

UNIVERSITY OF BIRMINGHAM

Occurrence, human exposure, and risk of microplastics in indoor environments

By

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A thesis submitted to the University of Birmingham

for the degree of

DOCTOR OF PHILOSOPHY (Ph.D.)

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December 2024

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Abstract

Microplastics (MPs), tiny plastic particles less than 5 mm in size, have become a significant environmental concern due to their widespread presence and potential health impacts, especially in indoor sites. This thesis investigates MPs occurrence, concentrations, and characteristics (i.e., size, shape, and polymer type) in indoor air (airborne and atmospheric deposition) and settled indoor dust in Birmingham, UK. The measured MPs concentrations are then used to estimate exposure of UK adults and toddlers to MPs via inhalation and inadvertent dust ingestion in UK homes and workplaces for the first time. Moreover, potential seasonal variation in indoor MPs concentration and estimated daily intakes were studied for the first time, over a 12-month monitoring period in 5 indoor microenvironments. In this thesis, analytical protocols have been developed and quality assured to minimize contamination during sampling/clean-up stages and accurately determine MPs in indoor air and dust samples. A digital fluorescence microscope has been applied to determine the size, shape, and count of MPs particles as physical properties. Additionally, Fourier-Transform Infrared Spectroscopy (μ-FTIR) was applied to identify the polymer types as chemical properties. The analyses revealed the presence of fibers, fragments, and foams of varying sizes. Polyethylene terephthalate (PET), polyvinyl chloride (PVC), and polypropylene (PP) were identified as the most prevalent polymer types in the studied samples. Particles in the size range (10 – 50 μm) were the most abundant in airborne MPs, while larger MPs (50 – 100 μm) were more frequently detected in atmospheric deposition and settled indoor dust samples.

The study of MPs in UK indoor air and inhalation exposure via active and passive air sampling from 30 homes and 30 workplaces is the first and most comprehensive study in this scope. 120 samples were investigated, and the mean concentrations of MPs in UK homes were significantly higher ($P<0.05$) than those in workplaces for both active and passive air samples. Airborne MPs concentrations were observed in homes (15.6 ± 5.4 MPs/m^3) compared to workplaces (13.1 ± 6.5 MPs/m^3). For atmospheric deposition samples, the average MPs concentrations were 3735 ± 1343 MP/m^2/day in homes and 3177 ± 1860 MP/m^2/day in workplaces. Similarly, higher concentrations of MPs were measured in settled indoor dust from homes (155 ± 222 MPs/mg), compared to workplaces (125 ± 209 MPs/mg) sampled in the present study. This was mainly attributed to carpeting, where all the sampled homes were carpeted, while 13 of the studied workplaces were uncarpeted.

The seasonal variation study revealed statistically significant differences ($P < 0.05$) in mean concentrations of MPs in indoor air and settled dust among the studied seasons for the first time. All sampling locations experienced the highest concentrations of MPs during the summer months, while significantly lower levels were observed in autumn and spring, which was reflected in significantly different estimated daily intakes of MPs in different seasons. This is of particular importance to exposure and risk assessment studies because it raises concern over the representativity and comparability of existing human exposure data from different studies based on single "spot" samples collected from various indoor microenvironments in different seasons.

Overall, Estimated daily intakes of UK toddlers to MPs were substantially higher than those for adults via both inhalation and inadvertent dust ingestion in all seasons and using different exposure scenarios. The is of concern due to toddlers' incompletely developed immune and nervous systems. Therefore, more effort and prompt actions are required to reduce MPs levels in UK indoor microenvironment to protect our population, particularly vulnerable groups like toddlers, from potential risks associated with exposure to MPs indoors.

Dedication

I dedicate this thesis with profound love and gratitude to my dear parents, beloved wife, beautiful daughters, supportive siblings, and loyal friends.

To my parents, your unwavering support, encouragement, motivational words, and wise advice have been the guiding light throughout this journey. This achievement is a testament to your influence. I hope you live long enough to see the fruits of the seeds you planted with so much love and care.

To my wife, your role as a remarkable mother to our daughters, a devoted wife, and a trusted partner has been the driving force behind my success. Your presence in my life empowered me to complete this degree.

To my sweet daughters, I hope this achievement fills you with pride and joy.

To my brothers and friends, words cannot express my gratitude and appreciation for your support throughout this journey, especially to the friends I met in Birmingham. Thank you sincerely for believing in me.

Acknowledgement

First and foremost, I would like to express my deepest gratitude to Allah, the Almighty, for granting me the strength, knowledge, and perseverance to complete this thesis. Without His blessings, none of this would have been possible.

I am profoundly grateful to my supervisors, Dr. Mohamed Abdallah Abou-Elwafa and Prof. Stuart Harrad, for their invaluable guidance and support throughout my doctoral research. Their insightful feedback and encouragement have been instrumental in shaping this thesis.

I would like to acknowledge the financial support provided by the Saudi Ministry of Education and the Royal Embassy of Saudi Arabia Cultural Bureau in London, UK, which enabled me to conduct my research effectively.

I owe an immense debt of gratitude to my beloved parents and family. Their belief in my abilities, love, and encouragement sustained me through the challenges of this PhD journey. I deeply appreciate their sacrifices and understanding.

I extend my heartfelt thanks to my devoted wife and sweet daughters for their patience and understanding throughout my absence on this long journey. My wife's love has been my greatest source of strength, motivation, and perseverance.

Many thanks to my colleagues in the Public Health Building and Molecular Science Building: Dr. Jingxi Jin, Dr. Misbah Alghamdi, Dr. Muneera Al-Mansoori, Dr. Yulong Ma, Dr. Oddny Ragnarsdottir, and Dr. Simeon Onoja, for their invaluable assistance with samples, data collection, and lab work.

Lastly, special thanks to my friend Dr. Abdullah Albalawi for assisting me with the data analysis of this thesis using the SPSS software program.

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Chapter 1

Large parts of this chapter are taken verbatim from the following published article: Ageel, H.K., Harrad, S., Abdallah, M.A.E. (2021) Occurrence, human exposure, and risk of microplastics in the indoor environment, Environ. Sci.: Processes Impacts, 24, 17 (1). 154 Citations, one policy document, 3 news articles.

1.1 Introduction:

Plastic contamination is considered a global threat to human and environmental health. The substantial increase in the worldwide annual production of plastics from 2 million tonnes in 1950 to over 300 million tonnes in 2018 has led to high levels of environmental plastic pollution (Hale et al., 2020). This is mainly due to the ability of plastics to accumulate in various environmental media owing to their resistance to chemical and/or biological degradation (Barnes et al., 2009a). Several types of polymers have been identified as the main sources of plastic pollution in different environmental compartments, including: polypropylene (PP), polyethylene (PE), polyethylene terephthalate (PET), polystyrene (PS), polyurethane (PUR), polyvinyl chloride (PVC), and polycarbonate (PC) (Li et al., 2016). While larger plastic waste items present an evident visible risk to the environment and biota, there is increasing concern over the environmental and health impacts of smaller plastic fragments. Based on size, plastics can be classified into five major categories: “Nanoplastic” (< 0.03 cm), "Microplastic" (< 0.5 cm), "Mesoplastic" (0.5 – 5 cm), "Macroplastic" (5 – 50 cm), and "Megaplastic" (> 50 cm) (Lebreton et al., 2018). Figure 1.1 displays the classification of plastic sizes (Wu et al., 2019b).

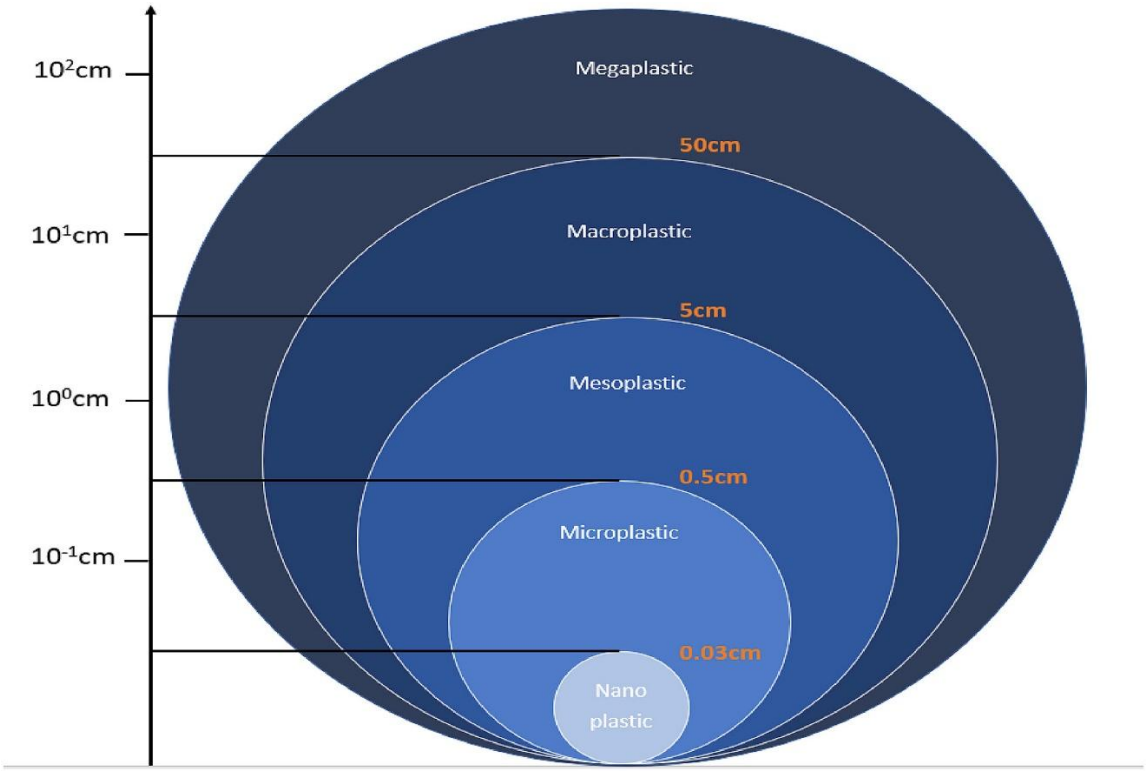


Figure 1.1 The classification of plastic sizes (Wu et al., 2019b)

Thompson (2006) was the first author to use the term "Microplastics (MPs)" for plastic particles such as fragments, pellets and fibres (Thompson, 2006). MPs is inclusive of all types of plastic polymers in different shapes with a longest dimension < 5 mm, including nanoplastics, which are <0.3 mm (Wu et al., 2019a). To further distinguish MPs from nanoplastics; Frias and Nash (2019) defined MPs as "synthetic solid particles or polymeric matrices, with regular or irregular shape and with size ranging from 1 µm to 5 mm, of either primary or secondary manufacturing origin, which are insoluble in water". Accordingly, MPs can have various shapes, such as, pellets, films, foam, fragments, and microbeads. Also, MPs can be classified into "primary" and "secondary" based on their origin. Primary MPs are those manufactured or released to the environment in the "micro" size. This includes microbeads used in cosmetics, microfibres shed from clothing and other textiles upon washing, as well as micronized particles emitted from the wear of rubber tyres (Browne et al., 2011a). Secondary MPs are produced from the breakdown of larger plastics in the environment as a result of

natural weathering processes including photo-, chemical-, and biological degradation (Andrady, 2011).

MPs have become emerging pollutants of increasing global concern due to their ubiquitous detection in several environmental compartments including air, water, soil, sediment, and biota (Wu et al., 2019c; Yao et al., 2019a; SAPEA, 2019a). However, most studies on MPs so far are centering on characterizing their occurrence, fate, and impact in the aquatic environment. Studies that have focused on MPs in terrestrial outdoor and indoor environments are limited compared to those in the marine/freshwater environment (Wright et al., 2013a; Gall and Thompson, 2015a). Therefore, very little is known about the magnitude, patterns, and associated risks of human exposure to MPs in the terrestrial environment, particularly indoors.

Plastics were long considered inert materials due to the covalent-bonded polymer structure that renders them of high molecular weight, size, and hydrophobicity; thereby unavailable for absorption and readily excreted unchanged. However, recent studies revealed that MPs (particularly those < 1 mm) have the potential to cause adverse health effects to living organisms by: obstruction, inflammation, and/or accumulation in organs after translocation (Wright et al., 2013a; Jiang et al., 2020; Wang et al., 2016). Moreover, there is further concern over the potential release of hazardous plastic chemical additives to the environment and biota upon exposure to MPs containing such additives. These chemicals are added to plastic polymers at varying concentrations (up to 70% by mass) to impart some desired physical (e.g. colour, strength) or performance properties (e.g. flame-retardancy, flexibility) (Hahladakis et al., 2018a). However, several classes of these plastic additives can cause serious adverse health effects (e.g. phthalates, bisphenols, organophosphates, biocides), as well as persistence and bioaccumulative properties (e.g. brominated flame retardants, chlorinated paraffins). This potential risk is aggravated by the high surface area to mass ratios of MPs, which may facilitate

the leaching of these chemicals out of the plastic polymer upon human exposure to these particles (Wu et al., 2019b; SAPEA, 2019a; Jiang et al., 2020a).

1.2 Background on Microplastics in the Environment:

There are several sources of MPs to environmental compartments, including the marine ecosystem, outdoor & indoor environments, soil, sediment, and air. The marine ecosystem is the environmental compartment most studied for MPs pollution, due to a multitude of input sources including the direct discharge of municipal waste, industrial discharge, and wastewater treatment plants (Cole et al., 2011; Alomar et al., 2016). Figure 1.2 shows MPs Sources, transport, accumulations, and fate in the different environmental media (Wu et al., 2019c).

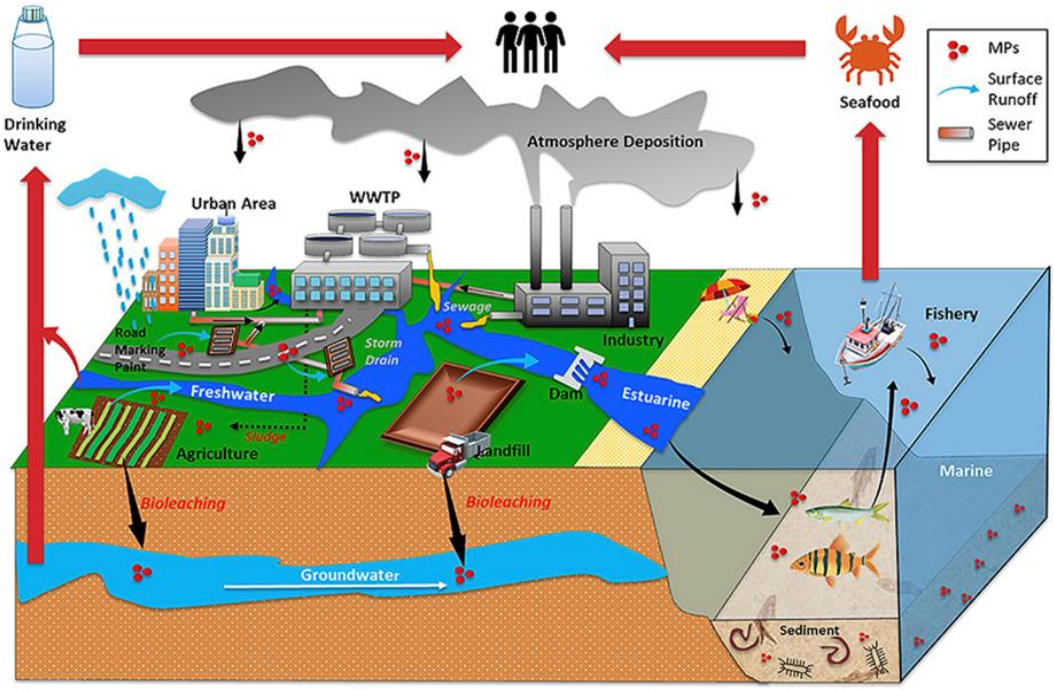


Figure 1.2 MPs sources, transport, accumulations, and fate in the environmental media (Wu et al., 2019c)

There are two major types of MPs, which are primary and secondary MPs (Waller et al., 2017). Primary MPs are manufactured in this “micro” size, to be incorporated in different products such as cleaning materials and cosmetics. Also, this definition extends to include MPs that enter the environment in the “micro” size range, such as microfibres originating from the

abrasion of synthetic textiles through washing yet entering the aquatic environment as microfibres. On the other hand, secondary MPs are derived from the break down of larger plastic objects into smaller plastic particles in the micro size range, due to degradation, abrasion, exposure to UV light or disintegration (Lassen et al., 2015).

There are several major input sources of MPs to the outdoor environment. Synthetic textiles have been identified as source of MPs to the aquatic environment. A study reported that item of clothing might be accountable for release around 1900 fibers per wash of a one blanket/fleece or several types of clothes, equating to >100 fibers/L of collected laundry effluent (Browne et al., 2011b; Vandermeersch et al., 2015). Another paper estimated that each blanket release around 1.1×10^5 fibers per wash (Carney Almroth et al., 2018). According to Napper and Thompson (2016), the overall emissions from laundry may release approximately 7×10^5 fibres per 6 kg load. Another investigation on MPs fibers release into air from garments made of synthetic textiles reported that MPs microfibres are released at range from 1 to 403 fibres per gram of textile with a length between 494 and 1034 μm (Cai et al., 2020).

Emission through plastic recycling processes, and from construction/demolition locations are sources of airborne MPs, which may affect the atmosphere (Huang et al., 2013; Hahladakis et al., 2018a). Also, traffic density may lead to the release of automotive tires wear particles contributing substantially to MPs in the atmosphere and road dust (Councell et al., 2004).

Several studies showed that the use of plastic films or mulching is a significant source of MPs to agricultural soils (Kyrikou and Briassoulis, 2007). Application of biosolid from wastewater treatments plants (WWTPs) to agricultural soils has been identified as another major source of MPs to soil (Wu et al., 2019c).

There are various potentials sources for MPs to the aquatic environment, such as, wastewater treatments plants (WWTPs) effluents to surface waters, run off from agricultural farms and storm water, discharge from domestic washing machines containing synthetic microfibres, industrial discharge and seeping leachates from landfills. According to Eunomia (2016), the total amount of secondary MPs released to the aquatic environment was estimated at 68,500–275,000 tons per year. This huge amount may be divided into coastal emission of 54,300–145,000 tons per year; inland emission of 500–20,000 tons per year and marine emission of 13,700–1,110,000 tons per year.

The small particle size and low density of MPs facilitate their distribution and transport in the environment. Therefore, when MPs particles enter the ecosystem, they will be transported among the environmental media, for example, from land to freshwater or/and from freshwater to marine environment, with different habitation periods in each compartment. For instance, the transportation from land to freshwater systems will rely on meteorological conditions, like wind speed and wind direction, in addition the distance between the sites and land cover type (Lambert et al., 2014).

The first worldwide detection of MPs was in aquatic habitats including open ocean, lakes, rivers, sea ice and estuaries (SAPEA, 2019a). MPs might be entering the aquatic environment from many sources such as harbours and shipyards, wastewater treatment plants, coastal tourism, urban runoff, and rivers (Stolte et al., 2015; Woodall et al., 2014). Also, the fate of MPs in marine environment including beach and bottom sediments is still unclear until now. While sediments are able to accumulate MPs, the relationship between MPs abundance and sediment grain size remains unclear as well (Browne et al., 2010; Alomar et al., 2016).

Several investigations suggested that the high concentrations of synthetic MPs, such as polyethylene (PE) and polystyrene (PS) over the short-term exposure may pose an important

risk to freshwater environment (Phuong et al., 2016; Wagner et al., n.d.). Furthermore, other studies expressed concern about the fate of MPs through the long-term exposure to the aquatic ecosystem. Subsequently, it could lead to bioaccumulation for submicron particles with wider implications of marine environment health generally (Farrell and Nelson, 2013; Setälä et al., 2014).

1.3 Microplastics in the indoor environments:

The indoor environment is important to modern human life due to the large proportion of time (up to 90% or more) that people spend indoors (e.g., homes, offices, transport…etc.) (Klepeis et al., 2001). Subsequently, indoor environmental quality has substantial implications for human health and wellbeing. This is further exacerbated by the various lockdown measures taken by several countries around the world to limit the spread of Covid-19, which may contribute to elevated exposure to emerging contaminants, such as MPs, indoors.

The following sections will summarise the current state-of-knowledge on the concentrations of MPs in environmental media relevant to indoor exposure. This includes indoor environment media (e.g. dust and air), as well as other exposure media relevant for indoor exposure (e.g. food and drinking water).

1.3.1 Settled dust:

Indoor dust is considered an important medium for human exposure to several pollutants such as, construction materials, painting products, microorganisms, heavy metals such as lead, and several organic chemical pollutants (e.g. flame retardants and plasticisers) (Mercier et al., 2011; Tan et al., 2016). The occurrence of MPs at various concentrations, shapes, and sizes in indoor dust has been recently reported. A study on three different indoor sites in Paris including two private apartments and one office at the University of Paris-Est-Creteil pointed out that the number of MPs fibres in dust sampled from vacuum cleaner bags ranged from 1 to 60 fibres/m^3.

These concentrations exceeded those detected in outdoor dust (0.3 to 1.5 fibres/m^3), indicating higher potential for human exposure to MPs via indoor than outdoor dust (Dris et al., 2017c). In a worldwide study on MPs in house dust from 12 different countries including China, Colombia, Greece, Japan, Kuwait, and Saudi Arabia. MPs were detected in all 286 dust samples, at concentrations of 38–120,000 μg/g (median: 5900 μg/g) (Zhang et al., 2020a).

A recent study investigated the presence of MPs in settled indoor dust collected from two offices, two schools, and two apartments in Surabaya, Indonesia. MP numbers measured in the two offices were: 334 particles on weekdays, 242 particles on a weekend (office 1); and 351 particles on weekdays, and 252 particles at the weekend (office 2). In schools they were: 290 particles on weekdays, and 239 particles at the weekend (school 1); and 321 particles on weekday, and 257 particles at the weekend (school 2), while in the apartments there were: 133 particles on weekdays, and 127 particles at the weekend (apartment 1); and 108 particles on weekdays, and 95 particles at the weekend (apartment 2). Despite the small sample size, it was evident that the number of MPs collected at each location during weekdays exceeded that found at weekends. This was attributed to the greater occupancy levels of the studied indoor environments on weekdays, which contribute to increasing MPs numbers through increased human activity (Bahrina et al., 2020).

Peng et al., (2020) developed a method to study nylon MPs (polyamide, PA6 and PA66) in different environment media including indoor dust, sludge, marine sediment, freshwater sediment, and fishery sediment in China. The results revealed that PA6 and PA66 MPs in indoor dust were 0.431−86.3 and 3.10−92.9 mg/kg, respectively. The authors also observed that the total concentrations of PA6 and PA66 MPs in indoor dust exceeded those in the other environmental matrices studied such as sludge, marine sediment, and freshwater sediment. Table 1.1 summarizes the current knowledge on the presence of MPs in settled indoor and outdoor dust.

Table 1.1 MPs in settled indoor and outdoor dust.

References	Samples number		Concentrations		Dominant size range (µm)	Polymer types	Shape
			Average	Range			
Soltani et al., (2021a)	32	Houses	3095 fibres/m^2/day	22 to 6169	200 to 400	PET, PA, PP, Cellophane	Fiber
Abbasi et al., (2022)	50	Schools	3771 MPs/g	81 to 55830	500 to 1000	PET, PP, PS	Fiber
Kashfi et al., (2022)	30	6 Houses	71.3 items/mg	27 to 112	>1000	PE 42%, PC 28%, PP 18%, PET 10%, Nylon 2%	Fiber, fragment, film
		6 Mosques	113 items/mg	64 to 157			
		6 Kindergartens	128 items/mg	93 to 196			
		6 Hospitals	60.3 items/mg	41 to 75			
		6 Universities	56.1 items/mg	41 to 94			
Nematollahi et al., (2022)	28	Schools	195 MPs/g	9.8 to 634.5	50 to 1000	PET 50%, PP 35%, PS 10%, Nylon 5%	Fiber, sheet, fragment
Zhang et al., (2023)	60	20 Farmland	20.3 mg/kg	1.17 to 80.45	100 to 5000	PE, PP, PET	Film, fiber, foam, fragment
		20 Forestland	19.5 mg/kg	0.55 to 65.51			
		10 Desert	8.1 mg/kg	0.22 to 30.25			
		10 Construction land	28.6 mg/kg	1.08 to 48.21			
Zhu et al., (2022)	242	47 Residential apartments	1174 MPs/g	150 to 3861	201 to 1000	PE, PP, polyester, PA, PC, PS	Fiber, fragment, film
		53 Hotels	843 MPs/g	74 to 2065			
		50 Offices	896 MPs/g	112 to 1470			
		48 University dormitories	775 MPs/g	99 to 1166			
		44 University classrooms	209 MPs/g	62 to 434			
Peng et al., (2023)	39	Houses	207,000 items/g	Not given	50 to 100	PU, PE, PVC, PP, PET	Fiber
Rabin et al., (2023)	144	36 Industrial areas	17.3 MPs/g	Not given	150 to 250	PE, PP, PET, PA, PS, PAA, Nylon	Fragment, line, fiber, film
		36 Commercial areas	16.8 MPs/g				
		36 Public Facilities	16.2 MPs/g				
		36 Residential areas	14 MPs/g				
Fan et al., (2024)	75	urban road dust in the megacity, Nanjing	143.3 particles/m^2	Not given	< 500	PS, PA, PVDF, Cellophane,	Fragment, fiber, film

						PVC, PP, PAE, PE, PTFE	
Valdiviezo-Gonzales et al., (2024)	60	Houses	3 MPs/mg	0.01 to 33.9	500 to 1000	PET, PP, Cellulose	Fiber

- (PP): polypropylene, (PVC): polyvinyl chloride, (PA): polyamide, (PE): polyethylene, (PET): polyethylene terephthalate, (PS): polystyrene, (PPS): polyphenylene sulfide, (PAA): polyacrylic acid, (PAN) Polyacrylonitrile, (PU): Polyurethane, (PPA): Polyphthalamide, (PES): Polyethersulfone, (PC): Polycarbonate, (PVDF): Polyvinylidene fluoride, (PTFE): Polytetrafluoroethylene.

1.3.2 Air:

Generally, there is limited data on the levels and types of MPs in air compared to other environmental media (Liu et al., 2019c). The presence of MPs in indoor air was attributed to emission and distribution from various sources including, furniture, municipal litter, and construction materials (Dris et al., 2016). Likewise, (Webster et al., 2009) identified several sources of MPs to indoor air such as, textiles (mats, clothing, curtains, mattresses), toys, rubber, kitchen items (plates, cups, utensils, bowls, bottles), electrical cables, electronics, indoor paint, and cleaning agents (Webster et al., 2009).

Dris et al., (2017a) measured atmospheric MPs concentrations at three different indoor sites including two private apartments and one office in Paris. The results showed the concentrations of indoor air fibres in the studied sites ranged from 4.0–59.4 fibres/m^3. The highest concentrations were observed in the office (4.0-59.4 fibres/m^3), followed by apartment A (2.5-18.2 fibres/m^3) and finally apartment B (1.1-16.3 fibres/m^3). The variation was attributed to differences in emission of MPs fibres from indoor sources like furniture and carpets as a result of daily usage and/or cleaning activities. Concentrations of MPs in indoor air from all 3 sites exceeded those measured in outdoor air (0.3–1.5 fibres/m^3). This was explained by the existence of indoor sources of the MPs fibres detected, as well as greater atmospheric dilution of MPs outdoors compared to indoors (Dris et al., 2017a). Moreover, the rate of MPs deposition from air (also known as atmospheric fall out) in different indoor sites was reported to range from 1586 to 11,130 particles m^{-2} day^{-1} (Dris et al., 2017a; Catarino et al., 2018a). These rates are substantially higher than those reported outdoors for a European metropolitan region (137−512 particles m^{-2} day^{-1}) (Klein and Fischer, 2019) and a remote conservation area in the United States (132 particles m^{-2} day^{-1}) (Brahney et al., 2020).

Zhang et al., (2020d) investigated concentrations of MPs in indoor air at three different indoor microenvironments, namely: dormitory room, office, and a lecture building at East

China Normal University. The results showed the highest average MPs abundance to be in the dormitory (9.9×10^3 MPs m^{-2} day^{-1}), followed by the office (1.8×10^3 MPs m^{-2} day^{-1}) then the corridor (1.5×10^3 MPs m^{-2} day^{-1}). The measured MPs existed mostly in the form of fibres, which had similar polymer compositions to the textile products used in the studied microenvironments. This led to the conclusion that textile quantity in an indoor microenvironment is one of the main factors affecting MPs abundance in indoor air, whereas high airflow turbulence can increase abrasion of textile fibres and MPs migration in indoor environments (Zhang et al., 2020e).

Another investigation of airborne MPs both indoors and outdoors in coastal California, showed higher concentrations of MPs in indoor air (3.3 ± 2.9 fibres and 12.6 ± 8.0 fragments m^{-3}) than in outdoor air (0.6 ± 0.6 fibres and 5.6 ± 3.2 fragments m^{-3}). Also, the results showed that indoor MP fragments (58.6 ± 55 μm) were smaller in size than outdoor MP fragments (104.8 ± 64.9 μm). The authors concluded that the risk of inhalation exposure to MPs indoors is higher than outdoors due to increased abundance and smaller size of MPs in indoor air (Gaston et al., 2020).

A recent investigation of indoor air MPs from 20 houses in Hull, UK revealed an average household MPs concentration of 1414 MPs m^{-2} day^{-1} over a 6 month monitoring period. MP fibres (5–250 μm) were the most abundant shape of MPs (90%), while PET accounted for 62% of the identified MPs. Moreover, results suggested substantially higher human exposure to MPs (1-45 times) via inhalation indoors than outdoors (Jenner et al., 2021a). Table 1.2 summarizes the current knowledge on the presence of MPs in indoor and outdoor air.

Table 1.2 MPs in indoor and outdoor air.

References	Samples number		Concentrations		Dominant size range (µm)	Polymer types	Shape
			Average	Range			
Jenner et al., (2021b)	118	Houses	1414 MPs/m^2	0 to 5412	5 to 250	PET, PA, PP, PE	Fiber, fragment, film, sphere, foam
Liao et al., (2021)	39	Apartments	1583 n/m^3	51 to 2887	5 to 4665	Polyester, PA, PP	Fragment, fiber
		Offices					
		Classrooms					
		Hospitals					
		Transit station					
Chen et al., (2022)	20	8 Living rooms	7.05×10^2 items/cm^2	1.47 to 21.4 × 10^2	5 to 5000	PET, Polyester, PP, PPA, Rayon, Cellophane	Fiber
		8 Student dormitories	8.66×10^2 items/cm^2				
		4 Offices	19.3×10^2 items/cm^2				
Perera et al., (2022)	18	7 Indoor sites	0.53 particles/m^3	0.13 to 0.93	100 to 300	PET, polyester, PA, PP, PS	Fiber, fragment
		11 Outdoor sites	0.11 particles/m^3	0.01 to 0.23			
Torres-Agullo et al., (2022)	15	6 Houses	4.8 MPs/m^3	Not given	43 to 4436	PA 51%, PES 48%, PP 1%	Fiber, fragment
		3 Buses	17.3 MPs/m^3				
		3 Subway car	5.8 MPs/m^3				
		3 IDAEA-CSIC Institute	4.2 MPs/m^3				
Xie et al., (2022)	232	75 Houses	62.6 particles/m^3	15.56 to 93.32	2.4 to 2181.5	PE 73.8%, Polyester 9.2%, Phenolic Resin 9.1%, PVC 3.1%, PP 0.6%, PU 0.2%	Fiber, fragment, bead
		157 Offices	27.6 particles/m^3				
Uddin et al., (2022)	25	11 Public buildings	8.9 MPs/m^3	3.2 to 27.1	0.45 to 2800	Polyester, nylon, PA	Fiber, fragment
		6 Houses	15.7 MPs/m^3				
		2 Mosques	14.3 MPs/m^3				
		6 Hospitals	4.2 MPs/m^3				

Yao et al., (2022)	Not given	Houses	1.96×10^4 fiber/m^2/day	Not given	35 to 1000	PS, PET, PE, PVC, PP	Fiber, film, fragment
		Hallway	1.44×10^4 fiber/m^2/day				
		Classrooms	6.20×10^3 fiber/m^2/day				
		Offices	6.43×10^3 fiber/m^2/day				
(Al-Hussayni et al., 2023)	270	Kindergartens	20.2 MPs/m^3	2.7 to 20.2	Not given	PS 40%, PET 25%, PE 14%, PP 15%, PA 5%, PVC 1%	Fiber, fragment
		Mosques	13.4 MPs/m^3				
		Schools	12.1 MPs/m^3				
		Shops	11.3 MPs/m^3				
		Hospitals	10.6 MPs/m^3				
		Offices	9.9 MPs/m^3				
		Dormitory	9.6 MPs/m^3				
		Barber salons	7.7 MPs/m^3				
		Cafeterias	6.8 MPs/m^3				
		Scientific laboratories	5.2 MPs/m^3				
		Pharmacies	2.9 MPs/m^3				
		Medical clinics	2.7 MPs/m^3				
Perera et al., (2023)	21	3 Childcare centre	2.25 particles/m^3	0.20 to 2.25	20 to 5000	PET 59%, PE 25%, PA 10%, PP 3%, PAN 2%, PVC 1%	Fiber, fragment
		3 Elder's home	0.87 particles/m^3				
		3 School	1.03 particles/m^3				
		3 House	0.47 particles/m^3				
		3 Office	1.20 particles/m^3				
		3 Vehicle	0.20 particles/m^3				
		3 Restaurant	0.70 particles/m^3				

- (PP): polypropylene, (PVC): polyvinyl chloride, (PA): polyamide, (PE): polyethylene, (PET): polyethylene terephthalate, (PS): polystyrene, (PPS): polyphenylene sulfide, (PAA): polyacrylic acid, (PAN) Polyacrylonitrile, (PU): Polyurethane, (PPA): Polyphthalamide, (PES): Polyethersulfone.

1.3.3 Drinking water:

MPs were detected in both tap water and bottled water (Kosuth et al., 2018; Mason et al., 2018a). One of the potential reasons for presence of MPs in drinking water is the possible erosion and/or degradation of the plastic components of the distribution networks (e.g., pipes, tubing) leading to fragments of MPs in drinking water (Mintenig et al., 2019). Also, using plastic bottles and caps as packaging for drinking water and potential degradation through the processes of manufacture, distribution, marketing, and use might be an important source of MPs in drinking water. Table 1.3 summarizes the current knowledge on the presence of MPs in drinking water.

Table 1.3 MPs in drinking water.

References	Samples number		Concentrations MPs/L		Detection frequency (%)	Polymer types	Shape
			Average	Range			
Kankanige and Babel, (2020)	105	10 spring water brands	140	Not given	100%	PET 28.4%, PE 24.2%, PP 18.1%, PA 7.2%, PVC 4.4%	fibres and fragments
		95 tap water bottles	52				
Zhang et al., (2020c)	7	Private households	0.7	0.3 to 1.6	100%	Rayon, PET, PE, PS, Polyester, PAA, PMPS, PI	fibres and fragments
Tong et al., (2020)	38	Private households	440	0 to 1247	95%	PE 26.8%, PP 24.4%, co PE-PP 22.0%, PPS 7.3%, PS 6.5%, PET 3.3%	fragments, fibres, and spheres
Mintenig et al., (2019)	24	9 Raw water	0.0007	0– 0.0007	42%	Polyester 62%, PVC 14%, PA and epoxy resin 9%, PE 6%	fragments
		15 Drinking water (supply chain)					
Zuccarello et al., (2019)	20	10 Mineral still	5.42 X 10^7	3.16X10^7 to 1.1X10^8	100%	Not given	Not given
		10 Sparkling					
Mason et al., (2018b)	259	253 plastic bottles	10.4[a], 315	0-14	93%	PP 54%, Nylon 16%	fragments, fibres, and films
		6 glass bottles					
Pivokonsky et al., (2018)	36 from WTPs[b]	12 WTP1	443	Not given	100%	PET 44%, PP	fragments, fibres, and spheres
		12 WTP2	338			PET 62%, PP	
		12 WTP3	628			PET 26%, PP, PE 24%	
Strand et al., (2018)	17	9 private households	< 0.58	Not given	24%	PP 50%, PS 25%, PET 25%	fragments
		3 private workplaces					

		5 private or public institutions					
Schymanski et al., (2018)	38 from mineral water	15 returnable plastic bottles	118	28-241	100%	PET 84%, PP 7%, PE 5%, PA 2%	fragments
		11 single use plastic bottles	14	2–44			
		3 beverage cartons	11	5-20			
		9 glass bottles	50	4-156			
Wiesheu et al., (2016)	1	1 mineral water brand	1 in the sample[c]	Not given	Cannot confirm contamination	PET	fibres

- (a): only particles ≥100 µm were verified with FTIR.
- (b): Water Treatment Plant.
- (c): only fibers counted.
- (PP): polypropylene, (PVC): polyvinyl chloride, (PA): polyamide (nylon), (PE): polyethylene, (PET): polyethylene terephthalate, (PS): polystyrene, (PTT): poly trim ethylene terephthalate, (PPS): polyphenylene sulfide, (PAA): polyacrylic acid, (PMPS): poly (methyl phenyl siloxane), (PI): poly (isoprene).

1.3.4 Food:

Fish and seafood are considered major sources of MPs in the human diet because of the relatively early detection of different types of MPs in various types of fish, shrimp, shellfish, and bivalve (Rummel et al., 2016a; Devriese et al., 2015a; Van Cauwenberghe et al., 2015a). Figure 1.3 shows the main potential pathways for the transport of MPs to human through the food chain in the marine environment (Wright et al., 2013b).

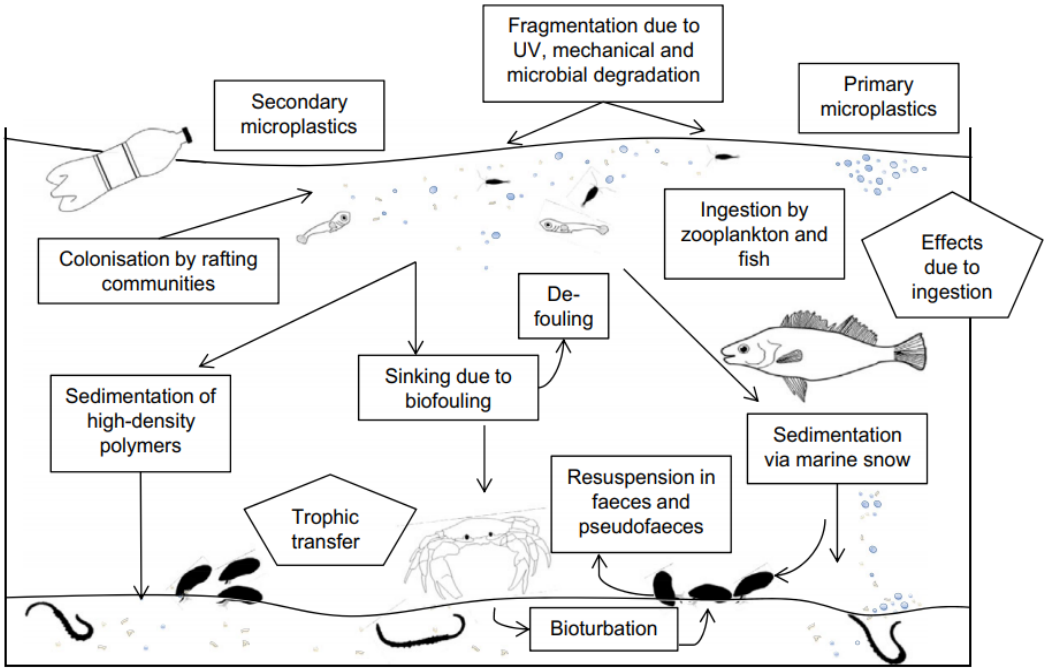


Figure 1.3 The main potential pathway for the transport of MPs to human during the food chain from marine organisms organisms (Wright et al., 2013b).

In addition, recent studies have reported the presence of MPs in different types of food and beverages such as sugar, honey, milk, table salt, soft drinks, and beer (Cox et al., 2019a; Diaz-Basantes et al., 2020a). The extensive use of plastic materials in food packaging may be a reason for the presence of MPs in packaged food (Schymanski et al., 2018b; Hernandez et al., 2019a). The extent of MPs release from food wraps/packages to packaged food remains unknown, although it is high based on the findings that one plastic tea bag can release millions of MPs into on cup of the hot beverage (Hernandez et al., 2019b). Moreover, the potential for ingesting MPs depositing from the atmosphere onto the surface of food items cannot be neglected as it has been estimated to lead to exposure in the range 1.4 – 6.8 × 10^4

MPs/person/year (Catarino et al., 2018b). Table 1.4 summarizes the current knowledge on the presence of MPs in food items.

Table 1.4 MPs in food and beverages.

References	Food type	Samples number	MPs average content	Polymer types	Size	Shape
Ghosh et al., (2021)	Fish	100	2.2 ± 0.89 MPs per individual	PET 55%, PP 33%, PEST 6%, PU 2%	< 500 µm	fibres, films, fragments, foams, and granules
Taghizadeh Rahmat Abadi et al., (2021)	Kutum fish	44	11.4 MPs items per fish (0.015 items per 1 g fish wet weight)	Not given	< 500 µm	fibres, fragments, and beads
Zakeri et al., (2020)	Fish	111	2.29 MPs particles/Fish	Not given	0.5 -1 mm to 2-4.75 mm	fibres, fragments, film, and beads
Diaz-Basantes et al., (2020b)	Milk	10	137 fibres/L 204 fragments/L	HDPE, LDPE 44%, PAAm 30%, PP 26%		fragments and fibres
Hernandez et al., (2019b)	Tea	4	~ 11.6 MPs/cup of the beverage	PA, PET	25 µm	N/A
Fang et al., (2019)	Bivalves	30	1.26 ± 0.39 to 1.56 ± 0.33 MPs items per individual^{-1}	PP, PA, PE, PS, PET, PVC, PAN	0.1 - 16.4 mm	fibres, fragments, and films
Baalkhuyur et al., (2018)	Red sea fishes	178	26 MPs particles found in fish stomach per species	PP 42%, PET 42%, PVC 8%, PS 4%, PAN 4%	2.39 ± 0.28 mm	fibres, fragments, and films
Karami et al., (2018)	Canned sardines and sprats	20	6 particles/item	PP 33.3%, PET 33.3%, PE 16.6%, PVC 16.6%	190 – 3800 µm	fragments, filaments, and films
Kim et al., (2018)	table salts	39	0−1674 particles /kg in sea salts 0−148 particles /kg in rock salt 28−462 particles /kg in lake salt	PP, PE, PS, PET, PVC,PA	100−5000 µm	fragments, fibres, sheets, and spherules

Li et al., (2018)	Mussels (Mytilus edulis)	162	0.7 - 2.9 MPs items/g of tissue (wet weight)	PP, PEST, PET	0.005 – 5 mm	fibres, fragments, spheres, and flakes
Naji et al., (2018)	Molluscs	123	0.2 - 21.0 MPs particles per g of soft tissue (wet weight)	PE, PET, PA	10 – 5000 µm	fragments, films, and pellets
Renzi et al., (2018)	Mussels	20	6.2–7.2 MPs items/g of tissue	Not given	1150–2290 µm	filaments
Renzi and Blašković, (2018)	Marine table salts	6 Italian brands 5 Croatian brands	1.57 – 8.23 items/g (Italy) 27.13 – 31.68 items/g(Croatia)	Not given	4–2100 µm (Italian salts) 15–4628 µm (Croatian salts)	fibres, films, fragments, granules, and foams
Bellas et al., (2016)	72 dogfish 12 hakes 128 red mullets	212 fishes	red mullets 1.75 ± 1.14 MPs practices per individual dogfish 1.20 ± 0.45 MPs practices per individual	Not given	0.38 - 3.1 mm	fibres, spheres, films, and fragments
Rummel et al., (2016b)	Fish	16	1–7 particles/fish	PE 40%, PA 22%, PP 13%, PS 10%, PET 5%, PU 5%, PEST 5%.		Not given
Tanaka and Takada, (2016)	Planktivorous fish	64	2.3 pieces on average and up to 15 pieces per individual	PE 52%, PP 43.3%	150 – 1000 µm	fragments and beads
Li et al., (2015)	Bivalves	9	2.1 - 10.5 particles/g	PE, PET, PA	5 – 5000 µm	fibres, fragments, and pellets
Devriese et al., (2015b)	Shrimp	165	0.68 ± 0.55 MPs/g wet weight	Not given		fibres, film, spherule, and fragment
Rochman et al., (2015)	Fish	76	0–21 particles per individual fish	Not given	> 500 µm	fragments, foam, and film
Van Cauwenberghe et al., (2015b)	Blue mussels (Mytilus edulis)	36	0.36 ± 0.07 items/g	Not given	5-25 µm	Not given
Yang et al., (2015)	Table salt	15	550–681 particles/kg in sea salts, 43–364 particles/kg in lake salts, and 7–204 particles/kg in rock/well salts.	PET, PE, CP		fragments and fibres

De Witte et al., (2014)	Blue mussel (Mytilus edulis)	30	2.6 - 5.1 fibres/10 g of mussel	Not given	200 - 1500 µm	fibres
(Liebezeit and Liebezeit, 2014)	Beer	24	0.025 fibres/mL 0.033 fragments/mL 0.017 granules/mL	N/A		fragments, fibres, and granules
Liebezeit and Liebezeit, (2013)	Honey	19	166 ± 147 fibres/kg 9 ± 9 fragments/kg	N/A	10 – 20 µm	fragments and fibres
Liebezeit and Liebezeit, (2013)	Sugar	5	217 ± 123 fibres/kg 32 ± 7 fragments/kg	N/A	10 – 20 µm	fragments and fibres

• (PP): polypropylene, (PA): polyamide (nylon), (PE): polyethylene, (PET): polyethylene terephthalate, (PS): polystyrene, (PEST): polyester, (CP): cellophane, (HDPE): high-density polyethylene, (LDPE): high-density polyethylene, (PAAm): polyacrylamide, (PU): Polyurethane, (PVC): polyvinyl chloride, (PS): polystyrene, (PAN): Polyacrylonitrile, N/A: not reported.

1.4 Human exposure to microplastics:

Currently, little is known about human exposure to MPs, especially indoors. This may be attributed to several reasons including the lack of standardized methods for quantitative analysis of MPs in different matrices relevant to human exposure (e.g., indoor dust, air, diet, water and personal care products). This is compounded by the reporting of results as number of MPs in the majority of the few available studies, while most exposure assessment models require the exposure doses in mass units. Nevertheless, current understanding is that human exposure to MPs occurs via a combination of inhalation (air), ingestion (food, drinks, dust), and dermal contact (dust, fabrics, cosmetics) (Wu et al., 2019c; Revel et al., 2018a). Figure 1.4 provides a schematic representation of the different pathways of human exposure to MPs and the risks associated with such exposure.

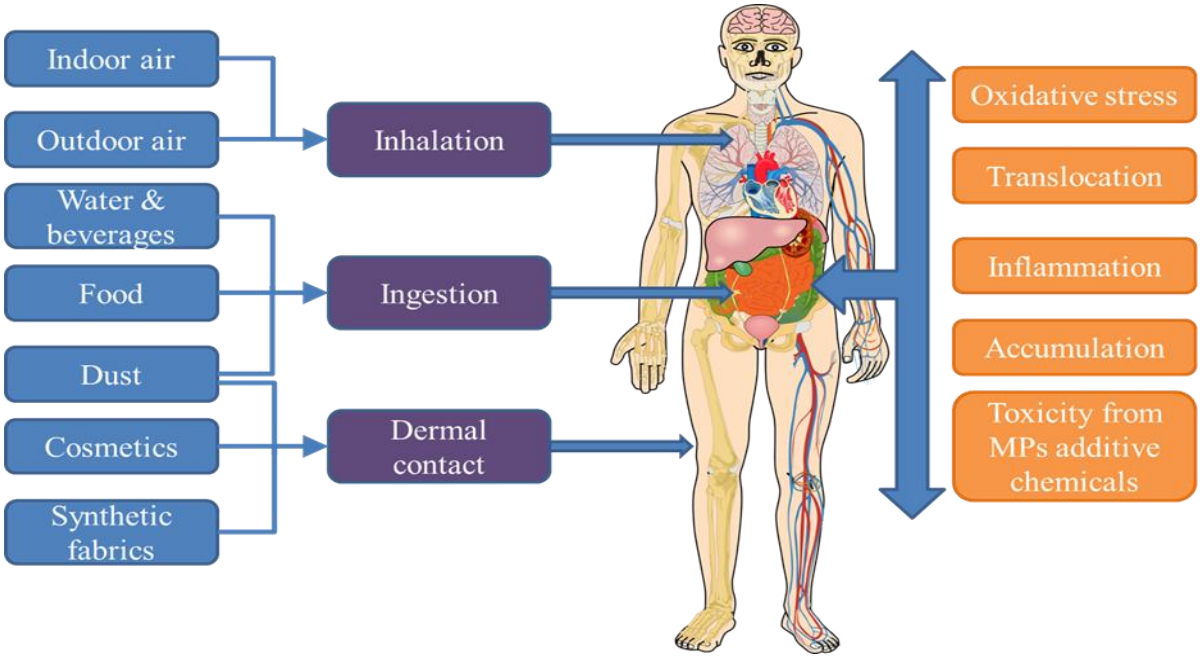


Figure 1.4 Pathways of human exposure to MPs and associated risks.

1.4.1 Inhalation:

Inhalation has recently emerged as an important route of human exposure to MPs; due to their consistent detection at considerable concentrations in both indoor and outdoor air (Zhang et al., 2020d; Liu et al., 2019a). Cox et al., (2019a) highlighted the significance of the inhalation

pathway through a meta-analysis of 26 studies on human exposure to MPs with a particular focus on the American population. Using an average concentration of 9.8 MPs/m^3, results revealed adult inhalation exposure of 170 and 132 MPs/day for males and females, respectively, while children were exposed to 110 and 97 MPs/day. These estimates constituted ~50% or more of the total daily exposure via all routes in the studied population groups indicating inhalation is the major pathway of human exposure to MPs (Cox et al., 2019b). A more recent study of MPs in Australian homes reported a mean inhalation intake of 0.2 mg/kg bw/year (equivalent to 12891 MPs fibres/year) with the highest intake in young children at 0.31 mg/kg bw/year (Soltani et al., 2021b). Domenech and Marcos estimated a relatively low global human daily inhalation intake of 5.9 MPs/day based on average global airborne MPs concentration of 0.685 particles/m^3 and breathing rate of 8.64 m^3/day (Domenech and Marcos, 2021a). In earlier studies, individual MPs inhalation exposure was estimated to be between 26 and 132 MPs/day (Prata, 2018a), while (Vianello et al., 2019a) reported mean inhalation of 272 MPs/day for average male adults doing light activity (Vianello et al., 2019a). Interestingly, a recent study highlighted the increased risk of MPs inhalation due to wearing different types of face masks during the COVID-19 pandemic. Fibres and spheres were the most abundant MP types detected, while activated carbon masks and N95 masks produced the highest and lowest risk of MPS inhalation, respectively (Li et al., 2021).

It is worth mentioning that meaningful comparison between the few available MPs inhalation exposure studies is difficult due to the different sampling, analysis and exposure assessment techniques. Such variation extends to the units for reporting concentrations of airborne MPs. Some studies report concentrations as number of MPs per m^2 (measured atmospheric fallout area), while others report number of MPs per m^3 (volume of air samples) (Huang et al., 2020). Therefore, development of standardised methods for analysis of MPs in air is crucial for accurate and comparable exposure assessment results.

Currently, very little is known about inhalation exposure to MPs via resuspension of settled indoor dust. Several studies have provided experimental evidence on the contribution of resuspended house dust to inhalable airborne particulate matter. Resuspension of indoor dust occurs when the previously settled particles are detached from surfaces (e.g. carpets, floor) to become re-entrained into indoor air by human activities such as walking, crawling and vacuuming (Hyytiäinen et al., 2018a; Wang et al., 2021). Interestingly, toddlers were reported to experience higher inhalation exposure to contaminants in resuspended dust particles via crawling, compared to adult exposure via walking (Hyytiäinen et al., 2018b). Several studies have assessed the inhalation intake of various resuspended particulate-phase contaminants in indoor dust (e.g. PM_{10}, $PM_{2.5}$, bacteria) (Mitro et al., 2016; Wu et al., 2018). Resuspension of house dust was found to be equivalent to a $PM_{2.5}$ source strength ranging from 0.03 to 0.5 mg/min. Moreover, resuspension from walking on carpets was reported as the most significant contributor to both $PM_{2.5}$ and PM_{10} exposure in the personal cloud (i.e. personal breathing zone) of retirement centre inhabitants (Rasmussen et al., 2018; Rodes et al., 2001). While MPs particles are expected to behave similarly, there exists no data on inhalation exposure to MPs via resuspension of settled indoor dust. This represents a significant knowledge gap suggesting that current reports of human inhalation exposure to MPs may be underestimated, particularly in toddlers.

1.4.2 Ingestion:

Until the last couple of years, ingestion was widely considered as the main route of human exposure to MPs (Galloway, 2015). This was led by few early studies that identified MPs in various food items, particularly fish and seafood (De Witte et al., 2014b; Van Cauwenberghe and Janssen, 2014). Moreover, evidence has now emerged of potential human exposure to MPs via inadvertent ingestion of indoor dust (Zhang et al., 2020b).

1.4.2.1 Diet:

Several studies have identified MPs at varying concentrations in different food items including fish, seafood, table salt, sugar, honey, milk and beer (Table 1.3). This will inevitably lead to human exposure to these MPs via diet. However, few studies have estimated such dietary exposure. According to (Cox et al., 2019b) the daily consumption of MPs via Americans male adults, female adults, male children, and female children to be 113, 142, 106, and 126 particles per day, respectively. Another study estimated European adult exposure to MPs via consumption of table salt to range between 37 to 100 MPs person^{-1} year^{-1} (Karami et al., 2017). Elsewhere, the measured concentrations of MPs in fish and shellfish samples from Iran could contribute to an average daily adult exposure of 5 particles per day (Abbasi et al., 2018).

The current state-of-knowledge on dietary exposure to MPs is far from comprehensive due to the lack of knowledge on MPs in major food groups (e.g., meat, vegetables, oil, dairy products etc.). This research gap was highlighted in a report by SAPEA (Science Advice for Policy by European Academies), which called for more comprehensive dietary studies for better characterization of the risk associated with human dietary exposure to MPs (SAPEA, 2019b). Another pertinent factor that requires further investigation is the release of MPs from plastic packaging to food and beverages which may result in elevated exposure. A study on hot beverages raised concern by reporting that brewing one plastic teabag at 95 °C releases approximately 11.6 billion MPs into a single cup of the beverage (Hernandez et al., 2019b). Although the methods and techniques applied in this study leading to such high estimate of MPs was questioned (Busse et al., 2020), the potential release of MPs from plastic teabags has been confirmed. Another alarming study showed that polypropylene infant feeding bottles may release MPs with values as high as 16 million particles per liter due to repeated sterilization and exposure to high-temperature water. The resulting exposure estimates ranged from 14,600–

4,550,000 particles per capita per day, which demonstrated that infant exposure to MPs may be higher than was previously recognized (Li et al., 2020). A recent study provided evidence on the contamination of meat products by MPs from the packaging polystyrene tray at concentrations of 4 - 18.7 MPs/kg of packaged meat. These particles were difficult to remove by rinsing and may probably be cooked before consumption (Kedzierski et al., 2020). Similarly, low concentrations of MPs (1-14 MPs/L) were detected in branded milk packages from Mexico, likely as a result of leaching from the thermoplastic sulfone membrane filters used during the manufacturing process (Kutralam-Muniasamy et al., 2020). Moreover, mechanical stress was identified as a factor that may influence human exposure to MPs via bottled drinks (Schymanski et al., 2018a). Likewise, Winkler et al. reported the frequent opening/closing of plastic bottles increases the number of MPs released into the bottled water via degradation; thereby increasing the chances of ingesting MPs (Winkler et al., 2019).

Collectively, these studies provide clear evidence on human exposure to MPs via diet in different countries and in different age groups from nursing infants to adults. There is even evidence on prenatal exposure to MPs via the placenta (Ragusa et al., 2021a). However, more comprehensive, large scale studies are required for further understanding of the magnitude, profiles and associated risk of human dietary exposure to MPs.

1.4.2.1 Dust:

The role of indoor dust as an important matrix for human exposure to various groups of hazardous chemicals has been extensively highlighted over the past 2 decades. This has spurred some recent studies to investigate potential human exposure to MPs via unintentional ingestion of indoor dust. A study on polyethylene terephthalate (PET) and polycarbonate (PC) MPs in both indoor and outdoor dust samples from 39 cities in China reported the estimated daily intake (EDI) for different age groups at 6,500−89,700 ng per kg bw per day. Children were the highest exposed age group with a mean EDI of 17,300 ng per kg bw per day, while

mean EDI of PET MPs in teenagers and adults were 7270 and 6500 ng per kg bw per day, respectively. Comparison between indoor and outdoor dust through EDI of PET MPs in all the studied age groups revealed higher exposure (almost double) via indoor dust (Liu et al., 2019b). The ubiquity of human exposure to MPs via indoor dust, combined with higher intake and subsequent risk in younger age groups was confirmed by another study of MPs in house dust from 12 different countries. High concentrations of MPs (38–120,000 μg/g) were detected; resulting in median EDI values of 4000–150,000 ng per kg bw per day for infants. Adults were less exposed with median EDI values of 360–12,000 ng per kg bw per day, which was mainly attributed to a combination of higher dust ingestion rate and lower body weight in infants (Zhang et al., 2020b). Similar findings were reported by a recent study on MPs in indoor dust from 32 Australian homes with estimated mean ingestion rates of 6.1 mg per kg bw per year (EDI = 16,712 ng per kg bw per day) for children and 0.5 mg/kg bw/year (EDI = 1,370 ng per kg bw per day) for adults (Soltani et al., 2021b).

Despite the small number of studies, it can be concluded that human exposure to MPs via ingestion of indoor dust and the potential risk of such exposure cannot be neglected, especially for children. More research is needed to understand the spatial and temporal variability of such exposure and the impact of individuals' time-activity pattern (e.g., the percent of time spent in homes, offices, outdoors, cars, schools and other types of microenvironments) on the overall daily exposure to MPs via dust ingestion in different age groups.

1.4.3 Dermal contact:

To date, there are no available studies assessing human dermal exposure to MPs and its associated risks. However, with the ubiquitous occurrence of MPs in indoor dust, atmospheric fallout from both indoor and outdoor air, as well as wide application of microbeads in cosmetics and continuous degradation of microfibers from textiles; it's reasonable to consider dermal

contact as a pathway of human exposure to MPs (Domenech and Marcos, 2021a). Microplastic beads (microbeads, generally < 1mm in diameter) have been widely applied in dermal exfoliation and cleansing products, as well as in toothpaste and denture fillings (Anagnosti et al., 2021). Few studies have attempted estimation of per capita consumption of microbeads using specific personal care products. A study on UK facial scrubs from different brands revealed a MPs content of 10 – 100 g /l resulting in per capita consumption of 40.5 – 215 mg/ day (Napper et al., 2015). Gouin et al. estimated an average consumption of 2.4 mg MPs/day per capita for the US population using liquid soap (Gouin et al., 2011).

While these few studies fall short of providing a comprehensive understanding on human dermal exposure to MPs, they provide evidence that the dermal route cannot be ignored. Although human skin can act as an efficient barrier against penetration of large particles and studies suggest that only particles < 100 nm (i.e. nanoplastics) can directly cross the dermal barrier (Revel et al., 2018b), there are other possible routes for transdermal penetration of larger particles through hair follicles, sweat gland or open skin injuries (Schneider et al., 2009). Moreover, skin damage as a result of inflammation and oxidative stress has been associated with dermal exposure to MPs (Wright and Kelly, 2017). Therefore, more research is required to assess human dermal exposure to MPs via contact with cosmetics, settled dust particles, fabric fibers, as well as evaluating the significance of this exposure pathway and the associated health risk.

1.5 Microplastics risk to human health:

Toxicological understanding of the potential impacts of MPs exposure on human health is still in its infancy (SAPEA, 2019a). This may be attributed in part to the lack of accurate, sensitive and standardized analytical methods for determination of MPs in different human tissues (e.g. blood, milk) and exposure-relevant matrices (e.g. food, air). On the other hand,

very little is known about toxicological endpoints of MPs and how they relate to environmentally-relevant human exposure doses (Rubio et al., 2020). It is also not clear if the sizes, shapes, and chemical composition of the wide variety of environmental MPs available for human exposure via different pathways will impact on their toxicity in humans (Amereh et al., 2020). Nevertheless, human internal exposure to MPs has been recently confirmed through the detection of MPs in human stool (Schwabl et al., 2019) and placenta (Ragusa et al., 2021b). This has spurred more research into the potential toxic implications of human exposure to MPs. Current understanding is that adverse health effects from MPs may be associated with particle toxicity and/or toxicity from associated chemicals (e.g. plasticisers, flame retardants, colourants etc.) (Shi et al., 2022; Vethaak and Legler, 2021).

1.5.1 MPs particle toxicity:

Evidence on the toxicological impacts of airborne MPs in human is sparse. It is widely conceived those MPs fibers < 20 µm are predominant in air (SAPEA, 2019a; Rahman et al., 2021). Therefore, entry of these MPS into the airway is possible, although this has not been quantified yet. Larger MPs might be deposited on the airway or trapped by the lung lining fluid and cleared out. However, smaller MPs may be engulfed by macrophages and epithelial cells (Geiser et al., 2005). Accumulation of MPs fibers in human pulmonary tissue has been previously reported (Pauly et al., 1998). Such accumulation of inhaled MPs in human airways may cause interstitial lung inflammatory and/or immune responses, and even cancer, although no large-scale epidemiological associations or toxicological endpoints have been estimated for these adverse effects in humans (Huang et al., 2021). Moreover, Small MPs (<2.5 - 5µm) were reported to be capable of entering the systemic circulation through endocytosis and can be translocated to mesenteric lymph nodes, blood circulation, liver and spleen (Urban et al., 2000). Increased permeability of the epithelial membrane due to inflammation was suggested as a plausible mechanism for this translocation of small MPs (Prata et al., 2020a). This was

supported by the observed increased hemolysis and subsequent release of histamine, a pro-inflammatory chemical, from human cell lines exposed to polypropylene MPs (20 -200 µm) (Hwang et al., 2019). Another potential toxicological impact may be caused by inhalation of MPs carrying microbial colonization. Pathogenic Vibrio Spp. colonies were identified on a number of MPs particles (e.g. polyethylene, polypropylene and polystyrene) from North/Baltic sea (Kirstein et al., 2016). In addition to the risk of pathogenic species infections, inhaled MPs with microbial colonization could cause a shift in the microbial community structure in the lung (SAPEA, 2019b).

Similar to inhaled MPs, current understanding is that intestinal absorption of ingested MPs is generally low with 90% or more of ingested particles excreted in faeces, depending on the particle size and shape (Smith et al., 2018). While an early study showed the mucus layer in the gut to present an effective barrier against the diffusion of 500 nm latex microbeads (Bajka et al., 2015), more recent studies in human cell lines and lab animals indicate that MPs < 10 µm can cross the gastrointestinal barrier and be translocated via the blood circulation to other organs (e.g. liver and kidney) (Banerjee and Shelver, 2021).

Controlled laboratory studies in human cell lines and laboratory animals have revealed several potential toxic implications as a result of MPs exposure. Cytotoxicity of MPs on cerebral and epithelial human cell lines was attributed to oxidative stress caused by increased reactive oxygen species to high concentrations (Schirinzi et al., 2017). This should be interpreted carefully though because in other human cell lines (Caco-2), no significant cytotoxic effects were observed upon exposure to polystyrene MPs, even at high exposure doses (Stock et al., 2019; Hesler et al., 2019). Other studies investigating the toxicity of MPs as particulate matter in air and water have reported on a range of potential toxicological impacts in animal models including immunosuppression and links to autoimmune diseases such as lupus erythematosus and autoimmune rheumatic disease (Prata et al., 2020a). Other studies suggested possible

neurotoxic effects due to increased activity of acetylcholinesterase enzyme in the brain and altered serum neurotransmitters levels in fish and mice (Barboza et al., 2018; Deng et al., 2017a). Exposure to MPs has been linked to obesity based on the increased food intake in mice as a response to higher energy demands or lower absorption efficiency associated with an observed decrease in liver weight and size (Deng et al., 2017b). Prata, (2018) suggested that chronic exposure to airborne MPs might be linked with cancer due to DNA damage caused by chronic inflammation and interstitial irritation.

It is worth noting that extrapolating results from these controlled laboratory animal studies to humans should be done carefully and interpreted with caution due to inter-species variability, more complex and sophisticated immune and nervous systems, as well as the differences in MPs exposure levels and pathways in humans.

1.5.2 Toxicity from associated chemicals:

A wide range of chemical additives are often incorporated into plastic polymers during manufacture at typical concentrations around 20% by weight (but can be up to 70% w/w) to impart specific properties or enhance the physical characteristics/appearance of the final product (Hahladakis et al., 2018b). Table 1.5 shows the common chemical groups used as plastic additives.

Table 1.5 Common chemical groups used as plastic additives.

Category	Group	Examples
Functional additives	Plasticizers	Phthalates, bisphenols, organophosphate esters
	Flame retardants	Brominated flame retardants, organophosphate flame retardants, chlorinated paraffins
	Stabilizers	Nonylphenols, cadmium and lead compounds.
	Biocides	Triclosan, Triclocarban
	Anti-static agents	Quaternary ammonium compounds, alkylsulfonates, and alkylphosphates.
	Slip agents	Metallic stearates, fatty acid amides and waxes.
Colourants	Soluble pigments	Azo dyes, Anthraquinones
	Organic pigments	Cobalt diacetate
	Inorganic pigment	Cadmium, zinc and chromium salts.

Reinforcements	Fibres	Carbon fibres, glass fibres.
Fillers		Zinc oxide, talk, clay, calcium carbonate.

Most of these additive chemicals (Table 1.5), particularly in the flame retardant and plasticizer groups, have been well-documented to cause adverse health effects in humans including: endocrine disruption, reproductive toxicity, neurotoxicity, hepatotoxicity and cancer (Jiang et al., 2020; Rubio et al., 2020; Rahman et al., 2021). In the case of MPs, the concern over exposure to these toxic chemical additives is exacerbated by the small particle size and large surface area of MPs. This is likely to allow more surface area for these additive chemicals to leach out from the plastic polymer to human body fluids (e.g. sweat, gastric, intestinal and lung fluids) upon exposure (i.e. higher bio-accessibility) (Prata et al., 2020a). While the plethora of evidence on the potential adverse effects associated with exposure to MPs additive chemicals is concerning, very little is known about the leaching potential of these different chemical groups from various polymer types and consequently their potential toxicological impacts in humans.

Another toxicological concern over MPs and associated chemicals is the so called "Trojan Horse effect", which refers to the potential for accumulation of hazardous environmental chemical pollutants on the surface of MPs (aided by their large surface area) and this will lead to an increase in the chemical toxicity within the organism as the chemicals are then released when the plastic is ingested and or inhaled (Trevisan et al., 2020a; Ramsperger et al., 2020). While recent studies have demonstrated the potential for increased toxicity of polycyclic aromatic hydrocarbons (PAHs) in zebrafish (Trevisan et al., 2020b) and heavy metals in Daphnia (Abdolahpur Monikh et al., 2020), the Trojan Horse theory still needs verification in humans and higher trophic level animals. It is also possible that adsorption of chemical pollutants to MPs can reduce their bio-accessibility in organisms, through a reduced

concentration in biological fluids, if the chemical co-pollutant is strongly bound to the MP and not desorbed easily, as reported in Northern Fulmars (Herzke et al., 2016).

1.6 Research gaps and future perspectives:

The extensive research on the identification and characterisation of MPs over the past decade has established their ubiquitous distribution in all environmental matrices. While the majority of this research was focused in the aquatic environment, few recent studies have raised concern over the existence of MPs in the indoor environment at concentrations relevant to human exposure. Current understanding is that human exposure to MPs in the indoor environment can occur through a combination of inhalation, ingestion, and dermal contact. The detection of MPs in indoor air and atmospheric deposition samples indicate inhalation as a significant exposure route, while the presence of MPs in indoor dust, food items, and beverages renders exposure via ingestion inevitable. Moreover, the use of MPs in cosmetics and personal care products, as well their release via the degradation of microfibres from domestic fabrics means the dermal exposure pathway cannot be neglected. This is alarming due to the fact that people spend up to 90% or more of their time indoors, recently exacerbated by COVID-19 pandemic lockdown measures. Critical evaluation of existing literature in the present review has revealed the paucity of information on human exposure to MPs indoors and the risk associated with it. The increasing public concern over MPs pollution and its potential detrimental effects on human health and the environment is escalating the pressure on regulators and policy makers to take decisive actions to mitigate such harmful effects. The knowledge base required for such informed decisions to protect public health against MPs pollution is still lacking. Significant research gaps exist in the current knowledge on both the sources and magnitude of human exposure to MPs, as well as the relative contribution of different exposure pathways to human body burdens of MPs. The few studies available on the

indoor environment mainly originate from developed countries, while very little is known about human exposure to MPs in low and middle-income countries (LMIC) and the impact of different climates (e.g. less time spent indoors in tropical and subtropical regions) and cultures on such exposure (e.g. consumption of plastic packaged takeaway food and beverages). Therefore, the following topics are recommended for prioritisation in MPs research:

(a) There is an urgent need for validated, standardised methods for quantifying concentrations of MPs in indoor air and dust samples. It is also essential to reach a consensus on the units for reporting these concentrations on a mass basis suitable for exposure assessment models. Of particular importance is the harmonisation of units used for reporting airborne concentrations of MPs. The discrepancy between studies reporting concentrations as MPs per m^2 (measured atmospheric fallout area) and those reporting MPs per m^3 (volume of air samples) in the existing literature is hindering comparability between various studies. Moreover, the use of atmospheric fallout data (MPs per unit area) to assess human inhalational exposure is challenging because existing exposure models are based on inhalation rates in volume (m^3) of air.

(b) Despite the few studies reporting MPs concentrations in human exposure-relevant media (e.g. air, dust and diet), very little is known about the magnitude of this exposure under real-life scenarios and for different age/sex groups. Therefore, comprehensive human exposure assessment studies are crucially required to estimate individual- and population- level exposure to MPs via total diet studies, market basket studies, dermal contact scenarios, and/or time-activity patterns (i.e. time spent in homes, offices, schools, nurseries, cars, outdoor exercise…etc) for different age groups in various countries.

(c) There is paucity of data on some sources and pathways that may contribute substantially to human exposure to MPs. More research is required to assess the resuspension of MPs

particles from settled indoor dust to the personal cloud of adults and toddlers; thereby assessing its contribution to human inhalation exposure of MPs. Studies should also address the factors influencing this exposure source, such as: personal activity (e.g. walking adult, crawling toddler), indoor features (e.g. carpeted vs. wooden floors), rate of vacuuming, temperature and ventilation.

(d) While few recent studies assessed human exposure to MPs via food items (mainly seafood), very little is known on the contribution of food packaging and food-contact materials on human intake of MPs. Future studies should investigate the contribution of packaging (e.g. plastic boxes, films, covers, plates and utensils) on MPs concentrations in food and subsequent intake by different age groups in different cultures. Similarly, more research is required to understand the contribution of plastic bottles and caps to MPs exposure via drinking and the impact of beverage composition (e.g. probiotic drinks, multivitamin drinks) and temperature (e.g. hot coffee vs cold juice) on the leaching of MPs from the bottle and consequent human exposure via drinking these beverages.

(e) Risk is assessed based on knowledge of both exposure and hazard. Recent studies have proposed risk assessment approaches for MPS in humans including probabilistic lifetime exposure models (Mohamed Nor et al., 2021) and multi-compartment exposure frameworks (Noventa et al., 2021). However, more information on the toxicological implications and endpoints of MPs in humans (i.e. hazard) is essential to assess the risk arising from their exposure. In particular, more research is required on the particle-induced toxicity of MPs in human at exposure-relevant concentrations and MPs particle size ranges. Similarly, studies on the potential leaching and bioavailability of toxic chemical additives from human ingested/inhaled MPs are urgently required. These studies should preferably be conducted in human cell-lines and tissues, or pertinent mammalian laboratory animals due to the large inter-

species variation that precludes meaningful extrapolation of toxicological endpoints from species like zebrafish and daphnia to humans.

1.7 Research Aim and Objectives:

The central hypothesis to be examined by this research is that MPs are present in indoor air and dust at concentrations leading to human exposure at levels that may be detrimental to human health. The main aim of this study is investigate human exposure to MPs in the indoor environment and assess the risk arising from such exposure. This will be addressed via determination of MPs occurrence, concentrations, and characteristics (i.e., size, shape, and polymer type) in indoor air and dust from several microenvironments in Birmingham, UK; as well as identifying potential seasonal variation in the measured indoor MPs concentration via a 12-month monitoring period in 5 indoor microenvironments (2 homes and 3 offices).

The objectives of this thesis are thus to:

(a) Develop and optimise accurate methods for sampling, quantifying and characterizing MPs in indoor air and dust samples.
(b) Apply the optimised methods to quantify and characterize MPs in air samples from several homes and workplaces (30 sites in each group), using both active and passive air sampling techniques in each site.
(c) Quantify and characterize MPs in indoor dust samples from different homes and workplaces across Birmingham, UK, 30 sites in each group.
(d) Assess human exposure to MPs in the UK indoor environment via inhalation and dust ingestion and identify potential factors influencing such exposure and the risk arising from it.
(e) Investigate potential seasonal variation in concentration of MPs in indoor air and dust and explore potential implications for human exposure and risk assessment.

Chapter 2

2.1 Methodology:

Indoor air and dust samples were collected and analysed for MPs to examine the hypotheses outlined in the first chapter, section 1.7. Both air and dust samples were collected from 30 homes and 30 workplaces. Two techniques, active and passive, were used for indoor air sampling. The analytical methodology for extracting and quantifying MPs consists of several distinct steps (sampling, preparation, extraction, and identification/quantification). It is challenging to detect airborne MPs indoors or outdoors. Currently, there is no standard procedure for MPs analysis in indoor environments. In addition, various analytical approaches have been employed depending on their traits, but no standardized method has been devised for their identification (Ebere et al., 2019; Bhat et al., 2023). Generally, MPs estimation is performed in two steps: physical characterization of potential plastics, such as microscopy, to identify the microplastic's shape and size, then chemical characterization, such as spectroscopy, to confirm plastic identity and determine the polymer types (Shim et al., 2017).

This chapter discusses the materials and methods used to collect indoor samples and quantify MPs in detail. It also presents the validation parameters of the applied analytical procedures and the quality assurance/ quality control measures employed to guarantee the reliability of generated data.

2.1.1 Sampling:

Sampling was conducted using the best available techniques and suitable methods for MPs sampling in different environmental media. MPs sampling should be performed using protocols that are extremely protective of contamination from the point of collection through to extraction and analysis.

2.1.1.1 Sampling sites:

The chosen sites for sample collection comprise two types of microenvironments: houses and workplaces. The sampling sites were randomly selected and constitute a sample of convenience for the research team, with enabled access during the required sampling times/durations. For the purpose of this thesis, private houses from several residential buildings and various workplaces, including offices, meeting rooms, and lecture rooms in Birmingham City, UK, were sampled throughout the period of May 2022 to September 2023. The ethics review number is (ERN_21-1117). 30 houses and 30 workplaces were sampled for indoor air (active and passive) and dust, providing a total of 60 passive air samples, 60 active air samples and 60 indoor dust samples.

To study seasonal variation in indoor concentrations of MPs, a subset of five microenvironments were chosen to collect indoor samples (airborne, atmospheric deposition, and settled dust) every month from January 2023 to December 2023. The total number of samples generated is 180. The selected sites included three offices from different buildings and two residential flats.

2.1.1.2 Air sampling:

In this thesis, the low-volume active air sampler comprised a low-volume pump (Capex L2X, Charles Austen, UK), operated at a flow rate of 6 L min^{-1} for 24 hours to yield a single sample comprising 8.64 m^3 air. The particulate phase was collected by employing a 47 mm diameter membrane glass fibre filter (1.0 μm pore size, Whatman, UK) housed in a stainless steel in-line 47 mm filter holder (Sartorius® 16254, Goettingen Germany) (Figure 2.1). The designated flow rate of 6 L min^{-1} was achieved and maintained by using a flow meter (Platon 12 L min^{-1}) connected to an adjustable valve. The flow meter was calibrated at the start and

the end of each experiment using a Gilibrator® air flow calibrator (Sensidyne, Inc., Clearwater, Florida, USA), which is classified as a primary standard device.

The passive air sampler was used to capture indoor airborne microplastics fall-out (atmospheric deposition), by applying an open glass Petri dish (20 cm diameter, Duran® soda-lime glass, VWR International ltd., Leicestershire, England) placed at head height (1–1.5 m) within the selected sites. The glass Petri dishes were only opened on site at a set date and time. In each sampling session, the Petri dishes were exposed to atmospheric fallout for 24 hours to collect dry and wet MPs atmospheric deposition fallout. (Figure 2.2), and participants were instructed to not change any of their habits or routines. At the end, the petri dish was closed with its specific glass lid, wrapped in clean aluminium foil, and secured with a paper-tape, then transferred to the laboratory for further analysis by the primary researcher. In the laboratory, Petri dishes were stored in a cool (5°C), dark place until sample processing was conducted.



Figure 2.1 The Active sampler

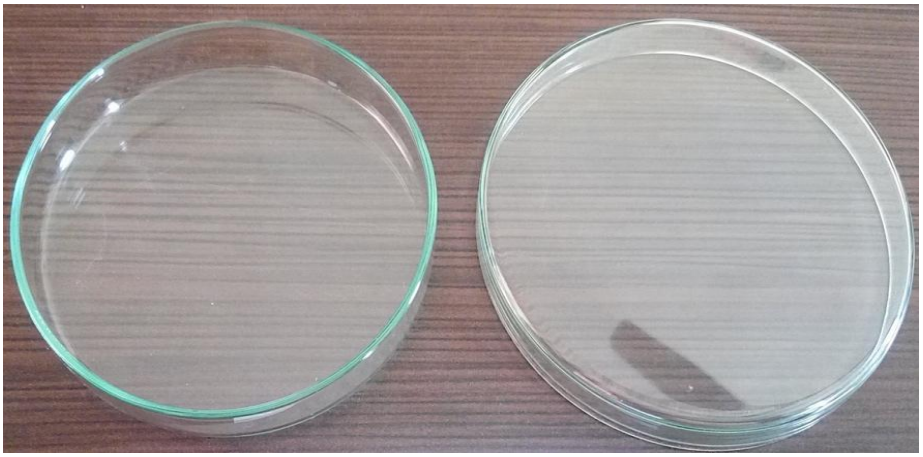


Figure 2.2 The passive sampler

2.1.1.3 Settled dust sampling:

In this study, the tools used to collect samples was a battery-powered, cordless vacuum cleaners (Karcher HV 1/1 BP CS 18v) with a specific design metallic tube attachment. Glass microfiber standard extraction thimbles (single thickness 25 mm × 100 mm, 1.0 μm pore size, Whatman, UK) were placed inside the vacuum tube to collect the dust samples and to avoid any chance of cross-contamination. Figure 2.3. The sampling session took exactly 15 minutes at each site. After collecting the samples, the thimble was removed carefully and emptied into a 47 mm glass Petri dish (BRAND®, Merck, UK) with the help of a disposable ECOFriendly® plastic-free brush with natural bristles, then moved directly to a clean Petri dish. The petri dish was closed with its specific glass lid, wrapped in clean aluminum foil, and secured with a paper-tape, then transferred to the laboratory for further analysis by the primary researcher. In the laboratory, Petri dishes were stored in a cool (5°C), dark place until sample processing was conducted.



Figure 2.3 Cordless vacuum cleaner for dust sampling

2.1.2 MPs extraction from dust samples:

In this study, extraction of MPs from indoor dust samples was adapted from the method developed at our UoB laboratories and described by Nel et al., (2019) to reduce sample contamination from the extraction process via the application of Zinc Chloride salt for density separation using the in-house built "Sediment Microplastic Isolation (SMI) units" (Figure 2.4).

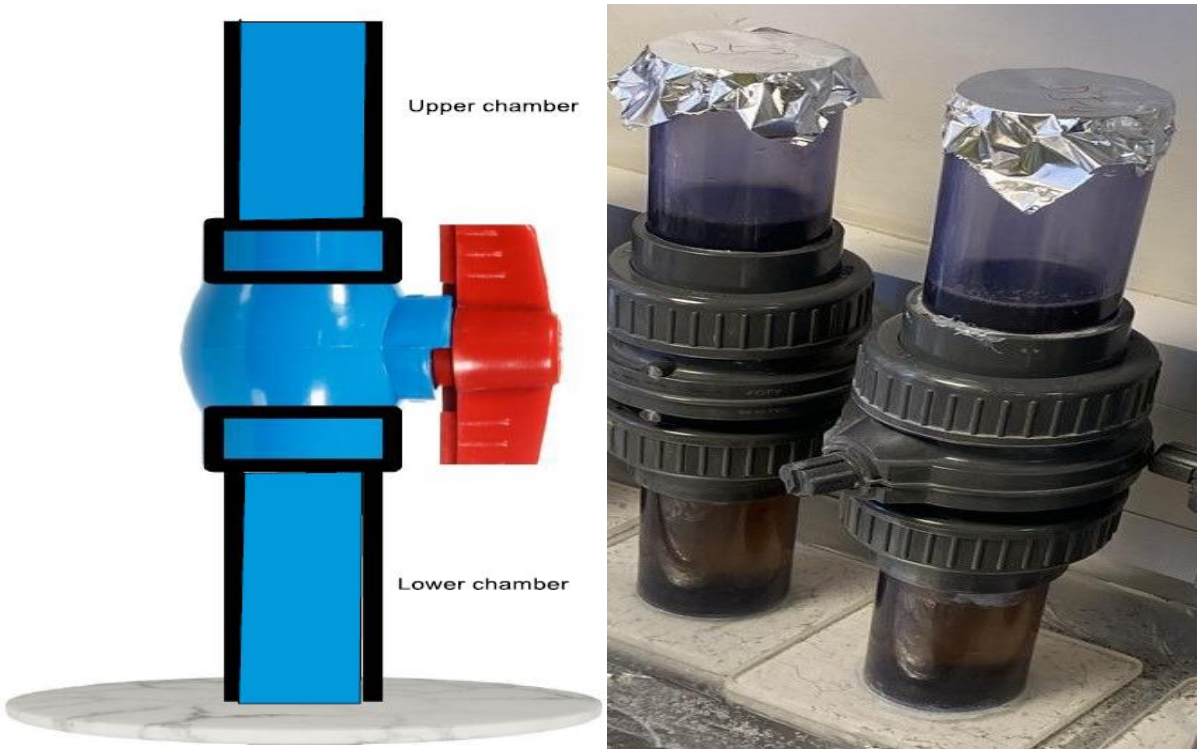


Figure 2.4 Sediment Microplastic Isolation (SMI) unit.

Before applying the extraction method, the collected samples were weighed on an accurate balance (LE324S, Sartorius, Germany) (Figure 2.5), and the weight of dust was recorded. The collected dust was then sieved using a stainless-steel sieve (width 125 µm, diameter 100 mm, VWR International Ltd) (Figure 2.6) and weighed again to record the size fraction relevant for human exposure via indoor dust ingestion (USEPA, 2011).



Figure 2.5 The first weight of the dust sample.



Figure 2.6 Stainless steel sieve (125 µm).



Figure 2.7 The final weight of the sieved dust sample.

2.1.2.1 Density separation:

The chemical reagent used in the density separation process is Zinc Chloride ($ZnCl_2$) (VWR International Ltd). Before starting the process, the chemical reagent must be prepared. For preparing zinc chloride (concentration = 700 g/L), 700 g of the powdered salt were accurately weighted on the sartorious balance and added to 1 litre of deionised Milli-Q water (18 MΩ.cm), then stirred using a magnetic stirrer (JENWAY, 1002 Stirrer) (figure 2.8) for 24 hours at room temperature to guarantee complete dissolution.

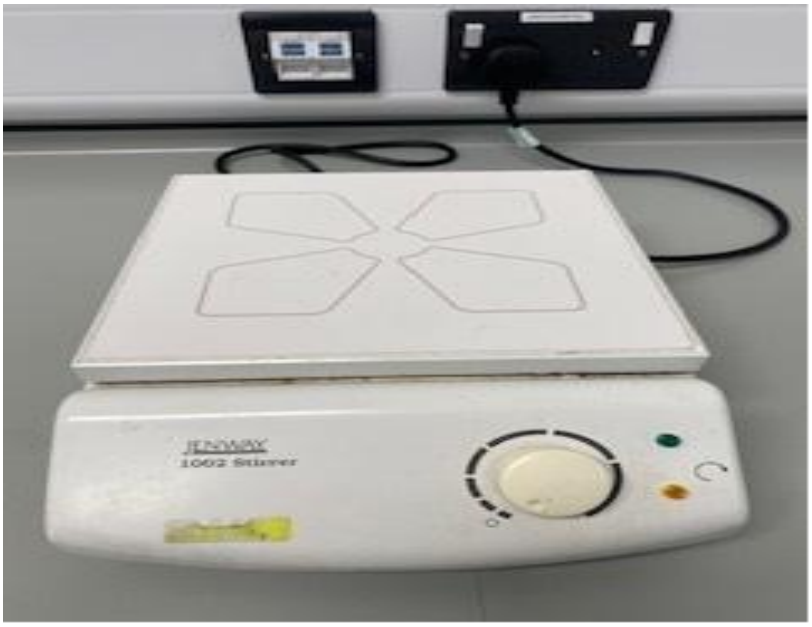


Figure 2.8 The magnetic stirring.

The sieved dust sample was added to the SMI unit then 50 mL of the prepared $ZnCl_2$ solution (700 g/L) were added. The SMI unit was then covered with a clean aluminium foil and placed on a magnetic stirrer and allowed to stir for 10 minutes at a rotation frequency of 5 to 8 revolutions per minute (rpm). Thereafter, the unit was left to settle for 12 hours. Once the denser particles had settled out, the supernatant was poured over a 25 μm stainless steel mesh into a beaker for filtration and re-use. The upper chamber of the SMI unit was also removed and rinsed 3 times (with 25 ml deionised water, each) over the mesh to ensure that all particles clinging to the surface of the equipment are collected (figure 2.9). The microplastics on the surface of the mesh were further washed 3 times (with 25 ml of deionised water, each) to remove any extraneous matter. The plastic samples were then transferred onto a clean glass microfiber filter (47 mm, 1.0 μm pore size, Whatman, UK) using a Millipore® All-Glass Filter Holder Kit and a Welch™ 2522 WOB-L piston vacuum pump (Merck, Dorset, UK). The filtration mesh was rinsed 7 times with 15 mL of deionized water from a squirt bottle over the same filter to ensure no MPs were lost during sample transfer.

Finally, the collected $ZnCl_2$ solution (and settle solids) were filtered through a clean microfiber filter (diameter 47 mm) using a sintered glass filter funnel connected to a similar

vacuum filtration system. The collected solids and the filter were discarded, while the recovered $ZnCl_2$ was stored in a glass bottle at 4℃ until discarded safely using the existing laboratory safety protocols for hazardous chemicals.



Figure 2.9 Transfer MPs particles from SMI through the stainless steel mesh.

The choice of 25 μm mesh for filteration was based on previous literature reporting 25 μm as the "commonly reported" lower size limit for reliable chemical analysis using micro-FTIR (Huang et al., 2023; Jung et al., 2021). However, it should be noted that smaller particles could be identified in our dust samples using microscopic analysis (see below). In addition, using our state-of-the-art micro-FTIR instrument in "spot analysis" mode (see below), enabled measuring particle sizes down to 10 μm. Detection of MPs sizes below the cut-off limit for filteration during sample preparation has been reported before and was attributed to potential agglomeration of small particles on the filter or the ability of smaller particles to cling to the filter mesh material at such small aperture size (Bhavsar et al., 2023; Jung et al., 2021; Agboola and Benson, 2021).

2.1.2.2 Digestion process:

Using 200 ml of 30% hydrogen peroxide (H_2O_2) (Fisher Scientific) to significantly reduce the particle count, a digest to remove biological (non-MPs) materials was conducted. The mesh was rinsed with 30% hydrogen peroxide (H_2O_2) into 250 mL glass beakers pre-washed with Milli-Q water. The beakers were then covered with aluminum foil and placed in a drying oven at 60 °C (Thermo Scientific, Fisherbrand® 65L, oven 230V, Germany) for 96 hours (figure 2.10). At the end of 96 hours, the content of the beaker was poured over a 63 μm stainless steel mesh, and the beakers rinsed over the mesh to ensure that all particles clinging to its surface were collected. The beakers used were rinsed and washed in a standardized approach every single use, which ensured that all sides of the beaker were cleaned thoroughly (Dris et al., 2015; Liu et al., 2019d). The last step is to transfer the potential MPs particles from the mesh surface to clean glass petri dishes (54 mm, BRAND®, Merck™, Dorset, UK), then place them in the oven again for 24 hours to be thoroughly dried in preparation for the identification and quantification process.

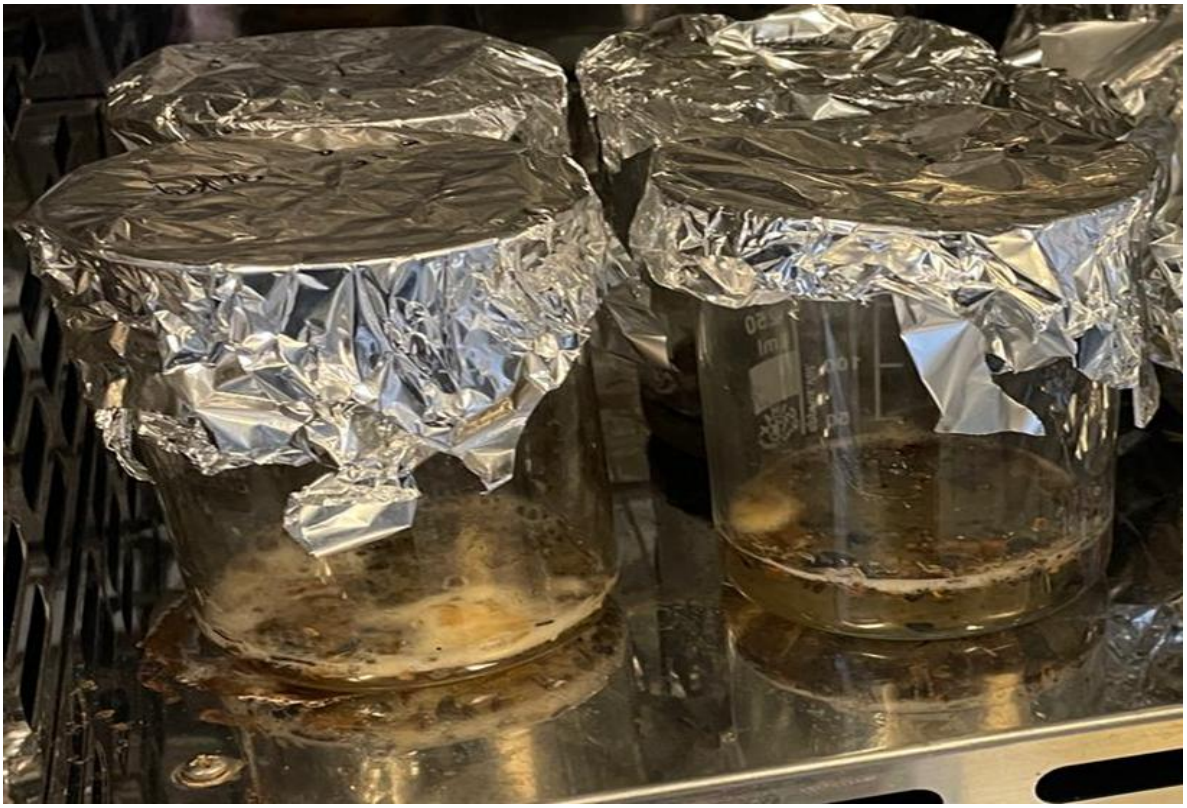


Figure 2.10 The digestion process.

2.1.3 MPs separation from air samples:

2.1.3.1 Active air sampling:

The glass microfiber filter (47 mm) was carefully removed from the stainless steel in-line 47 mm filter holder (Sartorius® 16254, Goettingen Germany), transferred to a clean glass petri dish (54 mm, BRAND®, Merck™, Dorset, UK) containing 1 ml of H_2O_2 solution (30% w/v, Merck™, Dorset, UK), which was enough to submerge the filter and particles on it. The petri dish was then placed in a plastic-free oven at 60℃ for 48 hours to oxidise any organic particles on the filter (digestion step). The digested filters were then covered by their pre-cleaned and dried lids in a clean dry place until further microscopic examination.

2.1.3.2 Passive air sampling:

After opening the glass petri dish (54 mm) used for sampling the deposition (atmospheric fallout) of MPs particles. 10 ml of Milli-Q water (18 MΩ.cm) were carefully

added and swirled around the dish, ensuring the lower sides of the dish are washed thoroughly, then placed on a slow mechanical shaker at 25 rpm for 30 min to release the particles. Then, the Milli-Q water was vacuum filtered through a glass fibre filter (47 mm, 1.0 µm pore size, Whatman, UK) using a Millipore® All-Glass Filter Holder Kit and a Welch™ 2522 WOB-L piston vacuum pump (Merck, Dorset, UK). The extraction cycle was repeated twice, with the 10 ml of Milli-Q water filtered on the same glass fibre filter (figure 2.11).

The filter was transferred to a clean glass petri dish (54 mm, BRAND®, Merck™, Dorset, UK) containing 1 ml of H2O2 solution (30% w/v, Merck™, Dorset, UK), which was enough to submerge the filter and particles on it. The petri dish was then placed in a plastic-free oven at 60℃ for 48 hours to oxidise any organic particles on the filter (digestion step). The digested filters were then covered by their pre-cleaned and dried lids in a clean dry place until further microscopic examination.

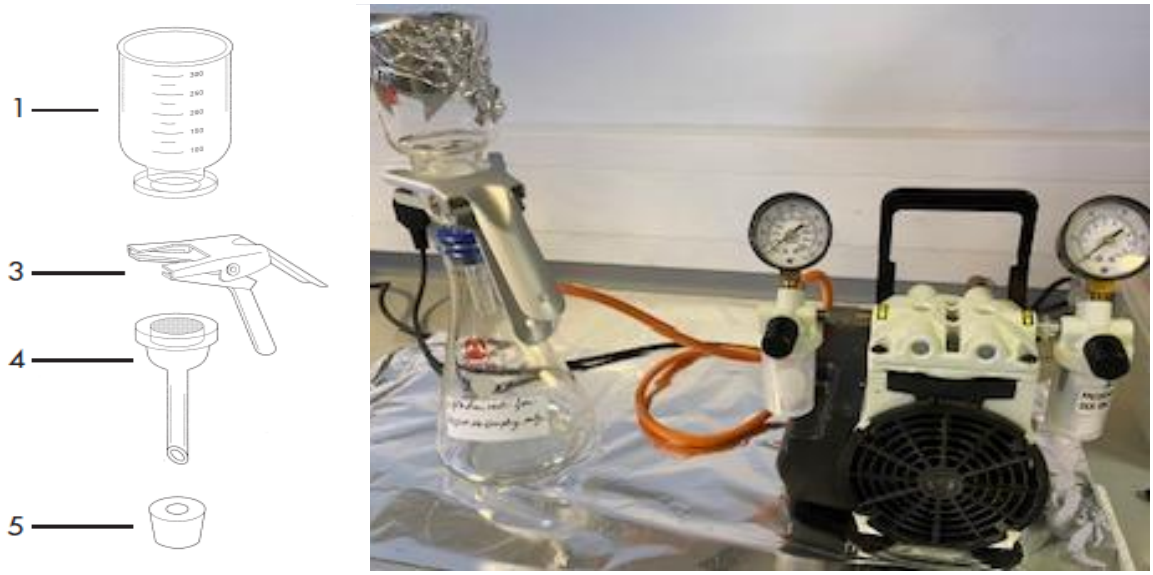


Figure 2.11 The filtration unit for passive samples.

2.1.4 MPs identification and quantification in indoor air:

In this thesis, two instruments were used to characterise MPs:

(a) Digital fluorescence microscope (OPTIKA™ IM-3LD4D PROVIEW, Version: x86,4.11.20351.20220226) with multiple lenses Plan Fluor (4×, 10×, 20×, 40×) has been

applied to identify the size, shape, and MPs particles counting as physical properties (Figure 2.12). The microscope is equipped with an X-LED cube 8 Watts illumination source, 6 LED fluorescence wavelength filters, a 250 x 290 mechanical stage mounted with a specific insert to hold 54 mm petri dishes, and a 6.3 megapixel USB 3.0 CMOS digital camera equipped with an IR filter range (380 – 650 nm). The digital microscope is operated through Microsoft Windows 7 system operating on a built-in 12-inch HDMI monitor. Visualisation and measurements of MPs were conducted using the bespoke "Optika™ ProView Image Analysis for fluorescence" software. The software requires regular calibration using a standard Optika supplied micromeasure on a standard slide, conducted weekly as the manufacturer advised (figure 2.13). As shown in (Figure 2.14) these are the shapes of MPs in indoor samples that appear by Digital fluorescence microscope. However, there is no standard or certified definition of MPs morphotypes (Kek et al., 2024b). The MPs shapes identified in the current study are morphology-based, as reported by (Baldwin et al., 2016). and followed widely by many researchers.



Figure 2.12 The digital Optika™ IM-3LD4D fluorescence microscope.

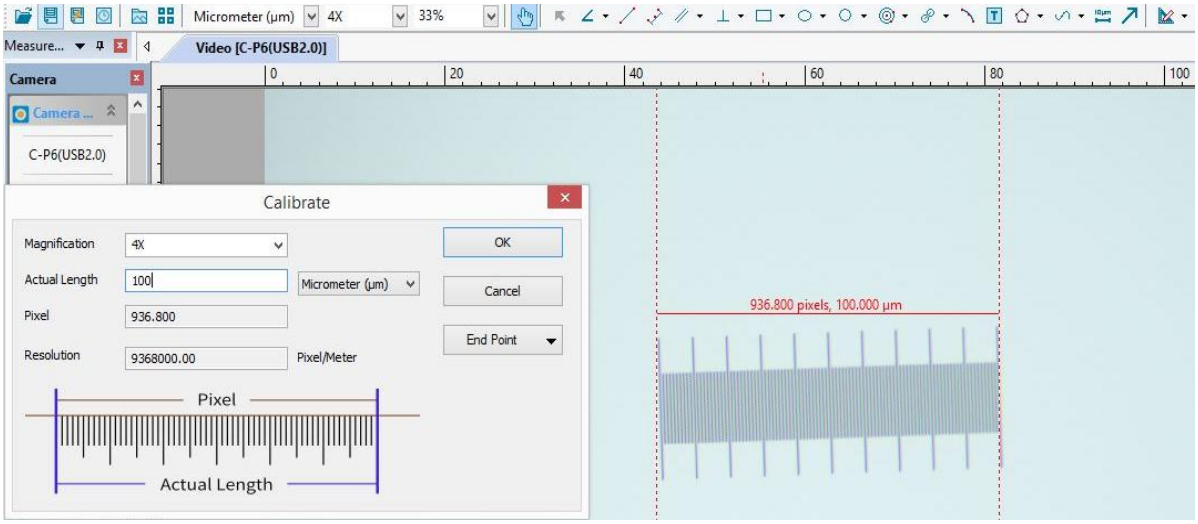


Figure 2.13 Standard unit calibration for the Optika™ IM-3LD4D microscope.

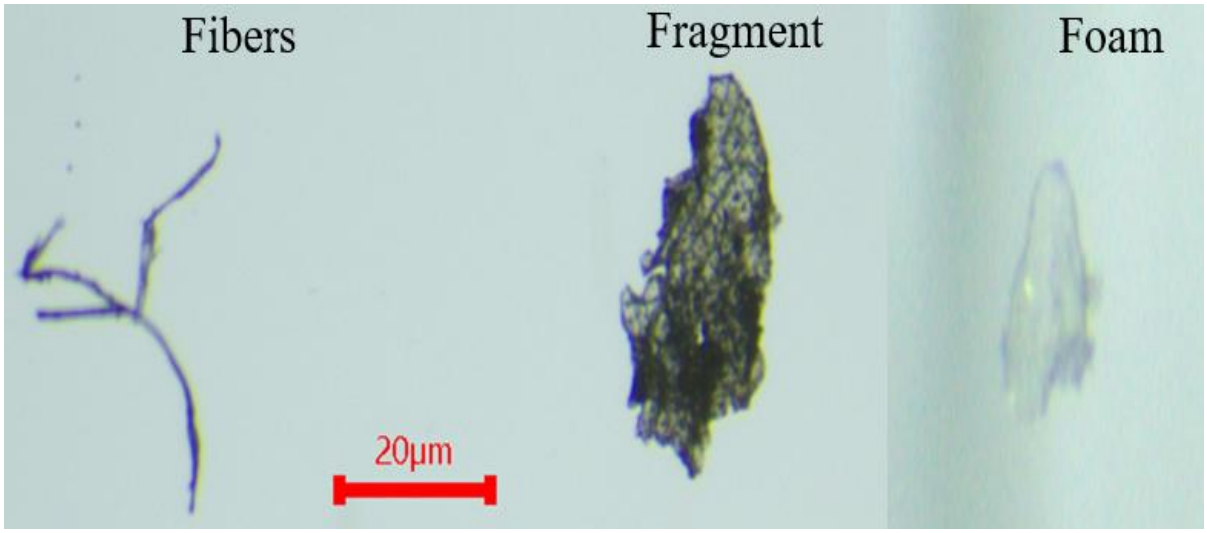


Figure 2.14 Representative examples of different MPs shapes observed in an indoor dust sample under the digital Optika™ IM-3LD4D fluorescence microscope.

(b) Perkin-Elmer Spotlight 400 imaging system has been applied to confirm identity of MPs particles. Also, Fourier-transform infrared spectroscopy (FTIR) has been used to identify the polymer types as chemical properties (Figure 2.15). A Perkin Elmer Spotlight^{TM} 400 Fourier-Transform InfraRed (µ-FTIR) imaging system with a remote-controlled stage and a Spectrum-3^{TM} FT-IR Spectrometer were used to analyze MPs. Spectrum MultiSearch^{TM} and SpectrumIMAGE^{TM} applications are installed on the system. Reflectance mode was used to image and map the whole filter.

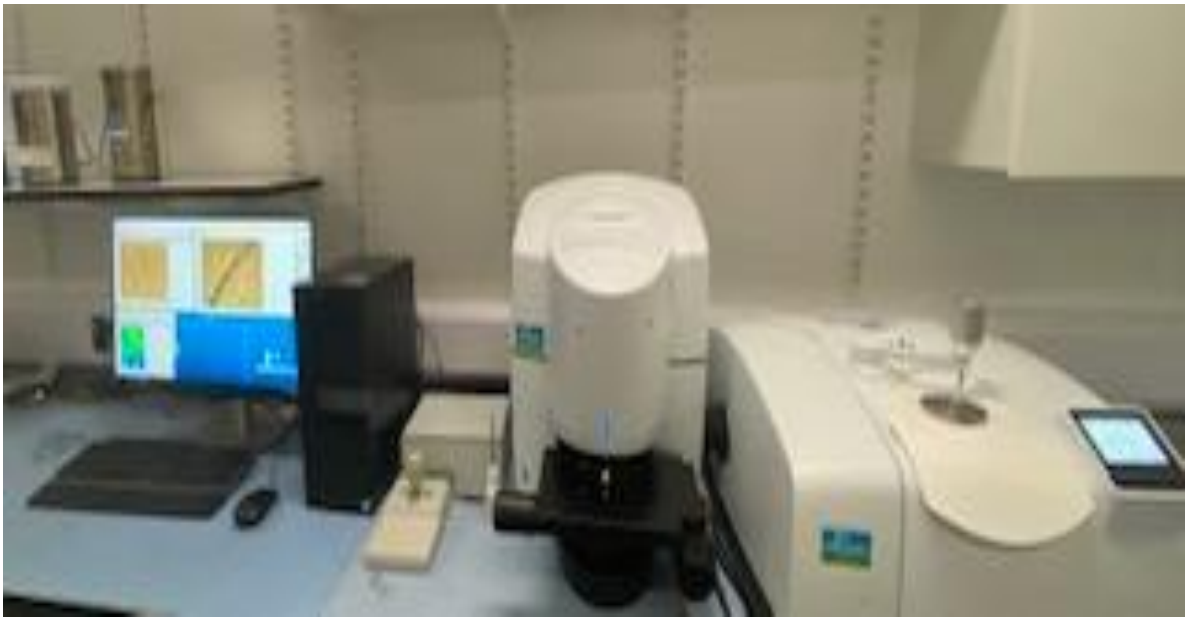


Figure 2.15 Perkin Elmer SpotlightTM 400 FTIR imaging system with a remote-controlled stage and a Spectrum-3TM FT-IR Spectrometer.

The size and type of glass microfiber filters (47 mm) used to collect MPs in the sampling/sample preparation processes (Section 2.1.3) are not suitable for µ-FTIR analysis in reflectance mode (Figure 2.16) and do not fit on the µ-FTIR instrument stage, which is fitted with holders for 13 µm and 25 µm filter holders. Hence, a further step has been taken to transfer the MPs particles to a silver membrane filter (diameter 13 mm) (STERLI TECH) through a vacuum filtration set (ADVANTEC, 1000 ml) (Figure 2.17). The reason behind selecting a silver filter (13 mm) is the excellent performance of silver filters under the whole energy range for µ-FTIR reflectance mode (figure 2.16). This is of paramount importance as it facilitates comparison and validation against various spectral libraries for different plastic polymers, which are all recorded in reflectance mode.

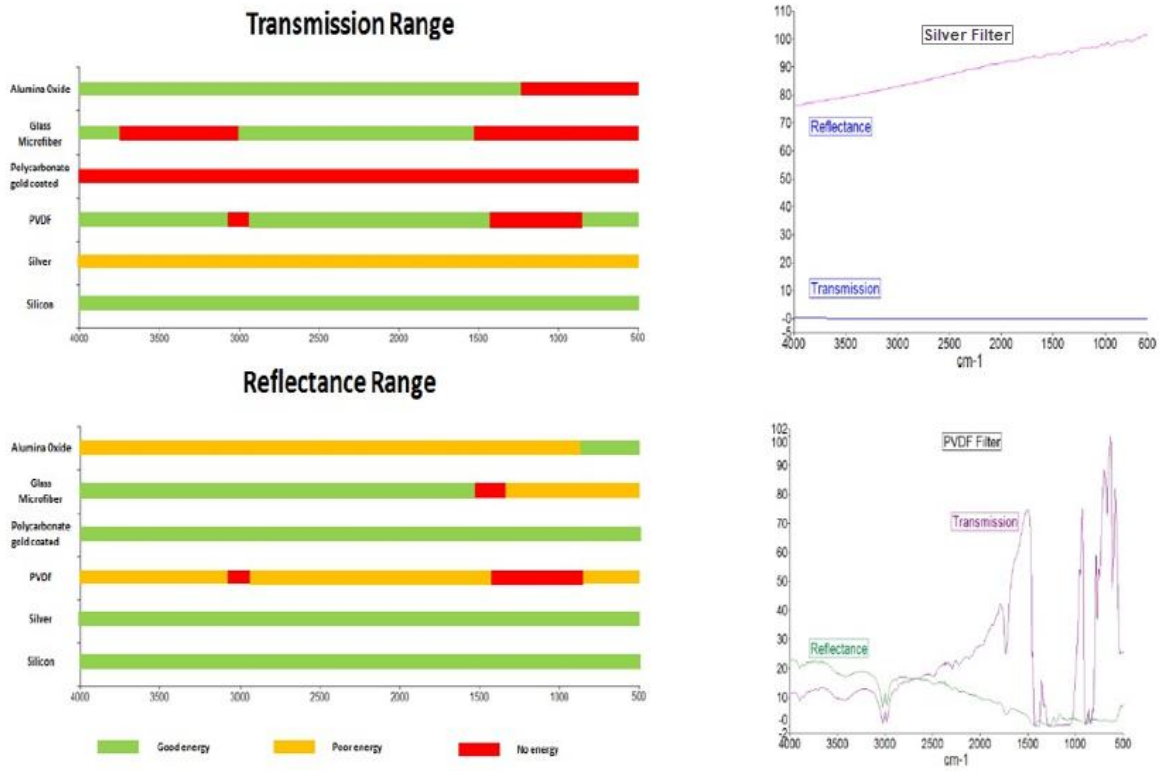


Figure 2.16 Energy efficiency for different filter types under full µ-FTIR transmission and reflectance ranges.

A 70% match criterion and visual peak diagnostics were used to compare the generated spectra to those from the Perkin Elmer Microplastics library and/or the independent software tool (siMPle®) to guarantee "best fit". Spectra were obtained with an aperture size of 20 × 20 µm, an accumulation of 16 scans, and a wavenumber range of 4000–600 cm^{-1} at a resolution of 8 cm^{-1}. When checked against the library spectrum, the particle was eliminated from the MPs count if the absorption spectra from 2850 to 3000 cm^{-1}, which originates from C–H vibrational stretching, did not occur. This indicates that the particle was not an organic molecule (Harley-Nyang et al., 2022; Mukotaka et al., 2021).



Figure 2.17 The filtration set used for transferring MPs particles from glass microfiber filters (47 mm) to silver filters (13 mm).

The number, shape, and size of MPs on each filter were determined using microscope images taken by the µ-FTIR microscope in imaging mode, with point mode used to verify MPs at or near the LOD of 10 µm in size. Figures 2.18 and 2.19 show a representative example of Perkin Elmer Spotlight^{TM} 400 FTIR imaging system and accompanaying software for detection and polymer type identification of MPs.

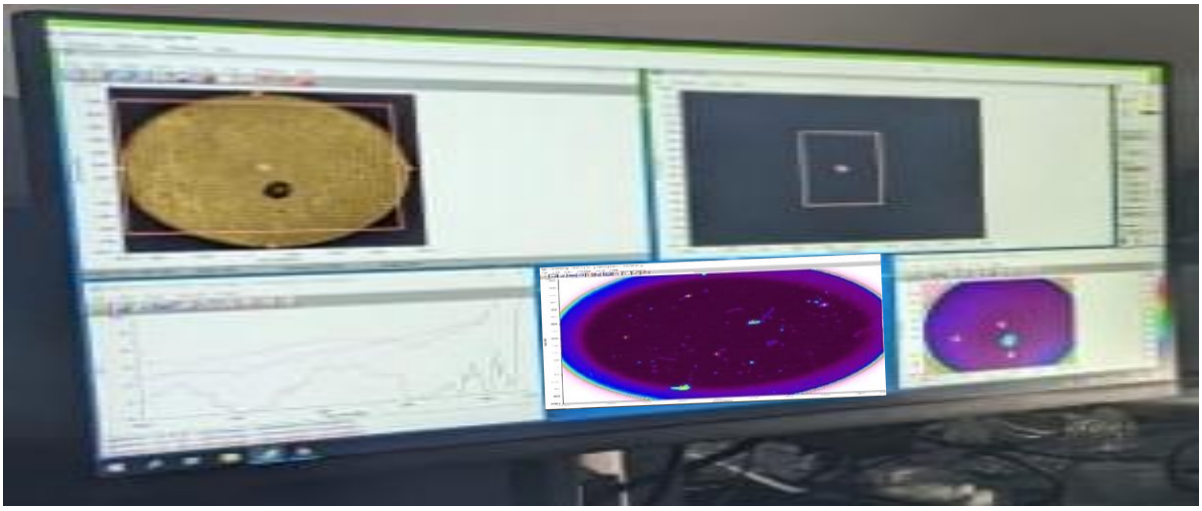


Figure 2.18 Perkin Elmer SpotlightTM 400 FTIR imaging system with Spectrum MultiSearchTM and SpectrumIMAGETM applications.

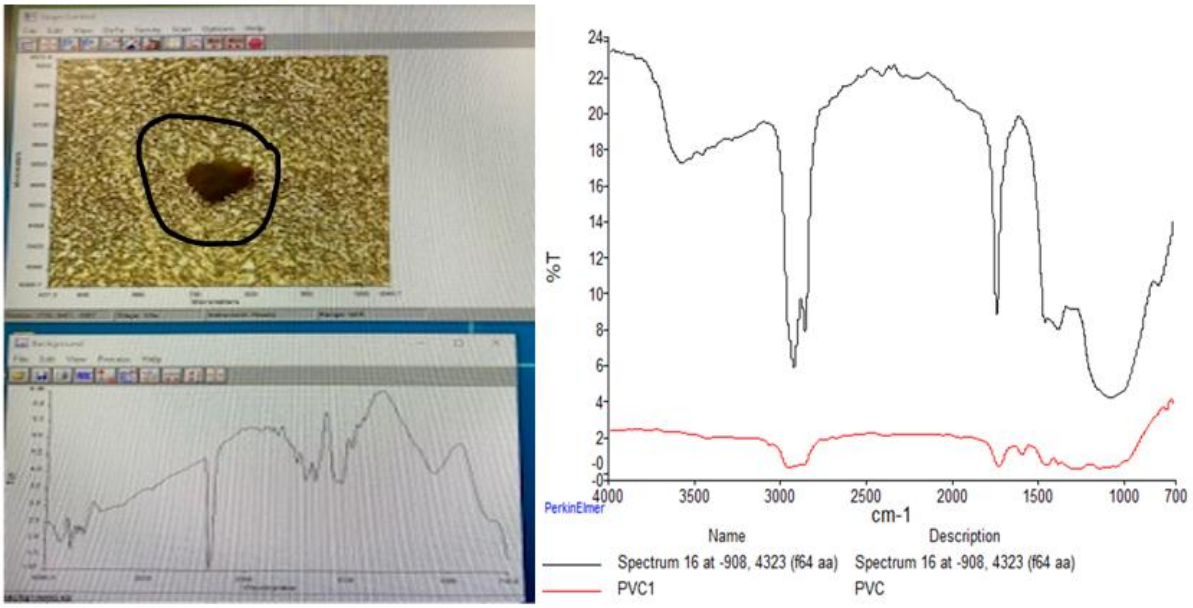


Figure 2.19 Representative example for polymer type identification of an MP particle.

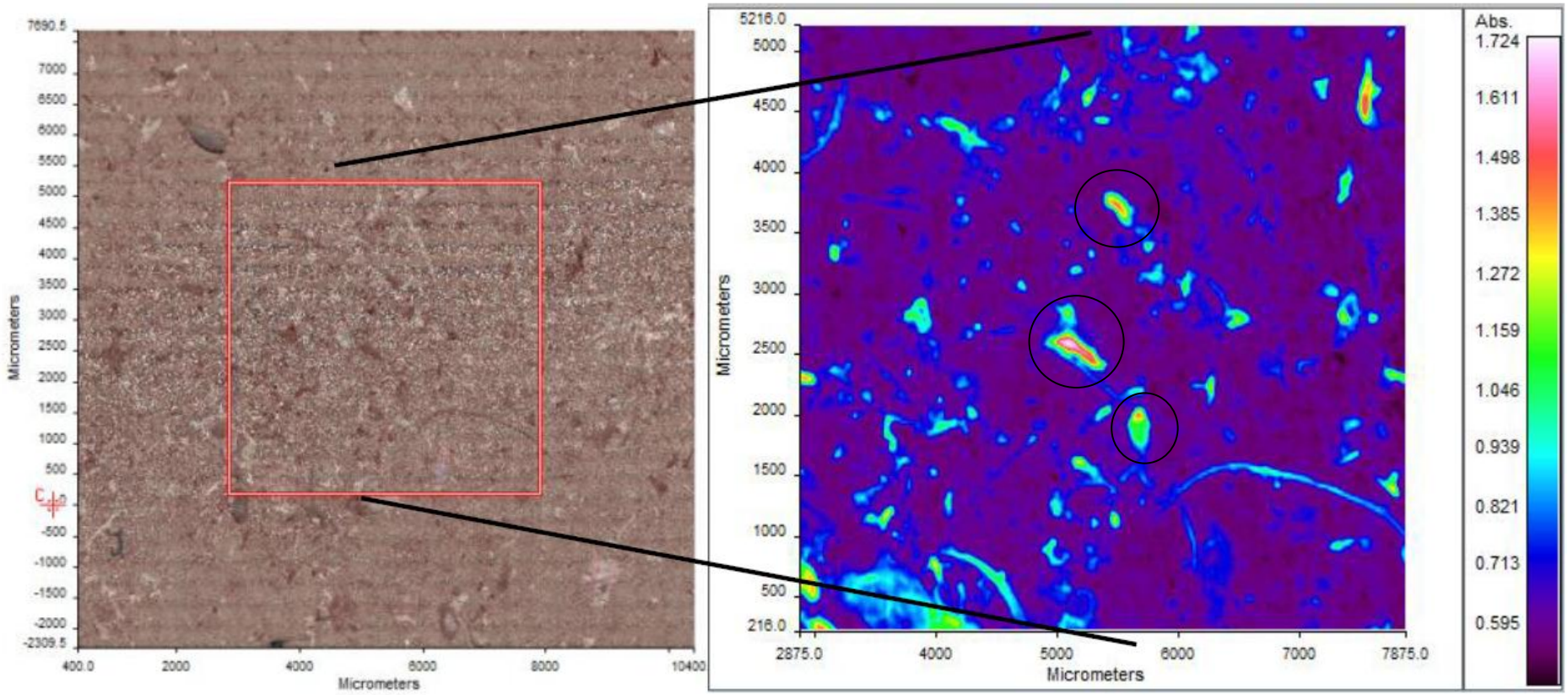


Figure 2.20 Representative example of μ-FTIR mapping for MPs on a silver filter.

2.1.5 Quality Assurance/Quality Control (QA/QC):

In the context of microplastics research, preventing background contamination by MPs particles in the laboratory is crucial. To achieve this, I wore lab coats of pure cotton and nitrile gloves throughout the experimental process. Additionally, all sample processing occurred on a laminar flow bench, which was regularly checked and wiped down with 90% ethanol. Furthermore, before use, all laboratory equipment and consumables were meticulously rinsed with Milli-Q water. In the investigation of potential contamination during the sample analysis (including preparation and filtration), dust samples were systematically analyzed in batches of five. A blank sample comprising an unused thimble, unpacked on the field (field blank) and treated like a sample (Section 2.1.1.3), was analysed alongside each batch of 5 dust samples. A "method blank" for the density separation method, comprising 15 mg of $ZnCl_2$ instead of indoor dust, treated as a sample (Section 2.1.2) was conducted alongside each 10 samples. Additionally, a "recovery sample", consisting of 50 mg of clean sediment (Sigma-Aldrich), intentionally spiked with a known quantity (20) of PE MPs particles (size fraction 38-45 µm, Cospheric™, California, USA), was processed alongside each 20 samples.

For air samples, A "field blank" was analysed alongside each batch of 5 air samples. For active air sampling, the field blank comprises an unused 47 mm GFF unpacked in the field and treated like a sample (section 2.1.1.2), whole for passive air samples, the field blank consisted of an unopened petri-dish treated like a sample (section 2.1.1.2). A "recovery sample" was processed alongside each 20 samples. For active air sampling, the recovery sample comprised an unused 47 mm GFF spiked with a known quantity (20) of PE MPs particles (size fraction 38-45 µm, Cospheric™, California, USA), while for passive air sampling, the recovery sample was an unused petri-dishspiked with a known quantity (20) of PE MPs particles (size fraction 38-45 µm, Cospheric™, California, USA).

Detailed results of blanks and recovery samples are provided in Table 2.1. Overall, the numbers of MPs in all blanks never exceeded 5% of the average concentration of MPs in the corresponding batch. Therefore, there was no need for blank corrections. The recovery of MPs in the recovery samples ranged between 80% and 110%, indicating good performance of the analytical methodology.

Table 2.1a Number of MPs in the analysed blanks.

Blank type/no.	Indoor dust	Active air sample	Passive air sample
Field 1	2	2	3
Field 2	5	1	2
Field 3	1	3	1
Field 4	3	1	2
Field 5	2	0	1
Field 6	3	0	3
Field 7	0	3	1
Field 8	0	1	3
Field 9	6	0	0
Field 10	2	0	0
Field 11	1	1	0
Field 12	2	1	0
Field 13	1	1	1
Field 14	0	0	1
Field 15	5	0	1
Field 16	2	2	3
Field 17	0	3	1
Field 18	0	1	2
Field 19	2	1	0
Field 20	3	0	3
Field 21	1	1	0
Field 22	4	1	3
Field 23	0	2	2
Field 24	1	3	2
Method 1	3		
Method 2	0		
Method 3	1		
Method 4	3		
Method 5	0		
Method 6	2		
Method 7	2		
Method 8	3		
Method 9	0		
Method 10	1		
Method 11	1		
Method 12	0		

Table 2.1b Recovery (expressed as %) of MPs in the QA/QC recovery samples.

Sample no	Indoor dust	Active air sample	Passive air sample
Recovery 1	85	95	80
Recovery 2	80	95	85
Recovery 3	110	80	105
Recovery 4	85	90	95
Recovery 5	110	105	80
Recovery 6	85	90	80
Average	93	93	88
SD	14	8	10
Median	85	93	83
Min	80	80	80
Max	110	105	105

2.1.6 Statistical Analysis:

Two programs were applied to analyze the data during this study: Excel version (Microsoft Office 365) and Statistical Package for the Social Sciences (SPSS) IBM version 29.0.1. Through Excel, the quantitative levels and distribution patterns were determined using parameters such as average, standard deviation, median, minimum, and maximum. To test the normality of the distribution to each data set using SPSS (Kolmogorov-Smirnov test). The outcomes were nonnormal distributed data. Also, the Independent-samples Mann-Whitney U Test was used to compare the differences between homes and workplaces in both groups separately (active and passive). For the seasonal variation sampling, ANOVA and the post hoc Tukey test were used to investigate differences among three or more dependent variables. The paired t-test was used to compare the means between the two datasets. Potential correlation between generated data was tested using Pearson correlations with a p-value of < 0.05 set as a level of statistical significance. All data are outlined as mean ±SD, and all "differences" are mentioned to indicate a statistically significant $P < 0.05$.

Chapter 3

This chapter is published from the following article: Ageel, H.K., Harrad, S., Abdallah, M.A.E. (2024) Microplastics in indoor air from Birmingham, UK: Implications for inhalation exposure. Environmental Pollution, Volume 362, 124960, ISSN 0269-7491. 6 Citations, 2 news articles.

3.1 Microplastics in indoor air from Birmingham, UK: Implications for inhalation exposure:

Abstract

Microplastics (MPs) are a group of emerging contaminants that attracted increasing scientific and societal attention over the past decade. So far, most studies on MPs focus on characterizing their occurrence, fate, and impact in the aquatic environment. Very little is known about the magnitude, patterns, and associated risks of human exposure to MPs, particularly indoors, despite people spending most of their time indoors. This paper provides the first and most comprehensive study hitherto of MPs in indoor air via both active and passive sampling from 30 homes and 30 workplaces in Birmingham, UK. The average concentration of MPs in the active air samples was 15.6 ± 5.4 MPs/m^3 in homes and 13.1 ± 6.5 MPs/m^3 in workplaces. For atmospheric deposition samples (passive sampling), the average MPs concentrations were 3735 ± 1343 MPs/m^2/day in homes and 3177 ± 1860 MPs/m^2/day in workplaces. Mean concentrations of MPs in UK homes were significantly higher ($P<0.05$) than those in workplaces for both active and passive air samples. This was mainly driven by carpeted floors in all the studied homes, while 13 of the sampled workplaces were uncarpeted. MPs concentrations in atmospheric deposition (passive) samples were significantly higher ($P<0.05$) than airborne (active) MPs samples in the studied microenvironments. Nonetheless, a strong correlation ($P<0.01$) was observed between the concentrations of MPs measured by active and passive sampling, indicating common sources of MPs to both active and passive samples collected from the same microenvironments. In terms of morphology, fibres were the dominant

shape of MPs, followed by fragment, constituting together ≥ 90% of the identified MPs in all samples, with the remaining minor percent made up by foams. Airborne MPs were dominated by particles in the size range (10- 25 µm), and the particles abundance decreased with increasing particle size. MPs in atmospheric fallout particles, were dominated by larger particles (50-100 µm) with lower contribution from smaller particles (10-25 µm) compared to airborne particles. Nonetheless, combined with the predominance of fibres, this raises concern over the risk from inhalation exposure because MPs fibres in the observed size ranges were observed to penetrate into human lung tissue. PET and PVC were the most abundant polymer types in the studied samples followed by PP and PE. UK adults and toddlers average daily inhalation exposure was estimated at 3.0 and 6.3 MPs/kg body weight/day, respectively. The higher inhalation exposure in UK toddlers raises concern over potential risk due to their incompletely developed immune and nervous systems.

Keywords:
Microplastics, Indoor air, Active sampling, Passive sampling, Human exposure

3.1.1 Introduction:

Global Plastic production has reached 300 million tonnes in 2018, and is likely increasing (Hale et al., 2020; Barnes et al., 2009b). The lack of efficient plastic recycling in many countries has caused an increase in plastic waste, as it was estimated that recycled plastic doesn't exceed 3% of plastic waste worldwide (Hale et al., 2020). In this vein, some estimates indicate that annual plastic waste will reach approximately 67.8 million tonnes in the environment by the year 2050, if the situation is not remedied (Wirnkor et al., 2019; Ebere et al., 2019). When introduced into the ecosystem, plastic materials undergo continuous processes of mechanical force, chemical weathering, photo-oxidation, and biological degradation, fragmenting into smaller, tiny pieces called microplastics (MPs) (Kubowicz and Booth, 2017).

MPs detection in several environmental compartments, such as marine ecosystems, soil, sediment, water and air, has become one of the most concerning global environmental pollution issues in the past few years; due to their persistence and ability to transport and accumulate in various environmental compartments (Yao et al., 2019b; Duis and Coors, 2016; Wu et al., 2019d; SAPEA, 2019c). Nevertheless, most recent studies in this field focused on the characterisation, fate, and behaviour of MPs in marine ecosystems (Ageel et al., 2022), while studies on MPs in indoor environments are generally scarce compared to those on MPs in aquatic and terrestrial outdoor environments. Consequently, the levels, patterns, and associated risks of human exposure to MPs in the indoor environment remain not fully understood (Wright et al., 2013c; Gall and Thompson, 2015b).

In principle, MPs are a heterogeneous class of emerging contaminants of different polymer types, sizes, shapes, densities, colours, chemical additives, and sources, with their longest dimension < 5 mm (Rocha-Santos and Duarte, 2015). Moreover, MPs have proven capacity to adsorbs various toxic chemicals present in their surrounding environment, including hydrophobic organic pollutants (e.g., polycyclic aromatic hydrocarbons, polychlorinated biphenyls, hexachlorocyclohexane isomers) and heavy metals (e.g., cadmium, zinc, nickel, lead, etc.) (Foulon et al., 2016). This has raised concern over threats to human health from exposure to MPs. Several studies reported on human absorption and accumulation of MPs, particularly in the smaller size range < 50µm, in vital tissues and organs including blood, ovaries, testes, placenta, spleen, liver, colon and lungs (Li et al., 2023a; Montano et al., 2024). However, according to the recent WHO analysis on the current research related to microplastics (WHO, 2022a), there is currently limited evidence to suggest microplastics are causing significant adverse effects on human health. This was attributed to major knowledge gaps in scientific understanding of the pathways and magnitude of toxicological impacts of MPs in various human tissues, as well as exposure thresholds required to trigger such impacts.

Current research indicates that MPs toxicity in humans and mammals may occur through inducing oxidative stress, DNA damage, metabolic disorders, immunotoxicity, reproductive toxicity and neurotoxicity (Li et al., 2023a).

Human exposure to MPs can occur through several pathways (via different environmental media), including inhalation (indoor and outdoor air), ingestion (diet, water and indoor dust), and dermal contact (Fabrics, personal care products, atmospheric deposition) (Prata et al., 2020; Ageel et al., 2022). Several studies have focused on human exposure to MPs via ingestion through diet, and to a lesser extent, drinking water and beverages (Karami et al., 2017;Sun and Wang, 2023). However, very little is known about human exposure to MPs via inhalation and dermal exposure pathways (Dris et al., 2017a; Yao et al., 2022). Moreover, few studies reported on the presence, abundance and characteristics of MPs in outdoor air, and to lesser extent in indoor air (Kek et al., 2024a; O'Brien et al., 2023). Collectively, these studies indicated higher MPs in indoor air, compared to outdoor air. This was attributed to a combination of factors including various indoor sources e.g., abrasion of synthetic textiles and deterioration of plastic materials, with factors like carpeting, airflow, and ventilation significantly impacting indoor MP levels, as well as relatively increased human activity in the indoor spaces, resulting in re-suspension of microplastic particles in the "personal cloud" this term refers to the localised area around an individual when re-suspended MPs particles tend to accumulate. (Kek et al., 2024a). Despite humans spending up to 90% of their time indoors, very little is known about human exposure to microplastics via inhalation in different indoor microenvironments. This is further compounded by existing challenges in comparing and modelling the outcomes of the few studies on the occurrence and characteristics of MPs in indoor air due to different sampling methods (e.g., active air sampling vs atmospheric deposition), analytical techniques, particle size range, polymer-type investigation, and

inconsistent reporting units (MPs/m^3 for active air sampling vs. MPs/m^2/day for atmospheric deposition) (O'Brien et al., 2023).

Against this backdrop, the aim of the current study is to provide first assessment of UK population's exposure to MPs via inhalation of indoor air from different microenvironments; thereby providing a basis for risk assessment from such exposure. This will be addressed via determination of MPs concentrations, and characteristics (i.e., size, shape, and polymer type) from several homes and office microenvironments in Birmingham, UK, using both active air sampling and atmospheric deposition. Possible correlations between MPs concentrations and characteristics in samples collected using both methods from the same microenvironments are investigated. Finally, exposure of different UK age groups to MPs via inhalation of indoor air is assessed, for the first time, and the potential risk from such exposure is discussed.

3.1.2 Materials and methods:

3.1.2.1 Sample collection:

Sampling was conducted between May 2022 and September 2023 in Birmingham, which is the second largest city of the United Kingdom, located in England's West Midlands region. The chosen sites for sample collection comprise two types of microenvironments: houses and workplaces. The sampling sites were randomly selected and constitute a sample of convenience for the research team, with enabled access during the required sampling times/durations. For the purpose of this study, private houses and flats from several residential buildings and various workplaces, including offices, meeting rooms, and lecture rooms. Two techniques were used in this study for collecting samples from each of the studied microenvironments: active and passive air sampling (Shao et al., 2022). 30 houses and 30 workplaces were sampled for indoor air, providing a total of 60 passive air samples, and 60 active air samples.

Active air Sampling:

The low-volume active air sampler comprised a low-volume pump (Capex L2X, Charles Austen, UK), operated at a flow rate of 6 L min^{-1} for 24 hours to yield a single sample comprising 8.64 m^3 air. The particulate phase was collected by employing a 47 mm diameter membrane glass fibre filter (1.0 μm pore size, Whatman, UK) housed in a stainless steel in-line 47 mm filter holder (Sartorius® 16254, Goettingen Germany). The designated flow rate of 6 L min^{-1} was achieved and maintained by using a flow meter (Platon 12 L min^{-1}) connected to an adjustable valve. The flow meter was calibrated at the start and the end of each experiment using a Gilibrator® air flow calibrator (Sensidyne, Inc., Clearwater, Florida, USA), which is classified as a primary standard device.

Passive air Sampling:

The passive air sampler was used to capture indoor airborne microplastics fall-out (atmospheric deposition), by applying an open glass Petri dish (20 cm diameter, Duran® soda-lime glass, VWR International ltd., Leicestershire, England) placed at head height (1–1.5 m) within the selected sites. The glass Petri dishes were only opened on site at a set date and time. In each sampling session, the Petri dishes were exposed to atmospheric fallout for 24 hours to collect dry and wet MPs atmospheric deposition fallout, while participants were instructed not to change any of their habits or daily routines. At the end, the petri dish was closed with its specific glass lid, wrapped in clean aluminium foil, and secured with a paper-tape, then transferred to the laboratory for further analysis by the primary researcher. In the laboratory, Petri dishes were stored in a cool (5°C), dark place until further processing.

3.1.2.2 Sample preparation and analysis:

Active air:

The glass microfiber filter (47 mm) was carefully removed from the stainless steel filter holder, transferred to a clean glass petri dish (60 mm, BRAND®, Merck™, Dorset, UK) containing 1 ml of H_2O_2 solution (30% w/v, Merck™, Dorset, UK), which was enough to

submerge the filter and particles on it. The petri dish was then placed in a plastic-free oven at 60℃ for 48 hours to oxidise any organic particles on the filter (digestion step). The digested filters were then covered by their pre-cleaned and dried lids in a clean dry place until further microscopic examination.

Passive air:

After opening the glass petri dish used for sampling the atmospheric MPs, 10 ml of Milli-Q water (18 MΩ.cm) was carefully added and swirled around the dish, ensuring the lower sides of the dish are washed thoroughly, then placed on a slow mechanical shaker at 25 rpm for 30 min to release the particles. Then, the Milli-Q water was vacuum filtered through a glass fibre filter (47 mm, 1.0 µm pore size, Whatman, UK) using a Millipore® All-Glass Filter Holder Kit and a Welch™ 2522 WOB-L piston vacuum pump (Merck, Dorset, UK). The extraction cycle was repeated twice, with the 10 ml of Milli-Q water filtered on the same glass fibre filter. The filter was transferred to a clean glass petri dish (60 mm, BRAND®, Merck™, Dorset, UK) containing 1 ml of H_2O_2 solution (30% w/v, Merck™, Dorset, UK), which was enough to submerge the filter and particles on it. The petri dish was then placed in a plastic-free oven at 60℃ for 48 hours to oxidise any organic particles on the filter (digestion step). The digested filters were then covered by their pre-cleaned and dried lids in a clean dry place until further microscopic examination.

3.1.3 MPs identification and quantification:

In this study, two instruments were used to characterise MPs: (a) Digital stereo fluorescence microscope (OPTIKA™ IM-3LD4D PROVIEW, Version: x86,4.11.20351.20220226) with multiple lenses Plan Fluor (4X, 10X, 40X, 100X) has been applied to identify MPs size, shape and particles count.

(b) Perkin Elmer Spotlight™ 400 Fourier-Transform InfraRed (μ-FTIR) imaging system with a remote-controlled stage and a Spectrum-3™ FT-IR Spectrometer was used to identify the polymer types and confirm identity of MPs particles. Spectrum MultiSearch™ and SpectrumIMAGE™ applications are installed on the system. Reflectance mode was used to image and map the whole filter. A 70% match criterion and visual peak diagnostics were used to compare the generated spectra to those from the Perkin Elmer Microplastics library and/or the independent software tool (siMPle®) to guarantee "best fit". Spectra were obtained with an aperture size of 20 × 20 μm, an accumulation of 16 scans, and a wavenumber range of 4000–600 cm^{-1} at a resolution of 8 cm^{-1}. When checked against the library spectrum, the particle was eliminated from the MPs count if the absorption spectra from 2850 to 3000 cm^{-1}, which originates from C–H vibrational stretching, did not occur. This indicates that the particle was not an organic molecule (Harley-Nyang et al., 2022; Mukotaka et al., 2021). The number, shape, and size of MPs on each filter were determined using microscope images taken by the μ-FTIR microscope in imaging mode, with point mode used to verify MPs at or near the LOD of 10 μm in size.

3.1.4 Quality assurance/quality control:

In the context of microplastics research, preventing background contamination by MPs particles in the laboratory is crucial. To achieve this, researchers exclusively wore lab coats of pure cotton and nitrile gloves throughout the experimental process. Additionally, all sample processing occurred on a laminar flow bench, which was regularly checked and wiped down with 90% ethanol. Furthermore, before use, all laboratory equipment and consumables were meticulously rinsed with Milli-Q water.

A “field blank” was analysed alongside each batch of 5 air samples. For active air sampling, the field blank comprises an unused 47 mm GFF unpacked in the field, then

transferred to clean glass Petri dish and treated like a sample. For passive air samples, the field blank consisted of an unopened petri-dish taken to the field then transferred to the lab and treated like a sample.

A "recovery QA sample" was processed alongside each 20 samples. For active air sampling, the recovery sample comprised an unused 47 mm GFF spiked with a known quantity (20) of PE MPs particles (Cospheric™, California, USA), while for passive air sampling, the recovery sample was an unused petri-dish, spiked with a known quantity (20) of PE MPs particles (Cospheric™, California, USA).

Detailed results of blanks and recovery samples are provided in Table SI3-1. Overall, the numbers of MPs in all blanks never exceeded 5% of the average concentration of MPs in the corresponding batch. Therefore, there was no need for blank corrections. The recovery of MPs in the recovery samples ranged between 80% and 110%, indicating good performance of the analytical methodology.

3.1.5 Statistical analysis:

Statistical analysis of data was conducted using Microsoft Excel (Microsoft Office 365 package) and Statistical Package for the Social Sciences (SPSS) IBM version 29.0.1. Microsoft Excel was used to determine statistical parameters such as average, standard deviation, median, minimum, and maximum of the generated datasets. The distribution of each dataset was examined using SPSS (Kolmogorov-Smirnov test). The generated datasets were log-normal distributed; therefore, further parametric tests (e.g., ANOVA) were conducted on log-transformed data. Potential correlation between generated data was tested using Pearson correlations with a p-value of < 0.05 set as a level of statistical significance.

3.1.6 Human Exposure Assessment:

Concentrations of MPs determined in indoor air from homes and offices using active air sampling were used to estimate exposures of UK adults and toddlers via inhalation of airborne MPs. In brief, two exposure scenarios were considered: a "typical" exposure scenario using median MPs concentrations and the second a "high-end exposure scenario", assuming inhalation of the 95th percentile MPs concentrations in the studied microenvironments. Daily inhalation rates, and average body weights for adults and toddlers were obtained from the USEPA exposure factors handbook (US EPA, 2011), while typical time activity patterns for UK population were adopted from previously published studies (Tao et al., 2016; Wemken et al., 2019a). Details of the exposure model and factors used in inhalation exposure calculations are provided in the supplementary information.

3.1.7 Results and Discussion:

3.1.7.1 Concentration of MPs in UK indoor air:

Active air sampling:

In this study, 60 UK indoor microenvironments were investigated. Microplastic particles were detected in all samples from all sites (Table 3.1). Using active air sampling, the average concentrations of airborne MPs were 15.6 ± 5.4 and 13.1 ± 6.5 MPs/m3 in the studied homes and workplaces, respectively. Comparing the means using a student t-test revealed MPs concentrations in the studied homes were significantly higher than in the workplaces ($P<0.05$). An investigation into possible causes of such discrepancy through potential sources of MPs in the studied microenvironments revealed that while all the sampled homes were carpeted, 13 of the studied workplaces were not carpeted (Tables SI3-4 and SI3-5). Indeed, statistical analysis revealed the mean concentration of MPs in the non-carpeted workplaces (n=13; mean = 9.5 MPs/m^3) was significantly lower ($P<0.05$) than the mean MPs concentrations in carpeted workplaces (n=17; mean = 15.8 MPs/m^3), and the studied homes (n=30; mean = 15.6 MPs/m^3).

Our results are in agreement with previous studies that reported carpeted floors as important sources of MPs to indoor air (Chen et al., 2022a; Dris et al., 2017b). However, a study from Kuwait reported the presence of carpets had no significant impact on MPs in indoor aerosols (Uddin et al., 2022b). This discrepancy may be explained by the finding of Yuk et al. (2022), who reported that MPs can originate from the deterioration of building and flooring materials to varying degrees due to friction, wear and tear (Yuk et al., 2022). This suggests that not only the presence of carpeted floors influence the concentrations of airborne MPs, but other factors such as: carpet material, age, type of flooring, frequency and method of cleaning and activity patterns, may also impact the levels and characteristics of MPs in indoor air (Kek et al., 2024a).

Our results show that airborne MPs concentrations in UK offices and homes fall within the range of indoor concentrations reported in the few studies available from other countries (Table SI3-3). Mean airborne MPs concentrations in UK houses and workplaces in the current study (Table 3.1) were higher than those reported from Barcelona's houses (4.8 ± 1.6 MPs/m^3) and workplaces (4.2 ± 1.6 MPs/m^3) (Torres-Agullo et al., 2022), South Korean apartment (3.02 ± 1.77 MPs/m^3) (Choi et al., 2022), Portugal house (6 MPs/m^3) (Prata et al., 2020b), and various Australian indoor microenvironments (0.20 – 2.25 MPs/m^3) (Perera et al., 2023). However, our results were lower than those reported from Denmark residential flats and workshops (92 – 548 MPs/m^3) (Maurizi et al., 2024), Taiwanese nail salon (46 ± 55 MPs/m^3) (Chen et al., 2022), and Chinese apartments (1583 ± 1180 MPs/m^3) (Liao et al., 2021). Notwithstanding the large differences in sampling and analysis of MPs in the above mentioned studies, which challenges meaningful comparison of results among different studies, the airborne MPs concentrations in UK indoor microenvironments reported in the present study are mostly in the same order of magnitude reported elsewhere in Europe, yet substantially lower than those reported from China (Liao et al., 2021). In addition to differences in sampling and analytical methods, the differences in indoor airborne MPs levels in various

microenvironments from different countries may also be attributed to different sources and lifestyle factors including: building and flooring material type and age, activity patterns, methods and frequency of cleaning, as well as number, age and condition of plastic items used indoors (Kek et al., 2024; Zhao et al., 2023).

Table 3.1: Statistical summary of MPs concentrations measured in the studied UK homes (n=30) and workplaces (n=30).

Statistical parameter	**Airborne MPs (MPs/m^3)**		**Atmospheric deposition (MPs/m^2/day)**	
	Homes	Workplaces	Homes	Workplaces
Average	15.6	13.1	3735	3177
Standard deviation	5.4	6.5	1343	1860
Median	14.6	12.4	3474	2951
Minimum	3.8	3.2	1623	706
Maximum	33.6	31.0	7513	9947

Passive air sampling:

Atmospheric deposition of MPs was observed in all the studied UK microenvironments with an average of 3735 ± 1343 MPs/m^2/day in homes and 3177 ± 1343 MPs/m^2/day in workplaces. Similar to airborne MPs, the mean atmospheric MPs deposition in the studied homes was significantly higher ($P<0.05$) than in offices, which could be mainly attributed to carpeted flooring in all the sampled homes, while 13 of the sampled workplaces were uncarpeted. The mean atmospheric deposition in the uncarpeted workplaces (2365 MPs/m^2/day) was significantly lower ($P<0.05$) than that in carpeted workplaces (3797 MPs/m^2/day) and homes (3735 MPs/m^2/day). This is in agreement with the results of Soltani et al. (2021) who reported a significant positive correlation between carpeted floors and increased indoor dust and MPs deposition in 32 Australian homes (Soltani et al., 2021c). A summary of results reported by the few previous studies on atmospheric deposition of MPs indoors is provided in Table SI-3. Notably, our results are largely in agreement with those

reported previously by a study from Yorkshire, UK, with an average of 1414 ± 1022 MPs/m^2/day from 20 houses sampled monthly for a 6-month period (Jenner et al., 2021c). Our results were generally in line with those reported from Australian homes (mean = 3095 MPs/m^2/day) (Soltani et al., 2021c), and an American office (6430 MPs/m^2/day) (Yao et al., 2022). However, MPs deposition in UK homes and offices in the present study was generally higher than that reported from School rooms from Sao Paulo, Brazil (307.2 ± 214.60 MPs/ m^2/day) (Amato-Lourenço et al., 2022), and lower than those reported from Chinese houses (78,839 – 96,367 MPs/ m^2/day) (Cui et al., (2022) and Indian offices (81,730 ± 1923 MPs/m^2/day) (Nandi et al., 2024). Understanding the underlying causes of these differences among studies from different countries is challenging given the wide variation in methodologies used in these studies, which include differences in particle size ranges of MPs investigated; chemical-based detection methods (e.g., laser direct infrared spectroscopy, micro FT-IR, and micro-Raman Spectroscopy); sample treatment processes (e.g., filter pore size, oxidation of organics); visual-based detection methods (e.g., Nile Red staining, SEM, fluorescence microscopy); and QA/QC protocols (e.g., blank corrections, recovery detection samples) (Table SI3-3). These differences can significantly influence the MPs concentrations reported in different studies, which can lead to uncertainty when comparing the outcomes (Ageel et al., 2022). Nonetheless, it could be inferred from available studies that MPs concentrations indoors are influenced by in-room characteristics including building and flooring materials, number, age and condition of plastic items, ventilation and frequency/method of cleaning, as well as personal activity and usage patterns within the sampled room (Soltani et al., 2021c; Ageel et al., 2022; Kek et al., 2024a; Zhao et al., 2023).

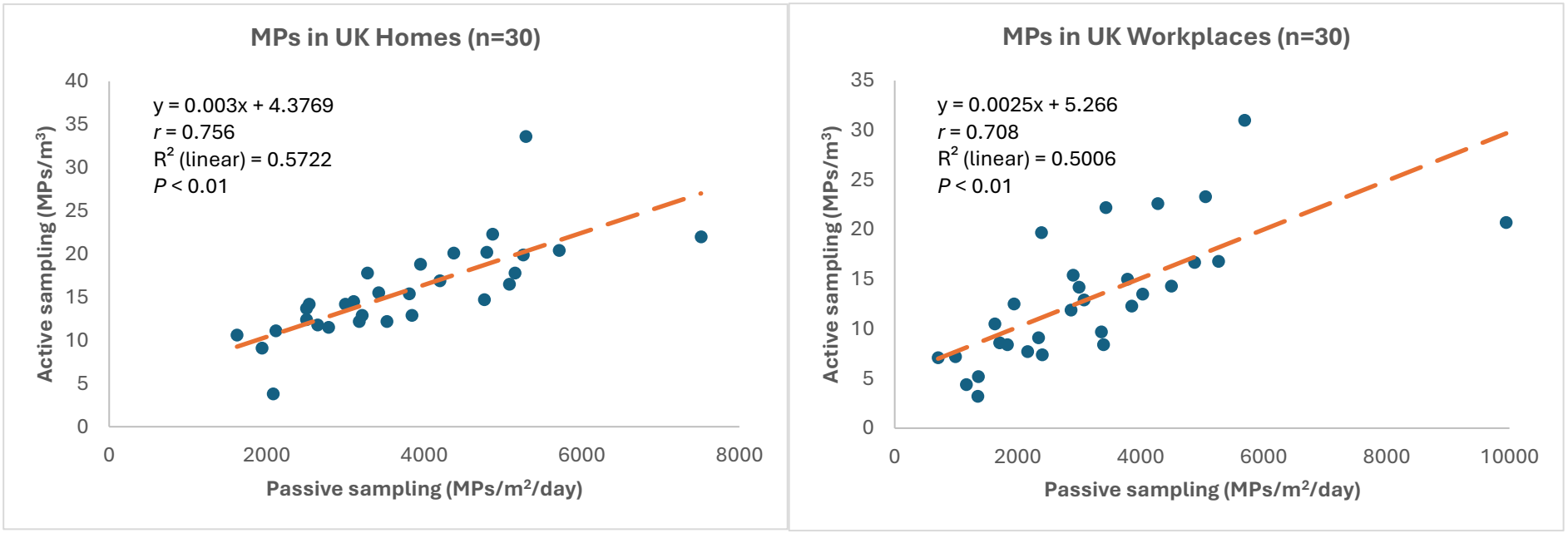
Active vs passive sampling:

In the present study, MPs were sampled from 60 UK microenvironments using both active and passive sampling techniques. While active sampling collects airborne particles, and

passive sampling measures atmospheric deposition of MPs, it is useful to compare the results obtained from the same microenvironments and with a reasonable sample size for the first time. Paired sample t-test revealed MPs in atmospheric deposition to be significantly higher (P<0.05) than airborne MPs in the studied UK microenvironments. The higher MPs concentrations in atmospheric fallout samples is expected given the variability in size and density of indoor MP particles, resulting in only a fraction of such particles being airborne for lengthy periods of time. This is compounded by the more confined nature of indoor microenvironments with less airflow and ventilation than outdoor air, which could facilitate MPs fallout indoors (Kek et al., 2024a). Nonetheless, A strong correlation (P<0.01) was observed between the concentrations of MPs measured by active and passive sampling in the studied homes and offices (Figure 3.1). This indicates common sources of MPs to both active (airborne) and passive (atmospheric fallout) samples collected from the same microenvironment.

On balance, passive sampling offers several advantages in terms of affordability, versatility, ease of use, and ability to sample over extended periods. However, passive samplers may have limitations in terms of the accuracy and precision of the results, related to extrapolation of their findings over unit space and time. On the other hand, active sampling offers better accuracy and precision (*i.e.,* known and fixed sampling rate), rapid sample collection, and is particularly efficient for assessing human exposure to MPs via inhalation as it samples airborne particles, but is more resource-intensive, requiring electric power and complex setup (Geng et al., 2023; Zhao et al., 2023).

Figure 3.1 Correlation between MPs concentrations determined via active (MPs/m^3) and passive (MPs/m^2/day) sampling in the studied UK microenvironments.



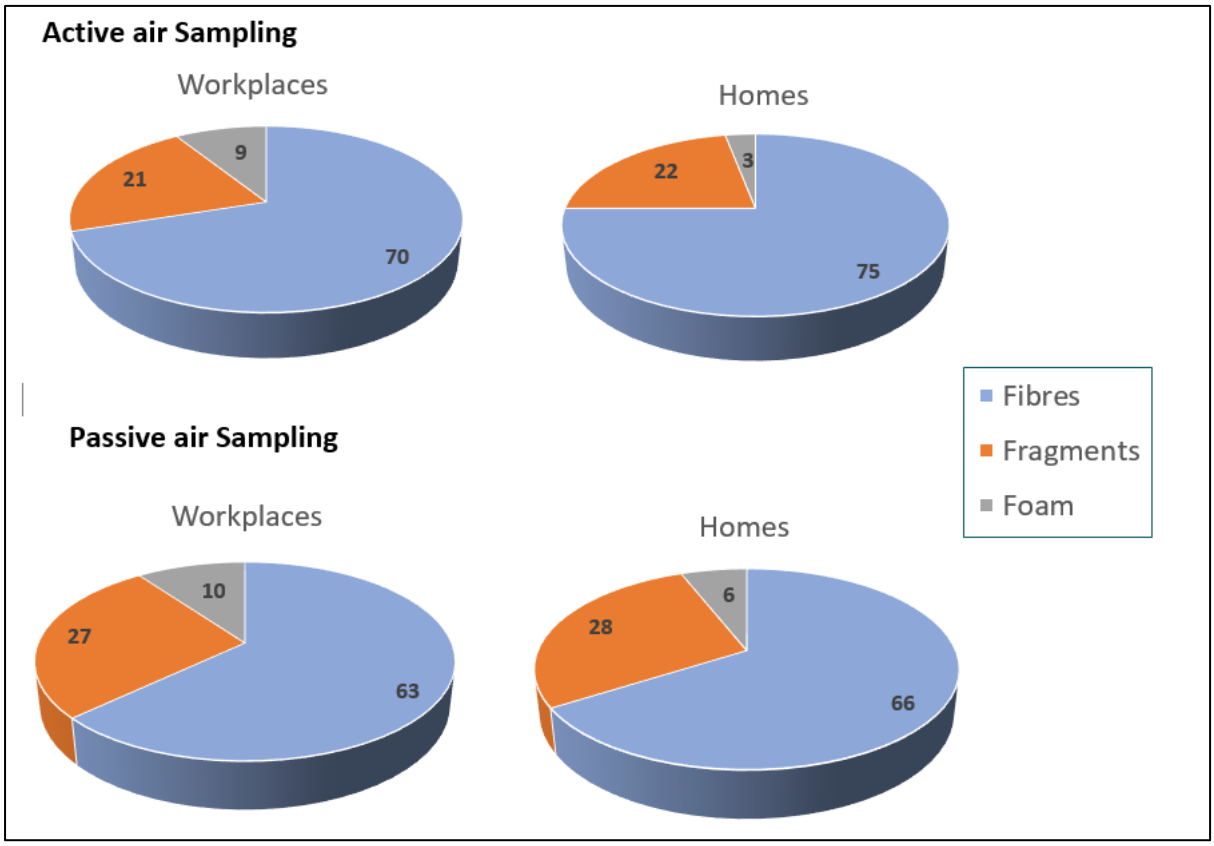
3.1.8 Characteristics of MPs in UK indoor air:

3.1.8.1 MPs shape:

In terms of morphology, the digital OPTIKA™ microscope was used to identify the shapes with the Perkin-Elmer™ Spotlight 400 imaging system applied to confirm particles plastic polymer type (discussed below). We observed that there were three different shapes of MPs in the analysed samples (Figure 3.2). In the UK home samples, the average composition of airborne MPs shapes was 75% fibres, 22% fragments and 3% foams, while in workplaces, the average composition was 70% fibres, 21% fragments and 9% foam. This was statistically indistinguishable from the average MPs shapes in atmospheric deposition (passive) samples which displayed an average of 66% fibres, 28% fragments and 6% foam in homes, and 63% fibres, 27% fragments and 10% foam in workplaces (Figure 3.2).

This is largely in agreement with previous studies, which reported fibres as the predominant MPs morphology in both active and passive indoor air samples from houses and workplaces from France (Dris et al., 2017b), Australia (Perera et al., 2023), China (Liao et al., 2021), and Sri Lanka (Perera et al., 2022b). Interestingly, one USA study reported fragments as the dominant MPs morphology in atmospheric deposition samples from a classroom and an office, while fibres were predominant (54% of total MPs) in atmospheric fallout samples from a family home (Yao et al., 2022). Overall, it could be concluded that MPs in indoor air can vary in morphology depending on the sources and types of MPs. Fibres, followed by fragments seem to be the predominant shapes of MPs in indoor air, which is largely reflecting their origins from carpets, textiles and domestic plastic items (Kek et al., 2024a).

Figure 3.2 MPs shapes (expressed as average percent contribution) in the studied UK microenvironments.

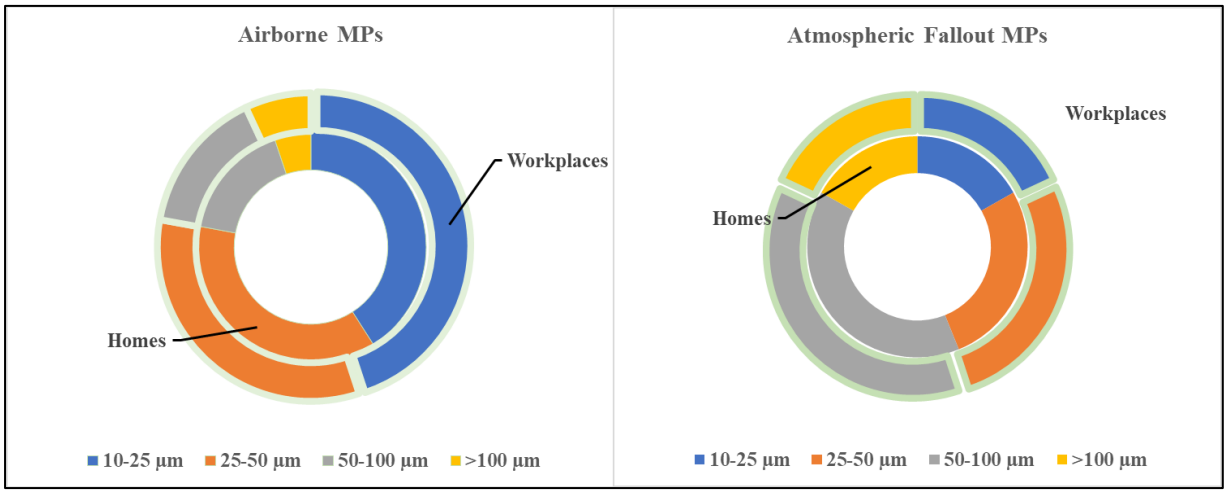


3.1.8.2 MPs Size:

The detected MPs in all the analysed samples were divided into 4 size ranges (Figure 3.3). In airborne MPs collected via active air sampling, particles in the size range (10- 25 µm) constituted > 40% on average in both homes and workplaces (Figure 3.3), and the particles abundance decreased with increasing particle size among the studied 4 size ranges. While the few existing studies on airborne MPs indoors have reported different size ranges, our results are generally in line with the previously observed trend of enrichment of smaller MP sizes in air (Kek et al., 2024a; Ageel et al., 2022; Torres-Agullo et al., 2022). This is largely concerning because smaller particles increase the potential risk of human inhalation. While our study could not measure MPs < 10 µm, a previous study from South Korea reported the size of airborne MPs originating from the deterioration of flooring materials could be below 2.5 µm, which poses a substantial threat to humans upon inhalational exposure (Yuk et al., 2022).

Interestingly, the MPs particle size distribution profile in atmospheric fallout particles, collected via passive sampling, was significantly different than airborne MPs from the same microenvironments (Figure 3.3). The predominant size fraction in atmospheric fallout particles was (50-100 µm) contributing >35% on average in both homes and workplaces. There was also significant higher contribution of larger sized particles (>100 µm) and lower contribution from smaller particles (10-25 µm) compared to the average particle size profile of airborne particles. This is in line with the findings of (Yao et al., 2022), who reported a median MPs size of 434 µm (range = 71 – 4950 µm) in atmospheric fallout particles collected from a classroom, hallway and office in USA. The difference in particle-size profiles between airborne MPs and atmospheric fallout particles from the same microenvironment may be attributed to a combination of factors including different polymer types, densities, as well as various MPs sources and atmospheric deposition rates.

Figure 3.3 Average particle size distribution of detected MPs in the studied UK homes (inner wheel) and workplaces (outer wheel).



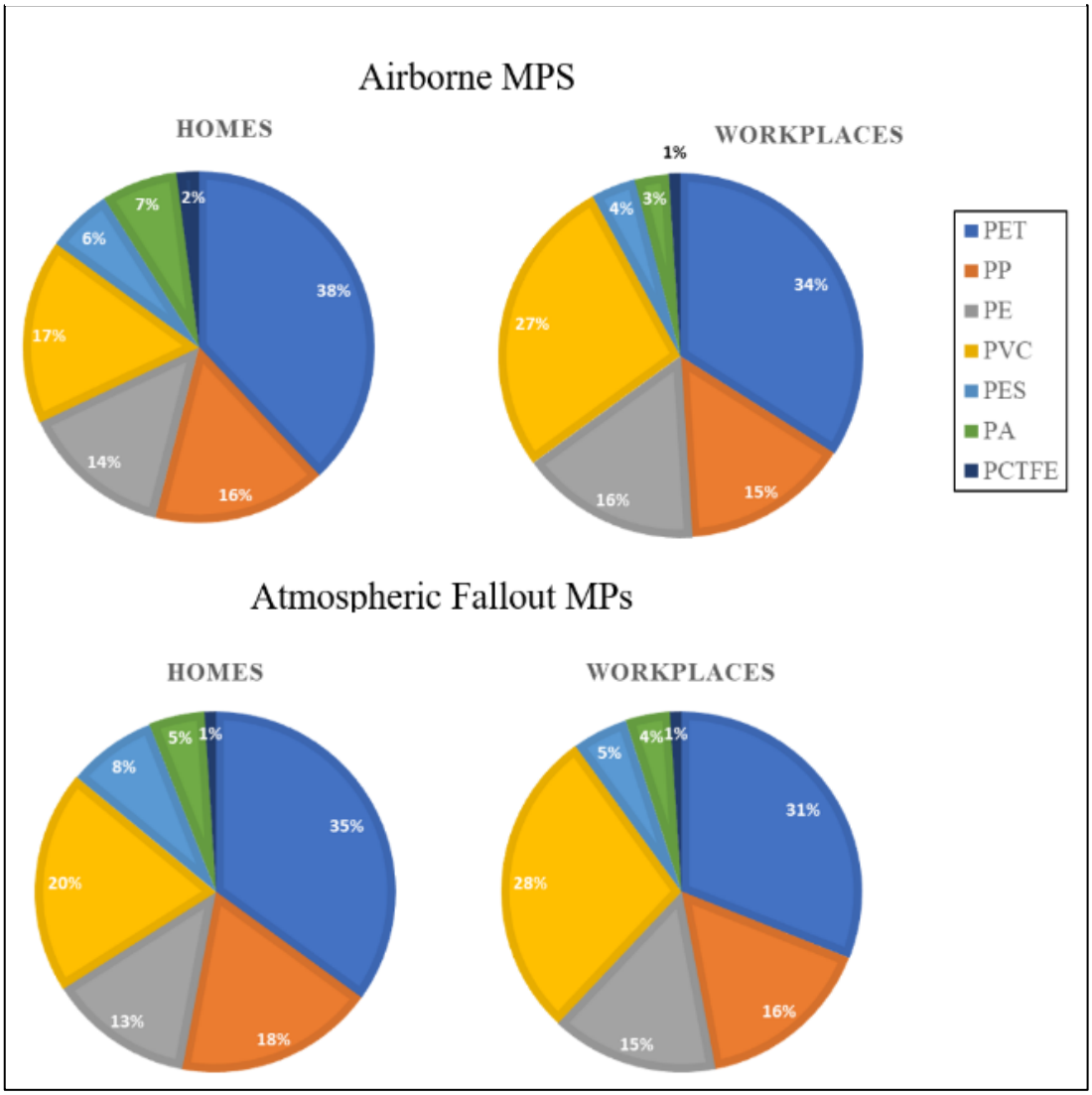
3.1.8.3 MPs Polymer types:

MPs particles were characterized by using µ-FTIR to identify polymer types. A total of 1400 particles were identified with polymer match >70%. Results showed that PET (polyethylene terephthalate) was the most abundant polymer type in all the studied samples (Figure 4). While PET constituted > 30% of airborne and atmospheric fallout MPs from UK homes and workplaces, PP (polypropylene), PE (polyethylene) and PVC (polyvinyl chloride) all contributed substantially (>10%) to the identified polymer types, while PES (polyester), PA (polyamide), and PCTFE (polychlorotrifluoroethylene) showed minor contributions (<10%) (Figure 3.4). Our results are in agreement with previous studies of MPs in indoor air, which highlighted PET as the major MPs polymer type indoors (Kek et al., 2024a; Ageel et al., 2022; Torres-Agullo et al., 2022; Perera et al., 2023). The identified polymer types in the current study are generally in line with previous studies on indoor MPs, which are often derived from synthetic textiles, furniture, clothing, paint, laminates, and household goods (Lim et al., 2022a; Soltani et al., 2021c). The identification of PCTFE in the identified MPs, albeit at low frequency, was interesting because this polymer has not been frequently reported previously. Further investigation revealed PCTFE to be applied for protection of Liquid Crystal Display

(LCD) units and other electronic components, as well as laminating PVC polymers, which may explain its presence in MPs from the studied UK microenvironments (Zou et al., 2022).

So far, the toxicological effects of MPs polymer types on human health are not fully understood, despite the noticeable concentration of MPs detected in several human tissues and excreta. Therefore, it's difficult to discern which MPs polymer type may exert more toxic impacts on human health upon inhalation (Shukla et al., 2024).

Figure 3.4 Average polymer type profiles (expressed as percentage distribution) of the detected MPs in the studied UK indoor samples.



PET = polyethylene terephthalate, PVC = polyvinyl chloride, PP = polypropylene, PE = polyethylene, PCTFE = polychlorotrifluoroethylene, PES = polyester, PA = polyamide.

3.1.9 Human exposure to MPs via inhalation of indoor air:

Recently, there has been a growing concern over human exposure to MPs in indoor air and the potential risk to human health (Wright et al., 2020). It's widely conceived that inhalation and ingestion are the major pathways for human exposure to MPs in indoor environments, while dermal contact may be a less considerable pathway (Zhang et al., 2020a, 2020c; Sharma et al., 2021; Prata, 2018). Although only particles with diameter ≤ 10 μm are considered inhalable theoretically, MPs fibres larger than that (width up to 29 μm, length up to 897 μm) have been observed to penetrate inside human lung tissue (Jenner et al., 2022). It was recently reported that the aerodynamic diameter of MPs, which is used to gauge the settling speed of particles of various densities and shapes, determines their settling depth in the respiratory system (Noorimotlagh et al., 2024). In the present study, we used the airborne MPs concentrations measured in the studied UK homes and workplaces to estimate inhalation exposure of adults and toddlers. The MPs concentrations were combined with typical time-activity patters for the UK population, as well as inhalation rates and body weights to provide a reliable estimate of inhalation exposure to MPs at a "Typical" (using average MPs concentrations) and "High-end" exposure scenarios (using 95th percentile concentrations of MPs in the studied microenvironments). For the ease of comparability with other studies, we provide our exposure estimates on a daily, annual and body weight bases (Table 3.2).

Our results show that on a daily basis, exposure of UK males (adults and toddlers) is higher than that of females (Table 3.2). This is in agreement with previous results from USA and Australia (Cox et al., 2019c; Perera et al., 2023), and can be explained by the higher respiration rates in males, which allows more air to the respiratory system with a mixture of particulate matter, including MPs (US EPA, 2011). Interestingly, when inhalation exposure is normalised to body weight, there remains hardly any difference in exposure between males and females because the higher respiration rates in males are offset by their higher body weights

compared to females (Table 3.2). On a body weight basis, inhalation exposure of UK toddlers to MPs was higher than that of adults (males and females), which is mainly driven by the lower body weights of toddlers. Nonetheless, this raises concern over the risk of such higher exposure to MPs in toddlers due to their incompletely developed immune and nervous systems (Kannan and Vimalkumar, 2021a).

On average, UK adults and toddlers (males and females) are exposed to 197 and 67 MPs/ day equating to 71,825 and 24,268 MPs/year via inhalation of indoor air. This is within the same order of magnitude to the exposure estimates reported for USA adults and toddlers based on an average airborne MPs concentration of 9.80 MPs/m3, which resulted in average inhalation exposure values for male and female toddlers of 40,225 and 35,338 MPs/year, and for male and female adults of 61,928 and 48,270 MPs/year (Cox et al., 2019c). In general, our results for UK population exposure to MPs via inhalation of indoor air are somewhere in the middle of the range of exposures reported worldwide (Table SI-5). Our average daily exposures are lower than those reported for Indian population (594 ± 269 MPs/day/person) (Yadav et al., 2022a) and higher than those reported for the Australian population (range = 1928 – 3187 MPs/year/person) (Perera et al., 2023). The differences between studies and the broad range of exposure estimates for MPs via inhalation of indoor air can be explained by a combination of factors including different sampling and analysis methodologies, various particle size ranges, different sources of MPs leading to large variations in concentrations among different microenvironments and microenvironment categories (e.g., homes, offices, schools, buses…etc) from different countries and even within the same country (Perera et al., 2023). Due to the lack of standard sampling and analytical method for determination of MPs in air, averaging MPs concentration data from different studies to obtain human exposure estimates can cause large uncertainties to the results (Cox et al., 2019; Kek et al., 2024).

It should also be noted that studies using passive sampling techniques (atmospheric fallout MPs) are more prone to measuring higher density and larger MPs that deposit on surfaces rather than remaining airborne for prolonged periods. While human activities indoors (e.g., walking, crawling, dusting) are likely to resuspend some of these settled particles to the human breathing zone (i.e., personal cloud), passive sampling data may overestimate the resulting human exposure (Ageel et al., 2022).

Table 3.2: Inhalation exposure estimates of UK adults and toddlers to MPs via indoor air.

	Male		**Female**	
Exposure (MPs/Kg Bw/day)	**Adult**	**Toddler**	**Adult**	**Toddler**
Typical	3.0	6.5	3.0	6.1
High-end exposure scenario	4.5	9.2	4.5	8.6
Exposure (MPs/Kg Bw/year)	**Adult**	**Toddler**	**Adult**	**Toddler**
Typical	1109	2368	1092	2211
High-end exposure scenario	1661	3365	1635	3142
Exposure (MPs/day)	**Adult**	**Toddler**	**Adult**	**Toddler**
Typical	217.3	68.8	176.2	64.2
High-end exposure scenario	325.3	97.7	263.8	91.2
Exposure (MPs/year)	**Adult**	**Toddler**	**Adult**	**Toddler**
Typical	79320	25101	64330	23434
High-end exposure scenario	118734	35672	96295	33303

Although the toxicological implications arising from human inhalation exposure to MPs are not yet fully understood, recent studies indicate the potential risk of such exposure. The risk of occupational exposure to Polypropylene (PP) MPs on respiratory health of workers was 3.5 times higher than that in the control group. TNF-α and serum IL-8 levels were increased in the plastic workers compared to the control group, while some workers also experienced diffuse ground glass attenuation and peribronchial thickening (Atis et al., 2005). In vitro studies on human lung cell lines revealed negative impacts on airway and alveolar epithelial linings upon exposure to nylon and polyester fibres, which negatively affected the

growth and development of human lung cells and organoids (Song et al., 2022). Moreover, the large surface area of MPs allows them to adsorb additional air pollutants, including heavy metals and polycyclic aromatic hydrocarbons, on their surfaces. This process may trigger the oxidative stress pathway, which may cause cytotoxicity, disrupts metabolism, causes inflammation in alveolar cells (Wright et al., 2020; Chen et al., 2023; Gasperi et al., 2018). In real-life exposure, the proportions of MPs cleared, deposited, or penetrated through an individual's lung not only depends on the properties of the MP particles but also on the physiology and condition of the individual's lungs (Carvalho et al., 2011). Therefore, vulnerable populations and individuals (e.g., babies, toddlers, and patients with impaired respiratory functions) may be at higher risk from inhaling airborne MPs compared to others.

3.1.10 Study limitations:

It is worth noting that indoor air sampling in the current study was conducted over 24 hours to facilitate comparison with previous studies and enable assessment of daily inhalation exposure. To our knowledge, there exists no data on daily, monthly or seasonal variations of MPs concentrations in indoor air. More research is required to study this variation and understand its potential impact on human inhalation exposure to MPs.

Our study is limited by the LOD of the μ-FTIR technique to 10 μm as the lower particle size limit. Smaller MPs (<10 μm) may exist in indoor air, which have the potential to penetrate deeper into human lungs; thereby pose high risk of potential adverse health effects (Kannan and Vimalkumar, 2021). Advances in sensitivity of analytical techniques will improve current capacity to measure MPs in the lower particle size range (<10 μm) and enable assessment of human exposure to these particles.

While our study identified carpeting as major source of MPs to indoor air, more research is required to investigate the impact of other factors (e.g., paintings, specific plastic materials,

textiles and lifestyle) on indoor MPs concentrations. Better understanding of such factors is crucial to advice policy on effective measures and to reduce human exposure to MPs indoors.

3.1.11 Conclusion:

This paper provides the first and most comprehensive study hitherto of MPs in indoor air via both active and passive sampling from 60 UK microenvironments. MPs were detected in all samples from all sites using both sampling methods. Statistical analysis revealed the mean concentration of MPs in UK homes was significantly higher than that in the studied workplaces. This was mainly attributed to carpeted floors in all the sampled homes, while 13 of the studied workplaces were uncarpeted. MPs concentrations in atmospheric deposition were significantly higher (P<0.05) than airborne MPs in the studied UK microenvironments. Nonetheless, a strong correlation (P<0.01) was observed between the concentrations of MPs measured by active and passive sampling in the studied homes and workplaces, which indicates common sources of MPs to both active (airborne) and passive (atmospheric fallout) samples collected from the same microenvironment. Fibres were the dominant MPs shape, contributing >60% to the detected MPS observed in all the studied samples. Fragments came second to fibres in terms of abundance, while foams were observed at a minor contribution (≤10%) in the analysed samples, which indicates the origins of these indoor MPs from carpets, textiles and domestic plastic items. Airborne MPs collected via active air sampling were dominated by particles in the size range (10- 25 μm), and the particles abundance decreased with increasing particle size. MPs in atmospheric fallout particles, collected via passive sampling were dominated by larger particles (50-100 μm) with lower contribution from smaller particles (10-25 μm) compared to airborne particles. Nonetheless, combined with the predominance of fibres, this raises concern over the risk from inhalation exposure to these particles because recent research found that MPs fibres in the observed size ranges can penetrate into human lung tissue. In terms of polymer types, PET and PVC were the most abundant in the studied samples followed by PP

and PE. Human exposure estimates revealed inhalation exposure of UK adults and toddlers to MPs in indoor air falls loosely in the middle of the range reported worldwide. On a body weight basis, UK toddlers were exposed to more MPs via inhalation of indoor air than adults, which raises concern over the potential risk to this vulnerable group with incompletely developed immune and nervous systems.

3.1.12 Supplementary Information:

Table SI3-1: Studies on airborne MP monitoring in various indoor environments.

References	Location	Indoor space	Sampling description	Abundance (unit)	Size range	Dominant MP's shape	Polymer type (%)
Maurizi et al., (2024)	Denmark	Meeting room, Workshop, New flat, Old flat	**Sampling:** Active pump sampler with a flexible plastic hose using a 13 mm ×200 µm Silicon filter **Sampling duration:** 24h **Analysis:** µ-Raman	• Meeting room: 92 MPs/m^3 • Workshop: 179 MPs/m^3 • New flat: 185 MPs/m^3 • Old flat: 548 MPs/m^3	1 – 67.5 µm	• Fragments (99.4%) • Fibers (0.6%)	• PA (21%) • PV (18%) • PE (16%) • PS (11%) • PEST (8%) • PP (6%)
Nandi et al., (2024)	Ranchi, India	Institute premises	**Sampling:** Active indoor air sampling with a flow rate of 2.9 L/min using a personal sampler **Sampling duration:** 24h **Analysis:** Stereomicroscopy & µ-FTIR	• 81730 ± 1923 particles/m^2/day	Fiber: 1.2 - 47 µm Fragment 0.9 - 16 µm Film: 4.1–9.6 µm	• Fibers • Fragments • Films	• PVC • Cellulose
Boakes et al., (2023)	Wardhurst, East Sussex, & London, UK	University College London & Private residence	**Sampling:** Active air sampling using a Burkard trap with an intake flow rate of 10L/ min on an adhesive-coated tape	• Rural: 344 Urban: 106 (MPs/m^3/day)	25 µm – 5 mm	• Fibre (77-88%)	• NA
Perera et al., (2023)	Australia	Childcare centre Elder's home School House Office	**Sampling:** Active air sampling with a stainless-steel air sampler with an intake air velocity of 100 ± 0.1 L/min and a 6	• 0.20 to 2.25 particles/m^3	20 – 5000 µm	• Fiber (98%) • Fragment (2%)	• PET (59%) • PE (25%) • PA (10%) • PP (3%)

		Vehicle Restaurant	cm diameter inlet point was used **Sampling duration:** 6h **Analysis:** Stereomicroscopy & μ-FTIR				• PAN (2%) • PVC (1%)
Torres-Agullo et al., (2022)	Barcelona, Spain	Public transport, workplace (storehouse/ laboratories), residential houses	**Sampling:** Active pump sampler with an intake flow rate of 10.0 ± 0.1 L/min and Nylon net filters (20 μm pore size, 25 mm diameter) **Sampling duration:** 1.5 hr-3 hr **Analysis:** Visual inspection & μ-FTIR	• Buses: (17.3 ± 2.4 MPs/m^3) • Subway: (5.8 ± 1.9 MPs/m^3) • Houses: (4.8 ± 1.6 MPs/m^3) • Workplace: (4.2 ± 1.6 MPs/m^3) •	<100 μm	• Public transport: Fiber (73 ± 9%) • Houses: Fiber (81 ± 4%) • Working areas: Fiber (53 ± 24%)	• PA (51%) • PES (48%) • PP (1%)
Field et al., (2022)	United Kingdom	Operating theatre, anaesthetic room	**Sampling:** Passive sampling of MP particle fallout with a glass beaker **Sampling duration:** 12hr **Analysis:** μ-FTIR	• Operating theatre: 0 – 9,258 (MPs/m^2/day) • Anaesthetic room:0 – 3,368 (MPs/m^2/day)	10–1000 μm; 10–250 μm (94%)	• Fragment (78%)	• PET (37 %) • PP (25 %) • PE (7 %) • Nylon (13 %) • PTFE (5%)
Cui et al., (2022)	Yangzhou, Jiangsu, China	Houses G1: Old folks + Young people + Child G2: Middle-aged + child G3: Young Couple G4: Old couple	**Sampling:** Passive sampling with a glass petri dish and filtered with 0.45 μm silver membrane **Sampling duration:** 24hr	• G1: 86,157 ± 36,524 • G2: 78,839 ± 28,765 • G3:88,877 ± 39,221 • G4: 96,367 ± 23,129 (MPs/m^2/day)	<50 μm	• Fragments (85%)	• PA (30.81–56.46 %) • PU (6.06–26.16 %) • PET (3.58–16.25 %) • PE (0.15–11.44 %)

			Analysis: LDIR & μ-FTIR				
Choi et al., (2022)	Seoul, South Korea	Apartment	**Sampling:** Active air sampling with a suction rate of 7 L/min **Sampling duration:** 48hr **Analysis:** μ-FTIR	• 3.02 ± 1.77 (MP particles/m^3)	20 - 6801.2 μm (Average size: 166.1 μm)	• Fiber	• PE (40%) • PP (24.6%) • PES (13.6%) • PA (9.9%) • PS (3.3%)
Amato-Lourenço et al., (2022)	São Paulo, Brazil	Medical school	**Sampling:** Passive sampling device with a glass surface of 0.48 m^2 coupled to a funnel, filtered with quartz fibre filter (1.6 μm) with a vacuum pump **Analysis:** μ-FTIR	• 307.2 ± 214.60 (MPs/ m^2/day)	100 - 200 μm	• Fragment (64%) • foam (23%) • film (9%) • granule (4%)	• PES fibre (100%) • PE (59%) • PP (26%)
Chen et al., (2022)	Taiwan	Nail Salon	Sampling: Cassette sampler with silver filter (25 mm, 0.2 μm) at an airflow rate of 9 L/min Analysis: FTIR	• 46 ± 55 (MPs/m^3)	<50 μm	• Fragment (NA)	• Acrylic (27%) • Rubber (21%) • PUR (13%)
Yao et al., (2022)	Newark, New Jersey, USA	Office, Hallway, Classroom, House	**Sampling:** Passive sampling of PM fallout with filter (2.2 μm, 47 mm) **Analysis:** Raman spectrometry, SEM	• House: $(1.96 \pm 1.09) \times 10^4$ • Hallway: $(1.44 \pm 0.07) \times 10^4$ • Office: $(6.43 \pm 2.54) \times 10^3$ • Classroom: $(6.20 \pm 0.57) \times 10^3$ (MPs/ m^2/day)	2-10 μm	• Office: Particles (86%) • Hallway & Classroom: Particles (67%) • House: Fibre (54%)	• PE (40%) • PET (26%) • PS (20%) • PP (7%) • PVC (7%)

Uddin et al., (2022)	Kuwait	Public buildings, mosques, hospital houses	**Sampling:** Active air sampling with a 6-stage compactor, at a flow rate of 30 L/min **Sampling duration:** 6hr **Analysis:** Nile red & Raman spectroscopy	• 3.2 – 27.1 (MP/m^3)	0.45– 2800 µm	• Fiber (91%) • Fragment (9%)	• PES (NA) • Nylon (NA) • PA (NA)
Soltani et al., (2021)	Sydney, Australia	Houses	**Sampling:** Passive sampling of dust fallout in a glass petri dish and filtered with glass fibre filter paper (0.6 µm pore size) **Sampling duration:** 30 days **Analysis**: µ-FTIR	• 22 to 6169 (MPs/m^2/day)	-	• Fiber (99%)	• PE (NA) • PES (NA) • PA (NA) • PS (NA)
Liao et al., (2021)	WenZhou City, China (urban & rural)	Apartments, offices, classrooms, hospital main corridors, transit station hall	**Sampling:** active pump sampler with an intake flow rate of 100 ± 0.1 L/min and glass microfiber filters (0.7 µm pore size, 90 mm diameter) **Analysis:** Nile Red staining/ imaging & µ-FTIR	• Apartment: 1583 ± 1180 • Hospital: 259 ± 67 (MP/m^3)	5–30 µm (60.4 ± 2.7%) 30–100 µm (28.5 ± 2.3%) >100 µm (11%)	• Fragment (89.6%) • Fibre (10.4%)	• PES (28.4%) • PA (20.54%) • PP (16.3%)
(Jenner et al., 2021c)	Yorkshire, UK	Homes and flats	**Sampling:** Passive sampling of dust fallout in glass beakers and filtered	• 902–14,551 MPs/m^2/day (mean 6204±3122)	5-5000 µm	• Fiber • Fragment • Film	• PET • PA • PAN

			with cellulose membrane filters (5 µm pore size). **Analysis:** Optical Microscope and µ-FTIR			• Foam • Sphere	• PMMA • PE • PP
Prata et al., (2020)	Aveiro, Portugal	Double-story house	**Sampling:** active air sampler with PM10 inlet at a flow rate of 5 L/ min on glass fibre filter (2.2 µm pore size) **Sampling duration:** 48hr	• 6 (MP/m^3)	17–3669 µm	• Fiber (NA)	• NA (NA)
O'Brien et al., (2020)	Australia	Laundry room	**Sampling:** Active sampling with a sampling volume of 55 m^3/h and glass filter (1.6 µm) **Analysis:** µ-FTIR & Pyr-GC/MS	• 1.6 ± 1.8 (MP/m^3)	764 µm	• Fiber (NA)	• PET (NA)
Zhang et al., (2020)	Shanghai, China	Office, Dormitory, Corridor	**Sampling:** Passive sampling using stainless-steel basins **Sampling duration:** 24 hr **Analysis:** µ-FTIR	• Dormitory: 9.9 × 10^3 • Office: 1.8 × 10^3 • Corridor: 1.5 × 10^3 (MPs/m^2/day)	50–2000 µm	• Fiber (NA)	• PES (NA) • Rayon (NA) • Acrylic (NA) • Cellophane (NA) • PP (NA) • PS (NA) • PA (NA)
Vianello et al., (2019)	Aarhus, Denmark	Apartments	**Sampling:** aluminum & glass fibre-made thermal manikin with breathing volume flow rate of 0.82 L/min	• 9.3 ± 5.8 (MP/m^3)	11–370 µm	• Fragment (87%) • Fibre (13%)	• PES (81%) • PE (5%) • Nylon (3%)

			Sampling duration: 24 hr **Analysis:** μ-FTIR				
Dris et al., (2017)	Paris, France	Private apartments, office	**Sampling:** Active air sampler pump with 8 L/min and quartz fibre filter (1.6 mm, 47 mm)	• 1-60 (MP/m^3) • 1586-11130 (MPs/ m^2/day)	50–3250 μm	• Fibre (NA)	• PP (NA)

*PA: Polyamide, PES: Polyester, PE: polyethylene, PET: Polyethylene terephthalate, PP: Polypropylene, PU: Polyurethane, PS: Polystyrene, PTFE: polytetrafluoroethylene; NA: Not Applicable

Detailed sample collection:

(a) Homes: The sampled homes comprised 21 houses (11 terraced, 8 semi-detached and 2 detached houses) and 9 residential flats (from 6 different building blocks).

The sampled homes were distributed in the following areas of Birmingham, UK: B17 (6 homes), B37 (1 home), B29 (7 homes), B32 (3 homes), B15 (3 homes), B31 (4 homes), B16(4 homes) and B23 (2 homes).

(b) Workplaces: The sampled workplaces comprised 13 shared offices, 10 individual (unshared) offices, 4 meeting rooms and 3 lecture rooms located within 22 different buildings distributed in the following areas of Birmingham UK: B15 (11 workplaces), B29 (5 workplaces), B4 (7 workplaces), B5 (3 workplaces), B6 (3 workplaces), and B24 (1 workplace).

Table SI3-2a: Number of MPs in the analysed blanks.

Blank type/no.	Active air sample	Passive air sample
Field 1	2	3
Field 2	1	2
Field 3	3	1
Field 4	1	2
Field 5	0	1
Field 6	0	3
Field 7	3	1
Field 8	1	3
Field 9	0	0
Field 10	0	0

Field 11	1	0
Field 12	1	0
Field 13	1	1
Field 14	0	1
Field 15	0	1
Field 16	2	3
Field 17	3	1
Field 18	1	2
Field 19	1	0
Field 20	0	3
Field 21	1	0
Field 22	1	3
Field 23	2	2
Field 24	3	2

Table SI3-2b: Recovery (expressed as %) of MPs in the QA/QC recovery samples.

Sample no	Active air sample	Passive air sample
Recovery 1	95	80
Recovery 2	95	85
Recovery 3	80	105
Recovery 4	90	95
Recovery 5	105	80
Recovery 6	90	85
Average	93	93

SD	14	8
Median	85	93
Min	80	80
Max	110	105

Human Exposure Assessment:

Concentrations of MPs determined in indoor air from homes and offices using active air sampling were used to estimate exposures of UK adults and toddlers via inhalation of airborne MPs. In brief, two exposure scenarios were considered: a "typical" exposure scenario using median MPs concentrations and the second a "high-end exposure scenario", assuming inhalation of the 95th percentile MPs concentrations in the studied microenvironments. Daily inhalation rates, and average body weights for adults and toddlers were obtained from the USEPA exposure factors handbook (US EPA, 2011), while typical time activity patterns for UK population were adopted from previously published studies in the UK and Ireland (Tao et al., 2016; Wemken et al., 2019a). Details of the exposure model and factors used in inhalation exposure calculations are provided below.

Daily exposure to MPs via inhalation was estimated for UK adults and toddlers by applying MPs air concentrations measured in this study in the following exposure algorithm:

$$\sum_{iex} = \frac{[(C_H \times F_H) + (C_W \times F_W) + (C_T \times F_T) + (C_P \times F_P) + (C_O \times F_O)]}{BW} \times R_i$$

where, $\sum_{iex}$ is the daily human inhalation exposure (MPs Kg BW^{-1} day^{-1}); C $_{H/W/T/P/O}$ is the concentration of MPs (MPs/m^3) in home/ workplace/methods of transport/public microenvironments/outdoors respectively; F $_{H/W/T/P/O}$ is the fraction for time spent in each microenvironment type; Ri is the daily inhalation rate (m^3/day); and BW is the body weight (Kg).

- The time-activity patterns for UK population were obtained from previously published studies, with reported typical activity patterns - i.e. for adults 63.8% home, 22.3% office, 5.1% PMEs, 4.1% car, and 4.7% outdoors; for toddlers 86.1% home, 5.1% PMEs, 4.1% car, and 4.7% outdoors (Tao et al., 2016; Abdallah et al., 2008).

Important to note is that human inhalation exposure estimated in the present study is based ONLY on the time fractions spent in home and workplace microenvironments. The other types of indoor microenvironments were NOT investigated in the current study; thereby were omitted from the calculations.

- The exposure parameters applied were obtained from the US EPA exposure factors handbook (US EPA, 2011), and are summarised in Table SI3-3 below:

Table SI3-3: Inhalation exposure parameters.

	Male		Female	
	Adult	**Toddler (1-2years)**	**Adult**	**Toddler (1-2years)**
BW (Kg)	71.5	10.6	58.9	10.6
Inhalation rate (m^3/day)	16.88	5.12	13.69	4.78

MPs concentration (MPs/m^3)	**Average**	**95th%ile**
Homes	15.6	22.17
Offices	13.1	22.99

- Two exposure scenarios were considered for both adults and toddlers: a "typical" exposure scenario using mean MPs concentrations and the second a "high-end exposure scenario", assuming inhalation of the 95th percentile MPs concentrations in the studied microenvironments.

Figure SI3-1 Representative examples of MPs shapes identified in UK indoor air samples.

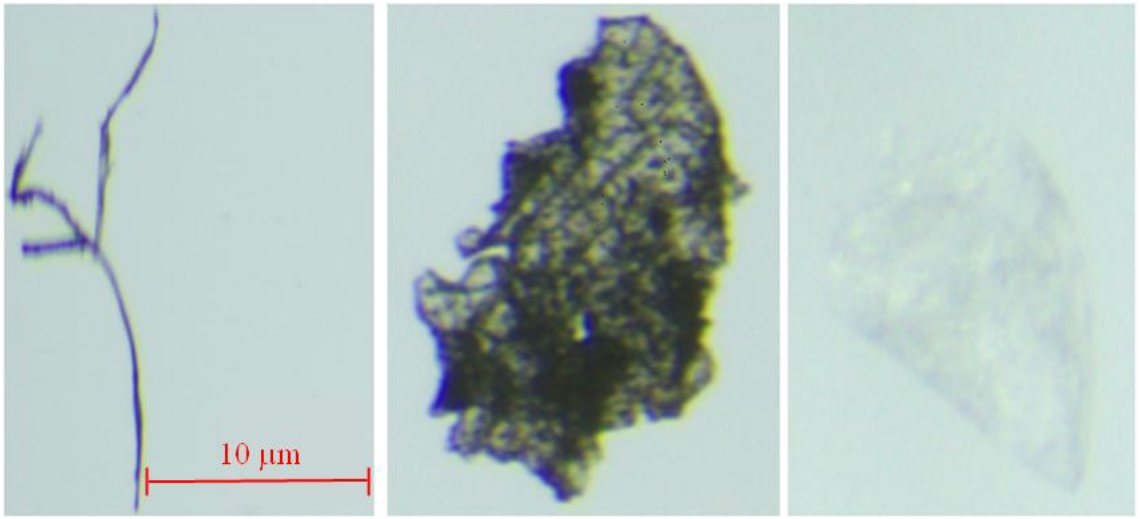


Table SI3-4: Airborne MPs concentrations (MPs/m^3) collected via active air sampling.

Homes Sample no#	Concentration (MPs/m^3)	Shapes percent (%) Fragment	Fiber	Foam
Home 1	20.4	27	72	1
Home 2	33.6	22	73	5
Home 3	13.7	23	77	0
Home 4	15.5	20	78	2
Home 5	22.3	16	83	1
Home 6	12.4	17	77	6
Home 7	14.2	16	66	18
Home 8	17.8	18	77	5
Home 9	19.9	20	75	5
Home 10	18.8	23	74	3
Home 11	16.5	20	69	11
Home 12	22	21	78	1
Home 13	14.5	25	69	6
Home 14	10.6	17	79	4
Home 15	12.2	22	77	1
Home 16	11.5	19	81	0
Home 17	11.1	25	75	0
Home 18	12.9	24	74	2
Home 19	20.2	26	69	5
Home 20	14.7	31	67	2
Home 21	12.2	29	70	1
Home 22	14.2	23	76	1
Home 23	11.8	22	74	4
Home 24	17.8	24	76	0
Home 25	16.9	19	79	2
Home 26	20.1	20	78	2
Home 27	9.1	21	76	3
Home 28	15.4	28	69	3
Home 29	3.8	24	74	2
Home 30	12.9	21	77	2
Average	**15.6**	**22.1**	**74.6**	**3.3**
STDEV	**5.4**	**3.78**	**4.19**	**3.67**
MAX	**33.6**	**31.0**	**83.0**	**18.0**
MIN	**3.8**	**16.0**	**66.0**	**0.0**
MEDIAN	**14.6**	**22.0**	**75.5**	**2.0**

Work Places Sample no#	Concentration (MPs/m^3)	Shapes percent (%) Fragment	Fiber	Foam	Carpet (Y/N)
Workplace 1	22.6	20	70	10	Y
Workplace 2	16.8	27	60	13	N
Workplace 3	12.5	26	74	0	Y
Workplace 4	16.7	8	78	14	N
Workplace 5	7.7	10	73	17	N
Workplace 6	13.5	22	59	19	N
Workplace 7	12.9	29	71	0	N
Workplace 8	7.4	19	65	16	N
Workplace 9	20.7	26	68	6	Y
Workplace 10	8.4	30	68	2	Y
Workplace 11	15.4	11	71	18	Y
Workplace 12	23.3	11	80	9	Y
Workplace 13	10.5	18	61	21	N
Workplace 14	7.2	26	74	0	N
Workplace 15	14.2	24	76	0	N
Workplace 16	8.4	32	66	2	N
Workplace 17	11.9	25	60	15	N
Workplace 18	3.2	15	82	3	N
Workplace 19	22.2	19	74	7	Y
Workplace 20	15	25	68	7	Y
Workplace 21	12.3	21	63	16	Y
Workplace 22	5.2	30	70	0	Y
Workplace 23	14.3	19	71	10	Y
Workplace 24	9.7	21	70	9	Y
Workplace 25	31	15	71	14	Y
Workplace 26	19.7	24	65	11	Y
Workplace 27	7.1	27	63	10	Y
Workplace 28	8.6	21	71	8	Y
Workplace 29	9.1	16	82	2	Y
Workplace 30	4.4	18	80	2	N
Average	**13.1**	**21.2**	**70.1**	**8.7**	
STDEV	**6.5**	**6.33**	**6.51**	**6.56**	
MAX	**31.0**	**32.0**	**82.0**	**21.0**	
MIN	**3.2**	**8.0**	**59.0**	**0.0**	
MEDIAN	**12.4**	**21.0**	**70.5**	**9.0**	

Table SI3-5: MPs atmospheric fallout concentrations (MPs/m^2/day) collected via passive air sampling.

Homes		Shapes percent (%)			Work Places		Shapes percent (%)			Carpet
Sample	Concentration (MPs/m^2/day)	Fragments	Fibers	Foam	Sample	Concentration (MPs/m^2/day)	Fragments	Fibers	Foam	(Y/N)
Home 1	5714	24	61	15	Workplace 1	4280	24	76	0	N
Home 2	5291	32	55	13	Workplace 2	5267	18	58	24	Y
Home 3	2504	27	64	9	Workplace 3	1939	32	56	12	N
Home 4	3422	21	75	4	Workplace 4	4879	27	72	1	Y
Home 5	4868	33	63	4	Workplace 5	2163	29	71	0	Y
Home 6	2504	18	77	5	Workplace 6	4033	16	77	7	Y
Home 7	2998	32	56	12	Workplace 7	3080	28	72	0	N
Home 8	3280	25	56	19	Workplace 8	2398	30	70	0	N
Home 9	5256	29	71	0	Workplace 9	9947	29	54	17	Y
Home 10	3951	34	66	0	Workplace 10	3398	35	64	1	Y
Home 11	5079	22	77	1	Workplace 11	2904	23	66	11	Y
Home 12	7513	19	77	4	Workplace 12	5056	35	53	12	Y
Home 13	3104	18	79	3	Workplace 13	1627	34	53	13	N
Home 14	1623	23	73	4	Workplace 14	987	28	54	18	N
Home 15	3527	23	74	3	Workplace 15	2998	25	57	18	N
Home 16	2787	37	55	8	Workplace 16	1834	27	54	19	N
Home 17	2116	34	55	11	Workplace 17	2869	22	73	5	Y
Home 18	3210	34	64	2	Workplace 18	1352	26	63	11	Y
Home 19	4792	24	71	5	Workplace 19	3434	34	58	8	Y
Home 20	4762	19	68	13	Workplace 20	3786	25	71	4	Y
Home 21	3175	31	65	4	Workplace 21	1362	26	55	19	N
Home 22	2540	33	67	0	Workplace 21	3856	32	53	15	N
Home 23	2646	28	68	4	Workplace 23	4500	31	56	13	Y
Home 24	5150	32	62	6	Workplace 24	3363	23	75	2	N
Home 25	4198	29	63	8	Workplace 25	5691	38	62	0	Y
Home 26	4374	29	61	10	Workplace 26	2387	21	70	9	Y
Home 27	1940	29	69	2	Workplace 27	706	16	55	29	N
Home 28	3810	31	69	0	Workplace 28	1705	26	74	0	N
Home 29	2081	28	65	7	Workplace 29	2340	25	54	21	Y
Home 30	3845	35	55	10	Workplace 30	1164	18	73	9	Y
Average	**3735**	**27.8**	**66.0**	**6.2**	**Average**	**3177**	**26.8**	**63.3**	**9.9**	
STDEV	**1343**	**5.56**	**7.38**	**4.92**	**STDEV**	**1860**	**5.73**	**8.63**	**8.25**	
MAX	**7513**	**37.0**	**79.0**	**19.0**	**MAX**	**9947**	**38.0**	**77.0**	**29.0**	
MIN	**1623**	**18.0**	**55.0**	**0.0**	**MIN**	**706**	**16.0**	**53.0**	**0.0**	
MEDIAN	**3474**	**29.0**	**65.5**	**4.5**	**MEDIAN**	**2951**	**26.5**	**62.5**	**10.0**	

Table SI3-6: Estimated human exposure to MPs via inhalation of indoor air by different studies.

References	Sampling Location	Indoor spaces	EDI of Human Exposure (Unit)
Maurizi et al., (2024)	Aalborg, Denmark	• Meeting room • New flat • Old flat • Workshop	3415 ±2881 Average daily exposure (MPs/day)
Perera et al., (2023)	Southeast Queensland, Australia	• Childcare Centers • Elder's Homes • Schools • Houses • Offices • Vehicles • Restaurants	• Ages 2 to 4 male: 2592 ± 551 • Ages 2 to 4 female: 2511 ± 532 • Ages 5 to 17 male: 2082 ± 601 • Ages 5 to 17 female: 1928 ± 549 • Ages 18 to 64 male: 3187 ± 594 • Ages 18 to 64 female: 2554 ± 471 • Age 65 or > male: 2978 ± 628 • Age 65 or > female: 2229 ± 324 Annual inhalation (n/year)
Yadav et al., (2022)	Mumbai, India	• Residential campus	594 ± 269 Average daily exposure (n/person)
Domenech and Marcos, (2021)	Data from multiple studies		2160 Annual intake (n/person)
Liao et al., (2021)	Eastern China	• Apartment • Office • Classroom • Hospital • Station	Adults: 103 Infants: 172 Newborns: 328 (MP/kg bw per day)
Cox et al., (2019)	Data from multiple studies		• Male children: 40225 ± 44730 • Female children: 61928 ± 68865 • Male Adults: 35338 ± 39296 • Female Adults: 48270 ± 53676 Annual inhalation (n/year)

Chapter 4

4.1 Human exposure to MPs via indoor dust ingestion; implications for human exposure:

Abstract

In recent years, microplastics (MPs) occurrence in indoor environments has raised significant concerns due to their distinctive physicochemical properties and potential toxicity. Indoor dust is well-documented as important vector for human exposure to emerging contaminants via inadvertent ingestion. Consequently, the main aim of the present study is to assess human exposure to MPs in settled indoor dust by determining concentrations, and characteristics (size, shape, and polymer type) of MPs in dust samples collected from houses (n=30) and workplaces (n=30) in Birmingham, UK. The average MPs concentrations were 155 ± 222 MPs/mg in homes, and 125 ± 209 MPs/mg in workplaces. Statistical analysis revealed the mean concentration of MPs in the studied homes was significantly higher ($P < 0.05$) than that in workplaces, which was attributed mainly to carpeting. In terms of morphology, various shapes of MPs (e.g., fiber, fragment, foam, foil) were identified, with fibres and fragments constituting ~90% of the identified MPs in all studied samples. Within the MPs particle size range (10 – 150 μm) investigated in the current study, the particle size fraction (50 - 100 μm) was predominant (> 49%) in all the studied samples, followed by particles >100 μm, with fibres having the largest average size (67 μm) among the detected MPs shapes. Polypropylene (PP) and Polyethylene Terephthalate (PET) were the most abundant polymer types in the studied samples, followed by Polyvinyl Chloride (PVC). Typical daily exposures of UK adults and toddlers to MPs through inadvertent ingestion of indoor dust were estimated at 56 and 103 MPs/day. The higher exposure of toddlers was exacerbated by their lower body weight with an estimated typical exposure of 9.7 MPs/Kg Bw/day, compared to 0.8 MPs/Kg Bw/day for adults.

The higher ingestion exposure to MPs in toddlers raises concern over potential adverse health effects due to their incompletely developed immune and nervous systems.

Keywords

Dust ingestion, Microplastics, Homes, Workplaces, Human exposure, Risk assessment

4.1.1 Introduction:

Because plastic goods are inexpensive, lightweight, chemically stable, impact-resistant, and resistant to corrosion, they are widely used in many aspects of contemporary life (Guo et al., 2020; Wong et al., 2020). According to Plastic Europe, 400 million tonnes of plastic were manufactured globally in 2022. The ubiquitous use of plastics will inevitably lead to environmental contamination. Plastic particles less than 5 mm are called "microplastics (MPs)," while those between 5 mm and 2.5 cm are referred to as "Mesoplastics", and those larger than 2.5 cm named "Macroplastics" (Jeyasanta et al., 2020). Because of their smaller size, recycling and removal of MPs from ecosystems, such as marine environments, is challenging (Thompson, 2006). Moreover, MPs can rapidly disperse into the atmosphere, water, sediment and soil, accumulating in different environmental compartments and reaching remote geographical regions. Consequently, MPs have attracted significant attention from the scientific community globally (Besseling et al., 2017; Sarker et al., 2020).

Based on their origin, MPs can be divided into two main classes: primary MPs, which are produced on purpose at a small size range for use in a variety of products like industrial abrasives and personal care products (like microbeads in face scrubs and toothpaste), and secondary MPs, which are created when larger plastic objects like bottles and packaging materials break down due to physical and environmental processes to produce smaller plastic particles in the MPs size range (i.e., <5 mm) (Bhat et al., 2023; Cai et al., 2018; Bhat et al., 2024).

Because plastic-based products are widely used in indoor residential space, this raises concern over MPs pollution in indoor settings due to the potential for generation of MPs from degradation/abrasion of plastic goods, furniture, and textiles. For example, fibres generated from synthetic polymer products—like textiles, clothing and carpets—due to regular household activities and natural wear and tear may constitute an important source of MPs in indoor environments (Lim et al., 2022). The possibility of MPs infiltrating into indoor dust results from a complex interaction between several factors. Dust is a prevalent element of the indoor environment and a proven vector of human exposure to different chemicals, microbes and particulate matter, including MPs (Kacprzak and Tijing, 2022). MPs can reach indoor dust from various input sources, such as abrasion of interior surfaces, textile, carpet and furniture fibre shedding, as well as infiltration of outdoor air, followed by deposition of MPs on indoor surfaces (Bhat, 2024). In addition, cooking can release particles containing microbeads into the air from non-stick cookware and plastic utensils and using personal care products can also introduce MPs directly into indoor surroundings (Guerranti et al., 2019; Fang et al., 2022).

Despite these possible routes for MPs to infiltrate indoor dust, there is still a lack of knowledge about their abundance, characteristics, and how they could affect human health (Eraslan et al., 2023; Bhat et al., 2022). This is compounded by the facts that humans spend almost 90% of their time indoors and do accidentally ingest indoor dust via inadvertent hand-to-mouth behaviour (Ageel et al., 2022). This is of further concern for toddlers due to their proximity to the floor, lower standards of personal hygiene, crawling movement, and increased hand to mouth behaviour, resulting in more inadvertent dust ingestion compared to adults (Ferguson et al., 2017). So far, only a small number of studies have investigated the presence of MPs in settled indoor dust from few countries around applied different analytical techniques, and used various assumptions to convert the measured MPs numbers to estimates human exposure via ingestion of indoor dust (Table SI4-1). Nonetheless, there remains crucial need

for further studies from different parts of the world, adopting the appropriate methodology and design to provide further information on human exposure to MPs via indoor dust ingestion and the risk arising from such exposure (Ageel et al., 2022; Kacprzak and Tijing, 2022).

The main aim of the present study is to assess human exposure to MPs via inadvertent ingestion of indoor dust and the risk arising from such exposure for the first time in the UK. The objectives of this study are: (a) to determining MPs concentrations, and characteristics (i.e., size, shape, and polymer type) from 30 homes and 30 workplaces in Birmingham, UK; (b) to assess human exposure of UK adults and toddlers to MPs via ingestion indoor dust; and (c) explore the potential risk arising from such exposure, based on the magnitude and characteristics of the estimated exposures. This investigation will assist in understanding the behaviour of MPs in the indoor environment and further evaluate the potential risk from exposure to MPs via indoor dust.

4.1.2 Materials and methods:

4.1.2.1 Sample collection:

A total of 60 settled indoor dust samples were collected from private homes (n=30), and various workplaces, including offices, meeting rooms, and lecture halls (n=30), in Birmingham, UK, between May 2022 and September 2023.

The tool used to collect indoor dust was a battery-powered, cordless vacuum cleaner (Karcher HV 1/1 BP CS 18v) with a specific design metallic tube attachment. Glass microfiber standard extraction thimbles (single thickness 25 mm × 100 mm, 1.0 μm pore size, Whatman, UK) were placed inside the vacuum tube attachment to collect individual indoor dust samples and to avoid any chance of cross-contamination. The sampling session took exactly 15 minutes at each site. After collecting the samples, the thimble was removed carefully and emptied into a 47 mm glass petri dish (BRAND®, Merck, UK) with the help of a disposable ECOFriendly®

plastic-free brush with natural bristles, then moved directly to a clean petri dish. The petri dish was closed with its specific glass lid, wrapped in clean aluminum foil, and secured with a paper-tape, then transferred to the laboratory for further analysis by the primary researcher. In the laboratory, petri dishes were stored in a cool (5°C), dark place until sample processing was conducted.

4.1.2.2 MPs extraction and density separation:

Extraction of MPs from indoor dust samples was adapted from the method developed at our UoB laboratories and described by Nel et al., (2019) to reduce sample contamination from the extraction process via the application of Zinc Chloride salt for density separation using the "Sediment Microplastic Isolation (SMI) units" (Figure SI4-1).

Prior to extraction, the collected dust samples were weighted on an accurate balance (LE324S, Sartorius, Germany), then sieved through a stainless-steel sieve (150 μm mesh size, 100 mm diameter, VWR International Ltd, UK), and the weight of the sieved dust was recorded as the size fraction relevant for human exposure via indoor dust ingestion.

The sieved dust sample was added to the SMI unit then 50 mL of the prepared $ZnCl_2$ solution (700 g/L) were added. The SMI unit was then covered with a clean aluminium foil and placed on a magnetic stirrer and allowed to stir for 10 minutes at a rotation frequency of 5 to 8 revolutions per minute (rpm). Thereafter, the unit was left to settle for 12 hours. Once the denser particles had settled out, the supernatant was poured over a 25 μm stainless steel mesh into a beaker for filtration and re-use. The upper chamber of the SMI unit was also removed and rinsed 3 times (with 25 ml deionised water, each) over the mesh to ensure that all particles clinging to the surface of the equipment are collected. The microplastics on the surface of the mesh were further washed 3 times (with 25 ml of deionised water, each) to remove any extraneous matter. The plastic samples were then transferred onto a clean glass microfiber filter

(47 mm, 1.0 μm pore size, Whatman, UK) using a vacuum filtration system comprising a Millipore® All-Glass Filter Holder Kit and a Welch™ 2522 WOB-L piston vacuum pump (Merck, Dorset, UK). The filtration mesh was rinsed 7 times with 15 mL of deionized water from a squirt bottle over the same filter to ensure no MPs were lost during sample transfer.

The choice of 25 μm mesh for filtration was based on previous literature reporting 25 μm as the "commonly reported" lower size limit for reliable chemical analysis using micro-FTIR (Huang et al., 2023; Jung et al., 2021). However, it should be noted that smaller particles could be identified in our dust samples using microscopic analysis. In addition, using our state-of-the-art micro-FTIR instrument in "spot analysis" mode enabled measuring particle sizes down to 10 μm. Detection of MPs sizes below the cut-off limit for filtration during sample preparation has been reported before and was attributed to the potential agglomeration of small particles on the filter or the ability of smaller particles to cling to the filter mesh material at such a small aperture size (Bhavsar et al., 2023; Jung et al., 2021; Agboola and Benson, 2021).

4.1.2.3 Chemical digestion:

Using 200 ml of 30% hydrogen peroxide (H_2O_2) (Fisher Scientific) to significantly reduce the particle count, a chemical digestion process was conducted to remove organic (non-MPs) particles. The mesh was rinsed with 30% hydrogen peroxide (H_2O_2) into 250 mL glass beakers (pre-washed with Milli-Q water). The beakers were then covered with aluminum foil and placed in a drying oven at 60 °C (Thermo Scientific, Fisherbrand® 65L, oven 230V, Germany) for 96 hours. At the end of 96 hours, the contents of the beaker were vacuum filtered on a glass microfibre filter, and the beakers rinsed to ensure that all particles clinging to its surface were collected (Dris et al., 2015; Liu et al., 2019d). The last step is to transfer the remaining MPs particles on the filter to clean glass petri dishes (54 mm, BRAND®, Merck™,

Dorset, UK), then place them in the oven again for 24 hours to be thoroughly dried and accurately weighted.

4.1.2.4 Instrumental analysis:

To determine MPs particle count, size and shape, a digital stereomicroscope (OPTIKA™ IM-3LD4D PROVIEW, Version: x86,4.11.20351.20220226) with multiple lenses Plan Fluor (4X, 10X, 20X, 40X) was applied. The microscope is equipped with a 6.3-megapixel USB 3.0 CMOS digital camera, operated through Microsoft Windows 7 system on a built-in 12-inch HDMI monitor. Visualisation and measurements of MPs were conducted using the bespoke "Optika™ ProView Image Analysis for fluorescence" software.

To confirm the identity and identify the polymer type of plastic particles, Perkin Elmer Spotlight^{TM} 400 Fourier-Transform InfraRed (μ-FTIR) imaging system with a remote-controlled stage was used. Reflectance mode was used to image and map the whole filter using the Spectrum MultiSearch^{TM} and SpectrumIMAGE^{TM} software. A 70 % match criterion and visual peak diagnostics were used to compare the generated spectra to those from the Perkin Elmer Microplastics library and/or the independent software tool (siMPle$^{®}$) to guarantee "best fit". Spectra were obtained with an aperture size of 20 × 20 μm, an accumulation of 16 scans, and a wavenumber range of 4000–600 cm^{-1} at a resolution of 8 cm^{-1}. When checked against the library spectrum, the particle was eliminated from the MPs count if the absorption spectra from 2850 to 3000 cm^{-1}, which originates from C–H vibrational stretching, did not occur. This indicates that the particle was not an organic molecule (Harley-Nyang et al., 2022; Mukotaka et al., 2021). The number, shape, and size of MPs on each filter were determined using microscope images taken by the μ-FTIR microscope in imaging mode, with point mode used to verify MPs at or near the LOD of 10 μm in size.

4.1.3 Estimated daily intake and Human exposure to MPs:

Human exposure to MPs in indoor dust was assessed specifically through the ingestion pathway (Nematollahi et al., 2022). To calculate the estimated daily intake (EDI; MPs/kg bw/day) of MPs ingested through indoor dust, the following general equation was applied (Dehghani et al., 2017; Mandin et al., 2017; Liu et al., 2019a).

$$EDI_{ing} = \frac{C \times f \times m_{di}}{BW}$$

where C is the determined MPs concentration in settled indoor dust samples (MPs/g); f is the time fraction spent in the sampled indoor microenvironment (*e.g., hours spent at home per day*)(Johnson-Restrepo and Kannan, 2009),; m_{di} (g/day) is the indoor dust ingestion rate for humans (USEPA, 2011); and BW is the body weight (kg).

The concentrations of MPs measured in indoor dust samples from UK homes and workplaces were used to assess UK adults' and toddlers' exposure via ingestion. Exposure was assessed based on two scenarios: first is a "typical exposure" using median MPs concentrations to reflect central tendency, and second is a "high-end exposure", assuming ingestion of MPs at the 95th percentile concentration in the analysed samples to reflect a worst-case scenario. Daily dust ingestion rates, and average body weights for adults and toddlers were obtained from the USEPA exposure factors handbook (US EPA, 2011), while typical time activity patterns for UK population were adopted from previously published studies in the UK and Ireland (Tao et al., 2016; Wemken et al., 2019a). Details of the exposure assessment model and different factors used for exposure estimations via indoor dust ingestion are provided in the supplementary information (table SI4-2).

4.1.4 Quality assurance and quality control:

In the context of microplastics research, preventing background contamination by MPs particles in the laboratory is crucial. To achieve this, researchers exclusively wore lab coats of pure cotton and nitrile gloves throughout the experimental process. Additionally, all sample processing occurred on a laminar flow bench, which was regularly checked and wiped down with 90% ethanol. Furthermore, before use, all laboratory equipment and consumables were meticulously rinsed with Milli-Q water. In investigating potential contamination during the sample analysis (including preparation and filtration), dust samples were systematically analyzed in batches of five. A blank sample comprising an unused thimble, unpacked on the filed (field blank) and treated as explained in the settled dust sampling section, was analysed alongside each batch of 5 dust samples. A “method blank” for the density separation method, comprising 15 mg of $ZnCl_2$ instead of indoor dust, treated as an MPs extraction and density separation section, was conducted alongside each 10 samples. Additionally, a “recovery sample”, consisting of 50 mg of clean sediment (Sigma-Aldrich), intentionally spiked with a known quantity (20) of PE-MPs particles (Cospheric™, California, USA), was processed alongside each 20 samples.

Detailed results of blanks and recovery samples are provided in Table SI4-3. Overall, the numbers of MPs in all blanks never exceeded 5% of the average concentration of MPs in the corresponding batch. Therefore, there was no need for blank corrections. The recovery of MPs in the recovery samples ranged between 78% and 114% (Table SI4-4), indicating good performance of the analytical methodology.

4.1.5 Statistical Analysis:

Two programs were applied to analyze the data during this study: Excel version (Microsoft Office 365) and Statistical Package for the Social Sciences (SPSS) IBM version

29.0.1. Through Excel, the quantitative levels and distribution patterns were determined using parameters such as average, standard deviation, median, minimum, and maximum. To assess whether each data set follows a normal distribution, the (Kolmogorov-Smirnov test) was used in SPSS. The outcomes were nonnormal distributed data. Also, the Independent-samples Mann-Whitney U Test was used to compare the differences between homes and workplaces. Potential correlation between generated data was tested using Pearson correlations with a p-value of < 0.05 set as a level of statistical significance. All data are outlined as mean ±SD, and all "differences" are mentioned to indicate a statistically significant $P < 0.05$.

4.1.6 Results and Discussion:

4.1.6.1 Concentration of MPs in indoor dust:

In this study, 60 indoor microenvironments (homes and workplaces) in Birmingham, UK were investigated. MPs were detected in all sites (Table 4.1). The average MPs concentrations were 154.7 ± 221.7 and 125.3 ± 209.1 (MPs/mg) in the studied UK homes and workplaces, respectively. Comparing the means by student t-test revealed the concentrations of MPs in the studied UK homes (n=30) were significantly higher ($P < 0.05$) than that in workplaces. This is in agreement with the findings of Zhu et al. (2022), who reported the mean concentration of MPs in residential apartments from Hangzhou City, China (1174 MPs/g; n=47) as the highest among other indoor microenvironments, including company offices (896 MPs/g; n=50), business hotels (843 MPs/g; n=53), University dormitories (775MPs/g; n=48), and University classrooms (209 MPs/g; n=44) (Zhu et al., 2022). Another study conducted in two cities from Iran reported higher concentrations of MPs in indoor dust from residential homes (74.0 ± 35.0 and 68.6 ± 36.1 MPs/mg; n=6) compared to university offices (63.6 ± 26.6 and 48.6 ± 8.6 MPs/mg; n=6) in Shiraz and Bushehr, respectively (Kashfi et al., 2022). The observed difference between MPs concentrations in indoor dust from homes and workplaces

in the present study may be attributed to carpeted flooring. while all the sampled homes were carpeted, 13 of the studied workplaces were not. This is confirmed by further statistical analysis showing the mean concentration of MPs in the non-carpeted workplaces was significantly lower (P <0 .05) than the that in carpeted workplaces (n=17) and the studied homes (n=30). This is also in agreement with Soltani et al. (2021) who documented a significant positive correlation between carpeted floors and increased concentrations of MPs in house dust from 32 Australian homes (Soltani et al., 2021a).

Table 4.1: Statistical summary of MPs concentrations (MPs/mg) in indoor dust measured in the studied UK homes (n=30) and workplaces (n=30).

Statistical parameter	Homes	Workplaces
Average	155	125
Standard deviation	222	209
Median	71	75
Minimum	8	10
Maximum	1120	1175

A survey of available literature revealed few studies have reported MPs concentrations in settled indoor dust on a mass basis (*i.e., MPs/mass of dust analysed*). Comparing our results to these relevant studies shows mean concentration of MPs in UK housed dust from our study is generally in line with those reported in 2 Paris apartments (Dris et al., 2017c) and 15 Chinese homes (Liu et al., 2019a) (Table 4.2). However, our results were higher than those reported in Peruvian houses Valdiviezo-Gonzales et al., (2024) and another study of Chinese apartments' indoor dust Zhu et al., (2022) (Table 4.2). In workplace dust, our results were higher than MPs concentrations reported in Chinese offices and Iranian University offices (Kashfi et al., 2022; Zhu et al., 2022). Elucidating the exact reasons for the observed differences in MPs concentrations in indoor dust among the few available studies is challenging. Understanding the underlying causes of these differences among studies from different countries is challenging

due to the large variation in sampling, extraction and analytical procedures for MPs detection/analysis. Combined with varying QA/QC protocols (e.g., blanks, matrix-matched recovery), these differences can strongly influence the particle size range of MPs reported in each study, as well as the accuracy of the results (e.g., μ-FTIR, and μ-Raman Spectroscopy vs. Nile Red staining), which can lead to uncertainty when comparing the outcomes (Ageel et al., 2022). Moreover, the current and previous studies have highlighted that MPs concentrations in indoor dust are impacted by room characteristics (*e.g., carpeting*), which may also be an underlying cause of variation among different studies, given the large variation in indoor microenvironment characteristics.

Table 4.2: Statistical summary of MPs concentrations (MPs/mg) in indoor dust from the present study compared to available relevant studies.

Location	Mean	SD	Median	Min	Max	Reference
UK, homes (n=30)	**155**	**222**	**71**	**8**	**1120**	**This study**
Iran, houses (n= 6)	71	36	NR*	35	102	(Kashfi et al., (2022)
France, apartments (n=2)	NR	NR	NR	190	670	(Dris et al., 2017c)
China, homes (n=15)	NR	NR	NR	17	620	(Liu et al., 2019a).
China apartments (n= 47)	1.2	0.9	0.8	0.2	3.9	(Zhu et al., (2022)
Peru houses (n= 60)	3	5	NR	0.1	34	(Valdiviezo-Gonzales et al., (2024)
UK workplaces (n=30)	**125**	**209**	**75**	**10**	**1175**	**This study**
China offices (n= 50)	0.9	0.8	1.1	0.1	1.5	(Zhu et al., (2022)
Iran offices (n= 6)	56	18	NR	43	76	(Kashfi et al., (2022)

**Not reported.*

4.1.6.2 MPs shape:

Our results show that four different shapes of MPs have been detected in the studied indoor dust samples, namely: fibres, fragments, foam, and foil (Figure SI4-2). Fibres and

fragments constituted more than 90% of the identified MPs in all studied samples (Figure 4.1). The existence of various morphotypes depends on both the sources of MPs and the mechanisms/processes leading to their formation from primary sources (Kek et al., 2024a) . In house dust, our results are in agreement with previous studies, with MPs shapes' abundance decreasing in the order fibres > fragments > foam > foil (Kashfi et al., 2022; Zhu et al., 2022; Abad-López et al., 2024; Nematollahi et al., 2022; Soltani et al., 2021a). A study conducted on dust from different Chinese indoor microenvironments identified MPs fibres, fragments, and films, where fibres were the predominant shape (> 80%), followed by fragments (~15%)

Interestingly, our results in dust from workplace sites showed the abundance of fragments was slightly higher than fibres (figure 4.1). This is to the contrary of the findings of Zhu et al. (2022) in dust from several Chinese microenvironments including offices, where the overall abundance of MPs shapes was dominated by fibres (> 80%), followed by fragments (~15%) (Zhu et al., (2022). However, our results are in agreement with a recent study on MPs in indoor dust from Saudi Arabia, where fragments were the predominant MPs shape (>60%) (Ahmad et al., 2024). This may be attributed to different primary sources of MPs to indoor dust, where, fibres likely originate from carpets, textiles, fabrics and clothing, while plastic products, including bags, toys, utensils and thicker plastic items are the likely sources of fragments (Zhang et al., 2020d).

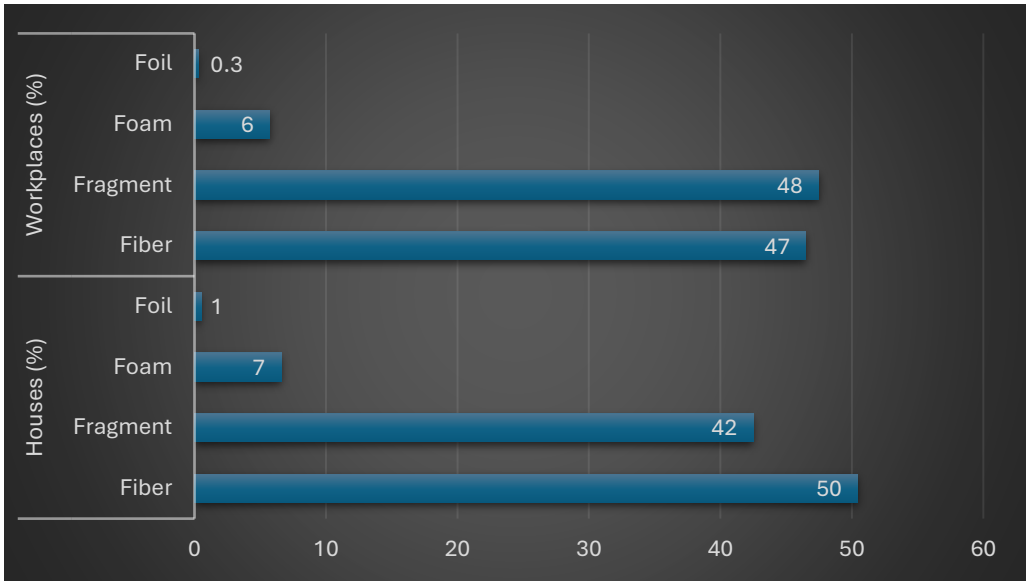


Figure 4.1 Average relative contribution (expressed as percent) of different shapes of MPs identified in dust samples from the studied UK homes and workplaces.

4.1.6.3 MPs size:

The detected MPs in the studied indoor dust samples were classified into 4 particle size ranges (Figure 4.3). Interestingly, similar MPs particle size distribution profiles were observed in dust samples from homes and workplaces, with predominant contribution from MPs in the range (50 – 100 µm), followed by particles >100 µm, while particles in the range (10 – 25 µm) had the lowest contribution (Figure 4.2). It should be noted that dust samples in the present study were sieved through 150 µm-mesh sieve, according to the USEPA exposure guidelines for "fine dust" which is more relevant for human exposure via inadvertent dust ingestion (USEPA, 2011).

On average, the largest size of all shapes in the studied samples was measured for fibers (mean = 67 µm; range = 20 to 132 µm). The smallest size range was observed for foam MPs (mean = 25 µm; range = 10 to 93 µm) (Figure 4.2, Table SI4-3). The average size of all MPs in home samples (50 µm) was greater than that in workplaces (37 µm), although not statistically significant ($P > 0.05$). Nonetheless, all the identified MPs particles in the present study were

in the “fine dust” range (<150 µm), with the potential for inadvertent ingestion by humans via hand-to-mouth behaviour (USEPA, 2011).

Large variation in MPs size ranges was reported by the few available studies in indoor dust. In Australia, a study conducted in 32 homes reported MPs in indoor dust in the size range of 200 to 400 µm (Soltani et al., 2021a). Another study on 26 workplaces from Shanghai, China identified MPs in the size range 50 to 2000 µm (Zhang et al., 2020). Also, a previous study in Surabaya, Indonesia on MPs in indoor dust from apartments, offices, and schools reported MPs to fall in the size range 3000 to 3500 µm (Bahrina et al., 2020). The variation in MPs particle size ranges reported by different studies from different countries may be, at least partly, attributed to a combination of factors including variations in sample collection and treatment procedures (e.g., sieving), MPs detection techniques (e.g., µ-FTIR vs SEM), as well as MPs sources to the microenvironments sampled (Kek et al., 2024a; Zhao et al., 2023).

Figure 4.2 Average particle size distribution of detected MPs in the studied dust samples from UK homes (inner wheel) and workplaces (outer wheel).

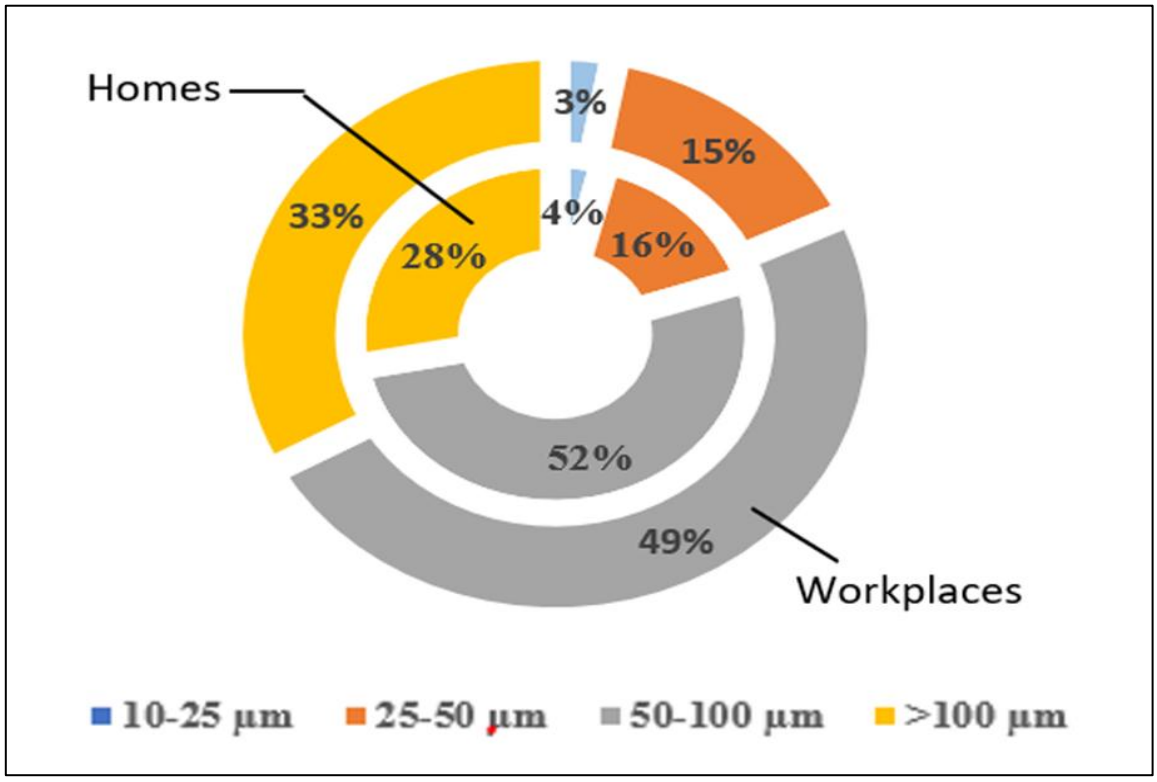
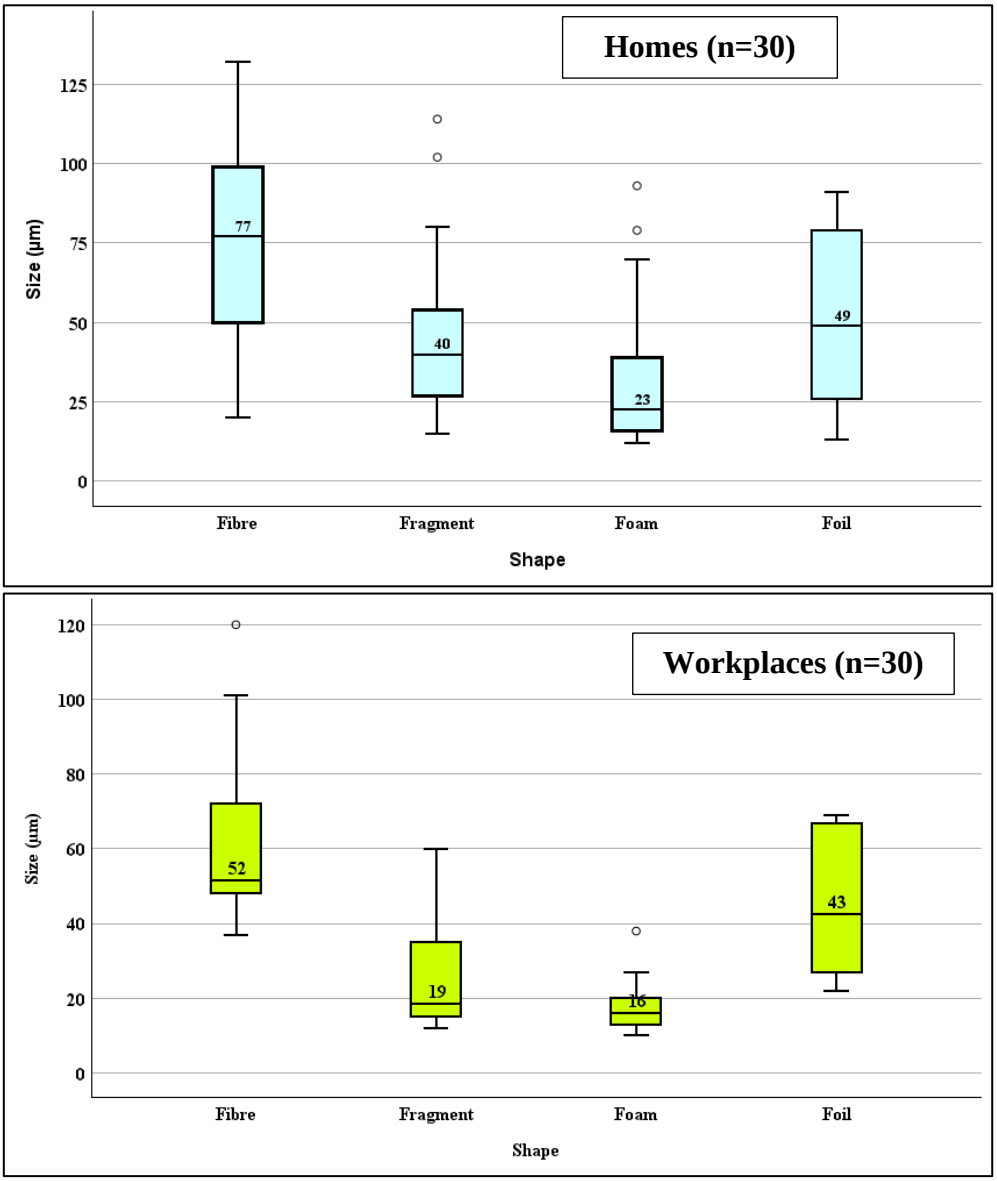


Figure 4.3 Boxplots of MPs size distribution for each shape category identified in the present study. Box lower and upper borders represent 25th and 75th percentiles, respectively, while the median is indicated by central line (and value).



4.1.6.4 MPs Polymer types:

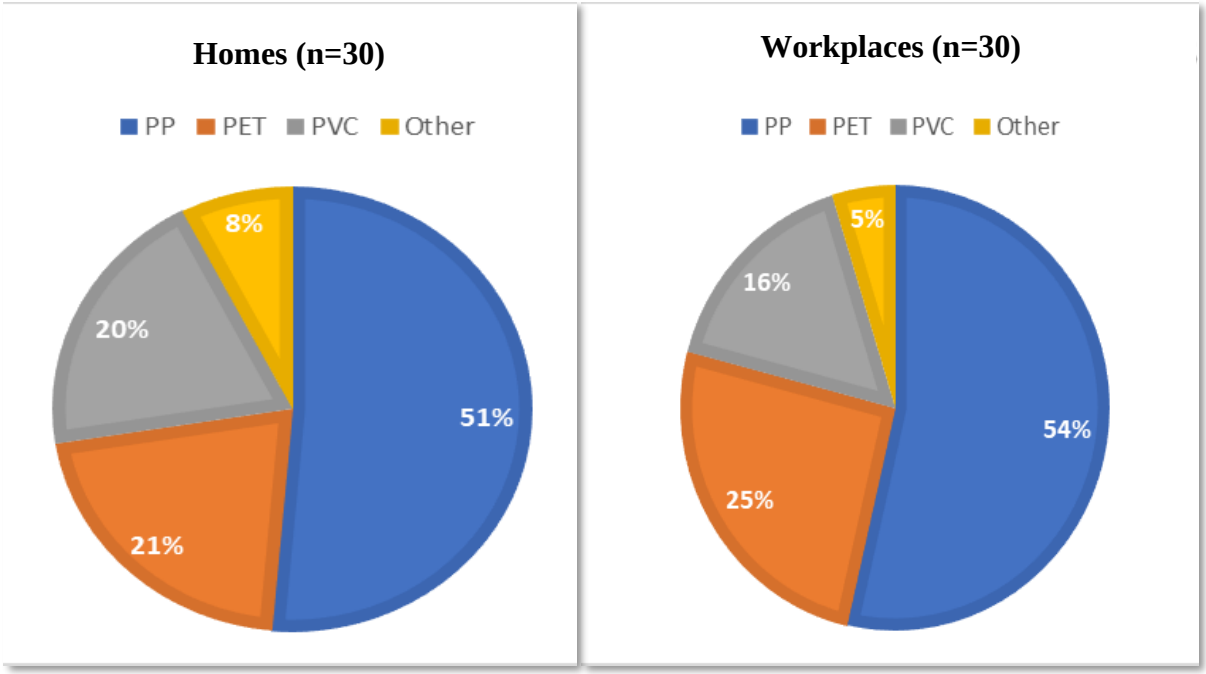
MPs particles were characterized by using µ-FTIR to identify polymer types. Results showed the most abundant polymer types of MPs in the studied indoor dust samples were polypropylene (PP), followed by polyethene terephthalate (PET) and polyethylene (PE), with smaller contributions from polyurethane (PU), polyvinyl chloride (PVC), polyester (PES), polyamide (PA), polyisoprene (PI) and PCTFE (polychlorotrifluoroethylene) (Figure 4.4).

These findings agree with previous studies of MPs in indoor dust, which identified similar polymer types as the current study (Soltani et al., 2021a; Nematollahi et al., 2022; Abbasi et al., 2022). In China, a study conducted in various indoor microenvironment categories indicated that PP was the most abundant polymer found in residential apartments and workplaces (Zhu et al., 2022). Another study performed in Iran showed that PET and PP had the highest contribution to the detected MPs polymers in indoor dust (Nematollahi et al., 2022). A recent study of MPs in house dust from Lima, Peru showed that PET, PP, and PVC were the predominant polymer types in the studied samples (Valdiviezo-Gonzales et al., 2024).

The polymer types identified in our current study align with findings from prior research and can be linked to potential sources of indoor MPs. PP is widely used in household items including kitchen utensils, toys, storage containers, appliances, food/beverage packaging and clothing, while PET is the main polymer in different types of packaging, as well as textiles and electronics. Similarly, other types of MP polymers identified in indoor dust in the current study can be tracked to various potential indoor sources including carpets, furniture foams, textiles, clothing, paint, laminates, electronic appliances and other household items (Kek et al., 2024a; Lim et al., 2022b; Soltani et al., 2021a)

It's worth noting that despite evident accumulation of MPs in various human tissues, understanding of the toxicological effects of different MPs polymers remains incomplete. Consequently, discerning which specific polymer type may have more pronounced adverse impacts on human health via a particular exposure pathway remains unclear (Shukla et al., 2024).

Figure 4.4 Average polymer type profiles (expressed as percentage distribution) of the detected MPs in the studied indoor dust samples.



4.1.6.5 Human exposure to MPs via indoor dust ingestion:

The concentrations of MPs in the studied samples were used to estimate daily intake of MPs via inadvertent ingestion of indoor dust for UK toddlers and adults at homes and workplaces. We used two exposure scenarios, one is "typical" based on average concentrations of MPs, and the other is "high-end" based on exposure at the 95th percentile concentration in the analysed samples (Table 4.3). Our "typical" estimates for UK adults (38 MPs/day) and toddlers (103 MPs/day) were slightly lower, yet within the same order of magnitude, compared to those reported for adults and toddlers from Punjabi cities (44 and 721 MPs/ day, respectively). On a body weight basis, our results for "typical" exposure of UK adults (0.8 MPs/Kg Bw/day) and toddlers (9.7 MPs/Kg Bw/day) were generally in line with those reported for Iranian adults (0.6 MPs/Kg Bw/day) and toddlers (3.2 MPs/Kg Bw/day) (Nematollahi et al., 2022), and slightly higher than those estimated for Chinese adults (0.23 MPs/Kg Bw/day) and toddlers (1.4 MPs/Kg Bw/day) (Zhu et al., 2022).

Interestingly, our estimated daily intakes for adults were higher at homes (typical exposure = 38 MPs/day), compared to workplaces (typical exposure = 18 MPs/day). This can be explained by the higher concentrations of MPs in home, compared to workplace dust in the present study, as well as the longer time spent at home. Interestingly, toddlers' exposure to MPs via indoor dust ingestion was higher than that of adults (Table 4.3). This is in agreement with previous studies on human exposure to MPs via indoor dust ingestion from different countries (Saygin et al., 2023; Aslam et al., 2022; Kashfi et al., 2022; Nematollahi et al., 2022; Soltani et al., 2021a), which may be attributed to a combination of factors including lower hygiene standards and proximity to the floor, leading to higher dust ingestion rates in toddlers, compared to adults. This is exacerbated by lower bodyweight in toddlers, leading to even higher exposure, compared to adults, on a body weight basis (Table 4.3).

While there exists hitherto no toxic thresholds, tolerable daily intakes or reference doses for human exposure to MPs via different exposure pathways, several *in vitro* models and laboratory animal studies highlighted potential adverse health effects arising from human ingestion of MPs (Blackburn and Green, 2022; Lin et al., 2023; Ageel et al., 2022). Ingested MPs can cause inflammation and irritation in the gastrointestinal tract, potentially leading to DNA damage and promoting cancer (Tang et al., 2024; Kadac-Czapska et al., 2024). Upon ingestion, MPs can interfere with production of gastric juices and mucous membranes, as well as gut microflora, leading to disrupted gastrointestinal functions (Fournier et al., 2021). Moreover, MPs can disrupt redox regulation of proteins in rats, indicating potential carcinogenic effect (Ding et al., 2024). Ingested MPs in mice were reported to cause damage to intestinal cell lining and trigger inflammatory reactions and oxidative stress, as well as increasing pro-inflammatory and pro-apoptotic factors in the colon, leading to overall weakening of the intestinal mucosal protective barrier (Thacharodi et al., 2024; Jia et al., 2023). Furthermore, some additives leached from MPs, such as phthalates, are known endocrine

disruptors that can impact the reproductive system (Ullah et al., 2023). Therefore, our results raise concern over the potential adverse effects arising from exposure to MPs via inadvertent dust ingestion, particularly the high exposure in toddlers due to incomplete development of their immune and nervous systems.

Table 4.3 Estimated daily intakes (MPs/kg bw/day) of MPs through indoor dust ingestion for UK adults and toddlers.

Scenario		Home	Workplace	∑Adult
Typical exposure[$]	Adult (MPs/day)	38.0	18.3	56.3
	Toddler (MPs/day)	102.6	-*	
	Adult (MPs/Kg Bw/day)	0.5	0.3	0.8
	Toddler (MPs/Kg Bw/day)	9.7	-	
High-end exposure	Adult (MPs/day)	157.8	39.9	197.6
	Toddler (MPs/day)	425.8	-	
	Adult (MPs/Kg Bw/day)	2.2	0.6	2.8
	Toddler (MPs/Kg Bw/day)	40.2	-	

[$]*Results presented in MPs/Day and MPs/Kg Bw/Day to facilitate comparison with other studies (Table SI-1). * Not estimated-irrelevant exposure for toddlers in the workplace.*

4.1.7 Conclusion:

In this study, 60 samples of settled indoor dust collected were investigated to determine the concentrations, and characteristics (*i.e., size, shape, and polymer type*) of MPs in UK homes and workplaces for the first time. MPs were detected in all samples with significantly higher concentrations in homes than in workplaces. This was mainly attributed to carpeting in all the sampled homes, while some of the studied workplaces were uncarpeted. Fibres and fragments were the dominant MP shapes, comprising >90% of the detected MPs in all the studied samples, while foams and foils were observed at a minor contribution (<10%). Fibres were the largest in size among the detected MPs shapes (mean = 67 µm), while foam MPs were the smallest (mean = 25 µm). In terms of polymer types, PP and PET were the most abundant in the studied samples, followed by PVC, which reflects potential indoor sources including carpets, textiles, household appliances, food/beverage packaging, furniture and clothing. Human exposure

estimates revealed exposure of toddlers to MPs via indoor dust ingestion was higher than that of adults, particularly on a body weight basis. Such high exposure to MPs raises concern over potential health risk to this vulnerable age group.

4.1.8 Supplementary Information:

Figure SI4-1 Sediment Microplastic Isolation (SMI) unit.

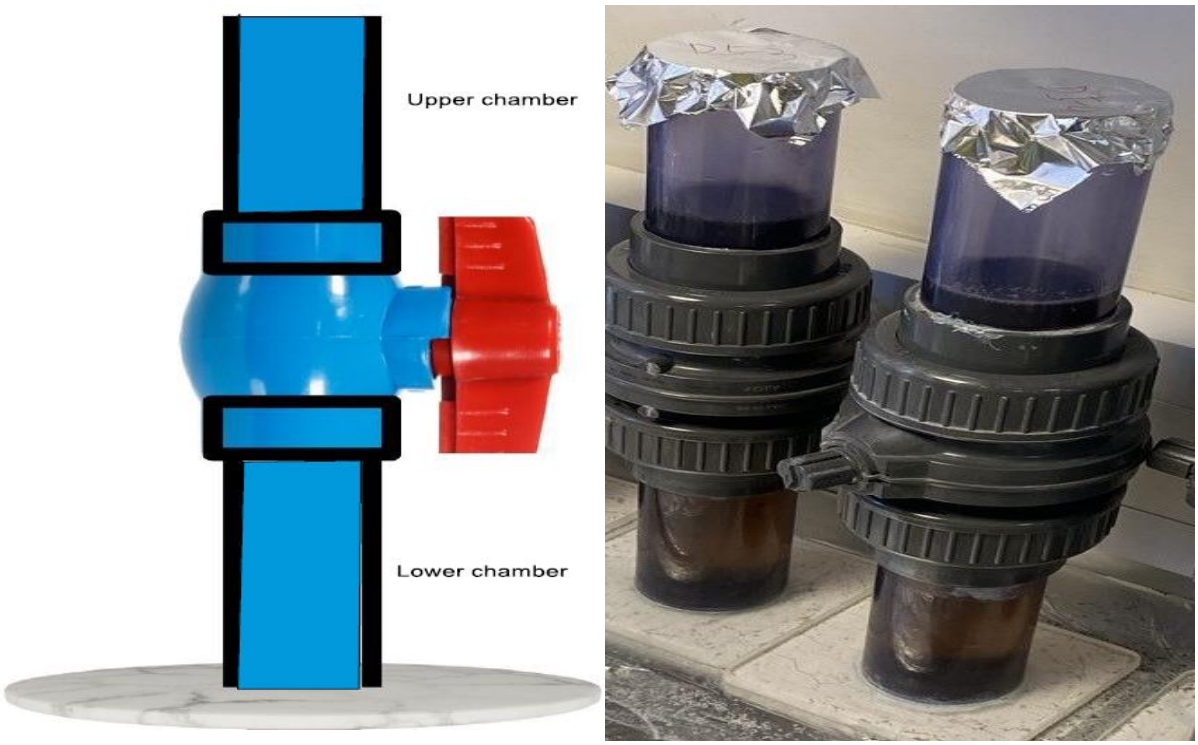


Figure SI4-2 Representative examples of MPs shapes identified in the present study.

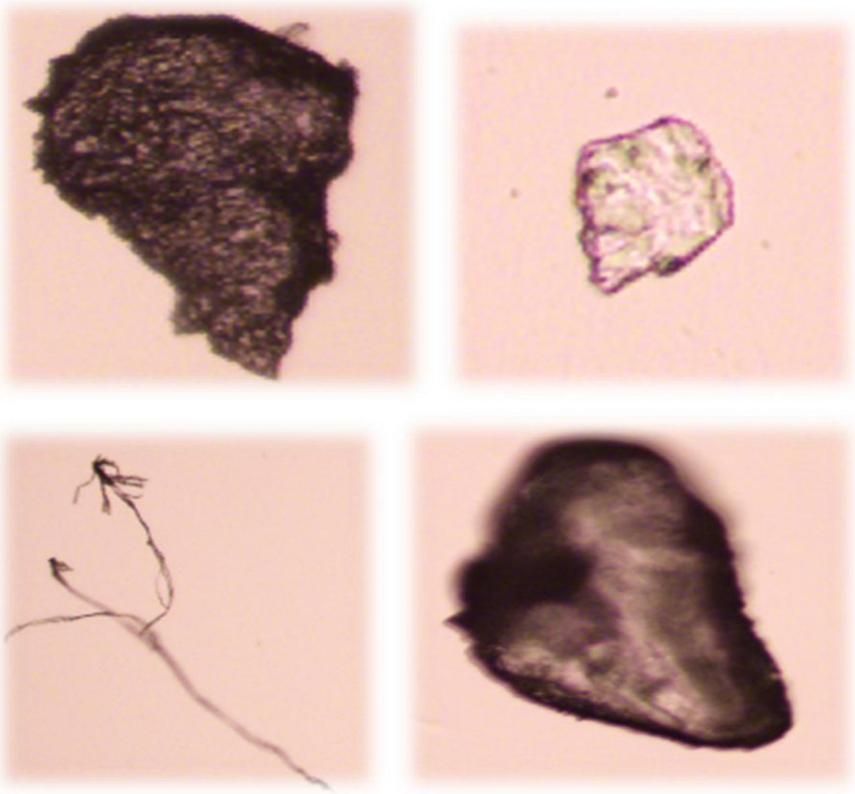


Table SI4-1: Studies on settled indoor dust MP monitoring in several indoor environments

References	Location	Indoor space	Analyzing methods	Mean Abundance (Unit)	Size range (µm)	Dominant MP's shape	Polymer type	EDI of Human exposure through ingestion (Unit)
Aslam et al., (2022)	Punjab, South Asia	• Houses in Lahore • Houses in Sahiwal	Stereomicroscope µ- FTIR	• 241.5 (items/m^2) • 159.1 (items/m^2)	100 – 350	• Fibers • Sheets • Films • Nurdles • Fragments	• PS • PET • PP • PE • PUR	**Ingestion** • Toddlers: 721 • Adults: 44 (particles/day)
Kashfi et al., (2022)	Bushehr and Shiraz, Iran	• Houses • Mosques • Kindergartens • Hospitals • Universities	SEM-EDS microscope	• 71.3 (items/mg) • 113 (items/mg) • 128 (items/mg) • 60.3 (items/mg) • 56.1 (items/mg)	>1000	• Fibers • Fragments • Fims	• PE (42%) • PC (28%) • PP (18%) • PET (10%) • Nylon (2%)	**Ingestion** • Infant: 0.48 • Toddlers: 0.11 • Children: 0.056 • Adults: 0.02 (items/kg-BW/day)
Nematollahi et al., (2022)	Shiraz, Iran	• Schools	SEM-EDS spectra	• 195 (MPs/g)	50 – 1000	• Fibers (99.7%) • Fragments (0.2%) • Sheet (0.1%)	• PET (50%) • PP (35%) • PS (10%) • Nylon (5%)	**Ingestion** • Infant: 13.7 • Toddlers: 3.2 • Children: 1.6 • Adults: 0.6 (MPs/kg/day)
Zhu et al., (2022)	Hangzhou City, China	• Apartments • Hotels • Offices • University Dormitories	Stereomicroscope µ- FTIR	• 1174(MPs/g) • 843 (MPs/g) • 896 (MPs/g) • 775 (MPs/g) • 209 (MPs/g)	201– 1000	• Fibers • Fragments • Films	• PE • PP • PES • PA • PC	**Ingestion** • Infant: 7.4 • Toddlers: 1.4 • Children: 0.49 • Adults: 0.23

		• University Classrooms					• PS	• University students: 0.22 (MPs/kg-BW/day)
Soltani et al., (2021)	Australia	• Houses	Olympus SZX12 stereomicroscope μ- FTIR	• 3095 (fibres/m^2 /day)	50 – 600	• Fibers • Fragments • Films	• PE • PA • PES • PAA • PS	**Ingestion** • Infant: 12.1 • Toddlers: 24.2 • Children: 59.7 • Adults: 64.5 (mg/kg-B_w/year)
Liu et al., (2019)	China	• Apartments	LC-MS/MS μ- FTIR	• 17 – 620 (fibers/mg)	50 – 2000	• Fibers	• PET • PC	**Ingestion for PET** • Infant: 89100 • Toddlers: 50400 • Children: 17300 • Adults: 6480 **Ingestion for PC** • Infant: 6.97 • Toddlers: 3.94 • Children: 1.35 • Adults: 0.51 (ng/kg-bw/day)

*PA: Polyamide, PAA: Polyacrylic acid, PES: Polyester, PE: Polyethylene, PET: Polyethylene terephthalate, PP: Polypropylene, PU: Polyurethane, PS: Polystyrene, PC: Polycarbonate, PTFE: polytetrafluoroethylene, PUR: Polyurethane, PVC: Polyvinyl chloride; NA: Not Applicable.

Table SI4-2: Number of MPs in the analysed blanks.

Blank type/no.	Indoor dust
Field 1	2
Field 2	5
Field 3	1
Field 4	3
Field 5	2
Field 6	3
Field 7	0
Field 8	0
Field 9	6
Field 10	2
Field 11	1
Field 12	2
Field 13	1
Field 14	0
Field 15	5
Field 16	2
Field 17	0
Field 18	0
Field 19	2
Field 20	3
Field 21	1
Field 22	4
Field 23	0
Field 24	1
Method 1	3
Method 2	0
Method 3	1
Method 4	3
Method 5	0
Method 6	2
Method 7	2
Method 8	3
Method 9	0
Method 10	1
Method 11	1
Method 12	0

Table SI4-3: Recovery (expressed as %) of MPs in the QA/QC recovery samples.

Sample no	Indoor dust
Recovery 1	85
Recovery 2	80
Recovery 3	110
Recovery 4	85
Recovery 5	110
Recovery 6	85
Average	88
SD	10
Median	83
Min	80
Max	105

Human Exposure Assessment

The time-activity patterns of the UK and Ireland populations were gathered from previously published research. Typical activity patterns were reported as follows: for adults, the patterns were 63.8% at home, 22.3% at work, 5.1% PMEs, 4.1% cars, and 4.7% outdoors; for toddlers, the patterns were 86.1% at home, 5.1% PMEs, 4.1% cars, and 4.7% outdoors (Tao et al., 2016; Wemken et al., 2019b).

The exposure parameters applied were obtained from the US EPA Exposure Factors Handbook (US EPA, 2011), and are summarised in Table SI-2 below:

Table SI4-4: Ingestion exposure parameters.

Age groups	Adult	Toddler
BW (Kg)	71.5	10.6
Ingestion rate (g/day)	0.05	0.10
MPs concentration (MPs/g)	**Average**	**95th%ile**
Homes	1191.7	4945.5
Workplaces	1644.2	3574.5

Table SI4-5: MPs concentrations (MPs/mg) and particle abundance (MPs/mg) in indoor dust samples.

Homes	Concentration (MPs/mg)	Shapes percent (%)				Workplaces	Concentration (MPs/mg)	Shapes percent (%)			
Sample		Fiber	Fragment	Foam	Foil	Sample		Fiber	Fragment	Foam	Foil
Home 1	45	85	14	1	0	Workplaces 1	41	73	24	3	0
Home 2	75	28	57	15	0	Workplaces 2	102	26	68	4	1
Home 3	212	66	32	2	0	Workplaces 3	49	70	27	3	0
Home 4	297	49	45	6	0	Workplaces 4	91	52	45	3	0
Home 5	17	61	33	6	0	Workplaces 5	40	53	44	2	0
Home 6	83	36	47	17	0	Workplaces 6	87	45	53	2	0
Home 7	188	43	43	15	0	Workplaces 7	26	28	69	3	0
Home 8	1120	56	38	5	1	Workplaces 8	10	43	46	11	0
Home 9	39	59	37	4	0	Workplaces 9	183	49	48	3	0
Home 10	88	48	37	14	1	Workplaces 10	13	49	45	6	0
Home 11	217	41	55	4	0	Workplaces 11	21	47	46	7	0
Home 12	321	44	51	4	1	Workplaces 12	145	34	56	9	2
Home 13	188	41	49	10	0	Workplaces 13	58	46	47	6	1
Home 14	51	80	15	3	2	Workplaces 14	113	64	32	4	0
Home 15	108	57	34	9	0	Workplaces 15	70	33	61	5	1
Home 16	297	32	63	5	0	Workplaces 16	100	53	41	3	2
Home 17	170	32	57	9	3	Workplaces 17	258	34	55	11	0
Home 18	16	43	53	4	0	Workplaces 18	160	52	43	5	0
Home 19	20	55	34	11	0	Workplaces 19	111	37	59	4	0
Home 20	280	35	56	8	1	Workplaces 20	32	40	56	4	0
Home 21	8	33	59	5	3	Workplaces 21	1175	41	56	3	0
Home 22	20	43	51	6	0	Workplaces 22	224	34	37	29	0
Home 23	12	65	31	4	0	Workplaces 23	39	45	47	7	0
Home 24	41	47	50	2	1	Workplaces 24	37	45	47	8	0
Home 25	19	43	45	10	2	Workplaces 25	16	36	48	17	0
Home 26	538	57	37	6	0	Workplaces 26	208	56	44	0	0

Home 27	49	67	32	1	0		Workplaces 27	70	57	42	1	0
Home 28	8	67	28	5	0		Workplaces 28	80	42	54	3	1
Home 29	50	53	43	4	0		Workplaces 29	157	48	48	3	0
Home 30	67	48	49	3	0		Workplaces 30	49	62	36	2	0
Average	**155**	**50**	**42**	**7**	**1**		**Average**	**125**	**47**	**47**	**6**	**0**
STDEV	**222**	**14**	**12**	**4**	**1**		**STDEV**	**209**	**12**	**10**	**6**	**1**
MAX	**1120**	**85**	**63**	**17**	**3**		**MAX**	**1175**	**73**	**69**	**29**	**2**
MIN	**8**	**28**	**14**	**1**	**0**		**MIN**	**10**	**26**	**24**	**0**	**0**
MEDIAN	**71**	**48**	**44**	**5**	**0**		**MEDIAN**	**75**	**46**	**47**	**4**	**0**

Table SI4-6: Average particle sizes (µm) of different MPs shapes identified in the studied indoor dust samples.

Homes	Concentrations (MPs/mg)	Average Shape size (µm)				Workplaces	Concentrations (MPs/mg)	Average Shape size (µm)			
Sample		Fiber	Fragment	Foam	Foil	Sample		Fiber	Fragment	Foam	Foil
Home 1	45	82	54	79	ND	Workplace 1	41	60	19	13	ND
Home 2	75	101	31	20	ND	Workplace 2	102	37	13	19	22
Home 3	212	79	50	22	ND	Workplace 3	49	47	19	14	ND
Home 4	297	99	40	23	ND	Workplace 4	91	52	12	13	ND
Home 5	17	99	45	93	ND	Workplace 5	40	59	16	16	ND
Home 6	83	20	102	70	ND	Workplace 6	87	39	14	10	ND
Home 7	188	28	114	66	ND	Workplace 7	26	61	19	10	ND
Home 8	1120	103	49	28	68	Workplace 8	10	49	15	11	ND
Home 9	39	24	19	17	ND	Workplace 9	183	48	15	16	ND
Home 10	88	101	42	21	79	Workplace 10	13	51	15	11	ND
Home 11	217	90	40	47	ND	Workplace 11	21	42	14	10	ND
Home 12	321	74	35	20	37	Workplace 12	145	97	40	19	29
Home 13	188	52	29	16	ND	Workplace 13	58	101	41	20	56
Home 14	51	77	71	60	91	Workplace 14	113	53	18	19	ND
Home 15	108	84	33	15	ND	Workplace 15	70	58	42	20	67
Home 16	297	77	80	39	ND	Workplace 16	100	72	60	38	69
Home 17	170	66	56	38	49	Workplace 17	258	73	27	18	ND
Home 18	16	132	37	16	ND	Workplace 18	160	120	45	18	ND
Home 19	20	40	19	15	ND	Workplace 19	111	51	15	14	ND
Home 20	280	50	23	15	13	Workplace 20	32	49	14	10	ND
Home 21	8	79	31	24	26	Workplace 21	1175	101	38	20	ND
Home 22	20	49	27	15	ND	Workplace 22	224	73	35	20	ND
Home 23	12	51	21	16	ND	Workplace 23	39	82	38	21	ND
Home 24	41	70	60	54	84	Workplace 24	37	66	35	27	ND
Home 25	19	55	27	12	20	Workplace 25	16	41	24	16	ND

Home 26	538	33	15	16	ND		Workplace 26	208	47	16	ND	ND
Home 27	49	103	56	32	ND		Workplace 27	70	46	20	15	ND
Home 28	8	100	41	38	ND		Workplace 28	80	50	15	23	27
Home 29	50	102	44	33	ND		Workplace 29	157	50	12	21	ND
Home 30	67	50	16	18	ND		Workplace 30	49	48	17	11	ND
Average	**155**	**72**	**44**	**33**	**52**		**Average**	**125**	**61**	**24**	**16**	**45**
STDEV	**222**	**28**	**24**	**22**	**30**		**STDEV**	**209**	**21**	**13**	**7**	**21**
MAX	**1120**	**132**	**114**	**93**	**91**		**MAX**	**1175**	**120**	**60**	**38**	**69**
MIN	**8**	**20**	**15**	**12**	**13**		**MIN**	**10**	**37**	**12**	**10**	**22**
MEDIAN	**71**	**77**	**40**	**23**	**49**		**MEDIAN**	**75**	**52**	**18**	**16**	**43**

Chapter 5

5.1 Seasonal variation of MPs in indoor environments from Birmingham, UK: Implication for ingestion and inhalation exposure

Abstract:

Over the last few years, there has been growing scientific and public interest in microplastics (MPs) as emerging contaminants. Previously, most research on MPs concentrated on their prevalence, transport, and fate in aquatic environments. Little is known about human exposure to MPs in the indoor environment despite the large fraction of time we spend indoors. This study investigates seasonal variation of MPs concentrations and characteristics in 5 indoor microenvironments for the first time and evaluates the impact of such variability on human exposure to MPs via inhalation and inadvertent dust ingestion. The average concentration of MPs airborne from the studied microenvironments was 9 ± 2.3 MP/m^3, while the average concentration of settled indoor dust MPs was 176.2 ± 96.7 MP/mg over 12-month monitoring period. Statistical analysis revealed significant higher concentrations ($P < 0.05$) of MPs in indoor air and dust in summer compared to autumn and spring, respectively. Moreover, substantial seasonal variation was observed in indoor MPs profile characteristics (shape and size distribution) in the 5 studied microenvironments, with several of these differences proving statistically significant at the 95% level. The observed differences may be attributed to a multitude of factors including ventilation, cleaning frequency, humidity levels, variable indoor sources and human activity leading to release of secondary MPs from wear and tear of indoor plastic items. Nonetheless, the observed differences have led to statistically significant seasonal variability in estimated daily intakes for adults and toddlers exposed to MPs via inhalation and inadvertent dust ingestion indoors. This indicates that prolonged monitoring studies are required to accurately represent human exposure to MPs indoors and raises concern over the

representativity and comparability of existing human exposure data from different studies based on single “spot” samples collected from various indoor microenvironments in different seasons.

Keywords:

Microplastics, Human exposure, Seasonal variation, Dust, Air, Indoor

5.1.1 Introduction:

Microplastics (MPs) are defined as plastic particles smaller than 5 mm in diameter (Baker et al., 2008). MPs pollution in ecological systems, encompassing marine environments, soil, and air, has emerged as a significant global contaminant in recent years due to its propensity to accumulate in various environmental compartments and the resilience of plastics to chemical and biological degradation (Yao et al., 2019b; Duis and Coors, 2016; Wu et al., 2019d; SAPEA, 2019c). However, contemporary research in this domain has predominantly focused on the characterization, fate, and behavior of MPs in marine ecosystems (Ageel et al., 2022). In contrast, investigations into MPs in indoor environments remain limited relative to studies on their presence in aquatic and terrestrial outdoor environments (Gall and Thompson, 2015a; Ageel et al., 2022; Bhat, 2023).

Plastic products are extensively utilized in indoor residential environments, leading to concerns that their excessive use may contribute to MPs pollution due to degradation and abrasion of items such as furniture, clothing, utensils and other plastic materials. For instance, fibers released from synthetic polymer products like textiles, apparel, and carpets through laundering, other domestic activities, and natural wear and tear, can be substantial sources of MPs in indoor settings (Lim et al., 2022b). The potential infiltration of MPs into indoor dust results from a complex interplay of several factors. Indoor dust, a ubiquitous component of the indoor environment, serves as a well-known conduit for human exposure to various agents,

including chemicals, microbes, and particulate matter. Among these contaminants, MPs are increasingly recognized as constituents of indoor dust, warranting further investigation (Kacprzak and Tijing, 2022). Additionally, cooking processes may emit secondary MPs into the surrounding environment from non-stick cookware and plastic utensils, while using some personal care products (PCPs), e.g., hair sprays and cleansing products can also introduce MPs directly into the environment (Guerranti et al., 2019; Fang et al., 2022). The levels of indoor MPs are further influenced by carpeting, ventilation, and airflow dynamics, which were reported to significantly impact MPs abundance and distribution within indoor microenvironments (Kek et al., 2024b).

MPs are diverse anthropogenic pollutants characterized by varying polymer compositions, sizes, forms, densities, colours, chemical additives, and by-products resulting from plastic degradation (Rocha-Santos and Duarte, 2015). Moreover, MPs have displayed substantial capacity to adsorb toxins within the ecosystem, including hydrophobic organic pollutants such as polycyclic aromatic hydrocarbons, polychlorinated biphenyls, and hexachlorocyclohexane isomers, as well as heavy metals like cadmium, zinc, nickel, and lead (Foulon et al., 2016). Concerns have been raised over the risks that MP exposure poses to human health. Numerous studies have documented the absorption and accumulation of MPs in humans, particularly those smaller than 50 μm, in vital tissues and organs such as the liver, colon, lungs, ovaries, testes, placenta, and spleen (Li et al., 2023; Montano et al., 2024), despite a recent analysis by the WHO indicates that evidence linking MPs to significant adverse effects on human health remains limited (WHO, 2022b).

This limitation is primarily due to substantial gaps in our understanding of the mechanisms and extent of toxicological impacts caused by MPs in various human tissues, as well as the exposure thresholds necessary to trigger such impacts. Current scientific knowledge suggests that MPs may exert toxicity in humans and mammals through oxidative stress, DNA

damage, metabolic disturbances, immunotoxicity, reproductive impairment, and neurotoxicity (Li et al., 2023b).

Human exposure to MPs can occur through multiple pathways, including inhalation of both indoor and outdoor air, ingestion via food, beverages, and settled indoor dust, and dermal contact with fabrics, personal care items, and atmospheric deposition (Prata et al., 2020b; Ageel et al., 2022). However, there remains a significant knowledge gap regarding human exposure to MPs through inhalation and ingestion within indoor microenvironments (Dris et al., 2017a; Yao et al., 2022). Additionally, only a limited number of studies have quantified the Estimated Daily Intake (EDI) of MPs across various exposure pathways in indoor settings. These studies have indicated that toddlers inhale and ingest higher quantities of MPs compared to adults across several microenvironments (Ageel et al., 2024; Al-Mansoori et al., 2025; Nematollahi et al., 2022; Abbasi et al., 2022).

While understanding of human daily intake of MPs via different exposure pathways, particularly indoors, remains at its infancy. Very little is known about how representative the existing few studies are in terms of assessment of such exposure. Currently available studies assessing human daily intake of MPs indoors are all based on “spot sampling”, which relies on collecting a given sample, or set of samples, from a particular indoor microenvironment at a specific point in time. The concentrations of MPs in these “spot samples” are then applied to estimate human exposure (Dris et al., 2017a; Yao et al., 2022; Ageel et al., 2024; Nematollahi et al., 2022; Abbasi et al., 2022). While such approach is informative, very little is known about seasonal variation of MPs in indoor microenvironments and its potential implications for human exposure. The current study provides first assessment of seasonal variation in MPs concentrations in indoor dust and air and its implications for human exposure via ingestion and inhalation.

The main objectives of this study are: (a) to investigate the seasonal variation of MPs in indoor environments, encompassing settled indoor dust and indoor air (both airborne and atmospheric deposition MPs); and (b) to assess the impact of seasonal variation on human exposure to MPs through inhalation of indoor air and ingestion of settled indoor dust. This will be achieved by determining the concentrations and characteristics (size and shape) of MPs on a monthly basis for a whole year (from January to December 2023), across five microenvironments (2 homes and 3 offices) in Birmingham, UK.

5.1.2 Materials and methods:

5.1.2.1 Sample collection:

Five sites were chosen for seasonal variation sampling to collect indoor samples, including air (airborne and atmospheric deposition) and settled dust. The chosen sites comprise two homes and three offices constituting a sample of convenience from Birmingham, UK. The sampling process was conducted every month from January 2023 to December 2023. The collecting samples date was the end of each month. The total number of indoor samples generated is 180 (5 sites × 3 sample types × 12 months).

Indoor air:

Two techniques were used in this study for indoor air sampling: active air sampling and atmospheric deposition passive sampling (Shao et al., 2022; Liu et al., 2019a). The low-volume active air sampler comprised a low-volume pump (Capex L2X, Charles Austen, UK), operated at a flow rate of 6 L min^{-1} for 24 hours to yield a single sample comprising 8.64 m^3 air. The particulate phase was collected by employing a 47 mm diameter membrane glass fibre filter (1.0 μm pore size, Whatman, UK) housed in a stainless steel in-line 47 mm filter holder (Sartorius® 16254, Goettingen Germany). The designated flow rate of 6 L min^{-1} was achieved and maintained by using a flow meter (Platon 12 L min^{-1}) connected to an adjustable valve.

The flow meter was calibrated at the start and the end of each experiment using a Gilibrator® air flow calibrator (Sensidyne, Inc., Clearwater, Florida, USA), which is classified as a primary standard device. The passive air sampler was used to capture indoor airborne microplastics fall-out (atmospheric deposition), by applying an open glass Petri dish (20 cm diameter, Duran® soda-lime glass, VWR International ltd., Leicestershire, England) placed at head height (1–1.5 m) within the selected sites. The glass Petri dishes were only opened on site at a set date and time. In each sampling session, the Petri dishes were exposed to atmospheric fallout for 24 hours to collect dry and wet MPs atmospheric deposition fallout, and participants were instructed to not change any of their habits or routines. At the end, the petri dish was closed with its specific glass lid, wrapped in clean aluminium foil, and secured with paper-tape, then transferred to the laboratory for further analysis by the primary researcher. In the laboratory, Petri dishes were stored in a cool (5°C), dark place until sample processing was conducted.

Settled indoor dust:

The tools used to collect samples were a battery-powered, cordless vacuum cleaner (Karcher HV 1/1 BP CS 18v) with a specific design metallic tube attachment. Glass microfiber standard extraction thimbles (single thickness 25 mm × 100 mm, 1.0 μm pore size, Whatman, UK) were placed inside the vacuum tube to collect individual indoor dust samples and to avoid any chance of cross-contamination. The sampling session took exactly 15 minutes at each site. After collecting the samples, the thimble was removed carefully and emptied into a 47 mm glass petri dish (BRAND®, Merck, UK) with the help of a disposable ECOFriendly® plastic-free brush with natural bristles, then moved directly to a clean petri dish. The petri dish was closed with its specific glass lid, wrapped in clean aluminum foil, and secured with a paper-tape, then transferred to the laboratory for further analysis by the primary researcher. In the laboratory, petri dishes were stored in a cool (5°C), dark place until sample processing was conducted.

5.1.2.2 Samples Preparation and MPs extraction:

Active air samples:

The glass microfiber filter (47 mm) was carefully removed from the stainless steel in-line 47 mm filter holder (Sartorius® 16254, Goettingen Germany), transferred to a clean glass petri dish (60 mm, BRAND®, Merck™, Dorset, UK) containing 1 ml of H_2O_2 solution (30% w/v, Merck™, Dorset, UK), which was enough to submerge the filter and particles on it. The petri dish was then placed in a plastic-free oven at 60℃ for 48 hours to oxidise any organic particles on the filter (digestion step). The digested filters were then covered by their pre-cleaned and dried lids in a clean dry place until further microscopic examination.

Passive air samples:

After opening the glass petri dish (54 mm) used for sampling the deposition (atmospheric fallout) of MPs particles. 10 ml of Milli-Q water (18 MΩ.cm) were carefully added and swirled around the dish, ensuring the lower sides of the dish are washed thoroughly, then placed on a slow mechanical shaker at 25 rpm for 30 min to release the particles. Then, the Milli-Q water was vacuum filtered through a glass fibre filter (47 mm, 1.0 μm pore size, Whatman, UK) using a Millipore® All-Glass Filter Holder Kit and a Welch™ 2522 WOB-L piston vacuum pump (Merck, Dorset, UK). The extraction cycle was repeated twice, with the 10 ml of Milli-Q water filtered on the same glass fibre filter.

The filter was transferred to a clean glass petri dish (54 mm, BRAND®, Merck™, Dorset, UK) containing 1 ml of H_2O_2 solution (30% w/v, Merck™, Dorset, UK), which was enough to submerge the filter and particles on it. The petri dish was then placed in a plastic-free oven at 60℃ for 48 hours to oxidise any organic particles on the filter (digestion step). The digested filters were then covered by their pre-cleaned and dried lids in a clean dry place until further microscopic examination.

Settled indoor dust samples:

Extraction of MPs from indoor dust samples was adapted from the method developed at our UoB laboratories and described by Nel et al., (2019) to reduce sample contamination from the extraction process via the application of Zinc Chloride salt for density separation using the in-house-built "Sediment Microplastic Isolation (SMI) units" (Figure SI5-1).

Prior to extraction, the collected dust samples were weighted on an accurate balance (LE324S, Sartorius, Germany), then sieved through a stainless-steel sieve (125 μm mesh size, 100 mm diameter, VWR International Ltd, UK), and the weight of the sieved dust was recorded as the size fraction relevant for human exposure via indoor dust ingestion (USEPA, 2011).

The sieved dust sample was added to the SMI unit then 50 mL of the prepared $ZnCl_2$ solution (700 g/L) were added. The SMI unit was then covered with a clean aluminium foil and placed on a magnetic stirrer and allowed to stir for 10 minutes at a rotation frequency of 5 to 8 revolutions per minute (rpm). Thereafter, the unit was left to settle for 12 hours. Once the denser particles had settled out, the supernatant was poured over a 25 μm stainless steel mesh into a beaker for filtration and re-use. The upper chamber of the SMI unit was also removed and rinsed 3 times (with 25 ml deionised water, each) over the mesh to ensure that all particles clinging to the surface of the equipment are collected. The MPs on the surface of the mesh were further washed 3 times (with 25 ml of deionised water, each) to remove any extraneous matter. The plastic samples were then transferred onto a clean glass microfiber filter (47 mm, 1.0 μm pore size, Whatman, UK) using a vacuum filtration system comprising a Millipore® All-Glass Filter Holder Kit and a Welch™ 2522 WOB-L piston vacuum pump (Merck, Dorset, UK). The filtration mesh was rinsed 7 times with 15 mL of deionized water from a squirt bottle over the same filter to ensure no MPs were lost during sample transfer.

5.1.2.3 The digestion process:

To remove unwanted particles such as organic matter (pollen, fungi, microbes, soot, etc.), a chemical agent for removal of MPs from this complex mixture often entrails chemical processing that includes digestion of organic matter, for instance, sodium hypochlorite (NaClO), sodium hydroxide (NaOH) or sodium hypochlorite (NaClO) (Uddin et al., 2021; Al-Salem et al., 2020).

Indoor air:

A digest was conducted using 200 ml of 30% hydrogen peroxide (H_2O_2) (Fisher Scientific) to significantly reduce the particle count and remove biological materials (non-MPs). After adding H_2O_2 to the samples, placed in a drying oven at 60 °C (Thermo Scientific, Fisherbrand® 65L, oven 230V, Germany) for 96 hours.

Settled indoor dust:

Using 200 ml of 30% hydrogen peroxide (H_2O_2) (Fisher Scientific) to significantly reduce the particle count, a chemical digestion process was conducted to remove organic (non-MPs) particles. The mesh was rinsed with 30% hydrogen peroxide (H_2O_2) into 250 mL glass beakers (pre-washed with Milli-Q water). The beakers were then covered with aluminum foil and placed in a drying oven at 60 °C (Thermo Scientific, Fisherbrand® 65L, oven 230V, Germany) for 96 hours. At the end of 96 hours, the contents of the beaker were vacuum filtered on a glass microfibre filter, and the beakers rinsed to ensure that all particles clinging to its surface were collected (Dris et al., 2015; Liu et al., 2019d). The last step is to transfer the remaining MPs particles on the filter to clean glass petri dishes (54 mm, BRAND®, Merck™, Dorset, UK), then place them in the oven again for 24 hours to be thoroughly dried and accurately weighted.

5.1.2.4 MPs identification and quantification:

To characterize MPs physically, the digital fluorescence microscope (OPTIKA™ IM-3LD4D PROVIEW, Version: x86,4.11.20351.20220226) with multiple lenses Plan Fluor (4X, 10X, 20X, 40X) was applied to identify the size, shape, and MPs particles counting.

5.1.3 Quality assurance and quality control:

Preventing background contamination by MPs particles in the laboratory is essential for microplastic research. To do this, researchers wore only pure cotton lab coats and nitrile gloves for the whole trial. Furthermore, a laminar flow bench was used for all sample processing, and it was routinely inspected and cleaned with 90% ethanol. All laboratory supplies and equipment were thoroughly cleaned with Milli-Q water before use to examine any contamination that may have occurred during the preparation and filtering of the sample for analysis.

Indoor air samples:

A “field blank” was analysed alongside each batch of 5 air samples. For active air sampling, the field blank comprises an unused 47 mm GFF unpacked in the field, then transferred to a clean glass Petri dish and treated like a sample. For passive air samples, the field blank consisted of an unopened Petri dish taken to the field, then transferred to the lab and treated like a sample.

A “recovery QA sample” was processed alongside each 20 samples. For active air sampling, the recovery sample comprised an unused 47 mm GFF spiked with a known quantity (20) of PE MPs particles (Cospheric™, California, USA), while for passive air sampling, the recovery sample was an unused petri-dish, spiked with a known quantity (20) of PE MPs particles (Cospheric™, California, USA).

Detailed results of blanks and recovery samples are provided in Table SI5-1. Overall, the numbers of MPs in all blanks never exceeded 5% of the average concentration of MPs in the corresponding batch. Therefore, there was no need for blank corrections. The recovery of MPs in the recovery samples ranged between 80% and 110%, indicating good performance of the analytical methodology.

Indoor dust samples:

In investigating potential contamination during the sample analysis (including preparation and filtration), dust samples were systematically analyzed in five batches. A blank sample comprising an unused thimble unpacked on the field (field blank) and treated as explained in the settled dust sampling section was analyzed alongside each batch of 5 dust samples. A "method blank" for the density separation method, comprising 15 mg of $ZnCl_2$ instead of indoor dust, treated as an MPs extraction and density separation section, was conducted alongside each 10 samples. Additionally, a "recovery sample" consisting of 50 mg of clean sediment (Sigma-Aldrich), intentionally spiked with a known quantity (20) of PE MPs particles (Cospheric™, California, USA), was processed alongside each of the 20 samples.

Detailed results of blanks and recovery samples are provided in Table SI5-1. Overall, the numbers of MPs in all blanks never exceeded 5% of the average concentration of MPs in the corresponding batch. Therefore, there was no need for blank corrections. The recovery of MPs in the recovery samples ranged between 80% and 110%, indicating good performance of the analytical methodology.

5.1.4 Statistical analysis:

Two programs were applied to analyze the data during this study: Excel version (Microsoft Office 365) and Statistical Package for the Social Sciences (SPSS) IBM version 29.0.1. Through Excel, the quantitative levels and distribution patterns were determined using

parameters such as average, standard deviation, median, minimum, and maximum. For SPSS, one-way ANOVA and Tukey HDS test were used to investigate differences among three or more dependent variables. The paired t-test was used to compare the means between the two datasets. Potential correlation between generated data was tested using Pearson correlations with a p-value of < 0.05 set as a level of statistical significance.

5.1.5 Results and discussion:

To investigate seasonal variation of MPs, we analysed 180 indoor samples, including (active air, passive air, and settled indoor dust samples) from five microenvironments (2 homes and 3 offices). Samples were collected monthly from January to December 2023, from each of the studied microenvironments and MPs were detected in all samples from all sites.

5.1.5.1 MPs concentrations:

Active air samples (airborne MPs):

Results showed that the mean concentration of airborne MPs in all studied sites ranged from 3 to 21 MPs/m^3. Mean concentrations of airborne MPs in all studied sites were in the order Summer > Winter > Spring > Autumn (Figure 5.1). One-way ANOVA revealed significant seasonal variability in airborne MPs concentrations with higher average concentrations in summer than autumn ($P = 0.015$), while the observed differences among other seasons were statistically insignificant. A closer look at seasonal variation in each of the microenvironments studied (Table 5.1) revealed MPs concentrations were not the highest in summer for Home-1, and that the observed higher MPs concentrations in summer was mainly driven by the sampled offices. When comparing the sampled sites' average concentrations, office-2 recorded the highest average concentration at 14.1 ± 5.3 MPs/m^3 in summer, while Home-1 had the lowest mean concentration of 5.0 ± 3.4 MPs/m^3 in autumn.

To date, there are no studies that investigated seasonal variation of MPs indoors, which makes it difficult to discuss our findings within a national or international perspective. To investigate a relevant context, a report from Japan studied the seasonal variation of volatile organic compounds (VOCs) in indoor air from 602 residences. The findings indicated that VOCs concentrations were significantly higher during the summer months compared to winter. This variation was mainly attributed to the influence of temperature on the emission rates of volatile pollutants from indoor materials, as well as increased ventilation rates during summer (Uchiyama et al., 2015). This was supported by the findings of recent study of indoor air quality in 5 low energy London apartments (Stamp et al., 2022). While this is less likely to influence seasonal variation of MPs which are non-volatile and have both indoor and outdoor sources, the Japanese study enlisted other contributing factors to the observed seasonal variation such as: differences in occupancy behavior *(e.g., personal activity impacting particulate matter resuspension)* (Uchiyama et al., 2015). Another study assessed indoor air quality from 12 working areas across three hospitals in Beirut, Lebanon investigated seasonal variation of particulate matter (PM_{10} and $PM_{2.5}$). The results indicated higher concentrations of indoor air pollutants during the warm months compared to the cold months, which was likely due to the high occupancy rates and human activities in the studied locations during the warm months (Chamseddine et al., 2018). In another study conducted in Alexandria, Egypt, average indoor concentrations of PM_{10} and $PM_{2.5}$ from 25 residential buildings decreased during summer, facilitated by increased ventilation and greater exchange with outdoor air (Abdel-Salam, 2021). Overall, the factors influencing seasonal variation of indoor air pollutants in the literature represent a combination of temperature, ventilation rates, indoor/outdoor air exchange, as well as occupancy rates and human activities which seem to be of particular importance for PM_{10} and $PM_{2.5}$ concentrations. MPs are non-volatile and have both indoor and outdoor sources so the impact of ventilation rates and indoor/outdoor exchange on their concentrations in indoor

air is unknown. Therefore, factors like occupancy rates and human activities are more likely to influence seasonal variation in MPs levels in indoor air, in addition to emission rates from indoor MPs sources (*e.g., carpets, clothing, as well as wear and tear of plastic items*).

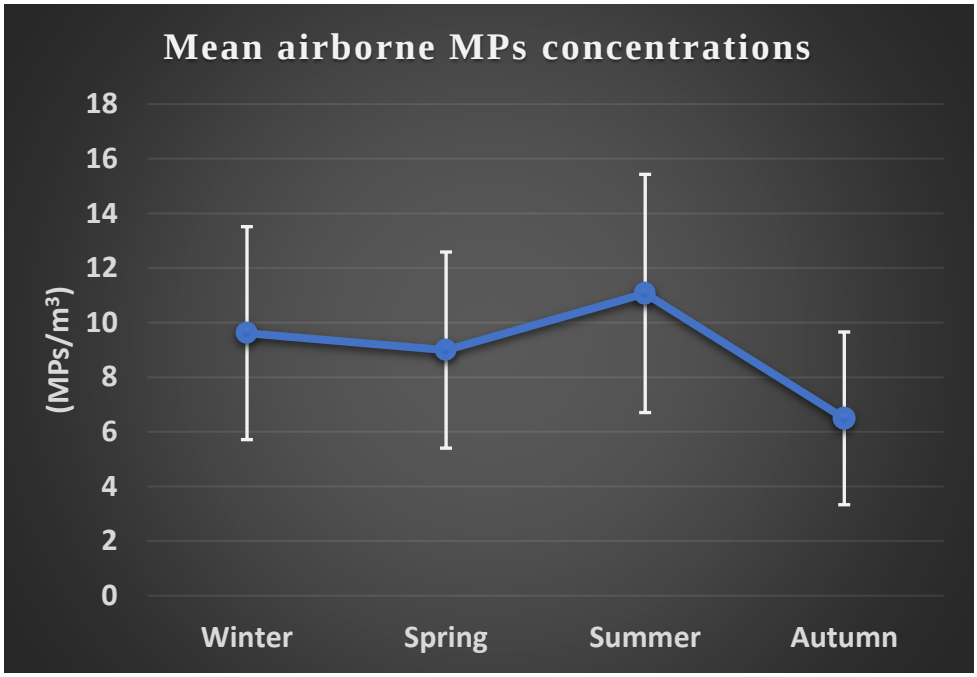


Figure 5.1 Mean concentration of airborne MPs in all studied sites.

Table 5.1: Mean concentration (± standard deviation) of airborne MPs in each of the studied indoor microenvironments.

Concentration of airborne MPs (MPs/m³)						
Season	**Home 1**	**Home 2**	**Office 1**	**Office 2**	**Office 3**	**All sites**
Winter	8.8 ± 6.3	11.0 ± 1.2	7.7 ± 3.6	9.6 ± 3.9	10.9 ± 5.1	9.6 ± 3.9
Spring	8.9 ± 2.6	9.5 ± 5.2	10.5 ± 6.1	9.5 ± 1.5	6.8 ± 2.1	9.0 ± 3.6
Summer	7.3 ± 1.5	11.2 ± 1.6	14.1 ± 5.3	10.4 ± 1	12.3 ± 7.8	11.1 ± 4.4
Autumn	5.0 ± 3.4	7.1 ± 3.5	7.6 ± 4.4	6.6 ± 3.5	6.2 ± 3	6.5 ± 3.2
All year	7.5 ± 3.7	9.7 ± 3.3	9.0 ± 2.8	10.0 ± 5.0	9.1 ± 5.1	9.0 ± 3.7

Passive air samples (atmospheric deposition MPs):

Concentrations of indoor atmospheric deposition MPs measured in all the studied microenvironments, ranged on average from 988 to 9947 MPs/m^2. One-way ANOVA showed significant (P = 0.026) higher levels of MPs in summer compared to spring, when the average concentrations of atmospheric deposition MPs in all the studied microenvironments were

considered. Mean concentrations of atmospheric deposition MPs in all studied sites were in the order Summer > Winter > Autumn > Spring (Figure 5.2).

Among the seasons at all sites, the highest average MPs concentration was reported in office-1 (6044 MPs/m^2 in Winter), while the lowest average was also measured in office-2 (2422 MPs/m^2 in Spring). Similar to airborne MP concentrations, the observed higher mean levels in summer, albeit statistically significant, were mainly driven by 3 of the studied microenvironments, while home-1 and office-2 had their highest mean concentrations in Autumn and Winter, respectively (Table 5.2). Notwithstanding the small number of microenvironments investigated in this pilot study despite the large number of samples generated (n = 180), the results collectively indicate that MPs concentrations in indoor air can show large variability depending on a multitude of factors not necessarily related to the change in temperature among seasons. These factors likely include potential MPs sources within each microenvironment, as well as ventilation, human activity and airflow dynamics reported to impact MPs abundance and distribution within indoor microenvironments (Kek et al., 2024b).

Similar to airborne MPs, no research has yet investigated seasonal variation of atmospheric deposition MPs indoors. However, a pertinent study employing air passive air sampling for persistent organic pollutants (POPs), specifically PCBs (polychlorinated biphenyls) and PBDEs (polybrominated diphenyl ethers), investigated their seasonal variation in indoor air from 92 UK indoor microenvironments. The results showed that PCB and PBDE concentrations in homes were highest during summer compared to other seasons, which may be attributed to higher indoor air temperatures and increased rate of room ventilation. However, the authors also noted that indoor air temperatures in the UK remain relatively stable throughout the year due to the use of heating systems during winter and air conditioning or enhanced natural ventilation during summer (Hazrati and Harrad, 2006). Another passive-sampling study examined the concentrations of PCBs and PBDEs across 20 residential sites in

the Czech Republic, in summer vs. winter. The findings indicated that the concentrations of all investigated POPs were higher in the summer compared to winter. This was primarily attributed to the influx of outdoor air on indoor environments, which is likely intensified during summer due to increased ventilation and the impact of seasonal temperature variation on indoor conditions (Melymuk et al., 2016). A study of seasonal variation in atmospheric deposition of VOCs across 50 homes in China indicated that all pollutants had significantly higher concentrations during the heating season compared to the non-heating season. This was attributed to low ventilation rates during the heating season (Duan et al., 2016).

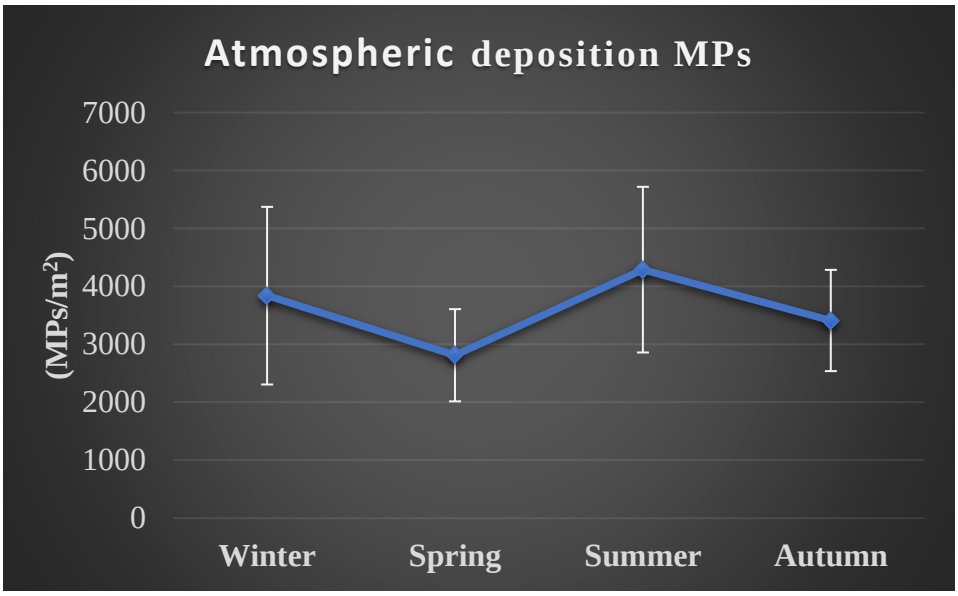


Figure 5.2 Mean concentration of atmospheric deposition MPs in all studied sites.

Table 5.2: Mean concentrations (± standard deviation) of atmospheric deposition MPs in each of the studied indoor microenvironments.

Concentration of atmospheric deposition MPs (MPs/m^2)						
Season	**Home 1**	**Home 2**	**Office 1**	**Office 2**	**Office 3**	**All sites**
Winter	3339 ± 1113	3915 ± 373	2669 ± 793	6044 ± 2477	3233 ± 1031	3840 ± 1532
Spring	3198 ± 371	2540 ± 907	2716 ± 1383	2422 ± 874	3186 ± 253	2812 ± 795
Summer	3516 ± 860	4292 ± 1183	3162 ± 1461	5891 ± 741	4585 ± 1642	4289 ± 1429
Autumn	3550 ± 824	3492 ± 337	3480 ± 240	3151 ± 1938	3386 ± 768	3412 ± 873
All year	3401 ± 792	3560 ± 700	3007 ± 970	4377 ± 1757	3598 ± 949	3588 ± 1034

Overall, several factors may contribute to the observed elevated concentrations of MPs in indoor air during the summer months. The practice of keeping windows and doors open for ventilation purposes facilitates the ingress of outdoor air, which may carry MPs into indoor environments (Eberhard et al., 2024). Additionally, weather conditions, including wind speed, can significantly influence the concentration of indoor air pollutants across different seasons. For instance, a study conducted in Athens, Greece, identified wind speed as a critical factor impacting the concentration levels of toluene and xylene within residential environments throughout various months (Alexopoulos et al., 2006). Also, elevated temperatures during summer can accelerate the degradation of larger plastic items into MPs (Perera et al., 2022c). Furthermore, higher humidity levels can promote the release of MPs from textiles and other materials through mechanisms such as wear and tear (Ageel et al., 2022). Human activities, which are more frequent in summer, such as cleaning, can disturb and resuspend MPs from surfaces. Air conditioning systems also may play a role in circulating MPs within indoor spaces (Boakes et al., 2023).

Settled indoor dust:

Concentrations of MPs in indoor dust samples from all the studied microenvironments showed large variation over the studied year, with average levels ranging from 10 to 1025 MPs/mg (Figure 5.3). Mean concentrations of settled indoor dust MPs in all studied sites were in the order Summer > Winter > Autumn > Spring (Figure 5.3). Although one-way ANOVA only revealed borderline significance between mean MPs dust concentrations in summer and spring ($P = 0.054$), this profile mirrors that observed for atmospheric deposition MPs, which is consistent with the nature of atmospheric deposition particles as proxy for settled dust particles (Heindel et al., 2020).

Among the studied seasons, the highest MPs concentrations (437 ± 319) were recorded in office-2 in summer, while the lowest concentrations (42.7 ± 24.8) were measured in home-1 during spring (Table 3).

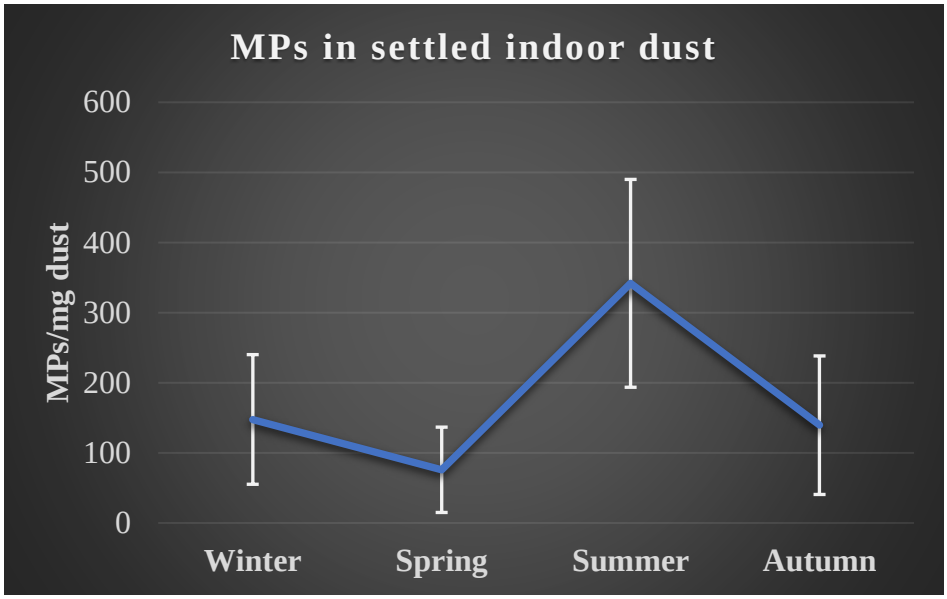


Figure 5.3 Mean concentration of dust MPs in all studied sites.

Table 5.3: Mean concentrations (± standard deviation) of MPs in settled indoor dust from each of the studied indoor microenvironments.

MPs concentration in settled dust (MPs/mg)						
Season	**Home 1**	**Home 2**	**Office 1**	**Office 2**	**Office 3**	**All sites**
Winter	43.2 ± 23.8	116 ± 76.9	206 ± 133	124 ± 84.8	82.2 ± 63.6	148 ± 92.5
Spring	42.7 ± 24.8	135 ± 120	102 ± 22.6	56.4 ± 56.8	79.5 ± 43.4	75.8 ± 60.9
Summer	265 ± 252	69.7 ± 53.5	138 ± 34.8	437 ± 319	101 ± 80.7	342 ± 148
Autumn	311 ± 187	352 ± 57.9	233 ± 75.3	452 ± 80.1	60.9 ± 24.7	139 ± 98.8
All year	166 ± 123	158 ± 77.2	170 ± 66.4	267 ± 135	81.1 ± 53.3	168 ± 90.8

Similar to indoor air, no studies have investigated seasonal variation of MPs in settled indoor dust. One of the logical reasons that may lead to higher MPs concentrations in the summer season is that increased airflow can resuspend settled dust, containing MP particles from surfaces; thereby redistributing MPs augmenting their concentration in indoor environments (Ageel et al., 2022). A pertinent study that investigated seasonal variation of phthalates (used as plasticizers) concentrations in settled indoor dust from 54 homes in Austin,

Texas reported frequently higher levels in summer than in winter (Bi et al., 2018). The increased concentration of MPs in indoor dust during summer was explained by the higher amount of dust particles that enter the indoor sites during the warm months through opened windows. This may also be relevant for MPs with increased influx from outdoor particles into indoor microenvironments during summer (Kim et al., 2021).

5.1.5.2 MPs characteristics:

5.1.5.2.1 MPs shape:

Three different shapes of MPs have been detected in the studied samples, namely: fibers, fragments, and foam. Fibers were the most abundant shape, with a percentage above 40% in all indoor sites during all seasons. Fragments came second contributing 35% - 61% to the detected MPs, while foams were the least abundant at <15% contribution in all the analysed samples (Tables 5.4 – 5.6).

Airborne MPs:

The average distribution of airborne MPs shapes in the studied microenvironments during all seasons is summarized in Table 5.4. Large variations were observed in MPs shape distribution between summer and autumn. One-way ANOVA revealed statistically significant differences ($P < 0.05$) in the percent contribution of fibers and fragments to airborne MPs between summer and autumn, while no significant variations were observed among the other seasons, nor for the percent contribution of foam, which was minimal (< 15%) in all seasons at all sites.

Table 5.4: Mean shape distribution of airborne MPs in the studied microenvironments.

Sites	Shapes	Winter (%)	Spring (%)	Summer (%)	Autumn (%)	All year (%)
Home 1	fiber	45 ± 24.6	40 ± 22.5	43 ± 21.4	44 ± 13.6	43 ± 21
	fragment	47 ± 20	46 ± 17.5	42 ± 20.8	41 ± 13	44 ± 18
	foam	8 ± 4.6	14 ± 5	15 ± 3.5	15 ± 5	13 ± 5
Home 2	fiber	52 ± 5.9	42 ± 3	53 ± 16.8	39 ± 24.8	46 ± 13
	fragment	39 ± 2	44 ± 7.6	28 ± 5.4	52 ± 26.1	41 ± 10
	foam	9 ± 4.5	14 ± 4.9	20 ± 12.2	9 ± 4.5	13 ± 7
Office 1	fiber	57 ± 5.3	52 ± 17.4	55 ± 8.1	34 ± 20	49 ± 13
	fragment	32 ± 4.4	31 ±12.3	30 ± 12	51 ± 18.8	36 ± 12
	foam	12 ± 2.6	17 ± 9.4	16 ± 6.8	17 ± 1.7	15 ± 5
Office 2	fiber	40 ± 13.1	36 ± 21.3	48 ± 12.8	35 ± 11.4	40 ± 15
	fragment	40 ± 5.9	50 ± 17.3	39 ± 11.2	55 ± 11.9	46 ± 12
	foam	20 ± 11.7	14 ± 4.3	13 ± 1.8	10 ± 1.5	14 ± 5
Office 3	fiber	51 ± 19	40 ± 13.5	55 ± 6	26 ± 7.5	43.5 ± 11
	fragment	35 ± 18.9	43 ± 8.6	33 ± 5	61 ± 10.8	43.5 ± 11
	foam	14 ± 1	16 ± 5.1	12 ± 4.5	12 ± 1.5	13 ± 3

Atmospheric deposition MPs:

The average distribution of atmospheric deposition MPs shapes in the studied microenvironments during all seasons is summarized in Table 5.5. Unlike airborne MPs, One-way ANOVA revealed the observed variation in MPs shape distribution among seasons was not statistically significant ($P > 0.05$).

Table 5.5 Mean shape distribution of atmospheric deposition MPs in the studied microenvironments.

Sites	Shapes	**Winter** (%)	**Spring** (%)	**Summer** (%)	**Autumn** (%)	**All year (%)**
Home 1	**fiber**	36.4 ± 12.6	38.3 ± 14.4	57 ± 10.3	50 ± 11.8	45.5 ± 12
	fragment	50.8 ± 10.7	48.2 ± 9.8	32 ± 7.1	42 ± 12.9	43.1 ± 10
	foam	12.8 ± 1.9	13.5 ± 7.1	11 ± 4.2	8 ± 2.1	11.4 ± 4
Home 2	**fiber**	56.8 ± 9.6	38.7 ± 20	70 ± 17.2	51 ± 33.8	54.2 ± 20
	fragment	26.9 ± 9.6	42.1 ± 8.8	20 ± 12.2	35 ± 29.2	31 ± 15
	foam	16.3 ± 2.1	19.2 ± 13.9	10 ±5.5	14 ± 6.7	14.8 ± 7
Office 1	**fiber**	53.5 ± 9.7	49.8 ± 7.8	50 ± 24.8	55 ± 9.4	52 ± 13
	fragment	27.9 ± 5.1	34.6 ± 10.7	35 ± 13.3	37 ± 7.7	33.6 ± 9
	foam	18.6 ± 9.1	15.6 ± 3.1	15 ± 11.5	8 ± 1.7	14.4 ± 6
Office 2	**fiber**	52.8 ± 2.5	51.3 ± 10.8	52 ± 9.6	68 ± 5.3	56 ± 7
	fragment	34.7 ± 9.6	33 ± 12.5	37 ± 9.7	23 ± 6.4	32 ± 10
	foam	12.4 ± 7.5	15.7 ± 2.1	11 ± 0.6	9 ± 2.3	12 ± 3
Office 3	**fiber**	56.4 ± 8.1	49 ± 23.1	61 ± 10	54 ± 7.9	55.2 ± 12
	fragment	28.7 ± 7.5	38 ± 25.9	26 ± 1.5	37 ± 8.7	32.3 ± 11
	foam	14.9 ± 3.1	13 ± 4.4	13 ± 9.2	9 ± 1.7	12.5 ± 5

MPs in settled indoor dust:

The average distribution of indoor dust MPs shapes in the studied microenvironments during all seasons is summarized in Table 5.6. One-way ANOVA revealed significant seasonal variation ($P < 0.05$) in MPs shape distribution between summer and autumn for fibers and fragments, while the variations in foam percent contribution were not significant statistically.

Overall, our results reveal that the shape distribution of MPs may display substantial variation among different seasons within the same indoor microenvironment in both air and dust. This may indicate variation of input sources for MPs to the indoor microenvironment in different seasons and/or the potential impact of human activity on the release/emission of secondary MPs *(e.g., cleaning frequency, contribution to wear and tear of plastic items by frequency of use or movement)*. It should be noted that while few studies in lower invertebrates and benthic organisms have reported different toxicological impacts for different MPs shapes, this has not been proven hitherto in humans or other apex predators (Bostan et al., 2023).

Table 5.6: Mean shape distribution of dust MPs in the studied microenvironments.

Sites	Shapes	Winter (%)	Spring (%)	Summer (%)	Autumn (%)	All year (%)
Home 1	**fiber**	41.8 ± 8.5	57.3 ± 11	57 ± 12.9	50 ± 21.2	51.4 ± 13
	fragment	52.8 ± 9.5	37.7 ± 10.3	40 ± 14.7	45 ± 22.4	44 ± 14
	foam	4.7 ± 2.1	5 ± 1	3 ± 2	6 ± 1.6	4.6 ± 2
Home 2	**fiber**	46.8 ± 30	51.4 ± 10	71 ± 4.5	57 ± 15.6	56.5 ± 15
	fragment	25.1 ± 11.3	42.8 ± 10.1	23 ± 1.5	39 ± 17.5	32.3 ± 10
	foam	4 ± 1.7	5.8 ± 2.1	6 ± 3.6	5 ± 2	5.2 ± 2
Office 1	**fiber**	47.7 ± 13.8	56.3 ± 11.5	66 ± 14	48 ± 12.2	54 ± 13
	fragment	47 ± 10.1	38.7 ± 14.2	29 ± 11.8	48 ± 12.5	41 ± 12
	foam	5.3 ± 4	5 ± 2.6	5 ± 3.6	4 ± 1.5	5 ± 3
Office 2	**fiber**	44 ± 11.3	52.8 ± 9.3	61 ± 11.2	49 ± 21	52 ± 13
	fragment	44.7 ± 1.6	41 ± 7.8	34 ± 10.6	47 ± 22.4	42 ± 11
	foam	11.3 ± 11	6.2 ± 1.8	4 ± 2.3	4 ± 2	6 ± 4
Office 3	**fiber**	49.8 ± 3.1	45.5 ± 10.3	70 ± 6.7	49 ± 9.9	53 ± 8
	fragment	44.1 ± 3.4	50.4 ± 10	27 ± 5.6	46 ± 13.1	42 ± 8
	foam	6.2 ± 3.3	4.1 ± 2.2	3 ± 3.1	5 ± 5.2	5 ± 3

5.1.5.2.2 MPs Size:

The largest MPs sizes among detected shapes were fibers with lengths ranging from 12.4 to 82.9 µm, with a mean of 30.1. The smallest size were fragments with longest dimension ranging from 3 to 4.3 µm with a mean of 3.7, while foams ranged in size from 4.4 to 6 µm with a mean of 5.1 µm (Figure 5.4).

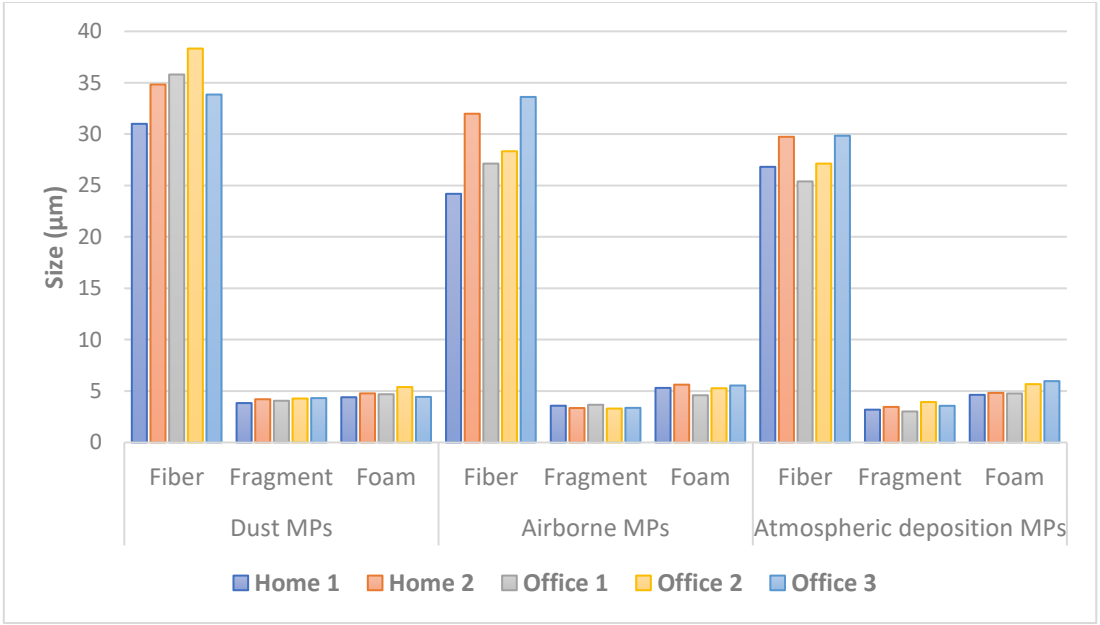


Figure 5.4 Mean size (µm) of different MPs shapes detected at the studied microenvironments over 12 months.

Airborne MPs:

The mean size distribution of airborne MPs in all studied microenvironments is provided in Figure 5.5. One-way ANOVA revealed significant seasonal variation ($P < 0.05$) in average airborne MPs sizes at all detected shapes. For fibers, the difference in average sizes were statistically significant between winter and spring ($P = 0.001$), summer and spring ($P = 0.002$), and summer and autumn ($P = 0.019$). In fragments, the identified differences were significant between winter and autumn ($P = 0.001$), spring and autumn ($P = 0.001$), spring and summer ($p = 0.004$), and summer and autmn ($P = 0.001$). Finally, for Foam MPs, significant differences in mean size were indicated between spring and all other seasons ($P = 0.001$), and between summer and autumn ($P = 0.008$).

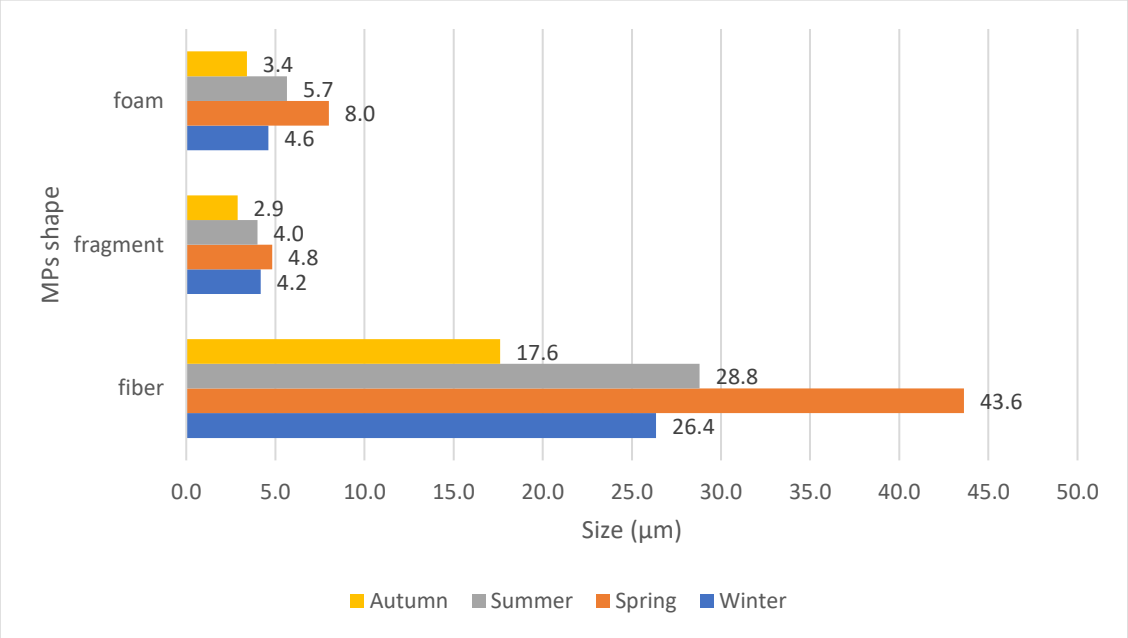


Figure 5.5 Mean size of airborne MPs from all microenvironments in different seasons.

Atmospheric deposition MPs:

The mean size distribution of atmospheric deposition MPs in all studied microenvironments is provided in Figure 5.6. One-way ANOVA revealed significant seasonal variation in MPs sizes of all detected shapes in atmospheric deposition samples. In fibers,

significant differences were observed between winter and autumn (P = 0.001), summer and autumn (P = 0.016), and spring and all other seasons (P = 0.001). For fragments, significant differences were identified between summer and autumn (P = 0.001), spring and winter (P = 0.031), spring and summer (p = 0.003), and spring and autmn (P = 0.001). In foam MPs, average sizes were significantly different between winter and spring (P = 0.011), winter and summer (P = 0.033), winter and autumn (P = 0.004), spring and summer (P = 0.001), and spring and autumn (P = 0.001).

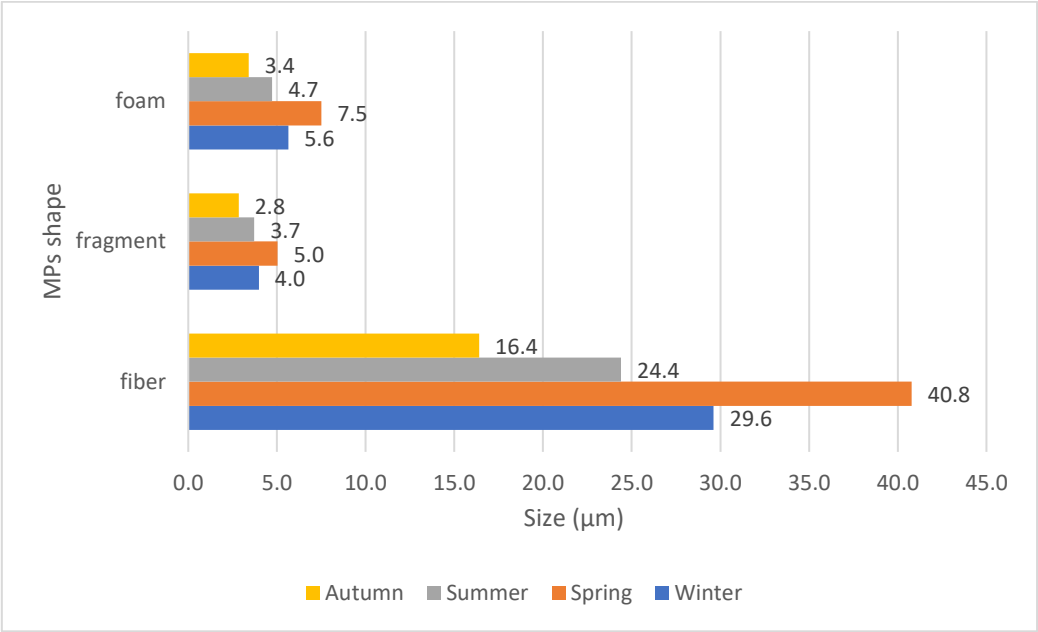


Figure 5.6 Mean size of atmospheric deposition MPs from all microenvironments in different seasons.

MPs in settled indoor dust:

The mean size distribution of indoor dust MPs in all studied microenvironments is provided in Figure 5.7. One-way ANOVA revealed significant seasonal variation in mean MPs sizes of all detected shapes in settled indoor dust samples. In fibers, significant differences were found between winter and spring (P = 0.001), winter and autumn (P = 0.001), spring and summer (P = 0.001), spring and autumn (P = 0.001), and summer and autumn (P = 0.031). For fragments, the identified significant differences were between winter and spring (P = 0.006),

winter and summer (P = 0.001), winter and autumn (P = 0.001), spring and summer (P = 0.001), and spring and autumn (P = 0.001). finally, in foams, the mean sizes were significantly different between winter and spring (P = 0.001), winter and autumn (P = 0.040), spring and summer (P = 0.001), spring and summer (P = 0.001), and spring and autumn (P = 0.001).

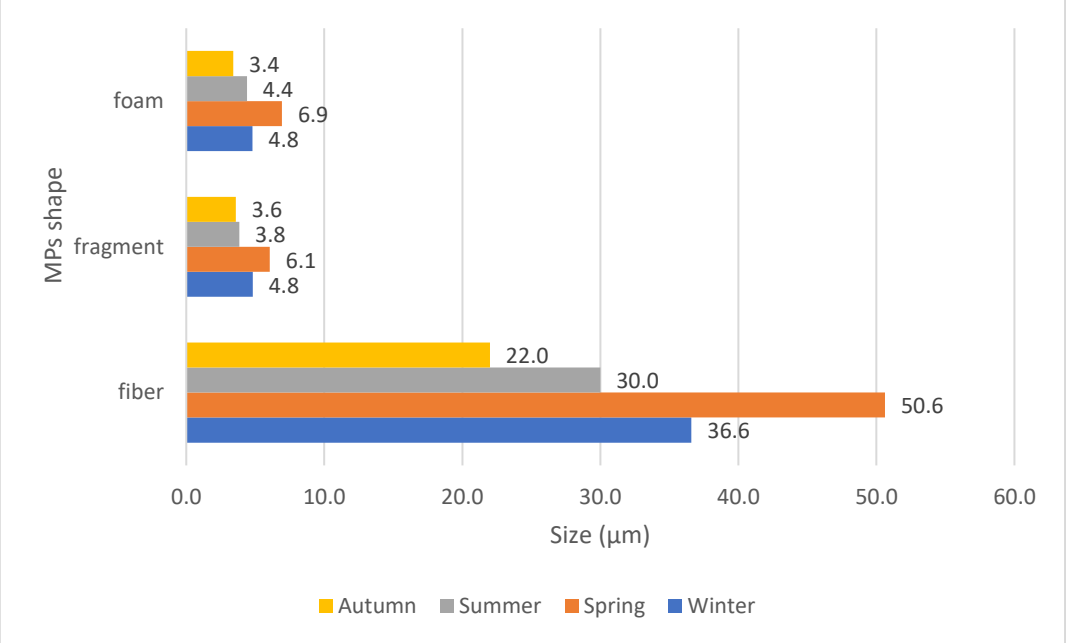


Figure 5.7 Mean size of dust MPs from all microenvironments in different seasons.

It is worth noting that average sizes for all MPs shapes in spring were higher than other seasons (figures 5.5-5.7). The reason behind this is not clear and further research is required to identify the factors influencing MPs size in the indoor environment.

Overall, MPs sizes can contribute to their adverse toxic effects. While the adverse effects of MPs on human health are not fully understood to date, smaller MPs (< 50 µm) are more likely to be absorbed, distributed and accumulated in different human vital organs (Ageel et al., 2022; Kek et al., 2024b). Therefore, the observed significant seasonal variation in sizes of indoor MPs may also influence the risk arising from exposure to these MPs via inhalation and ingestion.

5.1.5.3 Human exposure to MPs indoors:

Concerns over human exposure to MPs in the indoor environment and the possible health risks have recently grown (Wright et al., 2020). It is currently understood that the primary routes of human exposure to MPs in indoor environments are inhalation and ingestion, while dermal contact may represent a less significant pathway (Zhang et al., 2020a; Sharma et al., 2021; Prata, 2018). In theory, only particles with a diameter of less than 10 μm are deemed inhalable, yet MP fibers up to 29 μm in width and 897 μm in length have been shown to enter human lung and penetrate deep into the alveoli tissue (Jenner et al., 2021b). It was recently revealed that the settling depth of MPs in the respiratory system is impacted by their aerodynamic diameter, which measures the settling speed of particles with different densities and shapes (Noorimotlagh et al., 2024). Similarly, inadvertent ingestion of indoor dust via hand-to-mouth behaviour has been established as a major pathway of exposure to several persistent organic pollutants and emerging contaminants, particularly in toddlers and young children (Zhu et al., 2023). Combined with the frequent detection of MPs in indoor dust, this has recently raised concern over potential exposure via inadvertent dust ingestion, particularly in vulnerable age groups (Kek et al., 2024b).

In the current study, we used the mean concentrations of MPs in indoor air (airborne MPs) and settled indoor dust, measured in the studied microenvironments to estimate the daily intake of adults and toddlers to MPs via inhalation and ingestion of indoor dust. The MPs concentrations were combined with typical time-activity patters for the UK population, as well as inhalation and dust ingestion rates and typical “average” body weights from the US EPA exposure factors handbook to provide reliable estimates of the average exposure scenario. The focus in this paper will be on the impact of seasonal variation on estimated daily intakes (EDI)

of MPs, rather than estimating intakes from different exposure scenarios, which has been covered in our previous work (chapters 3 and 4 of this thesis).

5.1.5.3.1 Human exposure to MPs via inhalation of indoor air:

Mean EDI of inhaled MPs from indoor air in the studied microenvironments ranged from 0.3 MPs/kg bw/day in autumn to 1.7 MPs/kg bw/day in summer for adults, and from 2.1 MPs/kg bw/day in autumn to 4.8 MPs/kg bw/day in summer for toddlers (Table 5.7).

The lowest seasonal exposure for both adults and toddlers to MPs via inhalation in all the studied sites occurred in autumn. The highest exposure for adults was in summer, while for toddlers was in winter (Table 5.7). This discrepancy can be explained by the fact that toddlers are not exposed at offices; therefore their exposure in the present study was estimated based on the two homes sampled. Adults are exposed at both homes and offices; hence their exposure in the current study was based on the sampled 2 homes and 3 offices.

One-way ANOVA revealed the observed differences between the mean EDIs for MPs via inhalation in the highest and lowest season for both adults and toddlers to be statistically significant ($P < 0.01$).

Moreover, the mean EDIs over 12 months for both adults and toddlers in the 5 studied microenvironments showed substantial spatial variability, albeit not statistically significant (Figure 5.8). This can be explained by the different sources of MPs in different indoor microenvironments (*e.g., carpets, fabrics, plastic items*), combined with different human activities (*e.g., cleaning, ventilation, movement*), which can lead to substantial variation in MPs concentrations among different indoor microenvironments (Ageel et al., 2022; Kek et al., 2024b).

Table 5.7: Mean EDI (MPs/kg bw/day) to MPs via inhalation of indoor air from studied microenvironments in different seasons.

Season	Home 1		Home 2		Office 1	Office 2	Office 3	All sites	
	Toddlers	Adults	Toddlers	Adults	Adults	Adults	Adults	Toddlers	Adults
Winter	3.7 ± 2.6	1.3 ± 0.9	4.6 ± 0.5	1.7 ± 0.2	0.5 ± 0.2	0.4 ± 0.2	0.6 ± 0.3	4.1 ± 1.1	0.9 ± 0.1
Spring	3.6 ± 1.1	1.3 ± 0.4	4.0 ± 2.2	1.4 ± 0.8	0.5 ± 0.1	0.6 ± 0.3	0.4 ± 0.1	3.8 ± 1.0	0.8 ± 0.2
Summer	3.2 ± 0.6	1.1 ± 0.2	4.8 ± 07	1.7 ± 0.2	0.7 ± 0.1	0.9 ± 0.3	0.8 ± 0.4	4.0 ± 0.6	1.2 ± 0.2
Autumn	2.1 ± 1.4	0.8 ± 0.5	3.0 ± 1.5	1.1 ± 0.5	0.3 ± 0.2	0.4 ± 0.2	0.3 ± 0.2	2.5 ± 1.0	0.6 ± 0.1
All year	3.1 ± 1.5	1.1 ± 0.6	4.0 ± 1.4	1.5 ± 0.5	0.5 ± 0.2	0.6 ± 0.3	0.5 ± 0.3	3.6 ± 1.0	0.8 ± 0.2

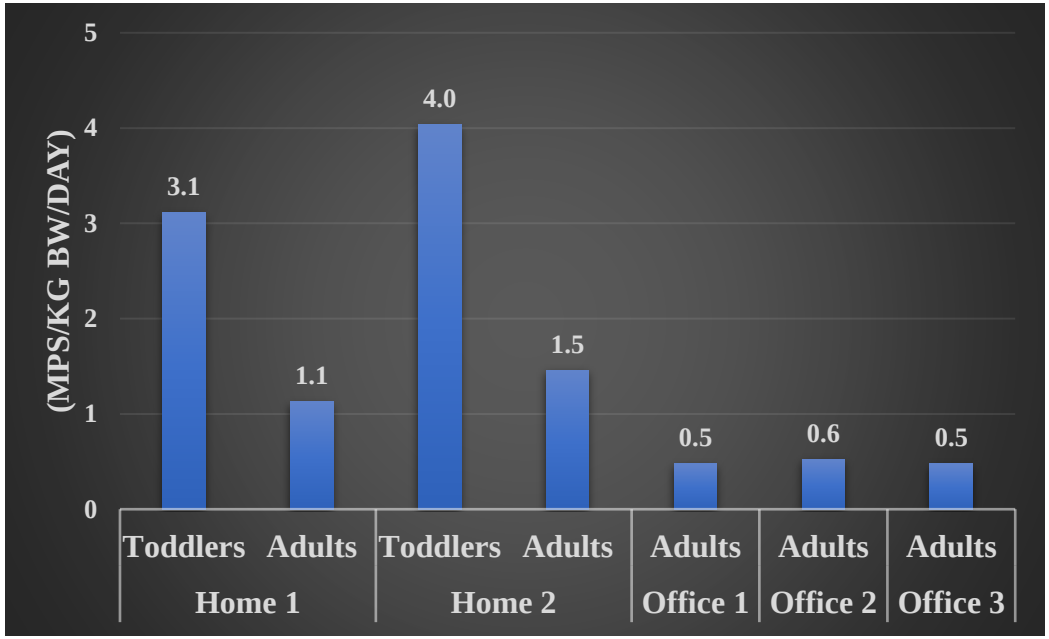


Figure 5.8 Mean EDI (MPs/Kg BW/day) to MPs via inhalation of indoor air from studied microenvironments over 12 months.

5.1.5.3.2 Human exposure to MPs via ingestion of indoor dust:

Similar to inhalational exposure, the mean EDI for MPs via inadvertent indoor dust ingestion showed large variation between 0.1 (spring) – 1.6 (summer) MPs/kg bw/day for adults, and 3.5 (spring) – 28.7 (summer) MPs/kg bw/day for toddlers (Table 5.8). The large difference between EDIs for adults and toddlers can be explained by lower hygiene measures in toddlers leading to increased dust ingestion via hand-to-mouth, combined with their lower body weight.

Summer was the season with the highest EDIs, while spring showed the lowest EDIs for both adults and toddlers. Unlike inhalation exposure, the results of one-way ANOVA did not show the observed seasonal variability in EDIs to be statistically significant at the 95% level ($P = 0.062$ which is significant at the 90% level). Large spatial variability was also observed among the estimated EDIs in the 5 microenvironments studied over 12 months, albeit not significant statistically (Figure 5.9).

Overall, the observed variations can be attributed to a multitude of factors, which can lead to significant differences in concentrations of MPs in different indoor microenvironments as discussed under section 5.1.5.1. However, the observed significant seasonal variation in EDIs is of particular importance to exposure and risk assessment studies. This indicates that studies based on “spot sampling” of indoor microenvironments at a single point in time may not accurately reflect personal exposure over a prolonged period. Moreover, the results may not be even comparable among studies when “spot sampling” was conducted in different seasons, or even within the same study if the sampling campaign was conducted over a prolonged period, whereby some microenvironments were sampled in summer and others in autumn.

Table 5.8: Mean EDI (MPs/kg Bw/day) to MPs via ingestion of indoor dust from studied microenvironments in different seasons.

Season	Home 1		Home 2		Office 1	Office 2	Office 3	All sites	
	Toddlers	Adults	Toddlers	Adults	Adults	Adults	Adults	Toddlers	Adults
Winter	25.3 ± 33.9	1.4 ± 1.9	4.0 ± 1.4	0.2 ± 0.1	0.3 ± 0.2	0.1 ± 0.1	0.1 ± 0.1	14.7 ± 17.3	0.4 ± 0.4
Spring	3.5 ± 2.0	0.2 ± 0.1	10.6 ± 9.4	0.6 ± 0.5	0.1 ± 0.02	0.1 ± 0.1	0.1 ± 0.04	7.0 ± 4.2	0.2 ± 0.1
Summer	21.6 ± 20.5	1.2 ± 1.1	28.7 ± 41.9	1.6 ± 2.3	0.2 ± 0.02	1.3 ± 0.8	0.2 ± 0.1	25.2 ± 16.8	0.9 ± 0.2
Autumn	3.5 ± 1.9	0.2 ± 0.1	3.6 ± 3.6	0.2 ± 0.2	0.3 ± 0.3	0.6 ± 1.0	0.1 ± 0.1	3.6 ± 0.9	0.3 ± 0.2
All year	13.5 ± 19.9	0.7 ± 1.1	11.8 ± 21.3	0.6 ± 1.2	0.2 ± 0.2	0.5 ± 0.8	0.1 ± 0.1	12.6 ± 13.6	0.5 ± 0.4

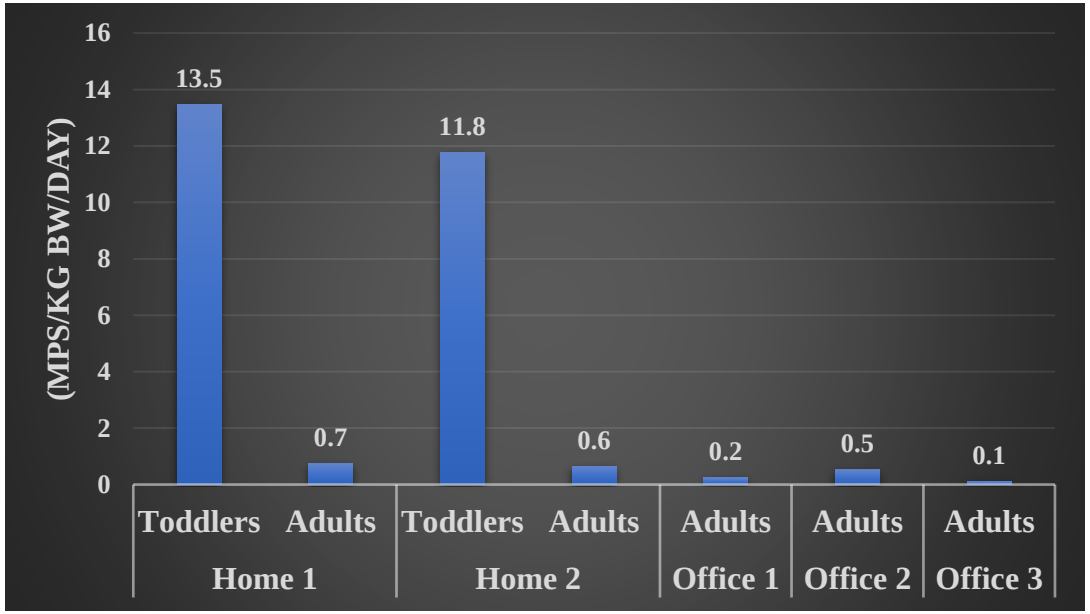


Figure 5.9 Mean EDI (MPs/Kg BW/day) to MPs via ingestion of indoor dust from studied microenvironments over 12 months.

5.1.6 Conclusion:

This is the first study to investigate seasonal variation in concentrations of MPs in the indoor environment and its implications on estimated daily intakes via inhalation and dust ingestion. The results indicated that all sampling locations experienced the highest concentrations of MPs during the summer months. This seasonal peak may be attributed to a combination of factors including increased indoor dust levels resulting from heightened outdoor activities, open windows, and greater activity *(e.g., foot traffic)*, which collectively introduce more particles indoors. Notably, One-way ANOVA revealed significant differences ($P < 0.05$) in MPs size and shape distribution as well as concentrations in indoor air and dust over the monitored seasons in the studied microenvironments. This was reflected in significant seasonal variability in estimated daily intakes of adults' and toddlers' exposure to MPs via inhalation and inadvertent dust ingestion indoors. This variability raises concern over the representativity and comparability of existing human exposure data from different studies based on single "spot" samples collected from various indoor microenvironments in different seasons.

5.1.7 Supplementary Information:

Quality assurance/quality control:

Table SI5-1a: Number of MPs in the analysed blanks.

Blank type/no.	Active air sample	Passive air sample	Settled indoor dust
Field 1	2	3	2
Field 2	1	2	5
Field 3	3	1	1
Field 4	1	2	3
Field 5	0	1	2
Field 6	0	3	3
Field 7	3	1	0
Field 8	1	3	0
Field 9	0	0	6
Field 10	0	0	2
Field 11	1	0	1
Field 12	1	0	2
Field 13	1	1	1
Field 14	0	1	0
Field 15	0	1	5
Field 16	2	3	2
Field 17	3	1	5
Field 18	1	2	1
Field 19	1	0	3

Field 20	0	3	2
Field 21	1	0	3
Field 22	1	3	0
Field 23	2	2	0
Field 24	3	2	6

Table SI5-1b: Recovery (expressed as %) of MPs in the QA/QC recovery samples.

Sample no	Active air sample	Passive air sample	Settled indoor dust
Recovery 1	95	80	85
Recovery 2	95	8	80
Recovery 3	80	105	110
Recovery 4	90	95	85
Recovery 5	105	80	110
Recovery 6	90	85	85
Average	93	93	88
SD	14	8	10
Median	85	93	83
Min	80	80	80
Max	110	105	105

Human Exposure Assessment to MPs of SVS:

Concentrations of MPs were determined in indoor air and dust during the seasonal variation period. Active air sampling was used to estimate exposures via inhalation of airborne MPs, and indoor dust was used to estimate exposures via ingestion of settled MPs for UK adults and toddlers.

Human exposure via inhalation of indoor air:

Daily exposure to MPs via inhalation was estimated for UK adults and toddlers by applying MPs air concentrations measured in this study in the following exposure algorithm:

$$\sum_{iex} = \frac{[(C_H \times F_H) + (C_W \times F_W) + (C_T \times F_T) + (C_P \times F_P) + (C_O \times F_O)]}{BW} \times R_i$$

Where $\sum_{iex}$ is the daily human inhalation exposure (MPs Kg BW^{-1} day^{-1}); $C_{H/W/T/P/O}$ is the concentration of MPs (MPs/m^3) in home/ workplace/methods of transport/public microenvironments/outdoors respectively; $F_{H/W/T/P/O}$ is the fraction for time spent in each microenvironment type; Ri is the daily inhalation rate (m^3/day); and BW is the body weight (Kg).

Human exposure via ingestion of indoor dust:

The estimated daily intake exposure (EDI; MPs/kg/bw/day) of MPs ingested through indoor dust was estimated for UK adults and toddlers by applying the following equation:

$$EDI_{ing} = \frac{C \times f \times m_{di}}{BW}$$

Where C is the determined MPs concentration in settled indoor dust samples (MPs/g); f is the time fraction spent in the sampled indoor microenvironment (e.g., hours spent); m_{di} (g/day) is the indoor dust ingestion rate for humans, (US EPA, 2011); and BW is the body weight (kg).

The time-activity patterns of the UK and Ireland populations were gathered from previously published research. Typical activity patterns were reported as follows: for adults, the patterns were 63.8% at home, 22.3% at work, 5.1% PMEs, 4.1% cars, and 4.7% outdoors; for toddlers, the patterns were 86.1% at home, 5.1% PMEs, 4.1% cars, and 4.7% outdoors (Tao et al., 2016; Wemken et al., 2019b).

The exposure parameters applied were obtained from the US EPA Exposure Factors Handbook (US EPA, 2011), and are summarised in Table SI-2 below:

Table SI5-2: Ingestion and inhalation exposure parameters.

Age groups	**Adult**	**Toddler**
BW (Kg)	71.5	10.6
Ingestion rate (g/day)	0.05	0.10
Inhalation rate (m^3/day)	16.88	5.12

Figure SI5-1: Mean airborne MPs shape distribution in the studied microenvironments over 12 months

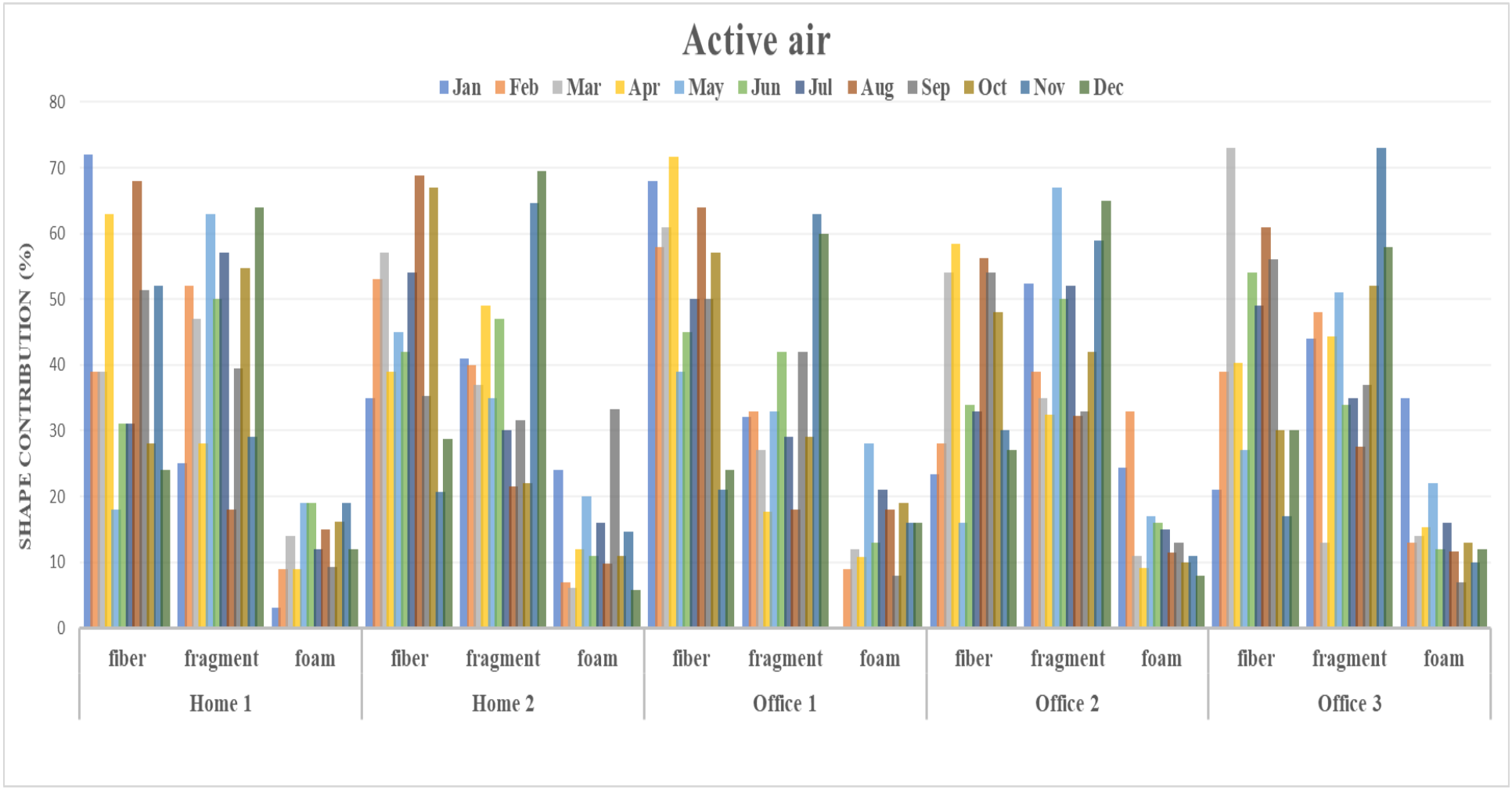


Figure SI5-2: Mean atmospheric deposition MPs shape distribution in the studied microenvironments over 12 months

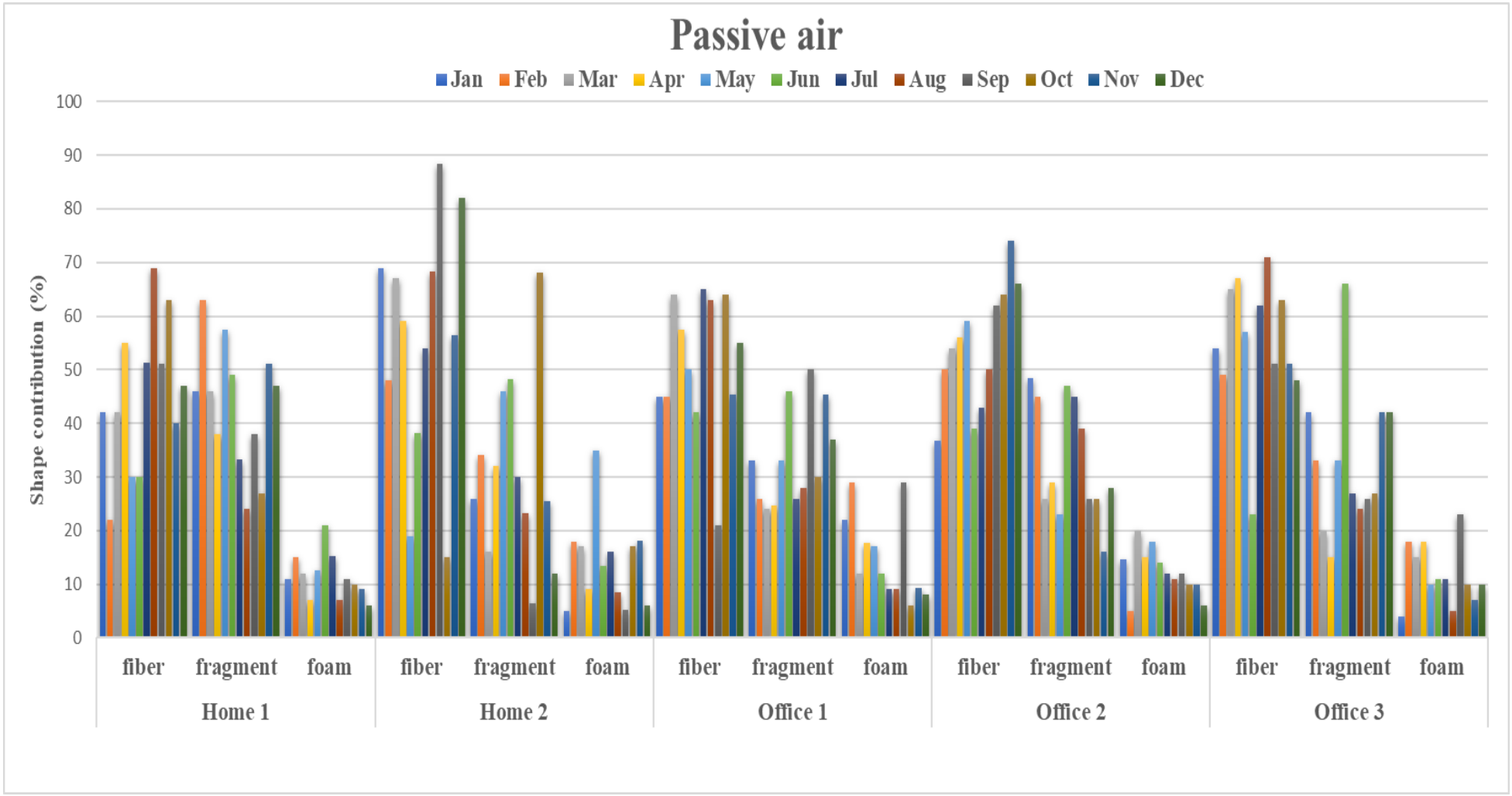


Figure SI5-3: Mean dust MPs shape distribution in the studied microenvironments over 12 months

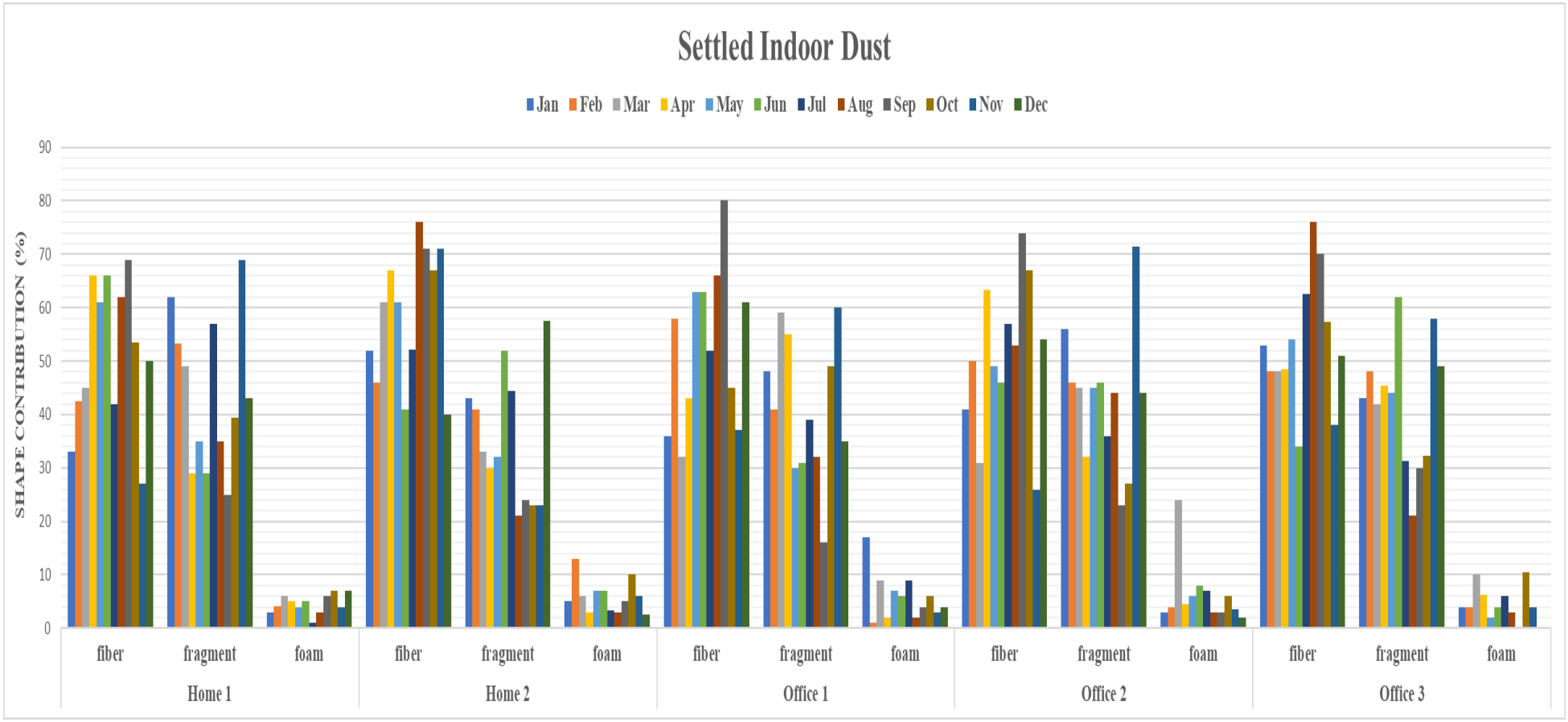


Figure SI5-4: Mean size of detected shapes in airborne samples over 12 months

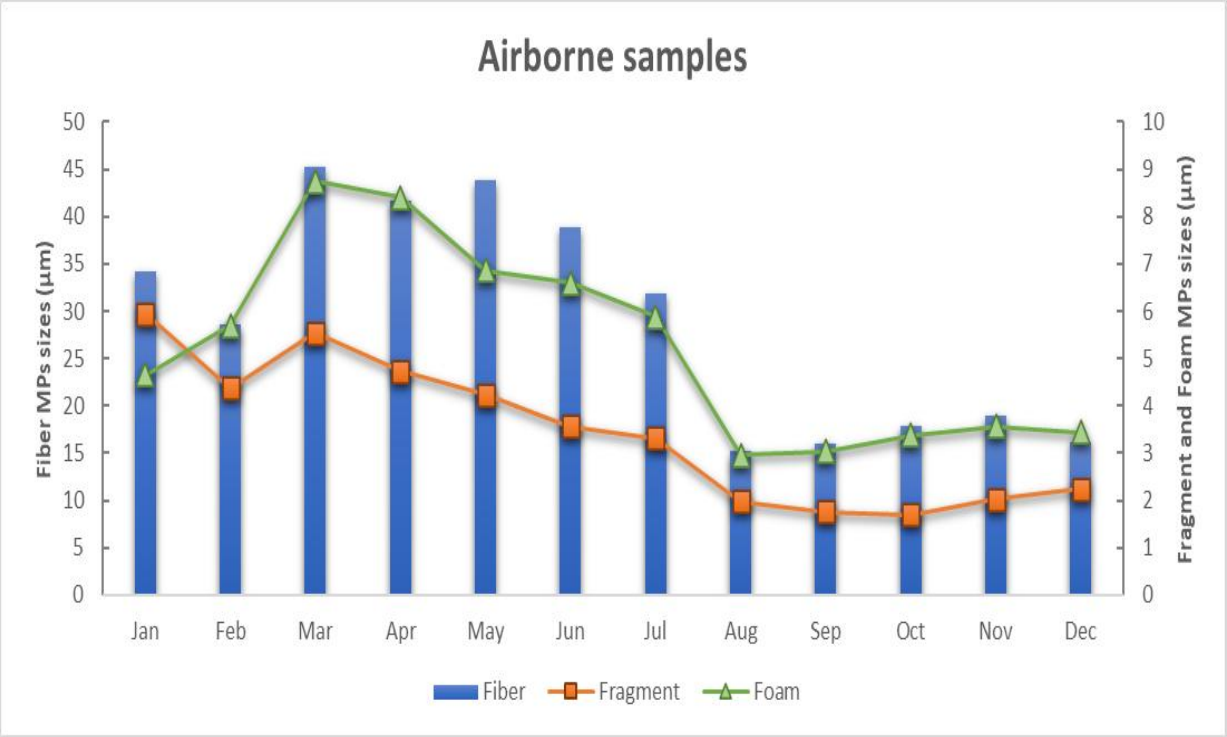


Figure SI5-5: Mean size of detected shapes in atmospheric deposition samples over 12 months

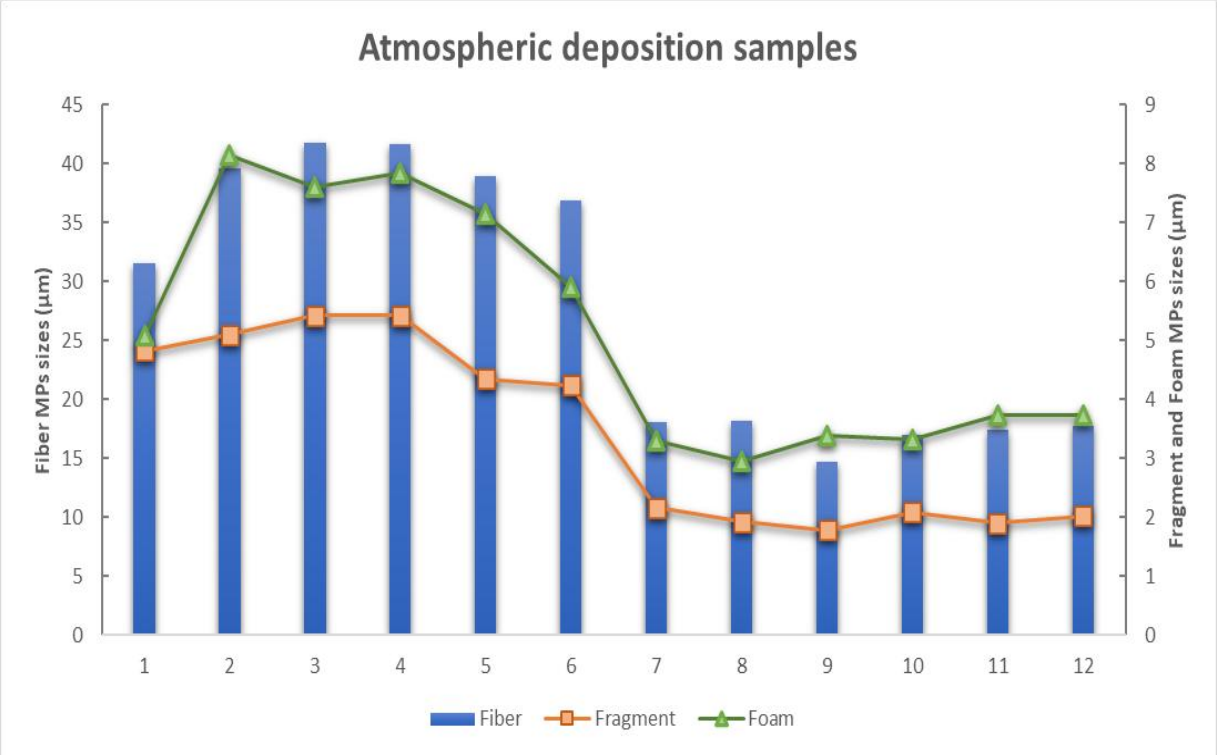
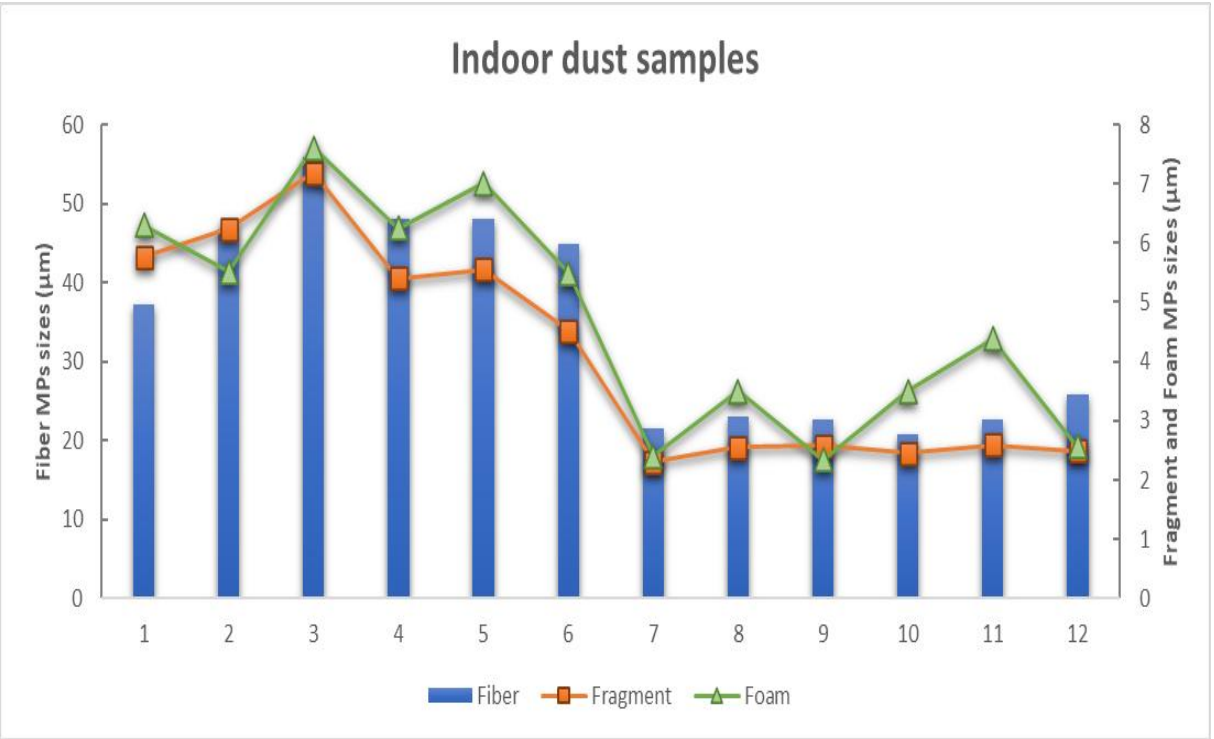


Figure SI5-6: Mean size of detected shapes in indoor dust samples over 12 months



Chapter 6

6.1 Summary and Conclusions:

Microplastics (MPs) are tiny plastic particles (<5 mm) that have become a significant environmental concern because of their widespread distribution, particularly in indoor microenvironments due to their presence in dust and air. As discussed in section 1.3, the primary sources of MPs originate from indoor sources such as furniture, synthetic textiles, and atmospheric deposition. They are also emitted from various household items like carpets, curtains, and cleaning agents. Studies show higher concentrations of MPs in indoor air and dust compared to outdoor environments.

Existing knowledge about human exposure to MPs in indoor environments is limited. The current understanding is that exposure occurs mainly via inhalation (air) and ingestion (food, drinks, dust). As discussed in section 1.4, inhalation is a significant exposure route, especially for smaller MPs particles that can reach the lungs and potentially cause respiratory issues. Settled indoor dust ingestion is no less important than the first pathway, particularly for exposure to various groups of hazardous chemicals, which has been extensively highlighted recently.

Little is known about MPs' toxicological endpoints and how they relate to environmentally relevant human exposure doses. As discussed in section 1.5, toxicity of MPs can cause oxidative stress, inflammation, and potentially translocate to organs. Smaller MPs (<20 μm) can be engulfed by cells and accumulate in different organs. Furthermore, chemical leaching in MPs can leach toxic additives like plasticizers and flame retardants, which pose additional health risks. In addition, Microbial Colonization of MPs can carry harmful microbes, posing further health risks.

The overriding aim of this thesis is to investigate human exposure to MPs in the indoor environment and assess the risk arising from such exposure. This was addressed via the determination of MPs occurrence, concentrations, and characteristics (i.e., size, shape, and polymer type) in indoor air and dust from several microenvironments in Birmingham, UK. It also identifies potential seasonal variation in the measured indoor MPs concentration via a 12-month monitoring period in 5 indoor microenvironments.

6.2 Key findings:

Our study of MPs in indoor air and inhalation exposure via airborne and atmospheric deposition samples from 60 UK microenvironments (homes and workplaces) is the first and most comprehensive study in this scope. The concentration of MPs was present in significant amounts, with higher concentrations often observed in homes compared to workplaces in both active and passive samples. This was mainly attributed to carpeted floors in all the sampled homes, while 13 of the studied workplaces were uncarpeted. MPs concentrations in atmospheric deposition were significantly higher than airborne MPs in the studied UK microenvironments. Nonetheless, a strong correlation ($P < 0.01$) was observed between the concentrations of MPs measured by active and passive sampling in the same microenvironments, which indicates common sources. MPs' shapes include fibers, fragments, and foam, with fibers being the most predominant shape detected in both environments, which may reflect the origin of these indoor MPs from carpets, textiles and domestic fabrics. The range of MP sizes (10 – 25 µm) was the most distributed in active samples and (50 – 100 µm) in passive samples from all microenvironments. Our results showed that PET, followed by PVC, were the most abundant polymer type in all the studied samples. The average daily inhalation exposure showed that toddlers inhaled MPs higher than adults. Regarding the potential health impacts, our study didn't directly measure health effects; it highlighted concerns about the potential for inhalation exposure to MPs and the need for further research

to understand the implications for human health. The study recommends reducing the use of plastic products and synthetic polymers to control potential sources of MPs in indoor environments and minimize human exposure to these particles.

Sixty samples of settled indoor dust collected were investigated to determine the concentrations, characteristics (i.e., size, shape, and polymer type) and ingestion exposure of MPs in UK homes and workplaces for the first time. In this study, the concentration of MPs was higher in homes than workplaces. This was mainly attributed to carpeting in all the sampled homes, while some studied workplaces were uncarpeted. The shapes detected were fibers, fragments, foams, and foil in all samples. The most distributed range of average particle size for MPs in the studied indoor dust samples was (50 – 100 μm). In terms of polymer types, PP and PET were the most abundant in the studied samples, followed by PVC, which reflects potential indoor sources including carpets, textiles, household appliances, food/beverage packaging, furniture and clothing. Human exposure estimates revealed exposure of toddlers to MPs via indoor dust ingestion was higher than that of adults, particularly on a body weight basis. Such high exposure to MPs raises concern over potential adverse health effects due to their incompletely developed immune and nervous systems to this vulnerable age group.

The seasonal variation study provides the first long-term monitoring of MPs concentrations and characteristics over a whole year indoors. The main objectives of this study were: (a) to investigate the seasonal variation of MPs in indoor environments, encompassing indoor air (both airborne and atmospheric deposition MPs) and settled indoor dust; and (b) to assess the impact of seasonal variation on human exposure to MPs through inhalation of indoor air and ingestion of settled indoor dust. This was achieved by determining the concentrations and characteristics (size and shape) of MPs on a monthly basis for a whole year (from January to December 2023), across five microenvironments (2 homes and 3 workplaces) in Birmingham, UK. The study's findings indicated that all sampling locations experienced the

highest concentrations of MPs during the summer months. The average concentration of airborne MPs from the studied microenvironments was 9 ± 2.3 MP/m^3, while the average concentration in settled indoor dust was 176.2 ± 96.7 MPs/mg over a 12-month monitoring period. Statistical analysis revealed significantly higher concentrations ($P < 0.05$) of MPs in indoor air and dust in summer compared to autumn and spring, respectively. This seasonal peak may be attributed to a multitude of factors, including ventilation, cleaning frequency, humidity levels, fluctuating indoor sources, and human activities that release secondary MPs due to the wear and tear of indoor plastic items.

In terms of human exposure, significant seasonal variations were observed in the estimated daily intakes of MPs by adults and toddlers, through both inhalation and inadvertent dust ingestion indoors. This heterogeneity raises concerns regarding the representativity and comparability of human exposure data currently available from multiple studies, which are often based on single "spot" samples collected at a certain point in time from diverse indoor microenvironments across different seasons.

6.3 Future research:

Microplastic pollution has recently garnered significant attention from the scientific community and the public due to its widespread presence in indoor environments. Therefore, advanced methods are required to determine the occurrence, concentration, and characteristics of MPs in indoor air and dust by fostering interdisciplinary collaboration among experts from diverse fields to improve the accuracy and reliability of measurements. Achieving reliable and reproducible results is crucial for understanding the full extent of the threat posed by MPs to human health. This necessitates a deep understanding of the benefits and limitations of the analytical techniques used in the MPs research field.

Further investigation is crucial to understand the fate, behaviour, and sources of hazardous MP chemical additives in indoor environments, as well as their potential risks to human health. Detecting MPs in microenvironments poses significant challenges. Additionally, there is currently no standardized procedure for the analysis of MPs in indoor settings. Various analytical approaches have been employed based on the characteristics of MPs, but a standardized method for their identification has yet to be developed.

Furthermore, it is essential to assess human exposure to MPs using comparable techniques over extended periods, such as those employed in our study during seasonal variations. Consistent methodologies are necessary to ensure data comparability across different studies. This requires the use of uniform sampling durations, analytical techniques, and quality control measures to accurately evaluate human exposure levels. Moreover, this thesis does not encompass several types of indoor microenvironments, such as hospitals, hotels, gyms, and places of worship. Therefore, conducting additional comparative studies is essential to assess MPs concentration levels in these settings and estimate human exposure more comprehensively to better understand the potential health risks associated with MPs in each specific indoor environment.

There is a critical gap in our current knowledge regarding the factors influencing human exposure to MPs through dermal contact. To date, dermal exposure remains less understood. In addition, the ability of MPs to penetrate the skin and enter the body is not well-documented. Factors such as particle size, shape, and the presence of chemical additives can influence the extent of skin absorption.

More comprehensive toxicological and epidemiological studies are required to better understand the risks associated with human exposure to MPs. Toxicological studies are crucial for assessing the potential health risks posed by MPs, as they investigate how MPs interact

with biological systems, including their potential to cause cellular damage, inflammation, oxidative stress, and other adverse effects. Additionally, MPs may contain chemical additives such as plasticizers, flame retardants, and stabilizers, which can leach out and pose further health risks. More reliable, robust and targeted toxicological studies are needed to help determine the bioavailability, toxicokinetic and adverse effects of these additives when leached from MPs. Epidemiological studies focus on understanding how humans are exposed to MPs through various pathways, including inhalation, ingestion, and dermal contact. These studies help quantify exposure levels and identify the most significant toxic effects of MPs. Furthermore, epidemiological research can identify correlations between MP exposure and health outcomes, such as respiratory issues, gastrointestinal problems, and chronic diseases.

In conclusion, by conducting more toxicological and epidemiological studies, researchers can provide the necessary data to establish safe exposure levels, inform policy decisions, and implement measures to protect public health from the potential risks associated with MPs.

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