Assessing microplastic characteristics in bottled drinking water and air deposition samples using laser direct infrared imaging

Jenny Nizamali, Svenja M. Mintenig, Albert A. Koelmans

PII: S0304-3894(22)01736-8
DOI: https://doi.org/10.1016/j.jhazmat.2022.129942
Reference: HAZMAT129942

To appear in: Journal of Hazardous Materials

Received date: 21 June 2022
Revised date: 2 September 2022
Accepted date: 5 September 2022

Please cite this article as: Jenny Nizamali, Svenja M. Mintenig and Albert A. Koelmans, Assessing microplastic characteristics in bottled drinking water and air deposition samples using laser direct infrared imaging, Journal of Hazardous Materials, (2022) doi:https://doi.org/10.1016/j.jhazmat.2022.129942

This is a PDF file of an article that has undergone enhancements after acceptance, such as the addition of a cover page and metadata, and formatting for readability, but it is not yet the definitive version of record. This version will undergo additional copyediting, typesetting and review before it is published in its final form, but we are providing this version to give early visibility of the article. Please note that, during the production process, errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.

© 2022 Published by Elsevier.
Assessing microplastic characteristics in bottled drinking water and air deposition samples using laser direct infrared imaging

Jenny Nizamali§, Svenja M. Mintenig§, Albert A. Koelmans§,*

§ Aquatic Ecology and Water Quality Management Group, Wageningen University, P.O. Box 47, 6700, DD, Wageningen, the Netherlands

Corresponding author: bart.koelmans@wur.nl

Abstract

There is an urgent need for data on microplastics (MPs) in uptake media relevant to humans. Here we apply Laser Direct Infra-Red (LDIR) spectroscopy for the rapid analysis of MP>10 µm in clean matrices such as bottled drinking water and (deposited) air samples, using strict QA/QC criteria. Nine water bottles of three different brands were found to contain on average 96 particles/L (range 7 to 364, median 44 particles/L) with polymer abundances in the order polyethylene terephthalate and/or polyurethane > polyamide > polyvinylchloride. Further, for laboratory and household rooms, MP deposition rates were measured, ranging from 0 to 573 particles m⁻²h⁻¹. For household deposition samples, these rates translated to 7 MP (range 0 –
16) consumed in an average meal. Polyethylene terephthalate and/or polyurethane > polyamide > polypropylene were the most abundant polymers. We find a statistically significant positive relationship between the total surface area of textile per unit of room volume (m²/m³) and the deposition of MP. Power law distributions for particle width, height, area, volume and mass were constructed for both sample types, with slopes ranging from 1.9 to 3.8. LDIR appears to be a relatively fast MP measurement method, with MP recovery during sample preparation as a potential improvement point.

1. Introduction

Microplastic particles (MPs; size 1 – 5000 µm) are all around us, so people are constantly exposed to them (Noventa et al., 2021; Wright & Kelly, 2017). MPs have been found across oceans, estuaries, rivers and lakes, in the soil and in the atmosphere (Akdogan & Guven, 2019; Enyoh et al., 2019; Wright et al., 2021) and recently also in food and beverages (Zhang et al., 2020).

The hypothesis is that the absorption of MP by the human body can lead to health effects (Coffin et al, 2022). For example, nanoscale MP can be absorbed in the gut and the fallouts from airborne MPs can be inhaled. MPs with diameters (D) < 10 µm and ≤ 2.5 µm cause the highest respiratory and cardiovascular hazard. MPs with D ≤ 10 µm can potentially reach and settle in the intrathoracic regions of the respiratory system, while MP with D ≤ 2.5 µm can even reach the alveolars of the lung (US EPA, 2019). Furthermore, MP of a few microns or less can be taken up directly by cells in the lungs or intestines. Thus, particle size and shape
play an important role regarding the bioavailability of MP (Cox et al., 2019; Vianello et al., 2019; Kooi et al., 2021). MP characteristics such as volume, area, aspect ratio and surface chemistry are also relevant with respect to actual toxicological mechanisms of effect (Kooi et al., 2021). For instance, MP surface area, chemical composition and release of oxidative species are factors triggering oxidative stress and cytotoxicity (Valavanidis et al., 2013; Schmid & Stoeger, 2016; Riediker et al., 2019). To define toxicologically relevant dose metrics (TRM) it is thus necessary to establish these characteristics of MP in food, air and drinking water. It has recently been described how this can be done by capturing the multidimensionality of MP features through probability density functions (PDFs) (Koelmans et al., 2020). These functions allow to extrapolate to the aforementioned smallest (< 10 µm) particle size classes, and to estimate their TRMs (Mohamed Nor, 2021; Kooi et al., 2021).

Airborne plastic fallouts are an important source of MP pollution that can be taken in by humans, e.g. as deposited MP during a meal (Catarino et al., 2018; Schwabl et al., 2019). Synthetic clothing is pointed out as an important source of airborne MPs (Chen et al., 2020; Prata, 2018). Next to this, sources of airborne MPs in the indoor environment may include plastic fibers and fragments from interior furniture (Prata, 2018). Therefore, it can be assumed that the MP concentration in air deposition samples is positively correlated with the total surface area (m²) of synthetic fabrics (carpets, curtains, upholstered furniture) per unit volume in a room (m³), e.g. expressed as fiber emitting fabric (FEF) density (m²/m³). This assumes that the fabric or textile surface is a source of MP, releasing MPs into the chamber volume. Thus, it is assumed that a large room with few textiles, such as a laboratory, has a low FEF value and MP deposition, while a small living room with a higher fabric surface area, i.e. carpets and curtains made of synthetic materials, would have a higher MP concentration.
In addition to inhalation, MP uptake can occur via ingestion (Thompson et al., 2009; WHO, 2019; Noventa et al., 2021). Ingestion can occur through consumption, for example when MPs are present in the food chain, but also through contamination of food and beverages during production or packaging (Cox et al., 2019; Schwabl et al., 2019). It has been shown that plastic bottles and caps can be an important source for MP in bottled water (Schymanski et al., 2018). It has also been found that friction between cap and bottleneck of plastic bottles may increase the number of MPs in a plastic water bottle (Winkler et al., 2019).

Although much research has been done on MP in the environment, knowledge about MPs in components of the human diet and in the atmosphere is still limited (Enyoh et al., 2019; Noventa et al., 2021). Studies typically use different methods, making the results difficult to compare (Kirstein et al., 2020; Koelmans et al., 2019; Mason et al., 2018; Winkler et al., 2019; Schymanski et al., 2018). In addition, Quality Assurance and Quality Control (QA/QC) requirements are important to identify MPs in environmental samples in the best possible way (Primpke et al., 2020). While crucial, they lead to lengthy procedures, in part due to slow data processing when characterizing all particles in a sample when using IR imaging. Recently, Laser Direct Imaging (LDIR) has been introduced as a relatively rapid way to analyse microplastic particle in environmental samples (Scircle et al., 2020; Cheng et al., 2021). LDIR spectroscopy uses a quantum cascade laser (QCL) to better focus IR light on the centre of MP particles in a sample, but its suitability for obtaining PDFs in media relevant to human exposure has not yet been tested.

With this study, we aim to for the first time apply LDIR spectroscopy to characterize concentrations, particle characteristics and TRMs for two sample types relevant to human exposure to MP: air deposition (particle fallout) samples and bottled drinking water samples. Secondary aims were to assess links between air deposition and room characteristics, such as abundance of fabric in a room, and to discuss human exposure via these sources. The study
design included 24 deposition samples taken in eight rooms, and 9 drinking water samples from three brands with previously applied cap friction to increase realism. Characterization was done for number and mass concentration, polymer type, length, width, area, volume and mass by means of PDFs constructed for all particles per sample type pooled. The latter averages the properties and is more representative for the heterogeneity of particles to which a human population is exposed over a longer period of time. To assure quality, this research followed recent QA/QC recommendations by Wright et al. (2021) and Koelmans et al. (2019).

2. Materials and Methods

2.1 Deposition samples

Indoor MP fallout (MP deposition) from six households in the cities Wageningen, Amerongen, Zetten and Utrecht (The Netherlands), and from two general laboratories in Wageningen University and Research were sampled between March 7th and 21st, 2021.

Each household received three cleaned and sealed glass Petri dishes (height 15 mm, diameter 100 mm) and a form to collect information on the households sampled. The Petri dishes were placed on the dinner table and exposed for 10 hours during the day to the indoor air. Hereafter, the dishes were capped, wrapped in aluminum foil and brought back to the laboratory for analysis (total n=18). Regarding the two laboratories: three deposition samples were taken in the instrument lab with the LDIR (lab#01), where the particle analysis was performed. Three other samples were taken in the general laboratory (lab#02), where samples were being prepared (total n=6). However, one replicate of lab#02 was considered invalid because of an error during focusing and scanning of the slide carrying the sample. Earlier, Catarino et al. (2018), took deposition samples using sticky-tape in the Petri dishes for a
reduced fibre loss and to increase colour contrast to facilitate visual particle enumeration. However, because we were interested in the net deposition after a much longer collection time (10 hours, versus 20 to 40 minutes in the study of Catarino et al. (2018)), and because particles were assessed spectroscopically using LDIR Imaging analysis, this was considered unnecessary.

All deposition samples were prepared in a laminar flow cabinet. In each Petri dish, 96 % ethanol (filtered 0.2 µm) was added with a glass Pasteur Capillary Pipette (230 mm) until the ethanol was completely spread on the surface of the dish. The dish was then gently tilted, and the ethanol dispersion was pipetted on a glass slide (7.5x2.5 cm, MirrIR, Kevley Technologies) to be analyzed with LDIR. Eventually, possibly remaining particles in the pipette were washed out with additional pure ethanol. Each glass slide was divided in two parts using a marker, each half containing one individual sample which required approximately 2-2.5 hours to be analyzed (depending on the number of particles in the sample). This way, preparation and analysis of the household, laboratory and blank deposition samples (n=27) approximately took 15 days.

2.2 Bottled water samples

Bottles with mineral water from three different brands (bottle volume 500 ml) were analyzed. This was done for each brand in triplicate (total n=9). Cap friction, i.e. the friction associated with opening and closing the cap of the bottle, was applied per water bottle for 15 times. Thereafter, the water was vacuum filtered over silver membrane filters (25 mm with a pore size of 3 µm, SPI supplies, Pennsylvania, US). Subsequently, water filters containing the MPs were held with a tweezer above a watch-glass, while MP on the filter surface were washed off with a Pasteur pipette, containing 96 % filtered ethanol. Following the procedure of the air deposition samples, the solution in the watch-glass was transferred with a Pasteur
pipette to half a Kevley glass slide for LDIR analysis. The preparation and analysis of the water bottle samples and controls (total n=12) took approximately 9 days.

2.3 Quality assurance and quality control procedures

To minimize contamination during the sample preparations, cotton laboratory coats with sleeves were worn, along with particle free nitrile gloves. Before every work step, the gloved hands were washed and rinsed with MilliQ water. Lab surfaces were wiped with 70 % ethanol and the needed equipment was rinsed three times with MilliQ before usage. All steps of the sample preparation were done in a laminar flow cabinet (Angelantoni LifeScience, STERIL POLARIS, Class ISO 3), wiped with 70 % ethanol before every use.

Negative controls per sample type (air and water) were carried out in parallel to actual samples to quantify and characterize background contamination in the samples. All sample handling was the same as for actual samples. For deposition samples, three Petri dishes freshly rinsed with 96 % ethanol (n=3) were treated. For water analysis, also three negative controls were handled, each with 500ml MilliQ water. To correct for background contamination, actual samples were corrected for polymer specific average MP numbers found in the blank samples. Polydisperse green fluorescent polyethylene microspheres (Product ID UVPMS-BG, 10 - 180 µm, lot number 130124-12, d=1.003 g/cm³, Cospheric, California, US) were used to determine particle recovery rates. Recovery rates were based on particle counts using pictures taken with a stereo microscope before and after the sample preparation. For the deposition sampling procedure, average recovery was 58.2 ± 8.7 % (n=5), and for the water bottle sampling procedure, average recovery was 59.3 ± 7.6 % (n=5) (Supporting Information, Table S1). As the LDIR is currently not able to analyse MP on filters and given the thus required more complex particle transfer procedure, this was deemed allowable. The results were corrected for recoveries. At a later stage, adjustments were made
to the sample transfer, and we report the results here as a perspective on the method. The inner and outer walls of the Pasteur pipette used to transfer the sample material were rinsed three times using filtered ethanol (96 %) from another pipette. This allowed us to increase the MP recovery to 79.3 ± 3.1 % (n=6) (Table S1).

2.4 Particle imaging and polymer identification

Polymer identity of water bottles and lids were characterized using ATR-FTIR spectroscopy (Agilent, Cary 630). Microplastic particle imaging and polymer identification were performed using an Agilent 8700 LDIR Chemical Imaging System. This is an infrared imaging microscope consisting of a Quantum Cascade Laser (QCL) combined with a single-point mercury cadmium telluride (MCT) detector and rapid scanning optics. Before each analysis, an IR scan at 1799 cm\(^{-1}\) of selected sample area on the kevley glass slide (75x25 mm, Kevley Technologies, Ohio, US) was performed to locate particles and measure their dimensions using a high-magnification visible-light camera. The instrument then targets the identified particles, collects a mid-IR range (1800 cm\(^{-1}\) to 975 cm\(^{-1}\)) spectrum for each particle, conducts a library search and provides a match for each identified particle. In this study the default particle sensitivity was used (medium) to locate particles, while the particle identification diameter range was extended to 10-5000 µm.

Given the reliability of MPs detected by the LDIR system, Wright et al. (2021) recommended a hit quality index (HQI) > 0.7 for (near) infrared spectroscopy methods, which is therefore applied to microplastic particles in this study. Only when reporting the total of detected microparticles (i.e. not necessarily polymer-based) per sample, all particles were included. Next to the particles’ chemical identity, their 2-dimensional shape (length, width and area) could be exported after completion of each measurement. The LDIR 8700 system was generally suitable for analyzing MP particles, although there were occasional artifacts typical
of 2D IR imaging. In particular, focus problems with particles of unequal height, e.g. of fibers, caused the software to detect one fiber as different particles, while 2 or more particles aggregated were counted as one particle, and got thus also only one measuring point assigned.

2.5 Data analysis

Spearman’s rank correlation tests were applied to interpret data statistically. Power law distribution parameters were estimated using a maximum likelihood estimation (MLE) method (Clauset et al., 2009; Newman, 2005), and plotted based on Kooi et al., 2021.

The source strength of Fiber Emitting Fabric (SS$_{FEF}$) per household was calculated as:

\[
SS_{FEF} = \frac{A_{FEF}}{V_r} \times 100
\]  
(Eq. 1)

Where $A_{FEF}$ is the total surface area of FEF in the room (m$^2$) and $V_r$ is the volume of the room (m$^3$).

To calculate MP number concentration, per polymer, the mean number of MPs found in the blanks was subtracted from the number of MPs of the same polymer in the collected deposition and water samples. Subsequently, these numbers were corrected for the determined recovery rates, resulting in $N_s$ (Eq. 2).

Microplastic deposition rates per meal ($N_{meal}$; average number of deposited particles m$^{-2}$ h$^{-1}$ per sample) were calculated as:

\[
N_{meal} = \frac{N_s}{Axt}
\]  
(Eq. 2)

Here, $A$ is the surface area of the Petri dish ($7.85 \times 10^3$ m$^2$) and $t$ the exposure time (10 h).

The amount of deposited particles consumed per meal was calculated following Catarino et al. (2018), assuming a duration of 20 minutes to finish a meal from an average dinner plate with a diameter (D) of 25 cm.
An estimated daily exposure ($N_{daily}$) was calculated as well, taking into account a breakfast bowl ($D_{bowl} = 15$ cm), lunch plate and dinner plate ($D_{plate} = 25$ cm) and 2x coffee, 2x tea and 2x glasses of beverage ($D_{glass} = 8$ cm), also assuming a duration of 20 minutes each.

3. Results and Discussion

3.1 Evaluation of applied methods

This is one of the first studies applying LDIR spectroscopy for the analysis of MP in air deposition and drinking water samples (Liu et al. 2022; Bäuerlein et al., 2022). For the rather clean drinking water and air deposition samples it was ideal as not the whole slides, but previously identified particles were targeted for analysis. Within 2-2.5 h measurements and analyses were completed. Until now, however, it is not yet possible to analyse MP concentrated on a filter which required pipetting the sample on the glass slide. Recovery rates of approximately 60 % were deemed acceptable.

Subsequent adjustments to the sample transfer, i.e. increasing the frequency of rinsing from one to three times, increased MP recovery to $79.3 \pm 3.1\%$ which improves generated results. In contrast to commonly applied FTIR spectroscopy, the LDIR works with a limited wavenumber range between $1800-975$ cm$^{-1}$. We here found that this was not sufficient to differentiate between PET and PU, and thus clustered these polymer types.

3.2 Microplastic deposition in laboratory and household rooms

3.2.1 Particle counts and polymer composition

Raw particle counts, identified materials and respective HQI values are provided as Supporting Information (Table S2 and S3). An HQI $> 0.7$ was required to accept the identification and differentiation into plastic and non-plastic particles. After doing so, total
Particle counts were significantly correlated with MP counts ($p=2.41 \times 10^{-9}$; Spearman Rank correlation, Table S2). Within linear regression plots (Supporting Information, Fig. 1) slopes indicate that around 15% of all particles found were also identified having a synthetic origin.

Fourteen polymer types were detected in the indoor deposition samples (Figure S2). The most abundant were polyethylene terephthalate (PET) and/or polyurethane (PU) in the households, which we found to be indistinguishable during the analysis of drinking water samples. Comparing polymer types found in households (n=18) versus those found in laboratory samples (n=5) revealed considerable differences. The three most commonly found polymers in household samples were PET and/or PU > Polyamide (PA) > Polypropylene (PP), whereas the ones from the laboratories were PA > Chlorinated PE (PE-C) > Polyoxymethylene (POM). Eight out of the fourteen polymer types were found in samples from the laboratories (Figure 1). The average number of polymers found in the blank samples are listed in SI, Appendix A.

Since our study was not designed to precisely identify the sources of the polymer particles, we can only speculate about the observed differences in polymer abundances. In domestic settings, we found a high prevalence of PET and PU, which are common plastics used in everyday life, including for carpets, clothing, bottles and packaging. We found a high prevalence of PA in both domestic and laboratory settings, which is also to be expected due to the application of this polymer in clothing and other textiles (eg carpets). The differences in relative presence between the two or three most dominant polymers were too small to allow meaningful further interpretation. We found an equally high prevalence of POM in laboratory and household rooms (9.9% vs. 8.2%), a polymer most commonly used in electronic devices and/or electrical wiring/insulation.

3.2.2 Deposition fluxes
At first, the amount of deposited MPs per m$^2$ per hour was calculated for each household (Eq 2). The microplastic deposition rate ranged widely, from 0 to 573 particles×m$^{-2}$×h$^{-1}$ with an average of 256 ± 253 particles×m$^{-2}$×h$^{-1}$ (Table S4). We calculated the uptake of MP from fall out on a 25 cm plate during a 20 min meal to be 7 (range 0 - 16) particles per meal (Table S5). A similar calculation was done in the study of Catarino et al. (2018), where 114 particles were calculated.

### 3.2.3 Source strength of Fiber Emitting Fabric (SS$_{FEF}$)

Data of fiber emitting fabric (FEF) was obtained from the six households and two laboratories where deposition samples were taken, to identify the relationship between the number of deposited MPs and the FEF surface (total n=8). For the laboratories it was estimated that an average of three people were present during the sample collection, each with a lab coat with an average surface of 0.5 m$^2$. The detected MP fall out ranged from 0 to 45 particles/m$^3$ (Table S6), indicating the differences in interior designs in laboratory and domestic rooms. The Spearman’s rank correlation (Rs) for household datapoints was 0.83 together with a p-value of 0.042, indicating a significant correlation between the abundance of fabric surface per unit of volume in a room and MP fall out (Table S6). Including the laboratory data, the correlation coefficient decreased to 0.66 (p-value of 0.076), indicating a difference in deposition characteristics in laboratories as compared to household rooms. As expected, the correlation for household rooms can be considered strong evidence that fabric from carpets and curtains is an important source of indoor particles, and thus also MP. More specifically, it supports the concept behind Eq. 1, implying that the surface area of fabric is an appropriate metric for source strength, whereas (MP) particles are subsequently diluted in the receiving air volume in a room.
Interior material and people’s clothing are thus important sources for MP in households, while the latter is not certain for laboratories. This also means that the difference between the deposition flux calculated from the current data and that reported by Catarino et al (2018), in addition to method differences, could probably also be influenced by such differences in fiber emissions.

Finally, while FEF was detected as an explanatory variable, a variety of additional factors apparently did not affect the correlation's significance, but probably contributed to the residual error. For instance, differences in turbulence, number of people in a room, type of garment used, variability in clothes being covered, or vacuum cleaner use (Neda et al., 2022).

3.3 Microplastic particles in three brands of bottled drinking water.

The polymer type of water bottle and lid of three water brands were identified using ATR-FTIR spectroscopy. All three bottle types were made of PET, and the lids of High-Density Polyethylene (HDPE) (Table S7).

As for the deposition samples, MP concentrations in bottled drinking water were determined with LDIR. The average amount of MPs found in water bottles was 96 particles/L (Table S8). However, number concentrations varied considerably between the tested brands. Water samples from brand B had a low amount of MPs/L (7 ± 3) compared to A (75 ± 70) and C (90 ± 109), even though the sampling procedure remained consistent. Regarding the bottle lids, both A and B had a screw-top lid, while C had a lid that could be flipped open. This, however, showed no remarkable difference in the MPs concentration.

Nine out of the fourteen identified polymer types were found in drinking water samples. In the water samples (Figure 2) and blanks (SI, Appendix A), by far the most abundantly identified polymer type was PET and/or PU (82%). This does not come as a surprise
considering that all bottles were made of PET. Still, we had to group these two polymer types because the recorded wavenumber range from 1800 to 975 cm\(^{-1}\) could not guarantee a correct polymer identification here. Further, PA and PVC were found in concentrations above 5 %, while hardly any particles made of polyethylene (PE), i.e. the material of the bottle caps, were found. This suggests that cap wear ultimately contributed little to the particulates in the water.

3.4 Comparison of characteristics of particles in air deposition and bottled water samples

It is important to understand human exposure to microplastics in terms of particle numbers and characteristics, also for the different relevant uptake pathways and dietary components. After all, this determines the potential toxicological profiles for these routes, here: exposure from drinking water versus exposure from air. Existing studies, however, do not allow such comparisons because they all use different methods and, for example, focus on different size classes. In contrast, in the following sections we provide the first particle-level comparison of microplastic concentrations and characteristics measured by the exact same method.

3.4.1 Probability density functions for characteristics of particles in air deposition and bottled water samples

Probability density functions (PDFs) were constructed based again on pooled MP characteristics, 1010 MP from air deposition samples and 285 particles from bottled water samples. The particle characteristics in air and drinking water thus vary in place and time. However, an interpretation on the level of exposure routes rather than on the level of individual samples is more relevant for estimating the average MP exposure for a human over a longer period of time.
PDFs were constructed in the form of power law distributions 
\[ P(X \geq x) = x_{min}[\text{characteristic}]^{-\alpha} \] of the length, width, area, volume and mass of the MPs detected in air deposition samples and bottled water samples (Figure 3; Table 1). Directly comparing the distributions of characteristics for these two sample types is possible because they were recorded using largely the same methodology and instrumentation. The regressions are linear to a lower bound in the size of the particles, probably because of losses or because of lower recoveries for the lowest size classes. Therefore, the regressions were applied to the linear part of the distribution (Figure 3).

For particles’ length and width, the slope (\(\alpha\)) hypothetically can range between 0 and 3 (Kooi & Koelmans, 2019). Values around 3 indicate a three-dimensional (3D) fragmentation pattern of the particles with full conservation of mass, while lower values could indicate 2D fragments. Consequently, with measured values around 3 (Table 1), 3D fragmentation is suggested for both sample types. For the air deposition samples, this is consistent with the formation of particles from erosion and wear on the surface of textiles. For the bottled water samples, this is consistent with erosion and wear caused by cap friction (Weisser et al., 2021; Gambino et al., 2022). While 3D fragmentation is related to 1D dimensions like length or width with an exponent around 3, it is related to 2D and 3D dimensions such as area and volume/mass, respectively, with a lower exponent value.

The power law slope for area is 0.47 log units higher (i.e. more negative) for bottled water samples than for air deposition samples. The bottled water samples thus had relatively more particles with a small surface area, a difference that remains unexplained at this time.

Power law slopes (alpha) for volume and mass are equal with error limits (range 1.87 – 1.94), which can be explained from the proportionality of particle volume and mass. Remarkably, the slopes are also equal for both sample types. Although the polymer composition (Figures 1
and 2) and shape (e.g., different slopes of the surface power law) are different for air deposition and bottled water samples, the difference in polymer densities is limited and the diversity in the particle mixture is such that the differences are averaged out and almost identical mass and volume distributions are obtained.

3.4.2 Polymer composition

When comparing domestic and laboratory samples, more polymer types were found in domestic samples and were slightly less evenly distributed. This indicates that households consist of more polymer sources compared to laboratories, and that some of these sources are more abundant. This is also to be expected looking at the different furniture items and materials in a living room compared to a laboratory. A review by Chen et al. (2020) showed that polymer types PET, PP, PS, and PE in general are dominant in atmospheric (indoor and outdoor) fallout. This is in accordance to our findings with PET and/or PU (and PP) being identified in highest abundances (see Fig. 1), as they are commonly used to produce polyester fiber, fabric, textiles, packaging material, and reusable products. In the present study, PA was also detected, a polymer typically used for clothing, furniture or other household equipment. In contrast, the variety of polymers in water bottle samples was lower compared to the deposition samples (see Fig. 2). PET, here reported in a cluster with PU due to analytical restrictions, was the most found polymer in water samples. Schymanski et al. (2018) and Wrinkler et al. (2019) already stated that plastic bottles and caps can be important MP sources (especially in combination with cap friction), which is confirmed by here presented results. Still it is rather surprising that hardly any PE particles, the lids’ material, were found. A small amount of PE was only detected in water samples from the water bottle with the flip-open lid.

4. Conclusions
The data and PDFs obtained in this study are relevant for MP human exposure assessment. Eventually, from the deposition samples, exposure to MP was assessed to be 7 MP consumed per meal with a duration of 20 minutes. For comparison, the study of Catarino et al. (2018) resulted in 114 particles per meal, a difference of a factor 16. This might have been caused by the differences in household characteristics, i.e. the fabric emitting surfaces, that can explain considerable differences in the MP fallout (Table S6). Considering the household with the highest fallout, the estimated consumption would result in 16 MP per meal, still a factor of 7 different to earlier results. However, it can also not be excluded that different methodologies applied caused some of these differences. Furthermore, the calculated daily exposure ($N_{\text{daily}}$) resulted in 513 MP per day. It should be noted that this MP uptake calculation only covers the contribution of MP fallout. MP contamination in food itself, and MPs released during food processing and from packaging should be considered as well. For the water bottle samples, determined MP number concentrations were generally lower than for air deposition samples. For future work it is recommended to examine higher sample volumes to obtain a more representative distribution of data. Mohamed Nor et al. (2021) indicated that MP in beverages occur in a range with 50th percentiles from 125 to 337 particles/L. In the present study, the average amount of MP per liter (96 MP/L) was below the range reported by Mohamed Nor et al (2021). The amount of MP found in the water bottle samples ranged much wider, from 7 to 364 MP/L.

We used Laser Direct Infra-Red (LDIR) spectroscopy imaging for the first time to identify MP concentrations in household and laboratory deposition samples and in water bottle samples. In both sample types, the polymer with the highest abundance was PET and/or PU. The deposition of MP was positively correlated with the surface area of the textiles present in the studied household rooms. Power law distributions for particle width, height, area, volume
and mass were constructed for both sample types. Human exposure to airborne MP through the uptake pathway of deposition on meals and beverages followed by ingestion, as well as exposure from drinking bottled water were quantified. Such data are important for modeling and assessing human exposure to MP.

**Declaration of competing interests**

The authors declare that they do not have competing financial interests.

**Acknowledgements**

We are grateful to Merel Kooi for her help in calculating the power law distributions.

**Author contributions**

AAK conceptualized and supervised the study. JN, SMM and AAK designed the methodology. JN and SMM carried out the data collection, as well as the formal analysis of the data. JN and AAK drafted the manuscript, which was reviewed and edited by SMM. AAK acquired financial support for this work.

**References**

Akdoğan, Z.; Guven, B. Microplastics in the environment: A critical review of current understanding and identification of future research needs. Environmental Pollution 2019;254:113011

Bäuerlein, P.S.; Hofman-Caris, R.C.H.M.; Pieke, E.N.; ter Laak, T.L. Fate of microplastics in the drinking water production. Water Research 2022;221:118790
Catarino, A.I.; Macchia, V.; Sanderson, W.G.; Thompson, R.C.; Henry, T.B. Low levels of microplastics (MP) in wild mussels indicate that MP ingestion by humans is minimal compared to exposure via household fibres fallout during a meal. Environmental pollution 2018;237:675-684

Chen, G.; Feng, Q.; Wang, J. Mini-review of microplastics in the atmosphere and their risks to humans. Science of The Total Environment 2020;703:135504

Cheng, Y. L.; Zhang, R.; Tisinger, L.; Cali, S.; Yu, Z.; Chen, H. Y.; Li, A. Characterization of microplastics in sediment using stereomicroscopy and laser direct infrared (LDIR) spectroscopy. Gondwana Research 2021;108:22-30

Clauset, A.; Shalizi, C.R.; Newman, M.E.J. Power-law distributions in empirical data. SIAM Rev. 2009;51:661–703.

Coffin, S.; Bouwmeester, H.; Brander, S.; Damdimopoulou, P.; Gouin, T.; Hermabessiere, H.; Khan, E.; Koelmans, A.A.; Lemieux, C.L.; Teerds, K.; Wagner, M.; Weisberg, S.B.; Wright, S. Development and application of a health-based framework for informing regulatory action in relation to exposure of microplastic particles in California drinking water. Microplastics & Nanoplastics 2022;2:12.

Cox, K.D.; Coverton, G.A.; Davies, H.L.; Dower, J.F.; Juanes, F.; Dudas, S.E. Human consumption of microplastics. Environmental science & technology 2019;53:7068-7074

Dris, R.; Gasperi, J.; Mirande, C.; Mandin, C.; Guerrouache, M.; Langlois, V.; Tassin, B. A first overview of textile fibers, including microplastics, in indoor and outdoor environments. Environmental pollution 2017;221:453-458

Enyoh, C.E.; Verla, A.W.; Verla, E.N.; Ibe, F.C.; Amaobi, C.E. Airborne microplastics: a review study on method for analysis, occurrence, movement and risks. Environmental monitoring and assessment 2019;191:1-17
Gambino, I.; Bagordo, F.; Grassi, T.; Panico, A.; De Donno, A. Occurrence of Microplastics in Tap and Bottled Water: Current Knowledge. Int. J. Environ. Res. Public Health 2022;19:5283

Kirstein, I.V.; Hensel, F.; Gomiero, A.; Iordachescu, L.; Vianello, A.; Wittgren, H.B.; Vollertsen, J. Drinking plastics?—Quantification and qualification of microplastics in drinking water distribution systems by µFTIR and Py-GCMS. Water Research 2021;188:116519

Koelmans, A.A.; Nor, N.H.M.; Hermsen, E.; Kooi, M.; Mintenig, S.M.; De France, J. Microplastics in freshwaters and drinking water: Critical review and assessment of data quality. Water research 2019;155:410-422

Koelmans, A.A.; Redondo-Hasselerharm, P.E.; Mohamed Nor, N.H.; Kooi, M. Solving the nonalignment of methods and approaches used in microplastic research to consistently characterize risk. Environmental science & technology 2020;54:12307-12315

Kooi, M.; Koelmans, A.A. Simplifying microplastic via continuous probability distributions for size, shape, and density. Environmental Science & Technology Letters 2019;6:551-557

Kooi, M.; Primpke, S.; Mintenig, S.M.; Lorenz, C.; Gerdts, G.; Koelmans, A.A. Characterizing the multidimensionality of microplastics across environmental compartments. Water Research 2021;202:117429

Liu, P.; Shao, L.; Li, Y.; Jones, T.; Cao, Y.; Yang, C.-X.; Zhang, M.; Santosh, M.; Feng, X.; BéruBé, K. Microplastic atmospheric dustfall pollution in urban environment: Evidence from the types, distribution, and probable sources in Beijing, China. Science of The Total Environment 2022;838:155989

Mason, S.A.; Welch, V.G.; Neratko, J. Synthetic polymer contamination in bottled water. Frontiers in chemistry 2018;6:407
Mohamed Nor, N.H.; Kooi, M.; Diepens, N.J.; Koelmans, A.A. Lifetime Accumulation of Microplastic in Children and Adults. Environmental science & technology 2021;55:5084–5096

Neda, S.S.; Mark, P.T.; Scott, P.W. International quantification of microplastics in indoor dust: Prevalence, exposure and risk assessment, Environmental Pollution, 2022;119957, https://doi.org/10.1016/j.envpol.2022.119957

Newman, M.E.J. Power laws, Pareto distributions and Zipf’s law. Contemp. Phys. 2005;46:323–351.

Noventa, S.; Boyles, M.S.; Seifert, A.; Belluco, S.; Jiménez, A.S.; Johnston, H.J.; Tran, L.; Fernandes, T.F.; Mughini-Gras, L.; Orsini, M. Paradigms to assess the human health risks of nano-and microplastics. Microplastics and Nanoplastics 2021;1:1-27

Prata, J.C. Airborne microplastics: consequences to human health? Environmental pollution 2018;234:115-126

Primpke, S.; Cross, R.K.; Mintenig, S.M.; Simon, M.; Vianello, A.; Gerdts, G.; Vollertsen, J. Toward the systematic identification of microplastics in the environment: evaluation of a new independent software tool (siMPlE) for spectroscopic analysis. Applied spectroscopy 2020;74:1127-1138

Schwabl, P.; Köppel, S.; Königshofer, P.; Bucsics, T.; Trauner, M.; Reiberger, T.; Liebmann, B. Detection of various microplastics in human stool: a prospective case series. Annals of internal medicine 2019;171:453-457

Schymanski, D.; Goldbeck, C.; Humpf, H.-U.; Fürst, P. Analysis of microplastics in water by micro-Raman spectroscopy: release of plastic particles from different packaging into mineral water. Water research 2018;129:154-162
Scircle, A.; Cizdziel, J.V.; Tisinger, L.; Anumol, T.; Robey, D. Occurrence of Microplastic Pollution at Oyster Reefs and Other Coastal Sites in the Mississippi Sound, USA: Impacts of Freshwater Inflows from Flooding. Toxics 2020;8:35

Thompson, R.C.; Moore, C.J.; Vom Saal, F.S.; Swan, S.H. Plastics, the environment and human health: current consensus and future trends. Philosophical Transactions of the Royal Society B: Biological Sciences 2009;364:2153-2166

USEPA. Integrated science assessment for particulate matter. Research Triangle Park: US Environmental Protection Agency, Center for Public Health and Environmental Assessment 2019;

Valavanidis, A.; Vlachogianni, T.; Fiotakis, K.; Loridas, S. Pulmonary oxidative stress, inflammation and cancer: respirable particulate matter, fibrous dusts and ozone as major causes of lung carcinogenesis through reactive oxygen species mechanisms. International journal of environmental research and public health 2013;10:3886-3907

Vianello, A.; Jensen, R.L.; Liu, L.; Vollertsen, J. Simulating human exposure to indoor airborne microplastics using a Breathing Thermal Manikin. Scientific reports 2019:9:1-11

Weisser, J.; Beer, I.; Hufnagl, B.; Hofmann, T.; Lohninger, H.; Ivleva, N.P.; Glas, K. From the Well to the Bottle: Identifying Sources of Microplastics in Mineral Water. Water 2021;13:841

WHO. Microplastics in drinking-water. Geneva: World Health Organization; 2019. Licence: CC BY-NC-SA 3.0 IGO

Winkler, A.; Santo, N.; Ortenzi, M.A.; Bolzoni, E.; Bacchetta, R.; Tremolada, P. Does mechanical stress cause microplastic release from plastic water bottles? Water research 2019;166:115082
Wright, S.L.; Gouin, T.; Koelmans, A.A.; Scheuermann, L. Development of screening criteria for microplastic particles in air and atmospheric deposition: critical review and applicability towards assessing human exposure. Microplastics and Nanoplastics 2021;1:1-18

Wright, S.L.; Kelly, F.J. Plastic and human health: a micro issue? Environmental science & technology 2017;51:6634-6647

Zhang, Q.; Zhao, Y.; Du, F.; Cai, H.; Wang, G.; Shi, H. Microplastic fallout in different indoor environments. Environmental science & technology 2020;54:6530-6539
Table 1: Power law slopes (alpha) of the power law distribution of the microplastic characteristics length, width, area, volume and mass for MPs in deposition and water samples

| Parameters       | mean $x_{\min}$ a) | SD $x_{\min}$ | mean alpha a) | SD alpha | $x_{\min}$ plot | N obs. |
|------------------|---------------------|---------------|---------------|----------|-----------------|--------|
| **Deposition samples** |                     |               |               |          |                 |        |
| Length (µm)      | 80.63               | 22.17         | 2.79          | 0.28     | 57.79           | 285    |
| Width (µm)       | 69.22               | 17.75         | 3.76          | 0.83     | 49.19           | 285    |
| Area (µm$^2$)    | 3551.45             | 1708.35       | 2.18          | 0.23     | 1877.58         | 285    |
| Volume (cm$^3$)  | 1869669.31          | 2151093.52    | 1.89          | 0.29     | 536710.27       | 285    |
| Mass (µg)        | 0.00                | 0.00          | 1.94          | 0.33     | 0.00            | 285    |
| **Water samples** |                     |               |               |          |                 |        |
| Length (µm)      | 86.39               | 31.21         | 2.83          | 0.16     | 44.41           | 1010   |
| Width (µm)       | 82.43               | 13.75         | 3.67          | 0.32     | 37.62           | 1010   |
| Area (µm$^2$)    | 3650.88             | 980.46        | 2.65          | 0.14     | 1677.03         | 1010   |
| Volume (cm$^3$)  | 957835.06           | 528622.08     | 1.87          | 0.07     | 236682.62       | 1010   |
| Mass (µg)        | 0.00                | 0.00          | 1.87          | 0.07     | 0.00            | 1010   |

a) $P(X \geq x) = x_{\min}[\text{characteristic}]^{-\alpha}$
Figure 1: Polymer types from six households (left) with a relative contribution ≥ 3 % and two laboratories (right). See Supporting Information, Appendix B, for household polymers with a relative contribution < 3 %.
Figure 2: Polymer types from nine water bottles with a relative contribution > 3%. See Supporting Information, Appendix B, for polymers with a relative contribution < 3%.
Figure 3: Power law distributions (PDFs) for length, width, area, volume and mass for detected MPs in air deposition samples (n=285) and bottled water samples (n=1010). The slope of the PDFs equates to $\alpha$ in $(P(X \geq x) = x_{\min}[\text{characteristic}]^{-\alpha})$ with [characteristic] being either length, width, area, volume or mass of the MPs detected in air deposition samples and bottled water samples.

Author contributions

AAK conceptualized and supervised the study. JN, SMM and AAK designed the methodology. JN and SMM carried out the data collection, as well as the formal analysis of
the data. JN and AAK drafted the manuscript, which was reviewed and edited by SMM.

AAK acquired financial support for this work.

Environmental Implication

Human exposure to microplastics is one of the biggest health problems today. We offer a new method for rapid assessment of microplastic concentrations in drinking water and air deposition samples using Laser Direct Infrared Spectroscopy, and strict QA/QC criteria. We apply the method to microplastics in drinking water and to microplastics that are deposited from the air in laboratory and household rooms. We show that the deposition flux is proportional to the surface area of textiles (e.g., carpets, curtains) per unit room volume. These new findings contribute to our understanding of the effects of microplastics on indoor air quality.

Declaration of interests

☒ 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.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:
We used LDIR spectroscopy to detect microplastic (MP) in air and water samples.

Deposition of MP from air appears proportional to the surface of textile in a room.

This demonstrates how the release of MP from textiles affects indoor air quality.

We provide probability density functions for all relevant particle characteristics.

All measured MP data is directly relevant for the assessment of MP human exposure.