



## Every breath you take

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Maurizi, Luca; Sanchez, Laura Simon; Vianello, Alvise; Nielsen, Asbjørn Haaning; Vollertsen, Jes

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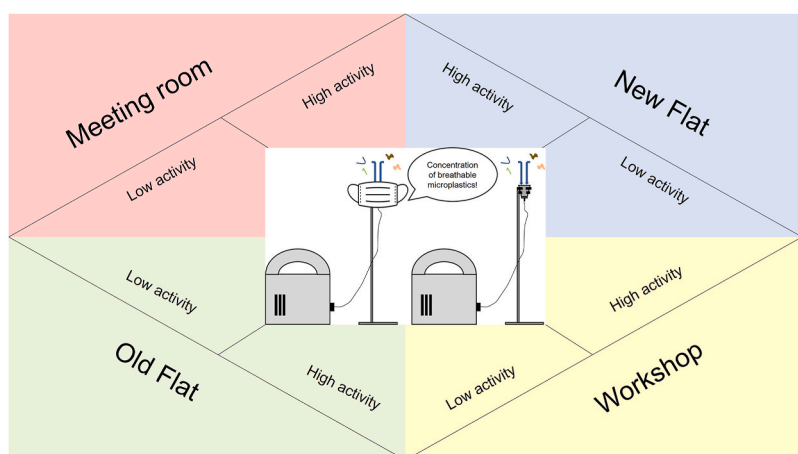


# Every breath you take: High concentration of breathable microplastics in indoor environments

L. Maurizi<sup>\*</sup>, L. Simon-Sánchez, A. Vianello, A.H. Nielsen, J. Vollertsen

Department of The Built Environment, Aalborg University, 9220, Aalborg, Denmark

## GRAPHICAL ABSTRACT



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## ABSTRACT

The widespread presence of microplastics (MPs) in the air and their potential impact on human health underscore the pressing need to develop robust methods for quantifying their presence, particularly in the breathable fraction ( $<5 \mu\text{m}$ ). In this study, Raman micro-spectroscopy ( $\mu\text{Raman}$ ) was employed to assess the concentration of indoor airborne MPs  $>1 \mu\text{m}$  in four indoor environments (a meeting room, a workshop, and two apartments) under different levels of human activity. The indoor airborne MP concentration spanned between 58 and 684 MPs per cubic meter ( $\text{MP m}^{-3}$ ) (median  $212 \text{ MP m}^{-3}$ , MPs/non-plastic ratio 0–1.6%), depending not only on the type and level of human activity, but also on the surface area and air circulation of the investigated locations. Additionally, we assessed in the same environments the filtration performance of a type IIR surgical facemask, which could overall retain  $85.4 \pm 3.9\%$  of the MPs. We furthermore estimated a human MP intake from indoor air of  $3415 \pm 2881 \text{ MPs day}^{-1}$  (mostly poly-amide MPs), which could be decreased to  $283 \pm 317 \text{ MPs day}^{-1}$  using the surgical facemask. However, for the breathable fraction of MPs ( $1\text{--}5 \mu\text{m}$ ), the efficiency of the surgical mask was reduced to 57.6%.

<sup>\*</sup> Corresponding author.

E-mail address: [lucam@build.aau.dk](mailto:lucam@build.aau.dk) (L. Maurizi).

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## 1. Introduction

Since the 1950s, the global use of plastic materials has dramatically increased (Andrady and Neal, 2009) due to their durability, adaptability to different purposes, and economic affordability. This, unfortunately, also led to a buildup of plastic waste in the environment due to mismanagement and increasing mass production (Ryan and Moloney, 1990).

In the recent years, the scientific community has focused on the pervasive presence of small plastic debris termed microplastics (MPs, 1–5000  $\mu\text{m}$ ; Gigault et al., 2018). MP pollution has ubiquitously been reported in natural and anthropogenic environments (Karbalaei et al. 2018), including the lower atmosphere (Allen et al. 2019; Brahney et al. 2020; Revell et al. 2021; Fan et al. 2022) and indoor air of households (Vianello et al. 2019; Xumiao et al. 2021), offices, educational institutes (Yao et al. 2022), and surgical environments (Field et al. 2022). Sources of indoor airborne MPs are synthetic clothes and textiles (Dris et al. 2015), tearing and weathering of building materials, abrasions from plastic products, landfilling, and waste incineration (Rahman et al. 2021; Kacprzak and Tijjing, 2022). Understanding the fate of airborne MPs is of pivotal importance, considering that nearly 90% of our life is spent indoors (Sarigiannis et al. 2019), and inhalation may represent humans' primary source of MP intake (Zhao et al., 2023; Wu et al. 2022).

Airborne MPs have recently been classified as Particulate Matter (PM) (Wang et al. 2021), which defines a mixture of solid particles and liquid droplets suspended in the air and associated with several environmental and health issues (He et al. 2022; Duffney et al. 2023). PM is classified according to the Aerodynamic Equivalent Diameter (AED) of the particles (Wieland et al. 2022), with  $\text{PM}_{10}$  (AED  $<10 \mu\text{m}$ ) and  $\text{PM}_{2.5}$  (AED  $<2.5 \mu\text{m}$ ) being intensely studied due to their ability to overcome mucociliary clearance in the upper airways (Warheit et al. 2001; Prata, 2018; Wieland et al. 2022). Particulates  $>5 \mu\text{m}$  are also called inhalable particulates, whereas the breathable (or respirable) particulates are represented by particles  $<5 \mu\text{m}$  (EN 481:1993) capable of reaching the alveoli in the lungs (Wieland et al. 2022). Finer particulates are more prone to cellular uptake, leading to stronger exposure response due to their surface charge (Silva et al. 2014), large surface-to-volume ratio (Schmid and Stoeger, 2016), and shape. Breathable MPs were recently reported by Amato-Lourenço et al. (2021) to accumulate in lung tissue. Particularly, airborne MP fibres might result in increased bioreactivity due to their shape (so-called “fibre paradigm”) as observed in other fibrous particulates e.g., asbestos (Shao et al. 2022). Fibrous MPs can accumulate in the lungs (Pauly et al. 1997) and may eventually cause long-term health effects (Wang et al. 2021, WHO global air quality guidelines, 2021). Specifically for airborne MPs, an increased cellular internalisation has also been observed after environmental exposure (Ramsperger et al. 2020). This may facilitate the penetration of other micropollutants (Wang et al. 2016) and pathogens (Kirstein et al. 2016) absorbed on the particles' surface. Moreover, additives, dyes, and pigments embedded in the polymeric matrix could leach out into the organism (Gasperi et al. 2018). Accordingly, in the field of occupational medicine, there is evidence indicating a link between exposure to particulates from the plastic industry and the occurrence of adverse health effects on humans (Pimentel et al. 1975; Soutar et al. 1980) e.g., chronic bronchitis (Miller et al. 1975), pneumoconiosis (Studnicka et al. 1995), and interstitial lung disease (Eschenbacher et al. 1999).

The measurement methods adopted for PM are generally gravimetric or real-time (Bo et al. 2017). In a gravimetric measurement, PM is collected on a surface or filter for subsequent analysis. Gravimetric methods can be performed either passively or actively i.e., by respectively employing a gravimetric collector (Eštoková et al. 2010; Baldelli et al. 2021) or a vacuum pump. Notably, the UNI EN 481:1993 and EPA 40 CFR PART 50 standards for  $\text{PM}_{10}$  and  $\text{PM}_{2.5}$  measurement recommend specific active sampling heads able to fractionate the PM according to its hydrodynamic diameter (Alfano et al. 2020). Real-time techniques normally employ optically based systems providing

instantaneous information on PM concentration, light scattering, and particle size. These devices are suitable for both spot measurements and long-term monitoring of various airborne pollutants e.g., volatile organic compounds (VOC) (Baldelli et al. 2020) and carbon dioxide ( $\text{CO}_2$ ) (Fromme et al. 2007).

For airborne MPs, only gravimetric methods are reported in the literature (Kek et al. 2024). While studies employing passive methods can be found (Cui et al. 2022), active methods are usually preferred since they provide more reliable data for concentration estimation (Torres-Agullo et al. 2022). The PM collected on the filters can subsequently be analysed with diverse techniques, depending on the information to be drawn from the samples besides chemical identification (i. e., number of MPs and morphology or MP mass). In addition to polymer identification, spectroscopic techniques such as Fourier-Transform InfraRed Micro-spectroscopy ( $\mu\text{FTIR}$ ) and Raman micro-spectroscopy ( $\mu\text{Raman}$ ) can provide data on MP morphology and number, while Pyrolysis-Gas Chromatography/Mass Spectrometry (Pyr-GC/MS) is used for the MP mass (Chen et al. 2020).

To bridge the current knowledge gap on the occurrence, distribution, and properties of airborne MPs in indoor environments (Vethaak and Legler, 2021), particularly in the breathable fraction, we investigated daily exposure to MPs in two private apartments and two workplace locations (a workshop and a meeting room). The study was driven by four hypotheses: i) Indoor air holds MPs down to the detection limit of the applied quantification technology ( $1 \mu\text{m}$ ); ii) Human activity affects the concentration and polymer types of airborne MPs; iii) A significant part of the airborne MPs are breathable ( $<5 \mu\text{m}$ ); and iv) commercial-type surgical masks reduce the exposure to airborne MPs.

## 2. Materials and methods

### 2.1. Investigated indoor locations

The indoor air sampling was conducted in four indoor environments in Aalborg (Denmark) in October–November 2022 (see also Supplementary Information, Figs. S1–S4). Briefly.

1. The workshop (hereafter “Workshop”,  $256.8 \text{ m}^2$ ) of the Department of the Built Environment, Aalborg University, where various materials (e.g., wood, concrete, metal, plastic, and electronic components) were stored, handled, and daily crafted by ten technicians. During weekends, no activity was carried out.
2. A meeting room (hereafter “Meeting Room”,  $8.1 \text{ m}^2$ ) in the same Department. It was furnished with three chairs, a table, a conference system, and a whiteboard. The room was actively ventilated by the building's ventilation system (G4 filter for particulates  $>10 \mu\text{m}$ , BS EN779). On workdays, the room's door was open, while it was kept closed on weekends.
3. An apartment located on the 1st floor of a renewed building from the late 19th century (hereafter “Old flat”,  $92 \text{ m}^2$ ). It comprised a studio, a kitchen, a living room, a bathroom, and a bedroom. The apartment was naturally ventilated and equipped with a wall radiator per room and a wooden floor.
4. An apartment on the 8th floor of a building from 2019 (hereafter “New flat”,  $70 \text{ m}^2$ ). It had four separate rooms (a bathroom, an open-plan kitchen/living room, a bedroom, and a studio). The apartment was equipped with a manually controlled air ventilation system (G4 filter for particulates  $>10 \mu\text{m}$ , BS EN779) and floor heating (installed beneath a wooden floor).

A workday was defined as one day between Monday morning at 8:00 a.m. and Friday afternoon at 4:00 p.m., while a weekend consisted of Saturday or Sunday. The level of activity in the investigated environments was categorised as high when humans were operating and low when they were not.

## 2.2. Indoor air sampling

Sampling was conducted with a stainless-steel funnel/filter holder (EMD Millipore Corporation, USA) placed at an average adult breathing height of 1.60 m by means of a laboratory stand (see also Supplementary Information) and connected to a laboratory air pump (Lab Logistics Group GmbH, Germany) with a flexible plastic hose. A 13 mm × 200 µm Silicon (Si) filter (Smart Membranes GmbH, Germany) of 1 µm pore size was placed at the bottom of the funnel and sealed using a polytetrafluoroethylene (PTFE) o-ring (approx. 10 mm inner diameter, 314 mm<sup>2</sup> inner area). The airflow (2 L min<sup>-1</sup>) was adjusted with the pump's flowmeter (RS Components A/S, Denmark) and was directed inward from the indoor atmosphere to the centre of the funnel. Each sampling session was conducted for 24 h (BS EN 12341:20231:2014), with a total filtered volume of approx. 2.9 m<sup>3</sup> per sample. Air samples were collected in each location on workdays and weekends (n = 8). Moreover, additional samples (n = 8) were taken under the same conditions with a type IIR surgical facemask mounted on the inlet of the sampling funnel with elastic rubber bands. After the sampling, the Si filters were carefully removed from the holder and stored in pre-cleaned Petri dishes until analysis.

## 2.3. Contamination prevention

The funnel's components, Si filters, metal labware, and facemasks were rinsed with pre-filtered ethanol (GF-F 0.7 µm) and flushed in a flow bench with pre-filtered nitrogen gas (1.5 Bar) before use to remove potential contamination from the external environment. All employed laboratory glassware was previously muffled at 500 °C for 4 h, and 100% cotton lab coats were worn during the entire operation. In addition, three clean 1 µm Silicon filters were analysed as procedural blanks to account for potential contamination. During data processing, particles identified as PTFE were not considered when calculating the MP concentration due to the potential contamination from the sampling setup.

## 2.4. Chemical analysis of the surgical mask fabric

The surgical facemasks used for the air sampling were analysed by Attenuated Total Reflection FTIR (ATR – FTIR, Cary 630 FTIR, Agilent, USA) to determine the main materials constituting its fabric. The spectral range was 600–4000 cm<sup>-1</sup>, collected at 4 cm<sup>-1</sup> resolution. Sixty-four scans were collected for the background. The raw spectra of the facemasks were automatically corrected (apodisation) by the instrument's software (MicroLab PC, Agilent, USA) and compared with the Agilent Polymer Handheld ATR library. The material of the facemask was identified as PP (Supplementary Information).

## 2.5. Chemical analysis of the indoor particulate

The filters were analysed with a Nano Xplora Plus confocal Raman microscope equipped with a Peltier-cooled 2048 × 70 pixel CCD Synchronicity detector, a 638 nm solid-state laser source (Horiba SAS, France), and a 50 × objective with a numerical aperture (NA) of 0.75 (Olympus, Japan). The software LabSpec6 (Horiba SAS, France) was used to set the analysis parameters, whilst the visible image analysis was performed with the module Particle Finder (Horiba SAS, France).

First, the visible montage of each filter was collected with the 50 × objective and automatically analysed with the built-in algorithm Max Entropy (intensity threshold values 89–256). The algorithm computed the size of each particle in the visible montage according to several parameters (e.g., area, perimeter, diameter, etc.) as well as the XY coordinates on the filter surface. Moreover, upon acquiring the visible montage, sub-micron items were excluded in Particle Finder by virtually pre-filtering all the particles with a maximum Feret diameter below 1 µm. A table gathering all the items with a max Feret diameter above 1 µm, their XY coordinates, and the relevant morphological parameters for

each of them was obtained (see also Supplementary Information).

The Raman spectrum of each particle was then automatically acquired by Particle Finder. The µRaman system was previously calibrated on Si first-order Raman emission (520.7 cm<sup>-1</sup>) by zero-order correction with a single-crystal Si wafer. For the spectral acquisition, the following instrument parameters were chosen: slit 100 µm, hole 300 µm, grating 600 l mm<sup>-1</sup> (spectral range 0–3500 cm<sup>-1</sup>), and laser wavelength 638 nm at 100% power (40.2 mW). The laser was automatically driven to each particle whose position on the filter was obtained in the visible montage analysis step. Moreover, the analysis of each filter was split into two sub-sessions according to the diameter range of the particles to be analysed. For the fraction above 10 µm in diameter, 100% of the particles on the filter were analysed (1 s × 6 accumulations per item).

For the fraction 1–10 µm, 10% of particles were randomly selected on the entire active area of the filter by choosing the “Random” analysis mode of Particle Finder, which subsequently performed the spectral acquisition of the chosen particles automatically (4 s × 2 accumulations per item). For the procedural blanks, 100% of the particles in both size fractions were analysed with the same analysis parameters as for the samples, given the relatively low number of particles on the blank filters. See also Supplementary Information for further details on the µRaman analysis process.

The spectral identification of the µRaman spectra was conducted with the software siMPle (Primpke et al. 2020) v. 1.3.1β. The signals from the Si filter (~519 and ~990 cm<sup>-1</sup>) were automatically removed, and the corrected spectra were compared with the SLOPP/SLOPP-E library (Munno et al. 2020) (Supplementary Information, section 5 for some examples of experimental Raman spectra from the air samples). The spectra presenting a matching quality above 0.50 were considered for further expert validation to assess the reliability of the identification.

For simplicity, the identified polymers were grouped into the following clusters: Other (cellulose acetate, ethylene vinyl alcohol copolymer (EvOH), poly-carbonate, poly-methyl-methacrylate, poly-formaldehyde, poly-urethane (PUR), silicone, and styrene – isoprene (SIS) rubber), PA (poly-amide), PE (poly-ethylene, poly-ethylene chloride, poly-ethylene-co-propylene), PEST (poly-butylene-terephthalate, poly-ethylene-terephthalate), PP (poly-propylene), PS (acrylonitrile-butadiene-styrene, poly-styrene), and PV (poly-vinyl acetate, poly-vinyl butyral, poly-vinyl chloride, poly-vinyl alcohol, poly-vinyl pyrrolidone).

## 2.6. Data analysis

MP concentrations were not blank-corrected and were expressed as number of MPs per cubic meter (MPs m<sup>-3</sup>). The limit of quantification (LOQ) for the polymers found in the blanks was calculated from equation (1):

$$\text{LOQ} = m_{\text{blank}} + 10 \cdot \sigma_{\text{blank}} \quad (1)$$

Where  $m_{\text{blank}}$  is the mean number of particles of each polymer identified in the blank filters and  $\sigma_{\text{blank}}$  its standard deviation. Hence LOQ was expressed as number of MPs per filter (MPs filter<sup>-1</sup>), and for those polymers found in the samples but not in the blanks, LOQ = 0.

The human exposure to airborne MPs per day (HE<sub>day</sub>) was calculated from equation (2):

$$\text{HE}_{\text{day}} = [\text{MPs}] \times I \times T \quad (2)$$

Where [MPs] is the MP concentration in MPs m<sup>-3</sup>, I is the inhalation rate equal to 16 m<sup>3</sup> day<sup>-1</sup> for an adult male (Stifelman, 2007), and T is the exposure time in hours day<sup>-1</sup> (8/24 = 0.33 for the two workplaces on workdays and 24/24 = 1 for the two flats on weekends).

The filtration efficiency (F) of the surgical facemask was calculated according to equation (3) for both the overall particles found on the filters and the MPs:

$$F(\%) = \frac{[\text{Particles}]_{\text{mask}} - [\text{Particles}]_{\text{nomask}}}{[\text{Particles}]_{\text{nomask}}} \times 100\% \quad (3)$$

Where  $[\text{Particles}]_{\text{mask}}$  is the total particle concentration in particle number per  $\text{m}^3$  obtained from the samples taken with the surgical facemask, and  $[\text{Particles}]_{\text{nomask}}$  is the corresponding total particle concentration from the samples taken without the facemask.

The software R v. 4.2.1 was used to analyse the datasets and perform the statistical tests. Shapiro-Wilk test was employed to assess the normality of the data. A Kruskal-Wallis test was performed to assess the significance of MP concentration, MP maximum Feret diameter, and the estimated human exposure, and a Dunn test was done for pair-wise comparison ( $\alpha = 0.05$ ). For the MP diameter and polymeric composition, a generalised linear model (GLM) was used to assess the significance of the data and for the pair-wise comparison among the crossed factors "Location" (Meeting room, New flat, Old flat, and Workshop), "Activity" (High, Low), and "Facemask" (Without Facemask, Surgical Facemask). The p-value of the GLM test was calculated from the corresponding Wald z value and was considered significant if  $< 0.05$ . A Principal Component Analysis (PCA) was conducted on the MP diameter distribution frequency data to explore how the use of the facemask influenced the diameter distribution of MPs. The results in section 3 were expressed as median or mean  $\pm$  one standard deviation (SD) as specified in the discussion.

### 3. Results and discussion

#### 3.1. Blank contamination

A mean of  $22 \pm 14$  MPs was found in the procedural blanks, corresponding to  $8.1 \pm 5.5\%$  of the mean MP number found in the samples ( $414 \pm 344$  MPs). The median maximum Feret diameter of the MPs found in the blanks was  $1.5 \mu\text{m}$  (min. – max.  $1.0\text{--}17.0 \mu\text{m}$ ). The main polymeric composition of these particles was PUR 73.8% (LOQ = 129 MPs filter<sup>-1</sup>), PA 10.8% (LOQ = 8 MPs filter<sup>-1</sup>), PP 7.7% (LOQ = 5 MPs filter<sup>-1</sup>), PS 4.6% (LOQ = 1 MP filter<sup>-1</sup>), and PE 3.1% (LOQ = 1 MP filter<sup>-1</sup>). The predominance of PUR was probably caused by remaining traces from the packaging material of the Si filters, made of a PUR foam layer. The number of MPs sampled on the filters at the investigated locations was always higher than the LOQ.

#### 3.2. Microplastics versus non-plastic materials

The mean MPs/non-plastic ratio for the samples taken without the facemask was  $0.2 \pm 0.1\%$ . Among the non-plastic particles, most of them were identified as cotton (52.2%) and cellulose (47.8%), but small amounts of calcium sulfate ( $\text{CaSO}_4$ ,  $< 0.1\%$ ) were also present.

Overall, the data showed a lower MPs/non-plastic ratio than the one reported by Vianello et al. (2019) (4%), whose dataset included only organic particles, and that of Torres-Agullo et al. (2022) (16%). Both studies used  $\mu\text{FTIR}$  for particle identification, which could limit a direct comparison with the present results. Moreover, the MPs/non-plastic ratio reported in the current study may well depend on the semi-randomised strategy chosen to analyse the filters and the settings employed for the spectral recognition (i.e., the spectral library and the quality threshold set in siMPLe).

#### 3.3. Microplastics in indoor environments

The presence of MPs in indoor environments has been widely documented (Yao et al. 2022; Uddin et al. 2022; Zhai et al. 2023; Zhao et al. 2023). Similarly, the current study also showed that airborne MPs were present at almost all investigated locations ( $58\text{--}684 \text{ MP m}^{-3}$ , median  $212 \text{ MP m}^{-3}$ ). Overall, the median MP concentration varied significantly with the level of human activity (Kruskal – Wallis  $p = 0.03$ ), ranging from  $85$  to  $335 \text{ MP m}^{-3}$ .

The two private apartments had the highest median MP concentrations, though there was a considerable variation between them (old flat:  $548 \text{ MP m}^{-3}$ , new flat:  $185 \text{ MP m}^{-3}$ ). This difference may be due to the presence of a mechanical ventilation system in the new flat. When equipped with adequate filters, these devices can indeed remove particulates, here among MPs, from the air (Isaxon et al. 2015; Stabile et al. 2019; Chen et al. 2022). The old flat was only naturally ventilated by opening the windows during weekends (high activity), which could explain the lower MP concentration measured during this period ( $412 \text{ MP m}^{-3}$ ). At low activity and when the windows were closed,  $684 \text{ MP m}^{-3}$  were measured.

The workshop held a lower median MP concentration ( $179 \text{ MP m}^{-3}$ ) than the two apartments, and no MPs were found in the sample collected during the weekend (i.e., low activity). As shown by Mølgaard et al. (2015), human activity can be a source of airborne particulate, especially in indoor environments where heavy work (e.g., cutting, sawing, and grinding of rough surfaces) is carried out. Hence, the temporary inactivity during the weekend may explain the observed outcome of the workshop. Moreover, the large area of this location ( $256.8 \text{ m}^2$ , section 2.1) and the height of the ceiling ( $3.5 \text{ m}$ ) may potentially have led to a dilution of the airborne MPs (Memarzadeh and Jiang, 2004). The lowest median MP concentration was found in the meeting room ( $92 \text{ MP m}^{-3}$ ), which also had a higher MP concentration at low activity than at high activity ( $112 \text{ MP m}^{-3}$  vs  $73 \text{ MP m}^{-3}$ ). Since the door was kept slightly open on workdays (i.e., high activity) and closed on weekends, the air exchange with the external environment may have diluted the airborne MPs during high activity (Hussein, 2017; Liu S. et al. 2022).

Previous studies reported lower indoor airborne MP concentrations for residential environments in Denmark ( $2\text{--}16 \text{ MP m}^{-3}$ ; Vianello et al. 2019), Portugal ( $0.7\text{--}1.6 \text{ MP m}^{-3}$ ; Xumiao et al. 2021), and Sri Lanka ( $0.1\text{--}0.9 \text{ MP m}^{-3}$ ; Perera et al. 2022). These studies generally targeted MPs above  $10 \mu\text{m}$  and employed different sampling strategies and analytical methods (Kacprzak and Tijing, 2022), challenging comparison among studies. Xie et al. (2022) used similar analytics to our study to investigate the occurrence of airborne MPs  $> 1 \mu\text{m}$  at outdoor and indoor locations in Shanghai (China) but reported lower indoor concentrations ( $16\text{--}93 \text{ MP m}^{-3}$ ). The difference might be explained by the low number of  $1\text{--}10 \mu\text{m}$  MP detected by Xie et al. (2022), who visually pre-screened particles  $> 1 \mu\text{m}$  prior to the  $\mu\text{Raman}$  analysis. As previously documented, this approach is prone to underestimate the smaller particles, as opposed to automatised and randomised strategies like the one presented in our study (Primpke et al. 2020; Schymanski et al. 2021; Duarte et al. 2022). Such automatised protocols eliminate the intrinsic subjectivity related to the sorting/pre-selection step and may potentially be applied to pollutants other than MPs (e.g., coatings (Bouchard et al. 2009), asbestos fibres (Rinaudo et al. 2010), and titanium dioxide (Mamedov, 2020)).

#### 3.4. Microplastic composition and morphology of the indoor air

##### 3.4.1. Polymeric composition

A total of 15 polymers were identified in the analysed samples (Fig. 1), with PA clearly dominating the polymer composition (mean 21%), followed by PV (mean 18%), PE (mean 16%), PS (mean 11%), and PEST (mean 8%). The sampling locations significantly influenced the MP polymeric composition (GLM  $p = 7 \cdot 10^{-4}$ ), as opposed to the level of human activity (GLM  $p = 0.49$ ).

PA, PE, PEST, and PS are the most common polymers recorded in indoor air (Habibi et al. 2022). Synthetic fabrics (Periyasamy et al. 2020), washing machines, and tumble dryers are generally considered relevant emission hotspots for PA and PEST (Periyasamy and Tehrani-Bagha, 2022; Kärkkäinen and Sillanpää, 2021). Accordingly, when tumble dryers were used in the flats (high activity level i.e., weekends), PEST showed higher frequency (new flat: 7.0%, old flat: 20.7%), as opposed to workdays (new flat: 0%, old flat: 7.6%). Similarly, a higher PA concentration was recorded in the "old flat" during high

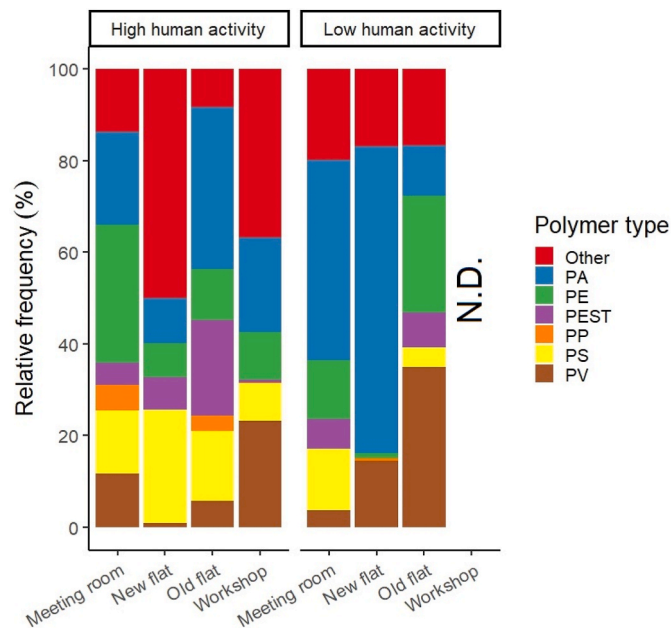


Fig. 1. Polymer relative frequency in the investigated indoor locations according to human activity.

activity (35.3% vs 10.8% in the weekend). PS was mainly recorded in the meeting room (mean 13.5%) and apartments (new flat mean: 12.4%; old flat mean: 9.7%). This polymer group is widely used in home appliances (Pious and Thomas, 2016), the packaging industry (Block et al. 2017), and as a concrete additive (Dixit et al. 2019).

Under high activity conditions, the category “Other” held the highest frequency in the new flat (49.9%), followed by the workshop (36.7%), meeting room (13.7%), and old flat (8.2%). Within this polymer cluster, cellulose acetate (mean 2.9%) and PUR (mean 14.5%) were the most abundant. Cellulose acetate is a common material used in textiles and yarns (Law, 2004). PUR is used within the furniture industry as a coating and sealant (Zia et al. 2007) in conjunction with flame retardants, most of which have been deemed potentially hazardous (U.S. Environmental Protection Agency, 2015).

### 3.4.2. Microplastic morphology

MPs with a length/width ratio  $>3$  were classified as fibres; otherwise, they were reported as fragments (Vianello et al. 2019). The detected MPs were mainly characterised as irregular fragments (99.4%), with the fibres representing only 0.6% (see also Supplementary Information). These findings contrast most studies investigating airborne MPs, where fibrous-shaped particles were commonly predominant (Dris et al. 2015, 2017; Jenner et al. 2021; Perera et al. 2022). However, most of these investigations relied on visual and manual pre-sorting of putative MPs, which is prone to bias due to the relative ease of recognising fibres (Song et al. 2015).

Overall, the maximum Feret diameter of airborne MPs were 1–67.5  $\mu\text{m}$ , with a median of 6.2  $\mu\text{m}$  ( $>10$   $\mu\text{m}$  median diameter: 13.6  $\mu\text{m}$ , 1–10  $\mu\text{m}$  median diameter: 3.2  $\mu\text{m}$ ). Most MPs were 1–10  $\mu\text{m}$  (72.6%), with 1–5  $\mu\text{m}$  MP representing the majority (mean 45.2%). In contrast, MPs  $\geq 50$   $\mu\text{m}$  were scarce (0.1%), most likely because larger MPs may settle to a greater extent than smaller ones (Zhang et al. 2020a,b). The frequency of 1–10  $\mu\text{m}$  MP was significantly higher (74.6%) under high activity conditions than during low activity (70.4%) (Kruskal-Wallis  $p = 0.03$ ). Hence, the level of activity might influence the MP diameter in the indoor air by either producing or resuspending airborne MPs. The occurrence of inhalable and respirable MPs seems, therefore, inevitable and generally associated with human activity (Prata et al. 2021). Furthermore, Amato-Lourenço et al. (2021) showed the presence of MP

fragments between 1.6 and 5.6  $\mu\text{m}$  in human lung tissue using  $\mu\text{Raman}$ , which indirectly corroborates our findings on the high frequency of 1–10  $\mu\text{m}$  MP in indoor air.

### 3.5. Surgical facemasks reduce the overall exposure but have low efficiency on the breathable fraction

Applying a surgical facemask to the inlet of the sampling device led to significantly lower MP concentrations (Kruskal-Wallis  $p = 0.03$ ), ranging from 4 to 196  $\text{MP m}^{-3}$ . Fig. 2 shows the median MP concentration (i.e., average of low and high activity) in the investigated locations for the samples taken without and with the surgical facemask.

The surgical facemask reduced the total amount of indoor airborne MPs by preferentially filtering out MP above 5  $\mu\text{m}$  (Fig. 3a). Accordingly, the median maximum Feret diameter of MPs found in the indoor air samples (collected without facemask) was significantly larger than in those taken with facemask for both the fractions  $>10$   $\mu\text{m}$  (13.6  $\mu\text{m}$  vs 12.7  $\mu\text{m}$ , Kruskal-Wallis  $p = 3.48 \cdot 10^{-5}$ ) and 1–10  $\mu\text{m}$  (3.2  $\mu\text{m}$  vs 2.2  $\mu\text{m}$ , Kruskal-Wallis  $p = 1.34 \cdot 10^{-6}$ ). Additionally, a PCA (Fig. 3b) showed that the 1–5  $\mu\text{m}$  diameter range mostly described the variance of the MP diameter related to the samples taken with a surgical facemask. Consequently, the two sets of samples (with and without facemask) showed a significant difference with regard to the MP diameter (GLM  $p = 6.11 \cdot 10^{-14}$ ).

The overall filtration efficiency of the surgical facemask for the indoor airborne MPs was  $85.4 \pm 3.9\%$ . Different efficiencies were found for each MP diameter range: 1–5  $\mu\text{m}$ : 57.6%, 5–10  $\mu\text{m}$ : 85.4%, 10–20  $\mu\text{m}$ : 89.0%, 20–50  $\mu\text{m}$ : 94.9%, 50+  $\mu\text{m}$ : 100%. Overall, these results indicate that a surgical facemask helps to reduce the exposure to MPs down to 5  $\mu\text{m}$ , but it loses efficiency on 1–5  $\mu\text{m}$  MP, which may represent an increased risk to human health (Wieland et al. 2022). These findings agree well with the work of Tcharkhtchi et al. (2021), who also reported a filtration efficiency of approx. 50% for commercial surgical facemasks tested with aerosols  $<5$   $\mu\text{m}$ .

### 3.6. Estimation of the MP human intake from indoor air

The potential MP human intake was estimated for the periods with

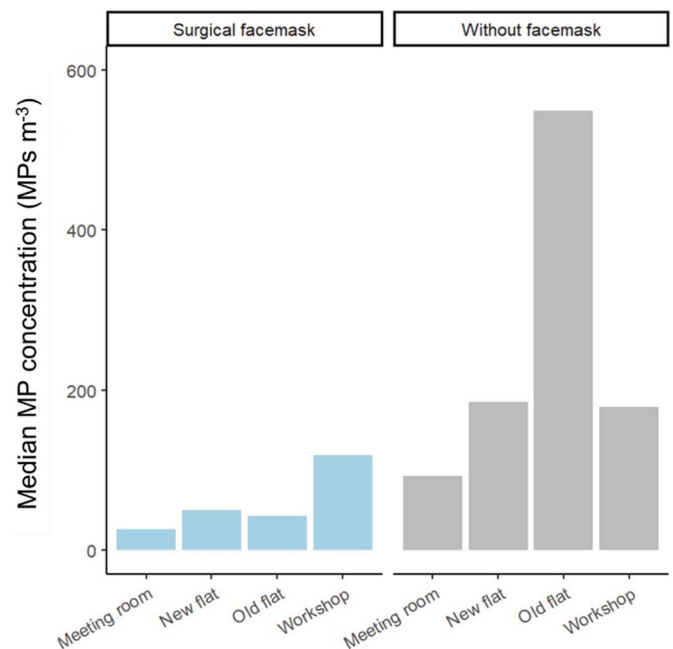


Fig. 2. Median MP concentration ( $\text{MPs m}^{-3}$ ) in the investigated indoor locations collected with a surgical facemask (blue) and without (grey).

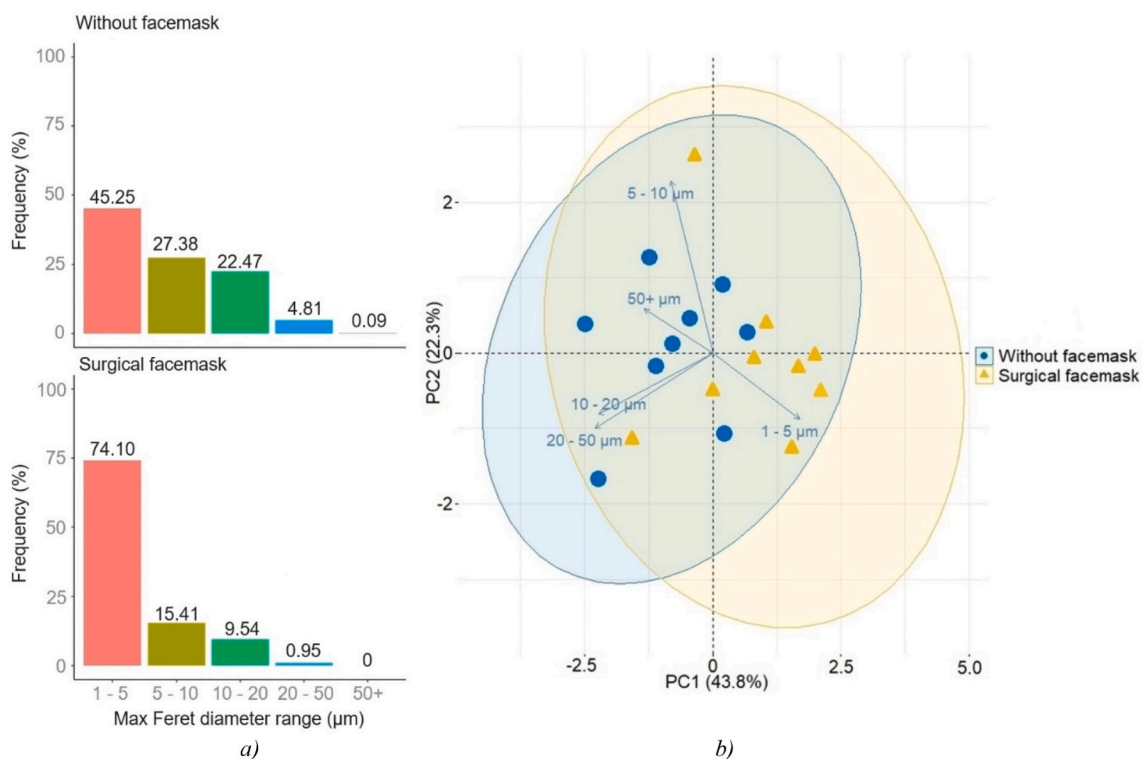


Fig. 3. a) MP relative frequency per diameter range in the investigated samples; b) Principal Component Analysis (PCA) of the MP diameter frequency (scores plot).

high activity (i.e., workdays for the two public settings and weekends for the two flats). Table 1 summarises the results of the MP intake estimation, both for the overall MP count and specifically within the breathable fraction (diameter 1–5  $\mu\text{m}$ ).

The values of  $\text{HE}_{\text{day}}$  obtained from the samples without a facemask are significantly higher than those obtained with the surgical mask (Kruskal-Wallis  $p = 0.04$ ). Most of the previous studies estimated the human exposure to airborne MPs above 10  $\mu\text{m}$  (Vianello et al. 2019: 272  $\text{MPs day}^{-1}$ , Cox et al. 2020: 170  $\text{MPs day}^{-1}$ , Soltani et al. 2021: 35  $\text{MPs day}^{-1}$ , Torres-Agullo et al. 2022: 37  $\text{MPs day}^{-1}$ ). With our approach, we estimated a mean  $\text{HE}_{\text{day}}$  of 867  $\text{MPs day}^{-1}$  for the MPs >10  $\mu\text{m}$  (high activity conditions), which suggests that past investigations may have underestimated the potential impact on human health of airborne MP pollution in indoor environments.

To date, only the work of Xie et al. (2022) reported the human intake of airborne indoor MPs between 1 and 10  $\mu\text{m}$ , obtaining values between 20 and 892  $\text{MPs day}^{-1}$ . In this size fraction, we found, on average, 2548  $\text{MPs day}^{-1}$  during high indoor activity. The difference might be due to factual differences in air concentrations between the two studies or to the analytical methods adopted. The fact that Xie et al. employed visual

Table 1

Daily human exposure to airborne indoor MPs in the four investigated locations without facemask and with surgical facemask (high activity periods).

Location	$\text{HE}_{\text{day}}$ to indoor MPs ( $\text{MPs day}^{-1}$ )		$\text{HE}_{\text{day}}$ to indoor 1–5 $\mu\text{m}$ MP ( $\text{MPs day}^{-1}$ )	
	Without surgical facemask	With surgical facemask	Without surgical facemask	With surgical facemask
Meeting room	352	123	150	67
New flat	4991	753	1915	632
Old flat	6600	67	3222	56
Workshop	1717	190	730	50
Mean $\pm$ SD	3415 $\pm$ 2881	283 $\pm$ 317	1504 $\pm$ 1360	201 $\pm$ 287

pre-screening of individual particles, while the current study applied an automatised and randomised approach, might explain the higher number of MPs detected in our study. Moreover, approx. 45% of our estimated value was represented by 1–5  $\mu\text{m}$  MP (1504  $\text{MPs day}^{-1}$ , Table 1), the finer and potentially more harmful size fraction of airborne MPs. Notably, the MP fraction 1–5  $\mu\text{m}$  would still be inhaled despite the use of a surgical facemask at a rate of 201  $\text{MPs day}^{-1}$ , accounting for 71% of the estimated total intake (283  $\text{MPs day}^{-1}$ , Table 1).

#### 4. Conclusions

This work expands the current knowledge on indoor airborne MP occurrence by providing novel insights into the inhalable and breathable fraction of this synthetic particulate. The concentration of indoor airborne MPs down to 1  $\mu\text{m}$ , assessed at four locations (two workplaces and two private apartments) during low and high-activity periods, indicates that human activity is a major source of MP pollution in indoor environments. At the same time, humans are also potentially more prone to MP intake during high-activity periods, highlighting that spending time indoors increases the risk of inhaling and potentially breathing airborne MPs. This intake may be partially reduced by using a surgical facemask, but the exposure to finer MPs (1–5  $\mu\text{m}$ ) would still be non-negligible since almost half of MPs in this size range were not retained by the device's fabric.

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#### CRedit authorship contribution statement

L. Maurizi: Writing – review & editing, Writing – original draft,

Visualization, Methodology, Investigation, Data curation, Conceptualization. **L. Simon-Sánchez:** Writing – review & editing, Visualization, Supervision, Methodology, Investigation, Data curation. **A. Vianello:** Writing – review & editing, Visualization, Supervision, Methodology, Conceptualization. **A.H. Nielsen:** Writing – review & editing, Supervision. **J. Vollertsen:** Writing – review & editing, Supervision.

### Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Luca Maurizi reports financial support was provided by Horizon Europe.

### Data availability

Data will be made available on request.

### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.chemosphere.2024.142553>.

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