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Identification and Quantification of Microplastics (MPs) in the Precipitation of Faribault City in Minnesota

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ABSTRACT

As one of the most successful synthetic materials, plastics have been produced at an exponential rate since its invention in the 1950s. Due to its strong durability, most disposed plastics can stay in the environment for a long time and experience degradation into microplastic particles (<5mm). Small microplastics particles have been found in fish, indicating seafood as a source of ingesting plastics; other studies have found atmospheric deposition as another potential microplastics particle transport route, as microplastics particles have been found in rain in many European cities and in some states in the US. Yet, there have been none trying to find microplastic particles present in precipitation in Minnesota. This study discovered the presence of microplastics in the rainwater in Faribault, a southern Minnesota city. The rain and snow was collected over night; photos and μ-Raman Spectroscopy graphs were taken for detailed analysis. Specifically, fibers and acrylic fragments (Type 1; Type 7) were present in the rain sample; no microplastic particles were found in the snow sample. The concentration of microplastic particles in the rain sample is approximately 75.34 particles m^{-2} d^{-1}, comparable to the concentration reported in other studies.

Background

Plastics are artificial polymers that were invented at the beginning of the 20th century. The first synthetic plastic that was made from fossil fuel, bakelite, was invented by Dr. Leo Baekeland in 1907 (Knight, 2014). Different plastics have distinct features for making certain types of products. For example, polyvinyl chloride (PVC) is widely used as a construction material due to its resistance to weathering, and polypropene (PP) is used in food wrappers and food packing due to its heat resistance (Hardin, 2021). Given their utility, plastics soon became a popular product after World War II. The annual production of plastics grew from 2.3 million tons in 1950 to 448 million tons in 2015, and is likely to double by 2050 (Parker, 2021). Meanwhile, disposal of plastics into the ocean has been an ongoing problem since their invention. Plastic products are disposed of in landfills, where they are carried by water (river or underground water), wind, and other factors, into the ocean.

Plastic's chemical structure–characterized by a series of carbon rings–shows its sturdiness to environmental weathering. Yet plastic will start to degrade slowly as soon as it is exposed to environmental factors such as wind, water, and heat. There are several mechanisms of plastic degradation, including photodegradation, thermal oxidative degradation, hydrolytic degradation and biodegradation. In addition to carbon, plastics also have hydrogen and other chemical additives. Hydrogen is the target of the most common type of degradation–thermal oxidative degradation. It works by converting hydrogen atoms in plastics into hydroperoxide, and the reaction can be catalyzed by heavy metals like copper (Martin, pg179). After this process, the plastics will shrink in size, and then can be categorized into megaplastics (>50cm), macroplastics (5-50cm), mesoplastics (5mm–5cm), microplastics (0.001mm–5mm), and nanoplastics (<0.001mm).

Microplastics' small size gives them the ability to "migrate" along natural circulations like wind and water. There are two categorizations of disposed plastics: primary and secondary. Primary plastics refer to those that seep into underground water which eventually flows into the ocean; secondary plastics refer to those plastics that break down from larger plastics that exist in the ocean. There, depending on the density and size, plastics can either float on the surface of the ocean, or sink deep into the bottom layer. Decades of investigations have found the ubiquitous existence of plastics–especially microplastics–throughout oceans globally. A study done in Kyushu University estimated that approximately 24.4 trillion pieces of microplastics are in the upper layer of the oceans globally, and they acknowledged that the actual number could be higher when considering plastics in all ocean layers and some ocean regions (Indian ocean and South China Sea) which lack data (Isobe et al, 2021). The transportation pathway of floating microplastics will be determined by the ocean current, bringing plastics to most places in the world. Often, plastics can end up being brushed on beaches, or get engulfed by marine organisms. Incidences of plastics have been reported in whales, seagulls, sea turtles, and many other marine organisms.

In addition to oceans, the atmosphere is another potential plastic reservoir, as studies have discovered microplastic fallouts in the Pyrenees mountains (Allen et al, 2019), Dongguan (Cai et al, 2017), Arctic regions (Bergmann et al, 2019), Hamburg (Klein&Fisher, 2019), and San Paulo (Amato-Lourenco et al, 2022). They may reach and stay in the atmosphere by blowing off the wind from surface or plastic aerosolization, a concept that refers to transport of microplastics from the surface of water bodies to the atmosphere. Then the microplastics will stay in the air until they are brought down by the gravitational effect (dry deposition) or precipitation (wet deposition).

Due to the prevalence of microplastics, their effects on human health have been a topic of investigation. Studies have found their existence in human blood (Leslie et al., 2022), lung tissues (Jenner et al, 2022), sputum (Huang et al, 2020), placenta (Ragusa et al, 2021), and human stool (Schwabl et al., 2019). Microplastics may cause oxidative stress in cells; this has been shown in T98G and HeLa cells (Schirinzi et al, 2017). Oxidative stress refers to the imbalance between the accumulation of oxygen reactive species and the rate at which the body is eliminating them out of the body. Oxidative stress can damage cell components, such as the plasma membrane and DNA, and can lead to subsequent problems like cardiovascular disease (CVD) and possibly cancer (Pizzino et al, 2017). One study found that microplastics can lead to the release of histamine, cytokine, and myokine (Danopoulos et al, 2021). Histamine and cytokine are immune cells that alter the body's defense against viruses and bacteria, and excessive release of histamine and cytokine could cause allergy and a cytokine storm. Myokine helps control the rate of metabolism, and the disturbance in levels may lead to obesity, diabetes and other problems (Ahima & Park, 2015). Another reason microplastics can be harmful to the human body is due to substances that attach to the microplastics. Previous studies have found the attachment of some metal elements (Al, Mn, Cu, and more) on polyethylene pellets sampled on the beach of west England (Holmes et al, 2011). In addition, harmful metals like mercury (a neurological toxin) that can affect human health significantly once absorbed have been found to adsorb onto the microplastics (Barboza et al, 2018). On beaches in north China, persistent organic particles (POPs) have been detected with plastics, and one of the dominant types is dichlorodiphenyltrichloroethane (DDT) (Zhang et al, 2015). DDT is a type of banned insecticide that may cause tremors, seizures and vomiting, and is postulated to be a carcinogen (CDC, 2021). While more research is needed to reveal additional effects of microplastics on the human body, it is clear that the presence of microplastics does pose health concerns.

As many studies have found microplastic fallouts in European and Asian regions, efforts are underway to discover whether there are microplastic fallouts in North America as well. While some studies have found the existence of microplastics in waterbodies of Minnesota and western Lake Superior (Conowall et al, 2023) (Hendrickson et al, 2018), there has been no research examining potential microplastics in precipitation. Thus, we hypothesized that there would be microplastics falling through the precipitation in the city of Faribault in Minnesota.

Method

Collection of precipitation

A bowl was used to collect rainwater. The bowl was made of stainless steel to prevent possible plastic pollution. The bowl was about 26.2 cm in diameter and 11.5 cm high. The opening area is 0.053 m^2. A metal mesh with openings size of 1mm^2 was attached onto the bowl by metal clips to prevent falling of twigs and leaves into the bowl. The bowl was placed in an open area, so the rainwater was collected directly from the sky, instead of from water on the ground or drops from the roof and gutters. Thus, we limited possible contamination from plastics on the ground or any building structures. After collection, the bowl was covered with aluminum foil to prevent exposure to light and evaporation. The bowl was stored in the refrigerator to minimize the effect of heat and inhibit microbe growth. The rain sample was collected on November 8th. Before the next collection, the bowl was placed in a drawer after being rinsed with water that has been filtered (using the same mesh we use in the study) to be MPs-free. Two samples of snow were collected from December 2022 to January 2023. Considering the larger volume of snow than water in the same amount of liquid, a vertical metal cylinder was applied to surround the bowl to accommodate more snow. The sample was collected from the night of January 16 to the morning of January 17, 2023.

Protocol One: Visual Identification

Dissecting scissors were used to cut the mesh into an approximate circle with a radius of 25 mm to fit a Swinnex 47mm filter holder. Then, a glass syringe was filled with sample water, attached to the filter holder by the syringe tip, and the plunger was pushed to move the water through the mesh. This attachment was based on the Luer-Lock system on the syringe. A beaker was placed under the filter holder to hold the sample water. Then, we removed the filter holder to siphon the sample water in the beaker into the syringe. Repeating this process multiple times would allow as many plastics to adsorb onto the mesh as possible.

Then, a tweezer was used to move the filter onto a glass slide which would be put under a Wolfe® Advanced LED Series Monocular Microscope (59-1002) to observe possible signs of plastics. The microscope had 4X, 10X, 40X, and 100X objective lenses, with 4X being used to locate plastics and 10X and 40X being used to record physical characteristics of observed objects.

Protocol Two: Lab Analysis

We shipped the 400 ml rain sample on November 9th and 300 ml snow sample on February 8th. Samples were shipped to the Environmental Molecular Science Laboratory (EMSL) in Tampa, Florida, for numerical data analysis. When the sample arrived at EMSL, a sonicator was used to break up any aggregates. The whole sample was filtered through a polycarbonate filter with pore sizes of 0.8 µm. Then, the filter was placed on a glass slide using double-sided tape. Two types of light microscopes (polarized light microscope: *Zeiss Universal Petrographic Microscope* and reflected light microscope: *Nikon DF Microscope*) were used. A Raman Spectrometer (*Horiba, Xplora Plus*) was used to analyze particles using red (785 nm) and green (532 nm) lasers; these lasers will bounce back from the particles. Photos with details of plastic shape, size, and color were taken, and the Raman spectroscopy was used to read and analyze the spectrum of the plastic particles to decide their categorization.

Results

Visual Clues at SSM

Rain Sample

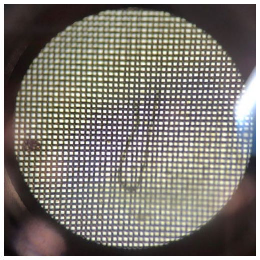

Figure 1: image showing a possible textile fiber with light green color

Figure 2: image showing an unconfirmed object that's likely to be plastic due to its bright color

Snow Sample

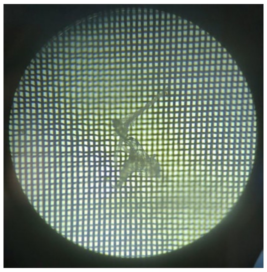

Figure 3: image showing a possible plastic foam (plastic bag) fragment

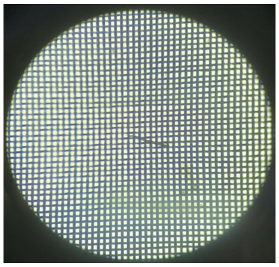

Figure 4: image showing a possible textile fiber

EMSL Lab Analysis: Rain Sample

Within the 400 ml sample, 6 plastic particles–4 polyester textile fibers (Type 1: polyethylene terephthalate) and 2 acrylic particles (Type 7: other types)–were found.

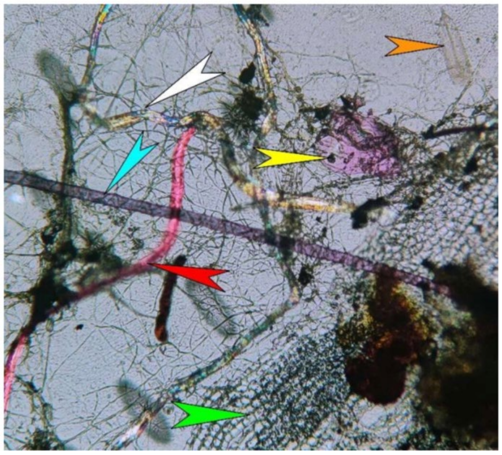

Figure 5: Image showing microplastics and exterior particles. Arrows with different colors show different particles; microplastics include polyester textile fiber (red arrow) and acrylic fragments (yellow arrow); other particles include animal hair (cyan arrow), cotton fiber (white arrow), plant matter (green arrow), and insect scale (orange).

In detail, the size of six microplastics particles falls within the range from 5μm to over 1000 μm (with maximum diameter of 1760 μm and length of 3500mm).

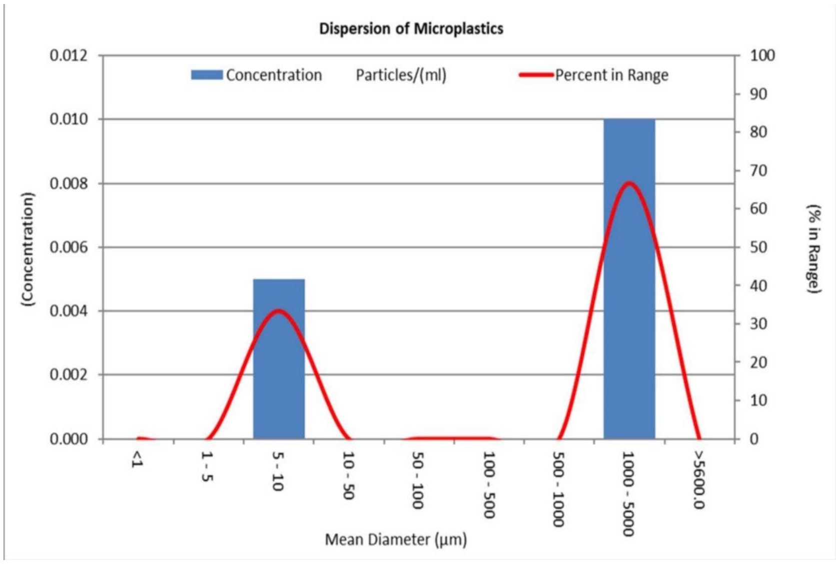

Figure 6: Histogram showing the size distribution of six particles; a mean diameter of 5-10 μm represents acrylic fragments, and 1000-5000 μm represents polyester fibers. The concentration is 0.005 and 0.01 pcs/ml, respectively. The percent in range represents the proportion of each type of MPs in total (there are 2 acrylic fragments detected among all 6, thus its percent in range is 33% which is shown as red lines on the left bar).

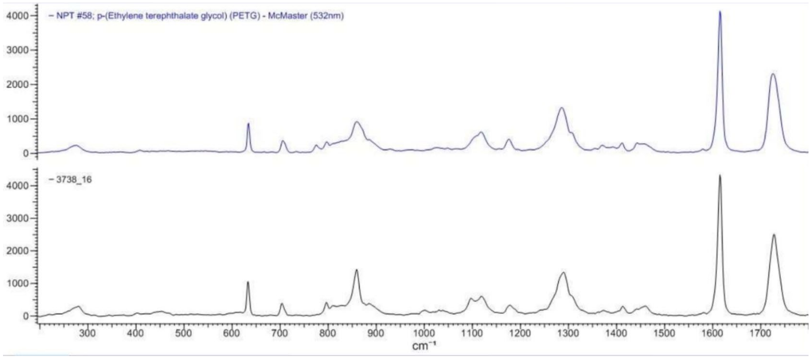

Figure 7: Raman spectrum of polyester textile fibers found in the rain sample

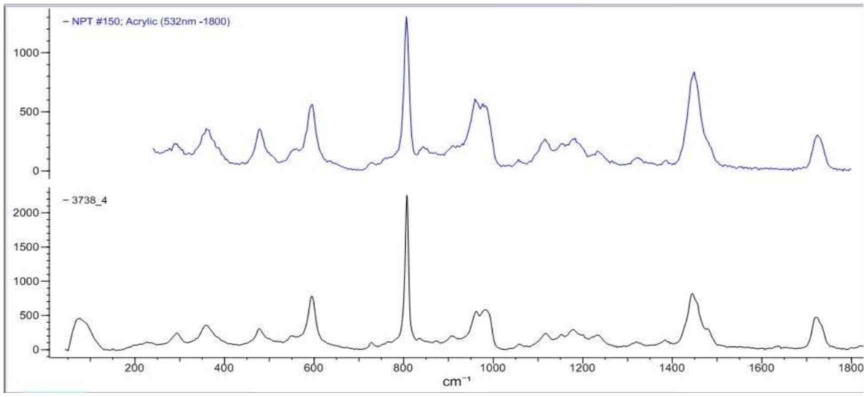

Figure 8: Raman spectrum of acrylic particles found in the rain sample

EMSL Lab Analysis: Snow Sample

Within the 300 ml snow sample, **no** signs of MPs were shown. Photos of plant matter and quartz in the snow sample are shown as below:

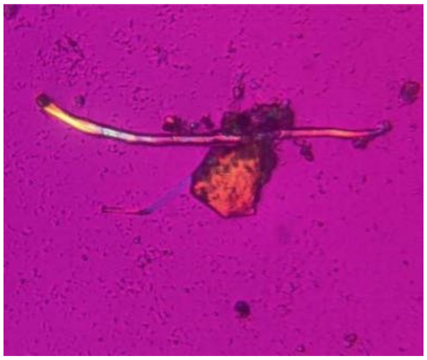

Figure 9: Image showing plant matter and quartz (sand)

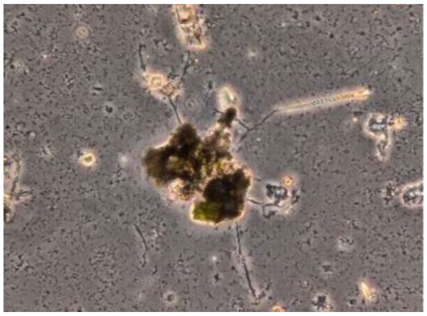

Figure 10: Image showing plant matter with chlorophyll (green)

Conclusion & Discussion

These results partially support the hypothesis: MPs were only found in rainwater in Faribault, MN. This study provides a first glance at the MPs concentration in precipitation in Minnesota. Previously, MPs were found in western states like Utah, Colorado, and Wyoming (Brahney et al, 2021), and in eastern states like New Jersey (Yao et al, 2021). These studies report MPs in $m^{-2}d^{-1}$; in order to make comparisons between this study and others', a conversion was needed. Equations for conversion between these two units are shown below (*Allen et al,* 2019):

$$\underline{X} = (\sum \quad X_{1-n}) / n$$

X¯ = the average MP count for a sample area (1370 mm^2), $X_{1,2,3}$ = the MP count for a sample area 1, 2, 3, etc. (sample area = 1370 mm^2) and n = sample area number (one sample area was investigated).

$$\mu p = (\underline{X} * Y/y) - \mathcal{E}$$

where μP = the total MP count per filter, y = the sample area (1370 mm^2 or 0.00137 m^2), Y = the total filter area (1370 mm^2) and ε = the sampling error, the number of MP particulates found on the blank samples (Ɛ=0).

$$MP = (\mu P/a) / d$$

where MP = MP count m^{-2} d^{-1}, a = the sample area of the atmospheric collector (m^2; a=0.053 m^2) and d = duration of the sampling period (days; d=0.66).

Based on these equations, our result is about 75.34 MPs m^{-2} d^{-1}, which is higher than the study done in Dongguan (36±7 MPs m^{-2} d^{-1}) (Cai et al, 2017), and smaller than the study done in Paris (118 MPs m^{-2} d^{-1}) (Dris et al, 2015) and New Jersey (327±19 MPs m^{-2} d^{-1}) (Yao et al, 2021). The population and human activities in the area local to the sampling site may be important contributors to the MPs deposition rate. Clearly, there is a significant difference in the population of Faribault (24,000) compared to Dongguan (7.5 million), New Jersey (9.26 million), and Paris (9.7 million). Nevertheless, it is possible that MPs may be transported from the closest urban setting–Twin Cities (approximately 3 million)–which may contribute to the high MPs in Faribault's rural setting. The effect of population on MPs deposition is supported by the study in Dongguan; *Cai et al* sampled 3 sites with a difference in population density (Cai et al, 2017). They found more MPs in a middle school compared to less populated areas such as gyms and waterworks. Another factor that may account for differences in MPs observed between these studies is the sampling height; our sampling height is 0m (on the ground), while other studies, such as Cai's (15m) and Yao's (25m), have their sampling device located at high heights such as a rooftop. In a study done by Purwiyanto in Jakarta, Indonesia, it was suggested that sampling height is a factor that influences the deposition rate: the higher the sampling device is placed, the more likely it will collect particles, resulting in a higher deposition rate (Purwiyanto et al, 2022). Other factors that contribute to these differences in MPs deposition may include meteorological factors like temperature and wind speed, and physical factors like terminal velocity and the size of raindrops (Zhao et al, 2015). However, this study lacks data on meteorological and physical factors that can be used for comparison.

In both EMSL and SSM microscopic observations, fiber is the most common type of observed particle (66% in EMSL sample); this is similar to results of other studies (Cai et al, 2017) (Dris et al, 2015) (Yao et al, 2021). In the 6 identified MPs, Type 1 (PETE) plastic accounts for the majority (66%), followed by Type 7. However, due to technology limitations, it is possible that fibers observed in SSM samples could be natural fiber, especially considering the high proportion of natural fiber found in Cai's study. In addition, since our visualization analysis was mainly based on the morphological features of objects, some objects were visually similar to natural matter identified by the Raman-spectroscopy in EMSL. For example, **Figure 4** is visually similar to **Figure 11** in shape and color; thus, unless the objects had pronounced features of MPs (bright color) we cannot assert that we have observed MPs in the sample.

Figure 11: root of Fungal Mycelium in snow sample taken by EMSL

The difference between rain and snow in the ability of catching solid particles is attributed to their difference in scavenging capacity. In previous studies, the difference in scavenging efficiency between rain and snow was measured. Zhao's study found that snow's scavenging efficiency can be up to 1000 times higher than rain (Zhao et al, 2015); the ability to scavenge particles for snow can be 50 times more efficient than rain (Bergman et al, 2019); Yet, in this study, rain appears to be more efficient in catching MPs than snow. As for our snow sample, the absence of MPs is likely due to the time of sampling.

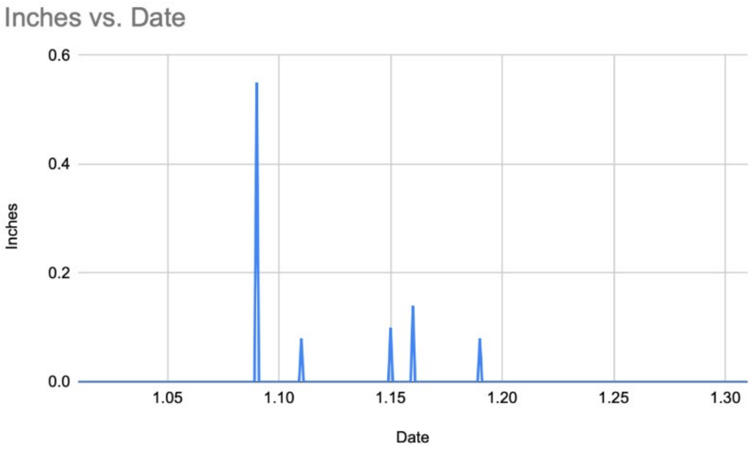

Figure 12: record of January precipitation. The snow sample was collected on Jan 16th, and is the fourth peak from left to right

As shown in **Figure 12**, there were multiple snow events in January prior to our collection date. Given the effective scavenging efficiency of snow compared to rain, we assumed that MPs in the atmosphere were mostly scavenged by the snow on Jan 15th or all three snow events prior to Jan 16th. Since the atmosphere cannot accumulate enough MPs after these snow events, our snow sample did not scavenge any MPs from the air. Another possible explanation for the difference between this study and others could be due to the sampling method and period. While our snow came directly from the sky, other studies collected snow by spooning the top layers of snow (Bergman et al, 2019) (Aves et al, 2022). This sampling method difference may also indicate the unit difference between rain and snow, as the opening area of the collecting device in rain collection is one factor that may affect the result. Thus, our snow sample has a much lower volume compared to these studies; meanwhile, EMSL's analysis accuracy is dependent on the volume of sample sent, where an ideal volume is at least 500 ml (compared to our 400 ml rain and 300 ml of snow). Our study considered the possibility of other MPs sources after snowfall, which can potentially increase the MPs being analyzed thus leading to a higher concentration; Bergmann's study shared the same concern, that their snow sample from Arctic regions might be exposed for an unknown time which may accumulate more MPs through dry deposition (Bergmann et al, 2019).

Summary

This study provides a first glance into the MPs in precipitation in Minnesota: as MPs are present only in rain samples, not in snow. When performing statistical analysis, we found that among many studies in this field, there has been no standardized way of collecting and analyzing the sample. In addition, the result of this study should be treated with caution, given the high variability accompanied with single-time collection. In the future, a collection over a long period of time is needed, and variables that may affect MPs deposition should also be recorded for comparison with other studies. In addition, a study investigating the influence of one precipitation event on later events in scavenged MPs is one of the future projects of the research.

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