Household indoor microplastics within the Humber region (United Kingdom): quantification and chemical characterisation of particles present

*Lauren C. Jenner[a], Laura. R. Sadofsky[a], Evangelos Danopoulos[a], Jeanette M. Rotchell[b]**

[a]*Hull York Medical School, University of Hull, Hull, HU6 7RX, United Kingdom;*

[b]*Department of Biological and Marine Sciences, University of Hull, Hull, HU6 7RX, United Kingdom*

Corresponding Author

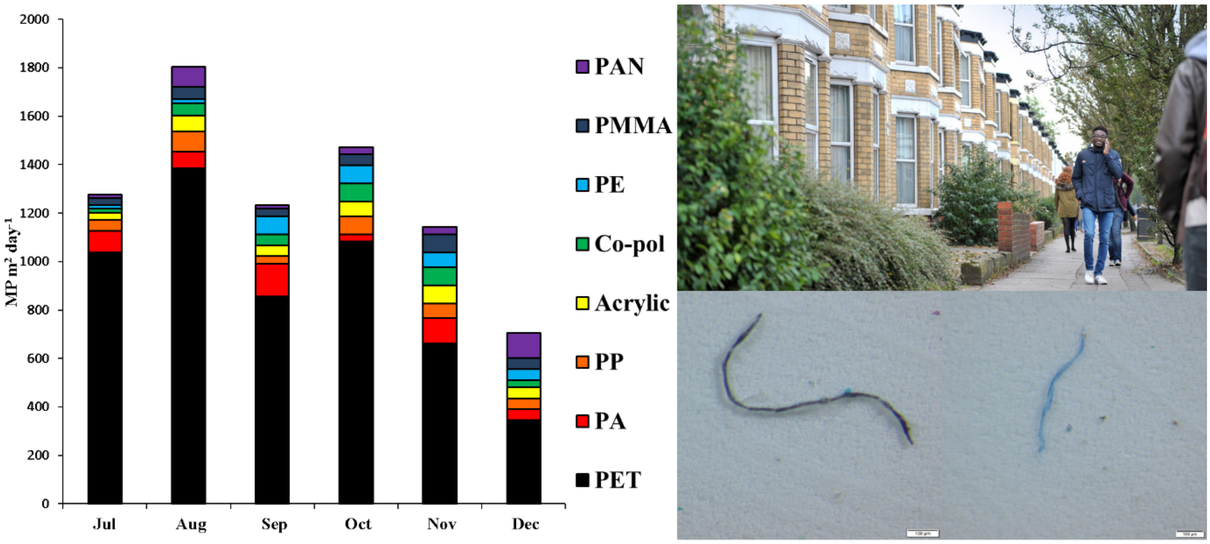
*Jeanette M. Rotchell, phone number: +44 (0) 1482 465333, e-mail: J.Rotchell@Hull.ac.uk

Abstract

Knowledge regarding the presence of suspended microplastics (MPs) within the air is lacking, especially indoors, yet the importance of indoor air quality and human health is rising. This study is the first to report MPs within multiple homes over a 6-month period, with concentrations exceeding previous outdoor studies. Twenty households, within the City of Hull and Humber region, U.K., were passively sampled, each month, collecting atmospheric fallout at head height for subsequent particle quantification, characterisation and μFTIR validation (n=3061). A household average of 1414 MP m^{-2} day^{-1} ± 1022 (mean ± SD) was observed. Smaller (5-250μm), fibrous, particles were the most abundant (90%), representing types most likely to enter the human body and cause physiological harm. Polyethylene terephthalate (PET) was present in 90% of samples and accounted for 62% of MPs. Additionally, polyamide (PA) and polypropylene (PP) were common. Results indicate that humans are exposed to significantly (1-45 times) higher concentrations, and ranges, of MPs within homes compared with the outdoor environment. In conclusion, the size range and types of MPs observed will inform laboratory experiments, using either human tissue culture or other approaches. This will allow determination of the wider implications on human health using realistic levels and representative types of indoor MPs.

Keywords: μFTIR, synthetic, indoor, home, air

Abstract art



Introduction

Plastic products are used in countless applications; from food packaging, textiles and electronics, to building, construction and medical devices (Geyer et al., 2017; Wright and Kelly, 2017). Yet, plastic waste mismanagement is stated to be a key burden in environmental literature (Murphy, 2017). Of the predicted 6300 million metric tons of global plastic waste generated prior to 2015, 79% has been estimated to aggregate within landfills and the environment, and only 9% recycled (Geyer et al., 2017). This results in the introduction of primary plastics, secondary degradation plastics and chemical leachates into aquatic, terrestrial and atmospheric compartments (GESAMP, 2015), and ultimately leads to global MP pollution. Research has previously been directed more towards routes of MPs into the environment and prevalence (Horton and Dixon, 2017), as well as dietary exposure via salt, seafood and drinking water (Catarino et al., 2018; Danopoulos et al., 2020[a]; Danopoulos et al., 2020[b]). However, there is now an emerging concern surrounding MP inhalation as another human exposure route (Wright and Kelly, 2017; Gasperi et al., 2018; Prata et al., 2020). Consequently, gaining a holistic view of MP pollution and human exposure is of increasing importance.

Since their initial identification (Dris et al., 2015), MPs have been consistently reported within atmospheric samples. MPs have been captured both passively and actively (Table S1), as well as within deposited dust samples, demonstrating their ubiquity (Abbasi et al., 2019; Huang et al., 2020). MPs (defined herein as between the size ranges of 1µm and 5mm (Hartmann et al., 2019)) are not yet considered an atmospheric pollutant. They are considered an emerging contaminant of concern and have been reported as a constituent of particulate matter (PM). Questions relating to human exposure rates and health consequences have since arisen (Wright and Kelly, 2017; Prata, 2018; Abbasi et al., 2019; Prata et al., 2020;). Passively sampled MPs are reported within literature as units of MPs m^{-2} day^{-1}, with publications from France (ranging from 53-365 m^{-2} day^{-1}) China (33-9900 m^{-2} day^{-1}), Germany (137-512 m^{-2} day^{-1}), and most recently, the UK (3-771 m^{-2} day^{-1}) (Dris et al., 2015; Dris et al., 2016; Cai et al., 2017; Allen et al., 2019; Klein et al., 2019; Wright et al., 2019; Zhang et al., 2020). In contrast, MPs captured actively are expressed in units of MP m^{-3}, with reports from France (0-59 m^{-3}), Denmark (9 m^{-3}), China (0-5 m^{-3}) and America (1-13 m^{-3}) (Dris et al., 2017; Abbasi et al., 2019; Liu et al., 2019[a]; Liu et al., 2019[b]; Li et al., 2020). To date, just four studies worldwide report on MPs within the home (Dris et al., 2017; Vianello et al., 2019; Zhang et al., 2020; Gaston et al., 2020). It is important to note that environmental sampling (duration, location, meteorological conditions, sample number), sample processing steps (digestion, purification), microscopic analyses (observational criteria, size and shape categories) and chemical analyses methodologies all vary, and it is difficult to conduct meaningful inter-study comparisons. Despite this, the majority of studies evidence that MPs increase in concentration with decreasing particle size (until an observational limit is reached) (Allen et al., 2019). MPs are prevalent in locations of high human activity e.g. urbanised city centres (Dris et al., 2016), and tend to be fibrous (Dris et al., 2016; Liu et al., 2019[a]). One passively sampled study recorded indoor atmospheric MP levels ranging from 1500-9900 MPs m^{-2} day^{-1}, depending on the room, a rate exceptionally higher than outdoor studies (Zhang et al., 2020). MP studies that have reported actively sampled indoor MP levels (ranging from 1-59 m^{-3}) (Dris et al., 2017; Vianello et al., 2019), again, exceed that of outdoor concentrations (which range from 0-6 m^{-3}) (Table S1). The significance of indoor air quality is highlighted by the fact that humans spend up to 90% of their time indoors (Klepeis et al., 2001) and as much as 60% within their homes (NICE, 2020). Whilst the health effects of exposure to indoor MPs are not yet defined, indoor PM has been linked to a number of health impacts including a decline in respiratory and cardiovascular health (Royal College of Physicians and Child Health, 2016) as well as specific MP types inducing toxic impacts in recent human lung cell culture exposure studies (Goodman et al., 2021; van Dijk et al., 2021).

To date, there are many unknowns: from the chemical composition, size or shape of the MP, to any chemical leachate or adsorbed pollutants. The possibility of MPs entering the human body and impact of such exposure on health is of increasing concern. This study aims to provide knowledge surrounding the indoor MPs that humans are most likely exposed to, by quantifying concentration rates, and determining particle dimensions, shapes and chemical composition.

Material and Methods

A total of 20 houses were selected for sampling (following a request to volunteer), all located within the city of Hull and wider East Riding of Yorkshire, U.K. region (Figure 1). Sampling covered a six-month timescale investigation from July until December 2019. Participants were given a short questionnaire (Supplemental Information) to gain details regarding their living conditions and routines. Additionally, each month, outstanding events such as ‘occupants away during sampling period’ or ‘building maintenance within the home’ were noted. Participant ages were categorised; 18-25, 26-35, 36-50, 51-65 and >65 (5%, 40%, 20%, 25%, 10%, respectively). Household occupancy was determined and ranged from 1-4 people (mean 2.2). The hours per day that participants spent within the home ranged from 10-21 h (mean 16 h). The hours spent within the room in which sampling was conducted ranged from 2-12 h (mean 7 h). Carpets and other furnishing samples were taken from the homes of willing participants to compare polymer composition to particles isolated from atmospheric samples.

Passive sampling

Identical 1L glass beakers (0.0095 m^2) were placed at head height (1-1.8 m) within the downstairs room that residents reported was the most occupied within the home. The beakers were opened by removing the foil lid on a set date and time. The beakers were exposed to atmospheric fallout for exactly 7 days to avoid weekend or weekday bias (Zhang et al., 2020), and participants were instructed to not change any of their habits or routines. A foil lid was placed tightly onto the beaker upon termination of the sampling period. Collection and transport to the laboratory was conducted by the primary researcher. Beakers were stored at room temperature in the dark until bulk processing was conducted.

Figure 1.A.

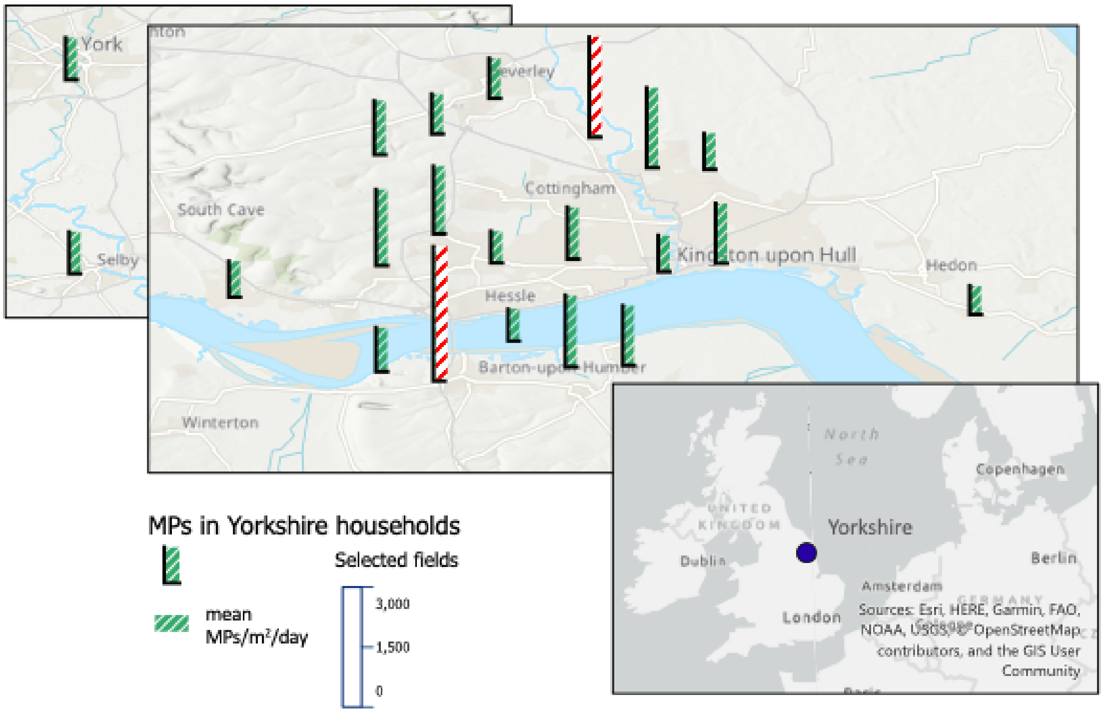


Figure 1.B.

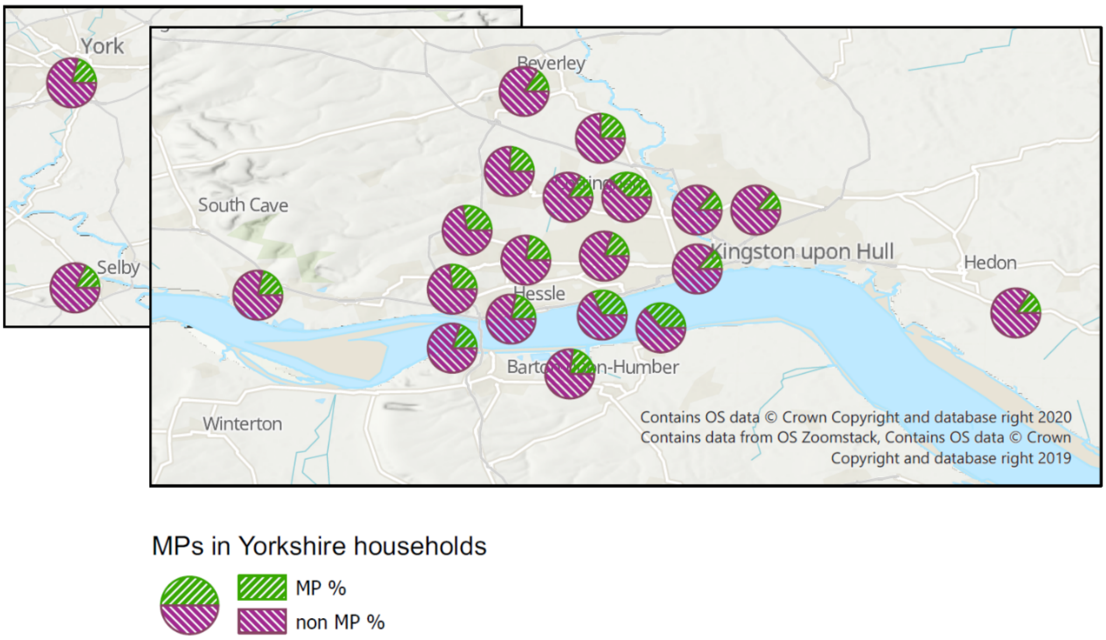


Figure 1. Location, levels and types of particles detected in the Hull and East Riding of Yorkshire region, U.K., with plotted points indicating each household. **A.** Mean MP concentration rate for each household (MP m^{-2} day^{-1}), red/white denotes households with high MP concentration rates relative to the majority of others, and **B.** Proportion of MPs to non-MPs at each household location.

Sample pre-treatment

Particle overload was a problem encountered during the pilot study. In order to significantly reduce the particle count, a digest to remove biological (non-MP) materials was conducted as follows. Each beaker was washed three times with 200 ml of 30% hydrogen peroxide solution (H_2O_2) (Fisher Scientific, Loughborough, U.K.). Beakers were rinsed in a standardised manner, ensuring the sides of the beaker were washed thoroughly (Dris et al., 2015; Liu et al., 2019[a]). The rinsate was collected into a 1L glass conical flask and sealed immediately with a foil lid. Each sample was placed in a shaking incubator at 55°C to ensure no destruction of polymers with a low degradation temperature (Allen et al., 2019), and rotated at 65 rpm for 3 days. This pre-treatment digestion step removed biological and other natural organic matter, such as skin and pet dander, before particle analysis (Clark, 1974; Vianello et al., 2019; Stanton et al., 2019), whilst not damaging any MP particles (Nuelle et al., 2014). Conical flask contents were then filtered using a glass vacuum filtration device and particulates collected onto mixed cellulose esters membrane filters (MCE), 47 mm diameter and 5 µm pore size, (MERCK, Gillingham, U.K.) Each conical flask was washed three times with 200 ml of MilliQ water and the sides of the glassware were rinsed to reduce particle loss. Filters were stored in covered petri dishes, labelled and allowed to dry at room temperature in the dark for a minimum of 24 hours.

Particle level quantification and characterisation

One quarter of each sample filter was randomly sectioned for stereomicroscope analysis (Olympus SZX10, Olympus Corporation, Japan), and extrapolation of the dataset was later calculated to represent the entire sample. Therefore, an assumption was made that even particle distribution had occurred during the filtration process, which may not always be the case. The length (largest side of particle) and width (second largest side of the particle) was recorded for each particle, as well as particle shape (Free et al., 2014) (Table S2). Particle dimensions were measured (CellSens software, Olympus Corporation, Japan) and categorised as follows; length: 5-249 µm, 250-500 µm, 501-1000 µm, 1001-5000 µm, and width: 5-10 µm, 11-20 µm, 21-30 µm, 31-40 µm, 41-50 µm and >50 µm. Particles with a length >5 mm were removed from the filter with tweezers in order to focus the analysis of the particles to a standard MP size range (Hartmann et al., 2019).

Particle chemical composition analysis

For each sample, 20% of the identified particles were analysed by µFTIR analysis. Particles were placed individually onto a DC-3 diamond compression cell (Specac Limited, Orpington, U.K.) and µFTIR analysis (using a Nicolet iN10, ThermoFisher, Waltham MA, U.S.A.) was conducted in transmission mode and using the liquid nitrogen cooling system. A blank area next to the particle being analysed was chosen as a background reference, before immediate analysis using the same spatial resolution (Omnic software package, ThermoFisher, Waltham MA, U.S.A.). All background and sample spectra were obtained using a scan number of 64 and a spectral range of 4000-675cm^{-1}. Each resulting spectrum was compared to a cluster of polymer libraries (Omnic polymer libraries). Three attempts were made to gain a spectrum with a match index ≥70%, and only these were included in the results.

Quality assurance and control

To optimise methodologies, a one-month pilot study was conducted in which the deposited dust from 10 households was continuously passively sampled and analysed, during June 2019. This facilitated subsequent standardised sampling, pre-treatment and analysis protocols to be developed.

MilliQ water and H_2O_2 were filtered three times with 47 mm glass fibre grade 6 filters (GE Healthcare Life Sciences, Marlborough MA, U.S.A.) using a glass vacuum filtration kit to reduce background particle contamination. Also, a procedural control was run alongside each batch of samples (n=26). A blank MCE filter was opened during stereomicroscope and µFTIR analysis in order to monitor atmospheric fallout contamination during laboratory particle analysis (n=118). All glassware were washed and rinsed three times with triple filtered MilliQ water before being immediately wrapped in foil. Foil lids were only removed when necessary and opening times kept to a minimum. Ventilation from windows, doors and airing devices was minimised and work was conducted at times of low activity to minimise particle suspension. 100% cotton laboratory coats were worn during all stages of the project and a fume cupboard used where possible. One individual researcher was responsible for all samples, ensuring standardised analyses throughout.

Limit of detection (LOD) and limit of quantification (LOQ) calculations (Horton et al., 2021) were applied to the three most common MP types identified within samples (Supplemental Materials Methods SM1 and Table S3).

Statistical analysis

MP quantity identified on each quarter was extrapolated to represent the entire filter and then the value adjusted to represent MP m^{-2} day^{-1} as follows: MP quantity per sample was multiplied by 105 which represents the factor to convert from the bottom of the beaker surface area to m^2, and then divided by 7 for a per day value (Supplementary Method 1). Statistical analyses were performed using SPSS. All data were determined not normally distributed with a Shapiro-Wilk test and either a Kruskal-Wallis test or Mann-Whitney U test applied. Statistical significance was accepted at a p <0.05 level and extreme significance at 0.01.

Results

6 samples were collected from each of the 20 households, with the exception of 2 samples. Therefore, a total of 118 passive samples were collected throughout the study. µFTIR analysis was

conducted on 3061 particles consisting of 2442 of ≥70% match index, 585 of 60-69% match, and 34 of <60% match.

Control and laboratory blanks

A total of 299 particles were identified from the 144 controls. From these, 25 were identified as MPs (ranging from 0-1 per sample, mean 0.2 ± 0.4), specifically polyurethane/ polypropylene copolymer (PUR/PP), polyvinyl chloride (PVC), PET and PP which were present in low levels (3, 2, 2, 2%, respectively). Non-MP particles consisted primarily of cellulose/ cellophane (86%). MPs identified within control samples were significantly different from MPs identified within samples ($p = 1.86 \times 10^{-41}$). Due to the low contamination rates of MPs within each sample it was decided not to subtract contamination rates from final results (Vianello et al., 2019) but LOD/LOQ results have been reported for the most common MP types observed (Table S3).

Total particle fallout

The number of particles collected during the sampling period varied, with a mean of 6204 ± 3122 particles m^{-2} day^{-1} (mean ± SD) and a range of 902-14,551 particles m^{-2} day^{-1}. The mean particle quantity for December was significantly lower than July ($p = 0.003$), August ($p = 0.005$) and September ($p = 0.05$), (Figure 2), however all other months were not significantly different in terms of their mean particle quantity.

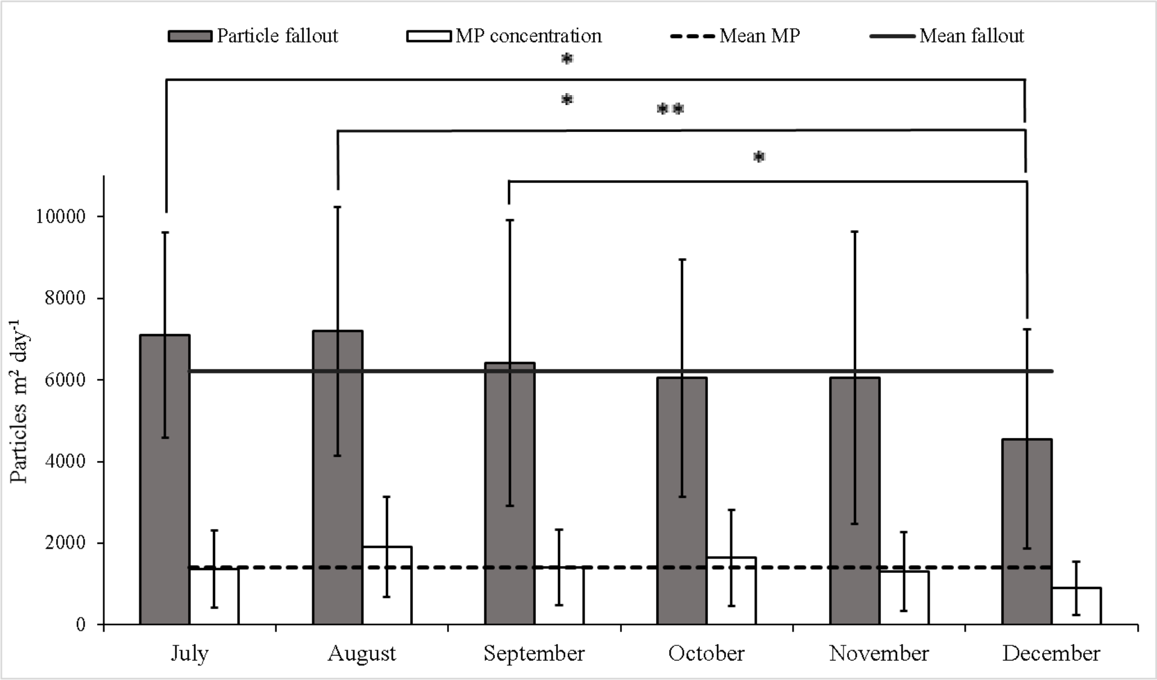


Figure 2. Bar chart displaying monthly (mean) particle fallout and monthly (mean) MP concentrations alongside the overall means. Particle fallout significant differences Dec/Jul (p = 0.003), Dec/Aug (p = 0.005), Dec/Sept (p = 0.05). No significant differences were recorded for MP concentration and month (p = 0.06).

Total particle characterisation

The majority of particles were of a fibrous nature (90%). The remaining 10% were comprised of fragment, film and sphere (8%, 1%, 1%, respectively) (Figure 3). No foam-like particles were identified within any samples.

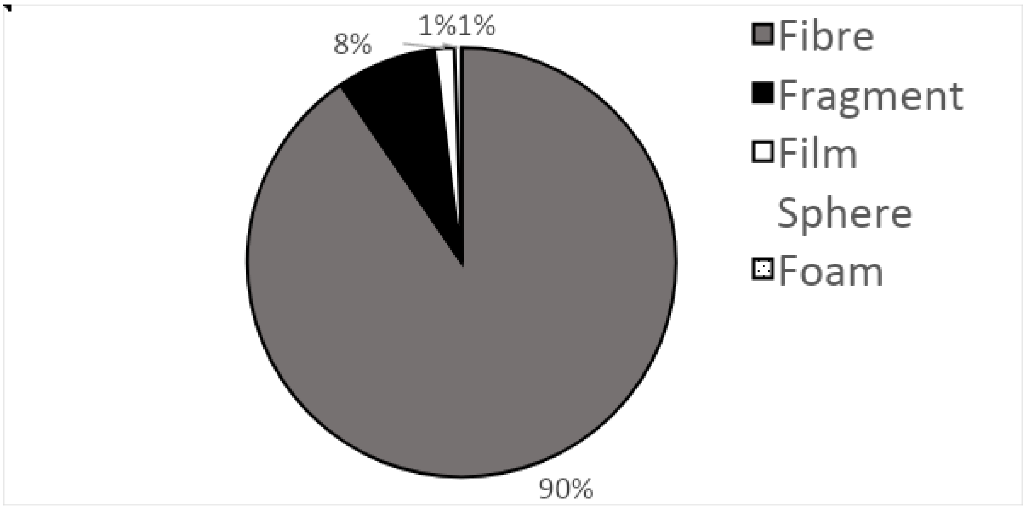


Figure 3. Pie chart displaying the total particle shape distribution.

An increase in particle concentration with decreasing particle length was observed (Figure 4.A.). Particles 5-250 µm accounted for 59% of particles, the remaining 41% were comprised of 250-500 µm, 501-1000 µm and 1001-5000 µm (18%, 13%, 10%, respectively). Particle width size categories were; 5-10 µm, 11-20 µm, 21-30 µm, 31-40 µm, 41-50 µm and >50 µm (29%, 63%, 6%, 1%, 0%, 1%, respectively) (Figure 4.B.).

Figure 4.A.

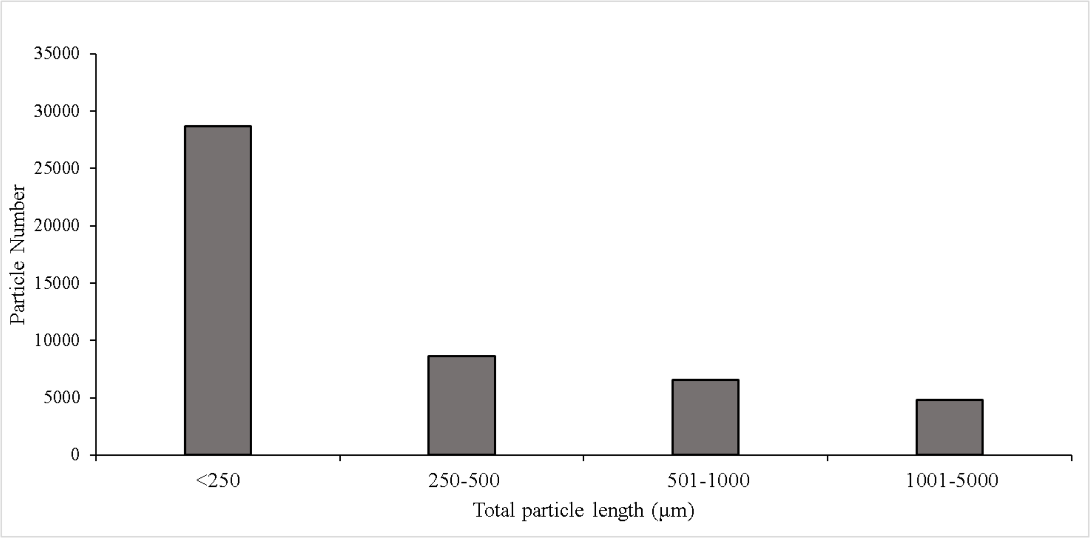


Figure 4.B.

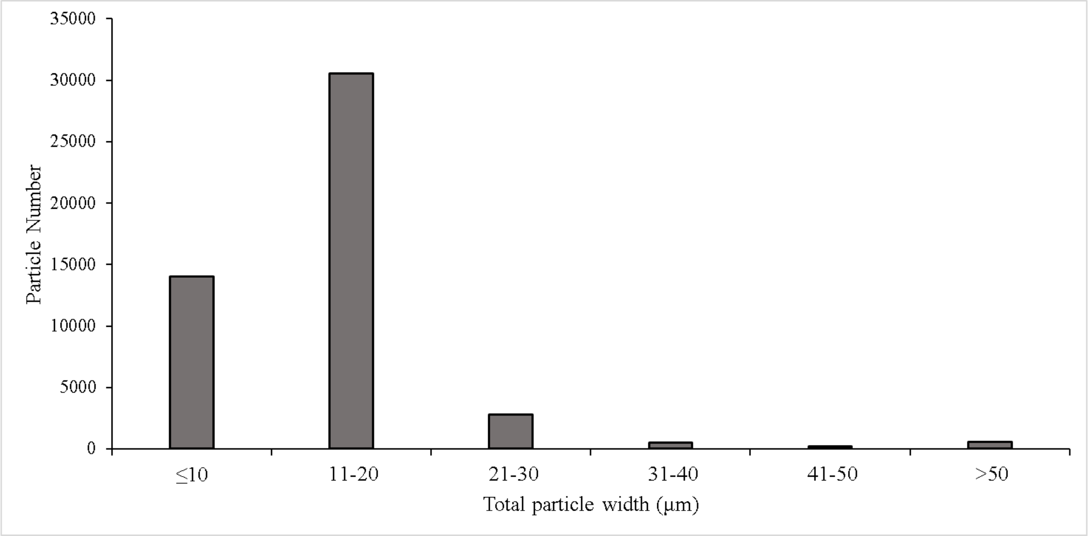


Figure 4. Bar chart showing the particle size distributions of the entire total particle fallout. **A.** Particle length categories and abundance, **B.** Particle width categories and abundance.

Non-MP particle composition

Non-MP particles (natural and artificial particles not of a petroleum derived nature) accounted for 77% of the total particle count (July 81%, August 74%, September 78%, October 73%, November 78%, December 80%). Non-MP fractions for each household ranged from 63-86% (Figure 1.B.). Of the non-MP fraction, cellulose/ cellophane accounted for 92% of particles. The remaining natural and artificial particles were comprised of zein, silk, cocamide and silicon dioxide (5%, 1%, 1%, 1%, respectively), and others (Figure S1).

MP concentration

The number of MP identified during the sampling period varied, with a mean of 1414 ± 1022 MP m^{-2} day^{-1} (mean ± SD), 0-5412 MP m^{-2} day^{-1} (range), 1203 MP m^{-2} day^{-1} (median). MPs were identified in all households and within 98% of samples (116/118). Lower concentration rates were noted for the month of December but there were no significant differences found between the sampling months ($p = 0.06$) (Figure 2). Two households had significantly higher MP concentration rates compared to others throughout the sampling period (Figure 1.A).

MP particle composition

Synthetic MPs particles accounted for approximately 23% of the total particle fallout across all households, which varied by month as follows: July 19%, August 26%, September 22%, October 27%, November 22%, December 20%. MP fractions for each household ranged from 14-37% (Figure 1.B.). PET was present in 90% of samples and accounted for 63% of the total synthetic fraction (Figure 5.A.). Other MP polymer types identified were PA, acrylates, PP, co-polymer blends (containing at least one petroleum derived polymer), polyacrylonitrile (PAN), polyethylene (PE), polymethacrylate (PMMA) (6%, 4%, 4%, 3%, 3%, 3%, 3%, respectively) and others (Figure 5.A.). Polymer type concentrations varied, with PET being the most abundant MP within every sampling month (Figure 5.B.) The level of PET recorded for December was significantly lower than July ($p = 0.005$), August ($p = 0.000$), September ($p = 0.004$) and October ($p = 0.001$) (Figure 5.B.). PET, PA and PP particle numbers identified within samples were above the LOD and LOQ (of 1.1 and 3.3 respectively, for PET as an example) (Table S3).

Figure 5.A.

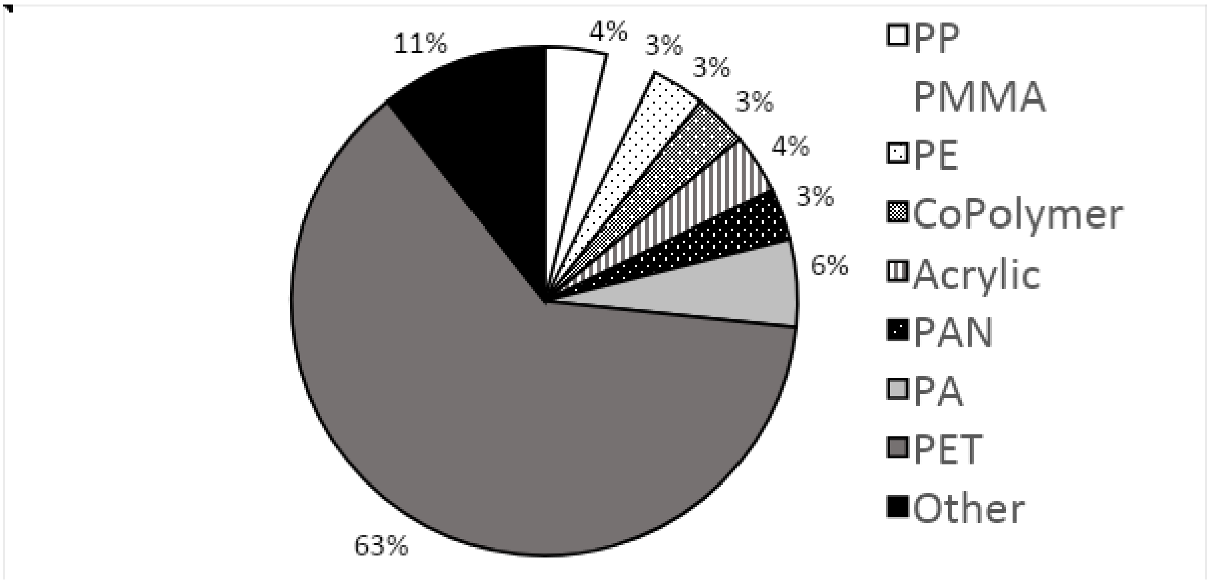


Figure 5.B.

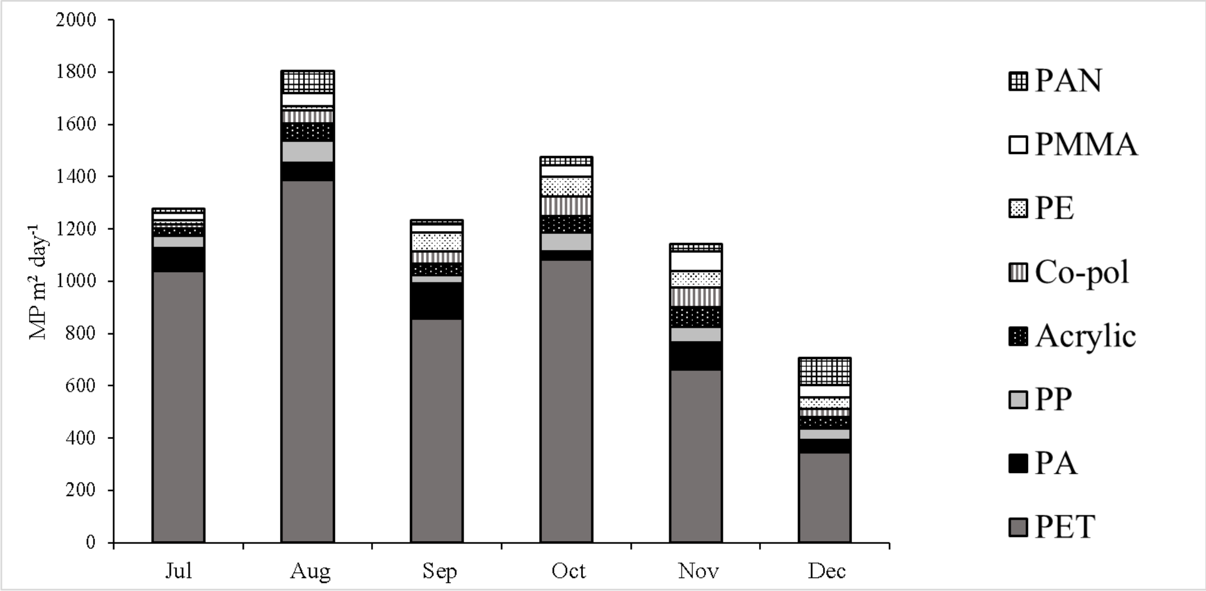


Figure 5. A. Pie chart displaying the synthetic MP polymer types alongside their abundance, **B.** Bar chart showing the monthly concentration (mean) of each MP polymer type. **Abbreviations:** PET = polyethylene terephthalate, PA = nylon, PP = polypropylene, Co-pol = co-polymer, PE = polyethylene, PMMA = polymethacrylate, PAN = polyacrylonitrile.

Discussion

This study addresses the limited knowledge on indoor MPs, specifically to outline what types and levels of MPs humans may be typically exposed to on a daily basis within the home. Of the three studies reporting MPs in a home environment, sampling was conducted on: one dormitory in China (2 days sampling per week, for 3 months) (Zhang et al., 2020), three apartments in Denmark (3 days consecutive sampling, totalling 9 samples) (Vianello et al., 2019) and two apartments in France (4-7 hours sampling, for 3 days in Feb, May, Jul, Oct) (Dris et al., 2017). This study provides a more substantial indoor MP dataset for 20 UK households over a 6-month period, with sampling durations of a week. Due to the increased importance of applying chemical analysis alongside MP data, this study developed 'best practice' by; providing a large number of particles analysed and validated by µFTIR. In doing so, the approach avoids selecting particles for analysis based on observational criteria alone and provides a larger sample size and duration to represent a more longitudinal dataset (Table S1). The approach used also takes into account any background contamination (Brander et al., 2020) and includes an LOD/LOQ consideration (Horton et al., 2021). These are important since the atmospheric microplastics research field is developing, and as such, no standardised procedures are in place. This approach provides a quality assurance threshold above that of simple polymer contamination subtraction, and values below the LOQ are not included in final concentrations. Within this study, MPs within samples were high and background contamination low, therefore final concentration levels were not significantly affected (Table S3). However, this LOD/LOQ technique provides an option for future studies to apply more stringent reporting, especially for datasets in which contamination is high or MP counts within samples are low.

MPs were identified within all households and 98% of samples, thus it is evident within an indoor setting that MPs are ubiquitous. An average household atmospheric MP concentration rate of 1414 ± 1022 MP m^{-2} day^{-1} (mean ± SD) (Figure 2) was observed. This is significantly higher than reported by all presently available passively sampled outdoor studies, and approximately 1-45 times higher than outdoor studies (Table S1). This reinforces claims of a higher contamination rate of atmospheric MPs within an indoor environment (Dris et al., 2017; Vianello et al., 2019; Zhang et al., 2020). It is also corroborated by active air sample concentration rates which range indoors from 1-59 MP m^{-3} and outdoors from 0-6 MP m^{-3} (Table S1). One study has investigated indoor atmospheric MPs in the comparable units of MP m^{-2} day^{-1} (Zhang et al., 2020), in which a dormitory (9,900 MP m^{-2} day^{-1}), office area (1,800 MP m^{-2} day^{-1}) and corridor (1,500 MP m^{-2} day^{-1}) were sampled discontinuously for 3 months, reporting concentrations similar to and higher than this current study. Importantly, cellophane and rayon, artificial polymers of natural origins were classified as MPs, unlike this study, and therefore comparable concentration rates are less than those stated. Despite this, there is a general consensus that indoor atmospheric MP levels significantly exceed that of outdoors (Dris et al., 2017; Vianello et al., 2019).

Fibres were the predominant particle form identified (90%), this is similar with the majority of other atmospheric MP studies which report a range from 67-90% (Dris et al., 2015; Cai et al., 2017; Wright et al., 2019; Zhang et al., 2020; Huang et al., 2020), yet contradicts others that report more fragments (Klein et al., 2019; Liu et al., 2019[b]; Vianello et al., 2019; Gaston et al., 2020). An 8% particle fragment rate was reported here, more than film (1%), sphere (1%) and foam (0%) (Figure 3). No foam-like particles were identified in any samples, regardless of a likely source being insulation, packing, furniture and carpet underlay (Hale et al., 2020; Sobhani et al., 2020). Discrepancies between fibre and fragment prevalence may be due to differing sampling locations e.g. remote versus urban, but could also be due to ambiguity within shape classification criteria, especially with decreasing particle size (Vianello et al., 2019).

The typical particle size dimensions for atmospheric MP studies observed is an increase in concentration with decreasing particle size (Figure 4.A.). Some studies report a fall in concentration before an observational size limit is reached (Liu et al., 2019[a]; Gaston et al., 2020), as seen within width categories in this study (Figure 4.B.). This may be due to an inability to detect smaller particles, or loss of smaller particles during sample processing. Particles in the size range 5-250 µm were the most abundant throughout all particle shape categories; fibre (55%), film (88%), fragment (99%) and sphere (98%). 92% of particles had a width ≤20 µm. This is consistent in terms of length (but not width) with selected studies reporting mean particle sizes of ~50 µm (Vianello et al., 2019), or 58.6 ± 55 µm for indoors and 104 ± 64.9 µm for outdoors (Gaston et al., 2020) but differs from another indoor study reporting a predominant size range of 50-2000 µm (Zhang et al., 2020). Particle size and shape, especially the finding of predominantly smaller fibres within households, is critical in terms of the potential health impacts discussed later.

Of the MPs identified within the households, whereby the mean value was 1414 ± 1022 MP m^{-2} day^{-1}, PET was found in 90% of samples and was 63% of the total synthetic fraction (Figure 5.A.). Other MP polymer types identified, to a lesser extent, were PA, acrylates, and PP. These findings differ significantly from previous studies of indoor MPs which report PES, PE, PA and PP as common synthetic MPs (Dris et al., 2017; Vianello et al., 2019; Zhang et al., 2020). There were more than 20 polymer types identified within this study, with PET being significantly higher in estimated concentration rate (887 ± 808 particles m^{-2} day^{-1}) compared to all other MPs (Table S3), despite PET having one of the highest densities of plastics (1.38g/cm^3) (Iñiguez et al., 2017). In the households studied, PET is the MP that occupants are therefore most exposed to. PET is a thermoplastic synthetic polymer used to produce drinking bottles and containers for cleaning products (Rodríguez-Hernandez et al., 2019). Its transparent and flexible nature, as well as adaptability makes it a high demand material in the packaging and textiles industry (Iñiguez et al., 2017; Hahladakis et al., 2018). In 2015 it was estimated that 27.8 million tons of PET were produced, globally, accounting for 8.6% of all polymer types (Rodríguez-Hernandez et al., 2019). The second most detected MP in the household samples, PA, specifically synthetic nylon particles, comprised 6% of the total MP fraction, with an average 79 ± 182 particles m^{-2} day^{-1} for each household (Table S3). Compared to PET, nylon has fewer indoor applications, and is found within indoor textiles and fabrics (Vianello et al., 2019). Interestingly however, Vianello et al. (2019)

reported the nylon particles captured were all fragments. PP, acrylic and PAN particles were also present in household samples (4%, 4%, 3%, respectively), and are often reported in atmospheric MP studies (Table S1). These are used in packaging, textiles and reusable plastic products (Allen et al., 2019). An outdoor study in London, United Kingdom, suggested winter clothing could influence the MP polymer concentration at different times of the year, for instance PAN was higher during winter months when warmer clothes were needed (Wright et al., 2019).

In terms of seasonal differences, no variation was detected for PA, PP or the acrylic categories ($p = 0.341$, $p = 0.749$, $p = 0.56$, respectively). However, during December, PET was significantly lower (compared with July $p = 0.005$, August $p = 0.000$, September $p = 0.004$, and October $p = 0.001$), suggesting a reduction in PET sources and suspension during the start of the winter period. PAN was higher in mean concentration during the months of August (mean 84 ± 139 PAN particles m^{-2} day^{-1}) and December (mean 105 ± 177 PAN particles m^{-2} day^{-1}) compared with July (mean 15 ± 67 PAN particles m^{-2} day^{-1}), September (mean 15 ± 67 PAN particles m^{-2} day^{-1}), October (mean 30 ± 93 PAN particles m^{-2} day^{-1}), and November (mean 30 ± 135 PAN particles m^{-2} day^{-1}). These months are when families are traditionally on breaks from education and work. PAN being significantly higher in December supports the claim that PAN increases in winter (Wright et al., 2019) due to clothing needs. Therefore, although indoor MPs are likely not directly affected by meteorological changes, it is possible that polymers can be indirectly influenced by the outdoor compartment, changes in household occupancy, activities and clothing.

It has been suggested that through observation of MP particle characteristics, it is possible to gain knowledge of their likely origin and history (Free et al., 2014; Cai et al., 2017). The exact sources of MPs are not yet well understood, non-fibrous MP even less so. Fragmented particles are thought to originate from ‘wear and tear’ of larger plastic products, resulting in an irregular shape, such as shards from plastic bottles (Free et al., 2014). Film shaped particles are likely from carrier bags and other thin transparent products, and spherical particles are used in personal care products (Leslie, 2014). The most frequently mentioned probable source of atmospheric fibrous particles is their release from textiles, clothing and furnishings. Either during wear or use of a fabric item, or during the washing and drying of fibrous products, resulting in shedding (Abassi et al., 2019; Dris et al., 2016; Dris et al., 2017; Cai et al., 2017; Wright et al., 2019; Zhang et al., 2020). It is also suggested that the more a fabric item ages, the more particles it sheds, and additionally an increase in plastic fabrics within the home is likely to increase atmospheric MP concentration (Zhang et al., 2020). When textiles, such as clothing and curtains, were taken from a single indoor setting, synthetic products accounted for <40% (56% cotton and 17% polyester (PES), with remaining constituents being a blend of co-polymers) (Zhang et al., 2020). Further, when the study compared these textile products to those collected during sampling, natural and PES particles were prevalent polymers. If MP concentration is influenced by the number and type of plastic textiles in the home, as well as how much they are used, it is likely that high variation would be seen when comparing household MP concentrations but also when investigating a single household over a period of time, and such wide ranges were reported here; 0-5412 MP m^{-2} day^{-1}. Within an indoor environment, fabrics are common, however, other sources cannot be ruled out, such as macroplastic degradation and release of synthetic fibres from carpets (Dris et al., 2016).

After the identification of atmospheric MPs, discussion has followed on sources and factors influencing their abundance and transport. These include precipitation and meteorological events, wind speed and direction, vehicular and road emissions, human activity and population density (Dris et al., 2016; Cai et al., 2017; Allen et al., 2019; Wright et al., 2019). Seasonal variance could also influence indoor polymer concentration and type, via an exchange of MPs from indoors to out, and vice versa (Royal College of Physicians and Child Health, 2016; Zhang et al., 2020). Differences in building materials, furniture, cleaning routine and human activity levels may also contribute (Vianello et al., 2019) making it hard to pinpoint sources. Of the recorded events within the home (derived from the participant survey), occupancy levels had no significant difference in particle fallout ($p = 0.562$) or MP concentration ($p = 0.719$) (Table S4). Building maintenance increased fallout and MP concentration in some households but not all (Table S4). Building work and maintenance has previously been reported to increase atmospheric MP concentration (Zhang et al., 2020) and to be a contributor to indoor air pollution (Royal College of Physicians and Child Health, 2016). Also, within the households studied, no significant difference was detected when investigating the use of regular indoor clothes hanging, tumble dryers, or presence of carpet (Table S5). The presence of pets (in 10 households reporting at least one) also had no significant impact in the total particle fallout detected (p = 0.862) or MP concentrations (p = 0.170). All households reported windows and doors being used as the most common method for airing. Particle fallout concentrations were highest during the summer months, with a significant decline observed during winter (Figure 2). It could therefore be suggested that ventilation increases particle suspension within the home, in the same way that wind suspends outdoor particles (Allen et al., 2019; Wright et al., 2019). A study investigating air conditioning units and MPs also supports this phenomenon (Zhang et al., 2020).

While it is unclear what the main drivers of indoor MP concentration and types are within the home, their levels are none-the-less high, especially PET, relative to outdoor environments (Table S1). Indoor air quality is a global cause for concern since humans spend the majority of their time indoors (Klepeis et al., 2001; NICE, 2020), with the participants in this study reporting that they spend a mean of 7h in the room sampled. The fibres detected in these households were mainly in the 5-20 μm length category, raising concerns relating to human ability to inhale particles of this size range, whether they enter lungs and what might be the health consequences. One published study highlights MP-induced human health impacts, caused by nylon fibers, in the occupational setting of nylon flock making (Burkhart et al., 1999). Workers displaying lung disease had been typically exposed to nylon fibres of size range 10-15 μm width and 1000 μm length at an average respirable particulate concentration of 2.2 mg m^3 (Burkhart et al., 1999), significantly more particles experienced in an average household environment yet consisting of the same particle size (width) range. In a controlled laboratory exposure study, human lung cell cultures have recently been exposed to nylon fibers (of approximate size shape 10 μm x 30 μm), at a level of 5000 fibers, and damage to the lung cell growth and development observed (van Dijk et al., 2021).

In conclusion, household environments contain significant levels of MPs, which range across homes and season from 0-5412 MP m^{-2} day^{-1}, with a mean of 1414 ± 1022 MP m^{-2} day^{-1} (mean ± SD). The most commonly detected MP was PET and the most abundant size dimensions detected was 5-250 μm, within the size range detected in nylon flock workers with lung disease and also found to harm lung cells in culture (Burkhart et al., 1999; van Dijk et al., 2021). The findings herein can inform laboratory exposures using human lung cell cultures, as part of our future work to investigate environmentally-relevant levels, using the most common chemical types of MPs detected, and determine any human health impacts.

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