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ORIGINAL ARTICLE

Airborne hydrophilic microplastics in cloud water at high altitudes and their role in cloud formation

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Abstract

Microplastic pollution is occurring in most ecosystem, yet their presence in high altitude clouds and their influence on cloud formation and climate change are poorly known. Here we analyzed microplastics in cloud water sampled at the summits of Japan mountains at 1300–3776 m altitude by attenuated total reflection imaging and micro-Fourier transform infrared spectroscopy. We observed nine microplastics including polyethylene, polypropylene, polyethylene terephthalate, polymethyl methacrylate, polyamide 6, polycarbonate, ethylene–propylene copolymer or polyethylene–polypropylene alloy, polyurethane, and epoxy resin. Microplastic were fragmented, with mean concentrations ranging from 6.7 to 13.9 pieces per liter, and with Feret diameters ranging from 7.1 to 94.6 µm. Microplastics bearing hydrophilic groups such as carbonyl and/or hydroxyl groups were abundant, suggesting that they might have acted as condensation nuclei of cloud ice and water. Overall, our finding suggest that high-altitude microplastics cloud influence cloud formation and, in turn, might modify the climate.

Keywords Cloud water · Airborne microplastics · µFTIR spectroscopy · Air pollution

Introduction

Plastics have become quite popular because of their low cost, light weight, and excellent malleability, which has increased worldwide pollution (Rochman et al. 2019; Zhang et al. 2020). Microplastics smaller than 5 mm are referred to as microplastics, which include microbeads used in cosmetics (Anderson et al. 2016), the ship-breaking industry (Reddy et al. 2006), and fertilizers (Katsumi et al. 2021, 2022, 2023), as well as in the degradation of larger plastics, such as plastic bags and plastic containers (Gesamp 2016; Song et al. 2017). Previous studies have detected MPs in various environments (e.g., oceans, rivers, and soil), in the digestive tracts of vertebrates and invertebrates (Rochman 2015; Green et al. 2016; Romeo et al. 2015; de Souza Machado et al. 2018; Chia et al. 2021), and in the lungs of wild birds (Tokunaga et al. 2023). MPs were also detected in the blood,

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lungs, and placentas of pregnant women (Amato-Lourenço et al. 2021; Ragusa et al. 2021; Leslie et al. 2022).

More than 10 million tons of plastic enter the ocean annually from land (Jambeck et al. 2015). When marine organisms inadvertently ingest these plastics, they can potentially obstruct the digestive tract and accumulate hazardous materials in tissues (Andrady 2011; Wright et al. 2013; Auta et al. 2017). Microplastics are also found in terrestrial ecosystems, with 79% of global plastic waste deposited in landfills (Geyer et al. 2017) because of their persistent nature, which can last for hundreds of years (Zhang and Liu 2018). The accumulation of MPs in the soil environment can reduce seed germination rates and shoot heights (Boots et al. 2019), as well as trigger changes in soil water-holding capacity and soil structure (de Souza Machado et al. 2018; Zhang et al. 2019).

Compared with microplastics studies in marine and terrestrial environments (Auta et al. 2017; Alimba and Faggio 2019; Prata et al. 2019), research on airborne microplastics has been limited. Early studies on airborne microplastics primarily focused on atmospheric deposition (Dris et al. 2015; Cai et al. 2017; Allen et al. 2019), but recent research has increasingly concentrated on atmospheric aerosols. However, comparing data across studies is difficult because of variations in the collection, pretreatment, and identification methods used by different researchers. In addition, most of the identified airborne microplastics are larger than the particulate matter with diameter smaller than 2.5 μm ($PM_{2.5}$) range of concern for health effects. Their potential sources are diverse; land-based sources include road dust (Dehghani et al. 2017; Abbasi et al. 2019), landfills (Nayahi et al. 2022), tire and brake wear (Carr 2017; Cai et al. 2021), artificial turf (Luo et al. 2021; Zhao et al. 2023), and clothing (Rogge et al. 1993). The ocean can also transfer airborne microplastics to the atmosphere through aerosolization processes (Allen et al. 2020). Sea salt aerosols, which are generated by wave action and bubble burst ejection, may also play a significant role in this process (Allen et al. 2020, 2022; Ferrero et al. 2022). Catarino et al. (2023) performed breaking wave experiments by adding fluorescent polystyrene beads (0.5–10 μm) to artificial seawater and reported that the 0.5-μm beads concentrated 20-fold in sea spray particles and the 10-μm beads concentrated twofold.

The free troposphere is an important pathway for the long-range transport of air pollutants owing to strong wind speeds; it has been observed that airborne microplastics are also transported in the free troposphere and contribute to global pollution (Evangeliou et al. 2020; Allen et al. 2021). In addition, airborne microplastics may act as cloud condensation nuclei and ice nucleus particles during transport in the free troposphere and atmospheric boundary layer, thereby potentially promoting cloud formation (Aeschlimann et al. 2022). Plastics are hydrophobic but become hydrophilic after prolonged exposure to ultraviolet light (Bain and Preston 2021). Furthermore, the adsorption of mineral particles and polycyclic aromatic hydrocarbons on their surfaces enhances their ice nucleation capacity (Ganguly and Ariya 2019). The first objective of this study was to determine the presence of airborne microplastics in cloud water in the atmospheric boundary layer and free troposphere. The secondary objective was to determine the properties of the identified airborne microplastics, that is, polymer type, shape, Feret size distribution, and concentration in cloud water. The tertiary, we aimed to estimate the origin of airborne microplastics in cloud water by combining major ion concentration and backward trajectory analysis of air masses. To the best of our knowledge, this is the first report on airborne microplastics in cloud water.

Experimental

Observation sites

The summit of Mt. Fuji (35° 35′ N; 138° 73′ E, 3776 m a.s.l.), which is the highest mountain in Japan and an isolated peak, is located in the free troposphere, whereas the southeastern foot of Mt. Fuji—Tarobo (35° 19′ N; 138° 48′ E, 1300 m a.s.l.) and the summit of Mt. Oyama (35° 26′ N; 139° 37′ E, 1252 m a.s.l.) are located in the atmospheric boundary layer (Fig. S1). Mount Fuji Research Station is located on Kengamine, which is the highest of the eight peaks of Mt. Fuji. The west side of the Mount Fuji Research Station is a sheer, unobstructed cliff, allowing for the collection of cloud water unaffected by climbers, mountain huts, or other human activities. Because Tarobo is surrounded by forests, the wind speed is low. Mt. Oyama is located 50 km southwest of Tokyo. The northern side of the summit is open and cloud (fog) event frequency about 30% per year in here (Wang et al. 2021).

Collection of cloud water

A string-type passive cloud collector was used at the summits of Mt. Oyama and Mt. Fuji (Fig. S3a and b), whereas a string-type automatic collector was used at Tarobo (Fig. S3c). To collect airborne microplastics from the cloud water, a 1.0-μm pore size polytetrafluoroethylene hydrophilic membrane filter (H100A047A; Advantec, Co.) was installed in the filter holder of each collector for sampling. Cloud water was collected monthly from Mt. Oyama and biweekly from Tarobo and only collected during the summer months (July and August) on Mt. Fuji. When the researcher was at the summit, cloud water was collected every few hours based on the cloud density. When the researcher was not at the summit, research assistants changed the collection bottles

three times a day: morning (6:00), noon (12:00), and evening (18:00). Tables S1, S2, and S3 in Supplementary Material show the specific sampling dates.

After sampling, the string-type nets of the passive and active collectors were cleaned using ultrapure water. The polytetrafluoroethylene hydrophilic membrane filter from which airborne microplastics were extracted was placed in a 50-mL glass centrifuge tube using stainless-steel tweezers, sealed with a screw cap, wrapped in aluminum foil, placed in a zipper bag, and transported to the laboratory in a cooler box. After measuring the liquid volume, the collected cloud water was transported to the laboratory in a 100-mL polyethylene bottle.

Pretreatment for analyzing airborne microplastics in cloud water

The specific pretreatment process for the samples is shown in Fig. S4.

Analysis of ionic components in cloud water

The cloud water was analyzed using ion chromatography (Dionex Corp., DX-1000 and DX-320). For details, refer to Supplementary Information.

Micro-Fourier transform infrared spectroscopy for detecting and identifying airborne microplastics

Airborne microplastics in the cloud water were identified by attenuated total reflection imaging using micro-Fourier transform infrared spectroscopy (μFTIR ATR imaging) (Spectrum3/Spotlight 400; PerkinElmer, Waltham, MA; hereafter μFTIR ATR imaging). For details, refer to Supplementary Information.

Backward trajectory analysis of air masses

Backward trajectory analysis was used to examine the transport processes of the air masses at the three sites. The analysis was based on the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT4) model (http://www.arl.noaa.gov/ready/hysplit4.html, NOAA) and reanalyzed meteorological data available from the National Centers for Environmental Prediction (http://www.ncep.noaa.gov).

Results and discussion

Airborne microplastics in cloud water

Nine polymers were detected in cloud water: polyethylene, polypropylene, polyethylene terephthalate, polymethyl methacrylate, polyamide 6, polycarbonate, ethylene–propylene copolymer or polyethylene–polypropylene alloy, polyurethane, epoxy resin, and one rubber were detected. For rubber specific identification method, please refer to Tables S2.

Among the identified airborne microplastics, polycarbonate, polymethyl methacrylate, polyurethane, polyethylene terephthalate, and polyamide 6 exhibited C=O stretching vibrations at 1770 cm^{-1}, 1725 cm^{-1}, 1720 cm^{-1}, 1710 cm^{-1}, and 1630 cm^{-1}, respectively (Perkin Elmer, FTIR Blog). Airborne microplastics other than polymethyl methacrylate and polycarbonate were identified by screening for C–H stretching vibrations; however, 94% of polyethylene terephthalate and 50% of polycarbonate were identified by screening for C=O stretching vibrations (1740–1710 cm^{-1}). A wider screening range for the C=O stretching vibrations could improve the detection of these types of polymers. The degradation level, carbonyl index, of polypropylene was calculated as the ratio of the intensity of the peak at 1715 cm^{-1} (carbonyl groups formed during degradation) to the peak at 2920 cm^{-1} (CH_2 asymmetric stretching). Five of the 14 polypropylene in cloud water were degraded, with medium and high degradation levels of 21.4% (Fig. S6). The hydroxyl index calculated from the ratio of the peak at 3467 cm^{-1} (hydroxyl formation due to degradation) to the peak at 2920 cm^{-1} suggested that most polypropylene (85.7 %) formed hydroxyl groups and became hydrophilic.

Figure 1 shows polyethylene terephthalate (fragment, 75.2 μm) in cloud water at Mt. Oyama, polymethyl methacrylate (fragment, 80.3 μm) and polypropylene (fragment, 27.2 μm) in cloud water at Tarobo, and polyamide 6 (fragment, 15.5 μm) in cloud water at Mt. Fuji as some examples.

Figure 2 shows the number of airborne microplastics detected in cloud water and their proportions of polymer types. There were 20 pieces at Mt. Oyama ($n=9$), 13 pieces at Mt. Fuji ($n=19$), and 37 pieces at Tarobo ($n=16$). There were number concentrations of 6.8 pieces L^{-1}, 6.7 pieces L^{-1}, and 13.9 pieces L^{-1}, respectively. The number concentrations in cloud water obtained in this study were very low compared with those in snow cover and ice sheets in the Arctic (10,700 pieces L^{-1}; Bergmann et al. 2019), Europe (1434 pieces L^{-1}; Bergmann et al. 2019), Everest (30 pieces L^{-1}; Napper et al. 2020), and Antarctica (29 pieces L^{-1}; Aves et al. 2022). The analytical methods used in each study were different; therefore, caution should be exercised when comparing the data. Using the same analytical method as that used in this study, we recently reported a number

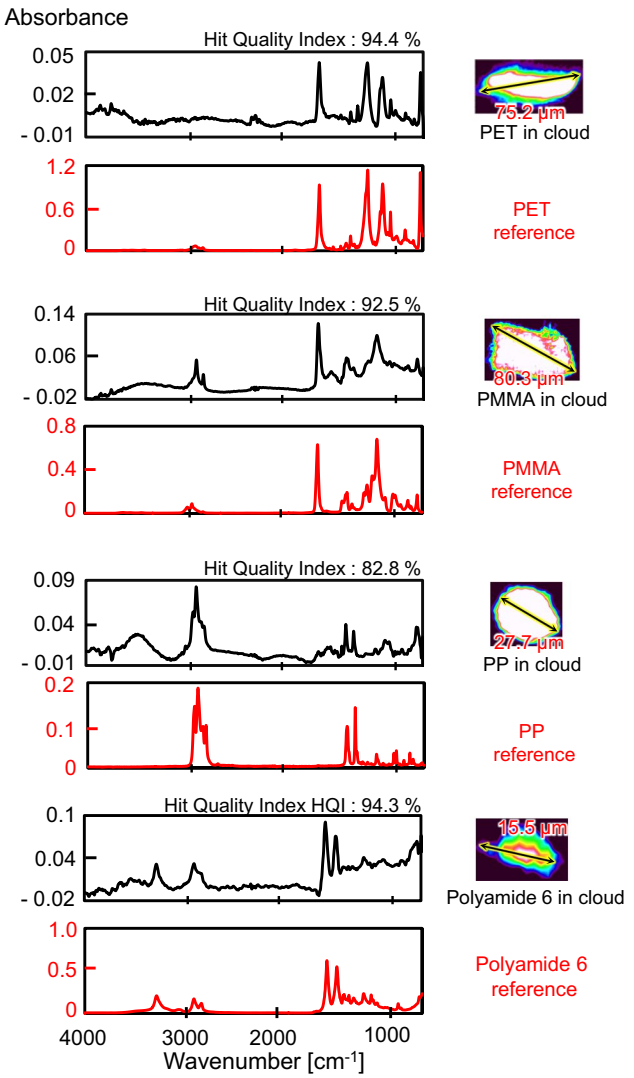

Fig. 1 Fourier transform infrared spectroscopy (FTIR) spectra of airborne microplastics detected in cloud water, including **a** polyethylene terephthalate; PET, **b** polymethyl methacrylate; PMMA, **c** polypropylene; PP, and **d** polyamide 6, as well as reference spectra and the Feret diameter of airborne microplastics

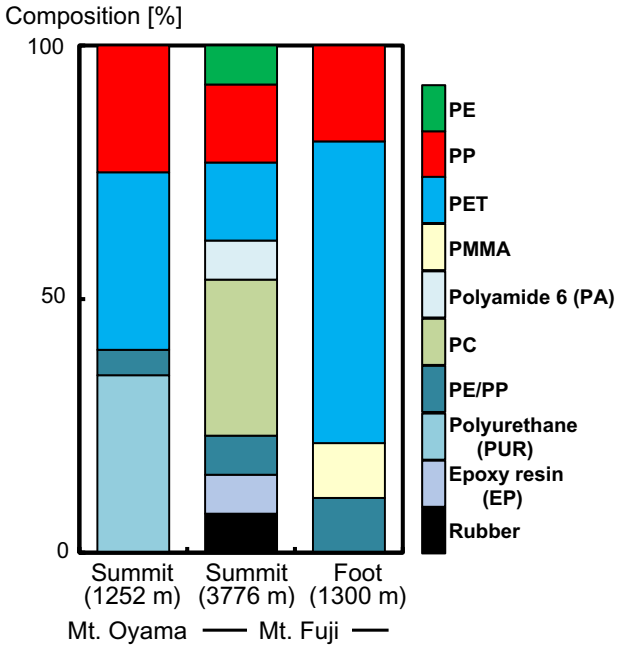

Fig. 2 Material composition of airborne microplastics (AMPs) in cloud water at the summit of Mt. Oyama, at the summit of Mt. Fuji, and at the foot of Mt. Fuji-Tarobo. Twenty pieces AMPs (sample numbers = 9) were detected in Mt. Oyama, 13 pieces (sample numbers = 19) at Mt. Fuji, and 37 pieces (sample numbers = 16) at Tarobo. The full names of the abbreviations in the legend are as follows: polyethylene (PE), polypropylene (PP), polyethylene terephthalate (PET), polymethyl methacrylate (PMMA), polyamide 6 (PA), polycarbonate (PC), ethylene–propylene copolymer or polyethylene–polypropylene alloy (PP/PE), polyurethane (PUR), epoxy resin (EP), and rubber

concentration of 119 pieces L^{-1} in snow cover at Mt. Fuji in 2022 (Tani et al. 2023), indicating that the number concentration of airborne microplastics in cloud water was low.

Polypropylene and polyethylene terephthalate were predominantly detected at Mt. Oyama and Tarobo in atmospheric boundary layer. Other polymers detected in the cloud water were polyurethane at Mt. Oyama, polymethyl methacrylate at Tarobo, and ethylene–propylene copolymer or polyethylene–polypropylene alloy at both locations. However, many types of polymers were detected in the cloud water at Mt. Fuji, with polycarbonate being the major polymer. The number concentration and polymer types of airborne microplastics in cloud water likely depend on the air mass. The origin of the air masses with airborne microplastics detected in the cloud water during the observation period was analyzed using backward trajectory analysis (Fig. S7). Air masses from northern China (NC), southern China (SC), peripheral maritime (PM), and northwestern (NW) directions prevailed at Mt. Oyama and Tarobo, where the types of airborne microplastics were similar. However, the air masses at Mt. Fuji in summer originated not only from the NC and SC directions but also from the southern maritime (SM), eastern maritime (EM), and southeastern (SE) directions. This explains why the similarity in the types of airborne microplastics in cloud water was higher than that in the other two locations. In Southeast Asia, the concentration of airborne microplastics is high and many types of polymers have been detected (Onozuka et al. 2023).

In interpreting the results of this study, it should be noted that the cloud water collectors used at the three sites were not identical. In particular, an active-type collector that can collect more cloud water was used at Tarobo. In addition, the string-type collectors used at all sites are used for the analysis of major ions in cloud water and are unsuitable for the analysis of suspended particles such as airborne

microplastics, as some of them may become trapped in strings and flow channels. This suggests that the number concentrations of airborne microplastics reported in this study were likely underestimated.

The Feret diameter distributions of airborne microplastics in cloud water at three sites, are ranging from 7.1to 94.6 µm (identified airborne microplastics number, $n = 70$) in Fig.S8. The mean Feret diameter was found to be 32.0 ± 20.6 µm at Mt. Oyama ($n = 9$), 29.9 ± 17.5 µm at Mt. Fuji ($n = 19$), and 38.3 ± 23.8 µm at Tarobo ($n = 16$), respectively. The proportion of Feret diameters between 10 and 20 µm in cloud water at Mt. Fuji was approximately twice that of the other two sites in this study. These results suggest that smaller airborne microplastics are more likely to diffuse (Evangeliou et al. 2020) and advect at cloud-forming altitudes (Aeschlimann et al. 2022). The shape of airborne microplastics in cloud water was nearly fragmented (more than 87%) at three sites, similar to the trend of airborne microplastics in wet and dry depositions measured in mountainous areas (Allen et al. 2019). In particular, the proportion of fragmented airborne microplastics in the cloud water was higher at Mt. Fuji than at the other two sites. Mt. Fuji is located in the free troposphere, whereas Mt. Oyama and Tarobo are located at similar altitudes in the atmospheric boundary layer. Our results suggest that small and fragmented airborne microplastics are more likely to be transported to higher altitudes than large and fibrous airborne microplastics because of their size. This indicates that the physical properties of airborne microplastics, specifically their size, play a role in their atmospheric transport.

Sources of airborne microplastics in cloud water

Figure 3 shows time-course change in the number concentrations of airborne microplastics and their composition in cloud water at Mt. Fuji during the sampling period between July 21–22 and 26–27, 2022. Backward trajectory analysis classifies air masses into three categories: northern (Continental), southwestern (Continental), and maritime (Pacific Ocean). Polyethylene terephthalate and ethylene–propylene copolymer or polyethylene–polypropylene alloy were detected in the cloud water from 16:40 to 21:12 on July 21 and from 21:12 on July 21 to 6:27 on July 22, respectively. At this time, the air mass came from the cities of Zhejiang, Fujian, and around the coastal sea of Hainan prefecture of China at a low height, gradually climbing to Mt. Fuji (Fig. S11 & S12), suggesting long-range transportation of air pollutants from the atmospheric boundary layer. The concentrations of NO_3^- and non-sea-salt sulfate (nss–SO_4^{2-}) as indicators of anthropogenic emissions and Na^+ as an indicator of oceanic origin were high during this period in Fig S10. This indicates that the airborne microplastics in cloud water originate from both anthropogenic and oceanic sources. The

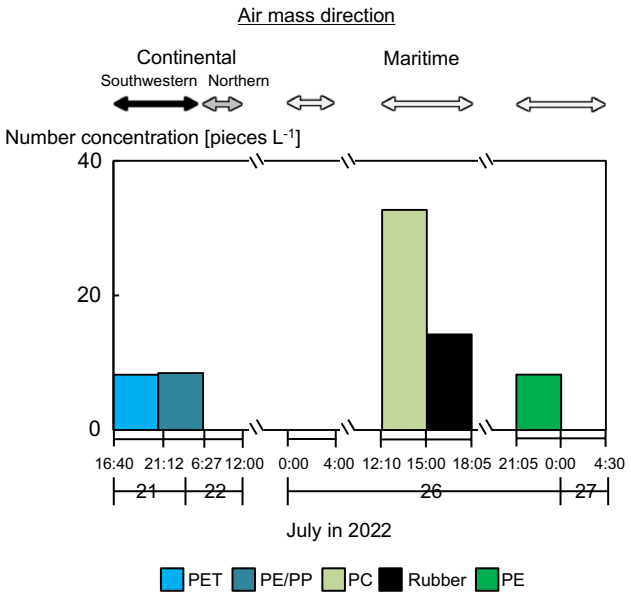

Fig. 3 Microplastics changes over time in cloud water at the summit of Mt. Fuji (Jul. 21 21–22, 2022; Jul. 26–27, 2022), the direction of air mass, the number concentration of airborne microplastics (AMPs) in cloud water, and their compositions. The full names of the abbreviations in the legend are as follows: polyethylene terephthalate (PET), ethylene–propylene copolymer or polyethylene–polypropylene alloy (PP/PE), polycarbonate (PC), rubber, and polyethylene (PE)

size of airborne microplastics was 26.3 µm for polyethylene terephthalate and 28.7 µm for ethylene–propylene copolymer or polyethylene–polypropylene alloy, respectively. Both were fragments. Although the air masses originated from the continental area in the sample from 6:27 to 12:00 on July 22, airborne microplastics were not detected. This is because the air mass originated only from within the free troposphere and did not pass through the atmospheric boundary layer, which was strongly influenced by the ground (Fig. S13).

Airborne microplastics were detected in the cloud water collected at 12:10–15:00, 15:00–18:05, and 21:05 on July 26 to 0:00 on July 27, 2022, when the starting point of the air mass was close to sea level, particularly from 12:10 to 18:05 on July 26 (Fig. S15, S16, and S17). During the studied period at Mt. Fuji, the highest number concentration of airborne microplastics was observed in cloud water collected from 12:10 to 15:00 (polycarbonate: 32.7 pieces/L, size: 10.7–17.8 µm, fragment), followed by in cloud water collected from 15:00 to 18:05 (rubber: 14.2 pieces/L, size: 28.7 µm, fragment). Polyethylene was detected in cloud water collected between 21:05 on July 26 to 0:00 on July 27, when the air mass came from below 850 hPa (approximately 2000 m a.s.l.) in the atmospheric boundary layer (8.2 pieces/L, size: 61.0 µm, fragment). Pan et al. (2019) reported that polyethylene is the predominant component of surface seawater in the Northwest Pacific Ocean. Moreover, polyethylene has been detected in cloud water transported by air

masses originating near the sea surface, suggesting that it may be derived from microplastics in the ocean. Although ocean air was present during these periods, the sodium concentration in the cloud water was low. Sea salt particles are typically found in coarse particle regions and are easily washed away by precipitation. Rainfall was observed around Mt. Fuji during the study period (Japan Meteorological Agency 2023). This is likely the reason for the reduced sodium concentrations.

An air mass passed through the free troposphere over the Pacific Ocean from 0:00 to 4:00 on July 26 and from 0:00 to 4:30 on July 27, without descending below 850 hPa (approximately 2000 m a.s.l.) within the atmospheric boundary layer. This is likely why airborne microplastics were not detected in cloud water (Fig. S14 & S18).

Conclusion

To the best of our knowledge, this study is the first to detect airborne microplastics in cloud water in both the free troposphere and atmospheric boundary layer. Polymers with carbonyl groups like polyethylene terephthalate, polyamide, and polycarbonate were abundant in the cloud water, and most of polypropylene were degraded and had carbonyl group and/or hydroxyl group. The high concentration of hydrophilic polymers in the cloud water suggested that they were removed as cloud condensation nuclei. The Feret diameter of airborne microplastics in cloud water was the smallest in the free troposphere. The shapes of the identified airborne microplastics in cloud water were mostly fragmented at the three sites, especially in the free troposphere. Backward trajectory analysis at the summit of Mt. Fuji in the free troposphere suggested that airborne microplastics in cloud water were originated primarily from the ocean.

Supplementary Information The online version contains supplementary material available at https://doi.org/10.1007/s10311-023-01626-x.

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Author contributions YW helped in data curation, formal analysis, investigation, resources, visualization, writing—original draft. HO contributed to conceptualization, investigation, methodology, resources, project administration, funding acquisition, writing—review and editing. YT and MT were involved in investigation, methodology, writing—review and editing. HH and YM performed investigation, writing—review and editing. NK, AS, KA, YI, and YF helped in methodology, writing—review and editing. MK and YI performed project administration, writing—review and editing. YN was involved in methodology, validation, software, formal analysis, writing—review and editing.

Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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