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Snow dumping station – A considerable source of tyre wear, microplastics, and heavy metal pollution

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ABSTRACT

Snow dumping stations can be a hotspots for pollutants to water resources. However, little is known about the amount of microplastics including tyre wear particles transported this way. This study investigated microplastics and metals in snow from four snow dumping stations in Riga, Latvia, a remote site (Gauja National Park), and a roof top in Riga. Microplastics other than tyre wear particles were identified with Attenuated Total Reflection Fourier Transform Infrared (ATR-FTIR) (>500 μm) and focal plane array based micro-Fourier Transform Infrared (FPA-μFTIR) imaging (10–500 μm), tyre wear particles by Pyrolysis Gas Chromatography-Mass Spectroscopy (Py-GC–MS), and total metals by Inductively Coupled Plasma with Optical Emission Spectroscopy (ICP-OES). Microplastics detected by FTIR were quantified by particle counts and their mass estimated, while tyre wear particles were quantified by mass. The concentrations varied substantially, with the highest levels in the urban areas. Microplastic concentrations measured by FTIR ranged between 26 and 2549 counts L^{-1} of melted snow with a corresponding estimated mass of 19–573 μg/L. Tyre wear particles were not detected at the two reference sites, while other sites held 44–3026 μg/L. Metal concentrations varied several orders of magnitude with for example sodium in the range 0.45–819.54 mg/L and cadmium in the range 0.05–0.94 μg/L. Correlating microplastic measured by FTIR to metal content showed a weak to moderate correlation. Tyre wear particles, however, correlated strongly to many of the metals. The study showed that snow can hold considerable amounts of these pollutants, which upon melting and release of the meltwater to the aquatic environment could impact receiving waters.

1. Introduction

Plastic use has increased substantially (Li and Zhang, 2021) due to its distinctive benefits over other materials, and the production has exceeded most other man-made materials, for instance, concrete, cement, and steel (Geyer et al., 2017; Ivleva et al., 2017; Li et al., 2018). The applications range from packaging materials (Chen et al., 2021) to highly engineered plastic components, for example, in aircrafts (Fette et al., 2015). However, inappropriate waste management has caused plastic to be a persistent pollutant, threating ecosystems, organisms, and even human beings (Galloway, 2015; Li and Zhang, 2021). Once plastic litter enters the natural environment, it starts to degrade and fragment into smaller pieces (Browne et al., 2011; Ragusa et al., 2021). The degradation of plastic can facilitate the release of additives, which then impact the nearby surroundings and organisms (Teuten et al., 2009). The additives associated to plastics (Chen et al., 2022) can interact chemically and may disrupt the endocrine system of living organisms (Ivleva et al., 2017; Teuten et al., 2009). In the case of small plastic fragments, they can easily disperse and be transported to remote locations far away from the source (K. Liu et al., 2019; Üstün et al., 2022).

The general consensus is that synthetic polymers between 1 and 5000 μm in their longest dimension are called microplastics (Hartmann et al., 2019). Some microplastics, commonly called primary microplastics, are produced to this size for various commercial purposes such as cosmetics. Other microplastics, commonly called secondary microplastics, are formed by fragmentation of large plastic debris due to physical and chemical processes. Microplastics have been investigated in all environmental compartments (Dalu et al., 2021; Ivleva et al., 2017; Lusher et al., 2017; Zhang et al., 2020) to understand the environmental and ecological risk associated to them. Examples are marine

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environments (Alimba and Faggio, 2019; Alimi et al., 2018; Liu et al., 2022; M. (Shabnam) Molazadeh et al., 2023), freshwater environments (Alimi et al., 2018; Eerkes-Medrano et al., 2015; Koelmans et al., 2019; Rasmussen et al., 2024; Molazadeh et al., 2024), and technical systems such as wastewater treatment plants (Browne et al., 2011; Cheung and Fok, 2017; Kazour et al., 2019). Other compartments have received less focus, here among the cryosphere including freshly fallen snow (Bergmann et al., 2019; Zhang et al., 2020; 2022). One microplastic type has been studied significantly less than others, namely particles of tyre wear (TWP). It has, though, received increasing attention over the later years. Most studies report that it tends to end up in soil (Giechaskiel et al., 2024), even though some is also found in remote areas (Goßmann et al., 2023). The reason for the comparatively few studies is that the most common analytical techniques, FTIR and Raman spectroscopy, struggle to identify TWP due to the content of carbon black (Järlskog et al., 2022; 2020). Here thermal decomposition, mainly pyrolysis GC–MS, is used instead. TWP contains many toxic additives and has been reported to cause detrimental effects in the environment (Wik and Dave, 2009), for example on aquatic organisms, such as, algae, crustaceans, and fish (Marwood et al., 2011).

Microplastics can be transported in the atmosphere over long distances and have been found in pristine terrestrial and oceanic environments, as well as high altitude locations (Wagner et al., 2018; Zhang et al., 2021). According to X. Wang et al. (2020), microplastics can be conveyed over distances as far as 1000 km. They can also be transferred from the ocean surface to the atmosphere (Allen et al., 2020; Goßmann et al., 2023). Atmospheric conditions, such as wind direction, wind speed, and precipitation could have a significant impact on the transport of these micropollutants (Allen et al., 2019; Roblin et al., 2020). The inherent properties of microplastics such as small mass, small size, and irregular shape could enhance the transport and distribution pattern (Li et al., 2018), and atmospheric transport and deposition hereof has gained increasing interest (Cai et al., 2017; Ding et al., 2021; Dris et al., 2015). Different levels of microplastic contamination have been reported at rural versus urban sites when analysing wet deposition (rainfall and snowfall) versus dry deposition (Klein and Fischer, 2019; Wright et al., 2020). Precipitation is important for conveying airborne particles to terrestrial and aquatic surfaces, and are likely also able to convey microplastics, and microplastic concentrations have been found to show moderate to strong correlation with both rainfall and snowfall events and intensities (Allen et al., 2019). Furthermore, significant amounts of microplastic fibres and particles have been identified in rainwater and snow (Abbasi et al., 2022; Bergmann et al., 2019; Dris et al., 2016; Roblin et al., 2020).

Microplastics interact with other pollutants such as metals and organic micropollutants (Sun et al., 2019; Tamis et al., 2021). Many of these are persistent (Li et al., 2022; Q. Wang et al., 2020) and can be detrimental to organisms and humans (Jaishankar et al., 2014). During aging in the environment, the adsorption capacity of microplastics can increase due to an increase in specific surface area and oxygen-containing functional groups (Liu et al., 2021; Wang et al., 2019). The oxygen-containing functional groups of microplastics are more prone to react actively with charged heavy metals and other pollutants. Likewise, other chemical and physical factors such as pH, temperature, dissolved organic matter and electrostatic interaction also have significant impact on the adsorption capacity of microplastics (Guo et al., 2020; Li et al., 2022).

In regions with prolonged cold temperature, snow can be subjected to atmospherically transported microplastics. Such snow is accumulated and afterwards moved to snow dumps till it melts when the weather gets warmer. We hypothesize that such dumps could be hotspots for microplastics and other pollutants, affecting the waters to which the snowmelt discharges. The pollutant level could depend on the land use, i.e. commercial areas, residential areas, urban centres, and rural areas. The objective of the present work is to evaluate this hypothesis by analysing microplastics in snow from snow dumps in the urban centre and residential areas of Riga, Latvia, and compare it to snow from sites less affected by human activity, in this case, a National Park. This study aims to understand the contamination level based on land use practices and human activities. It applied μFTIR imaging to investigate microplastics in terms of counts, size, and material types, and estimated their mass from the hyperspectral image. As TWPs cannot be identified this way, it used pyrolysis GC–MS for this microplastic fraction and used ICP-OES to quantify metals.

2. Materials and method

2.1. Study sites and snow sampling

Riga, the capital of Latvia, is covered by snow a quarter of the year. To support the city traffic, the urban snow is collected in the city and dumped at specific dump sites. Snow was collected from five sites in the city in December 2021, four of which were in the inner city: A central market (C1); a square in the old town with controlled car access (C2); a 65 m high rooftop of an academic science building (C3); and a parking lot of a shopping mall (C4). One residential site was chosen in a yard with limited car access (R1); and one site was chosen outside of the city national park, the Gauja National Park (P1) 50 km from Riga (Fig. 1, Table 1). Snow was taken to approximately the same thickness from all sites, which was 15–20 cm from the surface. Triplicates were taken from each site in 20 L metal buckets with a metal shovel, both pre-cleaned with distilled ultrapure water. In addition, a separate sample was taken in a plastic canister for metal analysis.

2.2. Sample preparation

2.2.1. Microplastic extraction

Upon arrival at the lab, the snow samples were kept at room temperature (22–23 °C) until melted, measured for volume, and filtered through a 10 μm stainless-steel filter (Ø167 mm) (Table 1). The cut-off size (10 μm) was restricted by the limit of detection (LOD) of the analytical method for the common polymers (section 2.3.1.2). This cutoff was also applied to TWP, even though the analytical method used for its detection (Pyrolysis Gas Chromatography-Mass Spectrometry, Py-GC–MS) in principle has no lower particle size detection limit, with the attempt to avoid sample splitting during extraction. Triplicate samples were collected and treated separately for each sampling site. Microplastics were isolated via a multi-step treatment (Rasmussen L.A. et al., 2023; Chand et al., 2021), in short: Density separation ($ZnCl_2$, 1.78 g cm^{-3}), enzymatic digestion (protease and cellulase, Sigma-Aldrich), catalysed oxidation (Fenton reaction), and a final density separation. The details of the procedure are in the supplementary information (section S 1.1). The extracts were then fractionated into two size classes: 10–500 μm, and ≥ 500 μm. The small size fraction was concentrated in a 10 mL glass vial with a known amount of 50 % ethanol (HPLC grade), whilst the large fraction was dried at 50 °C and stored in an aluminium tray until analysis. Field blanks (section 2.4) were collected in 5 mL 50 % ethanol.

2.3. Analysis of pollutants

2.3.1. Microplastics

The microplastic mass estimated by Focal Plane Array based Micro-Fourier Transform Infrared imaging (FPA-μFTIR) is not necessarily comparable to the mass determined by Py-GC–MS (Primpke et al., 2020b; Kirstein et al., 2020).. Furthermore, particle counts could only be obtained by FPA-μFTIR. To distinguish the two groups of microplastics, the abbreviation MP is used for microplastics determined by ATR-FTIR and FPA-μFTIR, while TWP is used for tyre wear microplastics.

2.3.1.1. Large fraction (≥500 μm). For the large size fraction, particles

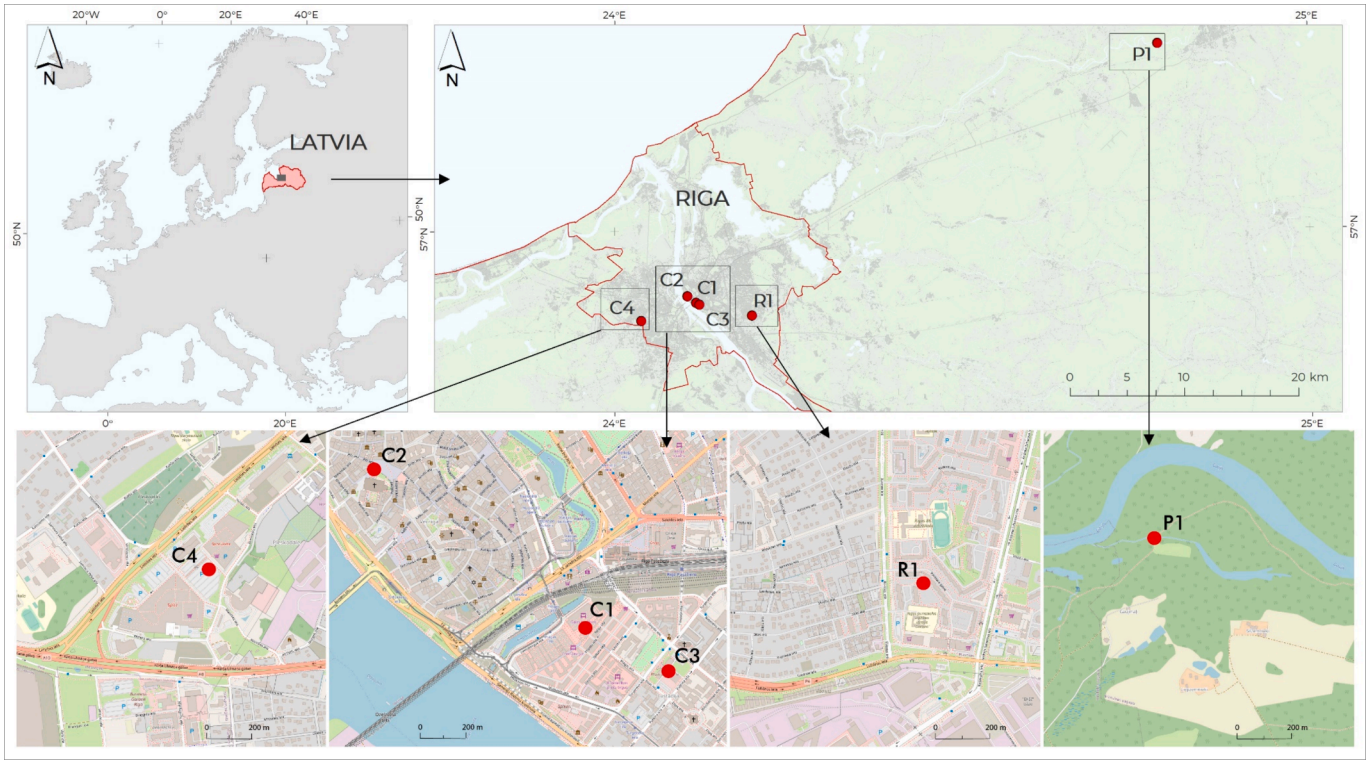

Fig. 1. Sampling locations. C1: Riga central market, C2: Old town centre (Doma Church square), C3: Latvian academy of science rooftop (65 m high), C4: Shopping mall "Spice home" parking lot, R1: Residential area (Plavnieki), and P1: Gauja National Park (appr. 50 km away from Riga city centre).

Table 1
Detail about the sampling sites, longitudinal and latitudinal position, and weather parameters and total volume of sampled snow.

Sampling sites	Land use	Longitude (E)	Latitude (N)	Humidity (%)	Wind speed (m/s) and direction	Temperature during sampling (°C)	Volume (L) of the melted snow of triplicate samples
C1	Central market and core business centre of Riga	24.11713°	56.94447°	55	1.3 Southeast	−7.1	i) 8.7 ii) 9.3 iii) 8 average: 8.7 total: 26
C2	Square in the old town with visitors throughout the year and controlled car access	24.10480°	56.94954°	32.9	2.3 South	−6.5	i) 9.1 ii)7.6 iii) 8.4 average: 8.4 total:25.1
C3	65 m high rooftop of the Latvian Academy of Science building	24.12207°	56.94298°	58.4	4.8 Southeast	−10.1	i) 6 ii) 6.1 iii) 5.8 average: 6.0 total:17.9
C4	Parking lot of a mall (Spice home) with 2000 parking spaces	24.03856°	56.93029°	51.6	1.0 South	−7.0	i) 11.5 ii) 10.9 iii) 11.7 average:11.4 total: 34.1
R1	Yard in a residential area with limited car access	24.19748°	56.93437°	52.8	1.5 South	−8.5	i) 7.7 ii) 7.9 iii) 7.9 average: 7.8 total:
P1	Gauja National Park, appr. 50 km from the city centre of Riga	24.78398°	57.14593°	57.7	0.0 South	−2.2	i) 7.1 ii) 7.3 iii) 7.1 average: 7.2 total:

were firstly characterised under a stereomicroscope (ZEISS, SteREO Discovery.V8) by measuring the surface area, circularity, major and minor dimension (software Zen2Core SP1), then analysed by Attenuated Total Reflectance-Fourier transform infrared (ATR-FTIR) spectroscopy for chemical composition (Agilent Cary 630 FTIR with a diamond ATR) Chand et al. (2022).

2.3.1.2. Small fraction (10–500 μm). For the small size fraction (10–500 μm), an aliquot was deposited from the concentrates onto a zinc selenide window after thoroughly vertexing the concentrates as described in (Chand et al., (2022,2021) and Simon-Sánchez et al. (2022). The window had an effective area of Ø10 mm and a thickness of 2 mm. For each sample, three windows were prepared by sequentially depositing aliquots to a minimum of 100 μl of 10 mL to 1 mL of 5 mL of concentrate, thus, to ensure a sufficiently populated window for analysis. Windows were dried at 50 °C between aliquoting. The deposited amount depended upon the particle content of the samples. The identification of MPs was performed by FPA-μFTIR (Agilent Cary 620 FTIR microscope equipped with a 128-pixel FPA and coupled with a Cary 670 IR spectroscope), resulting in a pixel resolution of 5.5 μm. Details of the instrumental settings are found in Simon et al. (2018), Vianello et al. (2019) and Chand et al. (2022).

2.3.1.3. TWP. The content of TWPs was measured by Py-GC–MS by subsampling 25–1000 μL from the concentrates using a glass capillary, then depositing it to a sample cup and drying it on a heating plate at 50 °C. In this way, TWP shared the same cutoff size with MPs, which was 10 μm. The Py-GC–MS system comprised a micro furnace pyrolyzer EGA Py-3030D (FrontierLab, Japan), an autoshot sampler AS-1020E (FrontierLab, Japan), a Thermo Scientific TRACE 1310 GC, and an ISQ™ single quadrupole GC–MS with helium as the carrier gas. The analysis employed a solution of 0.2 g/L deuterated anthracene (A-d10) in n-hexane as an internal standard. The Py-GC–MS method involved pyrolysis at 600 °C with an interface temperature of 280 °C. The GC was operated with helium gas as the carrier (1 mL min^{-1}) in split mode 30:1. The temperature program ranged from 40 °C (2 min) to 300 °C (5 min), with a temperature gradient of 10.5 °C min^{-1}. The MS was operated in EI positive mode (70 eV, *m/z* range 35–500), while the transfer line and ion source were maintained at 250 °C and 200 °C, respectively (Molazadeh et al., 2023; Rasmussen et al., 2024). The TWP concentration was assessed by examining extracted ion 54 chromatograms for 4-vinylcyclohexene, a well-known indicator of styrene butadiene rubber and butadiene rubber (Goßmann et al., 2021). The same marker was also used by More et al., 2023 and Mun et al., 2023. The quantification process involved utilizing an external calibration curve composed of a standard tire mixture obtained from Genan, Denmark. This company specializes in recycling Danish and Swedish tires. Genan acquires approximately 85 % of all used tires in Denmark, ensuring that their product for calibration is representative of the types of tires commonly in use in the region where the samples were from. Moreover, their tire mixture is homogenized during the manufacturing, contributing to the consistency of the calibration process. This choice of calibration standard not only reflects the diversity of tire compositions but also accounts for variations in tire makeup.

2.3.2. Metals

For each sampling site, 50 mL melted snow was mixed with 1 mL nitric acid (SupraPur quality, SCP Science, Quebec, Canada) and analysed for aluminium (Al), calcium (Ca), cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), iron (Fe), potassium (K), magnesium (Mg), manganese (Mn), sodium (Na), nickel (Ni), lead (Pb), and zinc (Zn) by Inductively Coupled Plasma – Atomic Emission Spectroscopy (ICP-AES). The choice of these metals was based on their wide occurrence in various environmental matrixes, including fresh snows (Veysseyre et al., 2001). The spectrometer was a Thermo iCap 6300 Duo (Thermo Fischer Scientific, Waltham, MA, USA) operated in axial view mode. Concentrations of each element were calculated as total metal, i.e., the sum of their various forms, for instance, ions, organic bound, and precipitated metals. The sample was introduced by a concentric glass nebulizer attached to a glass cyclonic spray chamber with a rate of 2 mL min^{-1}. The RF source was set at 1150 W and the plasma and auxiliary gas flows were 2 L min^{-1} and 12 L min^{-1}, respectively. The nebulizer gas flow was pressure controlled at 0.2 MPa. A series of matrix matched multi-element standards, ranging from 10–1000 mg/L were used for calibration. Each element was quantified by selecting one to five wavelengths. 1000 mg/L yttrium solution was added to each sample (0.2 mL min^{-1}) as the internal standards (reference), which was reflected on the wavelength of 360.073 nm. Over-range samples were diluted from 10 to 1000 times using a matrix matched solution (type I ultra-pure water (ELGA LabWater, High Wycombe, UK) and nitric acid) until the signal was within the range of the calibration standards.

2.4. Quality control

Measures were taken during sampling and in the lab to minimize contamination. Only non-plastic tools were used during sampling, and only cotton lab coats were allowed in the lab. All sample processing was done in a clean fume hood or a laminar flow bench (TelstarAV-100). The air in the lab was continuously filtered with a Dustbox® (Hochleistungsluftreininger, Germany) holding a HEPA filter (H14, 7.5 m^2). All reagents were filtered through 0.7 μm GF filters (Whatman). All glass wares and filters were muffled at 500 °C, or thoroughly cleaned with filtered demineralized water.

One field blank was collected for each sampling site by opening a clean petri dish (Ø167 mm) when the sampling started and closing it when it ended. The particles collected in the petri dish were concentrated into 5 mL of 50 % ethanol (HPLC grade), upon which approx. 20 % of the concentrate was analysed by FPA-μFTIR.

2.5. Data analysis

The IR spectra from ATR-FTIR were analysed by OMNIC (Thermo Fisher Scientific Inc., 8.2.0.387 version 1) for chemical identification. IR images from FPA-μFTIR were analysed by siMPle (previously called MPhunter, F. Liu et al., 2019). It compares the IR-spectrum of each pixel with a custom-made library, containing more than 450 spectra of natural and synthetic materials. siMPle provides particle dimensions, 2D projected areas, estimated volume, and estimated mass (Primpke et al., 2020; F. Liu et al., 2019). The major dimension of the particle was reported as the particle size and only particles consisting of at least two pixels were considered as MPs. Data analysis and visualization was performed using R (4.2.2) with package ggplot2. To assess whether there were significant differences in the size and mass of MPs among the sites, Kruskal-Wallis test was applied. One-way ANOVA test was performed to assess whether polymer composition varied significantly between sites. The significant level was in all cases set to 0.05. The location map was produced using ArcMap 10.3.

3. Results and discussion

3.1. Contaminations

For the analysed aliquots, 12 MPs belonging to seven polymer types were detected in five field blanks: five MP particles in C3, three in R1, two in C2, and one in each C4 and C1. No MPs were found in P1. The corresponding total mass of MPs in the blanks was 2692 ng, 1527 ng, 409 ng, 253 ng and 3 ng, respectively. Polyester (PES) and polyvinylchloride (PVC) were the major polymers, each contributing 25 % to the particle counts, followed by PAN acrylic with 17 % contribution. The remaining four types were polyamide (PA), polyacrylamide (PAA), polyurethane (PU), and epoxy-phenoxy resin, each contributing 8 %. Particles found in the blanks could originate from activities during the sampling, where MPs were released to the air then deposited onto the petri dish. The contamination level is site-specific; thus, it is reasonable to interpret the results as counts $site^{-1}$, or mass $site^{-1}$. Taking particle counts as an example, this corresponds to the highest pollution level as 25 counts $site^{-1}$ (C3), followed by 15 (R1), 10 (C2), 5 counts $site^{-1}$ (C1 and C4), and not detectable at P1.

3.2. MP and TWP abundance

3.2.1. MPs

MPs were found in all snow samples. The highest concentration was found in the central market (C1), both in terms of particle counts 2549 (±836) counts L^{-1} and estimated mass 573 (±318) μg L^{-1}, while the lowest was in the national park (P1) with 26 (±23) counts L^{-1} and 19 (±13) μg L^{-1}, closely followed by the rooftop (C3, 88 (±30) counts L^{-1}, 43 (±11) μg L^{-1}) (Fig. 2, Table S 1 and Table S2). Compared with the MP content in field blanks for each specific site, MP concentrations in snow samples were much higher, hence the results were not corrected for contamination. Within the urban environment, Rasmus et al. (2023) found that the stormwater runoff contained 0.3–1200 counts L^{-1} MPs using the same analytical method and cutoff size. This is slightly lower than the MP level found in the snow at the dumping sites, which makes the snow dumping sites a more important hotspot for MP pollution than stormwater runoff.

The high abundance of MPs in snow in populated areas (C1, C2, and C4, Fig. 2A and B) seems reasonable as these sites are close to microplastic sources. Some of the microplastics in the snow dumps may have been caught while the snow was falling, while others might have been caught after it was deposited (Zhao et al., 2015). The snow from the residential area (R1) held lower concentrations than the sites in the dense city (C1, C2, and C4), which may be due to a lower intensity of anthropogenic activity in the residential area. All in all, these results support previous observations that human activities and population density may influence the occurrence of microplastics (Bergmann et al., 2019; Chernykh et al., 2022; Dris et al., 2016).

MPs have also been reported in urban air by analysing of wet and dry deposition (Abbasi et al., 2017; Qiu et al., 2020), but only few studies have reported them in snow. Differences in sampling and analytical methods do however make comparation across studies problematic.

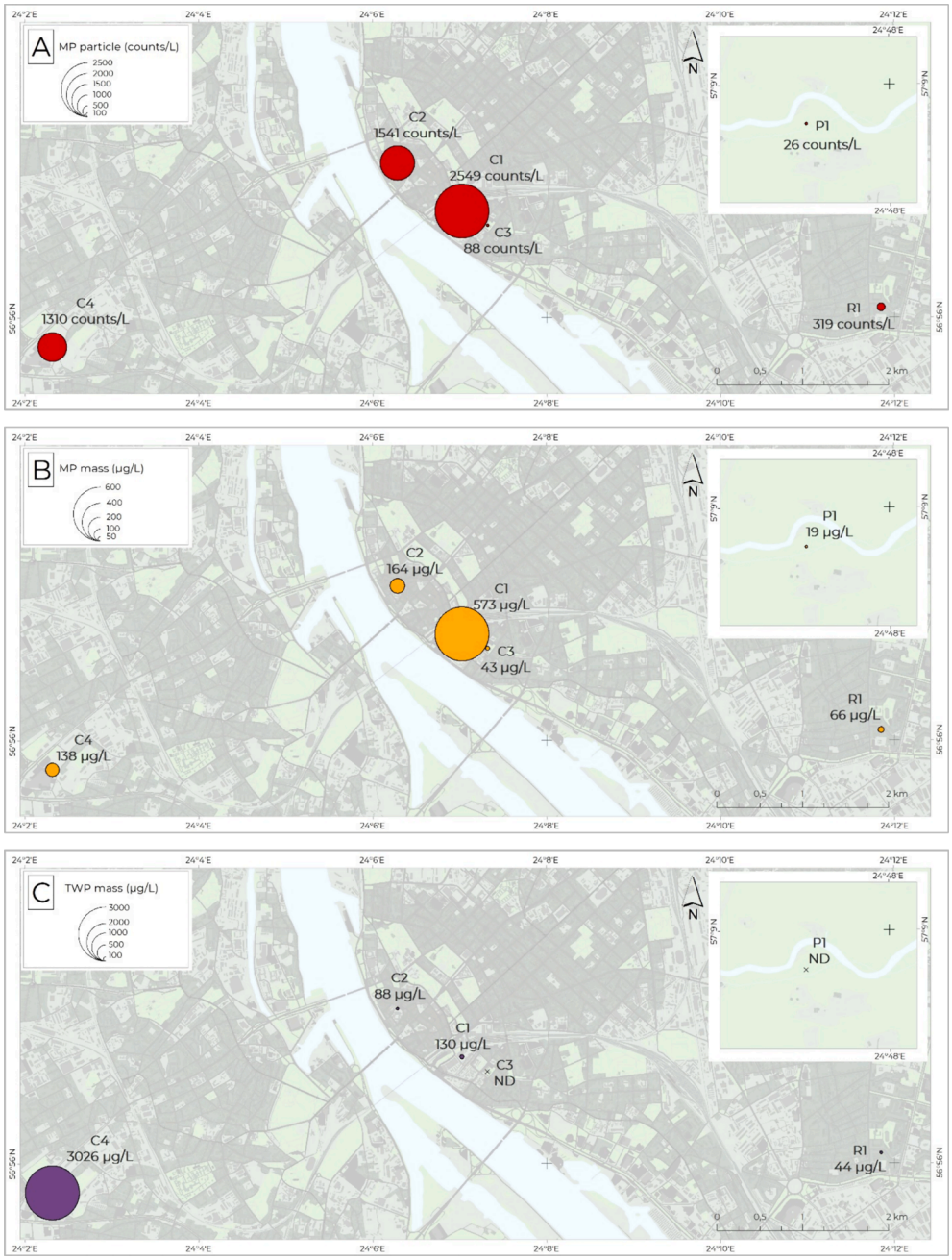

Fig. 2. Concentration: A) MP particle counts, B) MP mass, and C) TWP mass at the sampling locations. ND*: Not detected.

Malygina et al. (2020), for instance, investigated snow from Western Siberia employing visual identification coupled with fluorescent staining for MP identification, restricting the low cut-off size to 100 μm. They did not report concentrations, only that they found both particles and fibres. However, even if they had reported concentrations, such values cannot readily be compared to the current study, which applied a quite different instrumental approach and a lower size cut-off of 11 μm. Applying a similar hyperspectral imaging approach as the current study, and a similar size detection limit, Bergmann et al. (2019) found that snow from the Swiss Alps held significantly lower counts (0 to 14.4 × 10^3 counts L^{-1}) than urban sites from northern Europe (0.19 × 10^3 to 154 × 10^3 counts L^{-1}). The concentration difference between remote and urbanized regions was also notable in the current study, as MPs concentrations in snow from populated areas (C1, C2, C4, and R1) on average was 53 times higher than that of untouched snow in the Gaujas National Park (P1), which evidenced that MPs were less subjected to atmospheric transport.

MPs were also detected in snow from the 65 m high rooftop (C3). It held more than the snow in the national park (P1), but less than the snow on the city ground surfaces. As this site is publicly accessible for tourists, it is not possible to differentiate whether the identified MPs came from the visitors, for instance from synthetic clothing, or were transported to the snow via the atmosphere. The snow at P1 was, however, untouched by direct human activity, indicating that MPs were transported via the atmosphere to this remote area (Baztan et al., 2014; Kelly et al., 2020; Napper et al., 2020).

3.2.2. TWPs

TWPs were detected at four sampling sites (C1, C2, C4, and R1) but not in the snow from the national park (P1) and the rooftop (C3). In the four sites, the concentration ranged from 44 to 3026 μgL^{-1} (Fig. 2, Table S4). The highest abundance was found in snow from the parking lot (C4), while the lowest was in the residential area (R1). A study by Rødland et al. (2022) investigated TWP in snow along the roads in Norway, and reported concentrations 2–4 times higher than the most polluted site of the current study. TWP may be subject to atmospheric transport, as the deposited particles can be resuspended and brought up by airflow (Kole et al., 2017; Sun et al., 2022). However, in this study, TWP was absent in the snows from the rooftop and the national park. The reason could be that wind-driven transport of TWP was less pronounced than wind-driven transport of other microplastics. Another explanation could be that the concentrations were below the detection limit of the Py-GC–MS. All other sites than C4, the parking lot, held lower TWP mass than MP mass. This statement must be interpreted with caution, as the mass by FTIR is an estimation only, made by assuming that the imaged particle can be described by its equivalent ellipsoid, where the third dimension (thickness) equals 0.6 times its second dimension (width) (Simon et al., 2018), whilst the Py-GC–MS yields a direct and quantitative measurement. Hence, the two values are not necessarily comparable.

3.3. Polymer composition, size, and mass

3.3.1. Polymer composition

Twenty polymer types were identified by FTIR in the snows from the six sites. Polyester (PES), polypropylene (PP), and polyethylene (PE) were the most prevalent polymers in the samples. Some polymers were detected at a low frequency (1 % in terms of particle counts), and were lumped into a group termed 'Others' (Fig. 3A). Polymer composition though varied between sites, but with no statistical significance. Nevertheless, PES dominated at all sites except C4, both in terms of particle counts and estimated mass, and at R1 in terms of particle mass (Fig. 3B). Instead, most of MPs identified at C4 were PP, which dominated both as particle counts and mass. At R1, PP was the most abundant polymer in terms of particle mass, probably caused by the PP particles being comparably large.

PES was also a dominant polymer (56 %) in snow from Mount Everest (Napper et al., 2020). Likewise, Aves et al. (2022) found that 41 % of polymers identified in their 19 snow samples were polyethylene terephthalate (PET), which is a common thermoplastic resin of PES. As such PES is one of the polymers in high demand, and has been used extensively for textile filaments (Carr, 2017). This polymer can be shredded from the synthetic clothes and other textiles and end up in the snow. Other common polymers like PA, PE, PS, PU, and PVC were also observed in the snow but at lower frequency. With respect to mass composition, PES constituted over 90 % in both C3 and P1 (Fig. 3B), while PP dominated in R1 and C4 with 70 % and 54 %, respectively. There is little data on MPs in the Riga region, but MPs in adjacent marine areas, for instance the Baltic Sea, North Sea, Kattegat, and southern North Sea, showed a more or less similar trend with a high proportion of PE, PP, PES, and PS (Kirstein et al., 2016; Liu et al., 2022; Lorenz et al., 2019).

3.3.2. Size and mass and characteristics of identified polymers

A total 18,289 MP particles were identified in the size range 11–4427 μm when scanning all the snow samples by μFTIR and the larger particles by ATR-FTIR. The vast majority (17977 counts, 98.3 %) were small MPs (<500 μm) and only 312 particles (1.7 %) were above 500 μm. Considering the major dimensions of the MPs, the snow samples from different sites were statistically significant regarding particle size

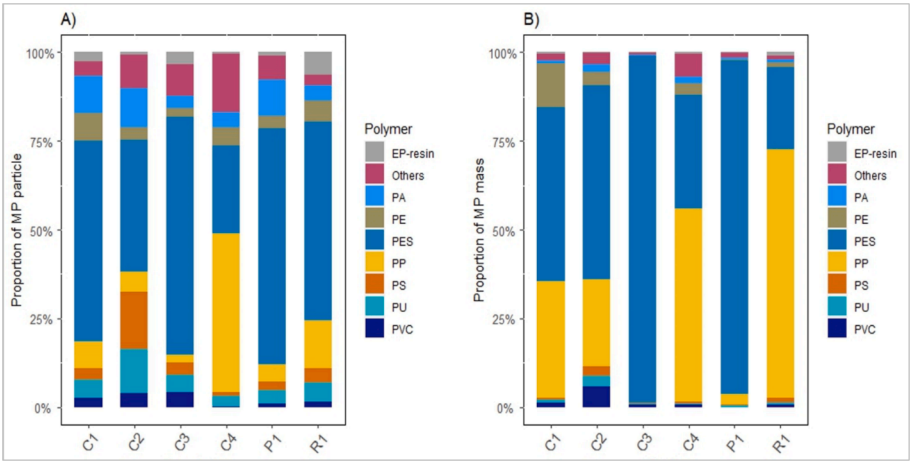

Fig. 3. Distribution of MPs based on: A) particle count and B) estimated mass. Others: ABS, AF Paint, Acrylic Paint, Acrylic, Alkyd, CE, EVA, PAM, PAN, PHB, VCC, and PC.

and mass (Wilcoxon rank sum test P-value < 0.05, Figure S 1A). Likewise, the estimated masses among the sampled snows were also significantly different (Wilcoxon rank sum test P-value < 0.05, Figure S 1B). The mass of individual MPs varied by 8 orders of magnitude, with the smallest particle having an estimated mass of 0.1 ng while the heaviest one weighted 1969 μg. The summary of the measured major dimension and estimated mass is provided in Table S 3 and figure S 3. The small MPs dominated the MP number concentration while the mass was governed by a few large particles, a finding which corresponds to e.g. that of Liu et al. (2022).

The identified MPs were further characterized as fibres and fragments based on their aspect ratio being above or below 3, that is, fibres have a ratio of major to minor dimension above 3 and fragments one below 3. In total, 16,611 (~91 %) of the detected MPs were fragments, while the remaining 1678 (~9%) were fibres. The ranking in terms of fibre content was P1 > C3 > C1 > R1 > C2 > C4 (Figure S 2). This study found a lower proportion of fibres than many other microplastic studies, which might be attributed to the detection of a large number of small MPs when using μFTIR with a low size detection limit combined with autodetection of particles using a dedicated software. For instance, 72 % of all MPs had a major dimension below 100 μm, while this is roughly the lower size limit that can be achieved when manually pre-sorting particles and applying visual detection.

3.4. Metals

The highest metal concentrations were found in snow from C4, except for Na and Pb which were high at C2 and C1, respectively. The high content of Na was likely from road de-icing salts, whilst Pb might be a legacy pollutant from both exhaust and non-exhaust traffic (Ye et al., 2022). The lowest concentrations were in snow from P1, except for Ca, which was lower in snow from C3. Metal concentrations tended to be high in densely populated and urbanized areas (Table 2 and Table S 5). However, the concentrations varied also between urbanised sites. The snow metal content was above that of several previous studies on snow from remote areas (Hur et al., 2007; Planchon et al., 2001), which probably was due to human activity at the studied sites (Hur et al., 2007; Li et al., 2022; Sakai et al., 1988; Sharma, 2014). For instance can Cd, Cr, Ni, Cu, and Zn come from traffic related activities close to the sampling sites (Pacyna et al., 2007). The metals in snow from P1 must, however, have come via long-range transport or originate from the local soils (Slukovskii et al., 2020; Valavanidis et al., 2011).

3.5. Correlation of MPs, TWP, and metals

There was a moderate positive corelation between MP counts and metals (Pearson's coefficient 0.4–0.67) (Fig. 4), while the estimated MP mass was only weakly to moderately correlated to metals (Pearson's coefficient 0.027–0.51). This is in contrast to findings of some laboratory studies reporting a positive correlation between MP concentrations and adsorbed metals (Q. Wang et al., 2020) and to a study on MPs in soil which showed a negative corelation between metals and MPs (Du et al., 2023). The latter could have been due to low organic content in their soil, as organic matter may increase adsorption of metals.

Except for Na, TWP and metal masses were on the other hand well correlated (0.70–0.98). Part of this might be caused by metals associated to tyres and traffic in general (Adamiec et al., 2016), which was particularly true for Mg, Ca, Co, Cu, Ni, and Fe with the Pearson coefficient all above 0.8 (Fig. 4). A study by Adachi and Tainosho (2004) stated that heavy metals in tire dust are not only attributed to TWPs, but also to road and traffic materials, for instance, brake linings and road paint, together enriching the diversity of metals released to the environment. Likewise, most metals were well correlated among themselves (Fig. 4). The exception was again Na, which generally was poorly corelated to other metals, a finding which agrees with a previous study on seasonal patterns of metals in snow (Hur et al., 2007). Whether there was an interaction between TWP, and metals is though not a given, as the co-occurrence of TWP and many metals might simply be due to similarity in sources.

MP counts and mass were, however, not correlated to TWP (Pearson's coefficient 0.2 and 0.0099, respectively). Considering the fact that metals correlated strongly with TWP, but only weakly to moderately with MPs, it is reasonable to assume that TWP and metals were likely to have originated from similar sources, which were distinct from those of MP. Since it is evident that the primary origin of TWP is traffic, which is also an important source for many metals (Rasmussen et al., 2023), it can be derived that MP were more associated with other types of human activities, a conclusion which is in line with findings by Bergmann et al. (2019) and Chernykh et al. (2022).

3.6. Fate of snowmelt

Upon melting, accumulated snow can convey microplastics and metals to surface waters. Riga city is situated on the Daugava River mouth on the south coast of the Gulf of Riga, Baltic Sea. Part of the

Table 2
Metals in the collected snow.

Sampling sites	Concentration [mg/L]				Concentration [μg/L]									
	Na	K	Mg	Ca	Cr	Mn	Fe	Co	Ni	Cu	Zn	Cd	Al	Pb
C1	80.4 (±28.5)	13.8 (±4.2)	12.7 (±7.6)	47.3 (±24.7)	24.0 (±15.5)	441 (±272)	8,910 (±6,145)	3 (±2.9)	14 (±8)	60 (±18)	525 (±60)	0.7 (±0.1)	3,350 (±2,129)	39 (±9)
C2	819. (±97.0)	8.8 (±1.7)	8.1 (±0.3)	36.9 (±3.3)	11 (±1.3)	177 (±14)	4,240 (±527)	2 (±0.3)	6 (±1)	58 (±9)	222 (±56)	0.3 (±0.5)	1,890 (±158)	21 (±3)
C3	1.1 (±0.4)	0.1 (±0.03)	0.1 (±0.04)	4.3 (±0.6)	1 (±0.6)	9 (±4)	105 (±40)	NQ	NQ	20 (±9)	94 (±71)	0.1 (±0.02)	50 (±10)	6 (±2)
C4	434.5 (±409.8)	37.7 (±30.2)	39.9 (±16.0)	132.5 (±55.5)	36 (±16.3)	622 (±246)	18,750 (±8,730)	11 (±5.5)	30 (±14)	130 (±50)	640 (±265)	0.9 (±0.4)	5,920 (±2,668)	33 (±15)
R1	33.7 (±53.5)	3.3 (±2.9)	5.4 (±6.4)	21.3 (±16.7)	6 (±6.8)	131 (±162)	2,190 (±3,084)	3 (±00)	4 (±5)	23 (±13)	154 (±182)	0.4 (±0.4)	1,460 (±1,757)	18 (±25)
P1	0.4 (±0.4)	0.05 (±0.01)	0.02 (±0.006)	5.9 (±2.9)	NQ	NQ	4 (0.3)	NQ	NQ	1 (±0.2)	1 (±00)	NQ	18 (±5)	1 (±0.1)

Ranked abundance of the specified metallic elements.
C1: Na > Ca > K > Mg > Fe > Al > Zn > Mn > Cu > Pb > Cr > Ni > Co > Cd.
C2: Na > Ca > K > Mg > Fe > Al > Zn > Mn > Cu > Pb > Cr > Ni > Co > Cd.
C3: Ca > Na > K > Mg > Fe > Zn > Al > Cu > Mn > Pb > Cr > Ni > Cd.
C4: Na > Ca > Mg > K > Fe > Al > Zn > Mn > Cu > Cr > Pb > Ni > Co > Cd.
R1: Na > Ca > Mg > K > Fe > Al > Zn > Mn > Cu > Pb > Cr > Ni > Co > Cd.
P1: Ca > Na > K > Mg > Al > Fe > Pb > Zn > Cu > Mn > Cr > Ni > Cd.
NQ: Non quantifiable, i.e., below the method quantification limit.

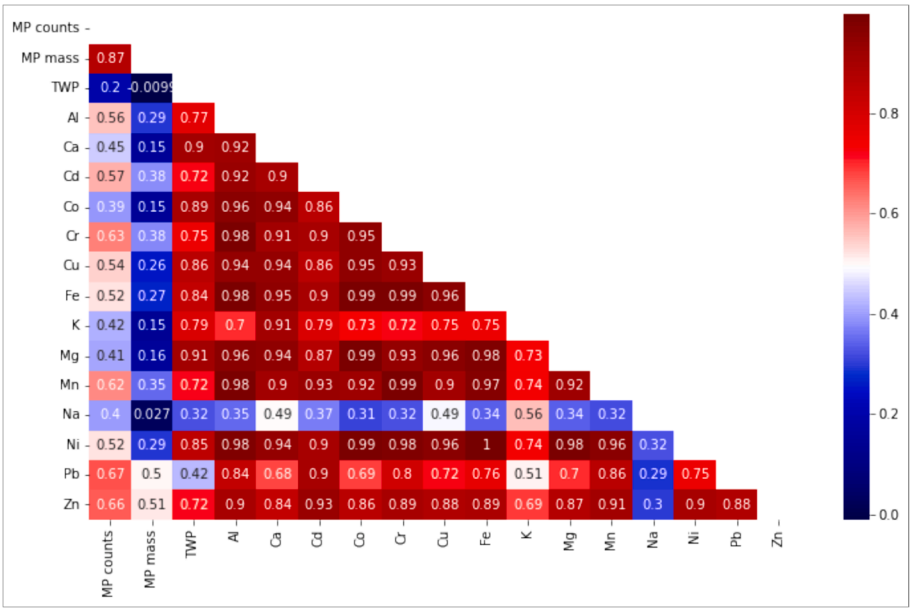

Fig. 4. Heat map of the correlation (R-coefficient) between MP counts, MP mass, TWP, and metals. The abundance of TWP was presented as mass.

snowmelt is collected by the city's combined drainage system and treated at its wastewater treatment plant, which discharges into the Gulf of Riga, and part goes directly into the Daugava via different watercourses. Snowmelts in Riga occur over quite short periods of time, leading to quite high pollutant loads being discharged during such events (Sakai et al., 1988). The microplastics and metals in the runoff may adversely affect the aquatic environment. A high Zn concentration, for instance, would negatively affect fish and invertebrates (Hagen and Langeland, 1973). Runoff from areas with dense traffic, for instance, highways, have been tested toxic towards various species (Kayhanian et al., 2008; McIntyre et al., 2015). Similarly, the marine fauna could be detrimentally affected by ingesting microplastics (Anderson et al., 2016), and additives in the microplastics, e.g., pigments, dyes, and plasticizers, might affect the organisms (Digka et al., 2018). Together, the findings of this study show that meltwater from urban snow should undergo treatment before being discharged to the aquatic environment.

4. Conclusions

Snow from snow dumping stations in Riga held significant concentrations of microplastics and metals compared to snow from an urban rooftop and a rural site in a national park. The latter were however also quite high compared to close by marine waters, as it held orders of magnitude more microplastics then previously reported for those. The content of tyre wear particles in the snow correlated strongly with most metals, while other types of microplastics showed little or no correlation. Consequently, tyre wear particle content did not correlate with the content of other microplastics. The snow dumps in the urban centre had higher concentrations of microplastics and metals than that of a residential area and those of a rooftop, indicating that urban activity influenced the pollution content of the snow. The quite high pollution level of the urban snow dumps proved that such dumping sites can be hotspots for microplastic and other pollution. If not treated prior discharge, it could impact receiving water quality. Urban snow melt should hence be treated prior to discharge to the environment, for instance, by sand filtration or sedimentation in a pond.

CRediT authorship contribution statement

Rupa Chand: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Data curation, Conceptualization. **Ieva Putna-Nīmane:** Writing – review & editing, Resources, Project administration, Investigation. **Elina Vecmane:** Writing – review & editing, Investigation. **Jeanette Lykkemark:** Writing – review & editing, Methodology, Data curation. **Jytte Dencker:** Writing – review & editing, Methodology, Data curation. **Asbjørn Haaning Nielsen:** Writing – review & editing, Methodology, Data curation. **Jes Vollertsen:** Writing – review & editing, Supervision, Funding acquisition. **Fan Liu:** Writing – review & editing, Methodology.

Declaration of competing 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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2024.108782.

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