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Citation for final published version:

Liu, Pengju, Shao, Longyi, Li, Yaowei, Jones, Tim , Cao, Yaxin, Yang, Cheng-Xue, Zhang, Mengyuan, Santosh, M, Feng, Ziaolei and Berube, Kelly 2022. Microplastic atmospheric dustfall pollution in urban environment: evidence from the types, distribution, and probable sources in Beijing, China. *Science of the Total Environment* 838 (1) , 155989. 10.1016/j.scitotenv.2022.155989

Publishers page: <https://doi.org/10.1016/j.scitotenv.2022.155989>

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1 **Microplastic atmospheric dustfall pollution in urban environment: Evidence from**
2 **the types, distribution, and probable sources in Beijing, China**

3

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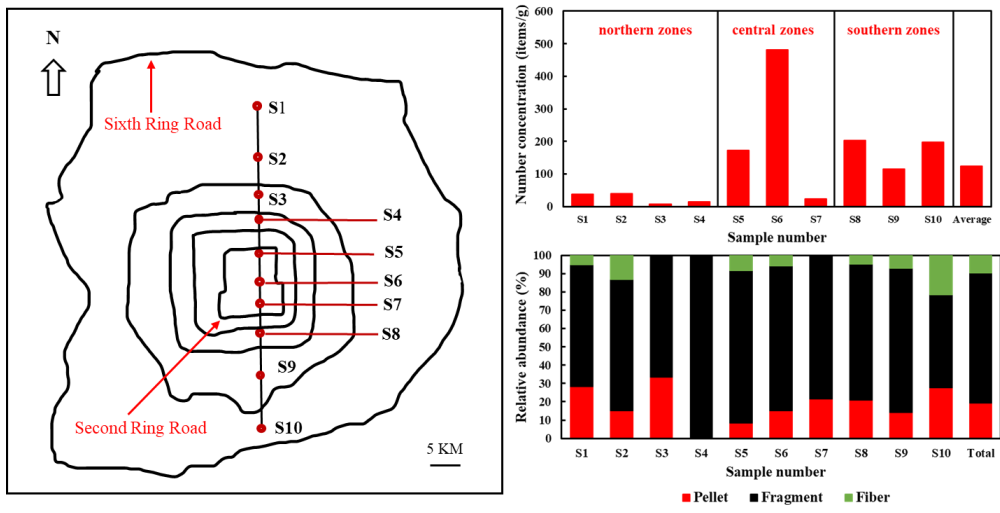
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20

21 Graphical Abstract



22

23

24 Highlights:

25

26 1. Microplastic pollution in the central Beijing is more serious than in the northern and southern
27 zones.

28 2. Nine different compositions of microplastics were identified with polypropylene being the most
29 abundant.

30 3. The morphologies of microplastics include fragments, pellets, and fibers; with fragments being
31 the most common.

32 4. The presence of aged microplastics was recorded in the dustfall samples.

33

34 **Abstract**

35 Airborne microplastics (MPs) pollution is an environmental problem of increasing concern, due
36 to the ubiquity, persistence and potential toxicity of plastics in the atmosphere. In recent years, most
37 studies on MPs have focused on aquatic and sedimentary environments, but little research has been
38 done on MPs in the urban atmosphere. In this study, a total of ten dustfall samples were collected in
39 a transect from north to south across urban Beijing. The compositions, morphologies, and sizes of the
40 MPs in these dustfall samples were determined by means of Laser Direct Infrared (LDIR) imaging
41 and Field Emission Scanning Electron Microscopy (FESEM). The number concentrations of MPs in
42 the Beijing dustfall samples show an average of 123.6 items/g. The MPs concentrations show
43 different patterns in the central, southern, and northern zones of Beijing. The number concentration
44 of MPs was the highest in the central zone (224.76 items/g), as compared with the southern zone
45 (170.55 items/g), and the northern zone (24.42 items/g). The LDIR analysis revealed nine
46 compositional types of MPs, including Polypropylene (PP), Polyamide (PA), Polystyrene (PS),
47 Polyethylene (PE), Polyethylene Terephthalate (PET), Silicone, Polycarbonate (PC), Polyurethane
48 (PU) and Polyvinylchloride (PVC), among which PP was overall dominant. The PP dominates the
49 MPs in the central zone (76.3%), and the PA dominates the MPs in the southern zone (55.86%), while
50 the northern zone had a diverse combination of MPs types. The morphological types of the individual
51 MPs particle include fragments, pellets, and fibers, among which fragments are dominant (70.9%).
52 FESEM images show the presence of aged MPs in the Beijing atmosphere, which could pose a yet
53 unquantified health risk to Beijing's residents. The average size of the MPs in the Beijing samples is
54 66.62 μm . Our study revealed that the numbers of fibrous MPs increase with the decrease in size.
55 This pollution therefore needs to be carefully monitored, and methods of decreasing the sources and
56 mitigations developed.

57 Keywords: Microplastics, LDIR, FESEM, chemical composition, morphology, health risk

58

59 **1. Introduction**

60 Microplastics (MPs), as an entirely anthropogenic type of pollution, are considered to be
61 stratigraphic markers of the Anthropocene Epoch (Corcoran et al., 2018). MPs are plastics with small
62 particle sizes, usually less than 5 mm (Arthur et al., 2009, Shao et al., 2022a), that originate from both
63 primary and secondary sources (Cole et al., 2011, Shao et al., 2022a). Primary MPs are mainly
64 sourced from common commercial products that contain microscopic plastics as part of their
65 manufacture; such as personal care, cosmetics, cleaning, and medical products (Wang et al., 2019).
66 Secondary MPs originate from the environmental degradation of larger-sized plastic products
67 (Akhbarizadeh et al., 2017). Plastic is widely used in numerous fields, including packaging,
68 construction, automotive, textile, medical, electronic, agriculture, sports, and safety equipment
69 (Andrady, 2011; Brahney et al., 2020; Gallagher et al., 2016; Mohammadizadeh et al., 2019). Plastic
70 has advantages that include low price, lightweight, strength, practicality, and durability (Moore, 2008).
71 Since the 1950s, approximately 8300 million metric tons of plastics have been manufactured
72 worldwide (Geyer et al., 2017). By 2025, the accumulation of plastic in the environment could reach
73 11 billion tons (Brahney et al., 2020). As the demand for plastics continues to grow, the rate of
74 accumulation of MPs in the environment has increased dramatically (Serranti et al., 2018). MPs
75 pollution is rapidly becoming a pressing global issue, which has attracted commercial, environmental
76 and public concern.

77 The accumulation of MPs in the environment can potentially exacerbate ecosystems and increase
78 health risks (Kvale et al., 2021). Although water treatment plants can reduce the concentration of MPs

79 in wastewater by up to 98%, large volume of MPs are still discharged into the receiving waters every
80 day (Murphy et al., 2016). Large amounts of MPs can be ingested by marine organisms with non-
81 selective filter-feeding behavior (Wang et al., 2021). In the scientific literature, MPs have been
82 detected in fish (Ding et al., 2018), shellfish (Ding et al., 2020), bivalves (Van Cauwenberghe and
83 Janssen, 2014), and earthworms (Jiang et al., 2020). Studies have confirmed that MPs can affect the
84 feeding, multiple molting, reproduction, growth, mortality, immune responses, and oxidative stress
85 of marine organisms (Bergami et al., 2016; Devriese et al., 2015; Jeong and Choi, 2019; Limonta et
86 al., 2019; Qiao et al., 2019, Ward and Kach, 2009; Zhang et al., 2021b). MPs are by definition very
87 small and therefore have a relatively large specific surface area; especially after aging and crushing
88 (Mao et al., 2020). A large specific surface area and hydrophobic characteristics make MPs more
89 susceptible to adsorption of toxic and hazardous substances, such as polycyclic aromatic
90 hydrocarbons (PAHs) (Klasios et al., 2021), organochlorine pesticides (OCPs) (Zhang et al.,
91 2021a), polychlorinated biphenyls (PCBs) (Pastorino et al., 2021) and heavy metals (e.g., Cd, Pb, Cr,
92 Cu, Zn) (Guo and Wang, 2021). Aged MPs can adsorb toxic and harmful substances, thus posing a
93 potential threat to the human body. The possible dangers posed by MPs on ecosystems and human
94 health needs to be better understood.

95 Recently, most research on MPs has focused on different aquatic environments such as rivers,
96 groundwater, lakes, and seawater (Bharath et al., 2021; Clayer et al., 2021; Kooi et al., 2021;
97 Woodward et al., 2021), and sedimentary environments such as island sediments, terrestrial, river,
98 and marine sediments (Braun et al., 2021; Saarni et al., 2021; Vermeiren et al., 2021; Yan et al., 2021;
99 Zhou et al., 2021). In addition, there are also studies on long-distance MPs transport. The studies
100 found that MPs can be transported to remote areas mostly unaffected by human influence, such as the
101 Tibetan Plateau (Liang et al., 2022), Arctic (Hamilton et al., 2021), Himalayas (Yang et al., 2021) and

102 western Italian Alps (Parolini et al., 2021).

103 In spite of the increasing studies on MPs, there is still a paucity of research on atmospheric MPs,
104 especially in megacities. MPs are recognized as widespread atmospheric pollutants due to their small
105 sizes and low densities (Revell et al., 2021). The distributions and characteristics of MPs in cities and
106 their influencing factors are still unclear. Due to limitations of available analytical techniques, there
107 is little information about the variations of concentrations, particle sizes and morphologies of MPs in
108 the atmosphere.

109 In this study, MPs in ten atmospheric dustfall samples were studied to elucidate the pollution
110 role of MPs in Beijing atmosphere. The morphological characteristics and compositional types of
111 MPs, and the regional distribution characteristics of MPs within Beijing were investigated. The
112 variations in the number concentrations, particle sizes, and morphologies of MPs within the
113 atmosphere are considered. The results of this study provide new insights into particulate pollution
114 compositions in the urban atmosphere of megacities.

115 **2. Materials and methods**

116 **2.1. Sample collection**

117 Samples were collected in urban areas of Beijing, China (Fig. 1). To understand the distribution
118 of MPs in the atmosphere, ten sampling sites were selected and the samples were collected at 2-7 km
119 intervals in a transect from the northern to the southern areas of the city. The atmospheric dustfall
120 was mostly collected on a smooth surface (non-plastic component). To minimize contamination, the
121 atmospheric dustfall was collected with an antistatic brush and dustpan, using a brush type that
122 minimizes potential brush fiber contamination. The bulk samples were stored in a sealed aluminum

123 foil bag. The collection data detailing the local environment of the sampling sites, and wind direction
124 is shown in Table 1.

125 Separated by the Second Ring Road, the sampling sites are divided into three zones (Fig 1). The
126 northern zone refers to the northern area outside of the northern Second Ring Road, including S1, S2,
127 S3 and S4. The central zone refers the area within the Second Ring Road, including S5, S6, and S7.
128 The southern zone refers to the southern area outside of the southern Second Ring Road, including
129 S8, S9, and S10.

130 **2.2. MPs separation**

131 In this study, ZnCl₂ solution was used as heavy-liquid to separate MPs by density flotation from
132 the bulk samples, which predominantly consisted of denser mineral particles. Previous research has
133 shown that this method is effective (Bellasi et al., 2021; Liu et al., 2019b; Shao et al., 2022a). The
134 steps are: (1) configure 1.7-1.8 g·cm⁻³ ZnCl₂ (premium pure) solution; (2) place a measured amount
135 of atmospheric fallout bulk dust into a 100 mL beaker, add 60 mL ZnCl₂ solution, stir the mixture for
136 two minutes, and then allow to stand over 72 hours; (3) transfer the surface floating component to
137 another beaker and add 60 mL of 30% H₂O₂ to digest the organic matter, which includes agitating it
138 on an oscillator for 10 min, and then standing for 24 hours to allow the H₂O₂ to fully digest the organic
139 matter; (4) MPs are collected by vacuum extraction filtration using a filter membrane (silver
140 membrane with a pore size of 0.45 μm), and then placed in a sterile petri dish for air-drying (5) place
141 the dried filter in ethanol and extract the sample off the filter into the solution, aided by ultrasound;
142 (6) remove the filter membrane from the ethanol and wash with ethanol several times until the filter
143 is clean, The ethanol was allowed to evaporate down to a volume of 200 μL, then a drop of the ethanol
144 is put on a glass coverslip. Once the ethanol has completely evaporated leaving the sample adhered

145 to the glass surface the samples are prepared for FESEM and LDIR analysis.

146 **2.3. FESEM analysis**

147 The projected image of the plastic particles provided by the LDIR imaging system is not clear,
148 and the size is larger than 20 μm . Therefore, FESEM was used to observe the microscopic
149 characteristics of the MPs (Li et al., 2020b). Studies have shown that FESEM is a very effective
150 method for the characterization of atmospheric particles (Shao et al., 2022a, Shao et al., 2022b). The
151 FESEM used in this study was a SUPRA 40 (Zeiss Germany) based at Henan Normal University. The
152 prepared sample on the glass coverslip was placed on the stub using conductive double-sided tape
153 and gold coated. The FESEM analysis was under 20 KV voltage and the working distance was less
154 than 5 mm.

155 **2.4. LDIR analysis**

156 The LDIR (Agilent 8700) analyzer was used to characterize the types and sizes of MPs. The
157 LDIR uses a Quantum Cascade Laser (QCL) as the light source, which has over 10,000 times the
158 energy of traditional Fourier Transform infrared (FT-IR) spectroscopy. The collimating laser
159 accurately aligns the light rays and directly irradiates the sample after optical path conversion (Li et
160 al., 2021). Even for micron-scale samples the infrared spectrum has a sufficient signal-to-noise ratio
161 to achieve accurate chemical characterization. Previous studies have confirmed that the LDIR
162 Analyzer is an advanced and reliable method for detecting plastics (Li et al., 2021; Ng et al., 2021).
163 In this study, fast single-wavelength (1800 cm^{-1}) light can scan the MPs on the slide, and the image
164 analysis software can measure the MPs sizes $>20\ \mu\text{m}$. Once the MPs were located, the LDIR
165 automatically moved around to scan particles on the slide, and the infrared median range spectrum of

166 each particle was collected, and compared with the standards in a plastics reference library (Li et al.,
167 2021). To ensure the reliability of the identification, only the results with a matching degree greater
168 than or equal to 0.8 were selected.

169 **3. Results**

170 **3.1. MPs number concentrations**

171 MPs were detected in all the samples. The number concentration in this study refers to the
172 number of MPs particles (items) per gram of dustfall. The abundance of MPs at all the sample sites
173 ranged from 7.25 items/g to 481.39 items/g, with an average of 123.6 items/g. The highest number
174 concentration of MPs was found at the sampling site S6, at 481.39 items/g, followed by S8 (202.29
175 items/g), S10 (197.03 items/g), S5 (172.73 items/g), S9 (115.19 items/g), S2 (38.85 items/g), S1
176 (37.62 items/g), S7 (22.95 items/g), S4 (13.64 items/g), and S3 (7.25 items/g) (Fig. 2).

177 The MPs distribution in the northern zone (S1, S2, S3, and S4), central zone (S5, S6, and S7),
178 and southern zone (S8, S9, and S10) show different patterns. In the northern zone, the average number
179 concentration was 24.42 items/g, in the central zone, the average number concentration was 224.76
180 items/g, and in the southern zone, the average number concentration was 170.55 items/g. The central
181 zone has the highest average concentration of MPs, 1.3 times that of the southern zone and 9.2 times
182 that of the northern zone. In the central zone, S5 was collected in Nanluoguxiang (South Luogu Lane)
183 with street food stalls, a developed fast-food service and a high population density. The S6 sample
184 site is in the center of the city, close to the world cultural heritage site the Forbidden City and shopping
185 centers. There are also many service sectors such as catering, hotels, and shops around the S6
186 sampling site. The number concentration of MPs at the S7 sampling site was only 22.95 items/g, and

187 this low concentration may be due to that the sampling site is not in a residential area and has not
188 been affected by road traffic (There are tall buildings between the sampling site and the road). The
189 average number concentration of MPs in the southern zone was nearly seven times higher than in the
190 northern zone. Sampling site S9 with the lowest number concentration (115.19 items/g) in the
191 southern zone was higher than that at the S2 with the highest number concentration (38.85 items/g)
192 in the northern zone (Fig. 2). In the northern zone, there are no other sources of MP pollution other
193 than MPs created by the residents. Construction, traffic levels, and industries in the southern zone
194 may be an important reason for the higher MPs number concentration in the southern zone compared
195 to the northern zone. It appears that areas with high human activity levels will produce relatively
196 higher number concentrations of MPs.

197 **3.2. MPs chemical types**

198 In our study, nine chemical types were recognized by LDIR, including Polypropylene (PP),
199 Polyamide (PA), Polystyrene (PS), Polyethylene (PE), Polyethylene Terephthalate (PET), Silicone,
200 Polycarbonate (PC), Polyurethane (PU) and Polyvinylchloride (PVC) (Fig. 3). The total samples
201 statistics reveal the relative abundances of different types of MPs. The PP and PA were the major
202 types, accounting for 56 items/g and 28.83 items/g respectively, followed by PE (8.82 items/g), PVC
203 (8.48 items/g), PS (7.46 items/g), PET (5.67 items/g), PU (4.81 items/g), Silicone (3.05 items/g), and
204 PC (0.48 items/g) (Fig. 4).

205 Our study found that the central, southern, and northern zones have different patterns in the
206 relative proportions of the different compositional types of MPs. In the central zone, PP was the
207 dominant component, accounting for 76.3%. In the southern zone, PA was the main component,
208 accounting for 58.86%, followed by PS (15.08%) and PE (7.36%). In the northern zone, PP was again

209 the dominant component at 32.4%, followed by PA (18.1%), PE (17.05%), PET (11.92%), and
210 Silicone (9.42%) (Fig. 5). Therefore, PP was the major contributor to the MPs pollution in the central
211 and northern zones, whereas PA was the main component of MPs pollution in the southern zone. In
212 the northern zone, MPs pollution consisted of diverse types of MPs.

213 **3.3. MPs morphological types**

214 Three morphological types of MPs were observed in this study: pellets/spheres (Fig. 6a),
215 fragments (Fig. 6b), and fibers (Fig. 6c). Pellet/Sphere refers to the morphology of individual MPs
216 with a rounded morphology. Fragment describes the morphology of individual MPs that are neither
217 rounded or fibers. Fiber describes the morphology of individual MPs that have a length: width aspect
218 ratio greater than 3. The fragments were the most common in all the sample sites. The number
219 statistics for all samples were fragments (70.9%), followed by pellets/spheres (19.53%) and fibers
220 (9.57%) (Fig. 7).

221 The study also identified that different chemical compositional types of MPs display different
222 morphologies. PS had fragment and fiber morphologies, but no pellet/spheres, whereas PC was
223 mostly pellets/spheres (Fig. 8). There was no apparent difference in the relative proportions of the
224 different MPs morphological types in the central, southern, and northern zones of Beijing.

225 **3.4. MPs size distributions**

226 The size of the individual MPs particle in the Beijing samples ranged from 37.7 μm to 95.78 μm ,
227 with an average of 66.62 μm (Fig. 9). Different compositional types of MPs had different average
228 sizes, with the PC being 95.78 μm , PP 78.08 μm , PET 69.7 μm , PE 63.23 μm , PS 66.18 μm , PVC
229 56.35 μm , PA 52.15 μm , PU 38.42 μm and Silicone 37.7 μm in descending order.

230 The size distributions are also different, with the average size of MPs in the central zone as 59.3
231 μm . In the southern zone, the average size of MPs is 57.52 μm . In the northern area, the average size
232 of MPs is 70.67 μm . It is noted that the higher number concentrations of MPs were associated with
233 the smaller average sizes in the central and southern zone.

234 We divided the size of MPs into three segments, 20-100 μm , 100-200 μm , and >200 μm . The
235 study found that MPs accounted for 84.63%, 13.34% and 2.83% in size range 20-100 μm , 100-200
236 μm and >200 μm (Fig. 10) respectively. The results showed that the number concentration of MPs in
237 the atmosphere increases with the decrease of size, with smaller MPs being associated with higher
238 number concentration. The greater abundance of MPs in smaller sizes may be attributed to the rapid
239 degradation of small plastic fragments (Zhang et al., 2016). In the >200 μm size segment, fibrous
240 MPs accounted for 5.26%. In the 100-200 μm size segment, fibrous MPs accounted for 8.16%, and
241 in the 20-100 μm size segment, fibrous MPs accounted for 10.39% (Fig. 11). The results indicate that
242 the amounts of fibrous MPs in the atmosphere increases with the decrease of size. We also found that
243 pellet/sphere, fragment, and fiber MPs are dominant in the 20-100 μm size segment (Fig. 12). These
244 results indicate that the MPs in the atmosphere of Beijing mainly come from the degradation of large
245 plastics.

246 **4. Discussion**

247 **4.1. MPs aging and health risk**

248 The aging processes and resulting MPs characteristics have been the subject of scientific
249 investigation (Lambert and Wagner, 2016). The formation of aged MPs is part of the processes that
250 will eventually lead to the breakdown of the plastics in the environment into non-plastic end products.

251 (Liu et al., 2019a). This degradation process can involve environmental weathering, ultraviolet
252 radiation, biodegradation, physical wear and chemical oxidation (Jahnke et al., 2017). MPs can age
253 more rapidly in the atmosphere than in water because of the availability of oxygen and higher levels
254 of ultraviolet radiation (Mao et al., 2020). The temperature, ultraviolet rays, ozone and other
255 substances in the atmosphere will directly act on MPs, resulting in their aging. In the Beijing dustfall
256 samples, we found that many of the MPs have undergone various degrees of recognizable aging.
257 Visual damage in the form of collapses, cracks, and structural embrittlement were observed in the
258 FESEM images of some MPs (Fig. 6).

259 A study of remote lakeshore sediments on the Tibet Plateau found that damage to MPs might have
260 resulted from collision with wind-mobilized sand grains (Zhang et al., 2016). Mineral particles are
261 common in Beijing's atmosphere (Wang et al., 2022). It is speculated that many of the damage
262 features seen at a microscopic level could have been the result of impact with atmospheric mineral
263 particles. However, cracking and embrittlement may not be the result of just mechanical weathering,
264 but the damage is likely to be a combination of ultraviolet radiation, oxidation, as well as the physical
265 weathering. As stated, a significant proportion of the MPs are secondary particles derived from larger
266 plastic pieces. Therefore, the regular observation of microscopic damage features is to be expected as
267 part of the process of converting the primary sources into secondary particles. As the particles become
268 smaller, the mechanisms of weathering are likely to subtly change as the smaller particles are less
269 prone to physical assault and become more brittle. Once the particles become exceedingly small it is
270 likely that most of the weathering damage is no longer physical, but rather driven by ultraviolet
271 radiation and atmospheric oxidation.

272 Ultraviolet radiation and oxidation are important factors that cause carbon-carbon bond breaks in
273 MPs as part of the plastic degradation process (Gewert et al., 2015). This change of chemical structure

274 and morphology will alter their macroscopic properties, with subsequent weathering leading to further
275 embrittlement and disintegration of plastics (Halle et al., 2017). A study on the degradation of PS
276 found that the size, surface morphology and microstructure will change with aging (Lambert and
277 Wagner, 2016). The aging processes are generally believed to be capable of enhancing the sorption
278 potential of MPs in soil, and mobility of the particles in groundwater (Ren et al., 2021). An isothermal
279 adsorption model shows that aging can significantly increase the adsorption of heavy metals by PS
280 (Mao et al., 2020).

281 In recent years, Beijing has experienced frequent haze events, and this atmospheric particulate
282 matter contains a large number of harmful substances (Feng et al., 2020; Li et al., 2020a; Shao et al.,
283 2021a). The dustfall samples have shown that the Beijing atmosphere contains MPs, which are part
284 of the pollution cocktail. Human exposure to toxic substances can be through three different pathways:
285 dermal, ingestion and inhalation (Cabral-Pinto et al., 2020). Dermal exposure is highly unlikely to
286 present a health risk as the MPs levels are so low, and the skin presents an effective barrier to the
287 MPs. Nanoparticles can cross the skin barrier, however the particles sizes recorded in this study are
288 much larger than nanoparticles. It is however possible that some atmospheric MPs could exist as
289 nanoparticles. Inhalation again depends upon the particle size, with PM_{2.5} (aerodynamic diameter less
290 than or equal to 2.5 µm) commonly considered to be the size that determines whether the particles
291 are capable of being respired into the deep lung (Shao et al., 2022b). The smallest of the MPs types,
292 Silicone, had an average size of 37.7 µm, and therefore is much larger than what is normally
293 considered to be the largest atmospheric particulate matter PM₁₀ (10 µm equivalent spherical
294 diameter). In this case the silicone particles would be considered to be a nuisance dust, which would
295 be filtered-out in the nose and upper airways. However, we also find MPs smaller than 2.5 µm (Fig.
296 6a) in this study. Although we cannot determine the type of MPs, these MPs have the same

297 aerodynamic characteristics as PM_{2.5} particles in the air, and they can reach deep lungs or alveoli
298 through respiration (Enyoh et al., 2019). The most recent research has found MPs in human blood
299 (Leslie et al. 2022). In the third potential pathway airborne MPs can be inadvertently ingested, causing
300 physical damage to the body. Study have found that MPs can be absorbed by human tissues through
301 phagocytosis and cell adsorption in the respiratory system and gastrointestinal tract, leading to
302 inflammation, cell necrosis and tissue tearing (Enyoh et al., 2019). Research has shown that MPs can
303 bioaccumulate by ingestion in a range of organisms (Prata et al., 2020), typically organisms such as
304 aquatic filter-feeders. Airborne MPs can also enter the body by eating contaminated food (Khalid et
305 al., 2020). Furthermore, recent study has found that Novel Coronavirus can be transmitted by aerosols
306 (Shao et al., 2021b) and can survive on the surface of aerosols for up to 72 hours (Salimi et al. 2022).
307 MPs are also a kind of particulate matter in the atmosphere, so MPs may also be used as a viral carrier.

308 Based on the above discussion, we recognize that aged MPs in the Beijing atmosphere may adsorb
309 toxic and harmful substances. Smaller MPs of undetermined component types in the study (Fig. 6b)
310 may form part of PM_{2.5}, therefore, MPs with smaller sizes could pose a yet unquantified health risk
311 to Beijing's residents.

312 **4.2. MPs possible original sources: proximal and distal**

313 The compositions and morphologies of MPs are controlled by the chemistry of the original
314 plastics that were used in their manufactured sources. In this study, PP accounted for the highest
315 concentration in MPs, and the central Beijing zone has a highest level. The size of the particles and
316 their distribution across Beijing supports the view that a significant proportion of these MPs were
317 created in central Beijing. In the central zone, numerous service industries produce large number of
318 packaging products, such as foam plastic boxes, PP plastic cups, food packaging bags and other

319 similar products; these when degraded are likely to be an important component of PP aggregation at
320 the S5 and S6 sample sites. In addition, PP is widely used in injection molded products (Li et al.,
321 2018), so injection molded domestic products may also be a source of PP. The overall second highest
322 number concentration and proportion of MPs is PA in the dustfall samples, with southern zone having
323 the highest number concentration. PA plastics are often found as fibers that are mixed with other types
324 of fibers to improve the wear resistance of fabrics (Liu et al., 2019b). With the breakdown or daily
325 wear of those fabrics the individual PA fibres would be released as MPs (Wright et al., 2020).
326 Research into indoor air at schools in Barcelona, Spain, has shown that the children's daily wear of
327 clothing releases fibers into the school rooms, therefore this type of fibrous MP would be expected in
328 any dense urban environment (Moreno et al., 2014). Investigations have shown that PA is widely used
329 in many fields, including industries such as pharmaceutical, beverage, furniture, domestic machinery,
330 transport, and clothing (Kasal et al., 2020; Welle et al., 2012). Since these industries are found in the
331 southern zone, this area may be an important original source of PA. Given the distribution of the MPs
332 within Beijing, their specific chemistries, and the probable original plastic sources, we can conclude
333 that a significant percentage of the MPs are created locally from daily life, service industries and
334 industrial emissions. Many of the waste plastic products will have been disposed of at ground level
335 and disintegrated at that level by a combination of chemical and physical degradation. It is probable
336 that a significant component of the dustfall MPs did not become sufficiently airborne to be inhalable
337 but were moved around at a near surface level either by natural wind or anthropomorphic generated
338 air movements, such as traffic air turbulence resuspension.

339 The morphologies of MPs provide important characteristics for tracing their possible sources.
340 Fragments are the dominant morphological type of MPs in the Beijing dustfall. Previous studies have
341 suggested that the fragmental MPs were created by the degradation of larger plastic objects (Müller

342 et al., 2018). The FESEM images show that larger fibrous MPs could splinter to create many more
343 fibrous fragments with smaller particle sizes as a result of aging (Fig. 6d). The pellets or spheres MPs
344 are generally thought to be primary particles released from personal care products, such as medicines,
345 and cosmetics (Alidoust et al., 2021). Routinely used in cosmetics these MPs have a number of trade
346 names, such micro-pearls, and nano-pearls, and given their microscopic size an individual application
347 of skin cream can contain many tens of thousands of particles, available for release into the
348 environment as the creams dry or are exposed to wind. As one of the largest megacities in the world,
349 Beijing has a huge population, a developed economy and advanced medical technology. This vast
350 population uses many medicines and applies MP-containing cosmetics on a daily basis, and this will
351 be a significant source of the pellet/sphere MPs found in the Beijing dustfall.

352 **5. Conclusions**

353 1) The number concentrations of MPs in the Beijing dustfall show an average of 123.6 items/g,
354 with the highest number being in the central zone, and the lowest number being in the northern zone.

355 2) Nine compositional types of MPs were identified in the Beijing dustfall, including PP, PA, PS,
356 PE, PET, Silicone, PC, PU and PVC. PA is the most common plastic type in the southern zone, PP
357 dominates the central zone, whereas the northern zone had a diverse combination of different
358 compositional types.

359 3) The morphologies of the MPs in the Beijing dustfall are of three basic types: fragments, fibers,
360 and pellet/spheres, with the fragments being the most common. There is no obvious distribution
361 difference in the morphological types of MPs in the central, southern, and northern zone of Beijing.
362 SEM images show the presence of aged MPs in the Beijing dustfall.

363 4) The average size of the MPs in the Beijing dustfall is 66.62 μm . The numbers of fibrous MPs

364 in the dustfall increases with the decrease of size. The results indicated that the MPs in the Beijing
365 dustfall mainly come from the degradation of larger plastics.

366 **6. Acknowledgments**

367 This study is supported by the National Natural Science Foundation of China (Grant No.
368 42075107), the Fundamental Research Funds for the Central Universities (2022YJSDC05) and the
369 Yueqi Scholar fund of China University of Mining and Technology (Beijing).

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605

606 **Table caption**

607 Table 1. The details of sample collection in Beijing.

608

609

610 **Figure captions**

611 Fig. 1. Location of the study area and distribution of the sampling sites.

612

613 Fig. 2. The number concentrations of MPs at the different sampling sites in the Beijing dustfall.

614

615 Fig. 3. Wavenumber and absorbance of different compositional types of microplastics collected in
616 Beijing dustfall. The solid line is the spectrum obtained by particle testing, and the dotted line is the
617 standard spectrum.

618

619 Fig. 4. The number concentrations of different compositional types of MPs in the Beijing dustfall.

620 Polypropylene (PP), Polyamide (PA), Polystyrene (PS), Polyethylene (PE), Polyethylene
621 Terephthalate (PET), Silicone, Polycarbonate (PC), Polyurethane (PU) and Polyvinylchloride (PVC).

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623 Fig. 5. Relative abundances of different compositional types of MPs at different sampling sites in the
624 Beijing fallout dust. Polypropylene (PP), Polyamide (PA), Polystyrene (PS), Polyethylene (PE),
625 Polyethylene Terephthalate (PET), Silicone, Polycarbonate (PC), Polyurethane (PU) and
626 Polyvinylchloride (PVC).

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628 Fig. 6. FESEM images showing different morphological types of MPS in the Beijing dustfall. a,
629 pellet/sphere; b, fragment; c and d, fiber; e, higher magnification of a stress embrittlement on the fiber
630 in image d.

631

632 Fig. 7. The relative abundances of different MPs morphological types at different sampling sites in
633 the Beijing dustfall.

634

635 Fig. 8. The relative abundances of different MPs morphological types for the different compositional
636 types in the Beijing dustfall.

637

638 Fig. 9. Averaged sizes by equivalent circular diameter of the different MPs in the Beijing dustfall.
639 The error bar stands for the standard deviation.

640

641 Fig. 10. The number of microplastics in different MPs size ranges in the Beijing dustfall.

642

643 Fig. 11. Relative abundance of different morphological types in different MPs size ranges in the
644 Beijing dustfall.

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646 Fig. 12. Relative abundance of pellets, fragments, and fibers in different size ranges of MPs in the
647 Beijing dustfall.

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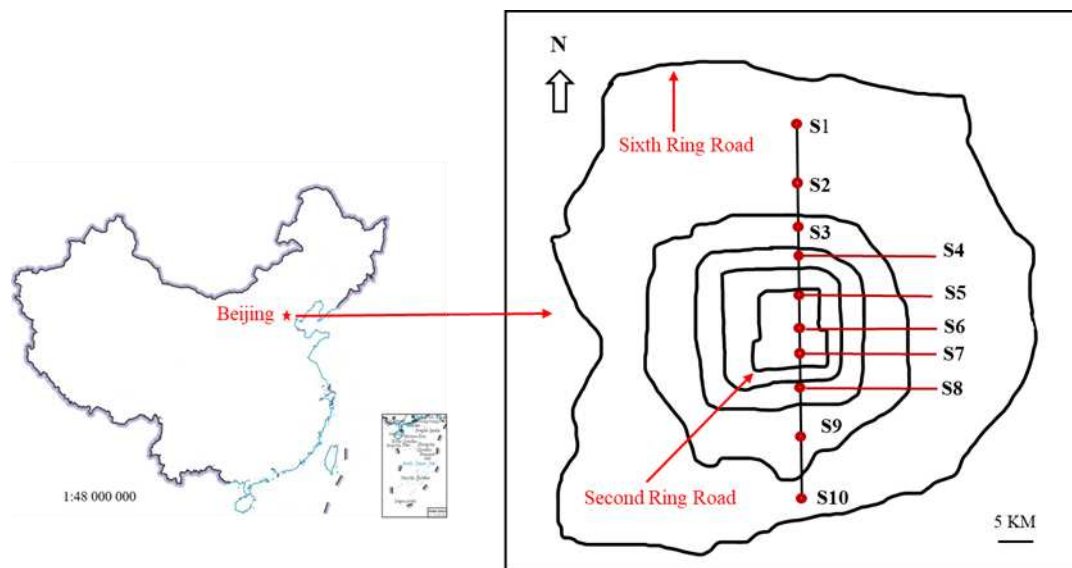
Table 1. The details of sample collection in Beijing.

Sample No	Sampling day	Sampling site environment	Wind direction
S1	2021.06.09	Residential area	North wind
S2		Park	
S3		Near the road (There are tall buildings between the sampling site and the road)	
S4	2021.06.10	Under the office building	West wind
S5	2021.12.10	Nanluoguxiang (residential areas, food stalls street, densely populated)	North wind
S6	2021.06.10	Residential area (close to tourism service industry)	West wind
S7	2021.12.11	Near the road (there are tall buildings between the sampling site and the road)	Northwest wind
S8	2021.06.10	Residential area (close to a pharmaceutical company)	West wind
S9	2021.06.15	Outside the residential area (close to construction activity)	North wind
S10		Residential area (close to beverage, furniture, machinery, and clothing companies)	

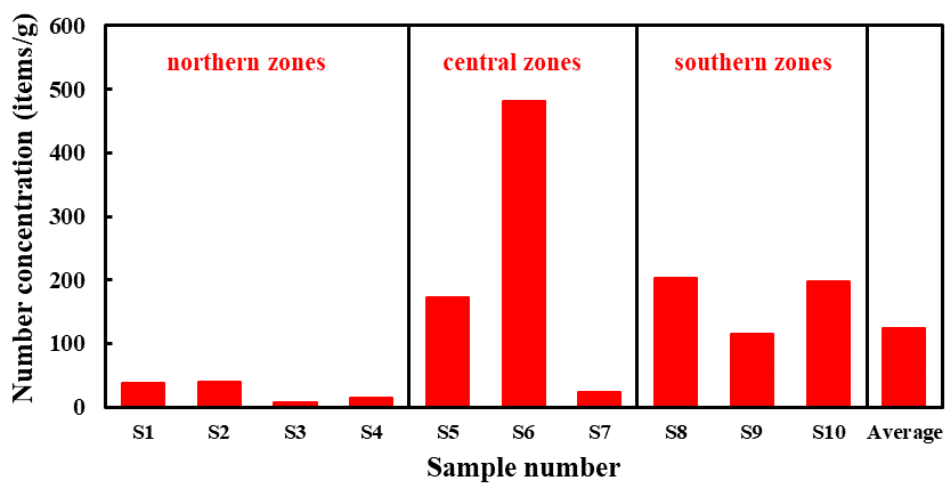
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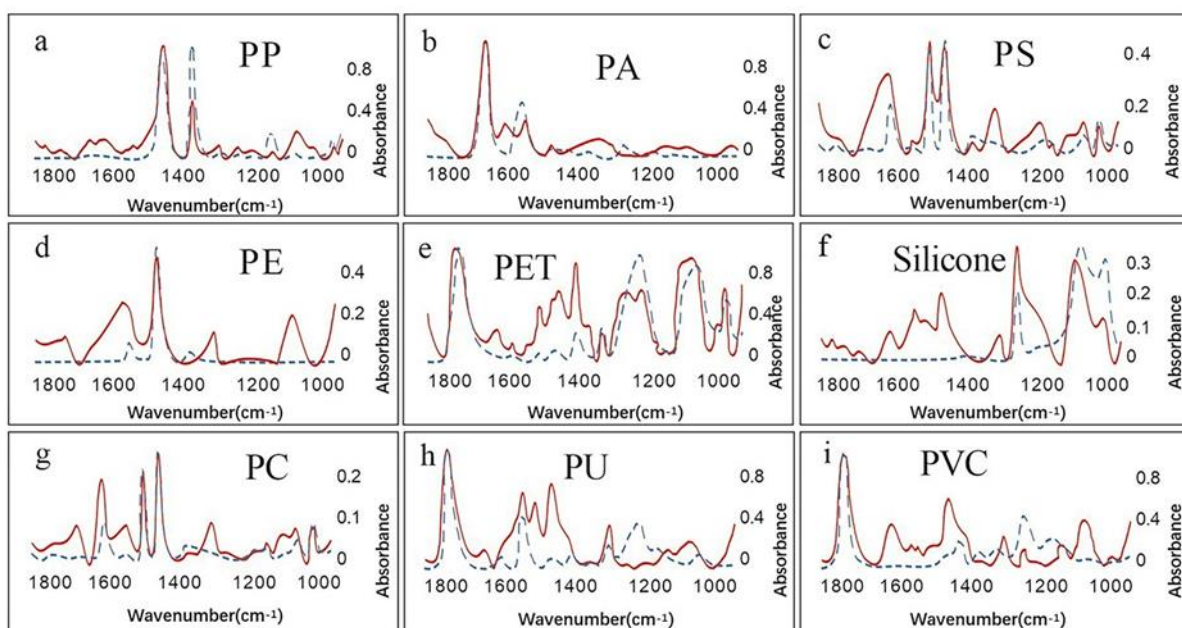
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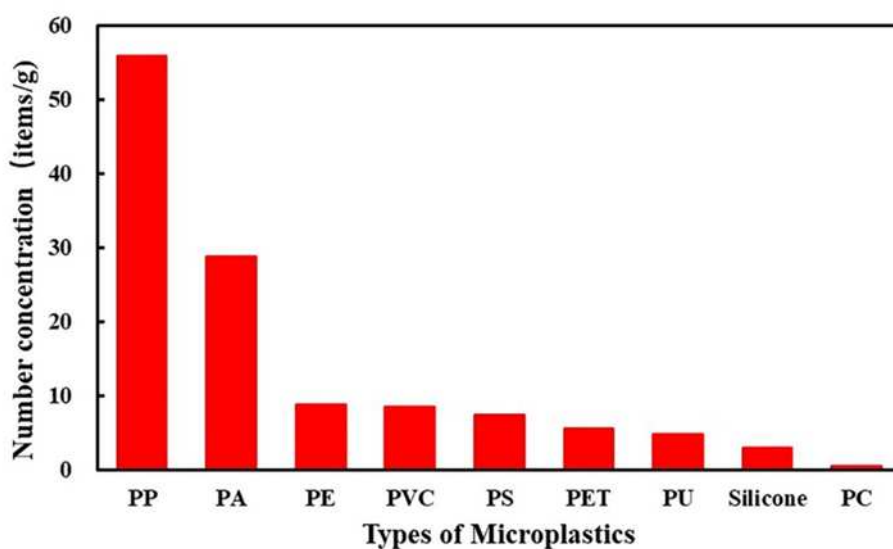
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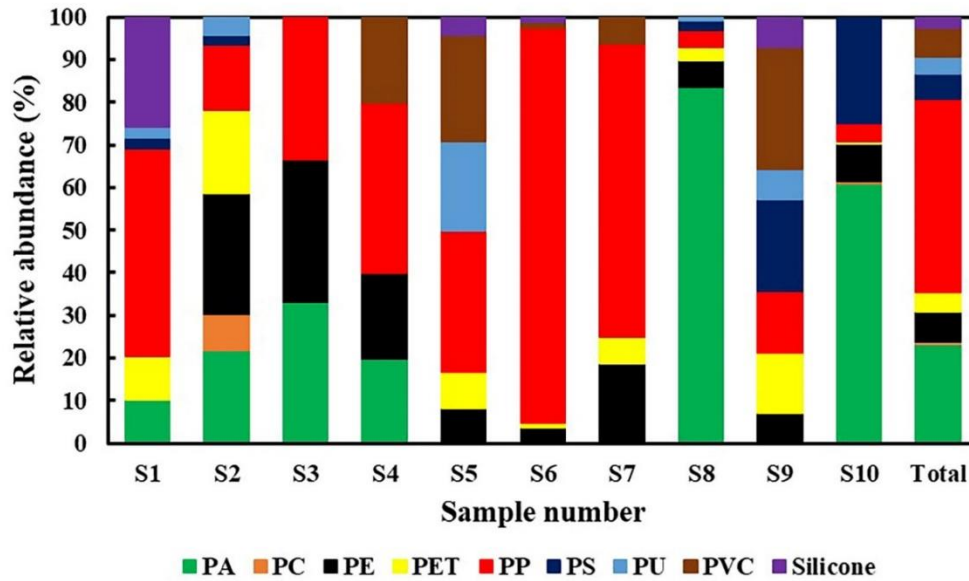
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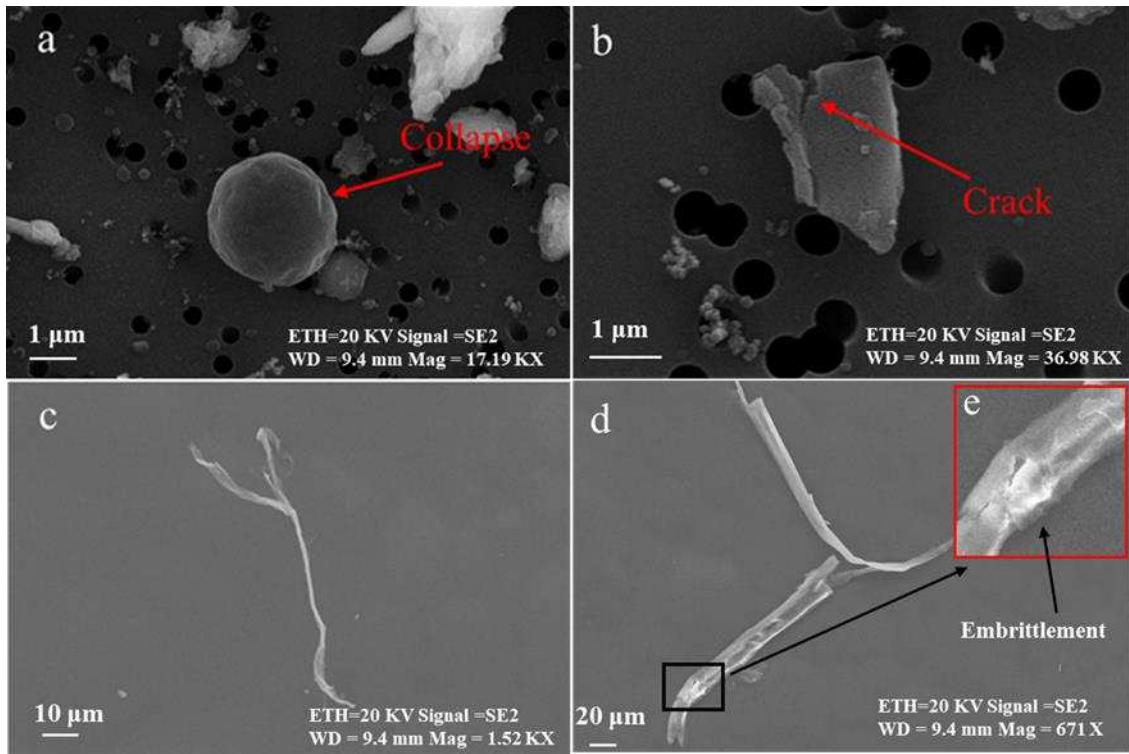


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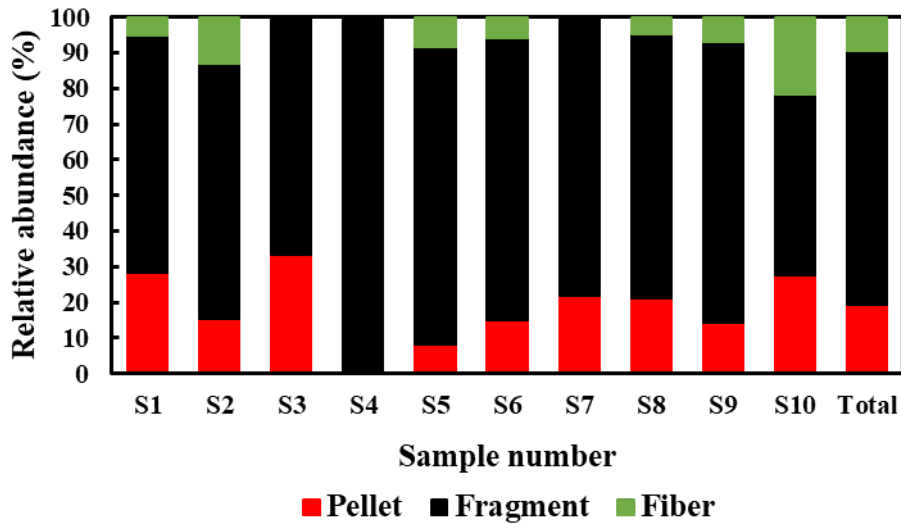
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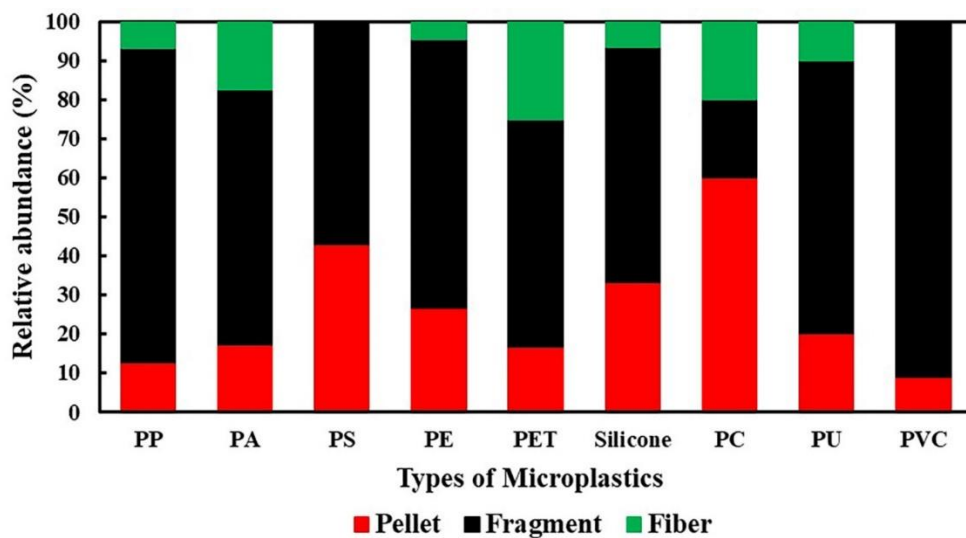
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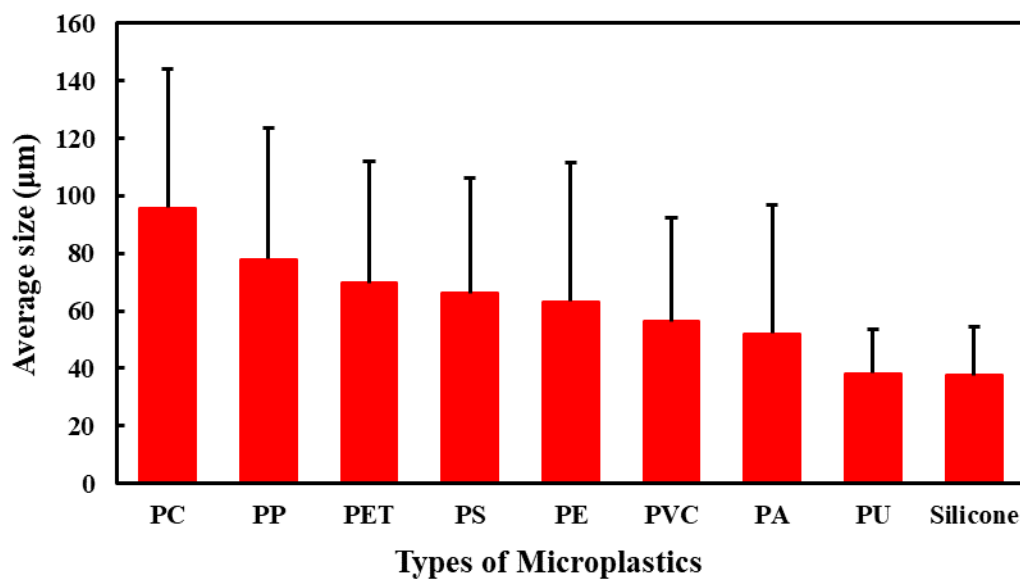
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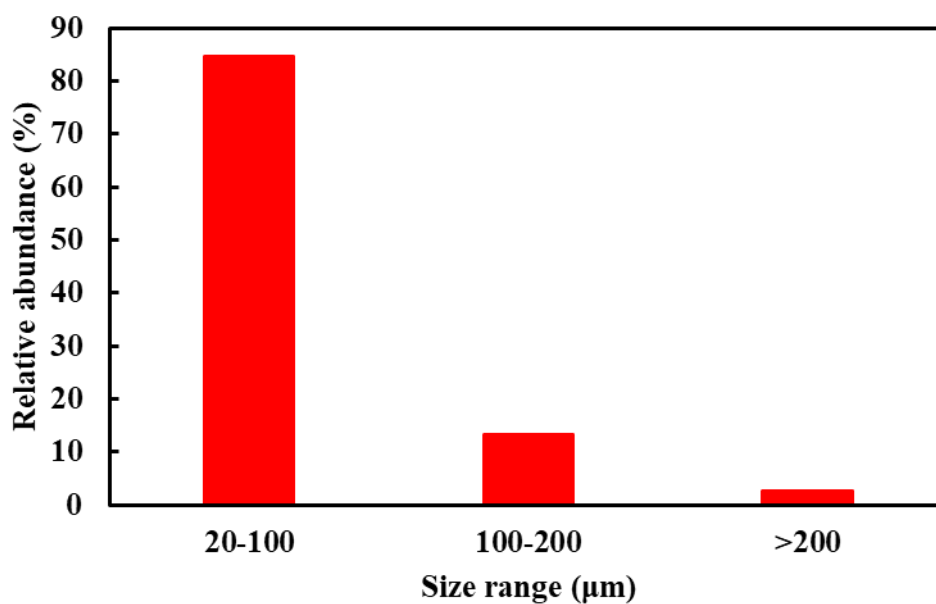
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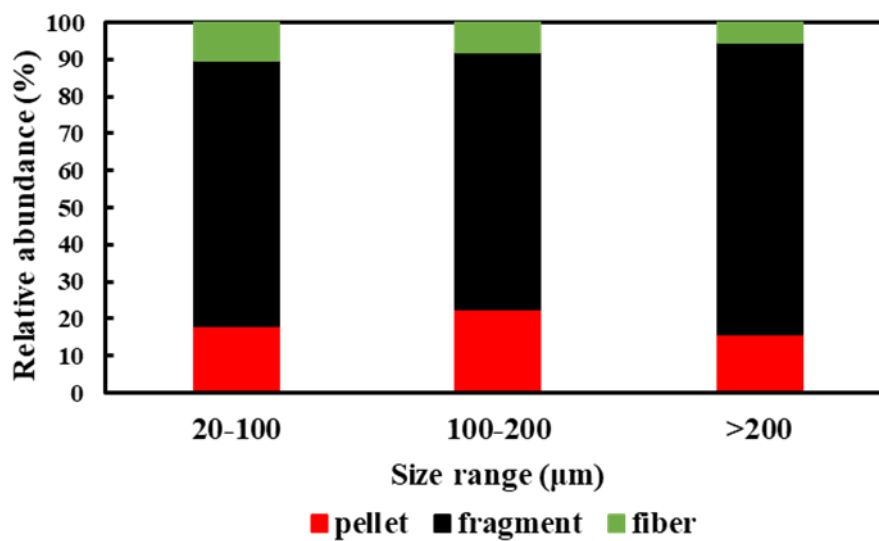
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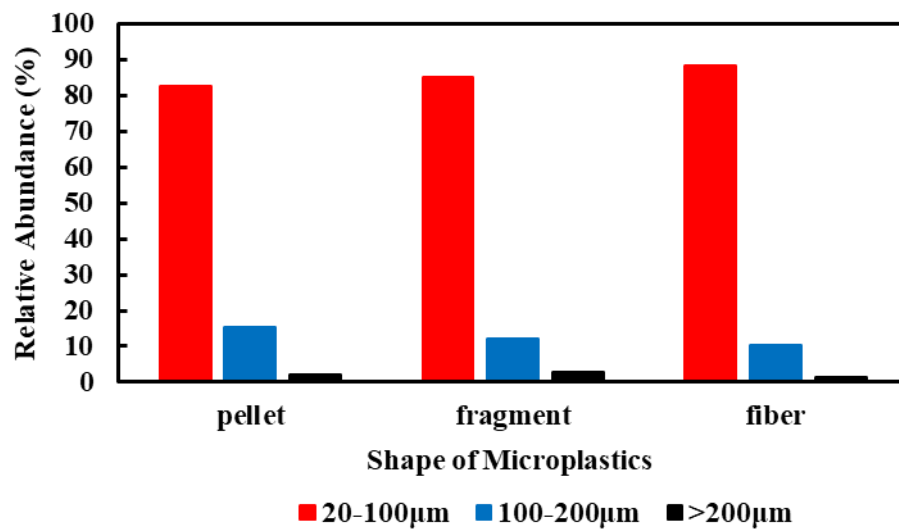
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