

**Minireviews** 

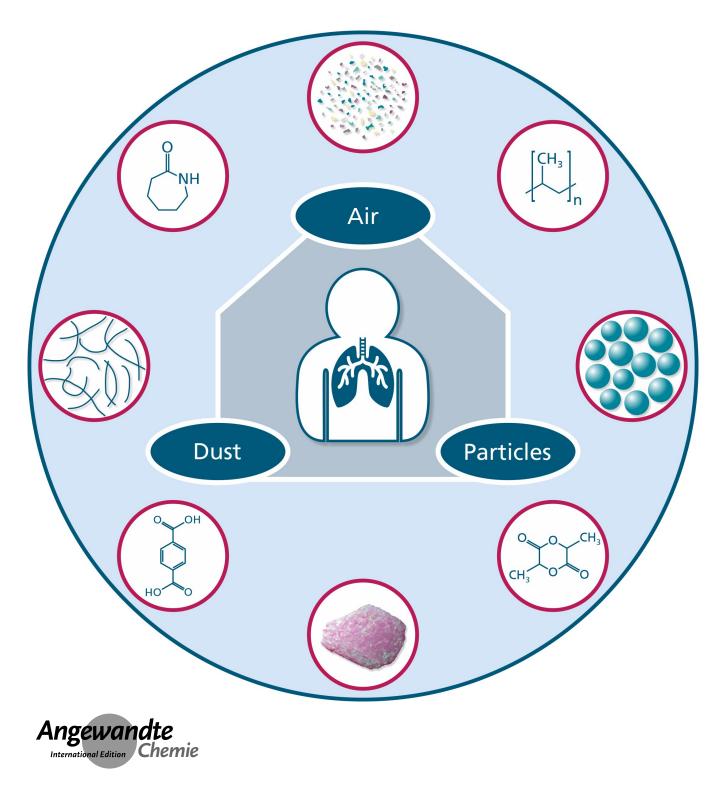


Environmental Chemistry

How to cite: Angew. Chem. Int. Ed. 2022, 61, e202205713International Edition:doi.org/10.1002/anie.202205713German Edition:doi.org/10.1002/ange.202205713

# Microplastics and their Additives in the Indoor Environment

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Angew. Chem. Int. Ed. 2022, 61, e202205713 (1 of 10)

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**Abstract:** Analyses of air and house dust have shown that pollution of the indoor environment with microplastics could pose a fundamental hygienic problem. Indoor microplastics can result from abrasion, microplastic beads are frequently added to household products and microplastic granules can be found in artificial turf for sports activities and in synthetic admixtures in equestrian hall litter. In this context, the question arose as to what extent particulate emissions of thermoplastic materials from 3D printing should be at least partially classified as microplastics or nanoplastics. The discussion about textiles as a possible source of indoor microplastics has also been intensified. This Minireview gives an overview of the current exposure of residents to microplastics. Trends can be identified from the results and preventive measures can be derived if necessary. It is recommended that microplastics and their additives be given greater consideration in indoor environmental surveys in the future.

### 1. Introduction

The accumulation of small plastic pellets and granules in the environment was already discussed at the beginning of the 1980s,<sup>[1]</sup> but it was not until 2004 that Thompson et al.<sup>[2]</sup> first formulated the term "microplastics". Today, microplastics as defined by the European Chemicals Agency (ECHA) are understood as solid plastic particles consisting of mixtures of polymers and functional components. Microplastics can be formed unintentionally, for example through abrasion, or intentionally when added to products to achieve certain properties. According to the European Food Safety Authority (EFSA),<sup>[3]</sup> the size of microplastics ranges from 0.1  $\mu$ m to 5000  $\mu$ m and nanoplastics from approximately 1 nm to 100 nm.

On a global scale, microplastics are primarily a problem in marine ecosystems.<sup>[4]</sup> In recent years, however, it has been recognized that microplastics are emerging particulate pollutants that can be found in all environmental compartments.<sup>[5]</sup> Because people living in industrialized areas spend much of their time indoors,<sup>[6]</sup> exposure via indoor air and house dust pathways needs considerable attention. It must also be noted that an indoor environment is not only understood as a private residence. Rather, indoor spaces, for example, include office workplaces, public buildings, schools, universities, sports halls, and passenger cabins.

Airborne particles and house dust have been intensively studied indoors for decades.<sup>[7]</sup> In the case of airborne particles, however, with the exception of asbestos and mineral fibers, measurements were largely carried out with regard to mass and number concentration. In the case of house dust, the focus is traditionally on the chemical and biological components.<sup>[8]</sup> Only recently has there been a move to specifically examining particles and dust for plastics.<sup>[9]</sup> This late realization is surprising, as there are

[\*] Prof. Dr. T. Salthammer
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◎ © 2022 The Authors. Angewandte Chemie International Edition published by Wiley-VCH GmbH. This is an open access article under the terms of the Creative Commons Attribution Non-Commercial NoDerivs License, which permits use and distribution in any medium, provided the original work is properly cited, the use is noncommercial and no modifications or adaptations are made. numerous direct and indirect sources of plastics and their additives in the indoor environment. These include personal care products and paints,<sup>[10]</sup> artificial turf in sports halls,<sup>[11]</sup> as well as abrasion from floorings, furniture, and textiles. 3D printing has significantly increased the processing of thermoplastics indoors.<sup>[12]</sup> Plastic materials can also be brought in from the outside via the air exchange. It is well known that the abrasion of car tires is one of the strongest sources of microplastics worldwide.<sup>[10]</sup> In addition, many plastics contain volatile and semivolatile organic compounds (VOCs and SVOCs) as additives that can be released during use.<sup>[13]</sup>

Microplastics are now ubiquitous. However, the available literature is still inconclusive with regard to possible health hazards from microplastics in the indoor environment.<sup>[14]</sup> This consequently results in the immediate need for further monitoring. A paradigm shift has recently taken place in indoor- and exposure-related studies insofar that emerging pollutants are now also being analyzed for preventive purposes.<sup>[15]</sup> This Minireview summarizes the current state-of-the-art on the subject of microplastics in indoor environments with the aim of including this aspect more closely in future investigations and surveys.

# 2. Analytical Techniques for the Characterization of Microplastics and Additives

Various tools for sampling, separation, and analysis are available to determine the chemical and physical properties of microplastics. Crawford and Quinn<sup>[4b]</sup> as well as Shim et al.<sup>[16]</sup> provide overviews. Under indoor conditions, some additional techniques are necessary for sampling and analysis of air constituents and house dust.<sup>[7,17]</sup> Note that the house dust matrix is not clearly defined. Active sampling on filters is common, as is the examination of vacuum bag contents. In the latter case, the dust is usually sieved to a fraction <63 µm or <150 µm before analysis. Table 1 shows a compilation of the most important analytical techniques; a brief discussion is presented below. For detailed information the reader is referred to the literature.

Chromatography is the classic technique for determining organic compounds in the environment. Microplastic additives, especially substances with a volatility (e.g. vapor pressure) higher than that of docosane (C22) can be routinely determined using thermal desorption gas chromatography/mass spectrometry (TD-GC/MS).<sup>[18]</sup> For substances

with lower volatility, classic extraction methods with analysis via GC/MS or liquid chromatography (HPLC-MS/MS) are suitable. Peng et al.<sup>[19]</sup> developed a depolymerization technique for the environmental analysis of the polyamides PA 6 and PA 6.6 via LC-MS/MS. Tian et al.<sup>[20]</sup> proposed in-matrix depolymerization and LC-UV for detecting polyethylene terephthalate (PET). Wang et al.<sup>[21]</sup> used LC-MS/MS for the analysis of polycarbonate (PC) and PET microplastics.

Pyrolysis gas chromatography (PYR-GC/MS) is a technique that thermally decomposes large molecules in an inert atmosphere or in a vacuum. The masses of the fragments are identified and provide information about the corresponding polymer. With a combination of thermal desorption, pyrolysis and DART-MS (direct analysis in real time), Velimirovic et al.<sup>[9d]</sup> presented a method that allows rapid screening of environmental microplastics and their additives without chromatographic separation.

Fourier transform infrared (FTIR) spectroscopy (typically 4000–400 cm<sup>-1</sup>) is a popular and widely used technique for analyzing microplastics in environmental samples. Most molecules absorb light in the infrared region of the electromagnetic spectrum, and FTIR is very accurate in identifying the type of polymer by providing specific infrared spectra that contain distinct band patterns, also allowing differentiation between synthetic and natural materials. The surfaces of samples can be analyzed directly using attenuated total reflection (ATR). Combined with microscopy, investigations in the micrometer range are also possible. Near-infrared spectroscopy (NIR) also uses a specific region of the electromagnetic spectrum (typically 12500-4000 cm<sup>-1</sup>). The overtones and combination vibrations occurring in this range allow plastic materials to be specifically identified. NIR is less sensitive than FTIR, but the penetration depth into materials is higher.

Raman spectroscopy describes the inelastic scattering of light on molecules or solids. The sample is irradiated with monochromatic light; in the scattered light other frequencies are observed in addition to the initial frequency. From the spectrum, conclusions can be drawn about the substance being examined. As with FTIR, micrometer resolution can be achieved by combining the technique with a microscope. Especially Raman spectral imaging is often used to charac-



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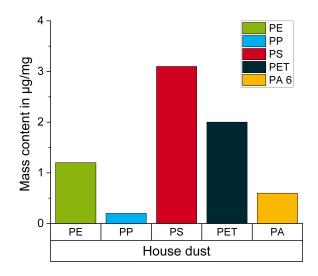
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terize microplastics.<sup>[22]</sup> Although IR and Raman provide similar spectra, the methods are supplementary or complementary.

Microscopic techniques are standard for characterizing the morphological and color properties of particles and fibers. Structures down to a size of 0.5 µm can be resolved with a light microscope. The use of fluorescence microscopes is also common. For this purpose, the sample is often stained with Nile Red dve to distinguish between fluorescent and nonfluorescent particles.<sup>[23]</sup> With a scanning electron microscope (SEM), the size resolution is in the lower nanometer range. In addition, this technique allows the coupling with energy-dispersive X-ray spectroscopy (EDX).<sup>[24]</sup> Atomic force microscopy (AFM) is able to achieve the nanoscale resolution often required for the analysis of polymers. However, this technique has disadvantages in terms of determining the chemical composition of materials. Therefore, AFM is often used in combination with other techniques such as IR.<sup>[25]</sup>

Thermal methods are also applied to characterize microplastics. Using thermogravimetry (TGA), differential thermal analysis (DTA), and differential scanning calorimetry (DSC), the phase transition temperature and degradation temperature of polymers can be determined against a reference compound.<sup>[27]</sup>

A new method combines TGA with TD-GC/MS. In a thermogravimetric analytical balance (TGA), the sample is heated to around 600 °C in the absence of oxygen. The decomposition gases are collected on a sorbent, then thermally desorbed (TD) and analyzed by GC/MS. The polymers are identified based on their characteristic decomposition products.<sup>[26]</sup> The method as described in an application note<sup>[28]</sup> is suitable for a large number of different matrices. Figure 1 shows the TGA-TD-GC/MS analysis of a house dust sample for components of microplastics conducted by the Bundesanstalt für Materialforschung und



**Figure 1.** Analysis of a house dust sample for microplastics via TGA-TD-GC/MS.<sup>[26]</sup> Picture by courtesy of Dr. Korinna Altmann, Bundesanstalt für Materialforschung und -prüfung (BAM), Berlin. See Table 2 for abbreviations of the polymers.

Angew. Chem. Int. Ed. 2022, 61, e202205713 (3 of 10)





Table 1: Analytical techniques (abbreviations given in the text) for the chemical and physical characterization of microplastics and microplastic additives.

Method	Remarks	
μ-FTIR	Standard method for the identification of polymers, reference spectra available.	
μ-ATR-FTIR	Surface examination of opaque materials.	
NIR	Low sensitivity but high penetration into material.	
µ-Raman	Identification of polymers, complements FTIR, analysis of samples down to 1 $\mu$ m.	
PYR-GC/MS	Identification of the thermal fragments of microplastics.	
DART-MS	Screening of microplastics and additives in combination with pyrolysis and thermal desorption.	
TD-GC/MS; HPLC-MS/MS	Identification of VOCs and SVOCs in air and dust.	
TGA; DSC	Phase transition, degradation.	
Light and Fluorescence Microscopy	Standard method for objects $>$ 0.5 $\mu$ m.	
SEM-EDX	High size resolution, often used in combination with X-ray elemental analysis.	
AFM	High resolution, often used in combination with IR.	

-prüfung (BAM). No further sample preparation is required for house dust, apart from sieving if the contents of vacuum cleaner bags are used.

### 3. Chemical Components of Microplastics

#### 3.1. Polymers

The spectrum of polymers, the properties, and the possible applications of plastics are far too diverse to be summarized here. Reference is therefore made to the monograph by Bonten<sup>[29]</sup> for classical plastics and to the monograph by Mather and Wardman<sup>[30]</sup> for textile fibers. Table 2 (see also for abbreviations) provides an overview of plastics that have been identified as components of microplastics in the air or in settled house dust indoors.

Wang et al.<sup>[21]</sup> detected PET and PC in house dust via the marker substances bisphenol A (BPA) and *p*-phthalic acid (PTA). Liu et al.<sup>[31]</sup> examined house dust samples from 39 Chinese cities. PET was detectable in high concentrations

**Table 2:** Components of microplastics identified in outdoor air, indoor air, and house dust (see text for details and references).

Symbol	Polymer	Remarks
ABS	Acrylonitrile butadiene styrene	thermoplastic
СР	Cellophane	made of cellulose
EVAC	Ethylene vinylacetate	thermoplastic elastomer
PA	Polyamide	thermoplastic
PAN	Polyacrylonitrile	degradation
PC	Polycarbonate	thermoplastic
PE	Polyethylene	thermoplastic
PET	Polyethylene terephthalate	thermoplastic
PLA	Polylactic acid	thermoplastic
PMMA	Polymethyl methacrylate	thermoplastic
PP	Polypropylene	thermoplastic
PS	Polystyrene	thermoplastic
PTFE	Polytetrafluoroethylene	thermoplastic
PUR	Polyurethane	duroplastic/elastomer
PVA	Polyvinyl acetate	thermoplastic
PVC	Polyvinyl chloride	thermoplastic
RY	Rayon (viscose)	made of cellulose
SBR	Styrene butadiene rubber	elastomer

in all samples, PC in 70% of the samples. The international study by Zhang et al.,<sup>[9a]</sup> in which 289 house dust samples from 12 countries were analyzed, also focused on PET and PC with the markers BPA and terephthalic acid (TPA). Zhang et al.<sup>[32]</sup> analyzed microplastics in Chinese house dust samples. PE (33-47%) and RY (44-60%) had the highest proportions, followed by significantly smaller proportions of polyacrylic, CP, PP, PA, and PS. Jenner et al.<sup>[9b]</sup> reported the chemical analysis of passively collected fiber fallout samples from 20 UK homes. On average, PET had the highest share with 63%; other common microplastics were PA, PAN, acrylic, PE, PMMA, PP, and co-polymers. For airborne microplastics, Liao et al.<sup>[33]</sup> found a more even distribution with the main components PA, PP, PE, PS, PVC, and miscellaneous polyesters. In an Australian house dust study, Soltani et al.<sup>[9c]</sup> found that with carpeting, PA, PE, PS, polyester, and polyacrylic were dominant. Conversely, in the absence of carpeting, PVC was the main component.

Other investigations come to analogous conclusions regarding the chemical composition of microplastics indoors.<sup>[34]</sup> In several house dust samples, microplastics were analyzed using EDX.<sup>[24,35]</sup> Again, the high proportions of carbon, oxygen, and nitrogen essentially confirm earlier results. However, the high amount of mercury (3–7%) in one Iranian study<sup>[35b]</sup> is surprising, but the authors do not discuss possible causes.

The publications mentioned so far essentially refer to living spaces. However, there are a number of areas assigned to the indoor environment that are intended for temporary stays, for example sporting activities. Here, artificial turf, which can be divided into "artificial grass" and "rubber mat",<sup>[10]</sup> is often used as the floor covering. Artificial grass mostly consists of organic polymers such as PE, PP, PA (nylon), PUR, or blends.<sup>[36]</sup> Recycling materials are used in the manufacture of rubber mats, in particular styrenebutadiene-rubber (SBR).<sup>[10]</sup> Mixtures of sand and recycled synthetic plastics (for example SBR) are common in horse riding arenas.<sup>[37]</sup> Due to the intensive mechanical stress on the ground in equestrian sport,<sup>[38]</sup> very high concentrations of airborne particles are often measured. The same applies to indoor motorsport events. There may also be emissions from brake and tire debris.<sup>[39]</sup>

Angew. Chem. Int. Ed. 2022, 61, e202205713 (4 of 10)

### 3.2. Additives

Most plastics contain additives to give the materials certain properties. In addition, residual monomers or their reaction products may sometimes be present. These substances are usually volatile and are released faster with increasing surface/volume ratio. It follows that the actual microplastic particles or fibers are often accompanied by organic compounds. A summary can be found in Table 3.

Best known are the plasticizers and flame retardants contained in many products. In the plasticizer sector, a shift from the classic phthalates to other substance groups such as adipates, terephthalates (DEHT, di-2-ethylhexl terephthalate), trimellitates (TOTM, tris-2-ethylhexyl trimellitate), or DINCH (1,2-cyclohexane dicarboxylic acid diisononyl ester) has been observed in recent years.<sup>[40]</sup> Nagorka et al.<sup>[41]</sup> compared the results of house dust analyses from 2003-2006 and 2014-2017, carried out as part of the German Environmental Survey (GerES). There was a clear decrease in DEHP (di-2-ethylhexyl phthalate) and a clear increase in DINP (diisononyl phthalate), DEHT, and DINCH. In the case of flame retardants, the trend is also moving away from so-called legacy substances towards emerging or novel substances.<sup>[13,42]</sup> House dust analyses are used for statistical classification based on reference values; biomonitoring is a valuable tool for determining the total body burden.<sup>[15]</sup>

A variety of additives are found in clothing materials as dyes, biocides, formaldehyde-releasing chemicals, for UV protection, etc. The list includes quinolines,<sup>[48]</sup> benzothiabenzotriazoles,<sup>[49]</sup> benzophenone derivatives,<sup>[50]</sup> zoles. formaldehyde,<sup>[51]</sup> and many other compounds.<sup>[47]</sup>

BPA is used in particular for the production of PC, the monomer is frequently found indoors. PET is made from terephthalic acid and ethylene glycol.

Styrene has long been known as an indoor pollutant. In the past, acrylic paints in particular, in which styrene was used as a reactive diluent, led to increased concentrations. In the field of plastics, polystyrene (PS) and various copolymers (ABS, SBR) are particularly well known. SBR is used indoors to prevent impact noise. In addition to the styrene monomer released, it is always the reaction products 4-phenylcyclohexene (4-PCH) and 4-vinylcyclohexane (4-

Table 3: (Micro)plastic monomers and additives detected in the indoor

Ref.

[12, 43]

[12, 43]

[43, 44]

[12, 43]

[40a]

[45]

[9a]

[9a]

VCH) formed by means of a  $_{\pi}4_{s} + _{\pi}2_{s}$  Diels-Alder reaction from styrene and cis-butadiene (4-PCH) or from cis- and trans-butadiene (4-VCH) that lead to problems in the indoor environment.<sup>[46]</sup> Styrene, other monomers such as caprolactam and lactide, as well as additives have recently been mentioned in the context of 3D printing.<sup>[12,43,44]</sup> This will be discussed in more detail in the next section.

#### 4. Indoor Sources of Microplastics

Particles and organic compounds are routinely monitored indoors today. It is therefore often difficult to distinguish which substances actually originate from microplastics and which come from other sources. Figure 2 gives a rough overview of possible microplastic sources as a basis for the subsequent discussion.

Outdoor environment-Various overviews on the occurrence of microplastics in the outdoor air are available.<sup>[5,10,52]</sup> Accordingly, the strongest sources are tire abrasion (here primarily SBR), waste disposal, road dust, macroplastic debris, sports fields, and construction work. Dris et al.<sup>[9e]</sup> characterized fibers based on their size. In addition to the size classification, Klein and Fischer<sup>[53]</sup> make a distinction between fragments and fibers according to their morphology. A comprehensive analysis of microplastics in terms of chemical analysis, color, shape, and size is provided by Xie et al.<sup>[54]</sup> Particles can enter the indoor environment via infiltration, tracking, and penetration. Infiltration refers to the direct exchange of air through open windows and doors and via mechanical ventilation systems without particle filters. Tracking describes the entry of particles, for example via footwear and clothing, and penetration is entry via small cracks in the building envelope. The respective efficiency of the mechanisms depends on the individual conditions.[55]

Abrasion is the mechanical wear of tissue components by rubbing against another surface. This leads to the release of fibers and particles (fragments), which usually accumulate in house dust. Today, many products are subject to certain minimum requirements in terms of abrasion resistance,<sup>[56]</sup> but only a limited statement can be made about their properties under real conditions. In addition, the stand-

Figure 2. Overview of possible sources of microplastics in the indoor environment.

PS Styrene PVC Phthalates, DINCH, TOTM, adipates, terephthalates, etc.

Styrene, 1,3-butadiene, 4-VCH, 4-PCH SBR [46] Fabrics Quinoline, bisphenols, benzothiazoles, [47] (clothing) benzotriazoles, formaldehyde, organophosphates

Angew. Chem. Int. Ed. 2022, 61, e202205713 (5 of 10)

environment or in test chamber emission studies. Additive/Monomer

Styrene

Lactide

Acetic acid

Caprolactam

**Bisphenol A** 

Terephthalic acid

Polymer

PA (PA 6)

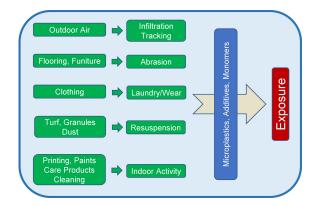
ABS

PC

PET

PLA

**PVA** 





ardized test methods essentially refer to the material surface and not to the release of fibers. However, it can be assumed that a large part of the microplastics identified in house dust is caused by mechanical stress on surfaces. This is especially true for floorings and furniture. The particles contained in the dust can be mobilized by resuspension. Thatcher and Layton<sup>[57]</sup> found the highest resuspension rates for particle sizes of 5–25  $\mu$ m.

**Clothing/laundry**—It is known that washing of textiles can heavily pollute the waste water with microplastics.<sup>[58]</sup> However, there are only a few investigations that deal with the effects of the washing process on air quality. O'Brien et al.<sup>[59]</sup> have studied mechanical drying processes and conclude that it is an emission source of microplastic fibers into ambient air. De Falco et al.<sup>[60]</sup> showed that the direct release of microfibers from garments to air as a consequence of daily wear is of equal importance to the release to wastewater by laundering.

**3D** printing is a relatively new source of indoor plastics, monomers, and additives that is currently gaining increasing attention. In the private sector, the Fused Filament Fabrication (FFF) technique, in which thermoplastic polymers are processed, is particularly common. ABS is the classic polymer used in FFF with the main emissions product being styrene. PLA is considered a sustainable alternative, whereby essentially lactide, the cyclic diester of lactic acid, is released. Other common thermoplastics for FFF are PA 6 (caprolactam), high-impact PS (styrene), and PVA (acetic acid). Overall, the number of organic compounds emitted during 3D printing is high, but only a few substances can be directly assigned to the polymers or their additives.<sup>[12,43,61]</sup> However, this also includes many new compounds, such as stabilizers, which were previously unknown as indoor air components.

Particulate emissions always occur with 3D printing. The typical size range is 10-200 nm, so the particles cannot be directly assigned to microplastics. In investigations with various polymers, Gu et al.<sup>[12]</sup> found that the particles evaporate when the temperature increases and have completely disappeared at 250-300 °C. The volatilization process begins around the glass transition temperature of the respective thermoplastic. Ding et al.[62] come to analogous results using thermogravimetric analyses. Potter et al.<sup>[44]</sup> pointed out that carbon nanotubes (CNTs) are added to 3D printer filaments to increase their conductivity. Bossa et al.<sup>[63]</sup> studied polymers for 3D printing containing MWCNTs (multiwall carbon nanotubes) and found that microplastics are released through abrasion. Overall, the chemical composition of particles produced by 3D printing is very heterogeneous and influenced by the filament, the chemicals in the printer itself, and its operating conditions. Therefore, the chemical fingerprint of such particles cannot be assigned to a specific polymer.<sup>[64]</sup>

**Sport arenas**—There are two important sources of microplastics here: artificial turf and sand/fiber mixtures. Modern artificial turf consists of a bonded substrate (base layer), a synthetic elastic layer (usually PP), fibers (PE, PP, PA), and granules as fillers (SBR, PUR, sometimes in combination with sand).

In equestrian arenas or in all arenas with a sand base, aggregates are often added in order to improve the durability of the floor. These are, for example, shredded carpets, non-woven fabrics, clothing, and car tire residues. They still contain the additives for the original use. It is an additional problem that they are further crushed by the intense mechanical stress. In recent times, it has also been customary to add biobased fibers and wood chips to sand in equestrian arenas.

Miscellaneous indoor sources-There are a variety of indoor products that intentionally or non-intentionally release microplastics. Dispersion paints essentially consist of pigments, a binder, usually PVA or acrylate, and additives. Microplastics are released particularly in the do-it-yourself (DIY) sector during application or when cleaning the devices. A strong source is spray paints and plastics for spray application in general. Microplastic substances (so-called beads) in cosmetics, personal care products,<sup>[4b]</sup> detergents, and cleaning agents are a much-discussed topic. Abrasion from shoes, the soles of which are mostly made of PUR, is also not negligible. The sources of fluorinated microplastics, especially PTFE, have declined significantly, but they are still used in non-stick coatings, water-repellent clothing, and cosmetics. Finally, it should be mentioned that people are also exposed to microplastics through drinking water and food.<sup>[65]</sup> However, that is not subject of this Minireview.

#### 5. Indoor Exposure to Microplastics

Based on the fact that terrestrial microplastics have moved into the focus of scientific interest for only a few years, a surprisingly large number of review articles have already been published, which underlines the great interest in this topic.<sup>[5,9d,14,34ac,52,65,66]</sup>

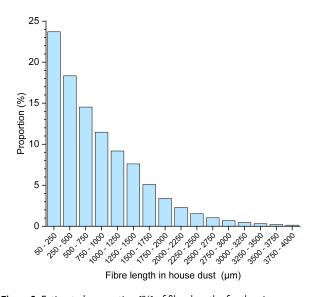
Table 4 summarizes published data on the occurrence of microplastics indoors. These are divided into house dust analyses, air analyses, fallout and emission studies. Since the analysis of microplastics is not standardized, the designations of the respective original publications have been retained. The data refer only to the components of microplastics, in particular fibers and fragments. These are large compared to airborne particles found indoors. Figure 3 shows the distribution of fiber lengths in house dust in the size range between 50  $\mu$ m and 4000  $\mu$ m; the data were taken from several publications (see figure caption). The percentage decrease from the longer to the shorter fibers is approximately exponential. Both Soltani et al.<sup>[9c]</sup> and Dris et al.<sup>[9e]</sup> used light microscopes with a lower observation limit of 50 µm. Nevertheless, Dris et al.<sup>[9e]</sup> found a significantly higher proportion of fibers in the 50-250 µm range.

The measurement of airborne particles usually refers to equivalent diameters  $< 10 \ \mu\text{m}$ . The reason is that the PM<sub>10</sub> fraction is considered respirable and the PM<sub>2.5</sub> fraction is alveolar (the indices refer to  $10 \ \mu\text{m}$  and  $2.5 \ \mu\text{m}$ , respectively).<sup>[7]</sup> Larger particles are therefore less important for inhalation and more important for ingestion (see Figure 3). Microplastics are part of the organic fraction of total suspended particles (TSP) in the air and are therefore

Table 4: Results of studies on the occurrence of microplastics indoors (units as provided in the original references).

Type of investigation	Location	Concentration/fallout/emission	Ref.
Indoor house dust	France	190–670 fibers mg <sup>-1</sup>	[9e]
	China	18–43 mg kg <sup>-1</sup> PA 6; 54–321 mg kg <sup>-1</sup> PA 6.6	[19]
	China	1550–120000 mg kg $^{-1}$ PET; $<$ 1– $\approx$ 100 mg kg $^{-1}$ PC	[31]
	China	$62-3861 \text{ MPs g}^{-1}$	[69]
	12 countries <sup>[a]</sup>	38–120 000 mg kg <sup>-1</sup> PET; < 0.11–1700 mg kg <sup>-1</sup> PC	[9a]
	Netherlands	$< 0.31 - 305 \text{ mg g}^{-1} \text{ PET}$	[20]
	Iran	81–55 830 MP g <sup>-1</sup>	[35a]
	Iran	10–635 MPs g <sup>-1</sup>	[35b]
	Iran	48.6–139 items $mg^{-1}$	[76]
	Pakistan	29–636 fibers m <sup>-2</sup>	[77]
Indoor air	France	1–60 fibers $m^{-3}$	[9e]
	USA	3-17 fibers m <sup>-3</sup>	[23]
	Portugal	6 fibers m <sup>-3</sup>	[73]
	China	$1583 \pm 1180 \text{ # m}^{-3}$	[33]
	China	16–93 N m <sup>-3</sup>	[54]
	Taiwan	$46\pm55~\mathrm{MPs}\mathrm{m}^{-3}$	[78]
	Denmark	$1.7 - 16.2 N_{MP} m^{-3}$	[68]
	South Korea	$0.49-6.64 \text{ MPs m}^{-3}$	[79]
Indoor fallout	France	1586–11 130 fibers m <sup>-2</sup> day <sup>-1</sup>	[9e]
	UK	1414 $\pm$ 1022 microplastics m $^{-2}$ day $^{-1}$ (mean for Jul.–Dec.)	[9b]
	Australia	22–6169 fibers $m^{-2} day^{-1}$	[9c]
	Brazil	$309 \pm 215 \text{ MPs m}^{-2} \text{day}^{-1}$	[34b]
	USA	$(6.20 \pm 0.57) \times 10^{3} - (1.96 \pm 1.09) \cdot 10^{4}$ fibers m <sup>-2</sup> day <sup>-1</sup>	[24]
	China	$(7.6 \pm 3.9) \times 10^5 \text{ MPs m}^{-2} \text{day}^{-1}$	[71]
Emission study	n.a.	Particle/VOC/SVOC emissions from 3D printers (ABS, PET, PC, PS, other)	[12]
	n.a.	Particle/VOC/SVOC emissions from 3D printers (ABS, PA, PLA, PS)	[43]
	n.a.	0.3 g m <sup>-2</sup> s <sup>-1</sup> abrasion rate for plastics used in 3D printing	[63]
	n.a.	1–403 fibers $g^{-1}$ fabric released to air	[60]
	n.a.	58 fibers/660 g blanket sample	[59]

[a] China, Colombia, Greece, India, Japan, Kuwait, Pakistan, Romania, Saudi Arabia, South Korea, USA, Vietnam.



**Figure 3.** Estimated proportion [%] of fiber lengths for the size range between 50 µm and 4000 µm in settled indoor house dust. The distribution is primarily based on data by Soltani et al.;<sup>[9c]</sup> data from other references were also considered.<sup>[7,9e,67]</sup>

measured indirectly. Vianello et al.<sup>[68]</sup> point out that in their measurements the average proportion of synthetic fragments and fibers was 4 % of the total identified particles. Due to the sophisticated sampling and analysis, the portion of microplastics is usually not identified, but many additives in microplastics have been routinely recorded using house dust analysis and biomonitoring.<sup>[15]</sup> In one house dust sample, the sum of five identified types of microplastics was around 10  $\mu$ gmg<sup>-1</sup> (1%) (see Figure 1), but this result cannot be regarded as representative. Zhu et al.<sup>[69]</sup> studied different indoor environments and found that residential apartments had the highest abundance of MPs in indoor dust samples.

Table 4 shows that the concentrations and deposition rates cover a wide range, making it difficult to predict exposure. This leaves a lot of room for interpretation. Various attempts have been made to calculate intake rates. Catarino et al.<sup>[70]</sup> estimate fiber ingestion from fallout to be between 13731 and 68415 particles/year per person. Fang et al.<sup>[71]</sup> come to considerably higher values with  $1.9 \times 10^5$ –  $1.3 \times 10^6$  MPs/year and person. Liu et al.<sup>[31]</sup> state that indoor dust is a non-negligible source of human exposure to MPs and assume a geomean daily intake of 17300 ng kg<sup>-1</sup>-bw of PET MPs in children. Zhang et al.<sup>[9a]</sup> calculated a median daily intake of PET-based MPs for infants in the range of 4000-150000 ng kg<sup>-1</sup>-bw from house dust. For children

Angew. Chem. Int. Ed. 2022, 61, e202205713 (7 of 10)

younger than 5 years, Soltani et al.<sup>[9c]</sup> calculate the mean annual inhaled fiber (50–250  $\mu m$ ) intake as  $12\,891\,+/$ –4472 fibers kg^-1-bw. The calculated median intake via ingestion is 6.1 mg kg^-1-bw/year for this age group.

Nematollahi et al.<sup>[35b]</sup> also found that children are more exposed to microplastics than adults. Furthermore, the authors point out that microplastic particles can also adsorb other contaminants such as metals and organic substances. Jenner et al.<sup>[9b]</sup> come to the general conclusion that humans are exposed to significantly higher concentrations of MPs within homes compared with than from the outdoor environment. This is in accordance with the results of Liao et al.<sup>[33]</sup> Ageel et al.<sup>[34a]</sup> also consider indoor exposure as important and state that the dermal route cannot be neglected. Torres-Agullo et al.<sup>[66a]</sup> make a connection between exposure to microplastics and COVID-19. The use of masks not only leads to more waste, but wearing them increases the inhalation of fibers.

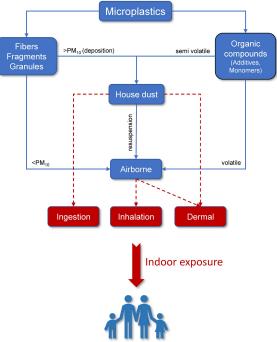
Some authors address the widely varying conditions and uncertainties that make health risk assessment difficult. Zhang et al.<sup>[32]</sup> describe the different fallout of microplastics depending on sampling location and season. Cox et al.<sup>[72]</sup> modelled the total intake of microplastic particles (indoor and outdoor) for adolescents and adults and discussed the high margins of error. As an example, the estimated intake via inhalation for adult males is  $61928 \pm 68865$  particles/year and person. Xie et al.<sup>[54]</sup> arrive at significantly higher values with improved models.

There are also only a few emission studies that relate directly to the release of synthetic materials and the possible contamination of the interior. Essentially, these are investigations into textiles and 3D printers (see Table 4).

Figure 4 shows the complex relationships that must be taken into account when estimating indoor exposure to microplastics. Some studies have attempted to carry out a risk assessment on this basis and under consideration of the available data.<sup>[5,14,34c,52,66b,c,73]</sup> However, this has not yet been achieved convincingly. Nevertheless, the following statements are unanimously made in the cited works: 1) indoor microplastics are an emerging concern; 2) the data situation is insufficient; 3) the proportion of microplastics in particles must be determined; 4) there is an urgent need for emerging data on human health impacts ;and 5) the overall situation is still unclear.

The guideline values currently available for indoor pollutants are only of limited use with regard to the evaluation of microplastics. The values of the World Health Organization (WHO) for airborne particles refer to  $PM_{10}$  and  $PM_{2.5}$ ;<sup>[74]</sup> guideline values for indoor air were derived for substances such as styrene, benzothiazole, and methyl methacrylate (see German Committee on Indoor Guide Values (AIR); https://www.umweltbundeamt.de) and some data (reference values) are available for substances accumulating in house dust.<sup>[13,41,75]</sup>

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*Figure 4.* Indoor exposure to microplastics and its components via air and house dust.

#### 6. Future Perspectives

The available literature on the occurrence of microplastics in the atmosphere is still limited compared to the studies on soil and aquatic systems,<sup>[5]</sup> but is now increasing significantly. This is due to a generally growing interest in the topic of microplastics and the attempt to estimate the total human exposure to microplastics and its components.

However, one has to realize that indoor pollution with microplastics is by no means a new topic, but has existed since plastics have been used in building materials and household products. One can even speculate that the exposure to microplastics and its components in the indoor environment is already decreasing. There are now alternatives for many of the classic plastics and fabrics; critical additives such as endocrine-disrupting plasticizers and flame retardants are being substituted. Potential problems such as artificial turf and sand/fiber mixes have at least been identified. On the other hand, there are new sources such as 3D printing, whose impact on indoor air quality definitely needs attention. With the help of house dust analysis and biomonitoring, trends can be identified with regard to decreasing or increasing exposure to target compounds.<sup>[15]</sup>

To date, it is not scientifically clear whether microplastics pose a serious threat to human health at all.<sup>[14]</sup> The analytical detection of fibers and fragments does not mean that they are also ingested/inhaled nor that they are bioavailable. Moreover, "intake" is in no way equal to "uptake".<sup>[15]</sup> However, when looking at the results shown in Table 4 and also taking into account related monomers and additives, one can certainly see a potential problem. It is therefore advisable to investigate the health effects of exposure to microplastics in a timely manner and in detail. In parallel, strategies must be developed to reduce the formation of indoor microplastics and the application of microplastics beads. In many household and personal care products, their use is not technically necessary.

From an analytical point of view, there is the problem that microplastics are not captured at all by the methods commonly used indoors. The detectable upper size range of particle counting devices ends at 10  $\mu$ m. The determination of larger particles and fibers in air requires sophisticated sampling techniques. The analysis of sedimented microplastics and house dust is significantly easier. Subsequently, the morphology of the fibers and fragments has to be characterized microscopically and the synthetic materials have to be distinguished from other particles, e.g. mineral fibers, using spectroscopic techniques such as FTIR and Raman. In the other direction, the problem also exists. The chemical spectrum of particles with a size  $<1 \,\mu$ m and nanoplastics in general can hardly be characterized.

Within the framework of environmental surveys, there are now intelligent strategies available for monitoring the exposure of the general population to pollutants in the indoor and outdoor environment. Today, these no longer only refer to long-known substances, but also have a preventive character. When looking at the wide use of plastic materials indoors and the fact that people spend a major part of their time in closed spaces for living, working, and leisure, it is recommended to include microplastics and their additives in such surveys in the future. If this is realized using easily accessible samples such as house dust and fallout combined with reliable analytical techniques such as TGA-TD-GC/MS, the benefit should more than justify the effort.

## Acknowledgements

I am very grateful to Dr. Korinna Altmann, Bundesanstalt für Materialforschung und -prüfung (BAM), for valuable discussions regarding the TGA-TD-GC/MS method and for providing Figure 1. I also would like to thank Manuela Lingnau, WKI, for the final design of the frontispiece. Open Access funding enabled and organized by Projekt DEAL.

### **Conflict of Interest**

The author declares no conflict of interest.

**Keywords:** 3D printing • artificial turf • fiber fallout • house dust • micro/nanoplastics

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Manuscript received: April 19, 2022 Accepted manuscript online: June 7, 2022 Version of record online: July 8, 2022

Angew. Chem. Int. Ed. 2022, 61, e202205713 (10 of 10)

