Turner

School of Geography, Earth and Environmental Sciences, University of Plymouth University Plymouth, PL4 8AA, UK

HIGHLIGHTS

- Microplastics (MPs) captured during 12 periods over 42 d from the atmosphere of SW England.
- \bullet MP concentrations in suspension ranged from 0.016 to 0.238 items m $^{-3}$.
- \bullet Depositional MP fluxes ranged from 0.47 to 3.30 m $^{-2}$ h $^{-1}$ and inversely related to rainfall.
- Synthetic polymers present but rayon fibres were dominant particle type.
- MP shape and polymer type exhibit fractionation between suspension and deposition.
- Calculated, local settling velocities of MPs ranged from about 7 to 180 m h⁻¹.

GRAPHICAL ABSTRACT



ARTICLE INFO

Handling Editor: Michael Bank

Keywords: Microfibres Paints Airborne Settling Scavenging Rayon

ABSTRACT

Atmospheric microplastics (MPs) have been sampled from coastal southwest England during twelve periods over a 42-day timeframe in late autumn. MPs were dominated by fibres, with foams, fragments and pellets also observed. The majority of fibres were identified as the semisynthetic polymer, rayon, while other shapes were dominated by various petroleum-based thermoplastics (including polyvinyl acetate, polyvinyl alcohol, polyamide and polyester) and paints. MP concentrations suspended in air ranged from 0.016 to 0.238 items per m³ but displayed no clear dependence on wind speed or direction. Total depositional fluxes ranged from 0.47 to $3.30~\text{m}^{-2}~\text{h}^{-1}$ and showed no clear dependence on wind conditions or electrical conductivity of precipitation (as a measure of maritime influence). However, the concentration of deposited MPs in rainwater was inversely related to rainfall volume, suggesting that incipient precipitation acts to efficiently washout microplastics. A comparison of deposited and suspended MPs by size, shape and polymer type suggests that larger fibres constructed of rayon, polyamide and acrylic are preferentially removed from the atmosphere relative to smaller, non-fibrous MPs and particles constructed of polyester. A quantitative comparison of deposited and suspended MPs provided estimates of location- and environment-specific net settling velocities of between about 7 and 180 m h⁻¹ and corresponding residence times for an air column of 5000 m of between about 30 and 700 h. The

E-mail address: aturner@plymouth.ac.uk (A. Turner).

^{*} Corresponding author.

1. Introduction

With regard to the concentration and distribution of microplastics (MPs; primary and secondary plastics in the size range 1 μ m-5 mm), the atmosphere has received considerably less attention than aquatic systems, and in particular the oceans (Zhang et al., 2020; Allen et al., 2021; González-Pleiter et al., 2021; Long et al., 2022). Aside from the potential health risks arising from airborne MPs via inhalation (Gasperi et al., 2018; Mehmood et al., 2021), evidence suggests that, given the right meteorological conditions and air mass movements, plastics emitted to the atmosphere, and in particular small MP fibres, are subject to long-range transportation. Moreover, the global atmospheric transport of MPs is more efficient than the oceanic or terrestrial pathways because of the much shorter timescales involved (Evangeliou et al., 2022). Consequently, particles have been detected in regions distant from any primary or secondary sources and major waterways that include subtropical deserts (Abbasi et al., 2021) and remote national parks and mountains (Allen et al., 2019; Feng et al., 2020).

MPs in the atmosphere are, ultimately, subject to removal at the Earth's surface by dry, gravitational deposition, or wet deposition through in-cloud scavenging and below-cloud interception (Aeschlimann et al., 2022). Although the significance of and mechanisms involved in wet deposition of MPs are not well understood, precipitation is considered as a key, positive driver for their removal in the lower troposphere that is likely related to the frequency, intensity and duration of rainfall or snowfall (Bergmann et al., 2019; Zhang et al., 2020; Abbasi, 2021; Purwiyanto et al., 2022). To this end, climatic factors are also important. For example, in semi-arid regions, wet deposition may be constrained to rainfall events in certain months of the year (Abbasi and Turner, 2021) while in temperate, maritime climates, wet deposition during persistent periods of rain may outweigh annual dry deposition (Roblin et al., 2020).

In order to improve our understanding of the transport of microplastics by air masses and their deposition, Zhang et al. (2020) suggest that multiple sampling methods are required over an extended period of time at a given location. Specifically, and in order to gauge the importance of removal relative to MP availability, it would be useful to consider both the concentrations of MPs in the atmosphere and the depositional fluxes from the atmosphere. This approach was recently adopted by Yuan et al. (2023) in a study of MPs in the atmosphere of Guangzhou, China, where a washout ratio was proposed to measure the removal efficiency by precipitation (but not dry deposition).

The present study sets out to address and compare the quantities and, through infrared spectroscopy, chemical characteristics, of MPs suspended in air and captured actively, but not necessarily deposited locally, and MPs subject to local deposition and captured passively. We hypothesise that the concentrations and signatures of MPs resident in the atmosphere might be different to those that are deposited through gravitational settlement and captured by rainfall capture. The study is conducted over an urban area of coastal southwest England for a period of about six weeks that encapsulates different meteorological conditions and rainfall events and intensities. We also hypothesise, therefore, that the concentrations and deposition of MPs are related to environmental conditions.

2. Methods

2.1. Sampling

Plymouth is a maritime city (population $\sim 260,000$) on the English Channel coast of southwest England. It has a temperate oceanic climate

and experiences a mean annual precipitation of about 1000 mm. The prevailing wind is from the west to southwest and the annual average wind speed at 10 m above ground level is about 5 m s $^{-1}$ (11.2 miles per hour; mph).

Sampling was undertaken from the roof of a teaching building (coordinates: 50.3751; -4.1385) at a height of 30 m above ground level on the University of Plymouth main (city centre) campus over a 42-day timeframe in late autumn (November and December of 2021). Specifically, material suspended in air and material deposited from the atmosphere were sampled during twelve, 70-to-100-h periods (Table 1), beginning at 12 noon. During each period, air was sampled though a Whatman GF/A, 25 mm-diameter, 1.6 µm-pore size glass filter housed in an adaptor with a circular aperture of 1 cm diameter. The adaptor was connected to one end of a 1 cm-diameter, 3 m-long PVC flexi-tube and was clamped to a metal railing at a forward angle of 45° in order to shield the filter from falling rain. Air was drawn through the filter at a rate of 9.0-10.5 L min⁻¹ (monitored with a 393-1130 rotameter; SKC Ltd) by a vacuum pump connected to the other end of the tube inside the building, resulting in, on average, about 42 and 60 m³ of air filtered for a 70- and 100-h sampling period, respectively. At the end of the sampling period, the filter was carefully retrieved with stainless steel tweezers and stored in aluminium foil in a metal cabinet.

Deposition during each period was sampled via 10-cm diameter glass funnels into two, 1 L amber glass bottles that were secured to a galvanised steel frame on the balcony of the roof. On completion of sampling, the combined contents of the bottles were vacuum-filtered through a 47-mm diameter, 0.45 μ m-pore size Whatman cellulose nitrate membrane filter housed in a glass-ceramic Buchner system. The filter was stored as above while the filtrate volume was determined in a series of 10 mL, 50 mL and 250 mL glass measuring cylinders before the contents were transferred to a narrow, 100 mL glass beaker. Provided sufficient volume was obtained, the electrical conductivity, κ (μ S cm $^{-1}$), as an indicator or marine influence, was measured with a WTW Tetra-Con electrode.

2.2. Identification and counting of suspected microplastics

Without further processing, all filters were examined under a Nikon SMZ800 stereomicroscope fitted with a 1x Achro objective (and attaining a magnification up to 63 X) that was connected to an Olympus SC30 camera operated by Olympus Stream software. Suspected MPs were identified with the aid of a metal probe and using criteria outlined in MERI (2014). That is, the response to the metal probe; lack of organic structures evident, except for biofouling; homogeneity of width (for plastic fibres); homogeneity in colour (except for possible partial bleaching of plastic fibres). For paint particles, identification was based on colour, brittleness and layering (Turner, 2021). Particles were classified according to colour, size (diameter or length; $<100 \, \mu m$ and with a detection limit of 30–50 μ m; 100–500 μ m; >500 μ m) and shape. With regard to the latter: fibres were defined as flexible, individual or intertwined particles with a ratio of length to diameter exceeding ten; fragments were irregular particles that included transparent and translucent films and layered structures, or more regular shapes that appear to have become detached from a larger structure; pellets were distinctly rounded or spherical entities; and foams were irregular but porous, sponge-like particulates.

2.3. Polymer identification by FTIR

With the aid of stainless-steel tweezers or the wetted tip of a sable hair paint brush (size 000) and under an OLYMPUS SZ40

G. Kyriakoudes and A. Turner Chemosphere 343 (2023) 140258

stereomicroscope, all suspected MPs that were large enough or not damaged or lost (n=160) were transferred from filters to a 2 mm-diameter Specac diamond compression cell. Individual particles were then analysed by Fourier transform-infrared (FTIR) spectroscopy in transmission mode with a Bruker Vertex 70 spectrometer coupled with a Hyperion 1000 microscope. Absorption spectra were recorded using OPUS software by averaging 32 scans in the region of 4000 to 600 cm⁻¹ and at a resolution of 4 cm⁻¹. Sample spectra were compared with various spectral databases of polymers and common materials, with a hit rate of >65% defined as our acceptance criterion for positive identification.

2.4. Cleanliness and controls

All glassware involved in sampling, measuring or analysis was washed three times with Millipore Milli-Q water (MQW) before being used or reused, and container openings and slide surfaces were covered with aluminium foil when not in use. Local laboratory surfaces were precleaned with MQW or ethanol and nitrile gloves were worn by the operator during microscopic analysis. Analysis of blank glass fibre and membrane filters under the stereomicroscope as above revealed no visible contamination from suspected MPs. As sampling controls, two 35-mL aliquots of MQW were poured into individual 1-L glass bottles via a glass funnel and the contents were subject to transference and filtration as above. Inspection under the microscope revealed the presence of four fibres in total and two on each filter, and subsequent analysis by FTIR identified rayon (n = 3) and cotton (n = 1). The total number of suspected MPs on sample filters that were observed under the microscope was subsequently corrected for this level of contamination (i.e., two fibrous particles) by subtraction.

3. Results

3.1. Environmental conditions

The meteorological conditions over the periods sampled in southwest England are summarised in Table 1. Thus, hourly wind speeds ranged from 2 to 29 mph, with directions most commonly from the north and northwest, and hourly temperatures ranged from 0 to 15 °C, with cooler temperatures generally associated with northerly winds. Rainfall over each sampling period ranged from <1 to about 14 mm, or 12–288 mL, and totalled about 60 mm, or 938 mL, with κ ranging from 55.7 to 329 μ S cm $^{-1}$. There were 18 days that were rain-free and the highest volumes of rainwater were generally associated with winds with a westerly component.

3.2. Number, concentration and deposition of suspected microplastics

Fig. 1 exemplifies a selection of suspected MPs identified under the

stereomicroscope that had been suspended in air (SMPs) and deposited from the atmosphere (DMPs) over southwest England. Overall, 45 particles of various colours, but mainly black or transparent, were identified in the twelve suspended samples, and, factoring in contamination from the controls, 117 particles of various colours, but mainly black, transparent or blue, were identified in the twelve deposited samples. Note also that deposited samples appeared to be cleaner (with regard to additional particulate matter on the filter) than suspended samples.

Table 2 shows the concentrations of suspected SMPs in the atmosphere and the depositional fluxes of DMPs. SMPs were calculated from particle number on each filter sampling suspended material, the duration of each sampling period (Table 1) and the time-averaged air flow, and resulting concentrations range from 0.016 m⁻³ during a period of southerly and easterly winds (period 12) to 0.238 m⁻³ during a spell of north/northwesterly winds (period 3). DMPs were calculated from the particle number on each filter isolating deposited material (after correction for contamination), sampling period duration (Table 1) and the capture area of the two funnels (157 cm²). Here, fluxes ranged from 0.47 m⁻² h⁻¹ during winds with a southerly or easterly component (period 12, and with a total rainfall of 1.53 mm and $\kappa = 66.9 \,\mu\text{S cm}^{-1}$) to $3.30 \text{ m}^{-2} \text{ h}^{-1}$ for a spell of north/northeasterly winds (period 6, and with a total rainfall of 0.96 mm and $\kappa = 82.3 \ \mu \text{S cm}^{-1}$). Although, overall, there was no significant relationship between the concentration of suspected SMPs and the depositional flux of suspected DMPs, there appears to be a group of seven points that are closely and significantly associated (Fig. 2). However, these data were not distinctive in terms of wind speed or direction, temperature, rainfall or κ .

Relative to the volume of rainfall captured, concentrations of suspected DMPs ranged from 36 to 1400 L^{-1} . While there was no clear dependence of suspected DMP concentration or deposition on wind speed or direction or on κ , there was a significant and non-linear (power law), inverse relationship between concentration in rainwater and rainfall volume (Fig. 3a). A weaker inverse relationship of the same form was also evident between the concentration of particles classified as fibres and rainwater volume (Fig. 3b).

3.3. Shape and size distribution of suspected microplastics

Table 3 shows the distribution of suspected SMPs and DMPs by shape and size sampled from the atmosphere of southwest England. Fibres were the dominant shape of particle in both cases. However, there was a greater proportion of fibres and a lower proportion of fragments subject to deposition than captured in suspension, and pellets were absent in deposited samples whereas foams were absent in suspended samples. With respect to size, there was a clear difference between particles suspended in and deposited from the atmosphere, with a lower proportion of fine particles ($<100~\mu m$) and a greater proportion of coarser particles ($>500~\mu m$ and up to 5 mm for fibres) in the latter.

Table 1
Dates and durations of the atmospheric sampling periods of southwest England, along with a summary of the (hourly) meteorological conditions (temperature range, wind speed range and principal wind direction). Rainfall was calculated from the volume of rainwater captured in the two collection bottles and specific conductance, κ , was determined by direct measurements of rainwater provided that sufficient sample was available.

Period	Start date	Duration, h	Temperature range, °C	Wind speed range, mph	Wind direction	Rainfall, mm	κ, mS cm ⁻¹
1	05/11/2021	70	2–14	2–11	N/NW	0.76	
2	08/11/2021	100	11–14	6–22	S/SW	7.71	55.7
3	12/11/2021	70	8–15	3–17	N/NW	0.76	
4	15/11/2021	100	9–13	2–11	W/NW	1.40	158
5	19/11/2021	70	4–12	2–17	N/NE	0.76	
6	22/11/2021	100	2–11	2–19	N/NE	0.96	82.3
7	26/11/2021	70	0–10	2–29	N/NW	3.69	329
8	29/11/2021	100	0–12	2–22	N/NW	12.36	115
9	03/12/2021	70	4–11	4–19	W/NW	14.14	42.2
10	06/12/2021	100	3–11	5–29	S/SW	11.97	260
11	10/12/2021	70	3–12	3–21	S/SW	3.69	92.6
12	13/12/2021	100	8–12	3–18	S/SE/E	1.53	66.9

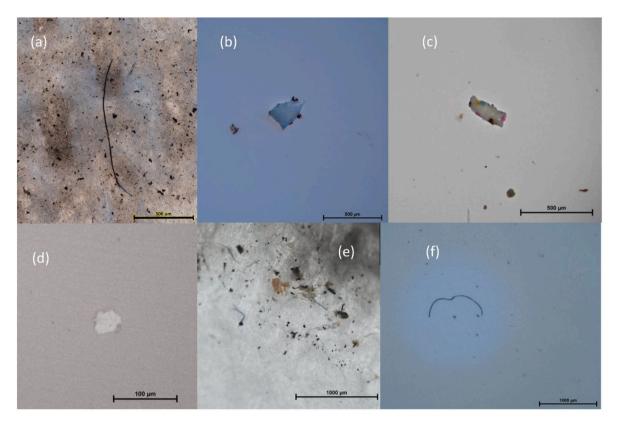


Fig. 1. Various suspected SMPs and DMPs sampled from the atmosphere of southwest England observed under the stereomicroscope. (a) A black fibre, (b) a translucent, blue film, (c) a multi-coloured fragment, (d) a white fragment, (e) various fibres and fragments, and (f) a blue fibre. Note the contamination of suspended samples (a) and (e) by dark particulate matter.

Table 2Number of suspected MPs and their concentrations suspended in air (SMPs), and the number, concentrations (relative to rainfall volume) and fluxes of suspected MPs deposited from the atmosphere (DMPs) during the twelve sampling periods of southwest England.

Period	No. SMPs	${ m SMP}~{ m m}^{-3}$	No. DMPs	${\rm DMP}\;{\rm L}^{-1}$	${\rm DMP}~{\rm m}^{-2}~{\rm h}^{-1}$
1	3	0.068	9	750	2.02
2	4	0.067	15	124	2.36
3	9	0.238	8	667	1.79
4	2	0.037	7	318	1.10
5	2	0.053	8	667	1.79
6	1	0.018	21	1400	3.30
7	5	0.119	7	121	1.57
8	3	0.055	10	52	1.57
9	5	0.119	8	36	1.79
10	6	0.095	18	96	2.83
11	4	0.095	3	52	0.67
12	1	0.016	3	125	0.47
mean	3.8	0.082	10	367	1.77
median	3.5	0.068	8	124	1.79
total	45		117		

3.4. Origin and polymeric makeup of suspected microplastics

Table 4 summarises the results arising from the FTIR analysis of suspected SMPs and DMPs sampled from the atmosphere, with examples of sample and matching reference spectra shown in Fig. 4. Here, DMP data have been corrected for the presence of two cellulosic fibres as contaminants in each sample; specifically, one cotton fibre and one rayon fibre, or two rayon fibres where cotton was not present, were subtracted from each tally. Overall, about 70% of particles analysed returned a positive identification, and of these about 46% were synthetic, and largely petroleum-based polymers. The most abundant

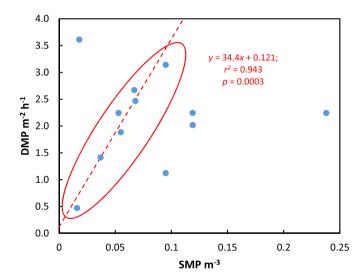
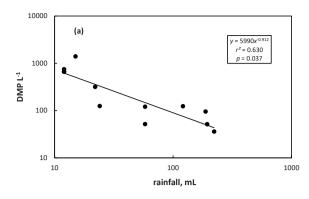


Fig. 2. Depositional flux of suspected DMPs versus concentration of suspected SMPs. Data highlighted by the red ellipse exhibit a significant relationship whose best-fit line and statistical parameters are annotated.

thermoplastics were polyamides (including nylon), acrylic, polyester (including polyethylene terephthalate) and polyvinyl alcohol, and about 30% of synthetic polymers were identified as paints. About 50% of positively identified particles were semi-synthetic fibres, with the regenerated cellulosic, rayon (viscose), present in all but one case, and $<\!5\%$ of particles were natural (including cotton).

There was a lower proportion of synthetic polymers and a higher proportion of semi-synthetic particles among DMPs than SMPs. Regarding the synthetic polymers, there appeared to be some



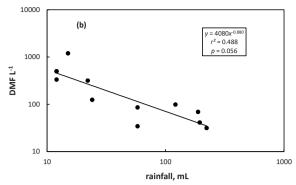


Fig. 3. Concentrations of (a) suspected DMPs and (b) suspected DMP fibres, DMF, per L of rainwater versus volume of rainfall captured. Also shown are the best power line fits through the dataset along with related statistical parameters.

Table 3Percentage distribution of suspected SMPs and DMPs by shape and size sampled from the atmosphere of southwest England.

	SMPs ($n=45$)	DMPs ($n = 117$)
fibres	64.4	80.3
fragments	24.2	12.8
pellets	11.1	0
foams	0	6.8
<100 μm	15.6	1.7
100-500 μm	42.2	24.8
>500 µm	42.2	73.5
synthetic	35.6	31.6
semi-synthetic	26.7	37.6
natural	0	4.3
unidentified	37.8	26.5

Table 4
Polymeric or material composition of suspected SMPs and DMPs that were positively identified based on FTIR analysis. Synthetic polymers are petroleum-based thermoplastics and paints (with the latter identified from resins and/or additives and pigments). Note that nylon is included under polyamide and

polyethylene terephthalate is included under polyester.

Polymer/material	SMPs $(n=28)$	DMPs $(n = 86)$
synthetic	16	37
acrylic	1	6
epoxy resin	0	1
expanded polyurethane	0	1
polyamide	3	9 ^a
polyester	5 ^b	1
polyetherimide	1	1
polypropylene	0	1
polyvinyl acetate	0	2
polyvinyl alcohol	1	4
polyvinyl chloride	0	1
other	5	10
paints	4	12
semi-synthetic	12	44
rayon	11	44
other	1	0
natural	0	5
cotton	0	1
Other	0	4

^a One sample returned a match for nylon and silk combined.

fractionation between the two media, with a greater proportion of acrylic being deposited and a greater proportion of polyester in suspension.

4. Discussion

Not all particles considered in the present study were petroleum-based MPs, although the semi-synthetic cellulosic that is shaped by extrusion, rayon, is often reported in the MP literature (Woodall et al.,

2014; Comnea-Stancu et al., 2017; Higgins and Turner, 2023). Nevertheless, all but at most four particles identified were anthropogenic in origin, and all anthropogenic particles that were not thermoplastics or paints (rayon, methyl cellulose and cotton) were fibrous. Thus, for the purposes of the discussion, we hereafter refer to MPs as petroleum-based particles and other anthropogenic microfibres, and with regard to any quantitative comparisons or calculations, we assume that all particles not analysed or unidentified by FTIR are MPs based on their visual characteristics.

The concentrations of MPs are heterogeneous in both suspended and deposited samples collected in Plymouth, southwest England, during twelve periods in late autumn. This observation reflects the multitude of local and more distal primary and secondary sources (including the ocean; Brahney et al., 2021) and variety of meteorological conditions and air masses encountered. In suspension, there is some evidence that wind direction and speed are partly responsible for this variation, while an inverse relationship between deposited MPs and volume of rainfall suggests that a high proportion of particles is washed out with incipient rain and that the abundance or availability of particles progressively declines as rainfall continues. Concentrations of particulate matter in the lower troposphere are known to be responsive to and/or negatively correlated with the length or intensity of precipitation (Ribeiro et al., 2003; Luan et al., 2019; Zhou et al., 2021), although confounding factors include time of day, wind speed and direction, air humidity and rainfall height (Dris et al., 2016; Kluska et al., 2020). Despite these observations, rainfall intensity or duration in the present study was not inversely related to the concentration of SMPs subsequently encountered in the atmosphere. This suggests that the stock of airborne MPs is relatively rapidly replenished by the passage of air masses of different origin and age.

The concentrations of SMPs and fluxes of DMPs sampled from the atmosphere over southwest England are compared with values reported in the literature in Table 5. Here, inter-study differences in the type of particle considered have been noted; specifically, whether cellulosic semi-synthetic fibres (e.g., rayon) and natural fibres (e.g., cotton) have been included (based on sample digestion), or whether all particle shapes or just fibres have been counted. With these differences in mind, concentrations of SMPs in Plymouth air (0.082 \pm 0.060 m $^{\text{-}3}$) are similar to concentrations reported for the coastal atmosphere of Pacific China and the French Pyrenees, greater than concentrations given for the open Pacific Ocean, and lower than concentrations measured in large cities (Paris, Shanghai, Guangzhou). The mean depositional flux of DMPs for southwest England (1.77 \pm 0.81 m $^{-2}$ h $^{-1}$) is on the same order of magnitude as most mean fluxes reported in the literature but is considerably lower than the value reported for the city of London.

Amongst the studies in Table 5, Yuan et al. (2023) have reported both concentrations of SMPs and depositional fluxes of DMPs. The authors propose that the two measures can be combined to calculate a dimensionless washout ratio, *W*, in order to evaluate removal efficiency,

^b One sample returned a match for polyester and cotton combined.

G. Kyriakoudes and A. Turner Chemosphere 343 (2023) 140258

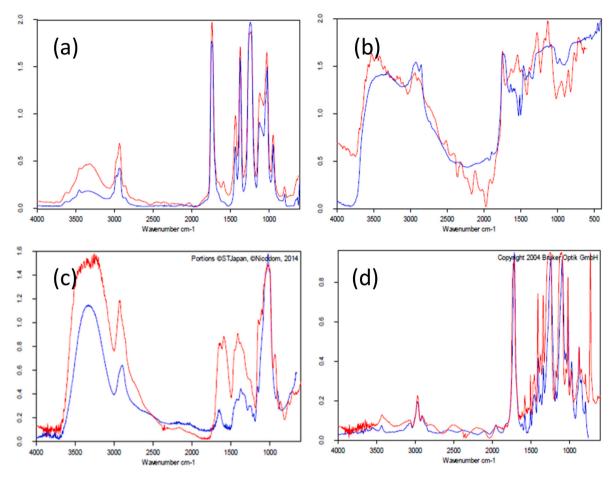


Fig. 4. Reference spectra (blue) and sample FTIR spectra (red) for (a) a polyvinyl acetate fragment, (b) a vehicle paint flake, (c) a rayon fibre and (d) a polyester fibre. Note the broad peak centred on 3300 cm^{-1} (O–H stretching) in (c) that distinguishes it from cotton (a narrower and less distinctive peak).

assuming that the majority of DMPs are subject to wet deposition:

$$W = DMP L^{-1}*10^{3}/SMP m^{-3}$$
 (1)

In the present study, both variables on the right-hand side of Eq. (1) are given in Table 2 and yield periodic values of W ranging from 3.0×10^5 (period 9) to 7.7×10^7 (period 6) and a median value of 1.8×10^6 . By comparison, Yuan et al. (2023) report a range for Guangzhou City from 2.2×10^3 to 1.5×10^6 . Thus, despite the present study not targeting specific rainfall episodes (that is, not discriminating dry and wet deposition), environmental conditions over southwest England appear to be more favourable at removing MPs from the atmosphere.

A similar approach can be adopted to evaluate the removal efficiency of MPs by size, shape (specifically, fibrous versus non-fibrous) and polymer type. Here, the data in Tables 3 and 4 have been used for the whole timeframe of sampling (1020 h) and summed volumes collected (593 m³ of air and 938 mL of rainwater) because many polymers or shapes were not detected during specific periods. Polymer-specific values of W shown in Fig. 5 reveal a range from about 1.3×10^5 for polyester to about 3.8×10^6 for acrylic, while values for microplastic fibres ($\sim 5 \times 10^6$) are considerably greater than values for non-fibrous MPs ($\sim 7 \times 10^5$) and values for larger (>500 µm) MPs ($\sim 2.8 \times 10^6$) are an order of magnitude greater than values for smaller (<100 µm) MPs ($\sim 2.3 \times 10^5$). That is, acrylic is considerably more prone to atmospheric removal than polyester, and removal favours larger particles that are fibrous in nature.

The reasons for these differences amongst polymer types are unclear but could be related to variations in their precise size or shape, or to more general aerodynamic properties (hence vertical distribution in the atmosphere) or hygroscopicity. Nevertheless, our observations suggest that polymers like acrylic have a tendency to be readily removed from the atmosphere whereas polymers like polyester have a greater propensity to remain airborne and, therefore, be transported longer distances. Partly consistent with this assertion, we note that Wei et al. (2022) reported a reduction in the proportion of total microplastics as polyethylene terephthalate (included as a polyester in Fig. 5) in the Qing River, Beijing, following a rainfall event.

Periodic data can also be employed to estimate, empirically, the net depositional velocities of MPs, ν_d (m h⁻¹), in the atmosphere of southwest England:

$$v_{\rm d} = \rm DMP \ m^{-2} \ h^{-1}/SMP \ m^{-3}$$
 (2)

The value of ν_d reflects the time integration of both dry and wet deposition of all MP shapes, sizes and polymer types and ranges from 7.1 m h $^{-1}$ for period 11 to 183 m h $^{-1}$ for period 6, with an overall periodic median of 37.5 m h $^{-1}$. Although wet deposition was incorporated into these estimations, ν_d was not correlated with rainfall, presumably because of the confounding effects of rainfall intensity and duration and other environmental variables. Nevertheless, and despite being specific to the location and conditions encountered, we note that Wright et al. (2020) provide a theoretical estimate of ν_d for representative fibres (400 μ m by 20 μ m and a density of 1.184 g cm $^{-3}$) falling through a dry atmosphere of 216 m h $^{-1}$ that is similar to our highest value. For MPs raised to an elevation of 5000 m (or about halfway up in the troposphere), the range in ν_d is equivalent to a range of residence times from about 27 h to 700 h.

An additional, important finding of the present study that is consistent with other recent investigations (e.g., Liu et al., 2019b; Roblin et al., 2020) is the significance of semi-synthetic polymers (fibres) in the

 $\label{thm:constraints} \textbf{Table 5} \\ \textbf{Mean $(\pm$ one standard deviation)$ or range of concentrations of MPs (including or excluding cellulosics) suspended in and deposited from the atmosphere that have been reported in the literature.}$

		3	
SMPs	Particle type	SMP m^{-3}	Reference
Paris, France	Fibres, including cellulosic	0.3 to 1.5	Dris et al. (2017)
Open Pacific Ocean	Excluding cellulosic fibres	~0.01	Liu et al. (2019a)
Shanghai, China	Including rayon	up to 4.2	Liu et al. (2019b)
Coastal Pacific Ocean	Including cellulosic	0.042 \pm	Wang et al. (2020)
of China	fibres	0.025	
Open East Indian	Including cellulosic	0.004 \pm	Wang et al. (2020)
Ocean	fibres	0.006	
French Pyrenees	Including cellulosic	0.09 to	Allen et al. (2021)
	fibres	0.66	
Guangzhou, China	Including cellulosic	$0.17~\pm$	Yuan et al. (2023)
	fibres	0.01	
Plymouth, UK	Including cellulosic	$0.082~\pm$	This study
	fibres	0.060	
DMPs		DMP $m^{-2} h^{-1}$	•
Paris, France	Including cellulosic	4.58 \pm	Dris et al., 2016
	fibres	4.00	
Dongguan, China	Including cellulosic	7.29 to	Cai et al. (2017)
	fibres	13.0	
French Pyrenees	Including cellulosic fibres	15.2	Allen et al. (2019)
Northern Germany	Excluding	11.5	Klein and Fischer
	cellulosic fibres		(2019)
Protected US areas	Including cellulosic	$5.50 \pm$	Brahney et al.
	fibres	0.25	(2020)
Irish Atlantic Coast	Fibres, including cellulosic	4.17	Roblin et al. (2020)
London	Including cellulosic	32.1 \pm	Wright et al. (2020)
	fibres	6.96	
Mount Derak, Iran	Excluding	$0.51~\pm$	Abbasi and Turner
	cellulosic fibres	0.20	(2021)
Shiraz, Iran	Excluding	2.65 \pm	Abbasi and Turner
	cellulosic fibres	1.44	(2021)
Gulf of Gdansk, Poland	Including cellulosic	0.41 \pm	Szewc et al. (2021)
	fibres	0.33	
South Central Ontario, Canada	Including cellulosic fibres	2.38	Welsh et al. (2022)
Coastal Indonesia	Excluding	0.63 \pm	Purwiyanto et al.
	cellulosic fibres	0.54	(2022)
Guangzhou, China	Including cellulosic	2.75 \pm	Yuan et al. (2023)
	fibres	0.31	
Plymouth, UK	Including cellulosic	1.77 \pm	This study
	fibres	0.81	

atmosphere, or a classification of anthropogenic particles that is often deliberately excluded through chemical destruction (Allen et al., 2019; Klein and Fischer, 2019; Liu et al., 2019a; Abbasi and Turner, 2021; Purwiyanto et al., 2022). Finnegan et al. (2022) recently reviewed existing literature on anthropogenic particles (mainly microfibres) reported in the atmosphere. Where polymer identification had been undertaken, the collective data revealed that 57% of particles were semi-synthetic (cellulosic) and 23% were petroleum-based. By comparison, data for suspended and deposited MPs in the present study yield respective values of 49% and 46%. Currently, synthetic polymers like polyester dominate the global production of textile fibres (>60%; Textile Exchange, 2022) and a higher proportion of semi-synthetic fibres in the atmosphere (and in particular, rayon), therefore, may be attributed to their lower durability and greater release from in-life and end-of-life products (Finnegan et al., 2022).

While both plastic and semi-synthetic types of fibre are predicted to pose similar health risks through inhalation (Athey and Erdle, 2021), the lower environmental persistence of cellulosic material means that semi-synthetic fibres may release chemical additives, like colourants, ultraviolet light stabilisers and antimicrobial agents, as well as any contaminants acquired from the environment, more readily (Ladewig

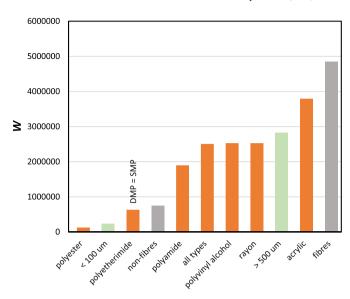


Fig. 5. Relative efficiency for atmospheric removal of MPs, *W*, by polymer type (and in grey, by shape, and in green, by size) over southwest England.

et al., 2015). In the present study, a maximum microplastic concentration in air of $0.238~\text{m}^{-3}$ is equivalent to the daily inhalation by an adult standing still of about five particles, at least within the size range detected microscopically.

5. Conclusions

SMPs and DMPs sampled from the atmosphere of southwest England during twelve periods over a 42-day timeframe exhibit temporal variations in abundance but fibres of the semi-synthetic cellulosic, rayon, were persistently the most common type of particle. Incipient rainfall appears to washout SMPs from air, with the volume of precipitation displaying an inverse relationship with DMP concentration. However, the combined periodic data reveal that SMPs are removed by deposition differentially and according to size, shape and polymer type, suggesting that these characteristics may be critical to the transportation and residence times of MPs in the atmosphere. The combined data also allow site- and environment-specific MP settling velocities to be calculated, with estimates ranging from about 7 to 180 m $\,h^{-1}$ and a similar order of magnitude to published, theoretical values. Although the precise results of the study are specific to the location and environmental conditions encountered, the broad findings are likely to be more generally applicable in improving our understanding of atmospheric MP occurrence, transport and removal.

CRediT authorship contribution statement

Giannis Kyriakoudes: Conceptualization, Methodology, Investigation, Formal analysis, Writing – original draft. **Andrew Turner:** Conceptualization, Investigation, Formal analysis, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgements

We are grateful to Mr Billy Simmonds and Mr Richard Hartley (University of Plymouth) for technical support. The comments of two anonymous reviewers greatly improved the manuscript.

References

- Abbasi, S., 2021. Microplastics washout from the atmosphere during a monsoon rain event. J. Hazard. Mater. Adv. 4, 100035.
- Abbasi, S., Turner, A., 2021. Dry and wet deposition of microplastics in a semi-arid region (Shiraz, Iran). Sci. Total Environ. 786, 147358.
- Abbasi, S., Turner, A., Hoseini, M., Amiri, H., 2021. Microplastics in the Lut and Kavir deserts, Iran. Environ. Sci. Technol. 55, 5993–6000.
- Aeschlimann, M., Li, G., Kanji, Z.A., Mitrano, D.M., 2022. Microplastics and nanoplastics in the atmosphere: the potential impacts on cloud formation processes. Nat. Geosci. 15, 967–975.
- Allen, S., Allen, D., Phoenix, V.R., Le Roux, G., Jimenez, P.D., Simonneau, A., Binet, S., Galop, D., 2019. Atmospheric transport and deposition of microplastics in a remote mountain catchment. Nat. Geosci. 12, 339–344.
- Allen, S., Allen, D., Baladima, F., Phoenix, V.R., Thomas, J.L., Le Roux, G., Sonke, J.E., 2021. Evidence of free tropospheric and long-range transport of microplastic at Pic du Midi Observatory. Nat. Commun. 12, 7242.
- Athey, S.N., Erdle, L.M., 2021. Are we underestimating anthropogenic microfiber pollution? A critical review of occurrence, methods, and reporting. Environ. Toxicol. Chem. 41, 822–837.
- Bergmann, M., Mützel, S., Primple, 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.
- Brahney, J., Hallerud, M., Heim, E., Hahnenberger, M., Sukumaran, S., 2020. Plastic rain in protected areas of the United States. Science 368, 1257–1260.
- Brahney, J., Mahowald, N., Prank, M., Prather, K.A., 2021. Constraining the atmospheric limb of the plastic cycle. Earth, Atmos. Planet. Sci. 118, e2020719118.
- 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. Control Ser. https://doi.org/ 10.1007/s11356-017-0116-x.
- Comnea-Stancu, I.R., Wieland, K., Ramer, G., Schwaighofer, A., Lendl, B., 2017. On the identification of rayon/viscose as a major fraction of microplastics in the marine environment: discrimination between natural and manmade cellulosic fibers using Fourier transform infrared spectroscopy. Appl. Spectrosc. 71, 939–950.
- 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.
- 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.
- Evangeliou, N., Tichý, O., Eckhardt, S., Zwaaftink, C.G., Brahney, J., 2022. Sources and fate of atmospheric microplastics revealed from inverse and dispersion modelling: from global emissions to deposition. J. Hazard Mater. 432, 128585.
- Feng, S.S., Lu, H.W., Tian, P.P., Xue, Y.X., Lu, J.Z., Tang, M., Feng, W., 2020. Analysis of microplastics in a remote region of the Tibetan Plateau: implications for natural environmental response to human activities. Sci. Total Environ. 739, 140087.
- Finnegan, A.M.D., Susserott, R., Gabbott, S.E., Gouramanis, C., 2022. Man-made natural and regenerated cellulosic fibres greatly outnumber microplastic fibres in the atmosphere. Environ. Pollut. 310, 119808.
- 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. Health 1, 1–5.
- González-Pleiter, M., Edo, C., Aguilera, A., Viúdez-Moreiras, D., Pulido-Reyes, G., González-Toril, E., Osuna, S., de Diego-Castilla, G., Leganés, F., Fernández-Piñas, F., Rosal, R., 2021. Occurrence and transport of microplastics sampled within and above the planetary boundary layer. Sci. Total Environ. 761, 143213.

- Higgins, C., Turner, A., 2023. Microplastics in surface coastal waters around Plymouth, UK, and the contribution of boating and shipping activities. Sci. Total Environ. 893, 164695.
- Klein, M., Fischer, E.K., 2019. Microplastic abundance in atmospheric deposition within the Metropolitan area of Hamburg, Germany. Sci. Total Environ. 685, 96–103.
- Kluska, K., Piotrowicz, K., Kasprzyk, I., 2020. The impact of rainfall on the diurnal patterns of atmospheric pollen concentrations. Agric. For. Meteorol. 291, 108042.
- Ladewig, S.M., Bao, S., Chow, A.T., 2015. Natural fibers: a missing link to chemical pollution dispersion in aquatic environments. Environ. Sci. Technol. 49, 12609–12610.
- Liu, K., Wu, T., Wang, X., Song, Z., Zong, C., Wei, N., Li, D., 2019a. Consistent transport of terrestrial microplastics to the ocean through atmosphere. Environ. Sci. Technol. 53, 10612–10619.
- Liu, K., Wang, X., Fang, T., Xu, P., Zhu, L., Li, D., 2019b. Source and potential risk assessment of suspended atmospheric microplastics in Shanghai. Sci. Total Environ. 675, 462–471.
- Long, X., Fu, T.M., Yang, X., Tang, Y., Zheng, Y., Zhu, L., Shen, H., Ye, J., Wang, C., Wang, T., Li, B., 2022. Efficient atmospheric transport of microplastics over Asia and adjacent oceans. Environ. Sci. Technol. 56, 6243–6252.
- Luan, T., Zhang, T., Guo, L., 2019. Below-cloud aerosol scavenging by different-intensity rains in Beijing City. J. Meteorol. Res. 33, 126–137.
- Mehmood, T., Hassan, M.A., Faheem, M., Shakoor, A., 2021. Why is inhalation the most discriminative route of microplastics exposure? Environ. Sci. Pollut. Control Ser. 29, 49479–49482.
- MERI, 2014. Guide to Microplastic Identification. Marine & Environmental Research Institute, Blue Hill, ME, USA.
- Purwiyanto, A.I.S., Prartono, T., Riani, E., Naulita, Y., Cordova, M.R., Koropitan, A.F., 2022. The deposition of atmospheric microplastics in Jakarta-Indonesia: the coastal urban area. Mar. Pollut. Bull. 174, 113195.
- Ribeiro, H., Cunha, M., Abreu, I., 2003. Airborne pollen concentration in the region of Braga, Portugal, and its relationship with meteorological parameters. Aerobiologia 19, 21–27.
- Roblin, B., Ryan, M., Vreugdenhil, A., Aherne, J., 2020. Ambient atmospheric deposition of anthropogenic microfibers and microplastics on the western periphery of Europe (Ireland). Environ. Sci. Technol. 54, 11100–11108.
- Szewc, K., Graca, B., Dolega, A., 2021. Atmospheric deposition of microplastics in the coastal zone: characteristics and relationship with meteorological factors. Sci. Total Environ. 761, 143272.
- Textile Exchange, 2022. Preferred Fiber & Materials Market Report. https://textileexchange.org/knowledge-center/reports/preferred-fiber-and-materials/.
- Turner, A., 2021. Paint particles in the marine environment: an overlooked component of microplastics. Water Res. X 12, 100110.
- Wang, X., Li, C., Liu, K., Zhu, L., Song, Z., Li, D., 2020. Atmospheric microplastic over the south China sea and east Indian Ocean: abundance, distribution and source. J. Hazard Mater. 389, 121846.
- Wei, Y., Dou, P., Xu, D., Zhang, Y., Gao, B., 2022. Microplastic reorganization in urban river before and after rainfall. Environ. Pollut. 314, 120326.
- Welsh, B., Aherne, J., Paterson, A.M., Yao, X., McConnell, C., 2022. Atmospheric deposition of anthropogenic particles and microplastics in south-central Ontario, Canada. Sci. Total Environ. 835, 155426.
- Woodall, L.C., Sanchez-Vidal, A., Canals, M., Paterson, G.L., Coppock, R., Sleight, V., Calafat, A., Rogers, A.D., Narayanaswamy, B.E., Thompson, R.C., 2014. The deep sea is a major sink for microplastic debris. R. Soc. Open Sci. 1, 140317.
- Wright, S.L., Ulke, J., Font, A., Chan, K.L.A., Kelly, F.J., 2020. Atmospheric microplastic deposition in an urban environment and an evaluation of transport. Enviro. Int. 136, 105411
- Yuan, Z., Pei, C., Li, H., Lin, L., Liu, S., Hou, R., Liao, R., Xu, X., 2023. Atmospheric microplastics at a southern China metropolis: occurrence, deposition flux, exposure risk and washout effect of rainfall. Sci. Total Environ. 869, 161839.
- Zhang, Y., Kang, S., Allen, S., Allen, D., Gao, T., Sillanpää, M., 2020. Atmospheric microplastics: a review on current status and perspectives. Earth Sci. Rev. 203, 103118.
- Zhou, B., Liu, D., Yan, Y., 2021. A simple new method for calculating precipitation scavenging effect on particulate matter: based on five-year data in eastern China. Atmosphere 12, 759.