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Microplastics captured by snowfall: A study in Northern Iran

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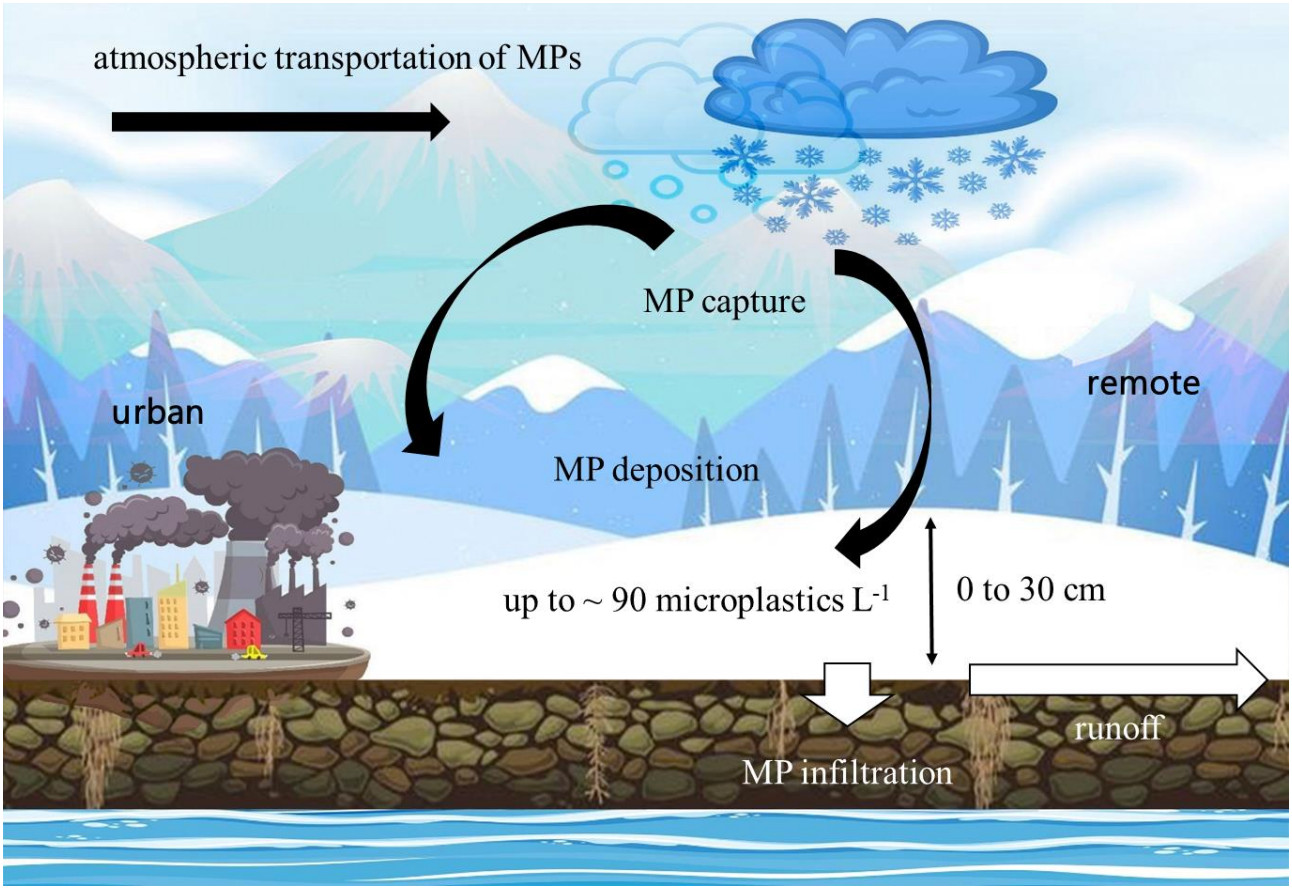
Graphical Abstract



Highlights

- Microplastics (MPs) have been observed in fresh snow in northern Iran
- MPs were heterogeneously distributed throughout the region and at different depths
- Mean and median concentrations were ~ 20 and 12 MP L^{-1}, respectively
- Snow appears to capture a greater diversity of MP sizes and shapes than rainfall
- The significance of snow to the global transport of MPs requires further study.

Abstract

Samples of fresh snow (n = 34) have been collected from 29 locations in various urban and remote regions of northern Iran following a period of sustained snowfall and the thawed contents examined for microplastics (MPs) according to established techniques. MP concentrations ranged from undetected to 86 MP L^{-1} (mean and median concentrations ~ 20 MP and 12 MP L^{-1}, respectively) and there was no significant difference in MP concentration between sample location type or between different depths of snow (or time of deposition) sampled at selected sites. Fibres were the dominant shape of MP and μ-Raman spectroscopy of selected samples revealed a variety of polymer types, with nylon most abundant. Scanning electron microscopy coupled with energy-dispersive X-ray analysis showed that some MPs were smooth and unweathered while others were more irregular and exhibited significant photo-oxidative and mechanical weathering as well as contamination by extraneous geogenic particles. These characteristics reflect the importance of both local and distal sources to the heterogeneous pool of MPs in precipitated snow. The mean and median concentrations of MPs in the snow samples were not dissimilar to the published mean and median concentrations for MPs in rainfall collected from an elevated location in southwest Iran. However, compared with rainfall, MPs in snow appear to be larger and more diverse in their shape and composition (and include rubber particulates), possibly because of the greater size but lower terminal velocities of snowflakes relative to raindrops. Snowfall represents a significant means by which MPs are scavenged from the atmosphere and transferred to soil and surface waters that warrants greater attention.

Keywords: snow; precipitation; atmosphere; deposition; scavenging; fibres; flux

1. Introduction

It is becoming increasingly evident that the atmosphere is a highly important reservoir and transporter of microplastics (MPs). Sources to the atmosphere include emissions from industry, urban-suburban centres and agriculture during all stages of the plastic life cycle (Zhang et al., 2020). Airborne MPs are eventually deposited in the terrestrial and marine environments through dry or wet deposition, although many MPs, and in particular those with favourable aerodynamic properties like fibres, may be transported long distances (100s to 1000s of km) with regional air masses before this takes place (Brahney et al., 2020; Allen et al., 2021). Consequently, MPs are a global problem, with plastic reported in a diversity of regions remote from any significant sources, including the Arctic (Bergmann et al., 2019), the central Pacific Ocean (Liu et al., 2020) and the Himalayas (Napper et al., 2020). Once deposited, MPs may be captured by soil, lost to ground water or entrained by surface water bodies, or may be resuspended by wind or through bubble burst ejection in breaking waves (Allen et al., 2020).

Atmospheric MPs are usually collected passively with a sampling device that provides a location and time specific indication of MP fluxes associated with rainfall and/or dry deposition (Dris et al., 2017; Klein and Fisher, 2017; Abbasi and Turner, 2021). However, despite its importance to global precipitation and the water cycle (Skofronick-Jackson et al., 2019) and evidence for an increase in frequency with climate change (Zhu et al., 2012), relatively little information exists on the mechanisms and significance of snowfall in capturing and depositing MPs. Moreover, most studies consider MP accumulation in Arctic or alpine environments (Bergmann et al., 2019;

Huntington et al, 2020; Parolini et al., 2021) where snow cover is permanent or occurs for long periods of the year and where MP capture (or scavenging) by falling snow and MP deposition on fallen snow may be difficult to distinguish. Snowflakes are larger, less dense and have lower terminal velocities than raindrops and one might expect them to capture and deposit different quantities and types of MPs. The gradual but continuous accumulation of snow during a single but protracted event also allows short-term temporal distributions of MP deposition and airborne MP contamination to be examined through different depths of snow accumulations.

In the present study, we describe, for the first time, the regional sampling of MPs in deposited snow arising from a single, sustained snowfall event. Samples were collected opportunistically from both urban and remote settings throughout northern Iran, and in some deposits snow was sampled at different depths. MPs were identified, quantified and characterised in melted samples using established techniques in order to better understand the role of this type of precipitation in scavenging and depositing MPs from the atmosphere.

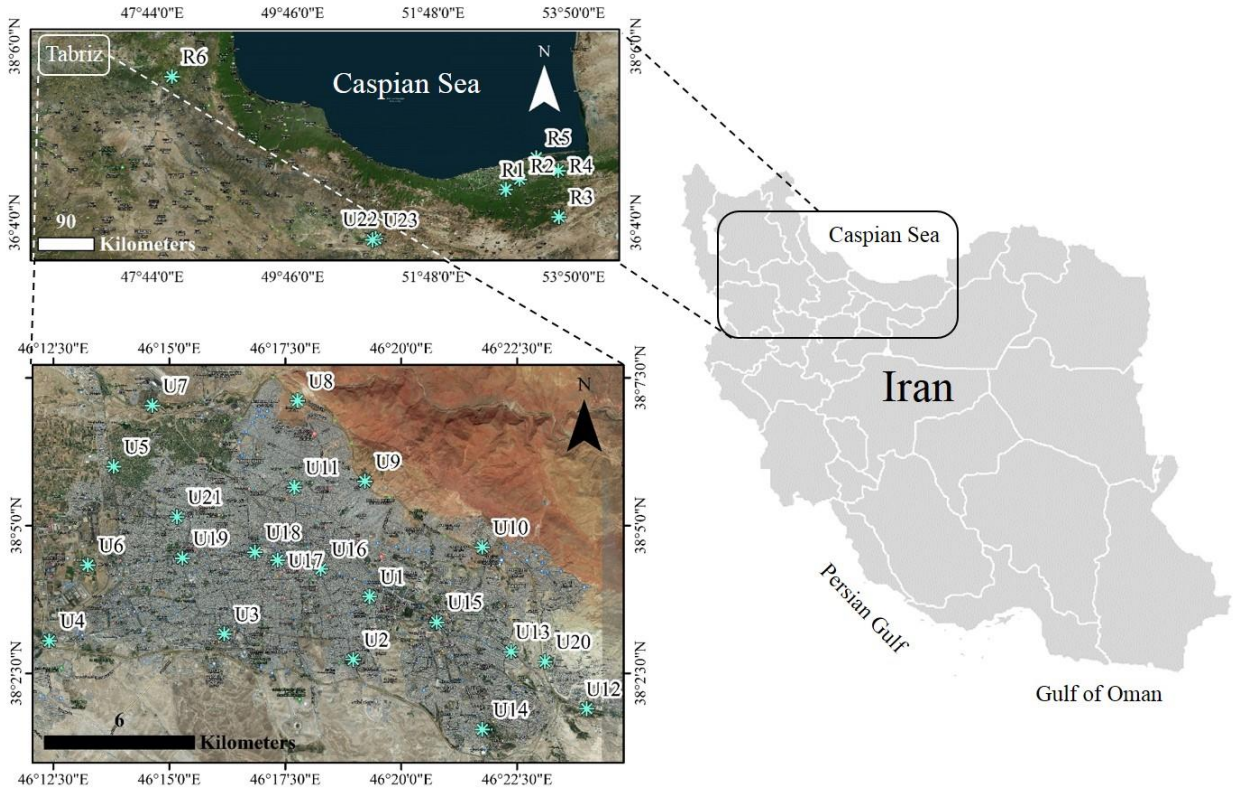


Figure 1: Map of the region under study. Locations pre-fixed with R and U are remote and urban, respectively.

2. Materials and Methods

2.1 Study region and sampling

Northern Iran encompasses various climatic types (including semiarid, cold semiarid and Mediterranean) but snowfall is a common occurrence in the region under study (shown in Figure 1) during winter and when influenced by maritime polar air masses from the northwest and continental polar air masses from Siberia to the north.

Twenty one "urban" locations (U1 to U21), including industrial zones, residential areas, transportation hubs and green spaces, were sampled in Tabriz, a city to the west of the region with a population of about 1.6 million and a mean elevation of about 1350 m above sea level. Tabriz is

118 home to various manufacturing industries, including food, chemicals, electronic equipment and
119 textiles, which contribute to anthropogenic pollutant emissions. Surrounded by mountains and
120 subject to thermal inversions, the city often experiences poor urban air quality (Parsa et al., 2019).
121 Two urban-industrial locations (U22 and U23) were also sampled in the hilly, northern reaches of
122 the Tehran metropolis (population ~ 15 million) to the south east of Tabriz. Six locations were
123 sampled from various, more "remote" areas to the south and south east of Tabriz (R1 to R6). These
124 included small towns and villages within forested areas or on mountain slopes, and a remote
125 location close to a thermal power station (R5).

126 Fresh, deposited snow samples were collected on 21st January, 2021, after a 12- to 15-hour period
127 of sustained precipitation caused by the convergence of polar air from the north and milder air
128 from the southwest. At each location, accessed by vehicle and/or on foot, and from the street, land
129 surface or rooftops, snow was scraped into a 2-L glass jar that had been triple rinsed with filtered,
130 distilled water (see below) using a stainless steel spoon by operators wearing cotton clothing and
131 facing upwind. The depth of snow sampled, determined using a metal ruler, was 5 cm or 10 cm,
132 but at four remote locations (R3, R4, R6) and one urban location (U23) where fresh deposits were
133 sufficiently deep, snow was scraped from the surface (0 to 5 cm) and subsurface (5 to 15 cm) and,
134 in one case (R6), from 15 to 30 cm. Sample jars (34 in total) were wrapped in aluminium foil to
135 avoid external contamination before being transported to the laboratory, thawed, and stored under
136 refrigeration at 4^{o}C pending MP isolation.

137 On the same day and after the precipitation event, a 25-cm diameter, pre-washed metallic pot
138 containing 500 mL of filtered, distilled water, was deployed for 12 h on the rooftop of a domestic
139 property near to site U1 in the city of Tabriz in order to evaluate the significance of dry deposition.
140 The sample was subsequently wrapped in aluminium foil and stored as above.

The potential source range of MPs associated with the period of snowfall was evaluated from 48-h back trajectories from 17th/18th to 21st January, calculated using the National Oceanic and Atmospheric Administration online software, Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) *v*4, and Global Forecast System (0.25 degree global) meteorological data. Backward trajectories of 48 h in duration were calculated at six-hour intervals and at a resolution of 1 degree, and at heights of 500 m and 2000m above ground level (and above the gradient height for open terrain and large cities). Trajectories were integrated as frequency distributions which are illustrated in Figure 2. Consistent with the meteorological conditions during and immediately before sampling, air was brought into the region principally from southwesterly and northerly directions, with a greater northwesterly component at the lower height of 500 m. The range of influence, defined by the distance of the 1% trajectory frequency from Tabriz, extends to about 750 km to the northwest and 2500 km to the southwest at a height of 500 m, and to about 300 km to the north and 3000 km to the southwest at a height of 2000 m.

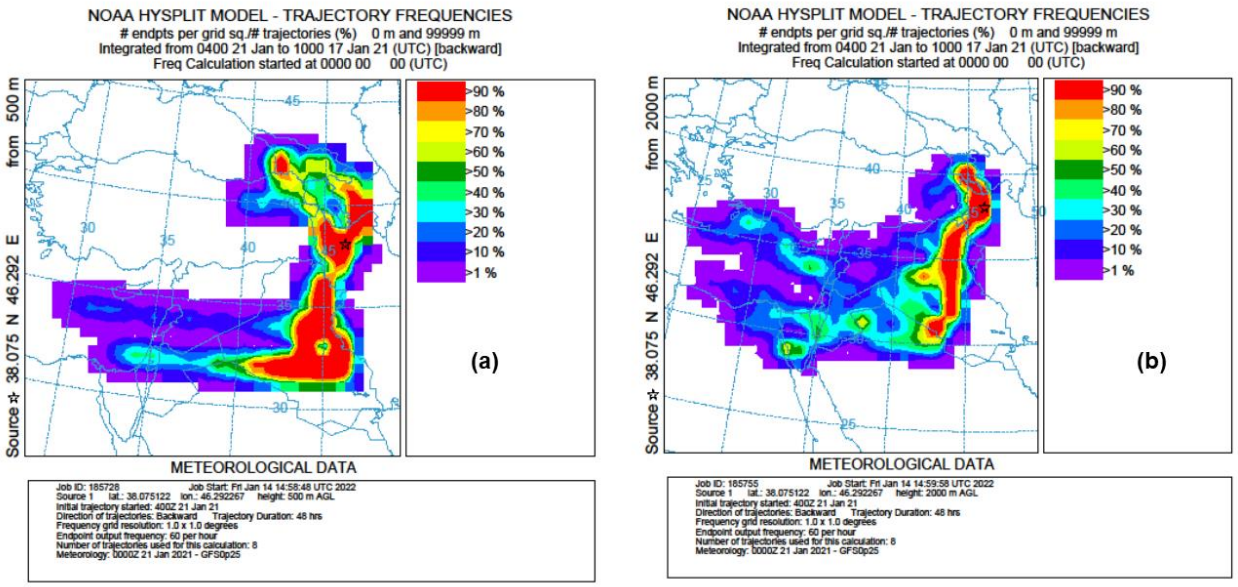


Figure 2: Trajectory frequencies calculated for Tabriz (starred) in northern Iran using HYSPLIT over a 48-h period and at six-hourly intervals for heights of (a) 500 m and (b) 2000 m.

2.2 Isolation of microplastics

To minimise contamination, all glass-ware and metal-ware used for sample processing, manipulation and analysis had been washed three times with filtered (S&S blue band, 2 μm), distilled water and stored in aluminium foil, and all sample processing was undertaken in a customized clean laboratory.

Thawed snow samples and the water capturing dry deposition were removed from refrigeration and allowed to reach room temperature. Using a graduated cylinder, 500 mL of each sample was transferred to a series of 1-L beakers. An appropriate quantity of filtered (< 2 μm) $ZnCl_2$ (Arman Sina, Tehran) was added to each beaker and the contents agitated with glass-covered magnetic stirrers to attain saturated solutions of density ~ 1.8 g cm^{-3}. Each solution, plus beaker rinses of ~ 20 mL saturated $ZnCl_2$, was then transferred to a series of 600 mL separation funnels and the contents were allowed to settle overnight. The upper 100 mL of each solution was retained in the funnels after discarding the remaining contents before aliquots of 100 mL of 30% filtered (< 2 μm) H_2O_2 (Arman Sina, Tehran) were added for 12 h to destroy organic matter. The contents of each funnel, plus triple 20-mL rinses of filtered, distilled water, were filtered through 1 μm Johnson cellulose test membranes to capture MPs, with filters subsequently dried in glass petri dishes and under desiccation for 48 h. Procedural blanks, processed likewise but using 500 mL of filtered (< 2 μm), distilled water in place of snow melt samples, revealed no contamination from exposure to laboratory conditions.

179

180 *2.3 Identification and characterisation of microplastics*

181 MPs retained on each filter were visually identified, counted and characterised under a binocular
182 microscope (Carl-Zeiss, Köln, Germany) at up to 200-x magnification using a 250 µm-diameter
183 stainless steel probe and ImageJ software. MP identification, and with a size detection limit of
184 about 30 to 50 µm, was based on shininess, thickness, hardness, surface and cross sectional
185 structures, and reaction (melting or curling) in the presence of the heated probe (Abbasi et al.,
186 2019). Classification was based on shape (fibre, film, fragment or spherule), length of the longest
187 axis, L ($L \leq 100$ µm, $100 < L \leq 250$ µm, $250 \leq L < 500$ µm, $500 \leq L < 1000$ µm, $L > 1000$ µm)
188 and colour (black-grey, yellow-orange, white-transparent, red-pink or blue-green).

189 The polymeric construction of 47 MPs of a range of shapes, sizes and colours and collected from
190 different locations was determined using a micro-Raman spectrometer (µ-Raman-532-Ci,
191 Avantes, Apeldoorn, Netherland) with a laser of 785 nm and Raman shift of 400-1800 cm^{-1}. Under
192 the microscope and with the aid of a needle and tweezers, MPs were transferred to conductive
193 copper adhesive and polymers were identified by comparing spectra with online, open access data
194 on hosted at Open Specy (https://wincowger.shinyapps.io/OpenSpecy/) (Cowger et al., 2021).
195 These MPs were subsequently mounted on microscope slides and gold-coated before being
196 analysed under a high vacuum scanning electron microscope (SEM; TESCAN Vega 3, Czech
197 Republic) equipped with an energy-dispersive X-ray microanalyser (EDX) and operated at a
198 distance of about 15 mm and with a resolution of 2 nm and an accelerating voltage of 20 kV.

199

200 **3. Results**

In total, 348 particles were identified microscopically as MPs amongst the 34 snow samples considered, with a selection illustrated in Figure 3. MP abundance in individual samples ranged from zero at site U8, the most sheltered (by mountains) and most northerly location in Tabriz, to over 40 at sites U12 and U18, located in the central and eastern reaches of the city, respectively. The mean and median numbers of MPs in the urban and remote samples were 11.3 and 6.0, and 8.7 and 4.0, respectively, and quantities were not significantly different between sample location type ($p = 0.299$) according to a Mann-Whitney test. Where samples were taken from the surface (0 to 5 cm) and subsurface (5 to 15 cm) ($n = 4$), the mean and median numbers of MPs were 9.3 and 10, and 6.5 and 5.0, respectively, and no significant difference ($p > 0.440$) was returned from a Kruskal-Wallis test.

A summary of the MPs by shape and colour is given in Table 1. Fibres comprised 90% of MPs and were represented by all colour categories but with black-grey the most abundant. Films and fragments contributed about 2% each to the total MP pool but while white-transparent and blue-green films and/or fragments were only detected in rural samples, most black-grey films and fragments were encountered in the urban setting. Significantly, all 16 spherules identified were black and originated from the rural sample in the vicinity of a thermal power station (R5).

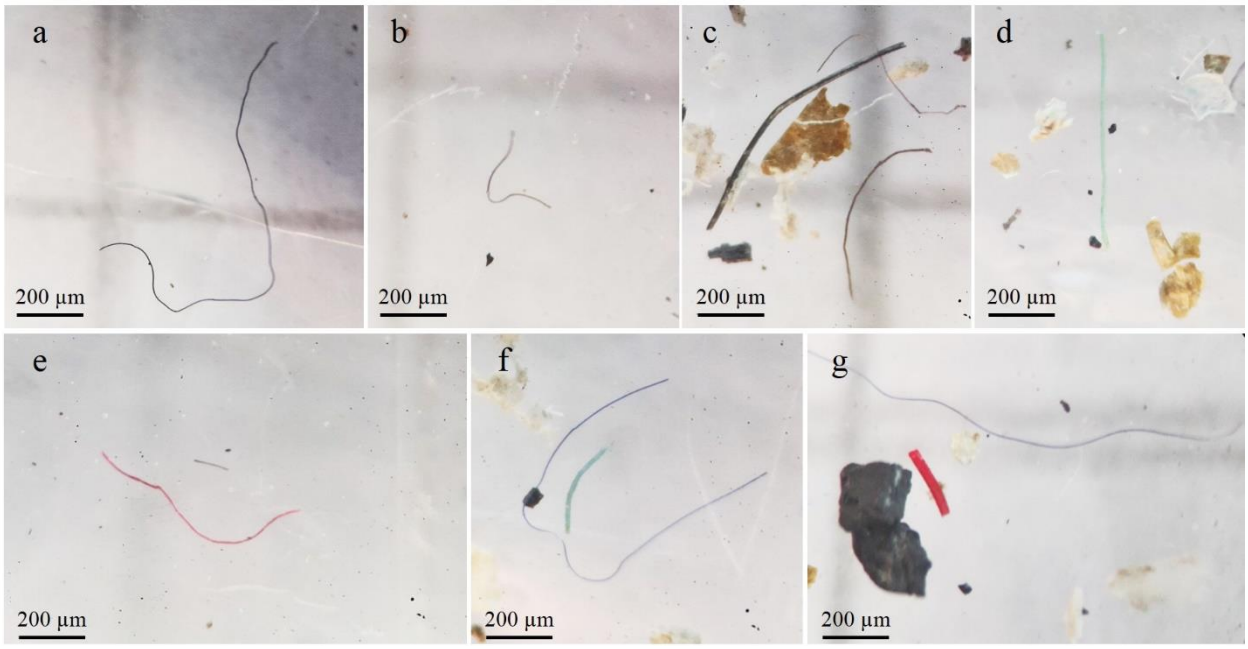


Figure 3: A selection of fibrous MPs in various snow samples identified under the microscope. Note the presence of biological debris in (c) and (d) and a black rubber particle in (g).

Table 1: Distribution of the MPs identified in the snow samples, including those below the surface, according to shape and colour.

	white-transp.	yellow-orange	red-pink	black-grey	blue-green
fibres (*n* = 312)	36	11	50	128	87
films (*n* = 11)	1	0	0	10	0
fragments (*n* = 9)	6	0	0	2	1
spherules (*n* = 16)	0	0	0	16	0
total (*n* = 348)	43	11	50	156	88

Figure 4 shows the size distribution of the MPs in the urban and remote snow samples, along with the frequency of non-fibrous particles in each category. The highest numbers of MPs occur in the 250 to 500 µm fraction for urban snow and the < 100 µm fraction for remote snow. Only one film-fragment was observed in the remote snow samples (or 1.2% of total MPs), while the frequency

of the 20 films and fragments in urban snows (or 7.4% of total MPs) is weighted towards the finer particle fractions. Note that the spherical particles referred to above were all below 100 μm in length.

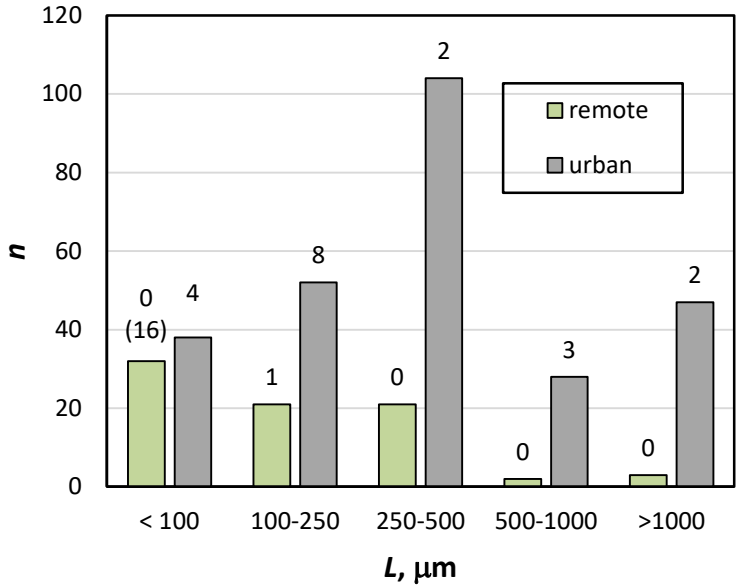


Figure 4: Distribution of MPs by size in the urban and remote snow samples, including those below the surface. Numbers above the bars correspond to the number of films and fragments in each category, and the single number in parentheses refers to the number of spherules observed at a single location.

Figure 5 illustrates the distribution of MPs according to polymer type identified by Raman spectroscopy. Here, and of the 47 samples analysed, 10 could not be matched with reference libraries and 13 polymer types were identified. Most samples were petroleum-based thermoplastics, but one was an elastomer (silicone), one was a natural polymer (silk) and seven

were cellulose-based. Since plant material is generally destroyed by H_2O_2 (Prato et al., 2019), we infer that the cellulose polymers were synthetic in origin.

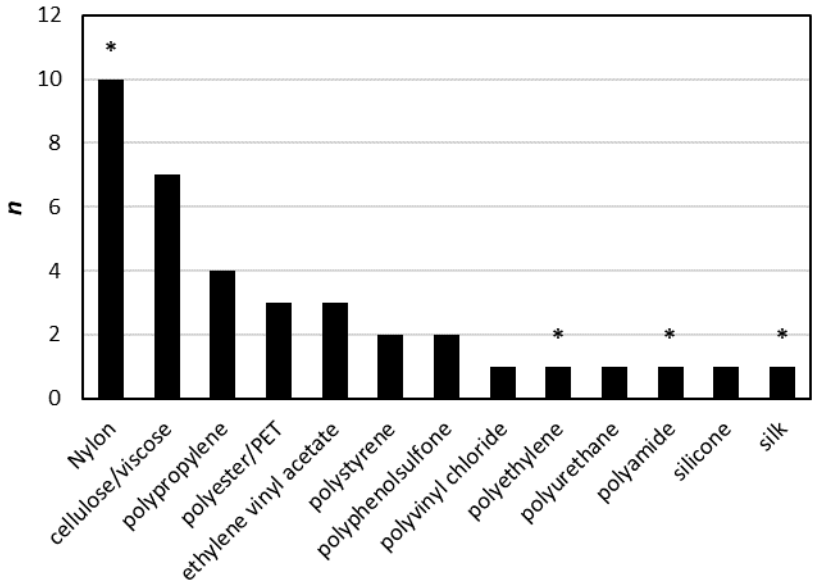
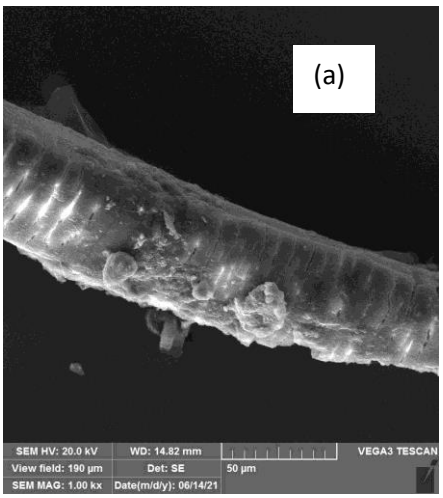
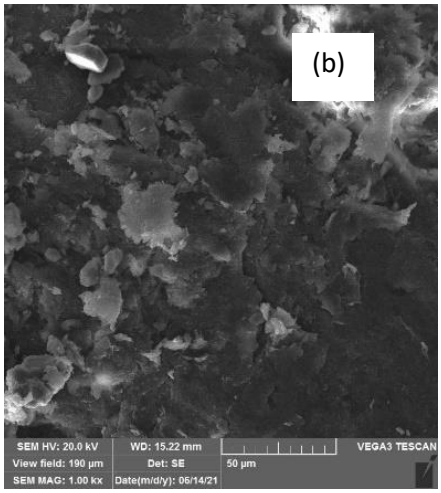


Figure 5: Distribution of MPs (*n* = 37) by polymer type. Asterisks denote that non-fibrous MPs (films and/or fragments) were amongst that category.


(a)
SEM HV: 20.0 kV
WD: 14.82 mm
VEGA3 TESCAN
View field: 190 µm
Det: SE
50 µm
SEM MAG: 1.00 kx
Date(m/d/y): 06/14/21


(b)
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VEGA3 TESCAN
View field: 190 µm
Det: SE
50 µm
SEM MAG: 1.00 kx
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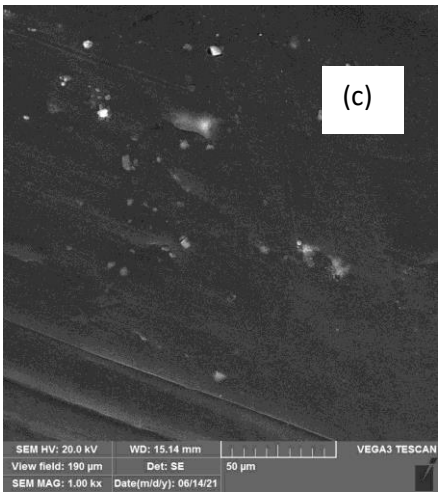
(c)
SEM HV: 20.0 kV
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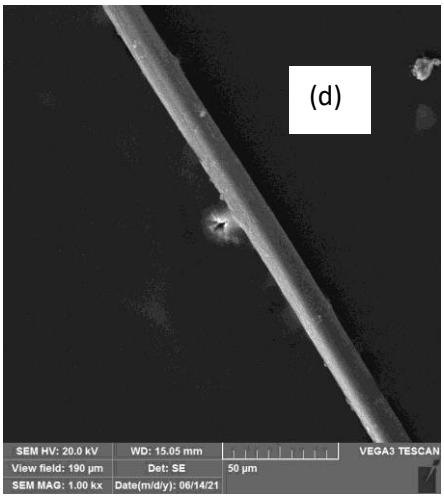
VEGA3 TESCAN
View field: 190 µm
Det: SE
50 µm
SEM MAG: 1.00 kx
Date(m/d/y): 06/14/21


(d)
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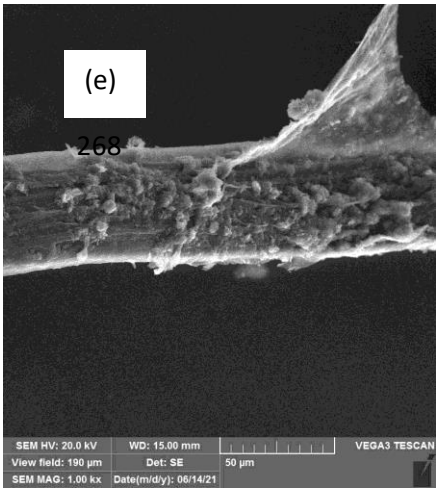
VEGA3 TESCAN
View field: 190 µm
Det: SE
50 µm
SEM MAG: 1.00 kx
Date(m/d/y): 06/14/21


(e)
SEM HV: 20.0 kV
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VEGA3 TESCAN
View field: 190 µm
Det: SE
50 µm
SEM MAG: 1.00 kx
Date(m/d/y): 06/14/21

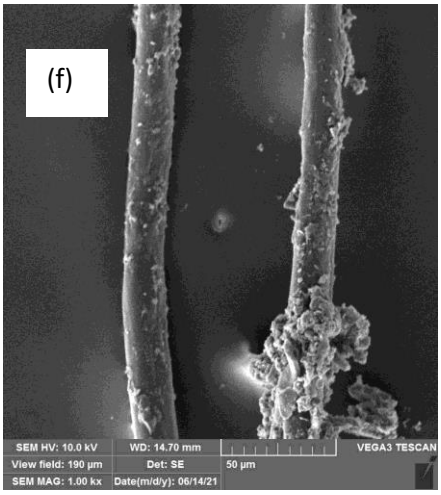
(f)
SEM HV: 10.0 kV
WD: 14.70 mm
VEGA3 TESCAN
View field: 190 µm
Det: SE
50 µm
SEM MAG: 1.00 kx
Date(m/d/y): 06/14/21

Figure 6: SEM images of selected MPs from the snow samples. (a) A blue, polyphenolsulfone fibre, (b) a white, polyethylene film, (c) a white Nylon film, (d) a red, polypropylene fibre, (e) a red, cellulose-based fibre, (f) a transparent, Nylon fibre.

SEM images of fibrous and non-fibrous MPs are exemplified in Figure 6. Some samples exhibit smooth surfaces (e.g. Figures 6c and 6d), with EDX detecting only C and O. Other samples, however, exhibit different degrees of weathering and photo-oxidation (flaking, cracking and fracturing) both at the surface and below the surface. Contamination is also visible, with EDAX revealing particulates composed of combinations of two or more of Al, Ca, Mg, Si and Ti. The scale bars on these and other images indicate that the diameters of fibres ranged from about 5 to 50 μm.

In contrast, the distilled water capturing dry deposition for a 12-h period following the precipitation event returned just one MP. This was a black fibre between 100 and 250 μm in length but that was not characterised by Raman spectroscopy or SEM.

4. Discussion

Despite the atmospheric transportation of MPs receiving considerable attention over the past few years (Dris et al., 2017; Allen et al., 2019; Klein and Fischer, 2019; Brahney et al., 2020; Evangeliou et al., 2020; Roblin et al., 2020; González-Pleiter et al., 2021; Huang et al., 2021; Wang et al., 2021), there is very little published information on the role of snow in capturing, depositing and preserving airborne MPs (Bergmann et al., 2019; Huntington et al., 2020; Parolini et al., 2021). The present study is the first to compare MPs in freshly fallen snow from the same

meteorological event in urban and remote areas, and the first to examine MP distributions with depth in snow deposits.

For continuous deposition, a constant wind speed and direction and no post-depositional MP migration in snow, it would be reasonable to expect an increase in MP abundance with snow depth as MPs are increasingly washed (or "snowed") out of the atmosphere over the period of precipitation. That our results only conform to this distribution in one case suggests that meteorological conditions and snow intensity may have been variable over the period of deposition, or that surface snow, through its roughness and porosity, acted as an accumulator of MPs through dry deposition shortly after the precipitation event. While the latter explanation is possible, the capture of only MP during the 12-h period following the precipitation event suggests that dry deposition was limited, possibly because snowfall acted to "clear" the atmosphere of suspended MPs.

There were no clear differences in the abundance or characteristics of MPs in snow samples between sites or between the different sample location types with two exceptions. Thus, firstly, the regular, black spherules observed at the sampling site close to a thermal power station suggests that combustion products were present that evaded destruction by peroxidation. Secondly, a greater fraction of grey-black fragments in the urban snows is consistent with greater capture or contamination by rubber tire-wear particles. The visual characteristics of these particles were often distinctly different from other MPs, being more elastic and less reflective.

SEM images indicated the presence of both "fresh", unweathered MPs and "aged" and significantly degraded and contaminated MPs amongst the snow samples. This suggests that sources of MPs in the urban and remote settings may be both local and recent, and distal and older. For instance, the stress fractures evident in the sample illustrated in Figure 6a are similar to those

observed in MPs retrieved from the Lut and Kavir deserts (Abbasi et al., 2021), suggesting exposure to the harsh conditions of the arid regions to the south and southwest (Figure 2), while the tire-wear particles noted above appear to have more local origins in the urban setting. Such a variety of sources and histories, coupled with variations in terrain, population density, traffic intensity, turbulence and altitude between the sites, results in a heterogeneous assortment of MPs across the region sampled.

Overall, and within the constraints and operationalities of our methods, the mean and median concentrations of MP in snow samples were about 20 and 12 MP L^{-1}, respectively, with no MPs evident in some samples and a maximum concentration of 86 MP L^{-1}. These concentrations are substantially lower than the mean values reported by Bergmann et al. (2019) for fresh European snow and Arctic snow of unknown age (about 2500 and 1800 MP L^{-1}, respectively). Here, however, particles were examined directly and without chemical separation by μ-FTIR spectroscopy and the authors classified both synthetic and natural fibres and a variety of rubbers as plastic. Moreover, about 80% of the MPs detected by their μ-FTIR were less than 25 μm in size, which is below the detection limit of our microscopic approach (about 30 μm).

More comparable with our study, Parolini et al. (2021) isolated MPs from subsurface snow in the Italian Alps by flotation in NaCl solution (density = 1.2 g cm^{-3}) and identified particles microscopically. The authors report a mean concentration of 2.3 MP L^{-1}, with fibres the most important type of MP but fragments and spherules also present. Pastorini et al. (2021) analysed three snow samples from the Italian Alps by μ-FTIR after staining with Rose-Bengal and only detected one MP fibre in total (equivalent to 1 MP L^{-1}). Napper et al. (2020) analysed fresh snow deposited on Mount Everest and found that concentrations identified by μ-FTIR were in the range 3 to 119 MP L^{-1} (mean = 30 MP L^{-1}). The majority of samples were fibrous and the most abundant

polymer was polyester, and while the authors suggested that tourists and climbers could make a significant, direct contribution to the pool of MPs, the sampling strategy adopted is more consistent with the collection of material deposited from the atmosphere.

An additional, useful comparison with the present results is with concentrations and characteristics of MPs in monthly precipitation (but mainly rainwater) sampled from a remote, elevated region of southern Iran (Mount Derak) and where plastics were defined, identified and quantified by precisely the same techniques (Abbasi and Turner, 2021). Here, and excluding one sample where the volume of precipitation was exceptionally small, concentrations ranged from 30 to 90 MP L^{-1} (mean = 66 MP L^{-1}; median = 60 MP L^{-1}). Concentrations here reflect multiple events and may have been augmented by intermittent periods of dry deposition but the overall measures of central tendency are not that dissimilar to those observed in the snow samples from northern Iran. It is also worth noting that all MPs observed in precipitation samples from southern Iran were fibrous, that the dominant polymers were polypropylene, polystyrene and polyethylene, and that no samples were encountered in the largest size fraction ($\geq$ 1000 μm). In our snow samples, however, there was a greater diversity of shapes, sizes and polymer types (with different densities), with nylon and cellulose-based materials most abundant. These comparisons suggest that the amount of MPs deposited by rainfall and snow may be similar, but that the latter, perhaps because of the greater size, lower density and slower terminal velocity of snow particles relative to raindrops, is able to capture a greater variety and size distribution of MPs. Consistent with these observations, snow is known to be a better scavenger of coarse (> 4 μm) aerosols than rainfall (Zhao et al., 2015). A greater scavenging capacity also renders snow as a more effective, as well as a simpler, synoptic indicator of atmospheric MP contamination than rainfall, and may reduce any confounding effects on precipitation fluxes associated with concurrent or intermittent dry deposition.

On melting of snow, MPs, along with other particulates and pollutants, will be carried into surface waters via runoff on impervious surfaces or into soil and groundwater and other vegetated surfaces (Zhu et al., 2012; Chen et al., 2018). However, it has also been suggested that MPs deposited with or after snowfall may accelerate the melting process itself (Ming and Wang, 2021). Here, coloured MPs, and in particular dark particles that have undergone weathering, may increase the absorption of solar radiation (or reduce the albedo) of fresh deposits of snow. More generally, this raises the possibility that MPs exert an impact on the Earth's radiative balance and climate.

5. Conclusions

This study has quantified and characterised MPs in fresh snow from a single precipitation event over a broad region of northern Iran. Concentrations of MPs were variable amongst samples and ranged from undetected to 86 MP per L of melted snow. There was no statistical difference in MP concentrations with depth of snow sampled or between urban and remote settings, with MPs reflecting a heterogeneous assortment of (mainly) fibres, fragments and films of different colours, sizes, polymer makeup and degrees of weathering. MPs appeared to be derived from both local and distal sources, with meteorological conditions and HYSPLIT modelling suggesting that MPs from the latter were brought into the area with northerly and southwesterly airflows. There appears to be a greater diversity of MPs in snow compared with rain sampled in southern Iran using the same protocols, and this may be attributed to the greater size and slower terminal velocities of snowflakes relative to raindrops. More studies are required to evaluate the mechanisms and global significance of snowfall in the capture and deposition of MPs from the atmosphere to soils and surface waters.

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