Freshwater microplastic concentrations vary through both space *and* time

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Abstract

Plastic pollution represents one of the most salient indicators of society's impact on the environment. The microplastic component of this is ubiquitous, however, microplastic studies are seldom representative of the locations they sample. Over 12 months we explored spatiotemporal variation in microplastic prevalence across a freshwater system and in atmospheric deposition within its catchment, in one of the most temporally comprehensive studies of microplastic pollution. Microplastics were quantified in low concentrations (max 0.4 particles L^{-1}) at all freshwater sites, including upstream of urban areas, and on rivers that do not receive wastewater treatment plant effluent. Extrapolated microplastic abundances at each site varied by up to 8 orders of magnitude over the course of the sampling campaign, suggesting that microplastic surveys that do not account for temporal variability misrepresent microplastic prevalence. Whilst we do not wish to underplay the potential impacts of microplastic particles in the environment, we argue that microplastic pollution needs to be placed in a more critical context, including assessment of temporal variability, to appropriately inform legislators and consumers.

Capsule

The main findings of this research are the extent to which freshwater microplastic concentrations are shown to vary with time, and the influence of this on flux calculations.

Keywords

Microplastic, Temporal variation, Freshwater, Atmospheric deposition

1. Introduction

Microplastic particles (<5 mm) are an environmental pollutant of substantial public and scientific concern. Functioning as pollutants in contemporary environmental systems, and demarking human activity for centuries to come as techno-fossils, microplastic particles are a widespread form of plastic waste. Their prevalence in the marine environment has been reported since the early 1970s (Carpenter et al. 1972; Carpenter and Smith, 1972), and their presence in estuarine systems (Zhao et al. 2015; Gallagher et al, 2016; Gray et al. 2018) and freshwater environments (Zhang et al. 2015; Klein et al. 2015; Peng et al. 2018; Mani et al. 2019; Watkins et al. 2019) has also been documented. However, whilst microplastics are thought to be ubiquitous beyond these systems (Rochman, 2018), records of microplastic pollution are often reported at low spatial and temporal resolutions.

Freshwater catchments are a key pathway in the transport of microplastic debris, which accumulates in marine environments (Wagner et al. 2014). Sources of freshwater microplastic pollution are known to be varied, including wastewater treatment plants (WWTPs), urban centres and road runoff (Horton et al. 2017), industry (Lechner and Ramler, 2015), the atmosphere (Dris et al. 2016), and the degradation of larger items of plastic waste. However, the predominate focus of freshwater microplastic studies has been on downstream reaches of large, highly developed rivers in China, Europe, and North America (Figure 1). Understanding how microplastic concentrations vary along a river's course is lacking, yet it is critical to understanding this key source and pathway of microplastic particles.

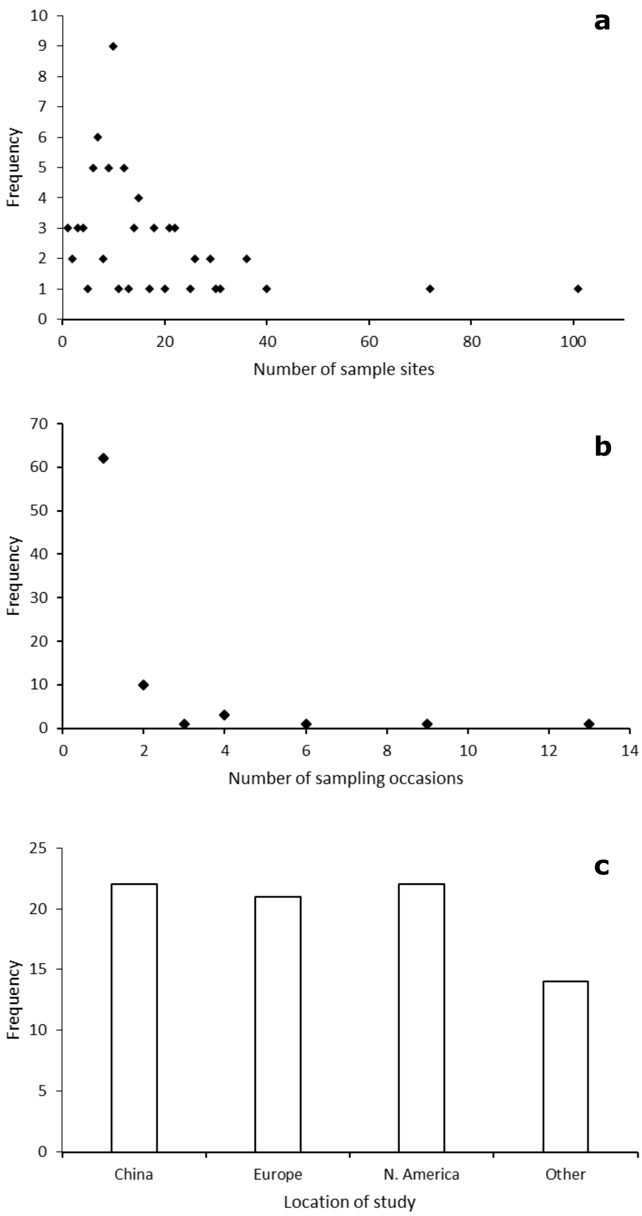

Figure 1: The number of sample sites (a), number of sampling occasions (b), and location (c) of 79 freshwater microplastic studies (see methods for Web of Knowledge search parameters used to identify relevant publications).

Freshwater sampling campaigns also rarely account for temporal variability in their sampling campaigns (Figure 1) (Schmidt et al. 2017), limiting the extent to which measurements are representative of that site beyond the time of sample collection. Whilst studies that quantify microplastics in different freshwater environments are of great value, they are not able to further our understanding of microplastic sources and distributions without careful consideration of the intra-site variability over representative time periods (Prata et al. 2019).

A comprehensive understanding of the sources and vectors of freshwater microplastic pollution is further limited by a lack of consideration of atmospheric deposition to what is a largely open system. Atmospheric deposition is a known source of anthropogenic particles found in both the benthic and suspended sediments of freshwater systems, including Spheroidal Carbonaceous Particles and Inorganic Ash Spheres (Rose et al. 2012). However, the study of airborne microplastic particles is limited to a few records of their presence in urban (Dris et al. 2016; Cai et al. 2017; Bergman et al. 2019; Stanton et al. 2019), and remote (Allen et al. 2019; Bergman et al. 2019) atmospheric deposition.

To address these research gaps, this study presents the findings of 12 months of freshwater and atmospheric sampling across the River Trent catchment, UK (Figure 2). We sampled the upstream reaches of the River Trent (RT), and the entire length of two of its tributaries, the River Leen (RL), and River Soar (RS), as well as atmospheric deposition within the Trent catchment. By sampling sites upstream of urban centres and at points without WWTP inputs, we assess the contribution of these previously cited sources of microplastic pollution to freshwater microplastic loads, and highlight the importance of accounting for temporal variation when disseminating microplastic findings.

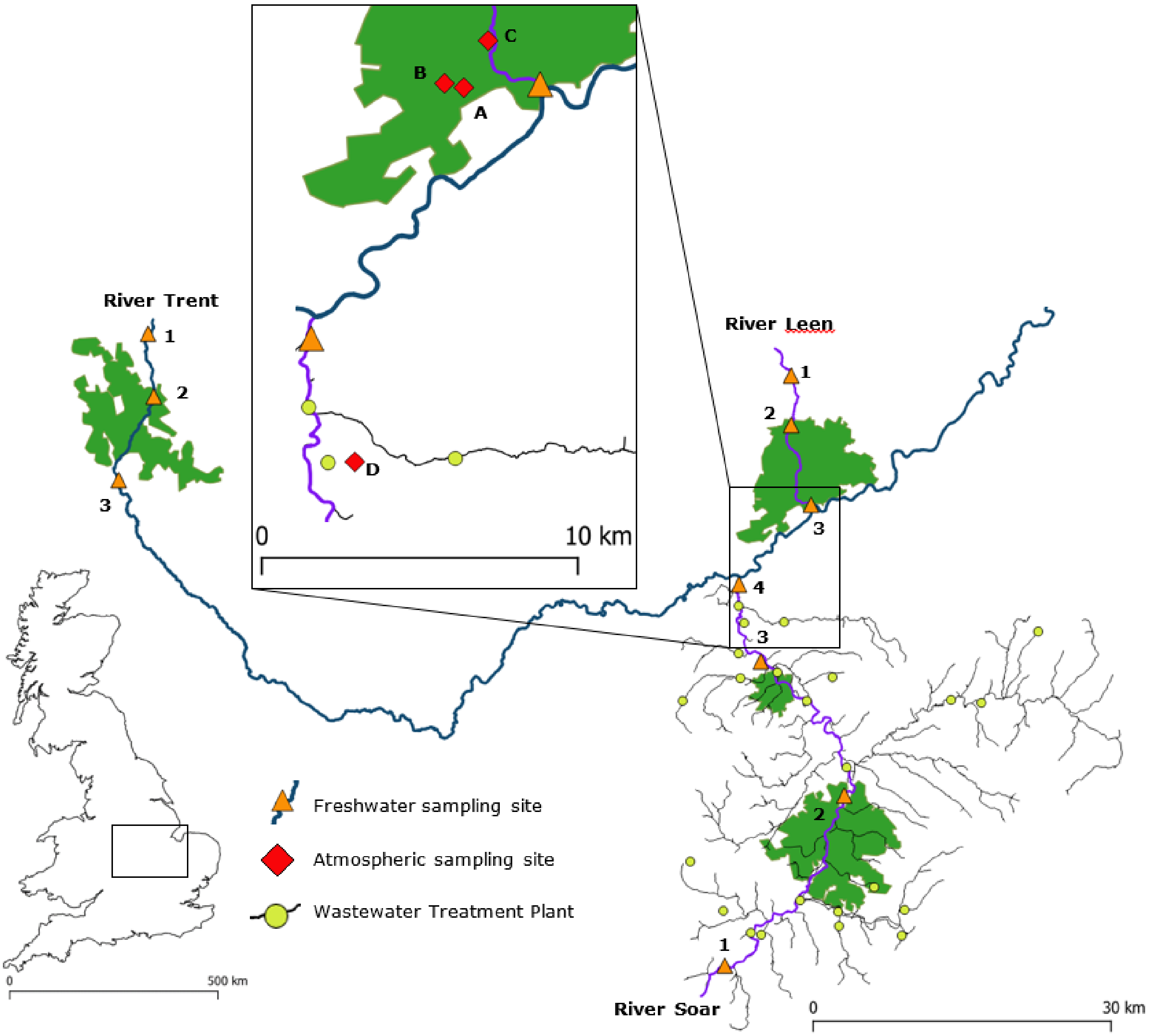

Figure 2: Locations of freshwater (numbered) and atmospheric (lettered) sampling sites within the Trent Catchment, UK. Green areas represent the urban areas of Stoke-on-Trent (River Trent), Nottingham (River Leen), Leicester (River Soar upstream) and Loughborough (River Soar downstream). The exact location of each sample site is provided in Table S1.

2. Methods

2.1 Literature search protocol for freshwater microplastic studies

Figure 1 was collated from the results of a Web of Knowledge publication search conducted on 24/07/2019 with the aim of collating the number of sampling sites, number of sampling occasions, and location of freshwater microplastic studies. This search was conducted using the following parameters:

Topic search for:

- Freshwater microplastic
  OR
- River Microplastic
  OR
- Lake Microplastic

Though these search terms are unlikely to have provided a complete coverage of all freshwater microplastic studies, they yielded 343 results from 2012 to 2019. All review articles and laboratory studies were excluded, leaving 93 studies (supplementary references), of which the authors had access to 79 that contained the necessary information to meet the above aim.

2.2 Sample sites, and sample collection and processing

Sample site locations (Figure 2), and the procedure for contamination control, sample collection and processing are described in detail in Stanton et al. (2019). In brief, every four weeks from 20/11/2017 to 23/10/2018 (12 months) 30 L of freshwater was collected from each of the 10 sites across three rivers within the Trent catchment (Figure 2). Samples were collected from the riverbank using a metal bucket on a telescopic pole. Each sample was concentrated onto a 63 μm sieve in the field, and the retained material was transferred into a glass sample bottle using distilled water. The location of freshwater sampling sites enabled this study to assess microplastic pollution near the source of rivers, up- and downstream of urban centres, and at locations that do and do not receive WWTP effluent. Exact locations were determined by site accessibility.

Atmospheric samples were collected using a scaled-down adaptation of the methods used by Dris et al. (2016). Atmospheric deposition was collected in 2.5 L amber glass bottles using a 12 cm diameter (0.0113 m^2) glass funnel. Where Dris et al. (2016) used a sampling surface area of 0.325 m^2, more recent research of microplastic deposition has favoured smaller sampling apparatus with a sampling surface area of 0.0177 m^2 (Cai et al. 2017), 0.014 m^2 (Allen et al. 2019), and 0.0113 m^2 (Klein and Fisher, 2019; Stanton et al. 2019). Atmospheric samples were collected fortnightly for 12 months from 23/11/2017 to 25/10/2018. To assess the potential influence of intra-site variation additional sampling was also conducted at site D, in which five replicate samples were collected from the same rooftop between 04/12/2018 and 11/12/2018. All buildings were 2 storeys (three floors) high.

Freshwater samples were treated with 30% H_2O_2 to remove organic matter before being vacuum filtered onto gridded cellulose nitrate filter papers with a 0.45 μm pore size (Whatman ME 25/41). The contents of the amber glass bottles used to collect atmospheric deposition was concentrated onto a 38 μm sieve in the laboratory, and the bottles triple rinsed with distilled water. The retained material was vacuum filtered onto the same gridded filter papers as the freshwater samples. Due to the mesh apertures of the sieves used to reduce the sample volumes, this methodology was unable to isolate all particles smaller than 63 μm for the freshwater samples, or smaller than 38 μm for the atmospheric samples.

2.3 Microplastic identification

Samples were initially visually inspected under a dissection microscope (Medline Scientific CETI Varizoom-10, Chalgrove, UK). Textile fibres were categorised according to Stanton et al. (2019), and the grid reference for all suspected non-fibrous microplastic particles was recorded. This grid reference aided the subsequent FTIR spectroscopy of these particles. Analysed particles were subjected to one of the following FTIR spectroscopy techniques: Attenuated total reflectance (ATR) FTIR spectroscopy (Bruker Tensor 27 FTIR spectrometer [Bruker Optics, Coventry, UK] equipped with a Golden Gate ATR accessory [Specac, Orpington, UK]), reflectance FTIR microscopy (Bruker Hyperion 2000 microscope [Bruker Optics, Coventry, UK]), or using an ATR-FTIR objective (Bruker Lumos microscope

[Bruker Optics, Coventry, UK]). Spectra were identified using Bruker's demonstration library.

2.4 Statistical analysis

Non-parametric tests were carried out on the freshwater dataset. Kruskal-Wallis tests were performed to determine whether microplastic concentrations over the sampling period were significantly different between sites on each river. Mann-Whitney U tests were performed to determine whether microplastic concentrations were significantly different between any two sites on the same river. A Levene's test was performed to assess the similarity of the variability between sites on each river.

3. Results and Discussion

3.1 Microplastic particles in the River Trent and its tributaries

Throughout the 12 month freshwater sampling campaign microplastic particles were identified at every site, including the most upstream sites. Identified microplastic particles included fragments, films and spherical beads, as well as extruded textile fibres. Extruded fibres include microplastic fibres (e.g. polyester) and regenerated cellulose fibres (e.g. rayon). Limitations of the analytical techniques available to this study, detailed in Stanton et al. (2019), meant that it was not possible to definitively categorise extruded fibres as plastic. It is possible that all of the extruded fibres identified were microplastics, and the data presented here assumes this for clarity and to present a worst case scenario. However, we recognise that as it was beyond the scope of this investigation to characterise extruded fibres this might not be the case.

A total of 178 plastic particles were identified in the freshwater samples collected for this study. 79 particles were identified as extruded textile fibres, and were present in similar concentrations at sites that do and do not receive WWTP effluent (Figure 2). Though WWTPs are widely cited as a source of textile fibres (Napper and Thompson 2016; Ziajahromi et al. 2017), by considering temporal variation, we show here that they do not always inflate textile fibre concentrations.

The remaining 99 plastic fragments included 95 microplastic particles and four plastic particles ≥5 mm, of which FTIR spectra were generated for 96. The use of ATR-FTIR spectroscopy is a common technique for the analysis of particles >500 μm in size (Biginagwa et al. 2016), but can be challenging for smaller particles. Alternative methods of FTIR spectroscopy, including the reflectance FTIR spectroscopy available to this study, can provide spectra for particles too small to handle for FTIR spectroscopy. By characterising a subsample of microplastic particles and extrapolating based on levels of error determined from this subsample, it is possible to infer the composition of microplastic populations within a study (e.g. Dris et al. 2016). Whilst this is a valid approach, we opted to attempt to identify each microplastic particle that was visually preselected using FTIR spectroscopy. However, the quality of reflectance FTIR spectra of particles <500 μm is often poor. The majority of microplastic particles were smaller than 500 μm in their largest dimension (Figure 3), but of the particles analysed, 21 (21%) could be identified by FTIR spectroscopy. Of these 21 particles, 20 were confirmed as plastic particle. Twelve were polyethylene, three were polypropylene, two were polystyrene, two were polyvinyl acetate (PVA), and one was identified as urethane alkyd (UA) (Figure 4). The PVA and UA particles may represent fragments of polymer-based paints. The remaining 77 spectra were too noisy to be identified confidently, but are thought likely to be plastic given the success of the visual identification of particles that could be confidently characterised.

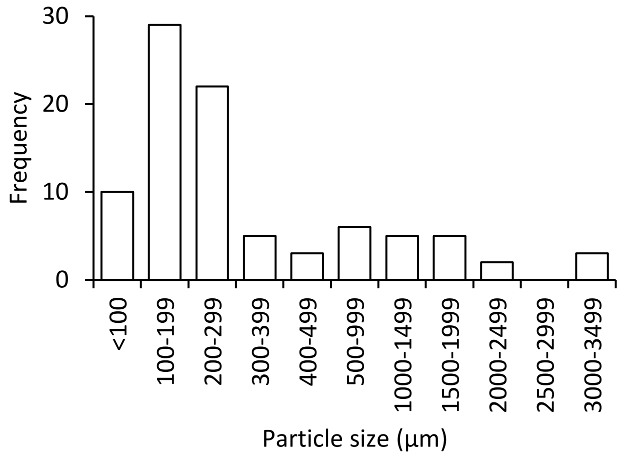

Figure 3: Distribution of non-fibrous microplastic particle sizes across all freshwater samples. 77% of particles had a largest dimension <500 μm.

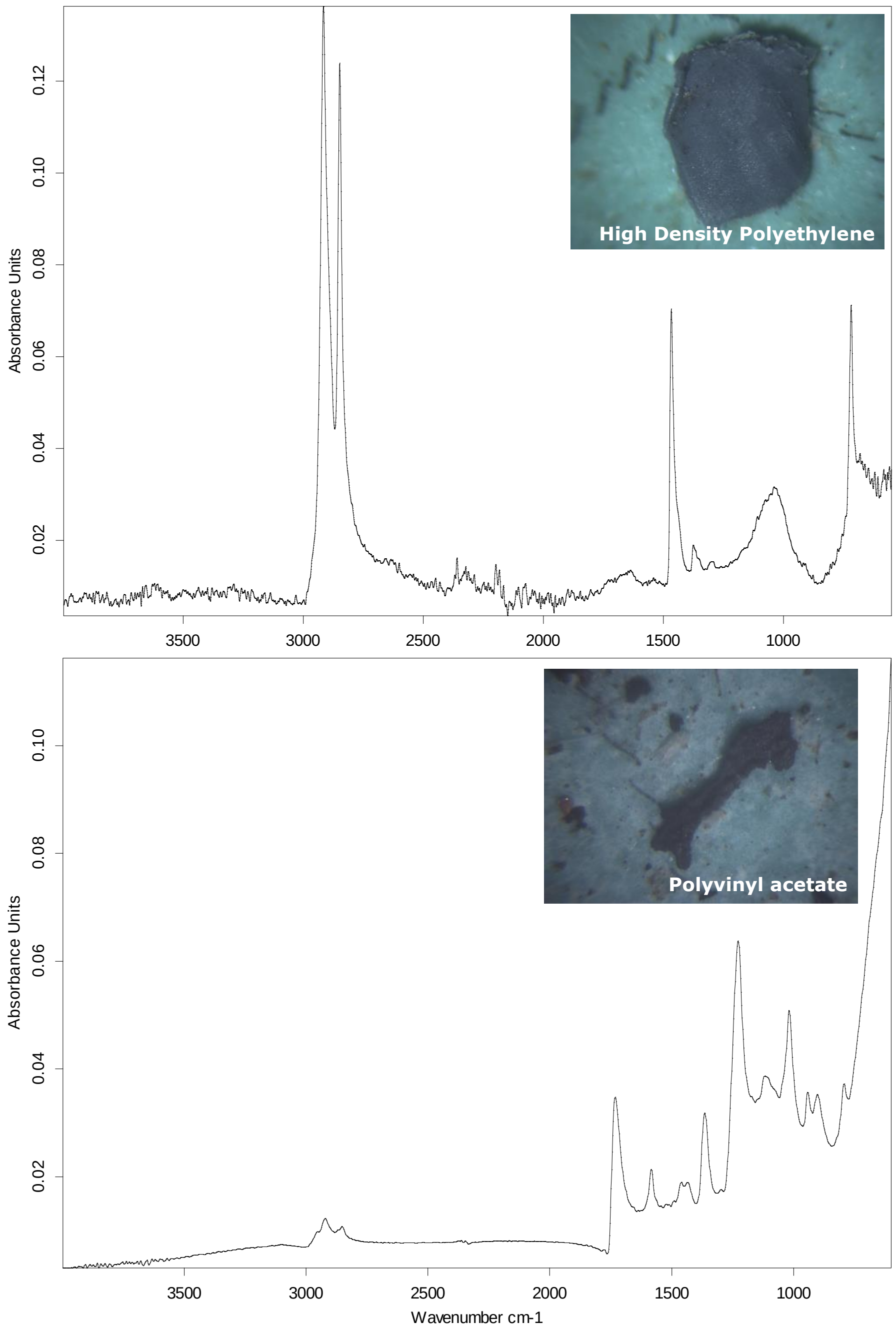

Figure 4: ATR-FTIR spectra of two of the microplastic particles identified during the twelve month sampling campaign.

The incidence of microplastic particles increased in both concentration and variability along the sampled reaches of the rivers Trent and Leen (Figures 5 and S1). The sampled reaches of these rivers do not receive WWTP effluent, but flow through the urban centres of Stoke-on-Trent and Nottingham, respectively.

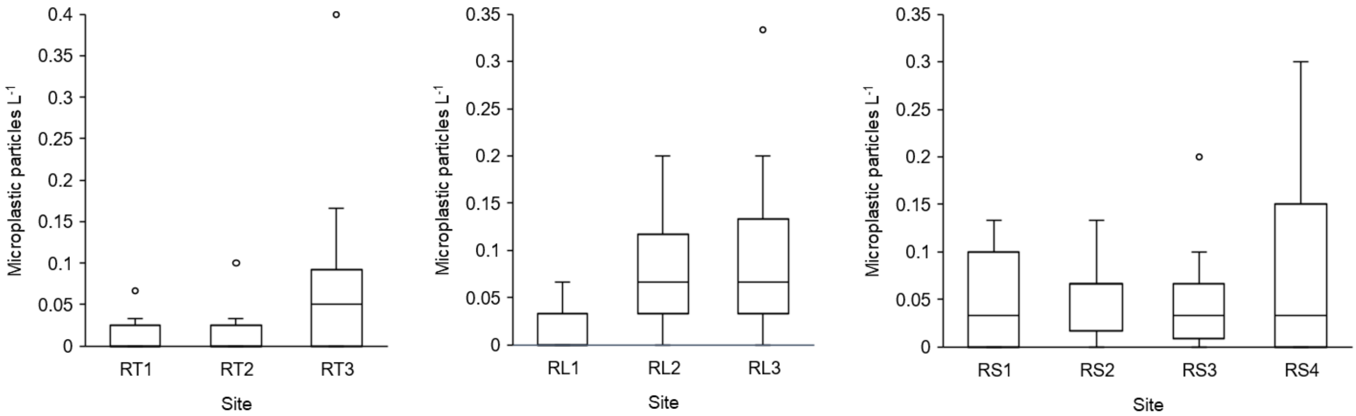

Figure 5: Box and whisker plots illustrating the median and range of microplastic concentrations at each of the ten freshwater sites sampled. Outlying points are more than 1.5 times the interquartile range above the upper quartile.

Across the 12 month sampling campaign, Kruskal-Wallis tests showed that the concentration of microplastic particles between sites on the same river was not significantly different for the River Trent ($p=0.88$) or the River Soar ($p=0.936$). Despite the WWTP input at sites RS2-4, this dataset shows that WWTP effluent does not always significantly increase microplastic and fibre concentrations. However, microplastic concentrations were significantly different between sites on the River Leen ($p=0.015$). Levene's tests showed that the variability of microplastic concentrations between sites on each river was significant for the River Trent ($p=0.027$), the River Leen ($p=0.026$), and the River Soar ($p=0.019$).

Mann-Whitney U tests were carried out to identify significant differences in microplastic concentrations between any two sites on the same river (Table S2). Significant differences were only found between sites RT1 and RT3 ($p=0.045$), RL1 and RL2 ($p=0.007$), and RL1 and RL3 ($p=0.022$). Therefore, the urban areas of Stoke-on-Trent, Nottingham, Leicester and Loughborough (Figure 2) did not significantly increase microplastic concentrations in the rivers that flow through them on the sampling occasions.

However, though not significant, mean microplastic concentrations (±SD) were almost four times greater downstream of Stoke-on-Trent at site RT3 ($\bar{x}$ = 0.075 ±0.11 particles

L^{-1}) than upstream of it at site RT2 ($\bar{x}$ = 0.019 ±0.04 particles L^{-1}). The influence of the Nottingham urban area on the microplastic concentrations of the River Leen was less stark. Mean microplastic concentrations (±SD) were comparable at site RL2 ($\bar{x}$ = 0.076 ±0.06 particles L^{-1}) and RL3 ($\bar{x}$ = 0.083 ±0.10 particles L^{-1}). On the River Leen, the greatest increase in microplastic concentration was observed between site RL1 ($\bar{x}$ = 0.019 ±0.03 particles L^{-1}) and site RL2 (Figure 5). Though located where the River Leen enters the urban area of Nottingham, anthropogenic activity near to site RL2 is extensive, highlighting the immediacy with which plastic debris associated with anthropogenic activity can enter the aquatic system.

Of the three rivers sampled in this study, the River Soar represented the largest system. There was no significant increase in microplastic concentration between any two sites sampled along the River Soar (Table S2). However, comparable microplastic concentrations between sites along the course of a river do not equate to comparable microplastic abundances, with water volume increasing along the river's course.

Many microplastic studies collect samples by trawling a net through surface waters (Rivers et al. 2019). Whilst this enables the sampling of large volumes of water, its application to freshwater systems imposes various limitations on the ability of studies to produce a comprehensive assessment of microplastic pollution within the surveyed catchment. The restrictions associated with coarse mesh apertures have been discussed previously (see Hidalgo-Ruz et al. 2012), however, the use of nets also limits the size of the system that can be sampled. Research that uses common Manta and Neuston nets to collect samples precludes itself from sampling small freshwater systems. The approach taken in the present study enabled the sampling of small, upstream sites to report microplastic pollution from close to the sources of the sampled systems (Figure S2). Moreover, concentrating samples onto a 63 μm sieve enable this work to account for smaller particle sizes than is normally possible using a net, as well as increasing the likelihood of fibre capture. In order to enable comparison between sites, this approach had to be followed at all sample sites, including those large enough that a net could have been used. The low concentration of microplastic particles recorded throughout the sampling campaign is not thought to be an artefact of the sample size, within which natural and extruded textile fibres were consistently reported and documented in Stanton et al. (2019).

3.2 Atmospheric deposition of microplastic particles

Throughout the 12 month atmospheric sampling campaign, microplastic deposition was sporadic and consistently low, with a total of 27 extruded textile fibres and eight microplastic fragments quantified across all four sites (Figure 6). Mean daily deposition (±SD) ranged from 1.14 ±2.4 to 3.16 ±4.9 particles m^{-2} day^{-1}, and the modal value for each site was 0 particles m^{-2} day^{-1}. Natural textile fibres were observed consistently across all sites throughout the sampling campaign (see Stanton et al. 2019). The additional sampling at site D showed little intra-site variation (Figure S3).

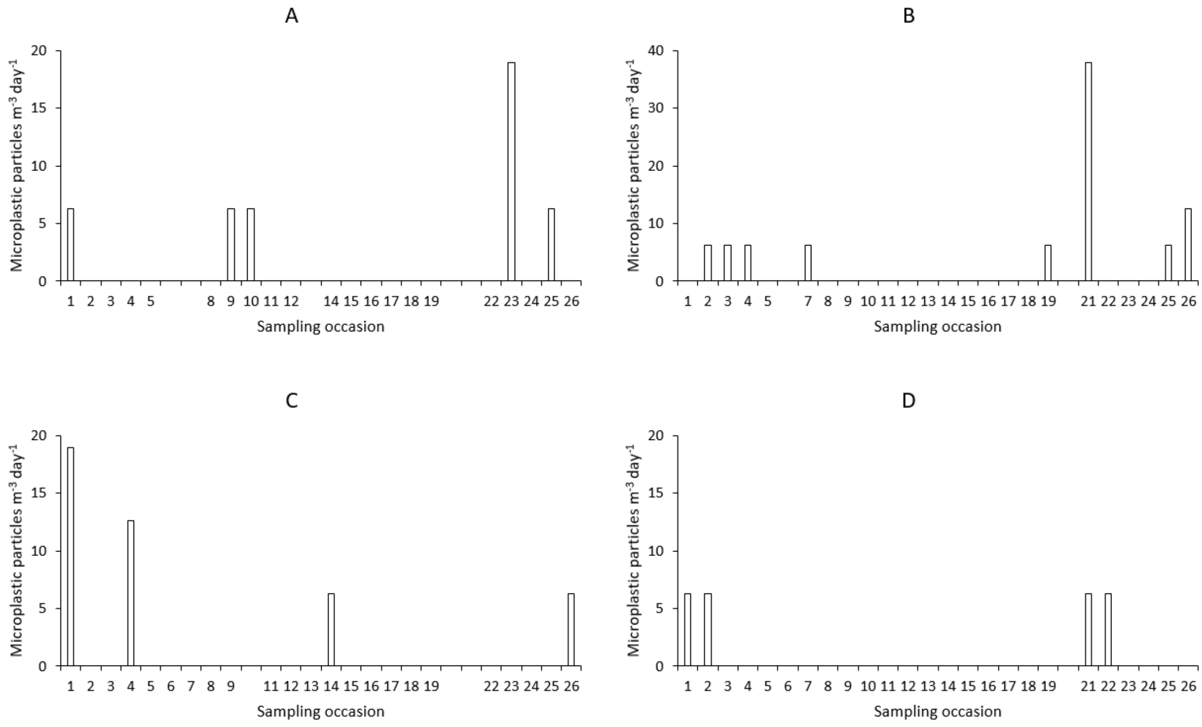

Figure 6: Microplastic deposition across the four atmospheric sampling sites throughout the 12 month sampling campaign

The atmospheric deposition of microplastic particles recorded in the present study is much lower than those reported previously. The sample sites in the present study represent areas of lower population density and urbanisation than those sampled by Dris et al. (2016), Cai et al. (2017), and some of those sampled by Bergmann et al. (2019), which is likely to contribute to the abundance of airborne particles. However, though the surface area of the atmospheric sampling device (0.0113 m^2) was similar to the largest device used by Allen et al. (2019) (0.014 m^2), they reported much higher mean microplastic concentrations (±SD) of 365 ±69 particles m^{-2} day^{-1} at their remote sampling sites. Multiple environmental and methodological factors could have influenced this discrepancy, including sampling height. The sampling reported here was undertaken on rooftops, as opposed to the sampling closer to ground level undertaken by Allen et al. (2019).

Here we show that atmospheric deposition is a source of microplastic particles in both rural and urban reaches of the freshwater system. However, the negligible deposition recorded throughout this sampling campaign indicates that atmospheric deposition it is not a major contributor to microplastic pollution at the sites of deposition sampled.

3.3 Temporal variation of freshwater and atmospheric microplastic particles

Microplastic particles are known to be present in the freshwater system from source to sea (Miller et al. 2017). However, freshwater and atmospheric microplastic concentrations varied considerably throughout the sampling campaign, and were absent from 41% (51 of 123) of samples collected across the 12 month sampling campaign. The modal microplastic concentration was 0 particles L^{-1} at six of the 10 freshwater sites samples (Figure S1).

Though recorded freshwater and atmospheric microplastic concentrations did vary at different points in time, no seasonal variation in microplastic concentration was observed (Figures 6 and S1). On sample occasion five (12th and 13th March 2018) samples were collected during a storm event (Table S3) which saw suspended microplastic concentrations increase at some sites (Figure S1). This increase can be explained by the in wash of microplastic particles via surface runoff (Wagner et al. 2014; Li et al. 2018), and the resuspension of sedimentary microplastic particles within the broader increase of the river's suspended solid loads during such an event (Hurley et al. 2018). However, the influence of this precipitation varied in the freshwater system, with microplastic concentrations at some sites also being present in similar, or lower, concentrations than their site average. Five of the 10 freshwater sites even recorded microplastic concentrations of 0 particles L^{-1} during this event. We therefore postulate that storm events can also dilute freshwater microplastic concentrations.

This temporal variation and inconsistent relationship between particle concentration and flow can lead to considerable misrepresentation of findings when particle fluxes are calculated. Microplastic fluxes were extrapolated at sites RT2, RL3, and RS4, which are located in close proximity to UK National River Flow Archive gauging stations. At site RS4 this flux extrapolation ranged from 0 to 643 000 000 particles depending on the sampling occasion (Table 1). These flux extrapolations are detailed for each sampling occasion throughout the sampling campaign in Table S3.

Table 1: Microplastic flux estimates, presented to three significant figures, at sites in close proximity to UK NRFA gauging stations. Numbers in brackets represent the codes for the NRFA gauging station used. Mean flow for each station is as stated by the NRFA on 31/07/2019, and was used to calculate mean microplastic flux from the mean microplastic concentration quantified for each site in the present study. Maximum microplastic flux was calculated using the mean flow rate for the day of sampling, as detailed in Table S3.

Site	Mean flow (m^3s^{-1})	Mean microplastic flux (particles / day)	Minimum microplastic flux (particles / day)	Maximum microplastic flux (particles / day)
RT2 (28040)	0.624	1 050 000	0	2 550 000
RL3 (28035)	0.684	4 920 000	0	88 400 000
RS4 (28074)	11.729	69 900 000	0	643 000 000

3.4 Implications for our understanding of microplastic pollution

Microplastic particles are ubiquitous in many environmental systems (Rochman, 2018). They are likely to be the most abundant form of plastic debris in the marine environment (Law and Thompson, 2014), with the freshwater system being a major source of marine plastic debris (Lebreton et al. 2017; Schmidt et al. 2017). However, the freshwater samples collected at discrete time points are not representative of the four weeks that separated them and, therefore, this 12 month dataset is not to be interpreted as a representation of the annual variation in microplastic concentrations at the sites sampled. Moreover, whilst microplastic particles are identified at every site, the consistently low concentrations at some sites and the repeated dominance of non-plastic anthropogenic particles in the form of natural fibres raises important questions about the relative risk that microplastics pose across some freshwater and atmospheric systems.

These systems are highly spatially and temporally variable, and by not considering this variability the findings of microplastic research risk being interpreted beyond the spatiotemporal context that they represent. Without such consideration, the subsequent public dissemination of such findings risks distracting attention from more pressing environmental concerns, including those whose harm has a stronger evidence base than that of microplastics.

4. Conclusion

The freshwater system is an important pathway for microplastic pollution to marine and lacustrine environments and it is concerning that microplastics have been found in even the most remote environments (e.g. Bergman et al. 2019). However, whilst the presence of microplastic particles is widespread, their abundance in the environment is harder to quantify. Here we show a clear need to increase temporal resolution of sampling campaigns, and for complementary work to assess the similarity of this variability in sedimentary and biotic matrices. Extrapolation from few samples in space or time, is likely to lead to substantial errors in assessment. This research also raises important questions about sources of microplastics to environments given its observation of plastic particles, including fibres, upstream of both the urban areas and WWTPs that are often thought to represent major sources of such particles. To this end, the findings of this work bring the authors to recommend that future research into the impacts of microplastic pollution generate longer term, high temporal resolution, records of microplastics in the environment, and that they assess risk at environmentally representative concentrations.

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Author contributions

T.S. conducted all field sampling, and all laboratory processing and analysis. W.M. assisted in the FTIR spectroscopy. All authors contributed to the interpretation of the presented data and the writing of the manuscript.

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