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Short Communication

Microplastic abundance in atmospheric deposition within the Metropolitan area of Hamburg, Germany

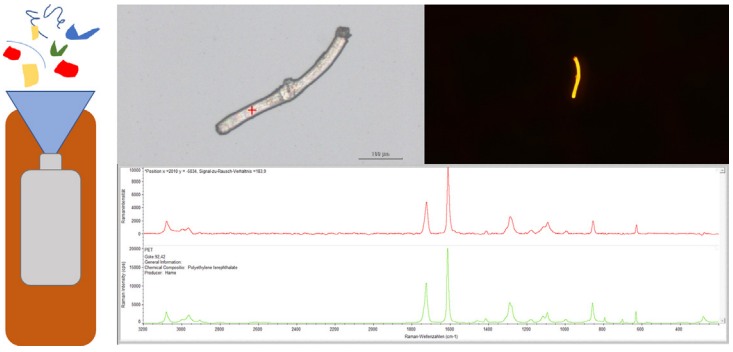
Malin Klein, Elke K. Fischer *

CEN - Center for Earth System Research and Sustainability, University of Hamburg, Bundesstrasse 55, 20146 Hamburg, Germany

HIGHLIGHTS

- Microplastics are ubiquitous in atmospheric deposition within the research area.
- Spatial patterns show high concentrations in rural sites.
- Microplastic particles were predominantly fragments.
- Nile Red staining proved to be highly efficient for small particle sizes.

GRAPHICAL ABSTRACT



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ABSTRACT

Only few studies investigated the input of microplastic particles via the atmosphere, so far. Here, we present results on microplastic concentrations in the atmospheric deposition in the metropolitan region of Hamburg. In total, six investigation sites were equipped with three bulk precipitation samplers each and sampled biweekly over 12 weeks (12/17–03/18). Three sites were located in a rural area south of Hamburg comprising one open field site and two throughfall sites under beech/oak and Douglas fir forest canopy, respectively. Three further sites were selected within the city following a transect from north to south representing urban sites of varying degrees concerning population, traffic and industrial pressures. Particles and fibers were counted under UV light within a photo box and under a fluorescence microscope (Axio Lab A.1, Zeiss). Results show that microplastic particles are ubiquitous at all sites. A median abundance between 136.5 and 512.0 microplastic particles per m^2/day has been found over the sampling period. This equals a mean microplastic abundance of 275 particles/m^2/day μRaman spectroscopy showed that polyethylenes/ethylvinyl acetate copolymers are dominating significantly (48.8 and 22.0%, respectively), 16 particles analyzed (14.6%) were identified as contamination from PE (polyethylene) samplers. In contrast to other studies, fragments were significantly dominating compared to fibers. The spatial distribution comparing the urban sites concentrations followed in the order from high to low: "north" (Henstedt-Ulzburg, low population density, suburb) – "center" (University; large population density) – "south" (Wilhelmsburg, middle population density, port and industrial facilities) with highly varying concentrations within the time series. Surprisingly, the rural sites in the southern part of Hamburg showed highest concentrations (Douglas fir > open field > beech/oak). This finding is most likely a result of factors such as the comb out capacity of the different forest types and/or direct input pathways from the agricultural areas and the nearby highway.

* Corresponding author.
E-mail address: elke.fischer@uni-hamburg.de (E.K. Fischer).

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1. Introduction

Since the beginning of the plastic production in the 1950s synthetic material is worldwide on the rise. With the demand for plastic continuously growing, microplastic accumulation rates in the environment have increased drastically (Rodrigues et al., 2018; Serranti et al., 2018). Microplastics can be found in all ecosystem components (Rodrigues et al., 2018; Cai et al., 2017) and are commonly described as particles smaller than 5 mm (Arthur et al., 2009). A large number of studies have been carried out to detect microplastics in marine and freshwater environments over the last years. These studies have reported a large quantity of microplastics in the waters, ice, biota, as well as in beach and bottom sediments of oceans (e.g. Serranti et al., 2018; Hengstmann et al., 2018; Tamminga et al., 2017), freshwater lakes (e.g. Baldwin et al., 2016; Fischer et al., 2016), rivers (e.g. Schmidt et al., 2017; Ravit et al., 2017; Dris et al., 2015b) and other terrestrial environments (De Souza Machado et al., 2018). Microplastics appear to be extremely mobile, being found from the Arctic to Antarctica (Kanhai et al., 2018; Waller et al., 2017; Chubarenko et al., 2018). Microplastics can enter the environment in different ways. Microplastic inputs to marine and terrestrial environments predominantly are present through the pathway of another ecosystem compartment such as rivers, sediment relocation from shorelines or atmospheric deposition (Chubarenko et al., 2018; Tamminga et al., 2017; Magnusson et al., 2016). Although there are a number of studies examining the role of rivers and shorelines, only few studies have been performed on the question, to which degree atmospheric deposition contributes to microplastics in marine and terrestrial ecosystems (Allen et al., 2019; Yurtsever et al., 2018; Cai et al., 2017; Dris et al., 2016). Atmospheric deposition is understood as the flux of substances from the atmosphere onto the earth's surface. It consists of dry and wet deposition, together making up the total atmospheric fallout (Hupfer et al., 2005). Following Schlesinger and Bernhardt (2013) wet deposition includes all forms of precipitation and the particulate and dissolved particles found in it are separated into rainout and washout. Rainout and washout are directly connected to each other with the rate of particles inversely related to the rate of precipitation. The longer the precipitation event, the smaller the contribution of the washout to the total wet deposition becomes. Dry deposition describes the gravitational sedimentation of solid particles floating in the air and gases, settling on the surface between precipitation events. Dry deposition deposited on any surfaces and thus also in the canopy of forest stands becomes part of the washout during precipitation events. Atmospheric deposition collected in forests is strongly influenced by this so-called comb-out effect.

It is important to investigate the amount of airborne microplastics in wet and dry atmospheric deposition to estimate the total load of microplastics input into the environment. Microplastics transported in atmospheric deposition add to stormwater, which infiltrates the soil and runs into freshwater lakes and the oceans (Magnusson et al., 2016). Microplastics in the air are assumed to pose a health risk for humans. Small plastic particles and fibers could be breathed in and may settle in the lungs of adults and children (Churg and Brauer, 2000; Prata, 2018; Atis et al., 2005). It has been found, that persistent organic pollutants and other harmful compounds accumulate on microplastics, such as trace metals and pathogens (Baldwin et al., 2016). According to Thorpe and Harrison (2008), airborne microplastic particles tend to have an upper size limit of 100 μm whereas particle matter in dust emission even peaks around 10 μm. In contrast, airborne fibers can be significantly longer, reaching a length of 1650 μm in outdoor air and 4850 μm in dust fall (Gasperi et al., 2018). Two studies from China and France show bigger particles to be present in atmospheric deposition as well, leading to the assumption, that local windblown debris plays a major role adding to the number of particles found in deposition bulk samples (Cai et al., 2017; Dris et al., 2015a). Both studies predominantly found fibers in their samples, covering the size range between <200–4850 μm (Cai et al., 2017; Dris et al., 2015a). A third and more extensive study (Allen et al., 2019) has been carried out in the French Pyrenees. The study found mainly fragments ≤50 μm. Due to the need for further research in this area, the present study has been performed. This study investigates the abundance and regional differences of microplastics in atmospheric deposition related to the metropolitan area of Hamburg, Germany.

2. Material and methods

2.1. Study area

The study was conducted within the Hamburg metropolitan area in the north of Germany. Hamburg is a city with a large port situated on the river Elbe which discharges into the North Sea. Its total population is 1.8 million with a total area covering 755 km^2 (Statistisches Amt für Hamburg und Schleswig-Holstein, 2018, see also Table 1). Sample sites were picked in the north, south and center of Hamburg. Six specific sites were selected due to differences in population density and type of development (Fig. 1).

The first sampling site is located in a small residential area in Henstedt-Ulzburg, a small suburb north of Hamburg. Henstedt-Ulzburg lies in the air corridor of the Hamburg Airport. Proceeding to the center of Hamburg, the second sampling site is situated on the campus of the University of Hamburg, a highly populated area (Table 1). The third sampling location is within the district of Wilhelmsburg, which is surrounded by the river Elbe. Wilhelmsburg is an island characterized by harbors, power plants and industries in the northwest. The samples were taken in the southeast of Wilhelmsburg, where a fairly large recreational area with parks and garden plots is located. The fourth, fifth and sixth sampling locations are all situated in the forested area of the Harburg Hills in the south of Hamburg. This area has a low population density. With an interest in the impact of tree cover on the abundance of microplastic, sampling locations in a beech/oak forest and a Douglas fir forest were picked. The third sampling location in the Harburg Hills is tree cover free. It is situated on the border between two agricultural fields.

2.2. Sampling process

Atmospheric deposition samples were collected biweekly during a twelve weeks period from December 2017 to February 2018. For the collection of the samples nine bulk samplers were installed. Additionally, nine samplers from a pre-existing ecosystem research area surveyed by the Institute of Geography of the University of Hamburg were integrated also. Bulk samplers were installed 100 cm above ground level in three parallels per site with a distance of at least 100 cm apart from each other. Each bulk samplers consists of a 150 cm long PVC-pipe, a PE-funnel and a 2 l PE-bottle. The pipe was buried 50 cm deep into the ground to collect atmospheric deposition 100 cm above ground level. The PVC-pipe doubles as a housing to protect the funnel and bottle from UV-radiation and abrasion. Inside the

Table 1
Population density in the districts of the 6 sampling sites.Data: StatistischesAmtfür Hamburg und Schleswig-Holstein 2018.

Sampling site	ID	Location	Population density (inhabitants/km^2)
Henstedt-Ulzburg	H	urban	702
University	U	urban	6121
Wilhelmsburg	W	urban	1531
Beech/oakforest	B	rural	199
Douglas firforest	D	rural	199
Open field	F	rural	199

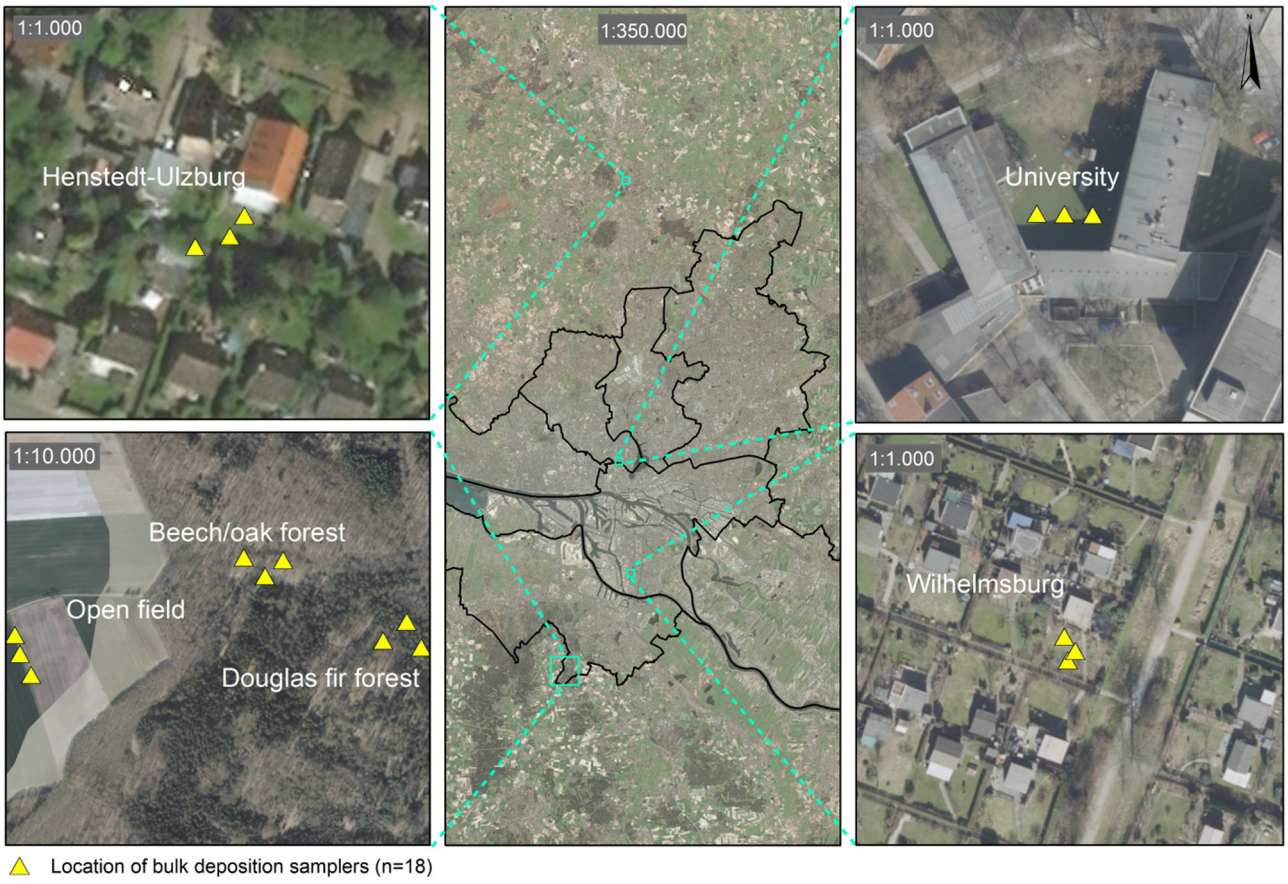

Fig. 1. Overview (center) and detailed views of the sampling area. Coordinate system: ETRS 1989 UTM Zone 32 N. Projection: Transverse Mercator. Vector data: Transparenzportal Hamburg, OpenStreetMap. Base map: Transparenzportal Hamburg, Esri.

PVC-pipe a base was created using wire to keep the PE-bottle in place. On the 2 l PE-bottle a PE-funnel with a diameter of 12 cm was screwed on firmly. The edge of the funnel aligns with the edge of the pipe. Through the funnel the atmospheric deposition could be collected inside the PE-bottle. To consider probable contamination with PE from the flasks, an additional bulk sampler with a procedural blank was run in parallel at the sampling location in Wilhelmsburg. The procedural blank was covered with aluminum foil at all times.

2.3. Laboratory analysis

In the laboratory the bulk volume of each sample was determined using a glass measuring cylinder. 200 ml of each sample were then transferred into a glass beaker and covered with a watch glass. If there was <200 ml deposition in the bulk sampler, the whole sample was transferred into the jar. To eliminate the organic matter, each sample was treated with sodium hypochlorite solution (NaClO, Merck Emplura, 6–14%) in a volume ratio 0.15:1 (30 ml NaClO per 200 ml sample) and left to stand for 24 h under a fume hood (Tamminga et al., 2017; Enders et al., 2016; Collard et al., 2015). Samples were filtered using a glass vacuum filtration device (Sartorius Stedim Biotech) with a 55 mm cellulose filter (413, VWR 516-0812, particle retention 5–13 μm). The glass beakers and the glass vacuum filtration device were thoroughly rinsed with MilliQ water to remove possible adhering microplastic particles. The filters were transferred to glass petri dishes, additionally covered with aluminum foil and left to dry for 24 h at room temperature under a fume hood. Subsequently all filters were stained with the lipophilic dye Nile Red following the method described by Tamminga et al. (2017). Therefore, Nile Red was mixed with Chloroform in the ratio 1 mg/ml. Each filter was dyed with 1 ml (two times 0.5 ml in order to ensure a thorough distribution) of the Nile Red solution (Tamminga et al., 2017; Fischer et al., 2016). To complete the staining process, the filters were covered with watch glasses and left to dry again at room temperature under a fume hood.

The staining method was developed and implemented within various studies on sediments, biota and water samples before and proved to be highly efficient (Fischer, 2019; Hengstmann et al., 2018; Tamminga et al., 2018). During the method development of the staining protocol several solvents were tested against each other on particles of virgin polymers, post-consumer products and biological organic material and their identification was evaluated under the fluorescence microscope. The best results were obtained for the solvent chloroform, even if it (partly) dissolves polystyrene. The disadvantage here is that the presence of PS can be detected on the basis of the melt residue, but the particle size cannot be determined. This disadvantage was accepted due to the better results, especially with regard to the clearer exclusion of biogenic organic particles. The method development and the mentioned tests are published in Tamminga et al., 2017.

2.4. Fluorescence microscopy

Stained filters of the first parallel and the laboratory and procedural blanks were examined under a fluorescence microscope (Zeiss Axio Lab A.1) coupled with a digital camera (Canon EOS 80D, exposure time 1″, ISO 500, ZEISS lens 2,5×/0.06 A-Plan). Microplastics appear bright yellow to white in contrast to an orange to red appearance of organic residues under the microscope after being dyed with Nile Red (Fig. 2). Relevant parts of the filter were photographed with a 4000 × 6000

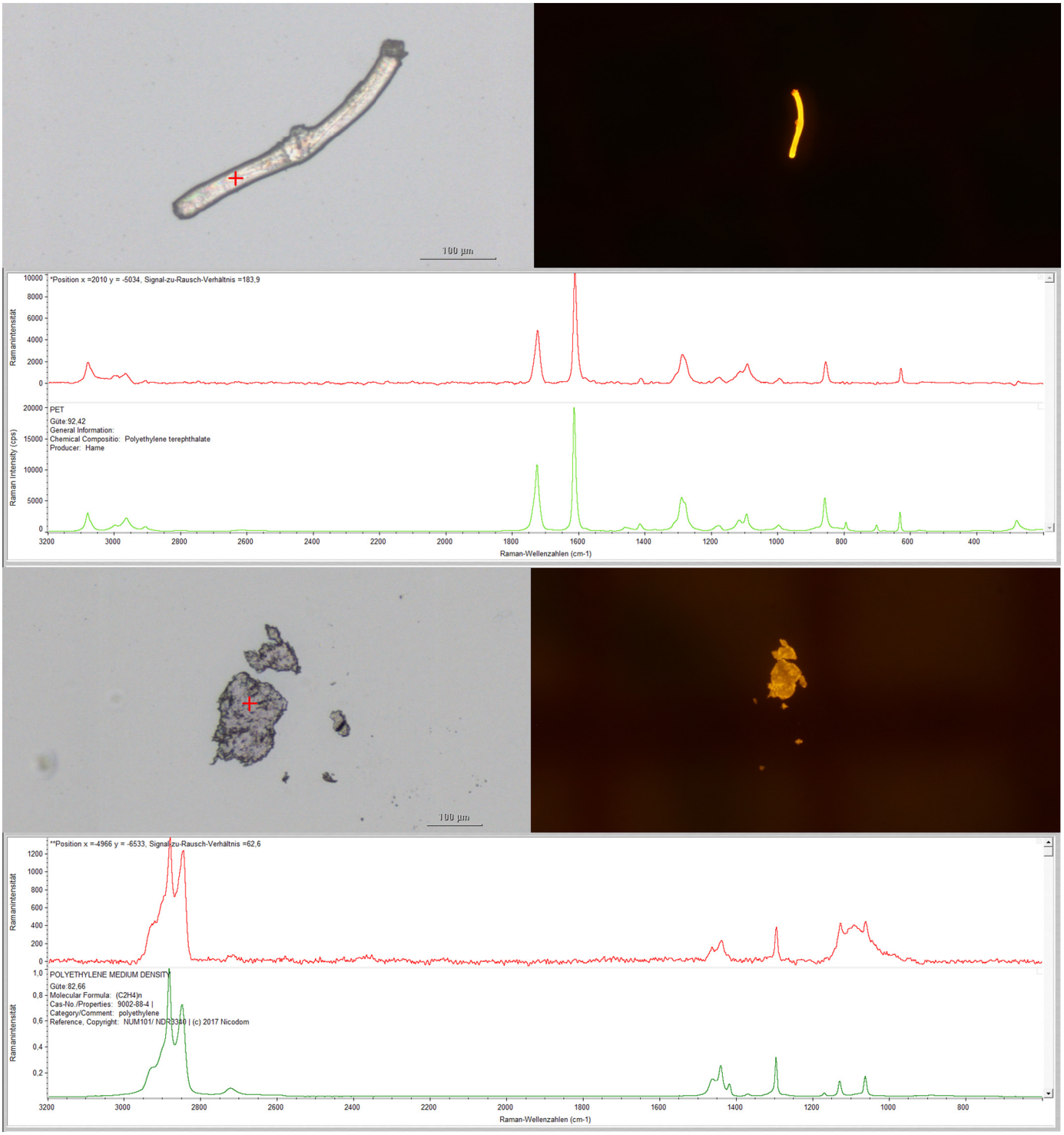

Fig. 2. Detail images taken under the fluorescence microscope (right), under the µRaman (left) and the corresponding µRaman spectrum (top: counting 1 fiber, verified as PET; down: counting 1 fragment, verified as PE).

pixel resolution. For comparison, all filters from each sampling location and the blanks were photographed in a self-built photo box under UV-light (Omnilux UV 18 W G13, 365 nm) with a digital camera (Pentax K-30, resolution 4928 × 3264 pixel, exposure time 2″, ISO 100). All images taken under the fluorescence microscope were transferred into a raster graphics editor (Adobe Photoshop CS5) and the amount of microplastics in each sample was counted. In addition to their fluorescence, particles were identified as microplastics accounting to the criteria described in Chubarenko et al. (2018): 1. no visible cellular or organic structure, 2. fibers are equally thick throughout their entire length and are not tapered at the end, 3. colored particles are homogeneously colored, 4. fibers are not segmented or appear as twisted flat ribbons, 5. particles are not shiny. Due to the chosen method, criteria 3 and 5 are not applicable here. If a particle could not be certainly

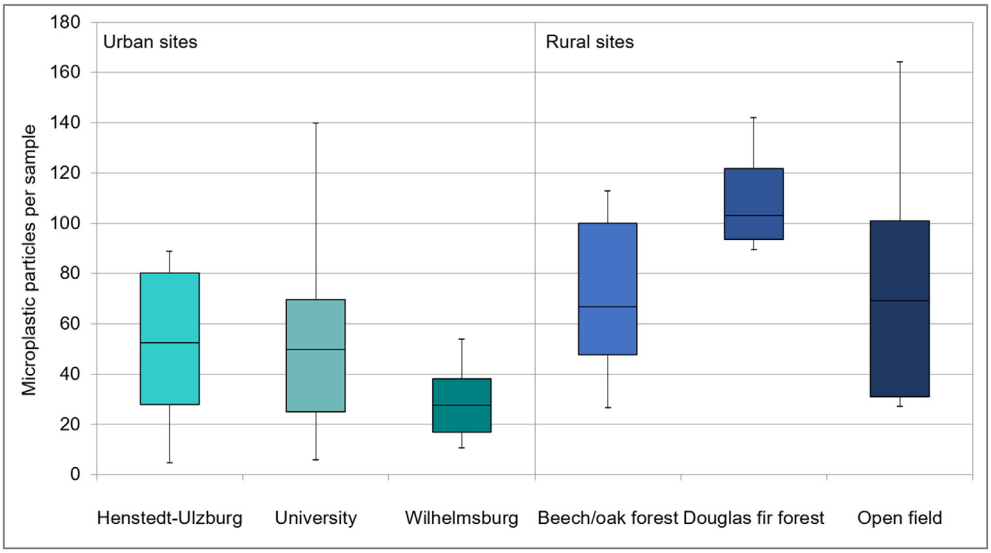

Fig. 3. Abundance of microplastic particles found per sample at each sampling site (12/17–03/18).

identified as a microplastic or the image was not clear enough, it was not classified as microplastic. The counted microplastics were grouped according to their shape into fragments and fibers and all particles were measured and classified according to three size classes sizes: >300 μm, 300–63 μm and < 63 μm.

2.5. Polymer composition

To determine the polymer composition of the identified fragments and fibers, a total number of 53 particles (>50 μm) were equally collected from the filters of one sampling series ($n = 6$ covering all investigation sites; sampling period = 29/12/17–12/01/18) and two filters from blank samples using tweezers and were transferred onto slides. The polymer composition was determined by μRaman spectroscopy (DXR2xi, ThermoFisher Scientific). To exclude the contamination of microplastic particles by the PE-bulk deposition collectors, three particles of the sampler material (new samplers and those already exposed for a longer time) were additionally examined prior and after treatment with NaClO and Nile red and their spectra were compared with the spectra of the particles found in the real samples. The particles from the deposition samples, which had a correspondence of >90% with the deposition collectors, were classified as contamination.

2.6. Background contamination

To determine the amount of background contamination, one laboratory and one procedural blank were processed alongside the samples during each sampling period of 14 days leading to a total number of six laboratory and procedural blanks each. The procedural blank consisted of 300 ml MilliQ water that was poured into an additional bulk sampler at the location Wilhelmsburg. It determines the amount of microplastic potentially being introduced into the sample by the bulk sampler itself and during the whole analysis process. The laboratory blank consists of 200 ml MilliQ water and offers information about the background contamination in the laboratory during digestion, filtration and staining. Measures were taken to keep the working environment and the samples clean. Laboratory coats made out of 100% cotton and Latex gloves were worn at all times. The samples were kept covered between the individual steps and stored under fume hoods. The glass vacuum filtration device, the glass jars, petri dishes and watch glasses were all rinsed with acetone prior to use. To reduce the risk of contamination due to airborne fibers, the humidity in the laboratory was increased using a pressure sprayer (Buerkle) and by wiping down all surfaces. Additionally, an air filter was used (Philips AC3256/10).

3. Results

3.1. Background contamination

The blank samples showed a median contamination of 8.5 (mean 7.3 ± 4.8, Q1 = 2.0) particles. The procedural blanks had a slightly higher contamination rate with a median of 11.0 (mean 11.5 ± 2.8, Q1 = 9.3) particles. The laboratory blanks median-value was 2.0 (mean 3.2 ± 2.3, Q1 = 2.0) particles. The proportion of fibers in the blanks was high, accounting for 51% of the found microplastics as opposed to the field samples in which fibers only made up for 5% of the total microplastics. The results were blank corrected accordingly.

3.2. Particle concentrations

Overall, 108 samples of atmospheric deposition in the Hamburg metropolitan were analyzed and microplastics were present in every sample. A subsample of 36 samples were processed under the fluorescence microscope for further investigation (one sample per sampling location every 14 days over three months) plus six procedural blanks and six laboratory blanks. The amount of found particles ranges from 5 to 164 microplastic particles per sample, with the lowest number observed in Henstedt-Ulzburg and the highest number found in the open field. Considering all samples taken over the three months sampling period, the sampling location Douglas fir forest accounted for the highest median abundance of microplastics with 103.2 microplastic particles per sample (total number 656; mean 109.4 ± SD 19.2; Q1 = 103.2), followed by the open field with a median of 69.2 microplastic particles per sample (total number 459; mean 76.5 ± 49.8; Q1 = 30.9). The third highest contamination was also found in this area, at the sampling site beech/oak forest with a median number of 66.8 microplastic particles per sample (total number 425; mean 70.9 ± 31.6; Q1 = 47.7). At Henstedt-Ulzburg a median number of 52.5 microplastic particles per sample was counted (total number 307; mean 51.2 ± 31.6; Q1 = 27.8) and at the University campus a median number of 49.8 microplastic particles per sample (total number 338; mean 56.4 ± 43.6; Q1 = 24.9). The sampling location Wilhelmsburg

Table 2
Shape and size distribution of the microplastic particles recorded at the six different sampling sites.

Sampling site	Median	Mean ± SD	Shape in %		Size in number of particles		
			Fiber	Fragment	>300 μm	300–63 μm	<63 μm
Henstedt-Ulzburg	60.0	51.2 ± 31.6	7.7	92.3	19	65	267
University	57.0	66.4 ± 43.6	7.6	92.4	25	108	249
Wilhelmsburg	35.0	29.1 ± 14.9	11.4	88.6	16	47	155
Beech/oakforest	74.0	70.9 ± 31.6	3.0	97.0	8	120	341
Douglas firforest	111.0	109.4 ± 19.2	2.9	97.1	23	138	539
Open field	77.0	76.5 ± 49.8	3.6	96.4	16	118	369

accounted for the lowest median with 27.5 microplastic particles per sample (total number 174; mean 29.1 ± 14.9; Q1 = 17.0). The results are shown in Fig. 3 and Table 2.

Median-values of microplastics were calculated as particles/m^2/day (Fig. 4). For the three urban sites the daily fallout is 260.6 particles/m^2/day (Henstedt-Ulzburg), 246.9 particles/m^2/day (University) and 136.5 particles/m^2/day (Wilhelmsburg). The three rural sampling sites in the Harburg Hills account for 331.4 particles/m^2/day in the beech/oak forest, 512.0 particles/m^2/day in the Douglas fir forest and 343.1 particles/m^2/day in the open field. During the sampling period, a median number of 275.0 microplastics/m^2/day was found considering all sampling sites together.

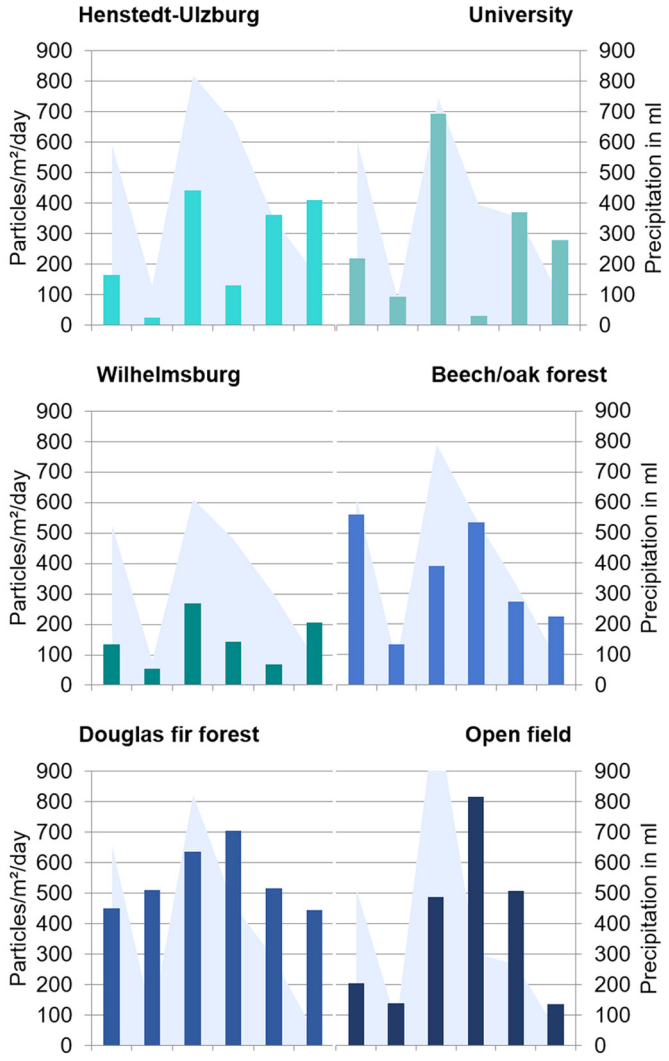

Fig. 4. Time series of the microplastic particles in atmospheric deposition at each of the six sampling sites (particles/m^2/day) compared to the total volume of bulk sample (ml). The columns represent the amount of microplastic.

To determine whether the abundance of microplastics in each sample is directly linked to the amount of precipitation as suggested in Dris et al. (2015a), the microplastic particles and the sample volume were checked for correlation. No strong correlation between the two variables was found at a significance level of 95% (p-value = 0.037). In order to consider a potential evaporation loss from the bulk samplers, the results also were correlated to precipitation data of Hamburg-Fuhlsbüttel (DWD, Deutscher Wetterdienst, 2018) which revealed no significant correlation (p-value = 0.084). Though, a significant correlation was found between changes in the dominant wind direction during the 14-days sampling period and the respective mean abundance of microplastic particles (p-value = 0.002).

3.3. Particle shape and size

The dominant shape of microplastics detected were fragments, making up 95% of the total particle numbers and only 5% comprised fibers. In total, 2625 microplastics have been recorded within this study (2492 fragments and 133 fibers). The majority of fragments belong to the smallest size class <63 μm with 1911 fragments, followed by 300–63 μm (564 fragments) and only 17 fragments larger than 300 μm (5000–300 μm) have been detected. Regarding fibers the size class between 5000 and 300 μm was dominant (90 fibers), 34 fibers belonged to the size class 300–63 μm and 9 were identified in the smallest class (<63 μm).

3.4. Chemical composition

A total number of 53 particles were examined with μRaman spectroscopy, 41 particles of them were assessed to be plastic polymers under fluorescence microscope, 12 particles were assessed to be biological organic material. Only 5 plastic polymer particles were identified as background contamination from the PE bulk sampler by μRaman spectroscopy. All other particles (n = 36) could be identified as polymers with polyethylene (n = 20, 48.8%) and ethylene vinyl acetate copolymer (EVAC) (n = 9, 22.0%) dominating. Only one fragment each was determined to be teflon (PTFE) and polyvinyl acetate (PVA), respectively. All fibers evaluated to be of synthetic origin (n = 4, 9.8%) were determined to be polyethylene terephthalate. In order to cross-check the results derived with staining and fluorescence microscopy 8 particles and 4 fibers that were not identified as plastics additionally were analyzed with μRaman spectroscopy. All fragments proofed to be organic residual material, the fibers were of cellulose origin.

4. Discussion

4.1. Methodology

A severe challenge in quantifying the amount of microplastics on a filter is the distinction between organic matter, other inorganic particles and the targeted microplastics (Tamminga et al., 2017; Erni-Cassola et al., 2017; Shim et al., 2016). In order to effectively stain the particles, the Nile Red staining protocol established by Tamminga et al. (2017) was followed. This method uses chloroform as solvent for Nile Red

due to best recovery results in pretests. However, chloroform has the disadvantage of dissolving PS (polystyrene) when not in its expanded form EPS (expanded polystyrene) (Tamminga et al., 2017). On the other hand, PS makes up only 6.7% of the European plastic demand (PlasticsEurope, 2018) and has been the smallest polymer group found in the atmospheric samples processed in the Dongguan City study performed by Cai et al. (2017) and it can still be detected due to its melt residue.

4.2. Spatial and meteorological influences

Wind-induced motions are important to understand the transport of plastic debris in the atmosphere and near ground level. Therefore, the microplastic data gathered in this study was compared to wind directions and wind speeds measured by the DWD (Deutscher Wetterdienst, 2018) during the sampling period. Days with a high maximum wind speed, categorized as stormy winds or storm (18.3–21.1 m s^{-1}), occurred on 6 days during the total sampling period. The dates of the storm events are conforming to the weeks of the highest abundance of microplastics detected in the atmospheric fallout. The data suggests that wind speed and microplastic abundance in atmospheric deposition are related. A significant correlation was found between the respective dominant wind direction and the number of microplastic particles measured during the same period. The dominating wind direction throughout the sampling period were westerly winds. A rise in the microplastic contamination has been found in weeks with a direction change to southerly winds. Though, this finding has to be regarded critically, since within the study setup it was not possible to consider wind fields at small scale, which play a major role in the wind-induced transportation processes (Loewe et al., 2005; Loewe, 2018). As described, no significant correlation could be determined between the microplastic abundance and precipitation. Similarly, no relation to the temperature profile was evident.

The most polluted sampling sites are those in the Harburg Hills. The main explanation for the higher concentration of microplastics in the Douglas fir forest of the Harburg Hills is the comb-out effect. The comb-out effect describes the ability of plants to filter particles from the dry atmospheric deposition. The load of particles getting combed-out by a tree or plant is defined over the leaf area index. The higher this index, the more particles are filtered out by the plants instead of reaching the ground (Hoelscher et al., 1998). In a precipitation event, the particles get washed off the leaves and add to the number of particles in the bulk samplers. The Douglas fir forest is an evergreen coniferous tree forest and its needles have a higher leaf area index compared to the leaves of the beech tree. The difference is especially noticeable during the winter months, when the beech/oak forest is free of leaves. In this study, the Douglas fir forest has the highest concentration of microplastics, which suggests a relation between the comb-out effect and the behavior of microplastics in atmospheric deposition. To validate this thesis, a comparison test should be performed in the summer months.

The samples from the open field sampling site had the second highest microplastic pollution (459 microplastic particles in total over the 3 months sampling period). A possible reason for the relatively high abundance of microplastic in the samples from this site could be the proximity to the highway interchange Hamburg-Southwest. Traffic related non-exhaust emission such as road dust and abrasion from tires, road paint and road surfaces are known as a source of secondary microplastics (Magnusson et al., 2016). According to Amato et al. (2014), these particles have a very local effect and could be blown off the highway and into the bulk samplers of the open field. Particles from local sources are usually bigger in size and travel only meters to some kilometers with the wind (Hoelscher et al., 1998). Following this argumentation, the reasons for Wilhelmsburg being the sampling site with the lowest abundance of microplastics could be similar. Despite Wilhelmsburg being closer to the city center and the port of Hamburg, it has a sheltered position in the Elbe river. The river could reduce the impact of wind transported particles.

A striking value with highest number of microplastics detected in the residential areas occurred in the sampling period following New Year's Eve. The results show that a connection between the high input of particulate matter into the atmosphere due to fireworks (Tandon et al., 2008) and the microplastics in atmospheric fallout is possible.

4.3. Comparison to other studies

The studies investigating microplastic in atmospheric deposition by Cai et al. (2017) and Dris et al. (2015a) have found microplastics in every sample as well. The study in Dongguan City, China by Cai et al. (2017) has found a mean microplastic abundance of 36 particles/m^2/day, the one in Paris, France, 118 particles/m^2/day (Dris et al., 2015a) and the French Pyrenees one 365 particles/m^2/day (Allen et al., 2019). In Hamburg the second highest amount of microplastics has been detected compared to the other studies, more precisely 275 particles/m^2/day. Whilst the dominant shape of microplastics identified in this study was fragments, fibers dominated the two studies from Paris and Dongguan City (Dris et al., 2015a; Cai et al., 2017). These differences are most probably due to different methodology comprising the detection of particles at a different cut-off size (Cai et al., 2017: 200 μm; Dris et al., 2015a: 100 μm, this study: 63 μm), installation heights of samplers (Cai et al., 2017 15 m height, Dris et al., 2015a "on roof top", this study 1 m above ground level) and detection methodology. The study by Allen et al. (2019) and this study both predominantly found fragments (88% and 95%, respectively) and both studies show a rise in the particle number with decreasing particle size. Both studies cover the time from December 2017 to February 2018 and were performed using common bulk deposition samplers with a similar opening size (this study: Ø 12 cm, Allen et al., 2019: Ø 13.5 and 20 cm). Concerning the detection methodology, the studies from Paris and China applied visual inspection with subsequent FTIR verification of a subset of samples and both studies found an overestimation of fibers solely based on visual inspection. Only 23 and 29% of fibers, respectively, that were identified as plastics through visual identification proved to be synthetic. Here, the staining protocol and pre-identification via fluorescence microscopy proved as a valuable decision tool. This study and the French Pyrenees study used μRaman for verification (Allen et al., 2019). The Paris study suggested a correlation between microplastic abundance and precipitation but could not find any significance (Dris et al., 2015a). This is concurring with the findings of Cai et al. (2017) and Allen et al. (2019). Even so a relation between these two variables with distinct rho-values occurring at 4 of the sample sites seems to be noticeable, no significance could be determined in this study either (Spearman's rank correlation). Allen et al. (2019) suggests an important link between microplastic deposition and wind speed and wind direction which seems to be visible in this study as well.

5. Conclusion

Microplastic is vastly present in the atmospheric deposition of the Hamburg metropolitan area. A total number of 2625 microplastic particles have been found in the sampling area, resulting in a median abundance of 275 microplastics/m^2/day. The dominant shape of microplastics were fragments, making up 95% of the total particle amount.

The use of fluorescence microscopy allowed for a cost efficient and fast way to detect microplastics and distinguish them from biological organic material.

The samples from the six different samplings sites varied in their microplastic contamination, with median concentration spans ranging from 136.5 to 512.0 microplastics/m^2/day. All six sites under investigation have different population densities and levels of forestation and

infrastructure, suggesting an influence of dust emissions and the comb-out effect of e.g. forest canopy on microplastic contamination.

A link has been made to the wind speed and storm events but no significant correlations to precipitation amount could be detected.

Further research is needed to determine the influence of wind speed and directions, precipitation amount and intensities on the spatial distribution and the amount of microplastic in atmospheric deposition. Within a next project phase these factors amongst others will be investigated in more detail.

In general the atmospheric deposition input pathway should receive increased scientific attention. Especially given the fact, that the interface between atmosphere and marine and freshwater water bodies and terrestrial surfaces is ubiquitous and thus there is a large area for potential microplastic input.

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