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Airborne Microplastics: Emission Mechanisms and Exposure Factors

Author

Leonard, Dona Jamie

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Airborne Microplastics: Emission Mechanisms and Exposure Factors

A dissertation submitted in partial satisfaction of the

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in Civil Engineering

by

Dona Jamie Leonard

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ABSTRACT OF THE DISSERTATION

Airborne Microplastics:

Emission Mechanisms and Exposure Factors

by

Dona Jamie Leonard

Doctor of Philosophy in Civil Engineering

University of California, Los Angeles, 2024

Professor Sanjay K. Mohanty, Chair

Microplastics are released from terrestrial surfaces into the atmosphere where they are conveyed via wind and may be deposited, remain in the atmosphere, or disperse across long ranges. Because of this high atmospheric contamination, inhalation is one of the primary exposure pathways for microplastics. Inhalation exposure is controlled by two key factors: a) the quantity of microplastics available to be emitted from terrestrial environments to the atmosphere, and b) the factors that influence the potential of this emission occurring. The reported concentrations of microplastics on terrestrial surfaces vary widely based on monitoring methodology and characteristics of the capture region, making it difficult to estimate potentially emitted microplastics. Concentrations of traditional atmospheric aerosols such as particulate matter (PM 10, PM 2.5), NOx, SOx, and heavy metals, vary based on region characteristics such as climate,

land use, socioeconomic vulnerability, and urban sources including landfills, wastewater treatment plants, and hazardous waste sites. However, microplastics have unique properties: low density and high hydrophobicity, which may alter their emission mechanisms and spatial distribution. Thus, it is unknown how fluctuation in these exposure factors could affect the emission potential of microplastics, influencing concentration levels in the atmosphere. Additionally, the fundamental emission mechanisms and factors that could affect the emission potential of microplastics into the atmosphere are unclear. A wide body of literature models the atmospheric emission of particulate pollutants; however, no model to date adapts these traditional emission models by accounting for the unique properties of microplastics. The overall objective of the dissertation is to improve the mechanistic understanding of microplastic emission into the atmosphere, and the factors which may affect the exposure of human populations to higher levels of emitted microplastics in the atmosphere.

The dissertation consists of six research chapters. Chapter 2 analyzes the global distribution of deposited airborne microplastics in terrestrial environments to estimate the relative importance of climate and land use, proving that the climate has a larger impact than land use on the abundance of deposited airborne microplastics. Chapter 3 compares the abundance, composition, and size of microplastics found in sediment cores and the water column in the Gulf of Mexico, proving that most microplastics have been deposited in the last two decades. Chapter 4 estimates the potential variability of the abundance of airborne microplastics on leaves in Los Angeles as a function of leaf height, leaf surface hydrophobicity, and land use, confirming a limited scope to use leaves as biomonitoring systems for urban atmospheric microplastics due to high uncertainty. Chapter 5 estimates the relative importance of socioeconomic status or proximity to known sources of microplastics on deposited airborne microplastics, revealing the difficulty of predicting exposure

risk and the ubiquitous spatial distribution of microplastic abundance in the atmosphere. Chapter 6 analyzes wind-borne sediments collected from wind tunnel experiments on biosolid-applied agricultural fields, finding preferential emission of microplastics from the soil when compared to the background soil. A theoretical model was constructed considering microplastics' low density and high hydrophobicity which verifies this experimental data and predicts a substantial underestimation of the wind events which exceed the fluid threshold necessary to emit microplastic from biosolid-amended solids. Chapter 7 quantifies the adhesion force between plastics and sand revealing a significant inverse relationship between ultraviolet (UV) weathering time and relative humidity (RH), which implies microplastic's emission potential into the atmosphere could be increased as residence time in the environment increases, or in more humid environments. Overall, the results help to: a) quantify the fundamental forces affecting microplastic emission, b) outline the factors that may influence the emission potential of microplastics, and c) identify research needs to better predict microplastic inhalation exposure.

The dissertation of Dona Jamie Leonard is approved.

Erik M.V. Hoek

Shaily Mahendra

David Jassby

Win Cowger

Sanjay K. Mohanty, Committee Chair

University of California, Los Angeles

2024

Dedication

To everyone who loves me enough to read past Chapter 1.

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VITA

EDUCATION

2021 M.S. in Environmental Engineering, University of California, Los Angeles

2020 B.S. in Environmental Science: Health & Engineering, University of California, Los Angeles

AWARDS

2024 UCLA Dissertation Year Fellowship (Awarded, Declined)

2024 UCLA Engineering Achievement Award for Student Welfare

2022 Expanded Shale, Clay, and Slate Institute John Ries Scholarship

2020 NSF Graduate Research Fellowship

2020 UCLA Graduate Opportunity Fellowship

2020 UIC Institute on Sustainability and Energy Summer Fellow

2021 UCLA Outstanding MS in Civil Engineering

2016 Sandpipers Foundation Scholarship

2016 Rotary International Women in STEM Education Scholarship

PUBLICATIONS

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1. CHAPTER 1 – MICROPLASTIC CONCENTRATIONS AND TRANSPORT IN THE ATMOSPHERE

1.1. Introduction

Microplastics, defined as plastic particles smaller than 5 millimeters, are pervasive environmental contaminants frequently found in terrestrial, aquatic, and more recently, atmospheric environments. Prolonged use of plastics has resulted in a rapid accumulation of microplastics on Earth's surface, from where they can be suspended by wind, transported through the atmosphere, and deposited back on lands across geographical boundaries (Bullard et al., 2021a; Habibi et al., 2022; Nafea et al., 2024; Rezaei et al., 2019, 2022a). The atmospheric pathway is particularly concerning as it represents a critical vector for human exposure, primarily through inhalation (Borthakur et al., 2021a; Chen et al., 2023; Gasperi et al., 2018; Geng et al., 2023; Saha & Saha, 2024). High levels of airborne microplastics can pose significant inhalation risks (Prata et al., 2020). The inhalation of small airborne microplastics, particularly those under 10 μm in size, may lead to negative health effects and decreased respiratory function in humans (Cao et al., 2024; Prata, 2018a; Prata et al., 2020; Vasse & Melgert, 2024; Xing et al., 2016). Moreover, some anecdotal studies have detected microplastics in the human bloodstream (Leslie et al., 2022a) and deep lung tissue (Amato-Lourenço et al., 2021a; Huang et al., 2024; Jenner et al., 2022), suggesting that respirable microplastics from the air can penetrate previously unforeseen regions where they are associated with adverse outcomes- such as cardiovascular disease (Marfella et al., 2024; Persiani et al., 2023). Given that many microplastics can adsorb heavy metals and toxic organic substances, leaching these chemicals from microplastics into the body could further heighten health risks (Borthakur et al., 2021a; Rodrigues et al., 2019). Inhalation risk is determined by the concentration of microplastics present in the atmosphere. This inventory is driven by two primary factors: (a) the amount of microplastics present in terrestrial environments for emission into the

atmosphere, and (b) the environmental factors that influence the emission of microplastics into the atmosphere. Thus, it is critical to understand which factors influence the emission and exposure of airborne microplastics.

Research on the atmospheric behavior of microplastics is still in its infancy, with many gaps in understanding the fundamental mechanisms driving their emission. The transport and deposition of microplastics in the atmosphere are influenced by their unique physical properties, including low density and high hydrophobicity. These properties distinguish microplastics from traditional atmospheric aerosols such as particulate matter (PM 10, PM 2.5), NOx, SOx, and heavy metals (Prajapati et al., 2022; Vieira et al., 2020). The low density of microplastics could influence their resistance to emission and dispersion, while their hydrophobic nature may affect the interparticle forces between plastics and terrestrial surfaces, as well as the cohesive interactions of water. These characteristics likely complicate the emission and transport of microplastics in the atmosphere compared to conventional particulate pollutants, and the extent to which they do needs to be investigated.

Furthermore, while the environmental and socioeconomic factors influencing the concentration and distribution of traditional atmospheric aerosols are well-documented- the effect of these factors on atmospheric microplastic concentrations is unknown. Such factors may include proximity to urban sources, regional climate variations, land use patterns, and socioeconomic vulnerability (Eeftens et al., 2012; Fiore et al., 2015; Jacob & Winner, 2009; Mikati et al., 2018; Rentschler & Leonova, 2023; Wu et al., 2015). To address this gap, it is essential to measure microplastic concentrations extensively in a region to feed into models that can evaluate which factors are more likely to affect exposure to microplastics.

This research aims to improve the mechanistic understanding of microplastic emissions into the atmosphere and the factors influencing human exposure to airborne microplastics. By quantifying the forces affecting microplastic emission and identifying the environmental and socioeconomic factors that impact their emission potential, this work seeks to inform public health exposure models and help develop strategies to mitigate the risks associated with airborne microplastics. This research is particularly relevant in the context of climate change, which is expected to alter regional climate zones and average wind and humidity levels, potentially affecting the emission and distribution of microplastics in the atmosphere. Understanding these dynamics is crucial for predicting future exposure risks and developing effective interventions to protect human health and the environment.

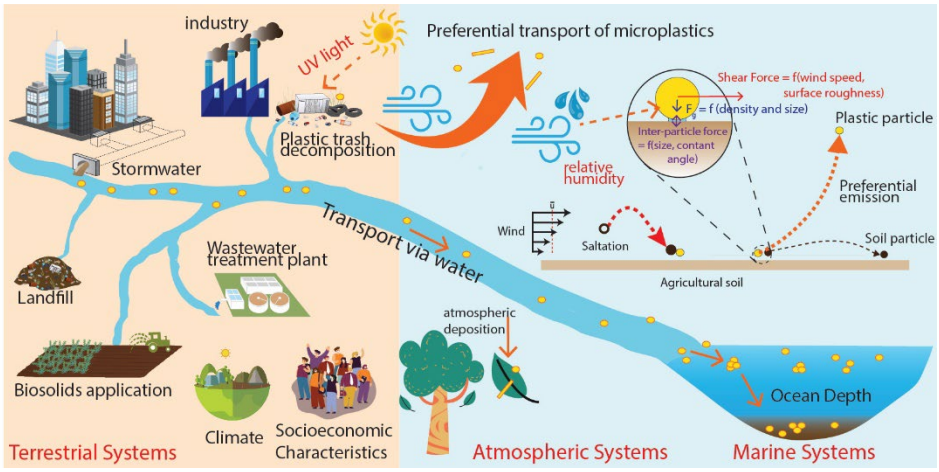

Figure 1-1: Emission and transport mechanisms of microplastics from terrestrial surfaces to the atmosphere, and factors influencing their emission including UV exposure, relative humidity, relative climate, land use, and socioeconomic characteristics of populations.

1.2. Research gaps

1.2.1. Impact of climate and land use on exposure to deposited airborne microplastic concentrations

Understanding the impact of climate and land use on the concentration of deposited airborne microplastics is crucial for assessing environmental exposure and developing mitigation

strategies. Despite the growing recognition of microplastics as an environmental contaminant, there is a significant research gap in understanding how regional climate and land use patterns influence their atmospheric deposition and subsequent exposure risks. The net amount of airborne microplastics in a region and their recorded deposition rates could be affected by a) the source region's land use, which provides the source or pool of microplastics for emission, and b) climate classification, which represents the available energy for microplastic disintegration, emission, and transport (Crossman et al., 2020a; Leonard et al., 2024). Climate plays a pivotal role in the dispersion and deposition of traditional airborne particles, and this may extend to microplastics. However, the specific impacts of different climatic conditions on microplastic concentrations remain underexplored. Research focusing on how variations in climate can help predict the concentrations of airborne microplastics could provide valuable insights into regional exposure risks. Land use patterns, including urbanization, agriculture, and industrial activities, also play a critical role in determining the concentration of microplastics in different regions (He et al., 2019; Koutnik et al., 2021; Lebreton & Andrady, 2019; Li et al., 2022). However, the interaction between different land use types and their combined effect on microplastic deposition is not well understood. Studies that examine the spatial distribution of microplastics within various land use categories could help identify hotspots of microplastic contamination and inform land management practices. No study to date has analyzed the relative importance of these factors on the overall deposition rate of airborne microplastics.

1.2.2. Lack of data for microplastics in deep sea sediment ocean cores

Waterways in terrestrial environments transport a large amount of microplastics into the ocean, where many of them break down into smaller particles and sink to the ocean floor. Microplastics deposited for several decades can change the composition of the ocean floor

(Cressey, 2016; Haward, 2018; Jambeck et al., 2015; MacLeod et al., 2021), which has a significant role in the health of marine species (Collard et al., 2019; Hall et al., 2015; Pannetier et al., 2020; Reichert et al., 2019) and gas exchange (Gregory, 2009; Shen et al., 2020). However, limited studies have measured microplastic concentration on the ocean floor due to difficulties in getting ocean core sediments (Cunningham et al., 2020; Loughlin et al., 2021; Martin et al., 2017a; Shruti et al., 2021). The scarcity of data on microplastic concentrations and distribution in deep-sea sediments hampers our ability to assess the full extent of marine plastic pollution and its ecological impacts. Comprehensive studies focusing on deep-sea sediment cores are essential to unravel the long-term deposition trends of microplastics, their interaction with deep-sea biota, and their potential role in the global carbon cycle. No study to date has constructed a depth profile for microplastics found in a sediment core from the deep sea, which can help estimate the onset of pervasive marine microplastic contamination. Addressing this gap is critical for developing a holistic understanding of marine microplastic pollution and informing effective mitigation strategies.

1.2.3. Lack of methods standardization for biomonitoring of airborne microplastics

Quantifying airborne microplastics often involves the installation of passive or active samplers- which are relatively tedious and labor intensive (Cai et al., 2017; Dris et al., 2016; Zhou et al., 2017). To overcome this limitation, researchers have explored the use of natural samplers such as leaves (Jiao et al., 2024; Liu et al., 2020; Xu et al., 2024), spider webs (Goßmann et al., 2022; Iordachescu et al., 2024), and lichen (Jafarova et al., 2022). Leaves in urban canopies naturally intercept wind flows and trap particulate pollutants on their surfaces, thereby improving the air quality in cities (Leonard et al., 2016; McPherson et al., 2011; Rai, 2016). Therefore, they have been used as passive samplers to measure the atmospheric deposition of a wide range of

pollutants including ozone (de Souza et al., 2022), particulate matter (Beckett et al., 2000; Hansard et al., 2012), persistent free radicals (Leonard et al., 2016; Oyana et al., 2017), and polyaromatic hydrocarbons (Wannaz et al., 2013). For leaves to be used as monitoring systems for airborne microplastics, it is critical to determine how microplastic concentrations may vary on the leaves based on leaf properties and leaf position so that a monitoring protocol can be standardized to reduce variability. One of the critical challenges in assessing the environmental and health impacts of airborne microplastics is the lack of standardized methods for biomonitoring. A few studies have quantified microplastic concentration on leaves (Koutnik et al., 2022a; Li et al., 2022; Liu et al., 2020; Liu et al., 2022; Xu et al., 2024), but they rarely account for the causes of concentration variability. Based on these studies, microplastic concentrations on leaves can vary by two orders of magnitude, from 0.1 to 50 pieces (n) per cm^2, even at the same spot. However, current studies on airborne microplastics often employ diverse sampling techniques, analytical methods, and units of measurement, leading to inconsistent and non-comparable results. This variability hampers the ability to accurately assess exposure levels, spatial and temporal trends, and the effectiveness of mitigation strategies. Thus, it is critical to investigate the sources of variability in microplastic concentrations on leaves and inform the development of standard sampling best practices to use leaves as a biomonitoring system. Standardization would enable more accurate assessments of exposure levels and trends; thus, addressing this gap is crucial for advancing our knowledge of airborne microplastics and for informing effective regulatory and policy responses.

1.2.4. Impact of socioeconomic factors and proximity to sources on exposure to deposited airborne microplastic concentrations

Socioeconomic factors and proximity to pollution sources have long been used to predict airborne particulate matter (PM) (Alastuey et al., 2006; Cheriyan & Choi, 2020; Mazzei et al., 2008; Mikati et al., 2018; Rentschler & Leonova, 2023; Yadav et al., 2020). However,

microplastics are a different class of particulate matter. It raises the question: Can socioeconomic factors and proximity to sources predict microplastic pollution just as they do traditional PM in the atmosphere? While a few studies have preliminarily explored how environmental factors contribute to microplastic distribution in air and terrestrial ecosystems (Corradini et al., 2021; Rafique et al., 2020; Yin et al., 2020; Zhang et al., 2022), none have systematically examined how socioeconomic status affects exposure levels. Understanding these dynamics is crucial as vulnerable communities, often situated near industrial zones or urban centers, may face higher exposure risks due to localized pollution. Moreover, socioeconomic factors such as poverty level and status as a disadvantaged community, likely influence airborne microplastic exposure patterns-just as they do traditional particulate matter (Morello-Frosch et al., 2001; O'Neill et al., 2003; Pastor Jr. et al., 2005). Addressing this gap is essential for developing targeted mitigation strategies and policies that can reduce disparities in exposure and protect public health effectively.

Furthermore, existing research often overlooks the complex interactions between geographical proximity to microplastic sources and exposure levels. While studies have identified potential sources like landfills, wastewater treatments plants, and industrial sites (Li et al., 2016; Mukherjee & Agrawal, 2017; Shi et al., 2020), it is unclear whether the concentration of airborne microplastics can be predicted based on proximity to source. Factors such as wind patterns, land use practices, and local climate variations may significantly influence how microplastics disperse and accumulate in different environments. This knowledge gap limits our ability to accurately assess exposure risks across diverse geographic settings and hinders the development of spatially informed policies and interventions. Therefore, filling this research gap is crucial for developing comprehensive risk assessment frameworks and targeted monitoring strategies that can effectively mitigate airborne microplastic exposure in communities worldwide.

1.2.5. Theoretical framework to predict microplastic emission by wind

Microplastic emission by wind occurs when the shear stress exerted by wind on the ground surface exceeds the shear strength of the aggregates and their resistance to detachment. The minimum wind shear stress required to cause erosion, commonly known as the fluid threshold, depends on several factors including particle characteristics, size and stability of the soil aggregates, field surface conditions, vegetation cover, and near-surface soil moisture (Kok et al., 2012; Ravi et al., 2011). Thus, microplastic properties such as size and surface properties could affect these forces, which in turn affects their emission potential. The emission potential of microplastics, especially under the influence of surface properties such as density and hydrophobicity, represents a critical area with substantial gaps in current knowledge. Density and hydrophobicity are known to influence the behavior of microplastics in aquatic environments, affecting their transport and bioaccumulation (Ben Stride et al., 2024; Y. Huang et al., 2024; F. Li et al., 2024; M.-M. Tan et al., 2024). However, their impact on airborne emissions, particularly in relation to the wind fluid threshold—the point at which particles become airborne due to wind action—is underexplored. Addressing this gap is essential for accurately predicting the environmental fate of microplastics and assessing their potential to contribute to atmospheric pollution. Current models for fugitive dust emission use well-established fluid thresholds for traditional particulate matter, however, these thresholds could differ for microplastics. As a result, there could be a severe over- or under- estimation of the number of wind events which are capable of resuspending microplastics, and thus expose human populations to microplastics- though this has never been evaluated. This gap hinders the development of robust predictive models and mitigation strategies tailored to minimize microplastic emissions and estimate microplastic exposure risks effectively.

1.2.6. Impact of UV weathering and relative humidity on emission potential of microplastics

The surface properties of particulate pollutants- like microplastics- are predicted to affect their interparticle forces and subsequent emission potential into the atmosphere; however, the influence of environmental conditions is yet unexplored. For example, microplastics accumulated in the environment are continually exposed to UV radiation in sunlight, which could alter the surface chemistry of plastic polymers (Lin et al., 2020; Mao et al., 2020; Sun et al., 2020), and affect the strength of the adhesion forces between microplastics and terrestrial surfaces as a result unclear (Burrows et al., 2024; Chen et al., 2024; Li et al., 2024; Mao et al., 2020). The adhesive force between two dry surfaces is made up of a variety of physical interactions: van der Waals, electrostatic and capillary forces, and hydrogen bonding. Due to atmospheric humidity, this dry scenario is rarely relevant. If there exists a surface water film resulting from atmospheric humidity, then the resulting capillary force is the most dominant force governing the interaction between plastic and soil surfaces. Thus, the adhesion force is known to be a function of the relative humidity (RH) (Ando, 2000; Christenson, 1988; L. R. Fisher & Israelachvili, 1981a; Yoon et al., 2003) because it is dominated by a wet bonding capillary force (ie. the meniscus force) caused by condensing water vapor in the gap between two surfaces. As a result, one needs to account for both the specific surface properties of microplastics, as well as the effect of environmental factors such as UV exposure and RH to accurately predict their emission potential. This will address issues raised in the previous chapter. Only two previous studies have looked at adhesion forces on plastics as a result of RH (Lai et al., 2018, 2024), and neither one used SiO_2 tips to proxy the relationship between microplastics and terrestrial surfaces. Additionally, no study has systematically analyzed the relationship between UV exposure, hydrophobicity changes, and changes in adhesion force for plastic. The influence of RH and UV exposure on the adhesion force between microplastics and

terrestrial surfaces, and thus, their emission potential is yet unclear. Thus, it is important to examine these environmental conditions which could influence the detachment of microplastics from soil, and their emission potential into the atmosphere. This could contribute to our fundamental understanding of the forces controlling microplastic emission, and consequently, their inhalation exposure risk. Understanding these dynamics is crucial for accurately predicting the release of microplastics from various environmental matrices, including soil, sediments, and water bodies, into the atmosphere.

1.3. Objectives

The overall objective of this dissertation is to improve our mechanistic understanding of microplastic emission into the atmosphere, and the factors that may affect exposure of human populations to higher levels of emitted microplastics. The dissertation consists of six research chapters that examine the distribution and accumulation of microplastics in terrestrial and atmospheric dust samples, to evaluate their exposure factors and microscale emission mechanisms. Chapter 2 analyzes the spatial distribution of deposited airborne microplastics in terrestrial environments to estimate the relative importance of climate and land-use, based on the analysis of global data. Chapter 3 quantifies and characterizes microplastics found in sediment cores and marine field samples. Chapters 4 and 5 estimate the spatial predictability of deposited airborne microplastics in Los Angeles based on a variety of variables: leaf height, leaf hydrophobicity, land use, socioeconomic status, or proximity to known sources of microplastics. Chapters 6 and 7 use theoretical frameworks and experimental data to investigate how the emission of microplastics from terrestrial surfaces differs from traditional sand emission, based on a wind tunnel experiment and AFM adhesion force measurements, respectively. Specific goals are described below.

Chapter 2 analyzes deposited airborne microplastic data reported in 24 studies from 15 countries to estimate the relative importance of climate and land use classification on their spatial distribution. This study proved that climate has a greater impact than land-use, perhaps due to long-range atmospheric transport of microplastics. This global analysis highlights a lack of standardization of sampling duration and availability of data in the global South, thus providing a framework to improve monitoring. The outcome of Chapter 2 is a peer-reviewed journal article:

> **Leonard, J**., El Rassi, L.A., Samad, M.A., Prehn, S., and Mohanty, S.K. (2024) The relative importance of local climate and land use on the dry deposition rate of airborne microplastics on terrestrial land. *Atmospheric Environment.* *https://doi.org/10.1016/j.atmosenv.2023.120212*

Chapter 3 quantifies and characterizes microplastics on the ocean floor of the Gulf of Mexico, using sediment cores and marine water column samples. Analysis of the depth profile of microplastic concentrations proves microplastic contamination may have accelerated in the last 2 decades. The outcome of the chapter 3:

> **Leonard, J.**, Koutnik, V.S. , Sankar, M. S., Katkar, A., El Rassi, L.A., Choy, M., Brar, J., Glasman, J.B., Cowger, W., Ravi, S., Li., C., Dash, P., and Mohanty, S.K. (202X) Microplastics in the ocean floor in the Gulf of Mexico reveal historic deposition. *In Preparation*.

Chapter 4 analyzes the variability of microplastic concentrations contributed by land-use type, leaf height, and leaf surface hydrophilicity which could inform the development of standard sampling best practices that use leaves as a biomonitoring system. The outcome of Chapter 4:

Leonard, J., Borthakur, A., Koutnik, V.S., Brar, J., Glasman, J., Cowger, W., Dittrich, T.M., Mohanty, S.K. (2023) Challenges of using leaves as a biomonitoring system to assess airborne microplastic pollution in urban areas. *Atmospheric Pollution Research*. 101651. https://doi.org/10.1016/j.apr.2023.101651.

Chapter 5 investigates the extent to which deposited atmospheric microplastic concentrations varies spatially based on socioeconomic variables such as poverty and Green Zone District status, as well as proximity to known microplastic sources such as landfills, wastewater treatment sites, and industrial sites. The results may assist future predictions of microplastic pollution or exposure risks to school children and communities in urban areas. The outcome of Chapter 5:

Leonard, J., Jing, W., Rodriguez, C., Tran, L., Lowe, L., Win Cowger, Patterson, R.F., and Mohanty, S.K. (2024) Spatial Distribution of Microplastics in Atmospheric Dusts Cross-referenced with Socioeconomic Status in Los Angeles, CA, USA. *In Preparation*.

Chapter 6 estimate the enrichment of microplastics in wind-blown sediments on agricultural soils with historic biosolid application and identifies the characteristics of microplastics which may cause this preferential emission using a theoretical framework. The results can update current models for fugitive dust emissions which underestimate the microplastic emission potential of biosolid-amended soils. The outcome of Chapter 6:

Leonard, J., Ravi, S., Mohanty, S.K. (2024) Preferential emission of microplastics from biosolid-applied agricultural soils: Field evidence and theoretical framework. *EST Letters*. https://doi.org/10.1021/acs.estlett.3c00850

Chapter 7 investigates the role that environmental weathering and humidity have on microplastic's adhesion force with terrestrial surfaces, and thus their resistance to emission. The results imply that microplastic's emission potential into the atmosphere could be increased as residence time in the environment increases, or in more humid environments. The outcome of Chapter 7:

> **Leonard, J.**, Lake, M., Stieg, A., and Mohanty, S.K. (202X) UV weathering and humidity effect on the adhesive force between microplastics and sand: Implications on microplastic emission by wind. *In Preparation.*

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2. CHAPTER 2 – THE RELATIVE IMPORTANCE OF LOCAL CLIMATE AND LAND USE ON THE DEPOSITION RATE OF AIRBORNE MICROPLASTICS ON TERRESTRIAL LAND

Leonard, J., El Rassi, L.A., Samad, M.A., Prehn, S., and Mohanty, S.K. (2024) The relative importance of local climate and land use on the dry deposition rate of airborne microplastics on terrestrial land. Atmospheric Environment. https://doi.org/10.1016/j.atmosenv.2023.120212

Abstract

Increasing concentrations of microplastics in the Earth's atmosphere could adversely impact ecosystems when deposited on land, and human health if inhaled. The deposition rate of airborne microplastics may vary widely depending on both land use and climate classification. While land use affects the pool of microplastics available for entrainment into the atmosphere, climatic conditions such as high wind speed or low humidity could increase the emission potential of the microplastics from land surfaces—both factors can synergistically affect microplastic emission. However, it is unclear which of these factors is more influential in affecting the deposition rate of airborne microplastics. Conducting a global analysis of deposition rates (n m^{-2} day^{-1}) based on 24 studies from 15 countries, I found that the deposition rate could vary by 5 orders of magnitude, but longer sampling durations (10 days) could reduce the uncertainty in the measurement of the deposition rate. The deposition rates were higher in arid and tropical climates, which was attributed to hot weather and wind turbulence. Interestingly, the deposition rates did not vary significantly with land use, possibly due to long-range atmospheric transport of microplastics. Overall, analysis of available data reveals that climate could have a greater impact than land use on the concentration and deposition rate of airborne microplastics, and a longer sampling duration could minimize measurement variability in deposition rates.

2.1. Introduction

Prolonged use of plastics has resulted in a rapid accumulation of microplastics on Earth's surface, from where they can be suspended by wind, transported through the atmosphere, and deposited back on lands across geographical boundaries (Bullard et al., 2021a; Habibi et al., 2022; Rezaei et al., 2019, 2022a). For this reason, microplastics have been frequently found in remote locations such as Arctic Sea ice (Peeken et al., 2018), remote mountain tops (Allen et al., 2019; Feng et al., 2020), and protected national parks (Brahney et al., 2020a). High concentrations of microplastics in the air can increase inhalation risks (Prata et al., 2020), and the inhalation of small airborne microplastics, particularly with sizes smaller than 10 μm, could potentially cause adverse health effects in humans (Prata, 2018a; Prata et al., 2020). Microplastics, like traditional particulate pollutants, are expected to remain in the oro-pharyngeal region if they have aerodynamics diameters between 10-30 μm, can reach tracheobronchial regions if they are between 2-10 μm, and are deposited in alveoli if they are between 0.5–2 μm (Thakur et al., 2020). The lower the penetrations in the lungs, the stronger the adverse effect on respiratory function (Xing et al., 2016). However, the aerodynamic diameter of microplastics has not been measured in the reported studies, although it is expected to vary widely due to variations in the density and shape of microplastics. Nevertheless, a few anecdotal studies have found microplastics in the human bloodstream (Leslie et al., 2022a) and lung tissue (Amato-Lourenço et al., 2021a; Jenner et al., 2022), indicating respirable microplastics from the atmosphere can find their way into the lower part of the lung. As many microplastics may adsorb heavy metals and toxic organic chemicals, leaching of the chemicals from microplastics into the body fluid could further elevate health risks (Borthakur et al., 2021b; Rodrigues et al., 2019). Microplastic inhalation risk can be determined based on the deposition rate of microplastics in an area. Additionally, these deposited microplastics

could adversely affect soil structure, bulk density, soil water evaporation rates, and nutrient immobilization in soil with nanoplastics finding their way into plant roots and soil microbiota (Rillig et al., 2019). Thus, it is critical to understand how major factors including climate and land use affect the deposition and accumulation rate of airborne microplastics.

The deposition rate in an area depends on the concentration of airborne microplastics, which in turn depends on the number of microplastics entrained from land into the atmosphere by wind and the fraction of airborne microplastics that remain suspended in the atmosphere (Crossman et al., 2020; Koutnik et al., 2021). The mass transfer of microplastics from land to the atmosphere is influenced by microplastic generation on the land surface, and exacerbated by land use and plastic waste management methods (Lebreton & Andrady, 2019; Lebreton et al., 2017). In urban areas, high concentrations of microplastics are generated due to a higher level of use of plastic-containing products, mechanical abrasion of tires and plastic products, and emissions from incomplete combustion of incinerated plastic wastes (Abbasi et al., 2017; He et al., 2019; Qiu et al., 2020). In rural or agricultural areas, the sources of microplastics are typically plastic mulch films (Huang et al., 2020; Li et al., 2022; Steinmetz et al., 2016) or biosolid fertilizers (Braun et al., 2021; Koutnik et al., 2021; Weithmann et al., 2018). Irrespective of microplastic sources in both types of areas, the released microplastics typically accumulate in the top 0-10 cm of soil (Koutnik et al., 2021), from where they can be preferentially suspended by wind into the atmosphere (Abbasi et al., 2023; Borthakur et al., 2021b; Crossman et al., 2020a). Additionally, airborne microplastics can migrate considerable distances from their sources, thereby diluting the contribution of land use on the deposition rate of airborne microplastics. This long-range transport has been attributed as the primary cause of elevated microplastic concentrations in remote natural habits (Brahney et al., 2020a).

Local climates could influence how fast macroplastics can break down to produce microplastics on land, their emissions from land into the atmosphere, their transport across land-use boundaries, and their deposition rate back on land surfaces—just as climates control emissions of natural particles including soil minerals (Reheis & Urban, 2011; M. Wu et al., 2020). Factors associated with specific climate classifications, including temperature, precipitation, wind, and UV irradiation, affect the physical and chemical processes that lead to the breakdown and transport of microplastics in the environment (Chamas et al., 2020; Corcoran, 2022; Zhang et al., 2021). In warmer climates, plastics may disintegrate faster due to increased exposure to heat and sunlight, leading to higher accumulation rates of microplastics on land (Corcoran, 2022). Wind can then entrain these microplastics into the atmosphere. Climatic conditions, such as rainfall and storms can cleanse the atmosphere of dust (Mbachu et al., 2020; Roblin et al., 2020)- and microplastics in dust conglomerates- and deposit them on land surfaces (Koutnik et al., 2021). Thus, the net amount of airborne microplastics in a region and their recorded deposition rates could be affected by a) the source region's land use, which provides the source or pool of microplastics for emission, and b) climate classification, which represents the available energy for microplastic degeneration, emission, and transport. Yet, no study to date has analyzed the relative importance of these factors on the overall deposition rate of airborne microplastics. Furthermore, data collected from different studies adopted different protocols such as dust collection duration, area of samplers, and size cut off, all of which could make it difficult to compare results between the studies. Analysis of deposition data and collection protocol could reveal any sampling biases related to the choice of sampling protocol.

This study identified trends in recorded microplastic deposition rates in regions of different land use and climate classification. Analyzing 24 studies that measured microplastic deposition

rates (particle m^{-2} day^{-1}) in 15 countries, I answer the following questions: To what extent do the deposition rates of microplastics vary globally? Can the deposition rate of airborne microplastics be predicted by a region's degree of dryness as defined by its climate classification? Does land-use pattern affect measured deposition rates? How does the sampling duration affect the reported variability of the deposition rates? In the last decades, many reviews on airborne microplastics have highlighted the sources, methodology techniques, models, and composition of airborne microplastics without using global data to inform their research questions (G. Chen et al., 2020; Hou et al., 2020; R. Li et al., 2020; Luo et al., 2022; Mbachu et al., 2020; O'Brien et al., 2023; L. Shao et al., 2022; A. Xu et al., 2022; H. Yang et al., 2021). In contrast, this study analyzes global data to confirm quantitatively if microplastic deposition rates on terrestrial lands can be predicted by a region's land use and Köppen-Geiger climate classification.

2.2. Data collection and analysis

I searched reported studies listed in Google Scholar and Web of Science prior to February 28, 2023, using keywords including "airborne microplastics", "microplastics in the atmosphere", "aeolian microplastics", and a combination of similar words. Only studies reporting the deposition rate (particles or n m^{-2} day^{-1}) were included because other units such as n g^{-1} or n m^{-3} provide the cumulative accumulation of microplastics for an unknown period of time without quantifying the deposition rate. A total of 61 articles have reported atmospheric microplastic concentrations but only 24 studies have reported the deposition rate (**Table 2-1**). These 24 studies were from 15 countries in North America, South America, Europe, and Asia (**Figure 2-1**). These locations span a wide range of climatic conditions as defined by Köppen-Geiger climate classifications at 0.083° spatial resolution (Beck et al., 2018). The reported studies were classified in 5 climatic groups (arid, continental, temperate, tropical, polar) based on spatial analysis performed in ArcMap

10.8.1, when overlayed over a public map of the Köppen-Geiger Climate Classification as published by World Bank. The reported studies are primarily located within around 30° of the equator, where UV irradiation levels are relatively high, thereby potentially accelerating the fragmentation of macroplastics into microplastics. A Tukey one-way test was used to compare microplastic concentrations as a function of climate and land use types, where a p-value < 0.05 is assumed as a statistically significant difference.

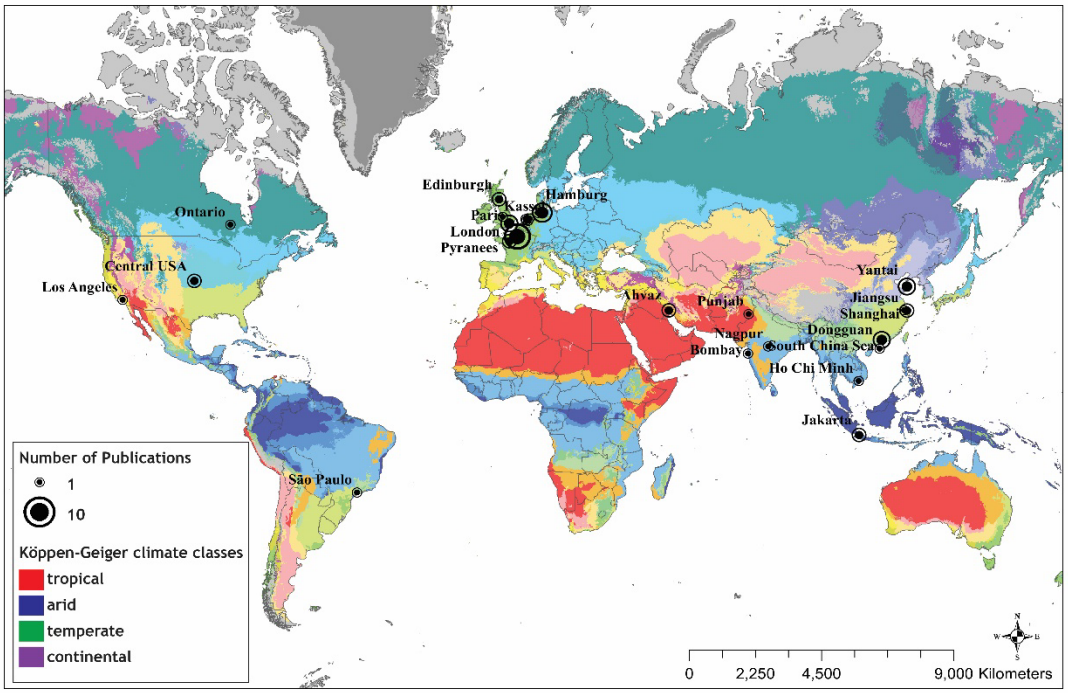

Figure 2-1: Locations of 24 studies that reported the deposition rate (n m^{-2} day^{-1}) of airborne microplastic, where n is the number of microplastics. The climate types and boundaries are based on present-day Köppen-Geiger climate classifications at 0.083° spatial resolution. Four of the major climatic zones were analyzed in this paper as most data were available in these regions.

Table 2-1: List of studies that reported microplastic deposition rates in different locations with different climates

Location	Climate	Deposition Rate (n m^{-2} day^{-1})	Sampling Time (days)	Detection Limit (µm)	Reference
Ahvaz, Iran	Arid	1754-3157	57	10	(Abbasi et al., 2023)
Pyrenees, France	Temperate	402-462	12-14	10	(Allen et al., 2019)
São Paulo, Brazil	Tropical	76.23-272.70	15	50	(Amato-Lourenço et al., 2022)
China, France, Germany, Indonesia, UK	Continental, Temperate, Tropical	2-2420	NR	NR	(Bao et al., 2023)
Protected National Parks, West USA	Temperate	48-435	30-60	5	(Brahney et al., 2020a)
Dongguan, China	Temperate	175-313	30	NR	(Cai et al., 2017)
Edinburgh, UK	Temperate	166666- 833333	0.0125-0.025	NR	(Catarino et al., 2018)
Paris. France	Temperate	2-355	~ 15	50	(Dris et al., 2016)
Paris, France	Temperate	1600-11000	0.42-1.67	50	(Dris et al., 2017)
Various Cities, Germany	Temperate	9.9-340	30	11	(Kernchen et al., 2021)
Hamburg, Germany	Temperate	136.5-343.3	15	50	(Klein & Fischer, 2019)
Los Angeles, USA	Temperate	1500-196000	NR	10	(J. Leonard et al., 2023)
Shanghai & Jiangsu, China	Temperate	700- 1900	NR	10	(K. Liu et al., 2020)
China, France, UK	Temperate & Tropical	36- 110	NR	NR	(Mbachu et al., 2020)
Wageningen & Weserbergland, The Netherlands	Temperate	2640-167280	0.42	NR	(Nizamali et al., 2023)
France, Germany, China	Temperate & Tropical	100-12300	NR	NR	(Prata et al., 2022)
Jakarta, Indonesia	Tropical	6-31	120	5 mm	(Purwiyanto et al., 2022)
Punjab, Pakistan	Arid	810667-5973333	0.021		(Qaiser et al., 2023)
China France, Germany, India, UK, USA, Vietnam	Arid, Continental, Temperate, Tropical	36-1801	NR	NR	(Sridharan et al., 2021)
London, UK	Temperate	575-1008	3-4	20	(Wright et al., 2020)
Ontario, Canada	Continental	4-9	3-48	50	(Welsh et al., 2022)
Bombay, India	Tropical	8101-59777	1	NR	(H. Yadav et al., 2022)
China, France, Germany, UK	Continental, Temperate, Tropical	36- 11000	NR	NR	(Q. Zhang et al., 2020)
Yantai, China	Continental	130-624	90-120	5	(Zhou et al., 2017)

***NR: not reported.**

2.3. Effect of land use

2.3.1. Effect of land use on the generation of airborne microplastics

The extent of plastic use on land varies with land use type, which in turn could affect the concentration of microplastics available for suspension by winds from terrestrial surfaces (**Figure 2-2**). Some of the emitted microplastics can be deposited back within the same locality in the absence of long-range microplastic transport, especially since aeolian microplastics including: PM 10, PM 2.5, brake wear/ tire wear particulates, tend to be suspended into the surface mixed boundary layer (∼1.5 km) where a residence time is typically 8.3-37 days (Evangeliou et al., 2020a). In that case, land use types could affect the total deposition rate of microplastics at a location. **Table 2-2** highlights existing studies that link land use types and plastic usage due to anthropogenic modifications to microplastic concentration. Due to a decrease in human activity in different lands, plastic usage is expected to decrease with the following order of land use: urban (residential, commercial, industrial), coastal, agricultural, grassland, forest, and mountains.

The level of plastic used and the method of their use in a region could affect the number of microplastics available for potential suspension into the atmosphere by wind. For instance, highly urbanized lands should have higher concentrations of microplastics in the atmosphere due to the increased use of plastic products during construction (Qiu et al., 2020), industrial, and household activities (Galvão et al., 2020; Guerranti et al., 2019; Hernandez et al., 2017). In urban areas, most microplastics released could be attributed to a rapid increase in single-use plastic products such as water bottles, cups, and plates (Laskar & Kumar, 2019; Nizamali et al., 2023; Oßmann, 2021) and the release of plastics from tires on roadways (Baensch-Baltruschat et al., 2020). Thus, a region's level of plastic usage and their accumulation on land could be predicted by its population density (Baldwin et al., 2016; Guan et al., 2016; Koutnik et al., 2022; Schmidt et al., 2017). For the same

reason, populated coastal regions could contain high concentrations of microplastics. The breaking of ocean waves could also release microplastics from ocean water into the atmosphere in the coastal regions (Chubarenko et al., 2018; Szewc et al., 2021).

In agricultural regions, microplastics can be released from plastic mulch or plastic covers typically used to control weeds, conserve soil moisture, and increase soil temperature (Steinmetz et al., 2016; Tian et al., 2022) and from wastewater biosolids (Borthakur et al., 2021b; Crossman et al., 2020a; Koutnik et al., 2021a; Mohajerani & Karabatak, 2020; Rezaei et al., 2022a; Zubris & Richards, 2005). These microplastics can be suspended by different agricultural activities such as crop-dusting and tilling (Abbasi et al., 2023; Maffia et al., 2020; Nordstrom & Hotta, 2004). Furthermore, unlike urban areas with high-rise buildings, agricultural lands offer limited resistance to wind flow, thereby increasing the potential of their emission. Collectively, these factors often contribute to an increased rate of dust emission in agricultural lands compared to urban lands (Katra, 2020; Lee et al., 2012). For the same reason, microplastic emissions from agricultural lands could be higher than urban lands within the same climate or regions. Because of vegetation, microplastic emission in bare agricultural land could be higher than that of agricultural land with vegetation (Barwise & Kumar, 2020; Gonzales et al., 2018; Helcoski et al., 2020). The vegetation coverage and height of plant species, which affect relative wind flux across landscapes, could vary based on land use types such as grassland and forests. Forests with higher canopies and thicker vegetation could lower ground-level wind speeds and thus lower emissions of dust and microplastic particles (Barwise & Kumar, 2020; Helcoski et al., 2020). Therefore, forests can often act as a net sink of microplastics. Consequently, measurements of vegetation covers by remote sensing (Jones & Vaughan, 2010; Myneni et al., 1995) and activities on ground that typically use plastic products (R. M. Harrison et al., 2012; Qiu et al., 2020) could determine the extent to which

land use type affect microplastic emissions and their depositions in an area. Future studies should integrate remote sensing data on vegetation with ground-level activities that may release microplastics to inform the potential of emission and deposition of airborne microplastics in a region.

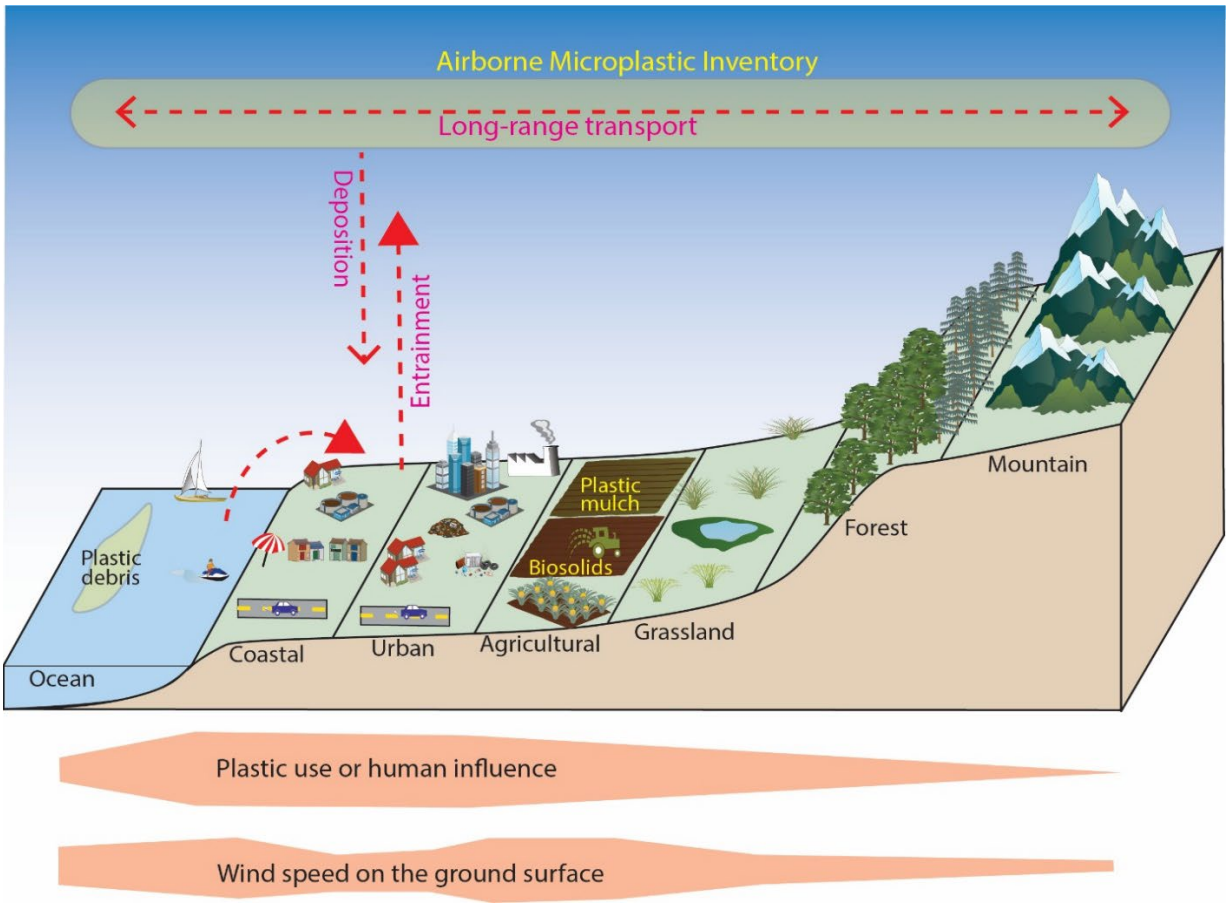

Figure 2-2: Illustration depicting the role of land use affecting the emission into and deposition of microplastics from atmospheric microplastic inventory.

Table 2-2: List of studies highlighting the effect of land use on the generation, emission and deposition of airborne microplastics

Land Use	Description	Sources	Relative Wind	Human Influence	Highlighted References
Residential, Commercial, Industrial Urban	High density of human populations for living and working, characterized by lots of buildings and structures.	- Roads/tyre abrasion - Mismanaged anthropogenic waste - Household products - parks/play structures - Lawn fertilizers - construction - Atmospheric transfer and deposition	Low	High	(Abbasi et al., 2018; Cai et al., 2017; Dehghani et al., 2017; J. Leonard et al., 2023; Zhou et al., 2017)
Coastal	Medium to high density of human populations, experiencing the strong wind fluxes across the ocean-land boundary	- Roads/tyre abrasion - Mismanaged anthropogenic waste - Marine waste -boating/harbor equipment - Marine plastic debris - Atmospheric transfer and deposition	High	High	(Abayomi et al., 2017; Allen et al., 2020; Purwiyanto et al., 2022; Qaiser et al., 2023; K. Zhang et al., 2017)
Agricultural	Low to medium density of human populations, focused on crop and cattle production across relatively flat landscapes	-fertilizers/ biosolids - lining/agricultural equipment - Atmospheric transfer and deposition	Medium	Moderate	(Crossman et al., 2020a; Lwanga et al., 2023; Mohajerani & Karabatak, 2020; Rezaei et al., 2022b; J. Yang et al., 2021)

Grassland	No to low human density with relatively flat landscapes and short, constant vegetative growth and high sunlight	- Atmospheric transfer and deposition	High	Low	(Corradini et al., 2021; Feng et al., 2020; B. Huang et al., n.d.; Y. Zhang et al., 2021)
Forest	No to low human density with tall, constant vegetative growth and shade due to canopy	- Atmospheric transfer and deposition	Low	Low	(Barasarathi, 2014; Choi et al., n.d.; Evangeliou et al., 2020b; Maghsodian et al., 2022; Ng et al., 2021)
Mountains	No to low human density with high winds due to high altitude, high UV, and high precipitation	- Atmospheric transfer and deposition	High	Low	(Allen et al., 2019; Bian et al., 2022; Bilal et al., 2023; Free et al., 2014; Padha et al., 2022)

2.3.2. Global data analysis of land use effects on deposition rates of airborne microplastics

Based on analysis of the available dataset, I showed that the deposition rates of airborne microplastic were not influenced by regional land use in temperate climates (**Figure 2-3**). The mean deposition rates in rural, remote, and urban areas were statistically similar ($p > 0.43$); however, variation was dependent on land use. Including all data, deposition rates varied by 5 orders of magnitude, although the variation was smaller within the subset of land use. The deposition rates in urban areas varied by three orders of magnitude, and the variation was reduced to one order of magnitude in rural and remote areas. This variation could be attributed to true variation in concentration, or from reporting errors due to a lack of standardized sampling protocols. Due to the high population density in urban areas, microplastic emissions in urban areas are expected to be higher than the rates in remote areas (Koutnik et al., 2021). High emissions of microplastics should correlate to high emissions of dust in urban areas (Amato et al., 2014; Guan et al., 2016). High emissions could increase the inventory of airborne microplastics and thus could influence their deposition rate. However, similar deposition rates indicate that any preferential effect of land use on microplastic emission is negated by the atmospheric transport of emitted microplastics between land use types. The data analysis indicates that geographical proximity to potential sources may not determine atmospheric concentrations of microplastics, since microplastics emitted from one place can be dispersed across the atmosphere-surface boundary layer and deposited elsewhere. However, the data set is small in remote and rural areas ($n = 21$) compared to urban areas ($n = 101$), which could skew the results. Thus, more studies should be conducted to measure the deposition rate of microplastics in these understudied remote and rural areas, to improve the strength of the limited conclusions presented in this study.

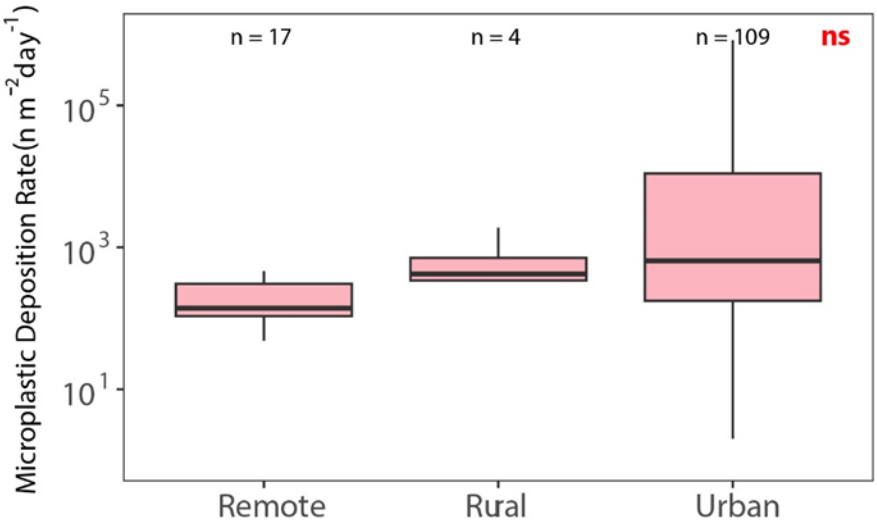

Figure 2-3: Deposition rate of airborne microplastic in temperate climatic regions across three different land use types: remote, rural, and urban. n is the number of data points for each category. Deposition rates reported in locations with different land use were not statistically different based on the pairwise Tukey one-way test.

2.4. Effect of Climate

2.4.1. Effects of climatic conditions on the generation and suspension of airborne microplastics

The Köppen-Geiger climate classification of a region indicates its relative temperature, UV radiation, relative humidity (RH), and wind energy. All these factors could play a significant role in determining atmospheric microplastic deposition rates (**Figure 2-4**). While temperature and UV radiation can affect the breaking down of macroplastics (Chamas et al., 2020a; Corcoran, 2022; Song et al., 2017), humidity and wind energy can affect the extent to which the microplastics can be suspended by wind (Katra, 2020; Ravi et al., 2004). Climate types with higher temperatures or UV intensity can increase the physical and mechanical breakdown of primary into secondary microplastics, which are finer and more likely to resuspend (Chamas et al., 2020a; Corcoran, 2022). This is especially true for plastic products that are exposed to UV irradiation from sunlight, which has been shown to increase the generation of smaller microplastics prone to emission (Cai et al., 2018; Song et al., 2017; Suhrhoff & Scholz-Boettcher, 2016).

Moisture films formed on soil due to atmospheric humidity could bind microplastics or other particles on land surfaces and reduce emission. Lower moisture levels in terrestrial environments can decrease cohesion and the ability of particles to stick together, which in turn amplifies dust emissions due to decreased threshold velocity (**Equation 2-1**).

$$u^* = \sqrt{\tau/\rho} = f\sqrt{|\Psi|A_c} = f\sqrt{|\ln RH|\left(\frac{\delta}{\delta_o}\right)^n} \quad (2\text{-}1)$$

where u^* is threshold wind friction velocity; τ is threshold shear stress force; Ψ is matric potential measured directly or estimated from Kelvin equation and RH; A_c is the contact area between adsorbed water and adjacent particle; RH is relative humidity; δ is the thickness of the adsorbed film; δ_o is the thickness of a monolayer of adsorbed water ($\delta_o = 0.3 \times 10^{-9}$ m) (McKenna Neuman & Sanderson, 2008). The humidity level varies based on local climate and location. This offers the opportunity in future to use remote sensing data (Cooper et al., 2021; Moghavvemi et al., 2005; Rhee et al., 2010) to record humidity and thus emission potential of microplastics in different climate regions.

I listed possible variations of climatic moisture across different climate classifications and its effect on microplastic emission and deposition in regions (**Table 2-3**). Storm events—characterized by high wind and precipitation conditions—vary widely by climate classification. In general, highly turbulent wind events can resuspend more microplastics into the air (Rezaei et al., 2019), while weaker wind conditions in other climates or seasons may not be sufficient to resuspend microplastics. Microplastics and other natural dust are emitted in large quantity when wind speeds in a region meet a threshold wind velocity u^* (**Equation 2-2**):

$$u^* = A_N\sqrt{\frac{\rho_p - \rho_a}{\rho_a} g D_p + \frac{\gamma}{\rho_a D_p}} \quad (2\text{-}2)$$

where u^* is the threshold wind velocity; ρ_a is the air density; ρ_p is the microplastic density; d_p is the microplastic diameter; g is the acceleration due to gravity; A_N is a dimensionless parameter; and γ is a parameter that scales the strength of the interparticle forces between the microplastics and soil surface (Cornelis et al., 2004; Mckenna Neuman, 2003; Ravi et al., 2004).

High precipitation surges may move microplastics on the Earth's topsoil surfaces, especially within the top 0-10 cm where smaller microplastics are concentrated (Besley et al., 2017; Koutnik et al., 2022b) and affect the availability of microplastics for suspension during the following dry weather. Precipitation events may also clear out airborne microplastics depositing them back onto the Earth's surface (Baatar et al., 2017; Speirs et al., 2023). Thus, the combined effect of wind and precipitation can either increase or decrease deposition rates. However, most studies do not report the humidity or date of the last storm event in the sampling location. Air quality has long since been linked to climatic factors particularly extreme dry and windy conditions. These conditions are expected to intensify during changing climate (Fiore et al., 2015; Jacob & Winner, 2009). Thus, long-term monitoring efforts should be carried out to estimate changes in atmospheric microplastic deposition due to climate change.

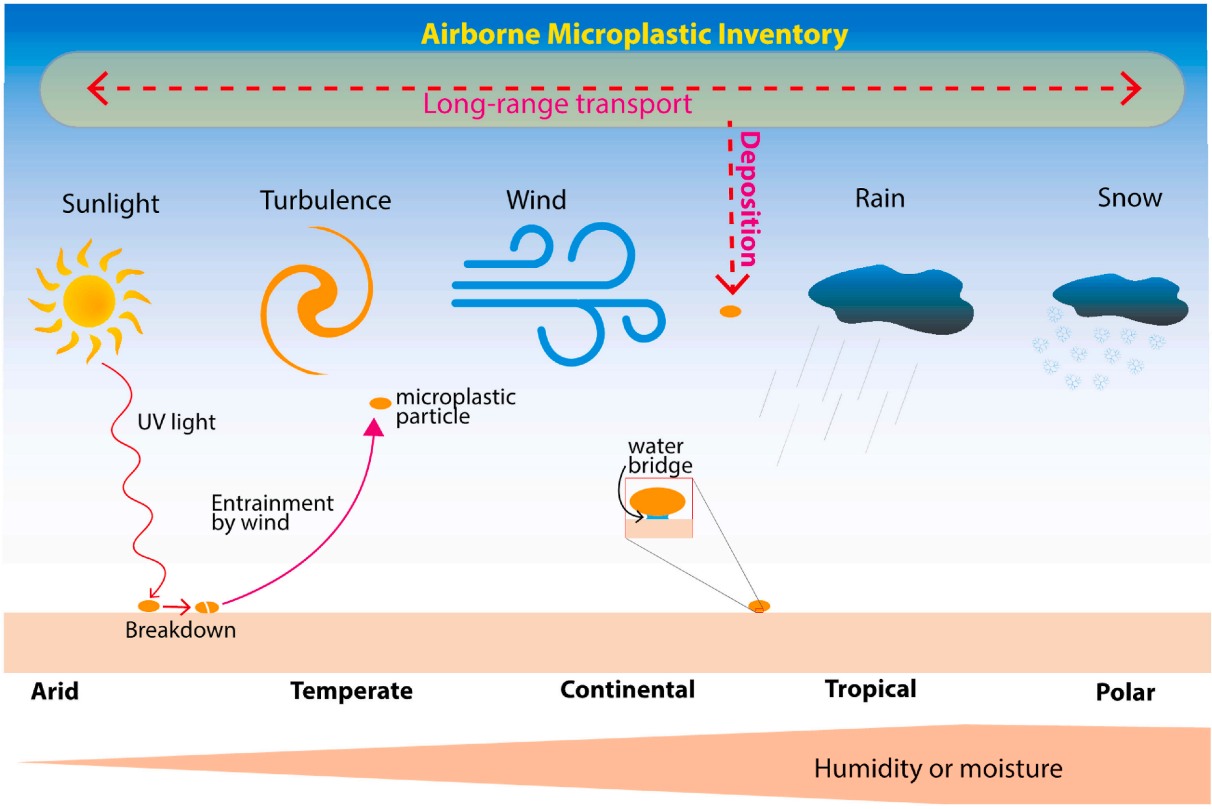

Figure 2-4: Illustration depicting the role of climate factors affecting the emission and deposition of airborne microplastics.

Table 2-3: List of studies highlighting the effect of climate factors on the generation, emission and deposition of airborne microplastics

Climate	Description	Relative Moisture	Relative Storm* Frequency	Highlighted References
Arid	Hot, dry climate receiving less than 10 inches (25.4 centimeters) of rainfall per year, often deserts	Low	Low	(Abbasi et al., 2023; Boakes et al., 2023; Chandrakanthan et al., 2023; Guan et al., 2016; O'Brien et al., 2020)
Temperate	Moderate seasonal rainfall, mild to warm summers and cool to cold winters	Medium	Medium	(Kernchen et al., 2021; Kyriakoudes & Turner, 2023; Nizamali et al., 2023; Roblin et al., 2020)
Continental	Regions with high seasonal variability, very warm summers and cold winters, usually inland	Medium-High	Medium-High	(Dong et al., 2023; Eo et al., 2021; Gasperi et al., 2015; Martin et al., 2017a; X. Wang et al., 2021)
Tropical	Hot, wet climate with constant temperatures of 18 °C (64.4 °F) or higher; and generally high annual precipitation, usually low elevations	High	High	(Y. Huang et al., 2021; Sridharan et al., 2021; Truong et al., 2021; Viet Dung et al., 2021; H. Yadav et al., 2022)
Polar	A very cold climate in which no monthly average goes above 10 °C (50 °F), oft at high latitudes	High	Medium	(Cunningham et al., 2020; González-Pleiter et al., 2020; Mishra et al., 2021; Peeken et al., 2018; Tirelli et al., 2020)

* Storm events are characterized by a period of high wind and precipitation

2.4.2. Data analysis on climatic conditions

My analysis showed that deposition rates in arid and tropical regions were statistically significantly ($p < 0.001$) higher than deposition rates reported in continental and temperate ones (**Figure 2-5**). I attributed the higher deposition rate of microplastics in arid regions to lower humidity. Lower humidity typically decreases the formation of moisture film between particles and the soil surface, increasing the potential for dust emission and, subsequently, their fall out back to land (Bisal & Hsieh, 1966). As airborne microplastics are simply a type of dust, their emission and deposition should follow similar principles that govern the emission of other soil minerals. Thus, it is expected that an increase in humidity could limit the emission of microplastics or their concentration in the atmosphere. Surprisingly, the deposition rate was higher in tropical climates, where conditions oscillate between humid and warm. I speculated that increased microplastic concentrations and deposition in the tropics could be linked to the increasing density of airborne microplastics from the adsorption of atmospheric moisture and the accelerated degradation of synthetic polymers on land supplying more microplastics for entrainment. Tropical climate types are ideal for the decomposition of organic material (Butenschoen et al., 2011; Davidson & Janssens, 2006). Thus, macroplastics may disintegrate or decompose in tropical regions faster than in other climate types, forming microplastics for emission during frequent energetic weather events. However, studies examining the decomposition rate of microplastics in different climatic conditions are limited. High humidity could also change the density of airborne microplastics due to the adsorption of water and increase deposition (Szewc et al., 2021). Future studies should measure the decomposition rates of plastics in tropical climates and the role of humidity on the deposition rate of airborne microplastics to evaluate the potential causes of high deposition rates in tropical regions.

The analysis shows that deposition rates did not vary statistically significantly between temperate and continental climates ($p > 0.9974$). These climates exhibit similar seasonal behavior with a slight difference in the timing of their dry season and the magnitude of their seasonal extremes. The results indicate that despite climate controlling the processes that affect the physical generation of microplastics and their emission by wind, the transportation of microplastics across climate boundaries could further complicate the prediction of deposition rates based on land use or climate. Microplastics have been reported to travel anywhere from 275 km to 10,212 km in the atmosphere (Allen et al., 2021), making conclusions based on geographical location somewhat inconclusive.

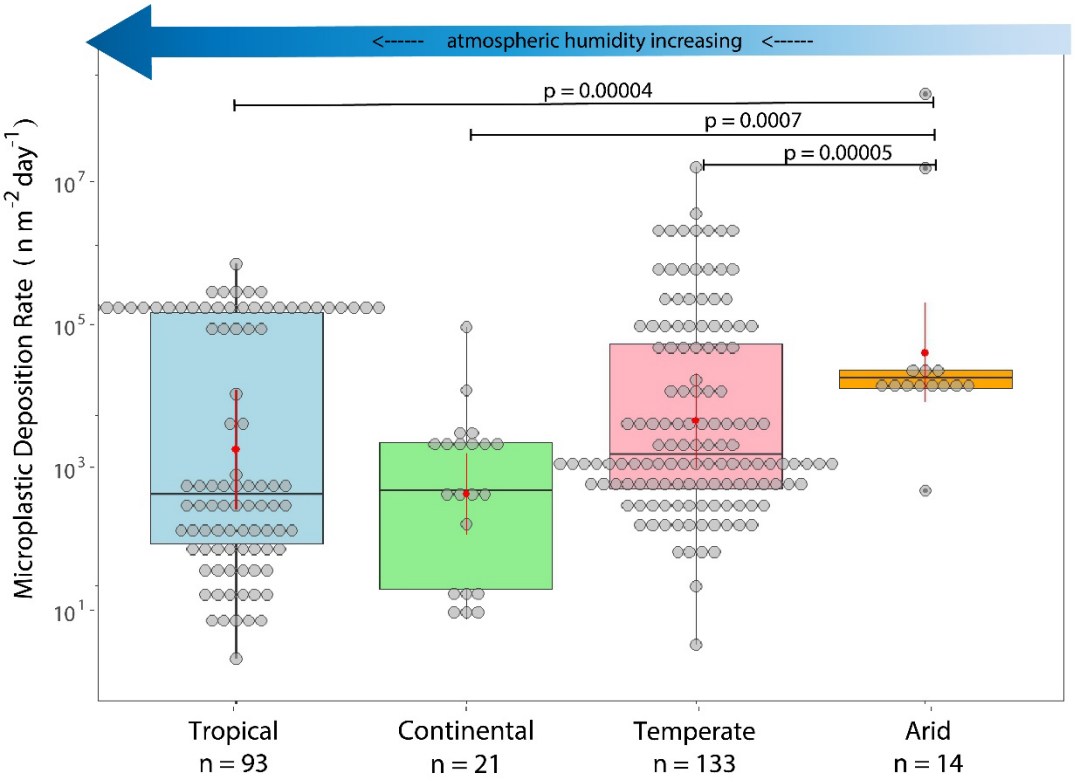

Figure 2-5: Deposition rate of airborne microplastics across four major climatic zones. Climatic zones are plotted from left (wetter) to right (drier) in terms of atmospheric humidity levels or the number of warmer or drier days. Statistically significant p-values from pairwise Tukey one-way tests are depicted on the top.

2.5. Challenges and Opportunities

Deposition data in diverse climates and regions: Global analysis identified regions or climates where data was not available, thereby informing the need for future studies in those specific areas. Most studies reported deposition rates in tropical and temperate regions, with limited data available from arid or continental climate zones, limiting the strength of the conclusions in this analysis. Similarly, the potential cause of the high deposition rate in tropical regions is unknown due to a lack of fundamental studies in the tropics. Extremely limited data was available in polar regions or regions further from the equator in the Southern Hemisphere, specifically in Africa (no study to date) and South America (one study).

Fundamental studies examining the effect of climate, land use, and surface properties on microplastic emissions: Numerous studies have examined the fundamental processes by which soil minerals or wind-blown sediments contribute to the pool of dust particles circulated in the atmosphere and deposited back on land (Gillette & Passi, 1988; Kok & Renno, 2006; Loosmore & Hunt, 2000). These studies involve manipulated wind tunnel experiments on the field and laboratory to simulate the effects of different variables including humidity (McKenna Neuman & Sanderson, 2008), land surface heterogeneity (Carmona et al., 2015; Darmenova et al., 2009; Gonzales et al., 2018), sediment properties (Reynolds et al., 2007; Z. Zhang et al., 2016), and wind conditions (Shao & Lu, 2000; Tian et al., 2023; Webb et al., 2016). However, similar studies are needed to improve the fundamental understanding of when, how, and to what extent microplastics may be emitted into the atmosphere or deposited back onto land, including their typical transport range. For instance, the effect of hydrophobicity and lower density of microplastic could play a significant role in the magnitude of microplastic emissions, and future models should be developed including these properties. Furthermore, future studies should report climate, land use, humidity,

and the most recent storm event in the sampling location to standardize the global dataset so that the contribution of each factor to microplastic deposition rates can be better assessed.

Improved sampling and reporting protocols: The literature also reveals a need for standardization of reported information in units, detection limit, and sampling protocols. The studies reported here rarely followed a uniform protocol leading to variations in sample collection methods, detection of microplastics, and reporting of data. For example, a uniform sampling duration was rarely followed when recording deposition rates. My analysis revealed that the use of a short sampling duration (< 1 day) could lead to an overestimation of the deposition rate by several orders of magnitude (**Figure 2-6**). For instance, sampling durations under 1 hour resulted in two to four orders of magnitude higher deposition rates than those observed for longer durations. In contrast, the deposition rate did not vary when the sampling duration exceeded 10 days, indicating any experimental artifacts or temporal variability can be averaged out when a sample was collected for a longer period. Thus, I recommend future studies to use a sampling duration of at least 10 days.

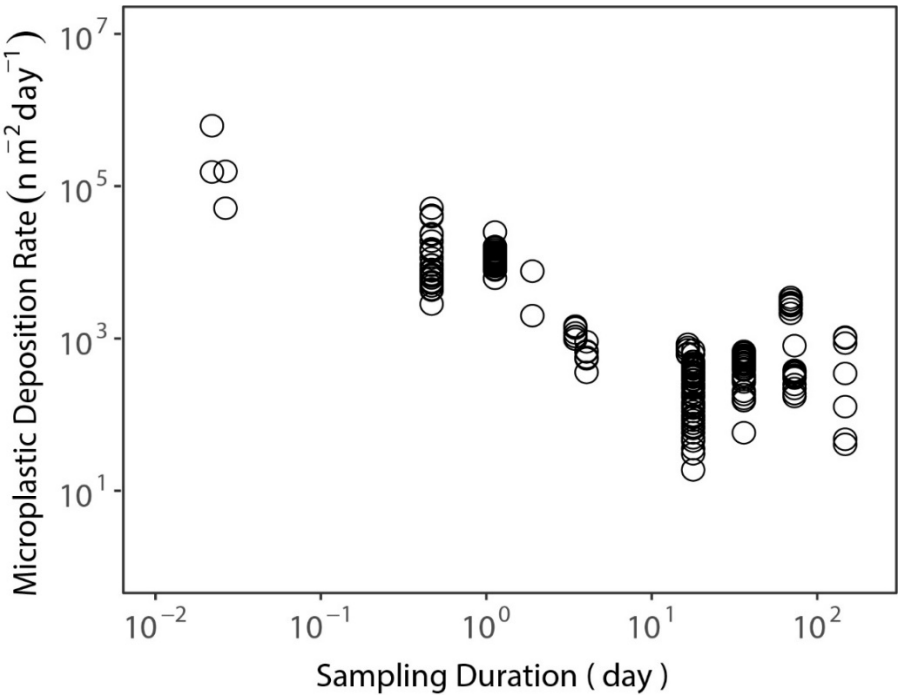

Figure 2-6: Deposition rates vary with the duration of sampling time, indicating the need for adopting a protocol that requires 10 days or longer duration for sample collection to minimize temporal variability in a short time.

A sensitive method to detect inhalable microplastics: Analysis reveals that most studies rarely reported the detection limit, or the smallest microplastics accounted for in the method. Only 52% of reviewed studies reported their detection limit (**Table 2-1**). As deposition rates could depend on particle size, all studies should clearly state the detection limit for the methodology used within. Most studies tend to underreport smaller microplastics (< 10 μm), which are more likely to cause adverse health effects. For transparency of data and to streamline future synthesis into a large-scale database, reporting protocol should include detection limits. Furthermore, more studies should use advanced tools to identify and detect microplastics with sizes less than < 10 μm. Additionally, microplastics reported in these reviewed publications were extracted from the dust particle agglomerates (Mahowald et al., 2014; Onyeagusi et al., 2023) that suspended them, and it is these agglomerate sizes that determines their inhalation risk, and which should be assessed in future inhalation risk publications.

2.6. Conclusions

This work analyzed global data to examine the relative importance of climate and land use on microplastic deposition rates. Analyzing the deposition rates (n m^{-2} day^{-1}) of airborne microplastics reported in 24 studies from 15 countries, I show that the deposition rate could vary by 5 orders of magnitude. The variation was attributed to the duration of sampling, climate, and land use. My analysis shows that the deposition rate could increase by orders of magnitude if the deposition rate was calculated based on sample collection within 1 day. Conversely, deposition rates did not vary when the sampling duration exceeded 10 days. Thus, the deposition rate should be estimated based on samples collected over 10 days or longer to minimize experimental variability or artifacts. Future studies could confirm the minimal sampling duration that can minimize variation. Higher deposition rates were observed in the arid and tropical climates. Deposition rates in temperate and continental climates were similar. Deposition rates did not vary significantly with land use, potentially due to the long-range transport of airborne microplastics across geographical boundaries. In summary, the analysis highlights that a region's climate classification could play a greater role than land use in predicting the deposition rate of airborne microplastics.

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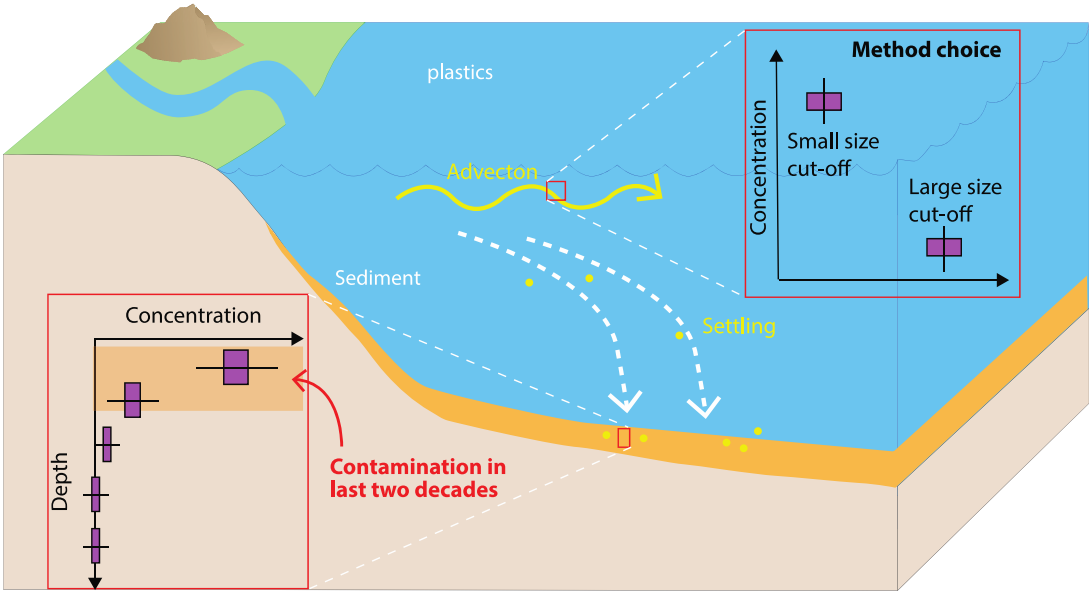
3. CHAPTER 3 – MICROPLASTICS IN OCEAN FLOOR SEDIMENTS IN THE GULF OF MEXICO: IMPLICATION ON THEIR HISTORIC DEPOSITION



Leonard, J., Koutnik, V.S., Sankar, M. S., Katkar, A., El Rassi, L.A., Choy, M., Brar, J., Glasman, J.B., Cowger, W., Ravi, S., Li., C., Dash, P., and Mohanty, S.K. (202X) Microplastics in the ocean floor in the Gulf of Mexico reveal historic deposition. In Preparation.

Abstract

The ocean floor in the Gulf of Mexico, particularly near the mouth of the Mississippi River, has been polluted with plastics ever since large-scale plastic production began in the early 1950s. Yet, the historic deposition of plastics on the ocean floor in this region has not been evaluated due to a lack of analysis of microplastic concentrations in different depths of sediment deposits on the ocean floor. This study aims to analyze microplastic concentrations in sediment cores up to a depth of 20-30 cm from the ocean floor in the Gulf of Mexico and compare the plastic type and size abundances of microplastics found in sediment cores and in the water column. Microplastic concentrations in the water column above the sediment cores averaged around 1.6×10^2 pieces per L, which is at least three orders of magnitude higher than that previously reported in the same region. We attributed the increase in concentration to a) the sampling location in the Mississippi River's region of influx and b) to a lower size cutoff (5-20 μm) for microplastic detection compared to that used in other studies (>100 μm). The sediment contained more diverse types of plastics, although polyethylene and rayon were the most abundant plastic types found in both the seawater and the sediments. Most microplastics were found in the top 6 cm of the sediment core, where their concentration decreased rapidly with increases in depth. Assuming an average sediment deposition rate of 3 mm per year in the Gulf of Mexico, the result implies that most microplastics have been deposited in the last two decades. The results highlight the need for wider studies using sediment cores to determine the historic deposition of microplastics in global ocean floors.

3.1. Introduction

Oceans make up more than 70% of Earth's surface, and play many critical roles across all global ecosystems including replenishing oxygen, mitigating carbon dioxide, regulating climate, and providing food and energy (Lovelock & Kump, 1994; Moran, n.d.; Riser & Johnson, 2008; Shen et al., 2020). The ocean floor alone has extensive properties that are pivotal to the global economy (Thrush & Dayton, 2002) and regulating climate (Shen et al., 2020; Visbeck, 2018; Visbeck et al., 2014). However, the composition of ocean floors is changing as a result of the plastic deposition (Cressey, 2016; Haward, 2018; Jambeck et al., 2015; MacLeod et al., 2021). Accumulation of microplastics on ocean floor could adversely affect the health of phytoplankton (Hope et al., 2020), benthic fish (Collard et al., 2019; Pannetier et al., 2020), corals (Hall et al., 2015; Reichert et al., 2019), and alter natural gas exchange from ocean floor (Gregory, 2009; Shen et al., 2020). Yet, limited studies have examined the historical accumulation of microplastics on the ocean floor, and no such study examined the deposition pattern in the Gulf of Mexico within the Region of Influx (ROI) of the Mississippi River.

Many studies have measured microplastic concentrations on the ocean floor by collecting sediments from the surface of the ocean bed (Cunningham et al., 2020; Loughlin et al., 2021; Martin et al., 2017b), but none of them measured these concentrations as a function of depth, particularly in the Gulf of Mexico within the ROI of the Mississippi River. This ROI into the Gulf of Mexico drains the fourth largest watershed in the world, including the surface waters and pollutants from about 40% of the continental United States (Milliman & Meade, 1983; Rabalais et al., 2002). The watershed receives runoff from many industrial sites (Bitter & Lackner, 2020; Deng et al., 2020; Wang et al., 2020) and vast agricultural lands in the USA (Braun et al., 2021; Corradini

et al., 2019) that could release microplastics into surface waters. Application of biosolids and plastic mulch on agricultural lands releases trillions of microplastics into surface water per year (Koutnik et al., 2021a), most of which could end up in the ocean via river flow (Koutnik et al., 2021). Many studies have confirmed the presence of high microplastic concentrations in the Mississippi River (Osorio et al., 2021; Rusinque-Quintero et al., 2022). After being discharged into the Gulf of Mexico, a fraction of this microplastic load could be removed from the seawater column by settling due to their large size and aggregation with other sediments (Galloway et al., 2017; Kane et al., 2020; Kooi et al., 2017; Pohl et al., 2020). Thus, the deposition of microplastics over several decades since the large-scale production of plastics from 1950s (Geyer et al., 2017) could change the composition of sediment deposits on the ocean floor in the Gulf of Mexico. As plastic's half-lives on average exceed 100 years (Chamas et al., 2020b), most plastics deposited historically could still be present in the sediment layers. Thus, it is important to estimate historic deposition of microplastics on the ocean floor.

Only one study has measured microplastic accumulation on the ocean floor in the Gulf of Mexico by collecting sediments from the ocean floor surface (Shruti et al. 2021). However, these results do not represent past deposits dating back to several decades. The average sedimentation depth in the Gulf of Mexico is ~3 mm per year (Rodriguez et al., 2020). Thus, the analysis of microplastic concentrations in different subsurface layers up to 21 cm could provide insight into the historic microplastic contamination levels in the last 70 years. A few studies have collected sediment cores up to a depth of 10 cm from different marine environment such as the North Atlantic Ocean (Courtene-Jones et al., 2020), mangrove tree forests in Vietnam (Viet Dung et al., 2021) and estuaries (Willis et al., 2017) in Australia. These studies provide insights on the short-term historic build-up of microplastics in those regions. A similar study on the ocean floor in the

Gulf of Mexico could record the extent of microplastic contamination on the ocean floor contributed by the largest river in the USA.

This study aims to determine the concentration of microplastics on the ocean floor of the Gulf of Mexico by collecting sediment cores and comparing the abundance of microplastics deposited in the sediment to that present in the seawater. To achieve the objectives, seawater and sediment cores up to a depth of 21 cm were collected from the Gulf of Mexico, and microplastic concentrations, abundance, and size distributions were analyzed in the water column and sediment cores at various depths corresponding to different sedimentary layers formed in the previous decades. Reconstructing the depth profile of microplastic concentration, assuming a constant sediment deposition rate of 3 mm per year determined in a previous study (Rodriguez et al., 2020), I provide insights into when microplastic contamination may have accelerated in recent history.

3.2. Method

3.2.1. Location for sampling

Seawater and a sediment core was collected from the northern Gulf of Mexico, within the region of freshwater influence of the Mississippi river (Simpson et al., 1993; J. H. Simpson, 1997). The river and its tributaries discharge water containing microplastics at a concentration ranging between 12–381 particles L^{-1} into the Gulf of Mexico, at an average rate of 16,792 m^3 s^{-1} from a nearly three million square km basin spanning over 32 states in the US (Shruti et al., 2021). Water samples and sediment cores were collected from a location (28° 46.693’ N; 90° 41.270’ W) approximately 32 km from the coastline in the northern Gulf of Mexico on 03/04/2021 during a research cruise onboard of R/V Pelican (**Figure 3-1**).

Figure 3-1: Sampling location of ocean water and a sediment core from the northern Gulf of Mexico, within the region of freshwater influence (ROFI) of the Mississippi river.

3.2.2. Seawater collection

Seawater samples were collected from the surface mixing zone from depths between 1.6 - 6.5 m using a rosette sampler. For each sample, 500 mL of seawater was pumped and filtered separately through clean pre-combusted (500 °C) 0.7-µm glass microfiber filters to capture debris containing microplastics. At least 25 samples were taken to get the average concentration of and type of microplastics found in present seawater so that the abundance in sediment cores can be compared. The seawater filter papers were folded, wrapped with aluminum foil, and stored in a freezer (-20°C) onboard the ship.

3.2.3. Sediment core collection from the ocean floor

The sediment core was collected using a benthos gravity corer containing a plastic liner (3 m length, 7 cm diameter) where the water depth was at least 18 m. The corer was released to fall freely from the ship's winch, and gravity led the heavy weighted pointed end into the sediment. For better results and to maintain the integrity of the core sample, a one-way check valve was installed at the top end of the corer, allowing the water to flow through the attached liner during its descent while blocking water during the ascent. Once the corer was retrieved, the liner containing the sediment core was removed from the corer. Using a metallic core cutter, the required length was cut out from the top part, capped on both ends, sealed with duct tape, and stored in a freezer (-20°C) onboard immediately after collection. The core was transported to the laboratory in an airtight cooler containing ice and stored in a freezer in the laboratory (-80°C) before subsequent analysis.

3.2.4. Extraction of microplastics from sediment samples

The frozen core containing sediment was split at every 2 cm increments using a metallic band saw machine. Immediately after splitting each section, the dirt at the top and bottom surfaces of the frozen subsections was removed using a clean brush with natural hair. Once the sediment was slightly thawed down, the samples were pushed out from each 2 cm increments using a clean metallic spatula into separate clean aluminum cups and covered using clean aluminum foil. To avoid any cross-contamination, saw, brush, and spatula were cleaned repeatedly during the sampling process. The outer rim of the core touching the plastic core was trimmed off and discarded. The subsamples from each depth were packed separately in aluminum foil and stored for analysis for the following cross-sectional layers of the core in centimeters 0-3, 3-5, 5-7, 7-9, 9-11, 11-13, 13-15, 15-17, 17-19.

Frozen sediments from each 2-cm cross-section were broken into smaller pieces using a hammer and metal spatula and separated into aluminum foil packets for microplastic identification following methods described in a previous study (Koutnik et al., 2022). To avoid contamination from the plastic core casing and cutting of the core, sediments from the edges and areas near the border were discarded. The spatula and hammer were thoroughly cleaned using deionized (DI) water and wiped between sampling to prevent cross-contamination. Briefly, 1 g of broken frozen sediment was soaked in DI water to make a homogeneous slurry and mixed with 40 mL of 1.6 g mL^{-1} potassium iodide solution for density separation. The suspension was centrifuged at 5000 rpm for 30 min to settle heavier soil particles and isolate lighter (density < 1.6 g cm^{-3}) particles including microplastics from the supernatant. The floating particles in the supernatant were vacuum filtered onto a 24 mm glass fiber filter paper with a 1.2 μm pore size. The filter was placed inside a glass Pyrex petri dish, covered with a glass cover, and left to dry.

3.2.5. Quantification of microplastics

The plastic particles among the dried particles on filters were identified and quantified using two methods: a rapid smartphone method using Nile Red for quick assessment of microplastic contamination (Leonard et al., 2022) and a traditional Fourier-transform infrared spectroscopy (FTIR) microscope for the confirmation of microplastic concentrations and characteristics (Cowger et al., 2021). While the FTIR method has the advantage of identifying plastic types, it can only identify microplastics larger than 20-50 μm and is susceptible to misidentification if the plastic surface is highly weathered. In contrast, the Nile Red method can identify microplastics as small as the resolution of the microscope (~2 μm), but the method cannot distinguish plastic particles from other organic matter, if any in the samples, that could also bind Nile Red (Maes et al., 2017a). To minimize false positives in the samples, samples were digested

using Fenton's Reagent system- hydrogen peroxide and iron hydroxide (Tagg et al., 2016) before quantifying them with the Nile Red method.

For quantification using Nile Red, digested filters in a glass petri dish were dyed with 0.17 mL of 0.5 μg mL^{-1} Nile Red in chloroform solution, air-dried with a glass cover for 24 h in the fume hood, transferred onto glass slides, and covered with a glass coverslip to eliminate dust deposition. The dyed particles were quantified from the images taken by a smartphone fitted with an external camera setup in a 3D-printed platform as described elsewhere (Leonard et al., 2022). The method can resolve particles as small as 4.5 μm despite a large field of view of 490 mm^2.

A subset of the samples was further analyzed using FTIR (Thermo Scientific Nicolet iN10) as described elsewhere (Cowger et al., 2021). The dry particles were scraped from the filters onto a gold-coated slide, and their FTIR spectra were recorded to confirm microplastic concentrations, size distributions, and abundance by polymer types. The FTIR microscope was used in the reflectance mode using the particle analysis wizard included in the OMNIC PICTA software to identify microplastics larger than 20 μm based on image analysis of particles spread on a 1 cm^2 area of the slide. When comparing sample spectra to spectra databases, 60% match criteria were used as a threshold to identify the particle. The following libraries were used for matching: FTIR libraries used to match spectra were: Polymer Laminate Films, Cross Sections Wizard, Hummel Polymer Sample, Polystyrene Quality Control, NIR Polystyrene QC, Synthetic Fibers by Microscope, HR Hummel Polymer and Additives, HR Aldrich Hydrocarbons, HR Nicolet Sampler Library, HR Polymer Additives and Plasticizers, HR Spectra Polymers and Plasticizers by ATR, HR Spectra Polymers and Plasticizers.

3.2.6. Quality Control

During the isolation of microplastics or handling of samples, the use of plastic materials or the possibility of cross-contamination was minimized. During lab work, clothing made from natural materials was worn to prevent cross-contamination of the samples. Pre-washed glass and aluminum containers were used for sampling, storage, and processing. All glassware and containers were rinsed with DI water three times to wash off any deposited particles. Samples were covered by glass covers or aluminum foil during the drying and storage period. The smartphone quantification methodology has an average recovery rate of 93.7% ± 13.7%, a human processing variation of 6.8% of the mean, and a sample processing variation of 9.1%, as estimated and reported in previous publications (Koutnik et al., 2021a). Therefore, the total maximum error for each of the microplastic measurements was estimated to be 22.2%- accounting for all experimental and human processing steps. All concentrations were visualized using boxplots generated in R, and statistical significance was assessed with t-tests and a 95% confidence level.

3.3. Results and Discussion

3.3.1. Microplastic concentration in seawater samples

Microplastic concentrations in water samples varied from 40-392 pieces L^{-1}, with an average of 160 pieces L^{-1} (**Figure 3-2**) using smartphone analysis. In contrast, the average concentration of previously reported studies was 3.8 pieces L^{-1} (Di Mauro et al., 2017; Rios-Mendoza et al., 2021). Results reveal that microplastic concentrations in seawater from the mid-shelf of the northern Gulf of Mexico in the region of the freshwater influx of the Mississippi River could be at least 3 orders of magnitude higher than concentrations previously reported in other studies from various sites in the Gulf of Mexico (**Figure 3-2**). Water samples were taken from the region of freshwater influx from the Mississippi River, where higher concentrations at river

mouths are expected (Osorio et al., 2021; Rusinque-Quintero et al., 2022). The concentrations of microplastics in the Mississippi River were found to be 12-381 particles L^{-1} using FTIR method with size cut-off >25 µm (Scircle et al., 2020). The concentration is expected to be diluted after mixing with ocean water. However, the concentration found in this study was still in a similar range to previously found in the Mississippi River with a large size cut-off. The results collectively imply that a decrease in cut-off size could increase the microplastics detected or reported in previous studies.

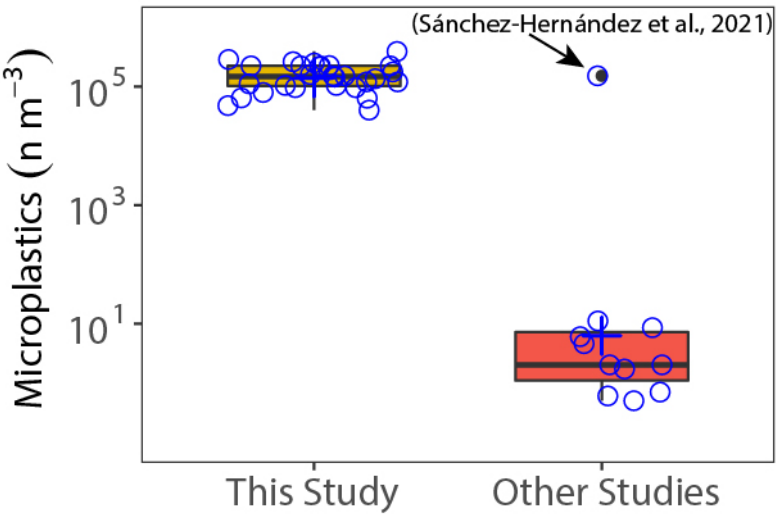


Figure 3-2: Microplastic concentrations (pieces, n, per m^3) in ocean water samples in this study in comparison to other studies in the Gulf of Mexico (Di Mauro et al., 2017; Sánchez-Hernández et al., 2021; Scircle et al., 2020).

To confirm the role of size cut-off, I measured the concentration of microplastics using two different methods with different cut-off sizes. Measuring microplastic concentrations using FTIR with the size cut-off at 20-50 µm resulted in a decrease in the concentration by one order magnitude, down to around 2-20 pieces L^{-1} (**Figure 3-3**). This confirmed the hypothesis that the size cut-off for microplastic quantification could disproportionally affect the microplastic quantification in seawater. The size cutoff in my smartphone/Nile red methodology is ~5 µm

whereas in other studies the cutoff size was orders of magnitude higher: 50 µm (Di Mauro et al., 2017) and 1.1 mm (Rios-Mendoza et al., 2021). My hypothesis is supported by another study (Sánchez-Hernández et al., 2021) where the size cutoff was 1.2 µm and the concentrations were in a similar range to that of this study. Thus, microplastic concentration in seawater could be 1000 times more than that previously reported, as global assessments typically have a size cutoff above 50 µm. It is important to note that other methodological differences, such as sampling volume, could also influence microplastic concentrations. Thus, future studies should measure microplastic concentration at smaller size ranges using a standard sampling volume.

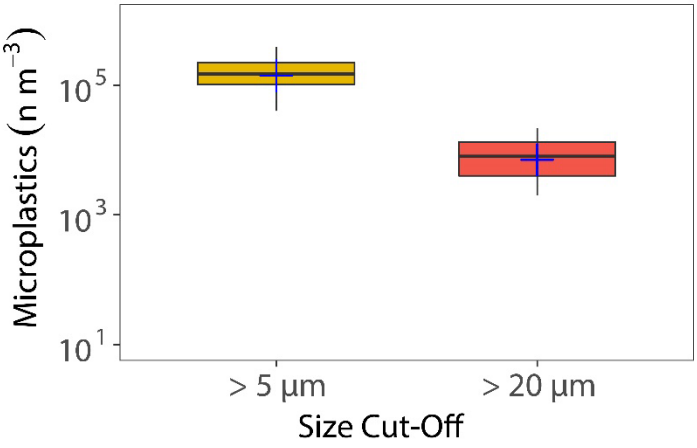


Figure 3-3: Microplastic concentrations (pieces, n, per m3) in ocean water samples in this study using the smartphone technique with a size cut-off of 5 µm compared to FTIR technology with a size cut-off of 20 µm.

3.3.2. Microplastic concentration in sediment cores

Microplastic concentrations in the ocean floor decreased rapidly with an increase in depth (**Figure 3-4**). Concentrations were the highest in the top 6 cm of the ocean sediment core samples with a mean concentration of 82.2 pieces g^{-1}, which is at least 5 times higher than the concentration measured in deeper layers. The concentration decreased to 11 pieces g^{-1} in the 10-15 cm layer, and nearly zero by 17-19 cm depth. A rapid decrease in microplastic concentration with increases in depth is also observed in other studies on the ocean floor (Loughlin et al., 2021; Martin et al.,

2017b; Viet Dung et al., 2021). To the best of my knowledge, this is the first study in the region to analyze the depth profile of microplastic concentrations. The concentration is at least three order of magnitude above the only other study in the Gulf of Mexico that reported microplastic concentrations (0.095 pieces g^{-1}) on the surface of the ocean floor near the mouth of the Grijalva-Usumacinta River (Osten et al., 2023). However, the microplastic loading could vary from that of the Mississippi River. The concentrations reported in this study are similar to concentrations (0.333-32 pieces g^{-1}) in sediment samples collected from other oceans (Courtene-Jones et al., 2020; Cunningham et al., 2020; Hammadi et al., 2022; R. Li et al., 2019; Loughlin et al., 2021; Martin et al., 2017b; McEachern et al., 2019; Qi et al., 2022; Su et al., 2020; X. Sun et al., 2021; Tang et al., 2018).

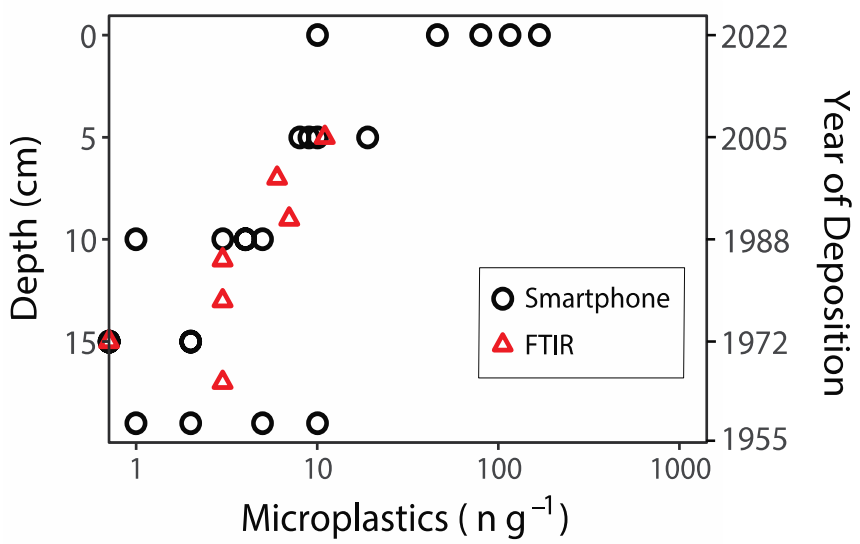


Figure 3-4: Microplastic concentrations (pieces g^{-1}) at different depths on the ocean floor. The layer in the core corresponds to sediment deposited in the past with an estimated sediment deposition rate of 3mm $year^{-1}$ (Rodriguez et al., 2020) in the Gulf of Mexico. Circles denote the concentration measured using Nile Red and a smartphone with a resolution of ~5 µm, whereas triangles represent the concentration reported using the FTIR with a resolution > 20 µm.

Microplastic concentrations in sediment cores vary widely, and this variation could be attributed to the size of microplastics, particularly the cut-off size for the method used (Baruah et

al., 2022; Lao & Wong, 2023; Nel et al., 2021). Most previous studies on microplastics in marine sediments rely on FTIR, which cannot detect microplastics smaller than 20 μm. Thus, actual concentrations of microplastics could be much higher. Microplastics smaller than 10 μm are typically associated with impaired feeding behavior, photosynthesis rates, reproductive success, and growth of phytoplankton, barnacle larvae, seagrass species, coral, and marine organisms at low trophic levels (Huang et al., 2023; Larue et al., 2021; Reichert et al., 2019; Lin, 2016). To examine if the size cut-off has any influence on the concentration of microplastics, I compared microplastic concentrations measured by a portable camera fitted on a smartphone (size cut-off 5 um) and by an FTIR microscope (size cut-off 20 um). Microplastic concentrations estimated using FTIR on a subset of sediment samples displayed similar concentrations to that measured by the Nile Red method, despite a difference in the size cutoff of both methods (**Figure 3-4**). The similar concentration of microplastics in sediments by both methods may indicate that most particles found in sediments were larger than 20 μm. Size distribution data reported later (**Figure 3-6B**) confirmed this assertion.

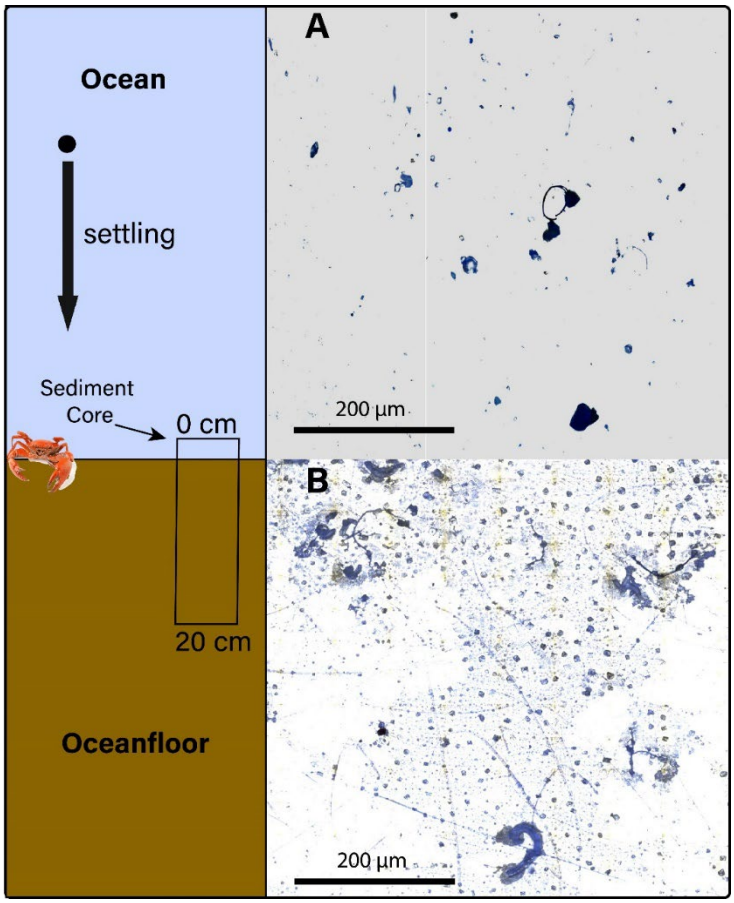


Figure 3-5: Images of particles found in ocean water (A) and ocean floor sediment cores (B).

3.3.3. Relative abundance of microplastics in seawater and sediment samples

A comparison of microplastic polymer types found in seawater and ocean floors **(Figure 3-5**) reveals that rayon and polyethylene were the most abundant microplastics in both seawater and marine sediments in the Gulf of Mexico (**Figure 3-6A**). Other plastic polymers, such as polyethylene terephthalate (PET) or polyvinyl chloride (PVC) were surprisingly not detected in ocean core sediment samples or seawater despite their extensive use in single-use plastic products. Plastic types were more diverse in sediment samples than in water samples. For example, polypropylene (PP) and polystyrene (PS) were present in ocean core sediments but not seawater samples. These plastic types are relatively light (910 kg m^{-3} and 1000 kg m^{-3} respectively) compared to seawater (1420 kg m^{-3}). However, they may settle out of the water column and deposit

on the ocean floor due to the formation of biofilms and aggregation with other soil minerals (Shen et al., 2023) . Contrastingly, cellophane was not found in sediment samples, but was present in seawater. This is of note since cellophane (1420 kg m^{-3}) is denser than seawater (1025 kg m^{-3}), yet remains suspended in the water column perhaps since it rapidly degrades within weeks (Kaplan et al., 1994) and was uncoverable in sediment samples. However, the results may be influenced by the limitation of FTIR in detecting heavily weathered microplastics, especially in seawater conditions (Phan et al., 2022). In my study, I used a 60% threshold to match the polymer spectra, which ruled out many particles as non-plastic due to degradation effects and artifacts introduced from baseline subtraction **(Figure 3-7**). Thus, the microplastic concentrations and abundance reported in this study are conservative estimates.

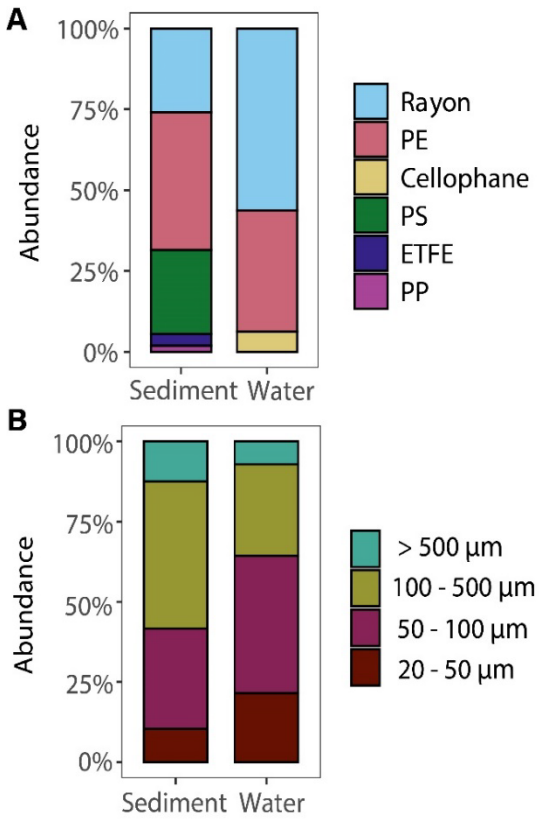


Figure 3-6: Comparison of trends in abundance for microplastics found in ocean water versus sediment based on (A) microplastic types and (B) size.

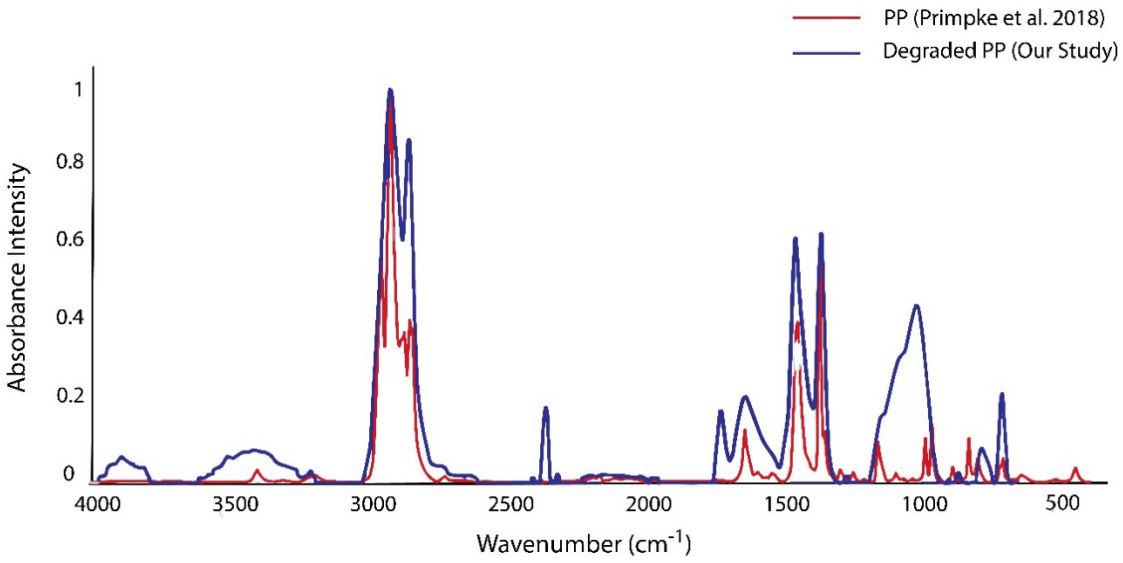


Figure 3-7: FTIR spectra highlighting regions that exhibit variability after weathering. The blue spectra represents samples taken from the Gulf of Mexico degraded after an unknown time in seawater and environmental conditions, compared to the red spectra representing pristine polypropylene (PP) spectra from established libraries (Primpke et al., 2018).

Characterization of microplastic size fractions revealed a higher prevalence of smaller microplastics in seawater than sediments, possibly due to size fractionation during settling. As settling favors large plastic particles, it can enrich larger plastic particles on the ocean floor. A higher abundance of larger microplastics within the 100-500 µm (29%) and 500 µm (43%) size fractions in ocean core compared to seawater confirmed this theory (**Figure 3-6B**). Furthermore, smaller microplastic fractions (50-100 µm and 20-50 µm) made up over 60% of seawater microplastics. The result confirms that settling could disproportionally enrich larger microplastics on the ocean floor, as predicted by Stokes' Law. Thus, size might be the main driver determining which microplastics end up on the ocean floor.

3.3.4. Implication on the historic microplastic deposition on the ocean floor in the Gulf of Mexico

To date, the historic deposition of microplastics on the ocean floor in the Gulf of Mexico has not been reported due to a lack of previous studies that reconstructed a depth profile from ocean floor sediment cores in the region. To roughly estimate the historic deposition of

microplastics based on their concentrations at different depths, I made two assumptions. First, I assumed a sedimentation rate of 3 mm $year^{-1}$ previously measured near the study site in the Gulf of Mexico (Rodriguez et al., 2020). Second, I assumed limited mixing of microplastics between layers in the deep ocean floor by near-bed thermohaline currents or bottom currents (Kane et al., 2020). Based on these two assumptions, 21 cm cores in this study could represent over 70 years of sediment deposits. By linking the sediment layer with the historic time in which the layer was formed, I observed that plastic deposition rapidly increased within the last two decades. Microplastic concentrations in sediment layers deeper than 15 cm—estimated to deposit prior to 1975—are almost negligible (not nonexistent) perhaps due to a lack of sufficient microplastics present in seawater for settling or because any deposited microplastics before that period were degraded beyond identification. Since plastic is highly durable with a typical lifetime of over 200 years (Chamas et al., 2020b), degradation could not explain negligible concentrations in sediments in deeper layers. Thus, the low microplastic concentration in deeper depth could reflect the low concentration of microplastics in the seawater at that time, indicating almost negligible deposition of microplastics around 60 years ago. High concentrations of microplastics within the top 6 cm layers imply that the ocean floor has been rapidly polluted with microplastics in the last two decades. This is proportional to plastic production rates which rose from 40 million metric tons of plastic in 1970 to 400 million metric tons by 2015, alongside population growth and anthropogenic activity (Geyer et al., 2017). Furthermore, this uptick in concentrations in sediment cores is mirrored by a parallel uptick in marine microplastic surface concentrations over the last 20 years (Eriksen et al., 2023)- lending further validity to our results.

It should be noted that the speculation on historic deposition rate has several shortcomings. This analysis of historic sediment layer formation is based on the critical assumption that the ocean

floor rises by 3 mm annually in the Gulf of Mexico, which may not be accurate for the precise location where the core was collected. The ocean floor could have risen much faster rate due to its proximity to the Mississippi River delta. Assuming a faster sedimentation rate, a rapid rise in concentration might have occurred in more recent history. Future studies should use the radio-isotope method to accurately estimate the age of the sedimentation layer (Long et al., 2022; Szmytkiewicz & Zalewska, 2014) to confirm the historical deposition of microplastics. It is also not clear if the current near the ocean floor could have affected the mixing of sediments between layers at the sites. Furthermore, microplastics exhibit both upward and downward migration in soil columns (Koutnik et al., 2022), which could affect their distribution. Nevertheless, historical microplastic deposition patterns in sediment cores on the ocean floor in the Gulf of Mexico could inform future studies to assess the current and historic contamination of the ocean floor sediments with microplastics.

3.3.5. Conclusion

I measured microplastic concentration in seawater in the Gulf of Mexico to be within the range of 40-392 pieces L^{-1}, and the sediment cores, particularly within the top 6 cm, to be 10-170 pieces g^{-1}. The observed concentration in seawater in this study was at least 3 orders of magnitude more than that reported in other studies from the same region. I attributed the discrepancy due to both influx from the Mississippi River and the high cut-off size for previous methodologies reporting microplastics. Thus, the extent of microplastic pollution and its negative effects on the marine ecosystem could be much higher than predicted based on previous studies that used a larger size cut-off. Measuring the depth distribution of microplastics in sediment cores from the ocean floor, I showed that most microplastics in sediment cores were concentrated in the top 6 cm. The concentration decreased rapidly with depth and became negligible below 15-21 cm of the ocean

floor. Thus, the contamination level can be assessed by collecting cores up to 20 cm depth. If the ocean floor rises at 3 mm yr^{-1} at the site in the Gulf of Mexico, the depth distribution data implies that microplastic pollution has accelerated in the last 20 years. As the extent of pollution is expected to rise, there is a greater need to enforce mitigation measures to limit the utilization of single-use plastic products and the removal of microplastics from surface waters in the terrestrial environment before they reach our oceans.

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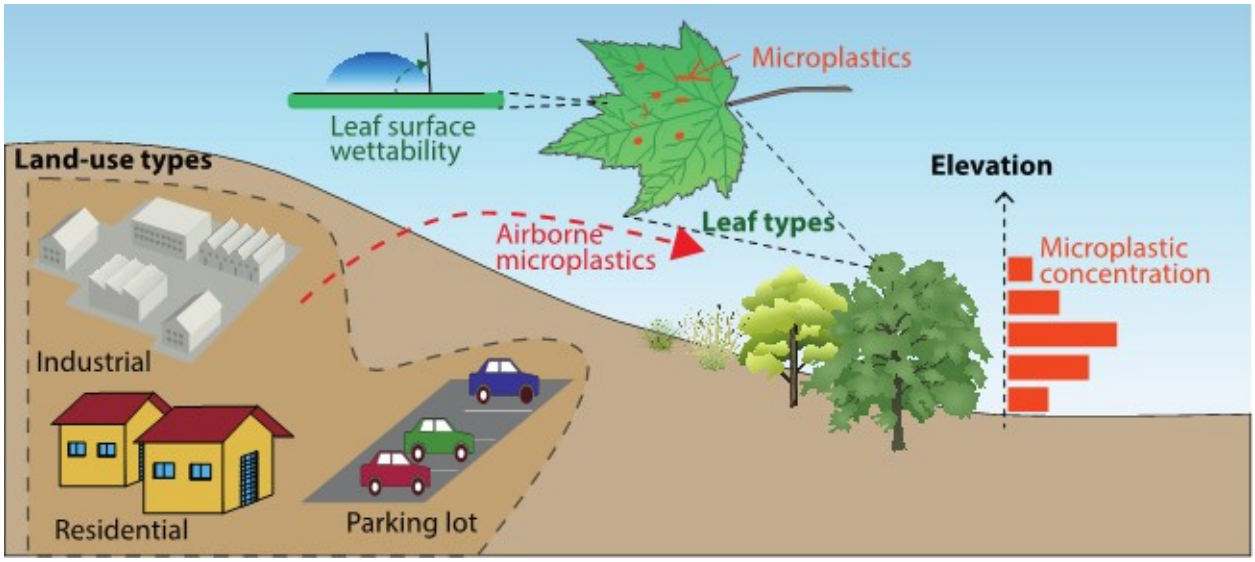
4. CHAPTER 4 – CHALLENGES OF USING LEAVES AS A BIOMONITORING SYSTEM TO ASSESS AIRBORNE MICROPLASTIC DEPOSITION ON URBAN TREE CANOPIES



Leonard, J., Borthakur, A., Koutnik, V.S., Brar, J., Glasman, J., Cowger, W., Dittrich, T.M., Mohanty, S.K. (2023) Challenges of using leaves as a biomonitoring system to assess airborne microplastic pollution in urban areas. Atmospheric Pollution Research. 101651. https://doi.org/10.1016/j.apr.2023.101651.

Abstract

Monitoring microplastic concentrations in the atmosphere is critical for assessing their inhalation risk in urban areas and other ecological impacts on natural environments. To assess these risks, leaves in tree canopies could be used as a biomonitoring system, but this requires knowledge of the potential variability of microplastic concentrations based on leaf properties and position in the tree. This study aims to quantify this potential variability by analyzing the concentration of airborne microplastics on leaves in Los Angeles as a function of leaf height, leaf surface hydrophobicity, and land use. Microplastic concentrations on leaves varied between 0.14 - 25 particles (n) cm^{-2}, but the concentration peaked within 0.6 to 1.2 meters height above ground, indicating the position of the leaf above the ground could matter. Microplastic concentration varied significantly between leaf types indicating leaf surface properties could influence the retention of microplastics. Contact angle measurements revealed that the hydrophilicity of leaves had a weak correlation with microplastic concentrations on leaves, indicating other factors such as surface roughness or leaf features could be important for microplastic retention. Land-use type did not affect microplastic concentration. The results confirmed a high uncertainty in predicting microplastic concentration on leaves in the urban canopy, which limits their ability to be used as biomonitoring systems for microplastic pollution in urban areas.

4.1. Introduction

Atmospheric transport has been identified as one of the major conveyors of microplastics in urban areas (Koutnik et al., 2022b; Sridharan et al., 2021; Zhu et al., 2021), where more than 56% of the world population currently live (Zhu et al., 2021). Thus, the majority of the population is exposed to these airborne microplastics, which may have human health implications at very high concentrations (Prata, 2018b). These airborne microplastics have been found unexpectedly in lower regions of the lungs (Jenner et al., 2022). The risk of microplastic inhalation could be exacerbated by their ability to carry toxic pollutants from land surfaces (Borthakur et al., 2021b). Therefore, it is critical to develop rapid monitoring systems to assess the atmospheric deposition of microplastics in urban areas.

Microplastic concentrations in urban areas were recently monitored by using passive samplers (Cai et al., 2017; Dris et al., 2016; Zhou et al., 2017). However, passive samplers are relatively cumbersome to deploy in a large area, and often require specific expertise to effectively install and execute sampling. In contrast, leaves in the urban canopy are readily accessible and could potentially be used as a biomonitoring system as they accumulate microplastics continuously throughout the year via dry deposition. Leaves in urban canopies naturally intercept wind flows and trap particulate pollutants on their surfaces, thereby improving the air quality in cities (McPherson et al., 2011; Rai, 2016). Therefore, they have been used as passive samplers to measure the atmospheric deposition of a wide range of pollutants including ozone (de Souza et al., 2022), particulate matter (Beckett et al., 2000; Hansard et al., 2012), persistent free radicals (Leonard et al., 2016; Oyana et al., 2017), and polyaromatic hydrocarbons (Wannaz et al., 2013). For leaves to be used as monitoring systems for airborne microplastics, it is critical to determine how microplastic concentrations may vary on the leaves based on leaf properties and leaf position

so that a monitoring protocol could be developed to standardize the sampling method and reduce this variability.

To date, only a total of 4 studies have so far quantified microplastic concentration on leaves (Koutnik et al., 2022a ; Li et al., 2022; Liu et al., 2020; Liu et al., 2022), but they rarely account for the causes of concentration variability. Based on these studies, microplastic concentrations on leaves can vary by two orders of magnitude, from 0.1 to 50 pieces (n) per cm^2, even at the same spot. This variability poses a challenge in using the data to compare microplastic exposure risks between sampling locations. Based on the variability of particulate matter (PM 2.5) on leaves (Sæbø et al., 2012; Zheng & Li, 2019), the cause of microplastic variability could be attributed to several factors: leaf height, leaf surface properties, and land-use type. Leaf height relative to the ground surface is critical in intercepting airborne particulates (Barwise & Kumar, 2020). The concentration of airborne particulate is also sensitive to sampling height due to the dilution and gravitation settling of particulates (Montoya & Hildemann, 2005; Rantio-Lehtimäki et al., 1991). A recent study demonstrated that microplastic concentrations on leaves in urban green infrastructures increased initially up to a critical height, and the concentration on leaves above that height decreased (Koutnik et al., 2022a). However, it is not clear if sampling leaves from the same height would reduce the variability. The effect of land use on the deposition of microplastics on nearby leaves is also unclear. Land use could affect plastic emissions (Barrows et al., 2018; Tanentzap et al., 2021; Y. Xu et al., 2020), since the proximity of the plant to emission sources could affect microplastic concentration on leaves (Liu et al., 2022). However, microplastics can also move away from the source, thereby diluting the effect of land use. Lastly, plant types could influence the concentration of particulate matter (Cai et al., 2017; Leonard et al., 2016; Sæbø et al., 2012). Leaf traits such as texture or surface roughness, hydrophobicity, and the presence of

waxy coating, could determine how much particulate matter will adhere to leaf surfaces after being intercepted from wind streams (Corada et al., 2021). The development of water films, which are sensitive to leaf hydrophobicity, on leaf surfaces may increase retention of particulate matters temporarily due to cohesive forces (Seville et al., 2000). However, no study to date has examined the effect of leaf surface hydrophilicity on microplastic concentration.

This study aims to quantify the variability of microplastic concentrations contributed by land-use type, leaf height, and leaf surface hydrophilicity. I hypothesized that leaf height and surface hydrophilicity would affect microplastic concentration more than land use due to limited constraints in the geographical boundary for atmospheric transport. To test this hypothesis, leaves were collected from five plants at different heights from the ground surface from locations with 3 different types of land use in Los Angeles, USA. The results could help improve the understanding of the sources of variability in microplastic concentrations on leaves and inform the development of standard sampling best practices to use leaves as a biomonitoring system.

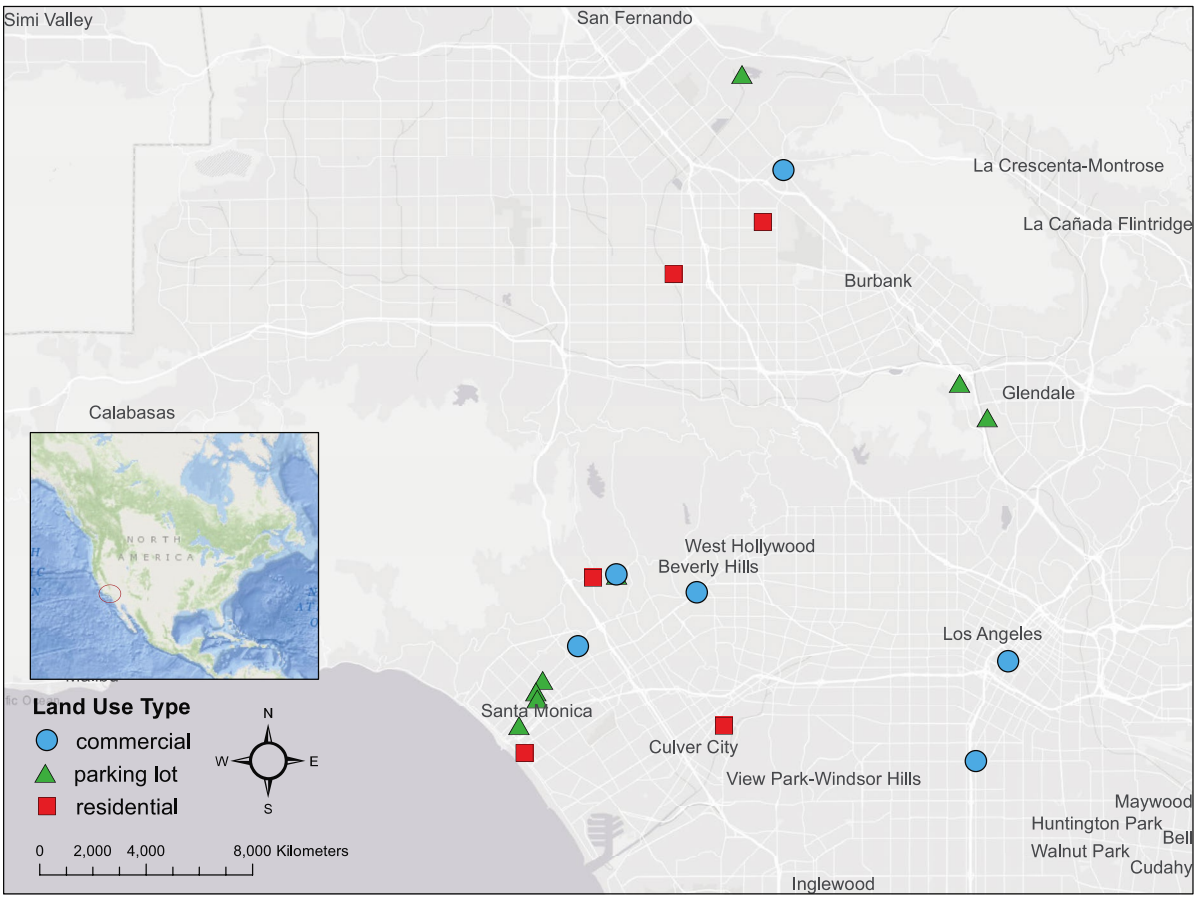


Figure 4-1: Sampling locations in Los Angeles, colored by the type of land use: commercial, parking lot, and residential.

4.2. Materials and Methods

4.2.1. Sample collection and processing

Leaves from the urban canopy were collected from 19 locations in Los Angeles, USA (**Figure 4-1**) on February 12, 2022, on a dry and sunlit day. The locations were categorized by three types of land use: residential, commercial, and parking lot. The sampling area last received ~ 2 mm of rainfall on January 17, 2022. Thus, the concentration on leaves would represent the accumulation of airborne microplastics deposited in 25 days. Healthy green leaves were collected at varying heights or at positions at varying elevations from the ground: < 0.6 m, 0.6 – 1.2 m, and > 1.2 m. Intact leaves were carefully collected from five plants that are frequently found in Los

Angeles: *Acer saccharum*, *Rhus ovata*, *Buxus sempervirens*, *Leymus condensatus*, and *Chamaerops humilis (***Figure 4-2***)*.

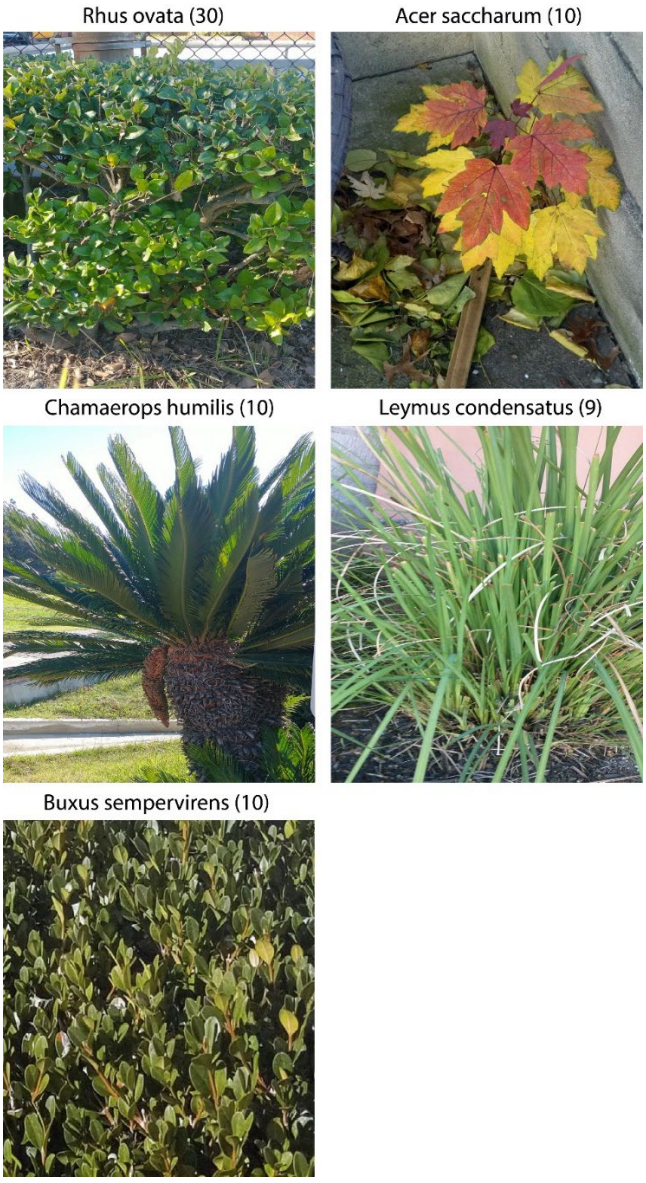


Figure 4-2: Figure depicts the species name (number of samples of that species) alongside a representative image of an urban canopy of that species where samples were taken.

Leaves were individually wrapped in aluminum foil and labeled by sampling location, height, and plant type. A total of 69 leaf samples were collected and analyzed for microplastic concentrations as a function of land use, height, and leaf type (**Table 4-1**). The complete details of

the locations and characteristics of samples are provided in **Table 4-2**. In the lab, leaves were cut into rectangular pieces with metal scissors so that their surface area could be calculated. Then, the pieces were wrapped in aluminum foil and stored. All samples were analyzed within 5 days of sample collection.

Some of the microplastics deposited on soil near the tree could be resuspended by wind and deposited on the leaf. To compare the relative abundance of microplastics on the leaf surface and in the soil near the tree, I collected soil from the top 0-2 cm surface of the ground near to the base of the tree from where leaves were selected. Samples of around 2-4 grams of surface soil were extracted using a pre-washed metal spoon into labeled aluminum packets. The spoon was thoroughly cleaned between sampling to prevent cross-contamination.

Table 4-1: Number of leaves (in parenthesis) collected per each criterium: land-use type, height, and leaf types.

Land use	***Height***	***Leaf type***
Commercial (37)	< 0.6 meter (19)	*Acer saccharum* (10)
Parking Lot (16)	0.6 – 1.2 meters (30)	*Rhus ovata* (30)
Residential (16)	> 1.2 meters (20)	*Buxus sempervirens* (10)
		Leymus condensatus (9)
		Chamaerops humilis (10)

Table 4-2: Table records the inventory for each qualitative and quantitative characteristic per each leaf sample: microplastic count, leaf height, land-use type, leaf species, leaf surface area, microplastic abundance, GPS location.

SAMPLE	COUNT (N)	LEAF HEIGHT (METERS)	LAND-USE TYPE	SPECIES	SURFACE AREA (CM^2)	MICROPLASTIC ABUNDANCE (N CM^{-2})	GPS LOCATION
10A'	26	2.3622	parking lot	*Buxus sempervirens*	6	2.166667	(34.26691118, -118.3865457)
10G'	58	0.4318	parking lot	*Leymus condensatus*	16.8	1.72619	(34.26691118, -118.3865457)
10O'	20	2.0066	parking lot	*Acer saccharum*	7.79	1.283697	(34.26691118, -118.3865457)
11C'	13	0.3302	parking lot	*Rhus ovata*	5.4	1.203704	(34. 075141, -118.444578)
11G'	102	0.635	parking lot	*Leymus condensatus*	19.2	2.65625	(34. 075141, -118.444578)
11O'	45	2.1082	parking lot	*Acer saccharum*	25	0.9	(34. 075141, -118.444578)
12A'	40	0.635	residential	*Buxus sempervirens*	9.24	2.164502	(34.074186, -118.45537)
12C'	57	1.3716	residential	*Rhus ovata*	9.62	2.962578	(34.074186, -118.45537)
12O'	47	2.2606	residential	*Acer saccharum*	16.74	1.403823	(34.074186, -118.45537)
13C'	38	1.6256	commercial	*Rhus ovata*	8.37	2.270012	(34.068534, -118.407462)
13P'	34	2.2606	commercial	*Chamaerops humilis*	19.6	0.867347	(34.068534, -118.407462)
14C'	153	1.4732	commercial	*Rhus ovata*	12.4	6.169355	(34.047802, -118.462296)
14O'	152	2.3114	commercial	*Acer saccharum*	11.22	6.773619	(34.047802, -118.462296)
15O'	9	0.4826	parking lot	*Acer saccharum*	30.82	0.146009	(34.034861, -118.478842)
16C'	227	1.0922	parking lot	*Rhus ovata*	7.2	15.76389	(34.030464, -118.481945)
17P'	38	0.5842	parking lot	*Chamaerops humilis*	4	4.75	(34.027714, -118.481319)
18C'	241	0.7366	parking lot	*Rhus ovata*	7	17.21429	(34.017517, -118.48982)
18P'	48	0.3048	parking lot	*Chamaerops humilis*	3.68	6.521739	(34.017517, -118.48982)
1C	221	1.62814	residential	*Rhus ovata*	23.46	4.710145	(34.00705466, -118.487221)
1G	151	0.635	residential	*Leymus condensatus*	4.6	16.41304	(34.00705466, -118.487221)
1P	166	1.1049	residential	*Chamaerops humilis*	13.32	6.231231	(34.00705466, -118.487221)
2C	93	1.8288	residential	*Rhus ovata*	16.77	2.772809	(34.01744577, -118.3948878)
2G	67	0.5588	residential	*Leymus condensatus*	6.66	5.03003	(34.01744577, -118.3948878)
2P	317	0.9144	residential	*Chamaerops humilis*	1.5	105.6667	(34.01744577, -118.3948878)
3A	105	0.9652	commercial	*Buxus sempervirens*	6.82	7.697947	(34.00368118, -118.2781737)
3G	163	0.4064	commercial	*Leymus condensatus*	7.68	10.61198	(34.00368118, -118.2781737)

3O	90	2.1844	commercial	*Acer saccharum*	15.36	2.929688	(34.00368118, -118.2781737)
3P	326	0.5588	commercial	*Chamaerops humilis*	1.6	101.875	(34.00368118, -118.2781737)
4A	192	1.2192	commercial	*Buxus sempervirens*	8.05	11.92547	(34.04212555, -118.2632089)
4C	210	0.4318	commercial	*Rhus ovata*	8.1	12.96296	(34.04212555, -118.2632089)
4G	392	0.889	commercial	*Leymus condensatus*	7.84	25	(34.04212555, -118.2632089)
5A	97	1.3462	parking lot	*Buxus sempervirens*	14.85	3.265993	(34.1354333, -118.273087)
5G	291	0.8382	parking lot	*Leymus condensatus*	7.48	19.45187	(34.1354333, -118.273087)
5P	92	0.6858	parking lot	*Chamaerops humilis*	1.76	26.13636	(34.1354333, -118.273087)
6A'	65	2.1082	parking lot	*Buxus sempervirens*	10.08	3.224206	(34.14859972, -118.2859005)
6O'	84	2.2606	parking lot	*Acer saccharum*	12.4	3.387097	(34.14859972, -118.2859005)
7A	106	1.1938	residential	*Buxus sempervirens*	7.8	6.794872	(34.190602, -118.4182163)
7G'	53	0.508	residential	*Leymus condensatus*	4.41	6.00907	(34.190602, -118.4182163)
7O	114	1.3208	residential	*Acer saccharum*	9	6.333333	(34.190602, -118.4182163)
7P'	169	0.2794	residential	*Chamaerops humilis*	7.14	11.83473	(34.190602, -118.4182163)
8A'	6	1.7018	residential	*Buxus sempervirens*	5.76	0.520833	(34.210473, -118.376875)
8O'	23	2.3622	residential	*Acer saccharum*	9.66	1.190476	(34.210473, -118.376875)
8P'	39	0.381	residential	*Chamaerops humilis*	1.84	10.59783	(34.210473, -118.376875)
9A'	36	1.651	commercial	*Buxus sempervirens*	5.4	3.333333	(34.2301655, -118.3671678)
9C'	22	1.1938	commercial	*Rhus ovata*	5.44	2.022059	(34.2301655, -118.3671678)
9G'	53	0.5334	commercial	*Leymus condensatus*	10.2	2.598039	(34.2301655, -118.3671678)
9O'	14	1.6002	commercial	*Acer saccharum*	21.15	0.330969	(34.2301655, -118.3671678)
9P'	53	0.7874	commercial	*Chamaerops humilis*	3.7	7.162162	(34.2301655, -118.3671678)
L21'	44	0.6096	commercial	*Buxus sempervirens*	6.66	3.303303	(34.075429, -118.444563)
L22'	55	0.6096	commercial	*Buxus sempervirens*	8.2	3.353659	(34.075429, -118.444563)
L22_1'	61	0.6096	commercial	*Buxus sempervirens*	8.8	3.465909	(34.075429, -118.444563)
L31'	62	0.9144	commercial	*Buxus sempervirens*	8.16	3.79902	(34.075429, -118.444563)
L32'	69	0.9144	commercial	*Buxus sempervirens*	9.6	3.59375	(34.075429, -118.444563)
L33'	50	0.9144	commercial	*Buxus sempervirens*	10.6	2.358491	(34.075429, -118.444563)
L41'	28	1.2192	commercial	*Buxus sempervirens*	7.98	1.754386	(34.075429, -118.444563)
L42'	33	1.2192	commercial	*Buxus sempervirens*	5.6	2.946429	(34.075429, -118.444563)
L43'	30	1.2192	commercial	*Buxus sempervirens*	5.28	2.840909	(34.075429, -118.444563)

L51'	63	1.524	commercial	*Buxus sempervirens*	9.8	3.214286	(34.075429, -118.444563)
L52'	44	1.524	commercial	*Buxus sempervirens*	6	3.666667	(34.075429, -118.444563)
L53'	26	1.524	commercial	*Buxus sempervirens*	6.15	2.113821	(34.075429, -118.444563)
L61'	54	1.8288	commercial	*Buxus sempervirens*	7.31	3.69357	(34.075429, -118.444563)
L62'	44	1.8288	commercial	*Buxus sempervirens*	4.42	4.977376	(34.075429, -118.444563)
L63'	33	1.8288	commercial	*Buxus sempervirens*	5.78	2.854671	(34.075429, -118.444563)
L71'	46	2.1336	commercial	*Buxus sempervirens*	7.41	3.103914	(34.075429, -118.444563)
L72'	54	2.1336	commercial	*Buxus sempervirens*	7.8	3.461538	(34.075429, -118.444563)
L73'	42	2.1336	commercial	*Buxus sempervirens*	9	2.333333	(34.075429, -118.444563)
LIA1'	24	0.3048	commercial	*Buxus sempervirens*	7.8	1.538462	(34.075429, -118.444563)
LIA2'	62	0.3048	commercial	*Buxus sempervirens*	7.65	4.052288	(34.075429, -118.444563)
LIA3'	73	0.3048	commercial	*Buxus sempervirens*	5.95	6.134454	(34.075429, -118.444563)

4.2.2. Leaf surface characterization and contact angle measurement

Leaf surface properties can affect whether or not the intercepted microplastics from the wind stream can stick on the leaf surface. As the surface of most plastic particles is hydrophobic, the hydrophobicity of the leaf surface might determine the retention of microplastics on the leaf after initial contact. To test this hypothesis, I measured the contact angle of water droplets on each type of leaf surface using a Contact Angle Goniometer (Rame-Hart 500). The contact angle was measured by placing a 3 µL water droplet onto a pre-washed leaf surface and measuring the angle 10 times using DropImage Advanced software. This process was repeated for 10 droplets on each sample to determine the average contact angle per sample type.

Some leaf samples were analyzed using Scanning Electron Microscopy (SEM) to qualitatively compare the size of stomata with that of deposited particles and measure the scale of the surface roughness, which can increase the retention of deposited microplastic on the surface. As SEM cannot distinguish microplastic particles from other particles, particles with sharp borders and lack of natural organic structures were assumed to be microplastics.

4.2.3. Microplastic isolation and quantification

Microplastics were isolated from leaves following the method outlined elsewhere (Koutnik et al. 2022a). Each rectangular leaf piece was washed thoroughly with 100 mL of deionized water in a glass beaker, and the suspension was vacuum filtrated on 24-mm G4 glass fiber filters with a 1.2-µm pore size (Thermofisher Scientific). To extract microplastics from the soil sample near the tree, 1 g of soil was mixed with 40 mL of 1.6 g mL^{-1} potassium iodide solution for density separation. The suspension was centrifuged at 5000 rpm for 30 min to settle heavier soil particles and isolate lighter (density < 1.6 g cm^{-3}) particles including microplastics from the supernatant.

The floating particles in the supernatant were vacuum filtered onto a 24 mm glass fiber filter paper with a 1.2 μm pore size, dried, and transferred on to a gold-coated slide and analyzed using FTIR.

The isolated microplastics or debris from leaves were quantified using a smartphone method where microplastics were first dyed using Nile Red (Leonard et al., 2022). The method can detect small microplastics (< 10 μm), which is relevant to the atmospheric transport of particles (PM_{10}). In contrast, the FTIR microscope can detect particles > 20 μm, which would miss most of the short particles that are more likely to stay suspended in the atmosphere for a long time. Nile Red has been used to identify microplastics, although it has limitations related to false-positive and the inability to color rubber particles (Erni-Cassola et al., 2017; Kang et al., 2020; Maes et al., 2017b; Prata, 2018b). However, Nile Red is non-selective and can introduce errors if environmental samples containing organic matter could also bind Nile Red (Stanton et al., 2019). To estimate the contribution of the non-plastic organic particles, I digested a subset of deposited dust using hydrogen peroxide and found that there was no significant difference in the concentration of particles detected with or without digestion. Thus, I assumed that the number of organic debris that could have interfered with microplastic measurement was limited in this study.

Details of filter preparation and microplastic counting in this study were described previously (Leonard et al., 2022). The same method was used recently to quantify microplastics in urban soil (Koutnik et al., 2022a; Koutnik et al., 2022b) and leaves (Koutnik et al., 2022a). Briefly, filters containing microplastics were stained with 0.17 mL of 0.5 μg mL^{-1} Nile Red in chloroform solution (Maes et al., 2017b) in a glass petri dish and dried for 24 hours in a fume hood to ensure low background fluorescence from the filter paper. The dyed filter paper enclosed in a LED light-illuminated chamber was then photographed using a smartphone equipped with an external camera. The acquired images of the filter membranes were stored in DNG format and analyzed

with a MATLAB algorithm that extracts the red channel of each acquired image, corresponding to the fluorescence emission wavelength of Nile Red. This method automatically quantifies the number of particles along with the pixel area. The analytical variance of this method is ~4% of the mean.

To measure the abundance of the microplastics on leaf samples, dust particles from some leaves were scraped onto a gold-coated slide for Fourier Transform Infrared spectroscopy (FTIR) analysis (V. S. Koutnik, Leonard, El Rassi, et al., 2022). Microplastics on the gold slides were characterized for their size distribution, shape, and polymer types using FTIR analysis (Thermo Scientific Nicolet iN10). The FTIR microscope was used in the reflectance mode using the particle analysis wizard included in the PICTA software. The FTIR microscope can identify the size distribution of microplastics larger than 20 μm based on the image analysis of particles spread on a 1 cm^2 area of the slide. When comparing sample spectra to spectra databases (which are listed in Section 3.2.5), 60% match criteria were required to identify the particle. Individual sample spectra were then visually examined against their database match to confirm the identification.

4.2.4. Quality assurance and quality control

Plastic cross-contamination was minimized during sampling collection, preparation, and processing steps by using non-plastic tools wherever possible. All glassware, containers, and filtration devices were rinsed with deionized water three times before use. Leaves were sampled with metal shears and aluminum foil, and pre-washed metal or glass containers were used to process samples. The filters were dried, stored, and transported in covered glass Petri dishes and slides. A subset of samples was randomly selected and analyzed using Scanning Electron Microscope (SEM) to study the morphology of microplastics and Fourier Transform Infrared spectroscopy (FTIR) analysis to characterize plastic composition. The morphology and elemental

characteristics of the colloids on gold-coated leaf samples were analyzed using SEM-EDS (Zeiss Supra 40VP). Polymer composition of microplastics were characterized using FTIR analysis (Thermo Scientific NicoletTM iN10) in reflectance mode (Brahney et al., 2020b), using a minimum 50% match criteria to identify the particle. In short, particles from leaf samples were scrapped off onto a clean gold-coated slide and dispersed with ethanol. A particle map of ~1 cm^2 area was analyzed using the particle analysis wizard included in the PICTA™ software. Image separation settings were tuned to best extract the particles from the gold slide manually. Spectra were measured using a normal resolution setting and 8 scans in 1 second, over a spectral range of 4000 - 675cm^{-1}. To identify particle composition and frequency of various polymers, the collected spectra were compared across all available commercial libraries (as listed in Section 3.2.5). An ANOVA test was used to test if microplastic concentrations significantly varied with leaf height or plant location. A Tukey one-way test ($p < 0.05$ indicates significance) was used to compare microplastic concentrations among five types of leaves (**Table 4-3**).

Table 4-3: Table records p-values results from Tukey one-way tests performed in all directions to determine if microplastic concentrations varied significantly by leaf species. P-values < 0.05 represent statistically significant differences and are bolded

	Rhus Ovata	*Acer saccharum*	*Buxus sempervirens*	*Leymus condensatus*	*Chamaerops humilis*
Rhus Ovata		0.9996	0.9817	0.8294	**0.0006**
Acer saccharum			0.9718	0.8379	**0.0045**
Buxus sempervirens				0.9924	**0.0273**
Leymus condensatus					**0.0979**
Chamaerops humilis					

4.3. Results

4.3.1. Microplastic concentrations and size on leaves

Microplastics concentration varies with leaf type (**Figure 4-3**). The concentration varied by two orders of magnitude between 0.14 - 25 n cm^{-2}. Compared to other leaf types, *Chamaeops humilis* had significantly ($p < 0.05$) higher microplastic concentration (**Table 4-3**). However, a large spread denoting the 95% confidence interval indicates a large spread in the concentration of microplastics on a particular leaf type. The result confirmed that leaves could capture a significant amount of airborne microplastics, but the concentration could vary within the same leaf type, possibly because of the elevation of the leaf position, which affects the wind speed and convective power of the wind to carry airborne microplastic (Koutnik et al., 2022a), and the location of the tree in the urban area, which determines the emission and concentration of microplastics in air based on local pollution level and land use.

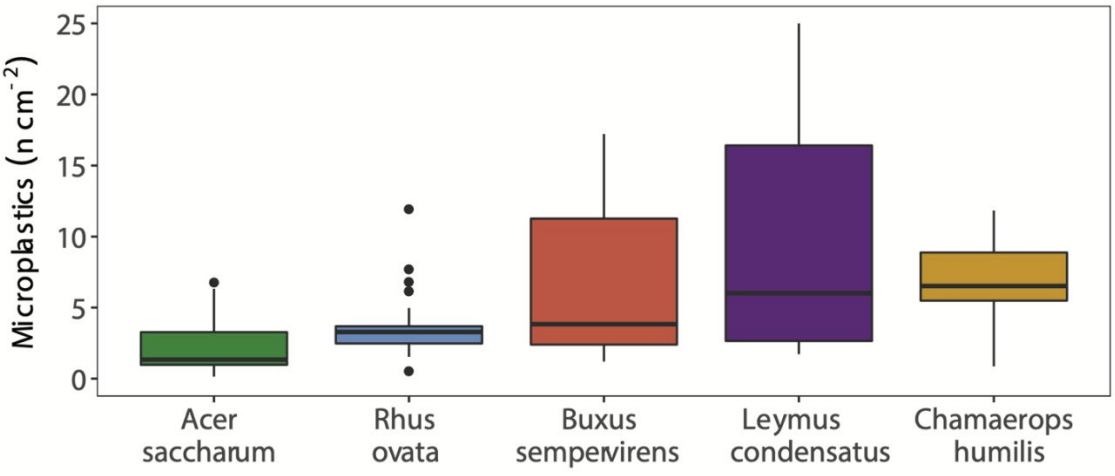


Figure 4-3: Microplastic concentrations range from 1-25 n cm^{-2} per leaf, depending on the leaf type. Compared to other leaf types, *Chamaerops humilis* had significantly ($p < 0.05$) higher microplastic concentration (**Table 4-3**).

SEM analysis confirmed particles or fibers greater than 100 µm (**Figure 4-4A**) could be airborne, deposited on leaves, and potentially cover the stomata. Smaller, inhalable particles (<10

μm) within the size range of stomata were observed on some leaves, but they cannot be positively identified as plastics (**Figure 4-4B**). The mean size range of stomata was around 40 μm, which was larger than some microplastics but not than these smaller microplastics (**Figure 4-4C and 4-4D**). The surface roughness features are in the order of 10-30 μm, which could provide adequate rough edges to prevent smaller microplastics from slipping off from the surface.

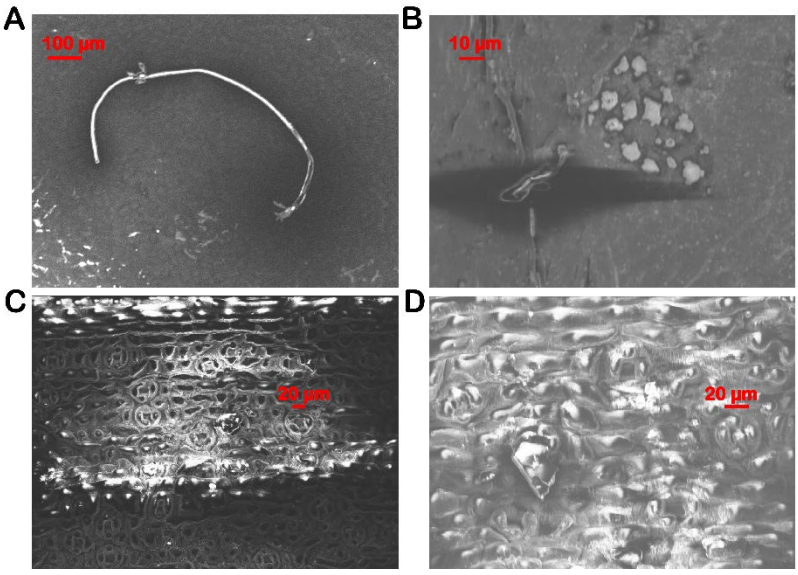


Figure 4-4: SEM analysis of leaf subsurface showing microplastic of various morphologies both A) larger than 100 μm and B) smaller than 10 μm. C) SEM analysis of the leaf subsurface revealed regular stomata patterns. D) Zoomed stomata entrance has widths of approximately 40 μm, which could permit the transport of micro- and nano-plastic particles.

4.3.2. Abundance of microplastics on leaves

FTIR analysis reveals that microplastics found on leaves were much more diverse in composition than the microplastics found in soils sampled near the tree from which leaves were collected (**Figure 4-5**). Both soil and leaf samples had polyethylene (PE) and rayon as the most dominant microplastic types. However, microplastics on leaves were less dominated by these polymers and had a more diverse range including polystyrene (PS), polypropylene (PP), polyvinyl chloride (PVC), and polyacrylic acid (PAA). The FTIR microscope used for microplastic

characterization in this study could not reliably detect microplastics smaller than 20 µm. Thus, the abundance of microplastics determined in this study did not account for the smaller microplastics.

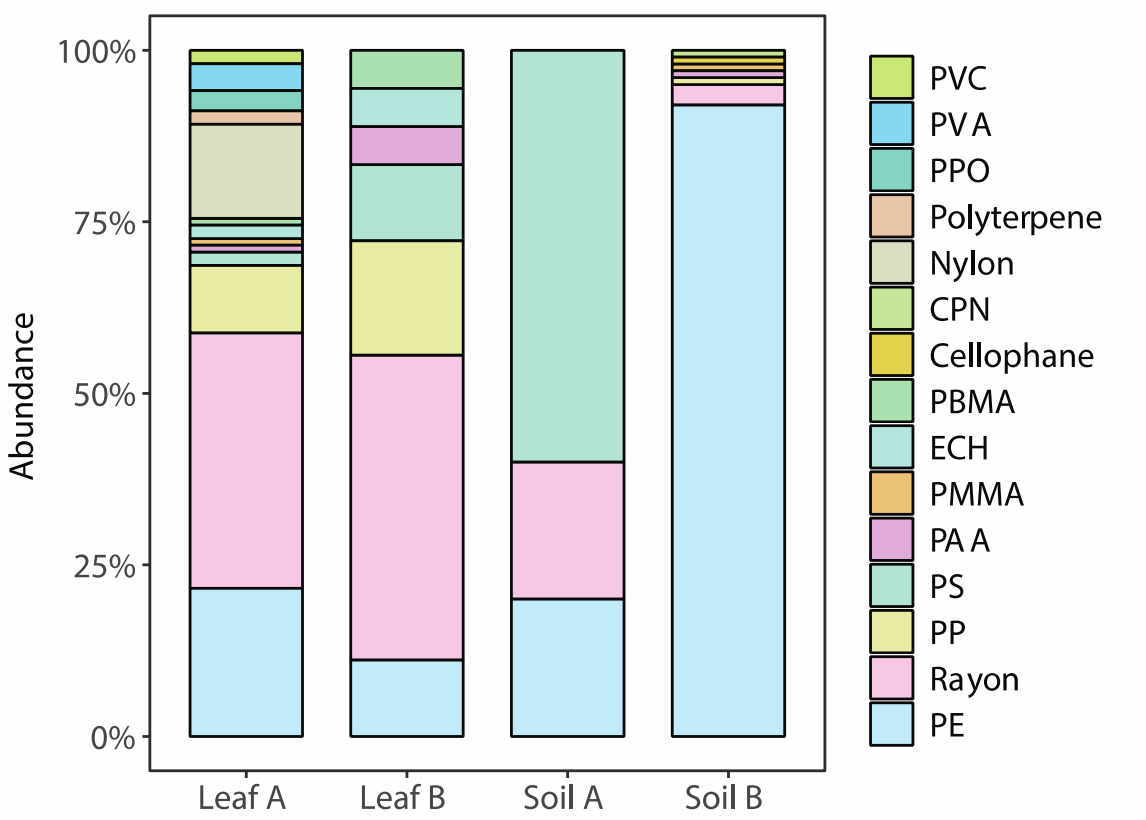


Figure 4-5: Abundance of various microplastic polymer types, as confirmed by FTIR analysis for 2 leaf samples, and 2 soil samples.

4.3.3. Effect of vertical position and land-use type on microplastic concentrations on leaves

Our results confirmed that the leaf position above the ground had a greater effect on the concentration of microplastics than the land-use type near the sampling site (**Figure 4-6**). Leaves positioned within 0.6-1.2 m above the ground had significantly ($p < 0.0006$) higher microplastic concentrations than the leaves collected below or above the critical height range (**Figure 4-6A**). In contrast, microplastic concentrations on leaves collected from sites with different land-use types were not significantly ($p = 0.78$) different (**Figure 4-6B**). Overall, the results indicate that the

position of the leaf above the ground has a greater influence on microplastic concentration than the land use in the area leaves were sampled from.

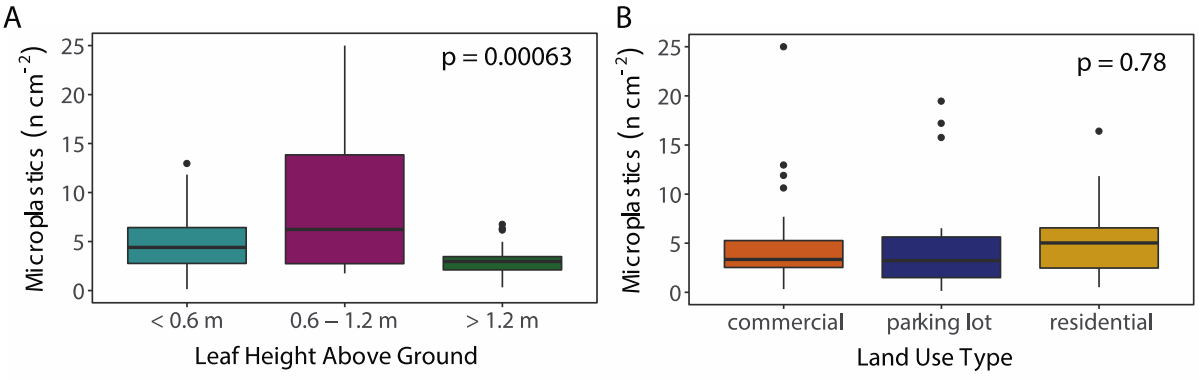


Figure 4-6: Microplastic concentrations on leaves at different as a function of (A) their height and(B) land-use types. A one-way ANOVA test was performed for both variables to determine significance (p-value < 0.05 was assumed significant).

4.3.4. Effect of leaf hydrophobicity on microplastic concentrations

The hydrophobicity of leaf surfaces, as measured by contact angle with water, was significantly different from each other. The contact angle of the leaf surface decreased in the following order: *Rhus ovata* > *Acer saccharum* > *Chamaerops humilis* > *Leymus condensatus* > *Buxus sempervirens* (**Figure 4-7A**). This increase in hydrophobicity slightly resulted in a decrease in microplastic concentrations, but the correlation was weak as these leaves were collected from all heights (**Figure 4-7B**). Only 12% (=R^2) of the concentration data can be predicted based on changes in hydrophobicity.

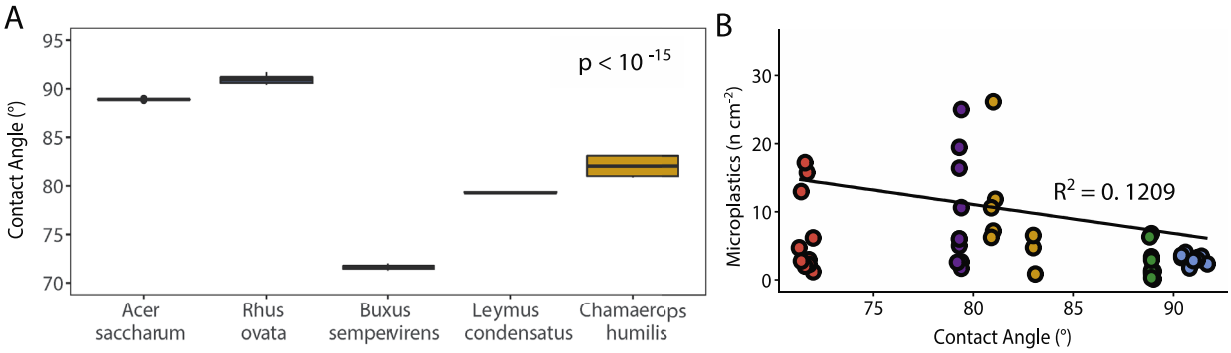


Figure 4-7: (A) Mean contact angle of leaf varied significantly with leaf types (B) Microplastic concentration was inversely related to contact angle or hydrophobicity, although the correlation was very weak (R^2 = 0.1209).

4.4. Discussion

4.4.1. Microplastic concentrations on leaves in urban canopy

The urban tree canopy is known to filter airborne pollutants in urban areas, so they are expected to trap microplastics in urban areas, where atmospheric concentrations are known to be high (Peñalver et al., 2021). In this study, microplastic concentrations varied between 0.14-25 n cm^{-2}. Only two other studies reported leaf concentration in urban areas (Liu et al., 2020; Liu et al., 2022), and the concentration was nearly one order lower magnitude (0.07 - 0.19 n cm^{-2}) than what was found in this study. We attributed the concentration difference to the methodology used, detection limit, and level of error associated with each method (Beaurepaire et al., 2021; Dos Santos Galvão et al., 2022; Prata et al., 2019). For instance, the smartphone method in my study detected microplastics as small as 10 µm, which is smaller than the detection limit of the method used in the previous study (Liu et al., 2020). However, all studies- including the current one- missed particles smaller than 10 µm (Leonard et al., 2022), which are relevant for inhalation toxicity. As future detection methods evolve to identify smaller microplastics (< 10 µm) and nanoplastics, the reported concentration of plastic particles on leaves will likely increase. Thus, the concentration found in this study could still underestimate the actual concentration of microplastics on leaves. More studies should quantify microplastics found at or below this size

range, as they are relevant for inhalation toxicity and phytotoxicity. For instance, a recent study observed foliar uptake of nanoplastics through the stomata (Sun et al., 2021). Thus, smaller microplastics or nanoplastics, which could be created by breaking down large microplastics, could enter the stomata or interfere with photosynthesis (Azeem et al., 2021).

SEM images on leaves confirmed the size of deposited particles, some of which could be microplastics, was smaller than 10 μm. Particulates of this size (PM 10) are not typically reported due to the limitation of optical microscopes and FTIR tools to identify smaller microplastics (Koutnik et al., 2021b). SEM results also confirmed the presence of fiber particles as large as 300 μm. Assuming the large particles with fibrous shape were microplastics or cotton fibers, soil particles of similar size were not found on leaves. Thus, these types of particles may be more susceptible to being suspended in the air than soil particles of similar size. I attribute the preferential emission of microplastic particles to their lower density compared with soil particles (Koutnik et al., 2021a). Based on colloidal emission models- both theoretical (Bagnold, 1941; Ravi et al., 2006) and empirical (Selah & Fryrear, 1995), density is relevant when particulates are greater than 100 μm. This explains this study did not find similarly sized soil particles as plastic ones, since the former's higher density makes them more difficult to resuspend into the atmosphere compared to microplastics. For the same reason, an enrichment of microplastics in dust was observed in recent wind tunnel experiments (Bullard et al., 2021b; Rezaei et al., 2019). However, these studies rarely analyzed microplastics in the size range (< 10 μm) relevant for long-range transport or inhalation toxicity in urban areas. Thus, future studies should examine the mechanisms for emission of smaller microplastics.

4.4.2. Microplastic concentration as a function of leaf position above the ground

The concentration of airborne particulate matter on leaves in the urban canopy can vary with their elevation above the ground since elevation affects wind speeds, and thus the concentration of airborne particulate matter. Thus, it is expected that a specific height above the ground would have a peak concentration of microplastics. In this study, I observed that microplastic concentrations on the urban canopy peaked between 0.6 – 1.2 m from the ground surface. The result confirmed that leaf position with respect to the ground surface plays a critical role in trapping microplastics from wind streams. The concentration of airborne particles at a certain height depends on their size or density. Density affects the downward gravitation pull and the wind velocity required to keep the particles suspended against the gravitational pull (Weathers & Ponette-González, 2011). Near the surface, wind speed is the lowest because surface roughness retards wind speed (Wizelius, 2007). With an increase in elevation, wind speeds increase (Bowen & Lindley, 1977; R. A. Schmidt, 1982) thereby increasing their capacity to keep particles suspended. However, an increase in elevation also increases the potential energy or gravitational pull (Weathers, 2011). Thus, the concentration is expected to be highest at a certain height.

4.4.3. Microplastic accumulation on leaves based on land-use

Our results showed that microplastic concentrations on leaves were not correlated with land use. Emissions from commercial, parking lot, and residential areas were expected to vary based on plastic uses and site environmental conditions (Järlskog et al., 2021). However, similar concentrations between different locations indicate that the concentration of microplastics on leaves may not be determined by their geographical proximity to potential sources. This is possible when emitted microplastics move far from the sources across geographical boundaries. In this study, the entire sample collection area spans over 276 km^2. The scale is still small compared to

the long-range transport scale of microplastics observed in recent studies (Allen et al., 2019; Evangeliou et al., 2020b; Feng et al., 2020). Thus, microplastics created in one place can be dispersed into the atmosphere and deposited uniformly across the urban canopy. Continuous deposition of atmospheric fallout could eventually result in uniform deposition of microplastics irrespective of land use. For the same reason, the concentrations of microplastics within stormwater control measures was found similar to concentrations outside stormwater control measures, even though stormwater control measures receive high loading of microplastics (Koutnik et al., 2022b). The results further confirm that the concentration of microplastics within a short distance scale may not be influenced by land use, due to the continuous deposition of airborne microplastics. Thus, while leaf microplastic concentration may serve as an indicator for exposure risks to airborne microplastics in the general region, they are not an indicator for their proximity to potential sources of microplastics, at least on the small scale used in this study.

4.4.4. Weak correlation between leaf surface hydrophilicity and microplastic concentration

Leaves have a diverse range of surface properties such as hydrophobicity, stomatal density (Zheng & Li, 2019), leaf hair, and wax cover (Sæbø et al., 2012). A variation in these properties could affect the retention of microplastics on leaves. This study examined the contribution of surface hydrophobicity to the retention of microplastics. I found that microplastic concentrations on leaves were negatively correlated with contact angle, although the correlation was weak (R^2 = 0.12). That is, decreasing contact angles or increasing hydrophilicity, appears to increase the concentration of microplastics on leaves (**Figure 4-7**). The results included concentration data from leaves of all heights, a factor that plays a major role in the concentration variability and which could have obscured stronger correlations. However, I lacked sufficient samples from one height range for all plants to rule out the variability associated with leaf position above the ground.

Despite variability based on heights, the existing weak correlation between microplastic concentration and hydrophilicity indicates that the formation of water film or liquid bridge based on atmospheric humidity could act as a glue to bind particulate matter such as microplastics (Seville et al., 2000). Other possibilities of variability are surface roughness such as leaf hair and leaf orientation against the winds. SEM data reveals that surface roughness on leaves has a relative dimension of 10-30 μm. Thus, microplastics with sizes lower than the roughness dimension would likely be affected to a greater extent. Future studies should examine how the roughness dimension affects microplastic interaction and retention on leaves.

4.5. Conclusions

The current study identified factors that could affect the concentration of microplastics on leaves to evaluate the use of leaves as passive samplers to assess microplastic pollution in urban areas. Among the three factors—the height of the leaf above ground, land-use type, and leaf surface hydrophobicity—height contributes most to variability in the concentration of microplastics on leaves. Leaf hydrophilicity, based on the contact measurement, is weakly correlated with microplastic concentrations on the leaves. Land use types have no influence on microplastic concentration on leaves, potentially due to continuous atmospheric deposition of airborne microplastics originating from distant sources. Thus, the concentration on leaves cannot precisely indicate the emission potential of sources near the canopy from where leaf samples are collected. The concentration on leaves merely indicates the total amount of microplastics deposited from the atmosphere, thereby informing potential inhalation risks in the region. SEM analysis confirmed that large microplastics (> 100 μm) could be transported by the wind. The same analysis also confirmed the presence of smaller (< 10 μm) microplastics, which can potentially enter leaf stomata, which were observed to have widths around 40 μm. Collectively, the results indicate that

leaves may not serve as a reliable passive sampler unless the sampling protocol is modified to account for uncertainty associated with concentration variation. The results could inform future efforts to develop consistent protocols for using leaves as a biomonitoring system for airborne microplastics in urban areas.

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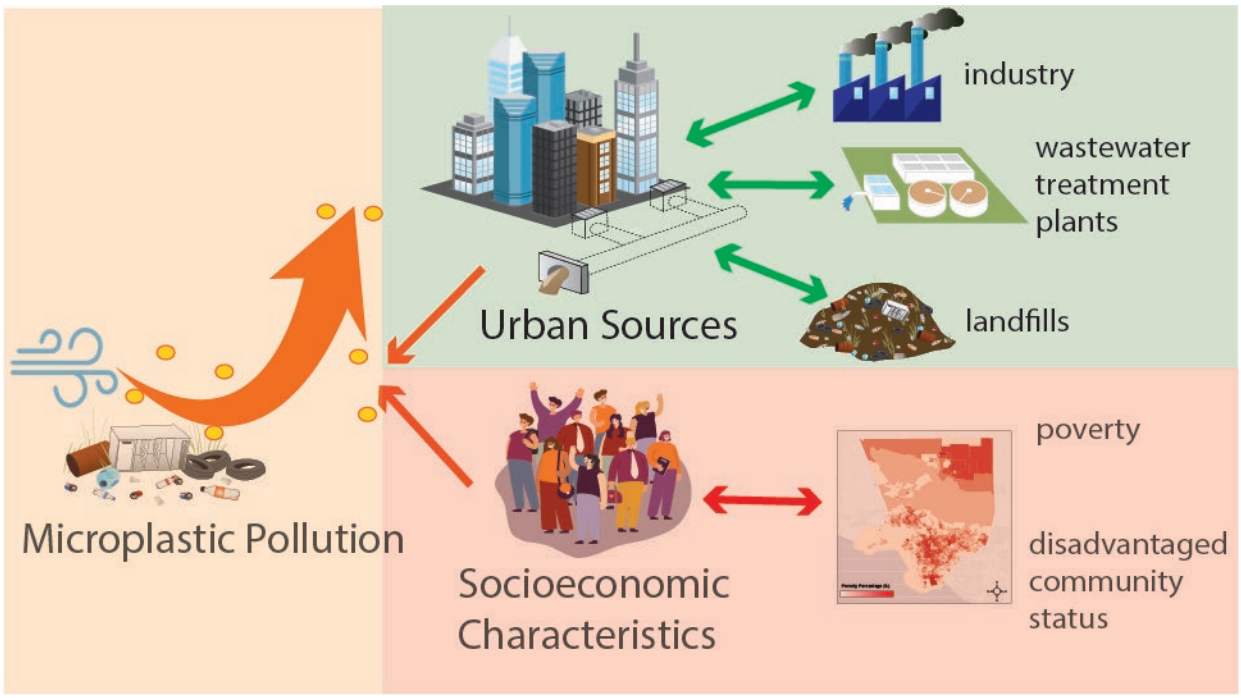
5. CHAPTER 5 – MICROPLASTICS EXPOSE EVERYONE: SOCIOECONOMIC AND LAND USE FACTORS DO NOT CORRELATE WITH MICROPLASTICS IN DEPOSITED DUSTS AT SCHOOLS IN LOS ANGELES



Leonard, J., Jing, W., Rodriguez, C., Tran, L., Lowe, L., Win Cowger, Patterson, R.F., and Mohanty, S.K. (2024) Spatial Distribution of Microplastics in Atmospheric Dusts Cross-referenced with Socioeconomic Status in Los Angeles, CA, USA. Environmental Pollution. In Review.

Abstract

Low-income communities are often disproportionately exposed to particulate matter due to their proximity to emission sources, such as near freeways. Microplastics are a new class of particulate matter found in the air, and they may exhibit similar patterns as particulate matter (e.g., PM 10) in response to changes in land use or socioeconomic factors. Yet, little is known if these factors could correlate with the likelihood of microplastic exposure in densely populated urban communities, particularly school children. To answer this, I measured outdoor microplastic concentrations in the deposited dust near schools in Los Angeles and correlated it with a) socioeconomic status, or b) their proximities to known sources of microplastics, such as wastewater treatment plants, landfills, and industrial facilities. Surprisingly, results showed no significant correlation with any of the socioeconomic variables and land use factors. The results confirmed that the spatial distribution of microplastic exposure via air does not map onto observed variations in socioeconomic status. I attributed the results to the transport and distribution of microplastics in the atmosphere in urban areas, thereby exposing everyone, rich or poor, irrespective of where they live, to a similar high concentration of microplastics. Thus, social indicators that are typically significant determinants of disproportionate particulate pollutant burden do not apply for microplastics. Predicting exposure risk to deposited microplastics may be challenging due to their ubiquitous and abundant presence.

5.1. Introduction

Microplastics, a new class of particulate matter, are frequently found in the air (Beaurepaire et al., 2021; Zhang et al., 2020), thereby increasing their inhalation risk and associated adverse health effects (Blackburn and Green, 2022). Once in the human body, microplastics can enter the bloodstream (Leslie et al., 2022), lungs (Amato-Lourenço et al., 2021), and placenta (Leslie et al., 2022), penetrating internal organs and causing oxidative stress (Zhu et al., 2024). A recent study correlated microplastic deposited in arteries to increased risk of cardiovascular disease (Marfella Raffaele et al., 2024). Humans can be exposed to microplastics from different pathways including water (Oßmann, 2021), air (Borthakur et al., 2021; Yuan et al., 2022), and food (De-la-Torre, 2020). Among different sources, airborne microplastics can travel across geographies, thereby making it challenging to predict the locations where one might inhale more microplastics. Understanding social and land use factors that are associated with increased exposure risk to microplastics could inform the development of management methods to minimize their exposure in urban areas.

In urban areas, microplastics are released from diverse sources including roadways, industrial facilities (D'Ambro et al., 2021; Lamichhane et al., 2023; Wright and Kelly, 2017), wastewater treatment plants (Brahney et al., 2020; Ding et al., 2021), and landfills (Njoku et al., 2019; Weinberg et al., 2011), and dispersed by wind (Chen et al., 2020; Rafique et al., 2020; Wu et al., 2022). Some of the airborne microplastics are filtered by vegetative canopy, and others are deposited back on terrestrial land based on the local climate conditions (Liu et al., 2020; Rochman, 2018; Zhang et al., 2020). The deposited microplastics can be used to understand exposure risk to microplastics in a region. Generally, it is expected that proximity to the known source of microplastics should increase the concentration of microplastics in the deposited dust.

Furthermore, land use factors such as green space or uncovered land roadways in urban areas could correlate with the amount of microplastics removed or deposited in an area.

Land use factors have been traditionally used to predict exposure to air pollutants such as PM10 or PM2.5 (Eeftens et al., 2012; Wu et al., 2015). For instance, communities living close to freeways are more likely to be exposed to particulate pollutants than communities living far from roadways and in locations covered with trees (Akhbarizadeh et al., 2021; Li et al., 2016). Additionally, racial residential segregation and socioeconomic status have been identified as important factors for disproportionate exposure to air pollution. Poverty is often utilized as a proxy for identifying disadvantaged communities with potentially higher exposure to atmospheric pollutants (Morello-Frosch et al., 2001; O'Neill et al., 2003; Pastor Jr. et al., 2005). In Los Angeles County, Green Zone Districts are defined to identify communities that bear a disproportionate pollution burden, and which are the focus of public policies to improve these conditions (LA County Planning, 2022). Together, these variables may lead to increased exposure to microplastic pollution, as well as increase vulnerability to its health risks. Many studies correlated PM with socioeconomic variables such as poverty (Bell and Ebisu, 2012; Mikati et al., 2018; Rentschler and Leonova, 2023) and land use factors such as proximity to landfills, wastewater treatment plants, and industrial sites (Li et al., 2016; Mukherjee and Agrawal, 2017; Shi et al., 2020). Understanding whether these factors also influence microplastic deposition in atmospheric dust is essential for assessing exposure risks and implementing effective mitigation strategies.

This study investigated whether deposited atmospheric microplastic concentration varied spatially based on socioeconomic variables such as poverty and Green Zone District status, as well as proximity to known microplastic sources such as landfills, wastewater treatment sites, and industrial sites. To achieve the objectives, deposited dust was collected from 66 sites (48 of which

were schools) in Los Angeles County and analyzed for microplastic concentrations, abundance, and size distributions. The result will inform the extent to which these land use and socioeconomic factors can help predict microplastic pollution or exposure risks to school children and communities in urban areas.

5.2. Method

5.2.1. Sampling sites selection

Two types of sites (n = 66) were selected for sampling within Los Angeles County: California Air Resource Board (CARB) air monitoring sites and school sites (**Figure 5-1**). CARB sites aim to monitor ambient air quality and particulate concentrations and were included to collocate with relevant regulatory sites within this region of interest. CARB site addresses and GPS coordinates were sourced from their online database, filtering by Los Angeles (LA) County (California Air Resources Board, n.d.). School sites in LA county were selected by first finding census tracts with varying poverty levels using CalEnviroScreen 4.0, which is based on US Census Data from the American Community Survey (Office of Environmental Health Hazard Assessment (OEHHA), 2021.) Ten communities were selected from each of the 5 poverty quantile groups: 100-80%, 80 – 60 %, 60 – 40 %, 40 – 20%, and 20 – 0 %, where poverty is defined as the percentage of that census tract's population with incomes below two times the federal poverty level as determined by the U.S. Census Bureau. A total of 48 schools in these census tracts were then selected for sampling.

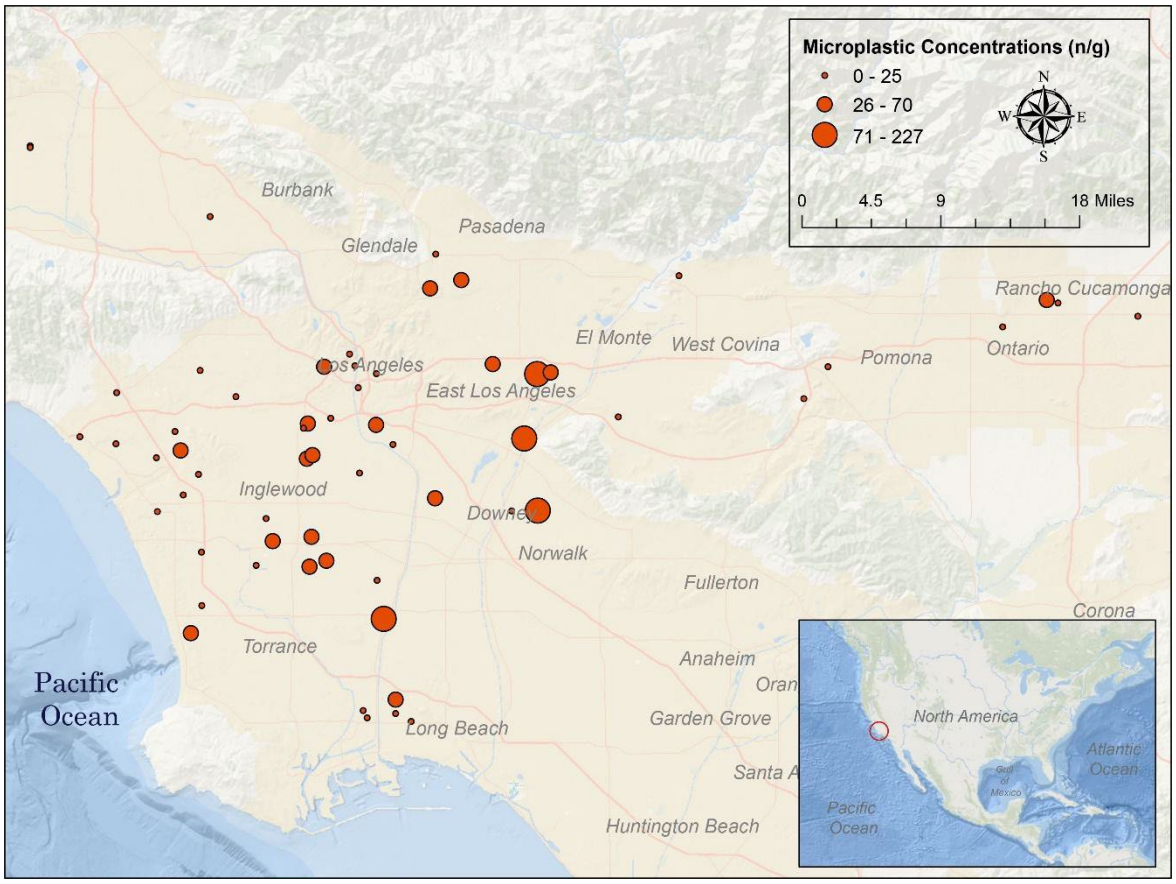


Figure 5-1: Deposited dust samples from sidewalks in Los Angeles were collected from two types of sites (n = 66): California Air Resource Board (CARB) air monitoring sites (18) and schools (48).

5.2.2. Social Vulnerability Index Variable Selection and Analysis

To explore the spatial correlation between microplastic concentrations in deposited dust and the factors that could affect the concentration, shapefile data specific to the region of interest were sourced from appropriate open-source data. Shapefile data was mined from county-level institutions to assign each sampling location a total of 4 land use attributes relevant for microplastic pollution: (1) green zone districts, LA County Planning, (2) landfills, LA County Department of Public Works, GIS Unit, (3) wastewater treatment plants (WWTP), LA County Sanitation District, and (4) industrial sites, LA County Location Management System. Data was analyzed in ArcGIS Pro for spatial analysis and mapping.

For geospatial analysis, I used near table analysis to estimate the distances between the sampling sites and the locations associated with the variables of interest. Then, an appropriate radius for each investigated variable was determined, and binary values were assigned to indicate proximity to specific variables of interest. These radius criteria were derived from established standards for residential proximity to environmental hazards (Brender et al., 2011; Njoku et al., 2019). Here, I adhered to a standard radius of 2 km or 5 km using ArcGIS to accurately estimate spatial proximity to sample sites. These binary tables, indicating proximity to risk factors, were analyzed in R using statistical t-tests and Fisher exact tests to examine the correlation between microplastic concentrations and the land use factors that may influence them.

5.2.3. Deposited atmospheric dust sample collection

Outdoor deposited dust samples were collected from all 66 sampling sites using a natural corn bristle broom, stainless steel dustpan, and stainless-steel spatula or spoons to avoid plastic contamination. Windowsills, rooftop gutters, sidewalks, and other non-plastic, impervious surfaces exposed to atmospheric fallout were brushed with the broom, and dust collected into the dustpan. The composite samples were wrapped in aluminum foil and labeled with site information. All equipment was thoroughly cleaned using deionized (DI) water and wiped between sampling to prevent cross-contamination.

5.2.4. Extraction of microplastics from dust samples

Microplastics were isolated from dust samples following the method described previously (Koutnik et al., 2022b). Briefly, samples were first sieved using a metal sieve (>600 μm) to remove large debris. To settle heavier soil particles and isolate lighter (density <1.6 g cm^{-3}) particles including microplastics, 1 g of sieved sample was mixed with 40 mL of 1.6 g mL^{-1} potassium iodide (KI, Thermofisher Fisherbrand, P410-500) solution and centrifuged at 5000 rpm for 30 min.

The supernatant was vacuum filtrated to trap floating debris on a 24 mm glass fiber filter paper with a 1.2 μm pore size (Thermofisher Scientific, 09-804-24C). The filter was then placed inside a glass Pyrex petri dish, covered with a glass cover, and left to dry before analysis using specific methods including smartphone-method (Leonard et al., 2022) and FTIR (Xu et al., 2019).

5.2.5. Quantification of microplastics

The concentration of microplastics on the filter paper was quantified by dyeing the filter with Nile Red and capturing an image of fluorescent particles with a smartphone-based fluorescence microscope as described in my earlier work (Leonard et al., 2022). This method has been previously used to assess microplastic concentrations in deposited dust on leaves (Leonard et al., 2023) and urban stormwater infrastructures (Koutnik et al., 2022b). Briefly, filters in a glass petri dish were dyed with 0.17 mL of 0.5 μg mL^{-1} Nile Red in chloroform solution and air-dried with a glass cover for 24 h in the fume hood. Dried filter membranes were transferred onto glass slides, covered with a glass coverslip to eliminate further contamination by dust deposition. The membranes were imaged using a smartphone-based fluorescence microscope. The method could detect microplastics as small as 10 μm in a large field of view of 490 mm^2, but has limitations associated with the selectivity of Nile Red to bind plastic polymers. Thus, this method was used for screening of samples and a subset of the samples was validated using FTIR.

All concentrations reported here within are from smartphone analysis. Samples from a subset of locations (n = 6) were then characterized with FTIR following the method described by Koutnik et al. (2022b) to extract size and abundance distributions. Isolated microplastics from filters were scraped onto gold-coated slides to confirm microplastic concentrations, size distributions, and abundance by polymer types using FTIR analysis (Thermo Scientific Nicolet iN10). The FTIR microscope was used in the reflectance mode using the particle analysis wizard

included in the PICTA software to identify microplastics larger than 20 μm based on the image analysis of particles spread on a 1 cm^2 area of the slide. When comparing sample spectra to spectra databases, 70% match criteria were set to identify the particle type.

5.2.6. Quality Control

At all experimental steps, the use of plastic materials or the possibility of cross-contamination was minimized. During lab work, clothing made from natural materials was worn to prevent cross-contamination of the samples. For sampling, storage, and processing, pre-washed glass and aluminum containers were used. All clean glassware and containers were rinsed with DI water three times. The DI water was analyzed following the methodology used for supernatant analysis for possible microplastic contamination. At all times when samples were processed, dried, or stored, they were covered by glass covers or aluminum foil. The methodology itself (which is externally validated and published) has an average recovery rate of 93.7% ± 13.7%, a human processing variation of 6.8% of the mean, and a sample processing variation of 9.1% (Koutnik et al., 2022a, Leonard et al., 2022). Therefore, the total maximum error for each of the microplastic measurements was estimated to be 22.2%. All statistical analyses were carried out using R software (Rstudio 2023.06.2). For each variable, t-tests with a confidence level of 95% (difference of means) and Fisher's exact tests were performed to verify whether concentrations varied as a result of the variable. Furthermore, a retrospective power analysis was performed, and for some of the source variables the power was < 0.80 (**Table 5.1**) indicating insufficient sample sizes for the null group to confirm the existence of a relationship. All significance, or lack thereof is discussed with this limitation in mind. Nevertheless, this is the first study to attempt to conduct spatial analysis of deposited airborne microplastic concentrations and is scientifically significant, even if statistical significance is limited.

Table 5-1: Retrospective (post-hoc) power analysis performed for proximity to source variables

Proximity to Which Source	*Power*
Green Zones Districts	0.6636794
Landfills	0.1586016
WWTP	0.8393154
Industrial sites	0.3381298

5.3. Results and Discussion

5.3.1. Microplastic abundance and size distribution in deposited dust

FTIR analysis reveals that microplastics found in deposited dust were diverse in composition with 10 species of polymer present (**Fig. 5-2A**). Dust samples were predominantly polystyrene (PS, 51%), olefin (26%), and polyacrylamide (11%) with relatively negligible amounts of polyethylene terephthalate (PET), polyethylene (PE), nylon, rayon, polypropylene (PP), polyamides, and polyvinyl chloride (PVC). This diverse distribution is similar to other studies looking at plastics in LA on leaves (Leonard et al., 2023) and in urban playgrounds (Koutnik et al., 2022), which found 9 and 15 species of polymers, respectively. Common in all three LA-focused studies, PS, PP, PE, and rayon dominate the distribution. Olefins are a class of plastic which includes both PS and PP, and thus the abundance of PS and olefins (PS and PP) in this study are consistent with other studies analyzing deposited microplastics in the same urban region. PP (930 kg/m3), PS (1005 kg/m3) and PP (910 kg/m3) are light polymers predicted to preferentially resuspend into the atmosphere due to their low density and high hydrophobicity reducing electrostatic binding forces (Leonard et al., 2024). Thus, it makes sense they are present in high concentrations in the deposited atmospheric dusts sampled in this study.

Microplastics found in samples were predominantly larger, with 80% of them being larger than 50 μm (**Figure 5-2B**). This could indicate that larger microplastics settle out of the atmosphere due to increased density (Leonard et al., 2024), while finer microplastics stay suspended in the atmosphere and are potentially spread further in the environment by wind. The abundance and size class of microplastics in the atmosphere depend on the characteristics of the source from which they were emitted. Using a theoretical framework (Leonard et al., 2024), I have shown that microplastics within the size range of 100-200 μm are more susceptible to emission by wind because smaller microplastics are easily bound to soil by interparticle force. This preferential emission of larger-sized microplastics is confirmed by existing literature (Abbasi et al., 2019; Allen et al., 2019; Boakes et al., 2023; Kyriakoudes and Turner, 2023; Liu et al., 2019; Zhou et al., 2017). Thus, my results indicate that this larger size range could also be the predominant size class found in deposited atmospheric dust. However, analysis of suspended particulates using passive and active samplers, not just deposited atmospheric fallout like in this study, could confirm which microplastic size class is prominent in dust samples. It should be noted that FTIR microscopy in this study could not detect microplastics smaller than 20 μm. Thus, the abundance of microplastics determined in this study did not account for the smaller microplastics.

The results reveal that microplastics in the samples were predominantly nonfibrous: 99% of the microplastics had an aspect ratio (length to width) below 5 (**Figure 5-2C**). Microplastics with a lower aspect ratio could exhibit a propensity for settling out of the atmosphere due to their comparatively greater mass-to-surface-area ratio, facilitating gravitational sedimentation which enriches atmospheric dusts (Yang et al., 2024). Because of the elongated shape of fibrous microplastics, they have lower aerodynamic diameters compared to spherical particles of identical volume (Xiao et al., 2023), which reduces their settling velocity (Dietrich, 1982; Zhang and Choi,

2022) and increases their range of atmospheric transport (Yang et al., 2024). In fact, recent studies suggest that the settling velocity of fibrous particles may be lowered by 76% compared to equivalent volume spherical particulates resulting in longer suspension times (Tatsii et al., 2023). While there are limited experimental studies analyzing the influence of shape on settling in air, rather than water, fibrous microplastics are more likely to stay resuspended, while similarly sized nonfibrous microplastics may settle out in atmospheric fallout (Dris et al., 2016). The results underscore the pivotal role of atmospheric dynamics and particle characteristics in governing the fate and distribution of microplastics in environmental compartments. The remaining findings discuss each social and environmental factor and its relationship with microplastic concentrations.

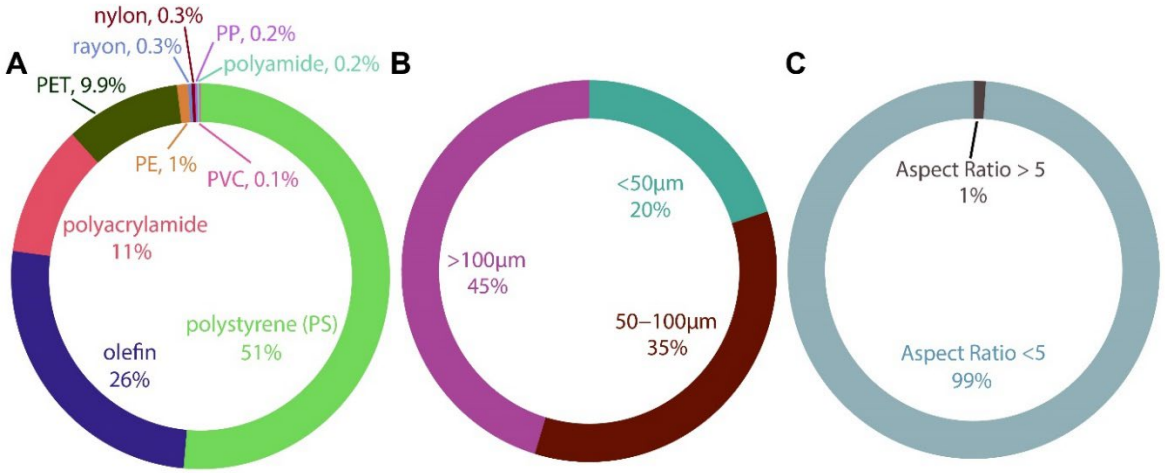


Figure 5-2: Abundance distributions for characterized microplastic properties: (A) polymer types; (B) size ranges, <50 µm, 50-100 µm, > 100 µm; (C) aspect ratio greater or less than 5 (used to determine fiber vs. nonfiber particles) as confirmed by FTIR analysis for 1048 plastic particles.

5.3.2. Poverty and microplastic concentrations

Microplastic concentration found in deposited dusts did not strongly correlate with the level of poverty of schools in the Los Angeles region (**Figure 5-3**). Microplastic counts (n g^{-1}) for different poverty levels were statistically similar ($p > 0.05$), however Fisher's exact test revealed

that the odds of having low microplastic concentrations is greater when having lower poverty rates than high poverty rates. Microplastic concentrations above 30 n g^{-1} are correlated with poverty percentages between 54-62% (**Figure 5-4**). The result indicates that the socioeconomic status of a community may not be a perfect determinant of exposure to microplastics, but that there is a relationship. This result differs slightly from existing literature, which show that lower socioeconomic status is strongly associated with increased exposure to PM 2.5 or PM 10 particles (Morello-Frosch et al., 2001; Rentschler and Leonova, 2023). It should be noted that the microplastics found in my samples are larger (**Figure 5-2**), and my analytical method (FTIR) could not detect samples below 20 µm. This could underestimate microplastics with diameters similar to PM 2.5 and PM 10. However, previous studies have identified the dominant size class of microplastics in wind-driven sediment samples to be greater than 100 µm (Leonard et al., 2024), which implies this detection limit should be sufficient. Thus, the results indicate that spatial variation of microplastic deposition has weak association with socioeconomic status. Atmospheric transport and deposition of particulate matter including microplastics is typically governed by wind transport that occurs across geographic boundaries. Thus, the scale of a city such as Los Angeles, 27,000 km^2, is small compared to the scale of the atmospheric transport of microplastics, and microplastic deposition rate could be similar across Los Angeles. Concentrations ranged from 0-44 microplastics per gram of dust sample, which is consistent with the median concentration of microplastics in atmospheric fallout from 8 other studies with similar units of n g^{-1}, 43.6 (Abbasi et al., 2023, 2019, 2017; Dehghani et al., 2017; Mbachu et al., 2020; Rezaei et al., 2022; Sridharan et al., 2021; Zhang et al., 2020). The standard deviation of microplastic concentrations for CARB sites was significantly smaller than that for school sites with varying poverty level. This may be the case since CARB sites are traditionally quite remote and away from the human activity that is

present at school sites- school drop off and pickup, recess on playgrounds, plastic packaging for lunch foods, etc.

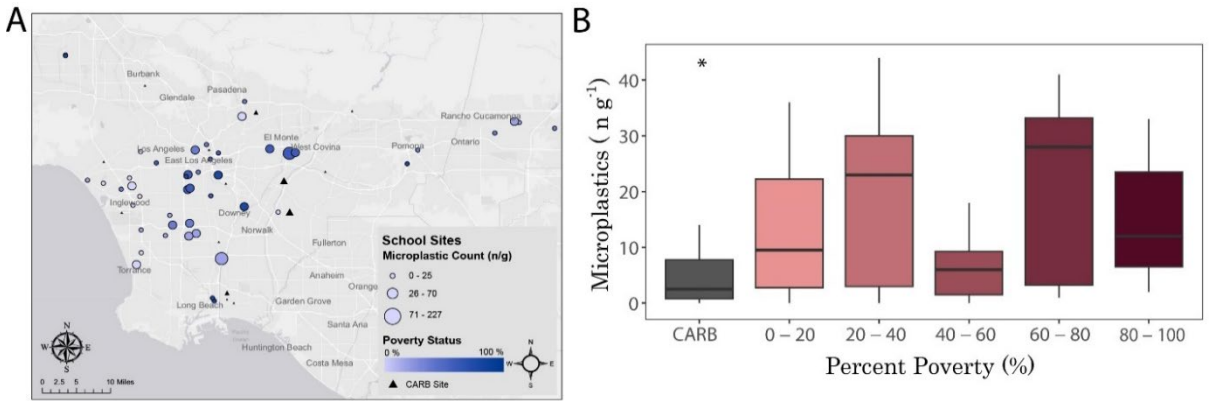


Figure 5-3: (A) Map showing the spatial location of sampling sites with increasing circles of graduated size representing increasing microplastic concentrations in deposited atmospheric dusts at these sites (B) Microplastic concentrations in deposited atmospheric dusts (n g^{-1}) do not vary across sampling sites with varying poverty levels: 100-80%, 80 – 60 %, 60 – 40 %, 40 – 20%, 20 – 0 %.

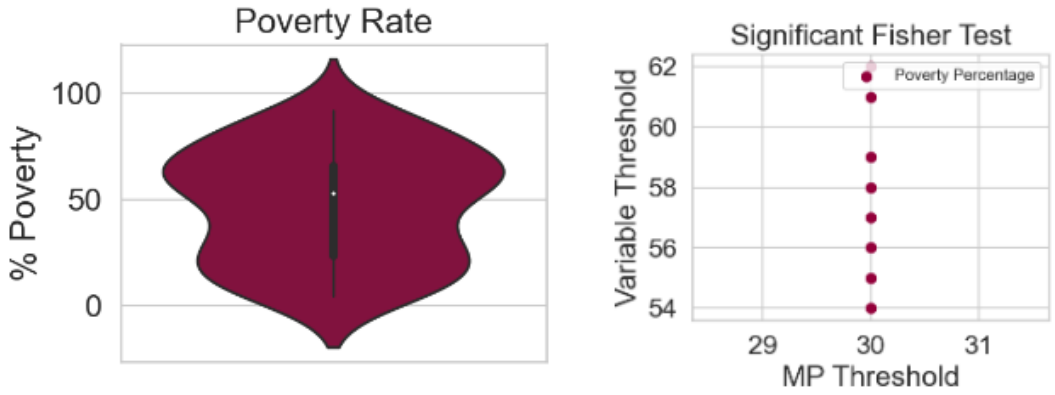


Figure 5-4: (A) Violin plot representing the distribution of poverty rate as a percentage, with a symmetrical distribution around the 50% median (B) Scatter plot indicating significant Fisher's test results for poverty percentages between 54-62%, with a threshold of microplastics concentrations higher than 30 n g^{-1}

5.3.3. Green Zone Districts and microplastic concentrations

Microplastics did not exhibit a statistically significant correlatation with proximity to Green Zone Districts, which are communities designated by Los Angeles County due to their disproportionate pollution burden (**Figure 5-5**). There was a difference in the statistics for concentrations within a green zone district (mean = 21.93 n g^{-1}, median = 24 n g^{-1}) and not within

(mean = 13.56 n g^{-1}, median = 6 n g^{-1}). However, these differences in mean microplastic count(n g^{-1}) were not statistically significant, though this conclusion is limited by the power of the study (power = 0.66). Proximity to a Green Zone district is correlated either positively with atmospheric pollutants due to the disproportionate pollution burden, or negatively with the pollutants due to higher environmental initiatives and community engagement (LA County Planning, 2022). In this study, these communities did not appear to statistically correlate with higher microplastics concentrations in the deposited dusts. This may be due to effective community clean-up projects and policies, or due to a lack of statistical power. This result differs from traditional environmental justice literature, which suggests their behavior, transport, and hotspots may differ significantly. Microplastic concentrations may not be proportional to, nor predicted by, regions of disproportionate pollution burden. This result prompts further investigation into microplastics, its spatial distribution in the environment, and the social and environmental factors that are associated with higher contamination levels.

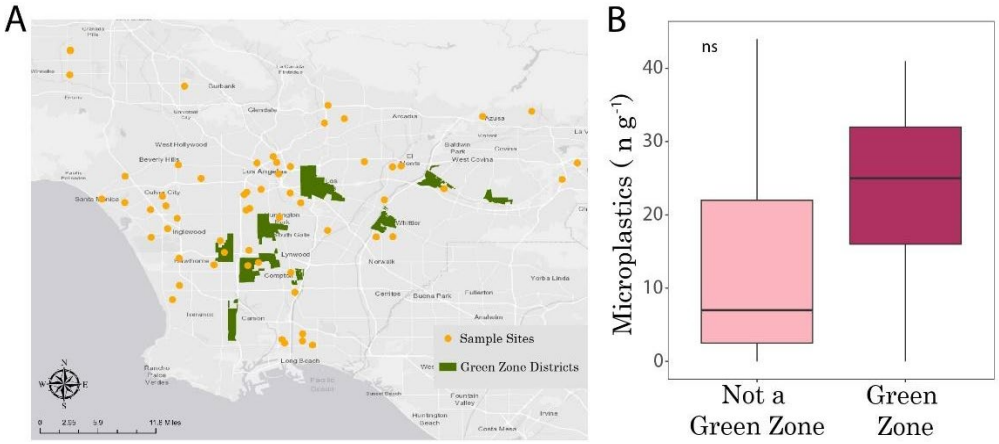


Figure 5-5: (A) Map showing the spatial location of sampling sites overlayed with green regions outlining Green Zone Districts within the region of interest (B) Microplastic concentrations in deposited atmospheric dust (n g^{-1}) do not vary across sampling sites within and outside of Green Zone Districts.

*5.3.4. **Proximity to landfills and microplastic*** concentrations

Landfills are a known source of microplastics, and they release microplastics into the environment via landfill leachate and wind (Upadhyay and Bajpai, 2021; Yadav et al., 2020). Analyzing the proximity of the sites to landfills, I found that microplastic concentrations in the deposited dust samples did not correlate with proximity to landfills within a 2 km radius (**Figure 5-6**). There was a difference in the statistics for concentrations within 2 km of a landfill (mean = 13. 57 n g^{-1}, median = 12 n g^{-1}) and not near a landfill (mean = 17.89 n g^{-1}, median = 8 n g^{-1}). However, statistical analysis revealed that microplastic counts (n g^{-1}) in sampling points located inside and outside the designated 2 km boundary encompassing landfill sites are statistically similar ($p > 0.05$), though this conclusion is limited by the low power of the study (power = 0.16). This result indicates that microplastic exposure from landfills may not follow the same pathways as other pollutants found in the air originating from landfills (Njoku et al., 2019; Salami and Popoola, 2023). Instead, the lack of correlation suggests that atmospheric microplastics do not stay proximal to their sources and that wind is a prominent vector for the long-range transport of microplastics (Evangeliou et al., 2020; Liu et al., 2019). Furthermore, these findings imply that microplastic distribution patterns are not solely governed by proximity to sources but are influenced by additional factors, warranting further investigation into the mechanisms underlying the spatial distribution of microplastics in terrestrial ecosystems.

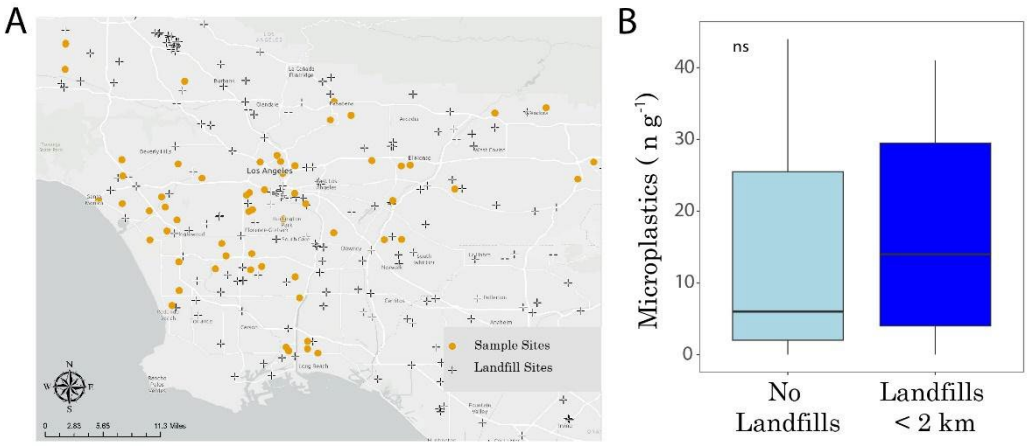


Figure 5-6: (A) Map showing the spatial location of sampling sites (orange circles) overlayed with crosses representing landfill sites within the region of interest (B) Microplastic concentrations in deposited atmospheric dusts (n g^{-1}) do not vary across sampling sites which are either proximal or non-proximal to landfill sites.

5.3.5. Proximity to wastewater treatment plants and microplastic deposition

Microplastic concentrations were not correlated with proximity to wastewater treatment plants (WWTP) within a 5km radius (**Figure 5-7**). There was a difference in the statistics for concentrations within 5 km of a WWTP (mean = 7.14 n g^{-1}, median = 2 n g^{-1}) and not near a WWTP (mean = 16.8 n g^{-1}, median = 12 n g^{-1}). Proximity to WWTP within 5 km appears to actually decrease the concentration of microplastics in the dust samples, but the increment was found to be insignificant ($p > 0.05$) with high statistical power (power = 0.84)WWTP sites are considered a significant source of microplastics in the aquatic environment, due to the high volumes of effluent contaminated with urban microplastics such as those from commercial products, personal care products, laundry, and tires (Carr et al., 2016; Koutnik et al., 2021). WWTP utilizes treatment processes such as ultrafiltration, coagulation, and membrane bioreactors that result in microplastic removal up to 50- 99%. However, microplastics between 20–300 µm in size persist with concentrations around 0.25 n per L in WWTPs effluent (Murphy et al., 2016) and as high as 39020 n per kg in WWTP sludge (Koutnik et al., 2021). As nearly 129 billion gallons of wastewater are treated and released daily, often contaminated with other pollutants such as

antibiotic, PFAS, and heavy metals which can adsorb onto microplastics, this presents a significant problem. These effluent waters make their way to our oceans, where recent studies suggest that microplastics are emitted from ocean surfaces to the atmosphere due to wind and wave-driven sea-spray aerosolization (Allen et al., 2020; Goßmann et al., 2023), responsible for global annual ocean-to-land emission of more than 0.8 million metric tons of microplastics (Brahney et al., 2021; Evangeliou et al., 2022). Additionally, wind can also pick up microplastics from recycled wastewater sludges, where removed microplastics are concentrated, which are reapplied to agricultural fields and landscapes for fertilization (Leonard et al., 2024). The lack of correlation between microplastics in the deposited dust and proximity of the sampling location to WWTP suggests that microplastic dispersion from WWTP may not be confined to the surrounding area, but could be equally contaminating a wider region of influence due to the circular recycling of effluent discharges and sludges. These findings underscore the complexity of microplastic distribution mechanisms and highlight the disconnect between land use factors and the distribution of microplastics.

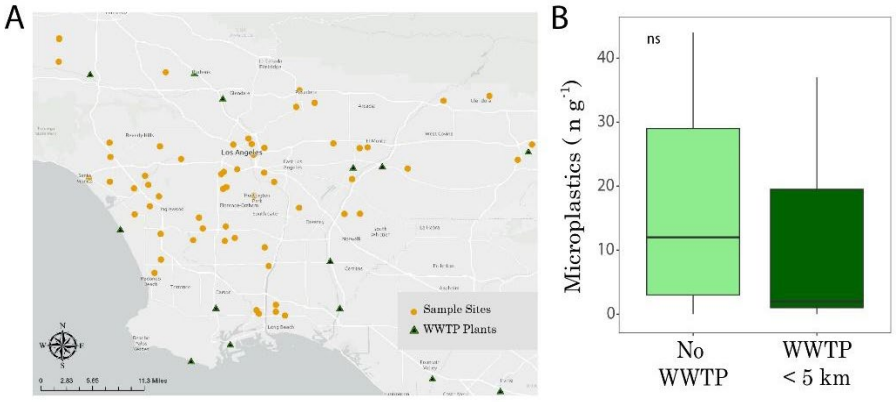


Figure 5-7: (A) Map showing the spatial location of sampling sites (orange circles) overlayed with triangles representing WWTP sites within the region of interest (B) Microplastic concentrations in deposited atmospheric dusts (n g^{-1}) do not vary across sampling sites which are either proximal or non-proximal to WWTP sites.

5.3.6. Industry and microplastic concentrations

Microplastic concentrations did not correlate with proximity to manufacturing or industrial sites within a 2 km radius (**Figure 5-8**). The prevailing assumption is that manufacturing activities directly influence particulate pollution- such as microplastics- due to exhaust emissions, improper disposal of packaging or waste, or even from the industrial processes themselves (Li et al., 2016; Shi et al., 2020; Wang et al., 2019). There was a difference in the statistics for concentrations within 2 km of an industrial site (mean = 11.22 n g^{-1}, median = 7 n g^{-1}) and not near one (mean = 16.44 n g^{-1}, median = 11 n g^{-1}). However, statistical analysis revealed no significant disparities in microplastic counts (n g^{-1}) between sampling points located inside and outside the designated 2 km boundary encompassing manufacturing zones, although this conclusion is limited by the low statistical power of the study (power = 0.34). This finding suggests that manufacturing or industry may not increase the concentration of microplastics in dust around the sites potentially due to improved methods to manage emission and dispersion of any accidental emission of microplastics. Manufacturing industries have implemented prevention strategies to reduce plastic packaging, improve production efficiency, analyze plastic product substitution, and improve disposal of waste by recycling and recovering plastic waste (Huang et al., 2020; Kibria et al., 2023; Interstate Technology Regulatory Council, n.d.). Furthermore, filtration can be used to remove microplastic from industrial exhaust with a) portable air cleaners or b) improved air filters in heating, ventilation, and air-conditioning (HVAC) systems (Interstate Technology Regulatory Council, n.d.; US EPA, 2014). Textile industries have reduced textile washing frequency, improved lint removal by dryers, or used non-vented condenser dryers to reduce fiber release from textiles to air (Moran et al., 2021). As education and public awareness of the risk of manufacturing and industry on atmospheric plastic contamination grow, so do efforts to combat this source of microplastics.

This result prompts a reevaluation of our understanding of microplastic distribution in urban areas and reinforces the ubiquitous contamination of microplastics in the atmosphere regardless of socioeconomic status, particulate pollution burden, or proximity to waste or industry.

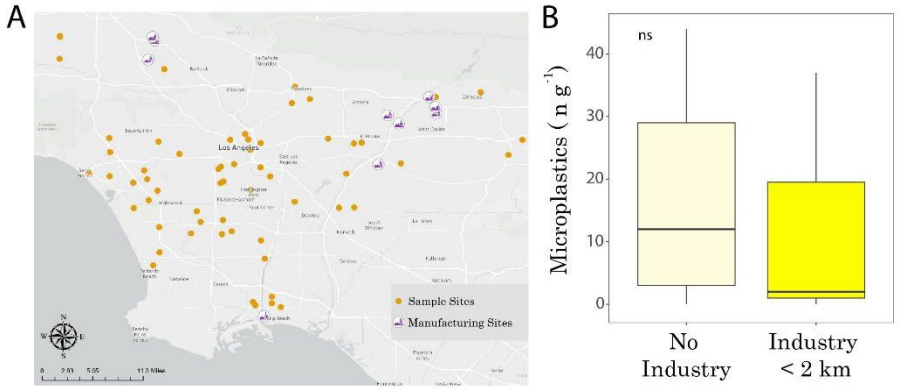


Figure 5-8: (A) Map showing the spatial location of sampling sites (orange circles) overlayed with purple sites representing manufacturing sites within the region of interest (B) Microplastic concentrations in deposited atmospheric dusts (n g^{-1}) do not vary across sampling sites which are either proximal or non-proximal to industry sites.

5.4. Conclusion

This study aims to answer whether microplastic concentrations in deposited dust in urban regions have a relationship with land use and socioeconomic factors of the regions, consistent with environmental justice literature that has shown that these factors are associated with higher levels of particulate matter in the air. I collected deposited dust samples from schools and CARB sites in Los Angeles and analyzed microplastic concentration in the dust. Despite extensive statistical testing (t-tests, Fisher exact tests), no statistical correlation was established between microplastic concentrations and social factors (i.e., poverty) or land use factors, such as proximity to known sources of microplastics such as landfills, wastewater treatment plants, and industry. Only poverty exhibited a significant relationship with microplastic concentrations over 30 n g^{-1}, albeit weak and not linearly proportional (**Figure 5-9**). These findings are limited by the relatively lower statistical power, as revealed by post-hoc testing. Median statistics for variables did show notable

differences, implying a need for larger-scale projects in the future. Nevertheless, these findings suggest these factors alone may not describe microplastic exposure risk near schools and that the spatial distribution of deposited microplastics does not correspond to variations in neighborhood socioeconomic status. This finding is in contrast with observations that communities living near pollutant sources are disproportionately exposed to air pollution. I attributed the results to the dispersion of suspended microplastics evenly throughout the regions by wind, thereby diluting any effect of local sources. Future studies should directly quantify concentrations in active or passive air samplers via long-term monitoring in these regions to examine if these conclusions hold for larger sample sizes. This novel study concludes that regardless of socioeconomic status and irrespective of where people live, they are all exposed to similar concentration of microplastics in the air. Thus, identifying significant determinants of differential exposures to deposited microplastics may be challenging given that they are ubiquitous and abundant.

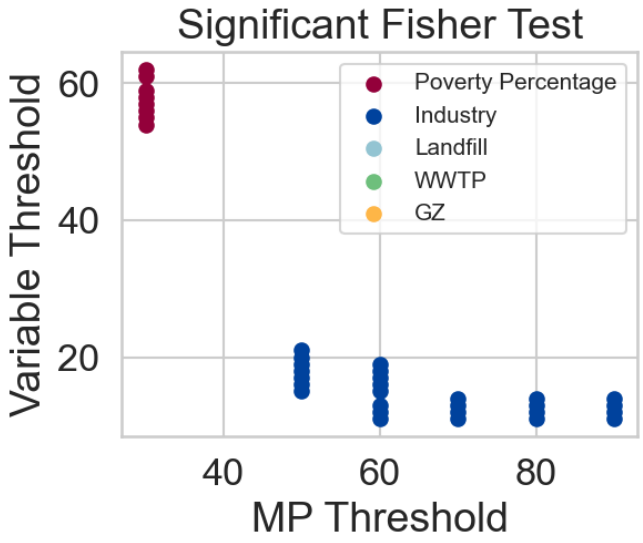


Figure 5-9: Scatter plot showing the Fisher test relationships between microplastic concentration threshold (0-90 n g-1) and the variable of interests: poverty percentage and proximity to industry, landfills, wastewater treatment plants (WWTP), and green zone districts (GZ).

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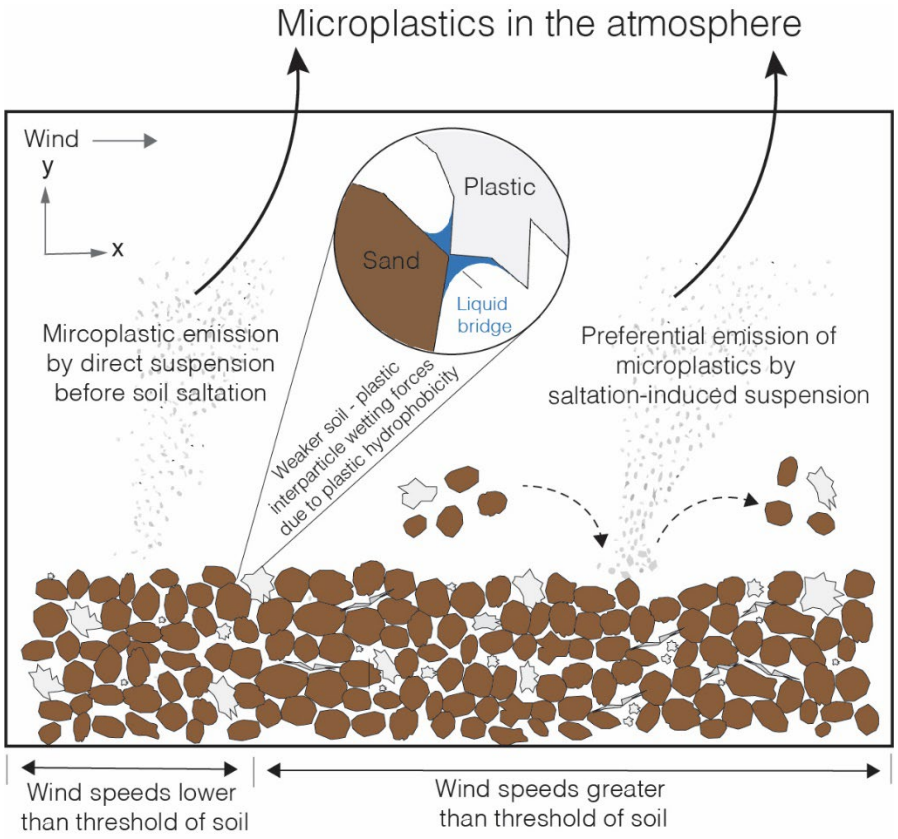
6. CHAPTER 6 – PREFERENTIAL EMISSION OF MICROPLASTICS FROM BIOSOLID-APPLIED AGRICULTURAL SOILS: FIELD EVIDENCE AND THEORETICAL FRAMEWORK



Leonard, J., Ravi, S., Mohanty, S.K. (2024) Preferential emission of microplastics from biosolid-applied agricultural soils: Field evidence and theoretical framework. EST Letters. https://doi.org/10.1021/acs.estlett.3c00850

Abstract

Land application of wastewater biosolids on agricultural soils is suggested as a sustainable pathway to support the circular economy, but this practice often enriches microplastics and associated contaminants in topsoil. Wind could transport these fine contaminated microplastics, thereby increasing their inhalation health risks. Analyzing wind-borne sediments collected from wind tunnel experiments on biosolid-applied agricultural fields, I prove an enrichment factor of 1.35 for microplastics in wind-blown sediments. I explain this preferential transport of microplastics from soil using a theoretical framework. This framework demonstrates how the combined effects of the low density of microplastics and weakened wet-bonding interparticle forces between microplastics and soil particles lower their fluid threshold – the minimum velocity necessary for wind erosion to occur. The results indicate that microplastics could be emitted at wind speeds lower than the characteristic fluid threshold of soil. Analyzing the windspeed distribution for 3 months of wind events over a bare soil surface, I showed that more than 84% of the wind events exceed the fluid threshold of microplastics, while only 23% of the wind events exceed the threshold velocity of background soils. Thus, current models for fugitive dust emissions may severely underestimate the microplastic emission potential of biosolid-amended soils.

6.1. Introduction

The application of wastewater biosolids on agricultural lands contributes to the circular economy by utilizing waste while reducing greenhouse gas emissions related to fertilizer use (Bogner, n.d.; Sharma et al., 2020). The annual production of biosolids is estimated to be 100 million tones globally, with a projected annual increase of 175 million tons by 2050 (Wijesekara et al., 2017). While this practice provides several benefits (Logan & Harrison, 1995; Singh & Agrawal, 2008), it could potentially introduce microplastics and other persistent pollutants adsorbed on microplastics (Corradini et al., 2019; Rolsky et al., 2020; Weithmann et al., 2018b). Biosolids application in the US alone could introduce trillions (Koutnik et al., 2021b) of microplastics on agricultural fields, resulting in microplastic concentration in the topsoil as high as 10 mg kg^{-1} after just 5 applications (Corradini et al., 2019). In these biosolids, more than 90% of microplastics are undetectable due to their small size (< 10 μm) and difficulty in identification using current methods (Koutnik et al., 2021b). Smaller microplastics have a higher potential to carry a wide range of pollutants due to their high surface area (Abbasi et al., 2020; Maguire & Gardner, 2023; Song et al., 2020; Ziccardi et al., 2016), and therefore pose a greater inhalation risk to humans (Amato-Lourenço et al., 2021a; Borthakur et al., 2021a). The inhalation risk is higher in arid and semi-arid agricultural lands because of increased wind erosion in the regions, coupled with the increase in biosolid application to improve soil quality. Wind-driven erosion is expected to accelerate because of recent increases in aridity, recurrent droughts, and seasonal disturbances (Ravi et al., 2011).

While many studies have measured microplastics in agricultural soils (Corradini et al., 2019; Crossman et al., 2020b; Kumar et al., 2020; Tran et al., 2023), limited field studies have examined the wind transport of microplastics from biosolid-applied agricultural lands (Bullard et

al., 2021b; Pi et al., 2018a; Rezaei et al., 2022a), and far fewer studies examine the conditions that affect the emission potential of microplastics using wind tunnel experiments in agricultural fields (Esders et al., 2022; Rezaei et al., 2019; M. Yang et al., 2022). Some of these studies added microplastics created in the laboratory at a concentration much higher than that expected in agricultural fields, thereby affecting interactions between soil grains and microplastics particles (Bullard et al., 2021b). Additionally, microplastics created in the laboratory by abrading plastic materials could differ significantly in shape, size, and surface properties compared to microplastics present in biosolids. Thus, previous conclusions about the enrichment of microplastics in agricultural dust emission could be inaccurate since they do not model environmentally relevant conditions.

The mechanisms of why microplastics may be enriched in wind-blown sediment are unclear. The physics of particle emission by wind is complex, as it involves atmospheric, soil, and land surface processes (Shao & Lu, 2000). Erosion by wind occurs when the shear stress exerted by the wind on the ground surface exceeds the shear strength of the aggregates and their resistance to detachment. The minimum wind shear stress required to cause erosion, commonly known as the fluid threshold, depends on several factors including particle characteristics, size and stability of the soil aggregates, field surface conditions, vegetation cover, and near-surface soil moisture (Kok et al., 2012; Ravi et al., 2011). At wind speeds beyond the fluid threshold, saltation-sized particles (70 - 500 μm) are entrained and carried by wind short distances away as a horizontal flux within the lowest 1 meter of the atmosphere (Bagnold, 1974). The saltating particles collide with other particles or aggregates on the surface and generate fine particles, which can also be resuspended (Gillette & Passi, 1988; Gillette & Walker, 1977; Ravi et al., 2020, p. 2). Most airborne sediments load to the atmosphere during the few events when wind speed exceeds the characteristic fluid

threshold necessary for soil saltation to occur (Bagnold, 1974). Thus, events when the wind speed is below this threshold are rarely considered while estimating the mass transfer of particles from the land surface to the atmosphere, though they could be critical for microplastic emissions. Compared to soil particles, microplastics could be preferentially resuspended by wind due to their low density and hydrophobic surface characteristics (Dike et al., 2023; Koutnik et al., 2021). While low density reduces the downward force exerted on the particle by gravity, higher hydrophobicity can lower the interparticle wet bonding forces (adsorption and liquid bridge bonding) that typically act as a glue against the liftoff force provided by the wind (Rao, 2004). Thus, a combination of both factors could make microplastics more likely to be entrained by wind even at lower fluid thresholds. The objectives of this study are to estimate the enrichment of microplastics in wind-blown sediments on agricultural soils with historic biosolid application and to identify the cause of this enrichment of microplastics using a theoretical framework.

6.2. Methods and Materials

6.2.1. Wind-blown sediment collection during a wind-tunnel experiment:

Soil and windborne sediment samples were collected from plots (~6 tons ha^{-1}) in Lind, Washington (47°00′N, 118°34′W) during a wind-tunnel experiment as described in a previous study (**Figure 6-1**) (Pi et al., 2018a). Briefly, a portable wind tunnel (Pietersma et al., 1996) was set up over two identical biosolid-applied and traditionally tilled (disk tilled method) plots within the same agricultural field, and a wind speed of 16 m s^{-1} was sustained over 10 minutes to collect around 5-10 g of wind-blown sediments using a vertically integrating isokinetic slot sampler (Pietersma et al., 1996). These wind-blown sediment samples were taken as representations of active saltation conditions that normally occur in the field during high wind events (Pi et al., 2018a, 2018b). Applied biosolid samples (n = 4) were taken from the supply stockpile, as a reference for

expected contamination levels since they are a source of microplastics in soil. To estimate the enrichment of microplastics in suspended sediments by wind, soil (n = 12) and wind-blown sediment samples (n = 8) were collected and analyzed for microplastics.

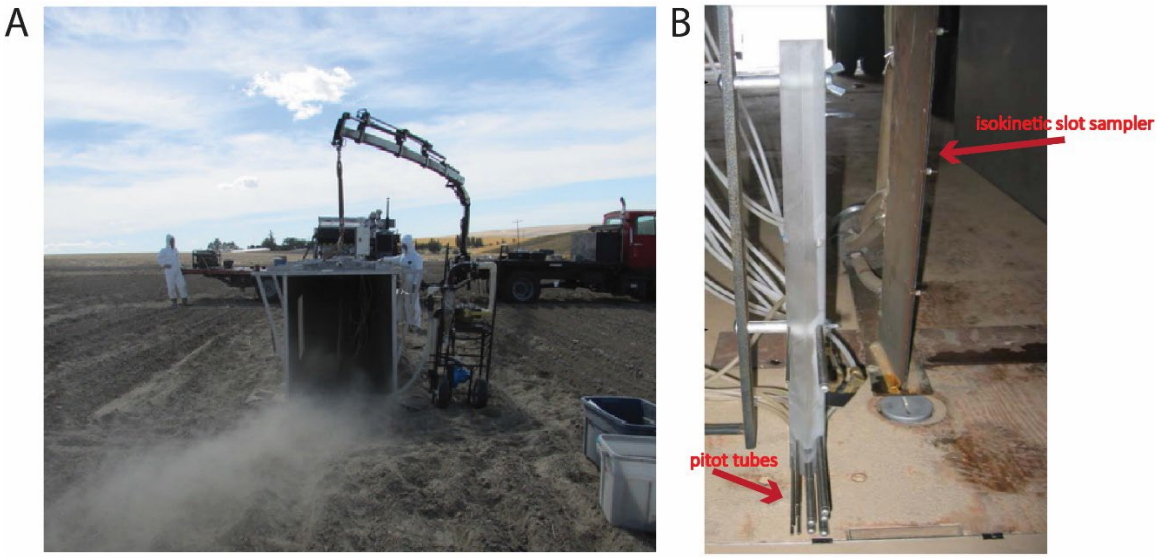


Figure 6-1: (A) Outside view of the wind tunnel used to collect windblown dust (0-75 cm height) (B) Inside view of the wind tunnel, with visual view of the apparatuses, and labeled pitot tubes and isokinetic slot sampler. Images are modified from a previous research publication.

6.2.2. Microplastic extraction and analysis:

Microplastics were isolated from all samples using density separation to remove denser soil particles followed by the digestion of natural organic matter as described in detail in previous sections (**Figure 6-2**). Briefly, 1 g of dry solid sample was mixed thoroughly with 40 mL of 10 M KI solution (density 1.7 g cm-3) so that microplastics and organic debris with a density lower than 1.7 g cm-3 would float. This mixture was centrifuged, vacuum filtered, digested using Fenton's reagent (2:1 ratio of 30% H2O2 and iron sulfate)(R. R. Hurley et al., 2018; Tagg et al., 2017), and acidified. A second round of density stratification and vacuum filtration followed. Microplastics isolated after the digestion of organics were counted manually using a compound microscope. Additionally, a subset of wind-blown sediment samples was characterized using Fourier-transform

infrared spectroscopy (FTIR, Thermo Scientific NicoletTM iN10) in reflectance mode, using a minimum 60% match criteria across all available commercial libraries to identify the microplastics' compositions and sizes.

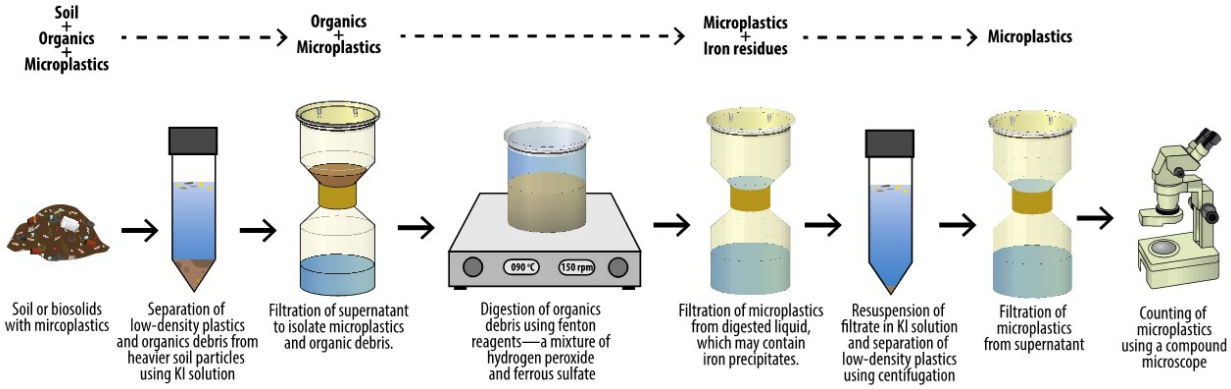


Figure 6-2: Methods for isolation of microplastics from biosolid, soil, and wind-blown sediment samples.

Plastic cross-contamination was eliminated during sampling processing steps by using non-plastic tools and containers wherever possible. All glassware, containers, and filtration devices were rinsed with deionized water three times before use. A Tukey one-way test ($p < 0.05$ indicates significance) was used to compare microplastic concentrations among three sample types: wind-blown sediment, soil, and biosolids.

6.2.3. Theoretical framework for emission of microplastics:

Under the influence of wind, a soil grain at the surface experiences several forces: aerodynamic drag, aerodynamic lift, and stabilizing forces like gravity and interparticle cohesive forces (Shao et al., 1993). The resulting force balance is used to estimate the fluid threshold (Shao & Lu, 2000). Wind speed controls the erosive action of wind, while field surface conditions, soil texture, size and shape of the aggregates, as well as near surface soil water content affect the threshold velocity. Several theoretical and empirical equations have been suggested in the past to express the threshold friction velocity as a function of these factors (Shao et al., 1993). Here, I

adopt a semi-empirical expression (**Equation 6-1**) for the saltation fluid threshold, developed by Shao & Lu (Shao & Lu, 2000).

$$u_* = A_N \sqrt{\frac{\rho_p - \rho_a}{\rho_a} g D_p + \frac{\gamma}{\rho_a D_p}} \quad (6\text{-}1)$$

where the ρ_a is the air density, ρ_p is the microplastic density, D_p is the microplastic diameter, g is the acceleration due to gravity, $A_N = 0.111$ is a dimensionless parameter (Kok et al., 2012), and $\gamma = 2.9 \times 10{-}4$ N m^{-1} is a parameter that scales the strength of the interparticle forces (Kok & Renno, 2006) (**Table 6-1**).

Table 6-1: Assumed Constants for Equation 6-1.

Constant	*Value*	*Units*
A_N	0.111	
g	9.8	m s^{-2}
γ	2.9×10^{-4}	N m^{-1}
$\rho_{p,\,PET}$	1397	kg/m^3
$\rho_{p,\,PE}$	930	kg/m^3
$\rho_{p,\,PS}$	1005	kg/m^3
$\rho_{p,\,PP}$	910	kg/m^3
$\rho_{p,\,sand}$	2650	kg/m^3
ρ_a	1.293	kg/m^3

The interparticle forces in soil include electrostatic forces, van der Waals forces, and capillary forces due to the presence of a liquid bridge between the particle and soil surface (Cornelis et al., 2004; Mckenna Neuman, 2003; Ravi et al., 2004). Among all forces, the capillary force exerted by the liquid bridge can be orders of magnitude higher than electrostatic and van der Waal forces. This adhesive force exerted by the liquid bridge depends on the contact angle, which in turn depends on the hydrophilic or hydrophobic nature of both surfaces (Jones et al., 2002). Thus, the adhesive force from the liquid bridge can be calculated using **Equation 6-2** (R. A. Fisher, 1926; Ravi, D'Odorico, et al., 2006).

$$F_i = 2\pi b T sin\,(\theta + \gamma) + \pi b^2 T \left(\frac{1}{c} - \frac{1}{b}\right) \quad (6\text{-}2)$$

where T = 0.07275 J m^{-2} is the surface tension of water, γ is the contact angle, θ = 40º depends both on moisture content (12.5%, expressed as percentage of pore space) and on soil packing (R. A. Fisher, 1926) (open packing of spherical particles), b is the radius if the fluid neck connecting two spherical grains, c is the radius of the meridian curve, and $\left(\frac{1}{c}-\frac{1}{b}\right)$ represents the total curvature of the surface at this point between the pore air and the water in the liquid bridge (**Table 6-2**).

Table 6-2: Assumed Constants for Equation 2.

Constant	*Value*	*Units*
c	Calculated from **Equation 6-3**	
b	Calculated from **Equation 6-4**	
T	0.07275	J m^{-2}
γ_{PET}	72.5	degrees
γ_{PE}	96	degrees
γ_{PS}	87.4	degrees
γ_{PP}	102	degrees
γ_{sand}	35	degrees
θ	40	degrees

For Equation 6-2, the following equations (R. A. Fisher, 1926) were used to calculate c and b, using the assumption that r = 0.000725 meters:

$$c = \frac{r\,(1-\cos\theta)}{\cos(\theta+\gamma)} \tag{6-3}$$

$$b = \mathrm{r}\sin\theta - c\left[1-\cos\left(\frac{\pi}{2}-(\theta+\gamma)\right)\right] \tag{6-4}$$

6.2.4. Calculation of the exceedance of the fluid threshold in an agricultural field:

To assess the frequency of microplastic emissions at typical wind speeds in an arid agricultural field in the US, I plotted a Weibull distribution of shear wind velocity records during a three-month windy season taken from Burger et al., 2023. Briefly, wind speed records were

collected between March 11, 2016 - June 30, 2016, from a 5 × 5 m bare soil plot at Sevilleta National Wildlife Refuge (N 34°23.961′ and W 106°55.710′). A 4-m high solar-powered meteorological tower empirically recorded average and max wind velocities at 4, 2, 1, and 0.5 m above ground every second and averaged the reading over 1-minute intervals (Burger et al., 2023). The average wind velocities at all 4 heights were used to fit the wind profile to the Prandtl–von Karman logarithmic law (Burger et al., 2023) (**Equation 6-5**).

$$u(z) = \frac{u_*}{0.4} ln \left(\frac{z-d}{z_0}\right) \quad (6\text{-}5)$$

where u(z) is wind velocity at height z, and $u_*$is the shear velocity (or friction velocity), d is the zero-plane displacement, z_0 is the aerodynamic roughness length, and 0.4 is the Von Karman constant (Campbell & Norman, 2000). The displacement height was assumed to be ~ 0 for the smooth field with no vegetation. The slope and y-intercept of the best-fit line relating ln(z) with u(z) in **Figure 6-3** were used to calculate z_0 = 0.049 and u_* (Campbell & Norman, 2000).

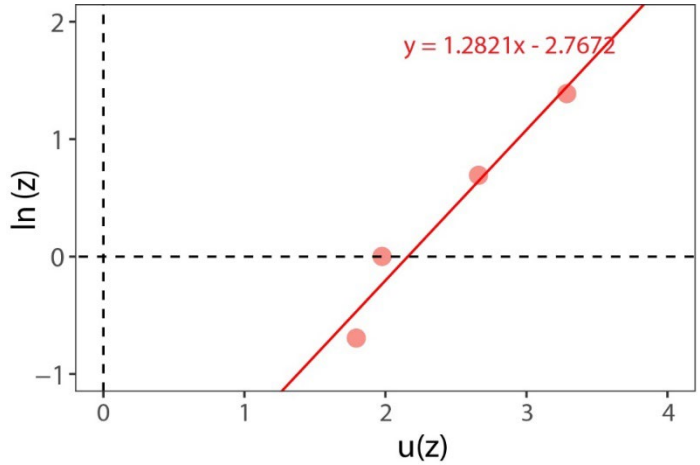


Figure 6-3: A plot of ln (z), where z is the height of a recorded wind velocity, versus the wind velocity, *u(z)*. The linear trendline has a slope of 1.2821 and y-intercept of -2.7672.

This fitted wind profile was used to convert recorded maximum wind velocities at 4 m, u(z = 4), into maximum wind speed at 2 m, u(z = 2), for each minute. I then generated a Weibull wind distribution to calculate the probability of windspeed exceeding any specific value. Using the

theoretical fluid threshold (**Equation 6-1**) for microplastics and experimentally measured fluid threshold for soil particulates at this site (Dukes et al., 2018), this distribution of wind speeds was used to estimate the number of one-minute wind events that exceed the fluid threshold of typical microplastics (PP). The fluid threshold for soil particles (Dukes et al., 2018) and microplastics (**Equation 6-1**) was converted into wind speeds at 2 m using **Equation 6-5** (**Table 6-3**), and the probability of windspeeds exceeding these critical wind speeds was calculated from the generated Weibull distribution. Here I represented microplastics as PP, the lowest density commonly used plastic with size *Dp*, 150 µm, which is the critical size range for emission estimated in my study.

Table 6-3: Fluid threshold (u*) values and conversion to wind speeds at 2 m (u(2))

Sample Type	$u_* (m\ s^{-1})$	*Source*	$u(2)\ (m\ s^{-1})$	*Source*
Soil	0.63	Experimentally derived at this site	5.841433	Calculated using Equation 6-5
Microplastic (PP)	0.176498	Theoretical Prediction from Equation 1	1.636511	Calculated using Equation 6-5

6.3. Results and Discussion

6.3.1. Enrichment of microplastics in wind-blown sediments:

The wind-blown sediments collected from the wind tunnel experiments contained more microplastics than either the source soils or the biosolids that released microplastics into the soil (**Figure 6-4A**). The mean microplastic concentrations in soil, biosolids, and wind-blown sediments were 635 n g^{-1}, 766 n g^{-1}, and 859 n g^{-1}, respectively. I measured microplastic concentrations in wind-blown sediments to be 78-12560 n kg^{-1}, which is comparable to concentration ranges reported in previous studies (Y. Chen et al., 2020; M. Liu et al., 2018). Furthermore, the result indicates microplastics were enriched in the wind-blown sediments by a factor of 1.35 when compared to traditional soil minerals such as quartz, aluminosilicate clays, and metal oxides.

Analyzing the size distribution of microplastics, I estimated the dominant microplastic size in the wind-blown sediments was > 100 µm (**Figure 6-4B**). This is consistent with the critical size of emission determined theoretically using **Equation 6-1** (**Figure 6-6A**). This critical size is where microplastics require a minimum fluid threshold and thus are most likely to be resuspended. In this study, FTIR was used to measure characteristic properties of microplastics (**Figure 6-5**) but it cannot positively confirm (> 60%) the composition of smaller particles due to a detection limit of 20 µm (Centrone, 2015). Thus, smaller microplastic fractions could be underestimated in this study. The dominant size range of microplastics in dust and wind-blown sediment samples varied in comparable studies (**Figure 6-4C**). However, the size reported in these studies could be affected by the microplastic quantification protocol (such as minimum detection limit or resolution of microscopy techniques) and collection methodology. Much of the data in previous studies were not from controlled wind tunnel experiments. Thus, future studies should conduct more comprehensive wind tunnel and field experiments to measure particle size distribution of emitted microplastics to confirm the preferentially resuspended particle size.

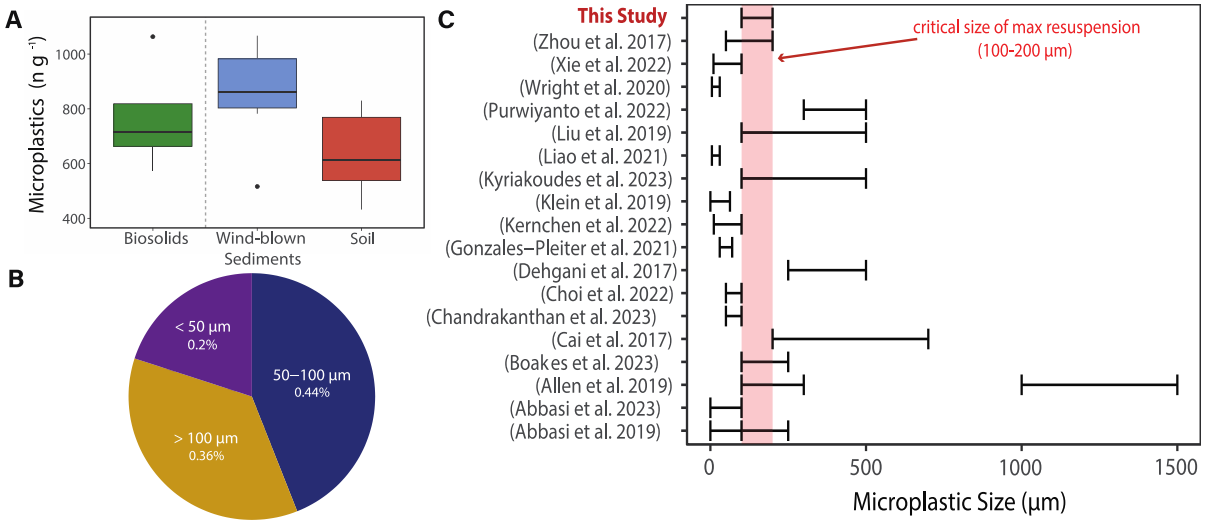


Figure 6-4: (A) Microplastic concentrations (pieces, n, per gram of solid) for each type of sample processed: biosolids, wind-blown sediments, and soil. Microplastic concentrations in the wind-blown sediments were significantly higher ($p < 0.05$) than those in the soil from where they are emitted. (B) Size distribution of

microplastics from wind-blown sediment samples based on the longest side of 35 identified microplastic pieces. (C) Dominant size ranges from 18 global studies (Abbasi et al., 2019, 2023; Allen et al., 2019; Boakes et al., 2023; Cai et al., 2017b; Chandrakanthan et al., 2023; Choi et al., n.d.; Dehghani et al., 2017b; González-Pleiter et al., 2021; Kernchen et al., 2022; S. Klein et al., 2015; Kyriakoudes & Turner, 2023; Liao et al., 2021; K. Liu et al., 2020b; Purwiyanto et al., 2020; Wright et al., 2020; Xie et al., 2022; Zhou et al., 2020) analyzing microplastics in wind-blown sediments, where the highlighted region is the theoretical estimate of the critical size for maximum emission.

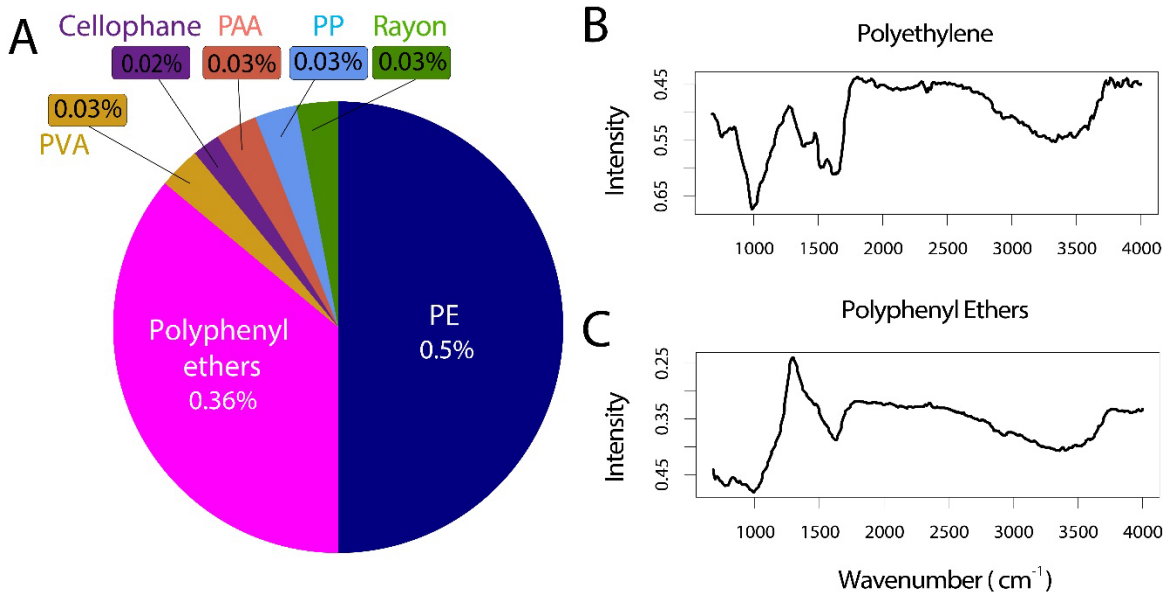


Figure 6-5: (A) Distribution of the plastic polymer compositions of microplastics identified using FTIR (B) FTIR spectra for polyethylene (PE), representing 50% of identified microplastics, and (C) FTIR spectra for polyphenyl ethers, representing 36% of identified microplastics.
*PVA: polyvinyl alcohols; PAA: polyacrylic acids; PP: polypropylene

6.3.2. Enhanced suspension of microplastic below threshold windspeed

Using a semi-empirical expression for the fluid threshold (**Equation 6-1**), I show that microplastics could be eroded at a lower fluid threshold than quartz sand, the most common mineral found in soil (Campbell & Norman, 2000) (**Figure 6-6A**). The fluid threshold depends on particle size: decreasing particle size until the critical size range (~100-200 μm) decreases the fluid threshold due to a decrease in interparticle force. Conversely, any further increase in particle size beyond the critical size range increases the fluid threshold due to the increasing effect of density and the resulting dominance of gravity. Thus, the effect of particle density becomes more apparent when the particles are bigger than 75 μm (Shao & Lu, 2000). The result indicates that for the

emission of small-sized microplastics (< 10 μm)—the relevant size for inhalation risk—interparticle forces could play a greater role than the density of plastic particles. For these small microplastics, a larger wind velocity is required to overcome the interparticle forces which become increasingly important as the particle size becomes smaller.

The surface adsorption of moisture is generally limited on microplastics, which are generally hydrophobic (Shao & Lu, 2000). Thus, the microplastic particles might be expected to retain less adsorbed water and to be consequently lighter and with a lower threshold velocity. When soil moisture (or humidity) increases in pore spaces, condensation starts to occur in the contact points between the particles, adsorbing onto the grain surface and thereby forming a "liquid bridge", which increases the interparticle forces between particles. The hydrophobic surfaces of microplastics can delay the formation of liquid bridges, or even prevent their formation in cases of extremely hydrophobic microplastics. Consequently, the interparticle forces associated with the liquid bridges between microplastics and soil particles (Wei & Zhao, 2007) are expected to be less adhesive than the bridges between soil particles due to increasing degrees of hydrophobicity (i.e. increasing contact angle (Chen et al., 2013)) (**Figure 6-6B**).The high hydrophobicity of plastic surfaces increases their contact angle, which results in a net repulsive interparticle force (**Figure 6-6B**). In contrast, a smaller contact angle on quartz particles allows the liquid bridge to act as a glue for quartz particles, resulting in a net attractive force. Thus, hydrophobic microplastics have weaker interparticle forces, or more repulsive force, than that experienced by quartz or other soil minerals with lower hydrophobicity. I conclude that a reduction in both adsorbed and liquid bridge bonding significantly reduces the interparticle forces associated with moisture bonding and makes microplastics more susceptible to erosion by wind than soil particles of the same size (Ravi et al., 2006).

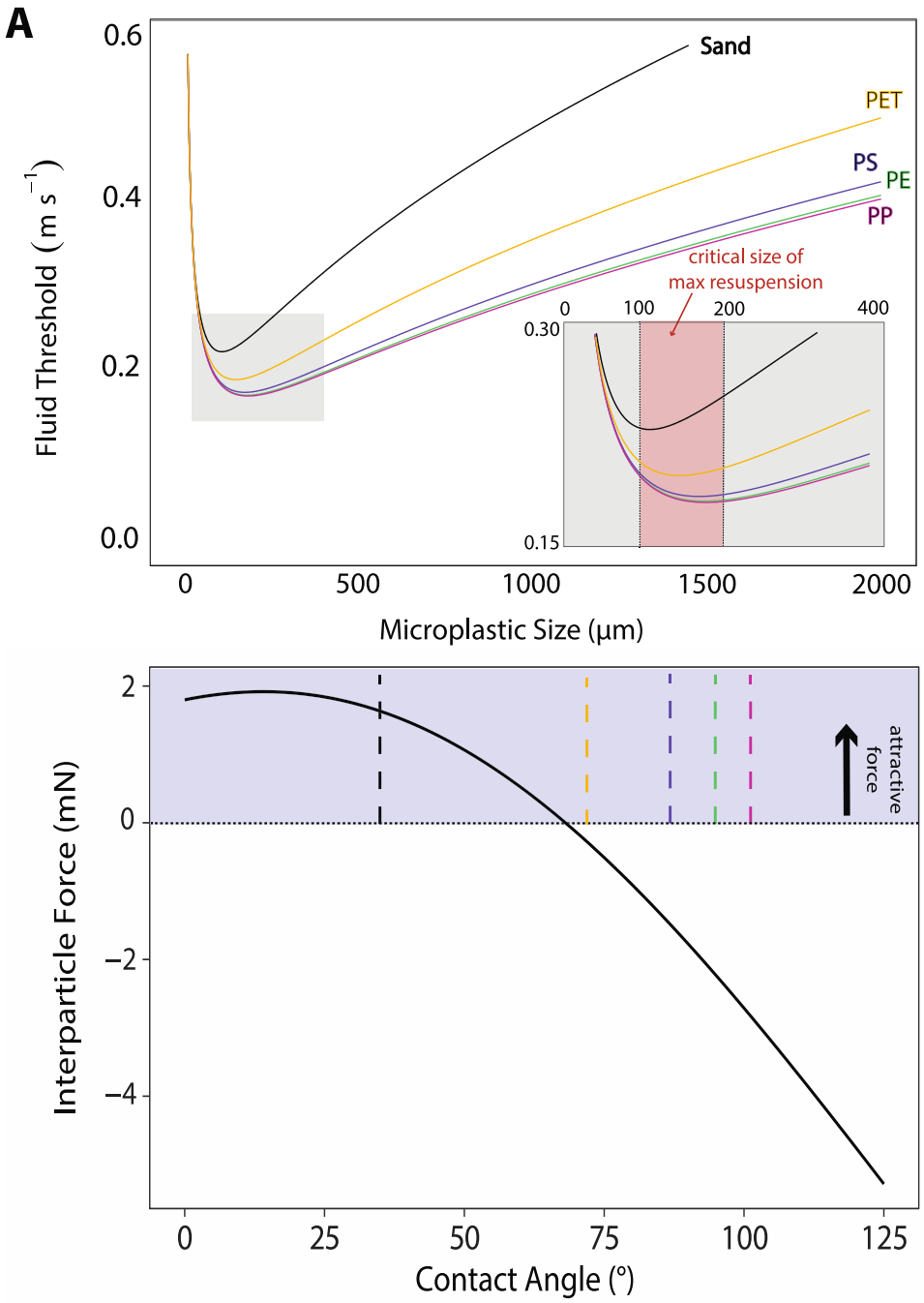


Figure 6-6: (A) Fluid threshold for initiating saltation (emission) for four types of microplastics with different sizes (0-2000 µm) and densities (0.91-1.005 g cm^{-1}). Within the region of interest in the inset map between 100-200 µm, microplastics require a minimum fluid threshold as visualized by the curves' minimums. (B) The interactive force from the water film on particles is calculated based on particle size and contact angle for an open packing system with 12.5 % moisture content.
*Abbreviations are defined as follows: PE: polyethylene; PET: polyethylene terephthalate; PS: polystyrene; PP: polypropylene.

6.4. Environmental Implications

Our analysis indicates that microplastics are more likely to be transported by wind below the characteristic fluid threshold of soil, due to their lower density and weaker liquid bridge bonding potential compared to traditional soil particles. To demonstrate the environmental implications of this current finding, I used a distribution for wind events at a typical arid region, where biosolids are typically applied to increase soil fertility. This analysis shows that microplastic's lower fluid threshold would result in a 269% increase in the number of events that would resuspend microplastics (**Figure 6-7A**). Based on a semi-empirical model to account for density effects, I estimated the fluid threshold for microplastic (PP) emission is around 0.176 m s^{-1}, resulting in a wind speed of 1.64 m s^{-1} at 2 m above the ground surface. This is much lower than an experimentally determined threshold velocity (5.84 m s^{-1}) of soil particles at this site (Dukes et al., 2018). The current models for fugitive dust emissions assume the soil surface to be stable with negligible dust emissions in the absence of wind speeds exceeding the fluid threshold of the soil or surface disturbances (EPA, 1997). With that assumption, current emission models may underestimate microplastic emission potential from agricultural soils—missing around 61% of wind events, which are too low to resuspend soil sediments but sufficient to initiate microplastic movement and drive their emission (**Figure 6-7B**) (Alfaro et al., 2022). Microplastic particles are readily emitted from soil surfaces under low wind speed scenarios, which are more frequent. The theoretical framework proposed in this study can be used to assess microplastic emissions more accurately by accounting for microplastic fluxes typically underrepresented. Thus, these emission events should be incorporated into fugitive dust emission models that inform environmental and human health risk assessments.

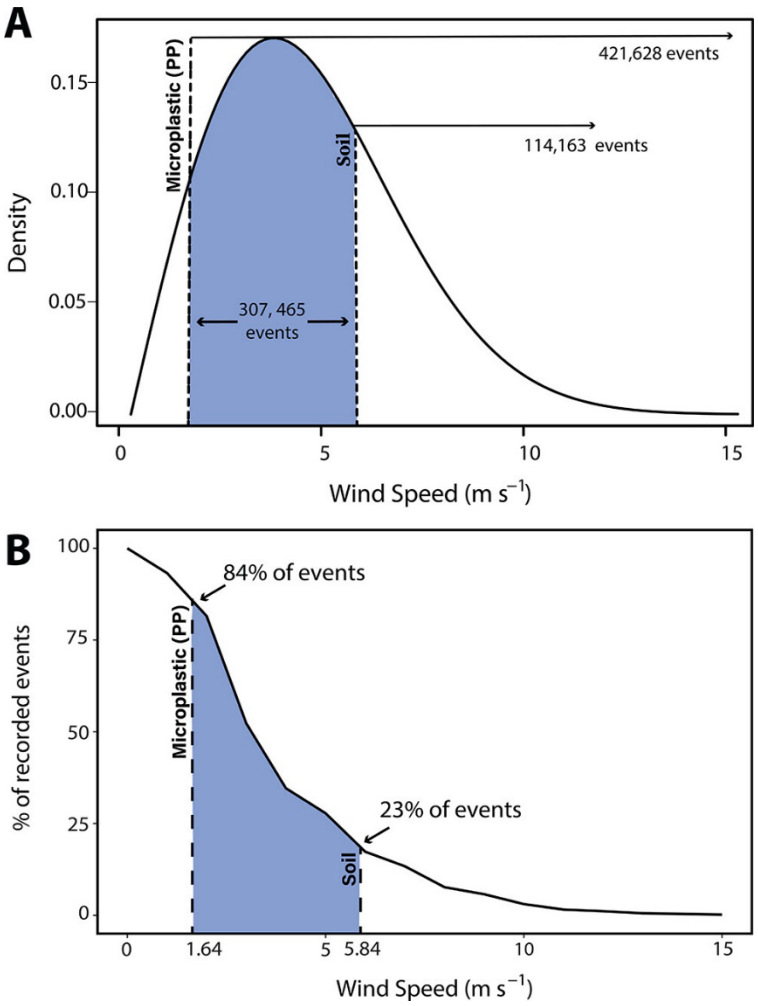


Figure 6-7: (A) Weibull distribution of wind speeds (at 2 m) for the windy season showing the threshold velocity range for microplastics (PP, 150 µm) and background soil at this site. (B) Percentage of wind events for each wind speed between 0-15 m s-1 for three months of wind data.

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7. CHAPTER 7 – UV WEATHERING AND HUMIDITY EFFECT ON THE ADHESIVE FORCE BETWEEN MICROPLASTICS AND SAND: IMPLICATIONS ON MICROPLASTIC EMISSION BY WIND

Leonard, J., Lake, M., Stieg, A., and Mohanty, S.K. UV weathering and humidity effect on the adhesive force between microplastics and sand: Implications on microplastic emission by wind. Environmental Science and Technology. In Preparation.

Abstract

Microplastics are emitted from terrestrial surfaces into the atmosphere when wind shear exceeds the net adhesive force between microplastics and soil surface. The adhesive forces between plastic and soil can be weakened or strengthened based on environmental conditions such as UV weathering and relative humidity (RH). Yet, the effect of these two, highly relevant conditions on the adhesion force between microplastics and soil has not been investigated. We use an atomic force microscope (AFM) to quantify adhesion forces between SiO2 AFM tips (a well-established proxy for the most common soil mineral) and polypropylene (PP) as a function of UV weathering time (0 to 30 min) and relative humidity (RH, 50 to 90%). Furthermore, we used a theoretical model to predict how these conditions can influence the adhesion force. Results showed that both these factors decreased the net adhesive force between PP and quartz tips, indicating an increase in the emission potential of the microplastics under weathered or humid conditions. Increase in UV weathering decreased surface hydrophobicity, as confirmed via contact angle measurement. A decrease in contact angle weakened the meniscus force between plastic and quartz under baseline humidity conditions (50% RH). Surprisingly, an increase in RH also decreased the meniscus forces between plastic and quartz. The reduced attractive forces between sand and plastic in both conditions can potentially increase the emission potential of microplastics into the atmosphere even in the humid environments. Thus, the results confirm that current fugitive dust emission models may be underestimating microplastic emission potential.

7.1. Introduction

Microplastics are present in extremely high, and ever increasing concentrations in all environmental compartments: terrestrial (Hurley et al., 2020; Machado et al., 2018), atmospheric (Beaurepaire et al., 2021b; Zhang et al., 2020), and marine (Ajith et al., n.d.; Hidalgo-Ruz et al., 2012). Humans are increasingly exposed to microplastics from these compartments through various pathways, including ingestion or inhalation. In fact, microplastics have been found in human bloodstream (Leslie et al., 2022b), lungs (Amato-Lourenço et al., 2021b), penis (Codrington et al., 2024), and placenta (Ragusa et al., 2021). Microplastics in the human body is associated with adverse health risks such as oxidative stress (C. Zhu et al., 2024), reduction in nutrient adsorption (Carlin et al., 2020; S. Chen et al., 2023; Lei et al., 2018; H. Tan et al., 2020), and most recently, an increased risk of cardiovascular disease (Marfella Raffaele et al., 2024). Inhalation is one of the primary exposure pathways for microplastics (Borthakur et al., 2021a; Chen et al., 2023; Gasperi et al., 2018; Geng et al., 2023). Inhalation exposure is controlled by two key factors: a) the quantity of microplastics capable of being emitted from terrestrial environments to the atmosphere, and b) the factors that increase emission potential, and thus exposure. These exposure factors include microplastic size and surface properties, wind conditions, atmospheric humidity, and soil conditions. Recently, microplastics were confirmed to have a higher emission potential that traditional soil particles and to be preferentially enriched in atmospheric dusts (Leonard et al., 2024). Thus, it is important to examine the conditions which could influence the adhesion force of microplastics on soil, which in turns governs their emission potential into the atmosphere.

The emission potential of microplastics is dependent on atmospheric conditions such as wind speed and humidity, and microplastic surface properties such as hydrophobicity and

roughness (Shao & Lu, 2000). Microplastic erosion by wind occurs when the shear force exerted by the wind on the ground exceeds the net adhesive force between plastic particle and soil. The adhesion force between two dry surfaces is made up of a variety of physical interactions: van der Waals, electrostatic and capillary forces, and hydrogen bonding. If there exists a surface water film resulting from atmospheric humidity, then the resulting capillary force is the most dominant force governing the interaction between plastic and soil surfaces. These hydration-induced interactions typically result in a net adhesive force, which can be measured by atomic Force Microscopy (AFM) (Harrison et al., 2015). To measure adhesion, AFM tips of various size, shape, and materials are lowered onto a surface until contact is established, and then retracted to quantify the force required to pull the tip from the sample surface. While a wide variety of hydrophobic and hydrophilic surfaces have been used in AFM studies, none have yet studied the adhesive force between sand (proxied by SiO_2) and plastic (PP) under varying humidity and UV exposure time. As adhesion force differs by particle size (Jones et al., 2002), shape (Sadao & Genji, 1967), and the chemical property of the surface (Fuji et al., 1999; Fukunishi & Mori, 2006), it is likely that plastic may experience difference microforces with sand when compared to other surfaces. Due to plastic's unique surface properties, the adhesive force between plastic and sand could vary significantly from previously studies of adhesion forces between traditional silicon AFM tips and surfaces like mica (Lai et al., 2022), silicon (Lai et al., 2024), silica (Lai et al., 2018), or aluminum (Ata, 2008). The adhesion force of plastic has been investigated very little, with studies only on polymers such as: polyethylene (Kim et al., 2016), polytetrafluoroethylene (Lai et al., 2024), and polycarbonate (Lai et al., 2024). However, no study has investigated polypropylene- one of the most abundant plastic polymers in the environment- nor investigated its microscale interaction under changing environmental conditions.

Environmental conditions such as relative humidity (RH) and UV weathering can affect adhesion forces. Adhesion force is known to be a function of the relative humidity (RH) (Ando, 2000; Christenson, 1988; L. R. Fisher & Israelachvili, 1981a; Yoon et al., 2003) because it is dominated by a wet bonding capillary force (ie. the meniscus force) caused by condensing water vapor in the gap between two surfaces. However, only two previous studies have looked at adhesion forces on plastics as a result of RH (Lai et al., 2018, 2024), and neither one used SiO_2 tips which are a better model for sand than the silicon tips used within. Thus, the relationship between microplastics and primarily sand terrestrial surfaces is yet unclear. Additionally, hydrophobicity- measured by contact angle- is a surface property that distinguishes plastics from other materials previously studied by AFM and could be influenced by weathering. Microplastics accumulated in the environment are continually exposed to UV radiation in sunlight, which could alter the surface chemistry of plastic polymers (Lin et al., 2020; Mao et al., 2020; Sun et al., 2020). UV radiation can cause oxidation of the plastic's surface, causing decreases in their hydrophobicity (Al Harraq & Bharti, 2021). However, no study to date has looked at the effect of UV weathering on the adhesion force between plastic and other substances.

This study investigates the effect of UV degradation and relative humidity on the adhesion forces between polypropylene (PP, plastic) and SiO_2 (sand) using AFM. To achieve the objectives, the effect of UV degradation on PP's hydrophobicity was verified using measurements of contact angle after exposure to specific time periods of UV (0, 1, 5, 15, 30 minutes). Then, AFM was used to quantify the changes in adhesion force between plastic and sand as a function of this UV exposure. Lastly, AFM was used to quantify the adhesive force between plastic-sand and silica-silica (control) as a function of relative humidity. The results inform the role that environmental

weathering and humidity on microplastic's adhesion force on the soil surface under environmental conditions and thus their resistance to emission.

7.2. Materials and Methods

7.2.1. Sample preparation

Transparent polypropylene (PP) plastic was cut into sample pieces (roughly 25 mm x 25mm) using metal scissors and glued in place to an AFM petri dish using epoxy resin. Since adhesion forces vary widely across a surface, partly due to its reliance on surface roughness, a 400 μm^2 subsection of the sample with limited scratches/blemishes was isolated using an optical microscope. For every set of measurements, AFM measurements were taken in this same isolated region to minimize the effect of sample surface heterogeneity. A spot on this region of interest was scored with a fine needle to serve as a visual reference point for AFM measurements (**Figure 7-1A**). To compare the interaction between quartz-quartz and quartz-plastic, a glass slide (silica) was used as a control sample. This sample was processed identically- cut into a 25mm x 25 mm planar piece and scored to create a visual marker.

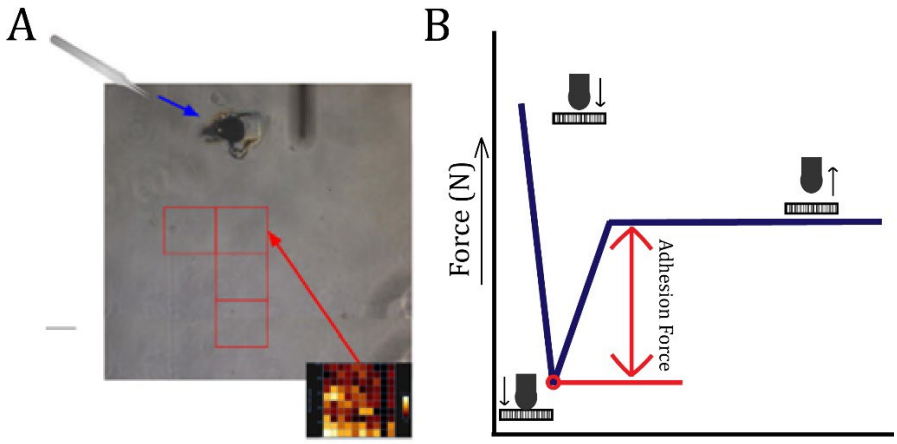

Figure 7-1: (A) Four 100 x 100 µm grids, each made up of hundred 1 µm2 force interactions were taken for each AFM measurement. The grids were consistently sampled a set distance away from a visual reference point created by scoring the sample with a needle tip to minimize the confounding effect of surface heterogeneity. An example image is shown above with the tweezer reference point, 4 grid outlines,

and an example AFM force map. (B) Schematic demonstrating typical adhesion force curve highlighting adhesion minima extracted for this study.

7.2.2. AFM and adhesion measurement

Adhesion forces were measured with a commercial AFM (NanoWizard® 4a NanoScience AFM) using pre-microfabricated tips with a 5 µm silica sphere attached to a tipless SiO_2 cantilever with a high spring constant designed for high force tapping measurements (AppNano ACT-SiO_2-A-5). The cantilever was 125 µm x 30 µm and the tip had a radius of 5 µm. The spring constant was calibrated at 22.71 N/m. The calibrated cantilever was used to collect force–displacement curves between quartz tip and PP samples under baseline humidity and temperature of 50 ± 1% RH and 24 ± 1°C, respectively. To check the repeatability and reproducibility of adhesion force, data points were collected in four replicated 100 x 100 µm grids, adding up to 400 force curves per measurement (**Figure 7-1A**). For each of the 400 approaches, the AFM probe was brought close to the cell surface, indented into the sample surface, and then retracted. Standard parameters used were as follows: Z length of 15 µm (maximum length to address long-range effect), approach and retraction speeds of 2 µm s^{-1}; force limit of 3000 nN (30 N m^{-1} levers). Adhesion forces were determined by extracting the minimum of the retract force curve, as described in previous studies (Cross et al., 2008; Farshchi-Tabrizia et al., 2008; Jones et al., 2002). The minimum represents the retraction force required to separate the probe from the sample surface, referred to hereafter as the adhesion (**Figure 7-1B**). An in-house analysis program using JPK Data Processing Version 6.4.12 and R Studio 2023.06.2 Build 561was developed to automate preprocessing and analysis of the large dataset of force curves efficiently and calculate adhesion force.

7.2.3. Theory

The net adhesion force is composed of several surface forces: van der Waals force, electrostatic force, meniscus force, and the force due to chemical bonds (Butt et al., 2005; Leite et al., 2012). However, the adhesion force between two surfaces is dominated by an attractive meniscus force, and thus is the focus of this model. The strength of this force has been modelled in detail by a previous study (Farshchi-Tabrizia et al., 2008) and been shown to depend highly on AFM tip geometry among other factors. This study uses a relatively large (5 µm) spherical silicon dioxide AFM tip compared to typical nanometer scale, thus the geometry of the interaction is most similar to flat cylindrical tip models and visualized by a plane to plane schematic (**Figure 7-2**).

Figure 7-2: (A) General schematic of the AFM tip used in this study approaching a planar plastic surface (B) Zoomed into the nanometer scale, the micron sized spherical tip becomes more planar from the perspective of the plastic surface (C) Once you reach the 10 nm scale, the radius of the AFM tip used in this study, it is essentially a plane-to-plane interaction (D) Using a planar assumption due to the micron scale (relatively large) AFM tip used in this study, this interaction can be approximated by a standard flat cylinder tip-planar sample surface schematic where the meniscus force is a result of the blue shaded region (Farshchi-Tabrizia et al., 2008).

For this tip geometry, the meniscus force (F_m) can be estimated with **Equation 7-1** (Farshchi-Tabrizia et al., 2008):

$$F_m = 2\pi\gamma r_c \cos\theta - \pi {r_c}^2 \frac{R_G T}{V_m} \ln\frac{P}{P_0} \qquad (7\text{-}1)$$

where γ is the surface tension of water (0.072 N m^{-1}), r_c is the radius of the AFM tip (5 µm), θ is the known contact angle of water on the AFM tip and sample plane, assumed to be equal

(54º for silica, 104.9º for PP), R_G is the gas constant (287.05 J kg^{-1} K^{-1}), T is 298 K, V_m is the molar volume of water (18×10^{-6} m^3 mol^{-1}), and *P/P0* is relative humidity. As demonstrated by the equation, meniscus force is a function of contact angle (θ) and relative humidity (RH, *P/P0*). Contact angle is a measure of the hydrophobicity of a surface and is a surface property known to change due to environmental weathering, such as exposure to UV light. UV light exposure could also affect surface roughness. Relative humidity is known to affect adhesion forces. Yet the effect of humidity on interaction between microplastics and geological material such as quartz has not yet been studied. Thus, this model was adapted from Farshchi-Tabrizia et al. 2008 to better analyze the effect of UV weathering and humidity on the adhesion forces which prevent the emission of microplastics from terrestrial surfaces into the atmosphere.

7.2.4. Methods to investigate the influence of UV Weathering

Environmental weathering, simulated through UV radiation, can inversely affect the surface hydrophobicity and surface roughness of microplastics— both parameters could affect the adhesion force (Al Harraq & Bharti, 2021). To simulate the UV radiation of polypropylene (PP), the sample was weathered in stages in a UV chamber (Novascan PSD-UV Ozone System) emitting at both 185 nm and 254 nm adding up to a total of 1, 5, 15, and 30 minutes of UV weathering. After each UV exposure, adhesion forces and changes in contact angle were measured.

Contact angle measurements are used to indicate changes in the hydrophobicity of a material (Al Harraq & Bharti, 2021). To examine changes in surface hydrophilicity of the exposed plastic surface, the contact angle was measured using a Contact Angle Goniometer (Rame-Hart 500) after UV exposure for 0, 1, 5, 15, and 30 minutes. The contact angle was measured by placing a 3 µL water droplet onto the weathered surface of the plastic and

measuring the angle 10 times using DropImage Advanced software. This process was repeated for 2 droplets on each sample to determine the average contact angle per sample type.

7.2.5. Methods to investigate the influence of RH

We created an in-house system to adjust humidity within an AFM dish as a controlled humidity chamber (**Figure 7-3**). The humidity was controlled into the inlet of the system using a commercial automated humidifier (Humidifier Fogger 4 L, Reptizoo), and connected to an outlet chamber with a commercial sensor (Growhub Controller E42A, Vivosun). A stream of air of known humidity from humidifier was funneled into the AFM dish, which was sealed with an O-ring and silicon cover, and had both an inlet and outlet. The outlet was fed into a sealed glass chamber, which was maintained at a known humidity with real-time feedback. The entire closed humidity circuit was linked with silicon tubing (0.8 mm inner diameter). The system was especially designed to allow for a fast change of the relative humidity (~ 15 min), using only commercial equipment. Humidity was increased in 10% (+/- 1%) increments starting with room RH of 50% up to 90%. At 95% RH, the interspace between AFM tip and sample was completely submerged. As the goal of this study is to measure the adhesion force between plastic and sand in air to link the force with atmospheric emission of microplastics, RH was capped at 90%. This experiment was repeated for both a PP and a silica (glass) sample. At each humidity stage (50%, 60%, 70%, 80%, 90%), adhesion forces were measured.

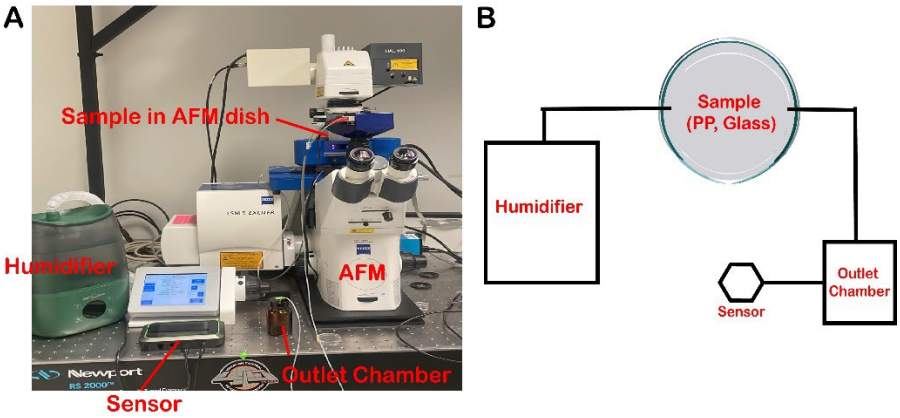

Figure 7-3: Schematic of the experimental setup used to adjust the humidity.

7.2.6. Quality assurance and quality control

Plastic cross-contamination was minimized during sample preparation and analysis by using non-plastic tools. Polypropylene sheets were commercially sourced and kept in a dark cupboard to prevent non-experimental UV weathering. Additionally, 0 minutes of UV weathering was served as the control/baseline. Polypropylene and glass samples were cut with metal scissors, scored with metal tweezers, and stored in pre-washed glass containers. Data was visualized as boxplots (minimum, first quartile, median, third quartile, maximum). Statistical significance of differences in mean values was assessed a Tukey one-way test with 95% confidence level where $p < 0.05$ is assumed to be significant.

7.3. Results and Discussion

7.3.1. Effect of UV exposure on contact angle of PP

An increase in UV exposure significantly ($p < 0.05$) decreased the contact angle of PP (**Figure 7-4**). After exposure to UV for 0, 1, 5, 15, and 30 minutes, the contact angles were decreased from 112.0° (control, no UV exposure) to 112.7°, 112.2°, 108°, and 94°, respectively. Up to 5 minutes of exposure to UV, contact angle did not change. However, after a minimum of 5 minutes of UV exposure, contact angle (4° to18°) on PP decreased, suggesting that the UV

exposure easily changes the surface hydrophobicity of microplastics. We attributed the results to changes in molecular properties of the plastic surface under UV radiation. UV radiation induces chain scission of plastic surfaces (Y. Shi et al., 2021), and the result of these free radical chain reactions includes the breaking of C-C bonds and formation of hydroxylated chemical groups, such as C=O, C-OH and -COOH (Cai et al., 2018; Lin et al., 2020). These hydroxyl and carboxyl groups are polar, decreasing the hydrophobicity of the plastic surface. The result is consistent with other studies that found the contact angle to non-linearly decrease with increased UV exposure for PET (Gunther et al., 2023) and steadily decrease for PS, PE, PET and PVC alongside increased radiation dosage (Lin et al., 2020). The latter study found the speed of contact angle reduction varied with plastic type, likely due to more unstable chemical bonds for different polymer types which expedites the chain scission-induced hydrophilicity. This can explain why some studies found relatively non-linear reduction in contact angles, while others found more consistent trends with increasing UV exposure. Many studies confirm the surface degradation of plastics after UV exposure (Cai et al., 2018; Gewert et al., 2015; Mao et al., 2020), making it very likely that changes in contact angle are indeed a result of chemical changes on a plastic's surface from reactions with UV light. Overall, the results imply that UV light may alter the surface properties of plastics very quickly, after at least 5 minutes of simulated UV exposure. This is not a perfect simulation of real environmental degradation, as natural UV exposure is not as highly dosed or concentrated. Therefore, these laboratory doses can be equivalent to many months of sunlight exposure. Nevertheless, the results show that UV weathering rapidly corrodes microplastic surfaces, decreasing the surface hydrophobicity.

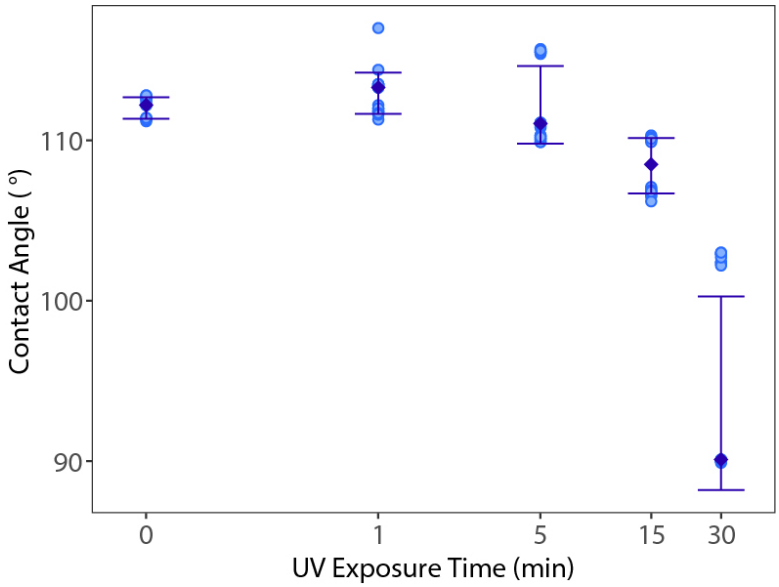

Figure 7-4: Contact angle (°) decreases significantly with increases in UV exposure time: 0, 1, 5, 15 ,30 minutes.

7.3.2. Increases in UV exposure to microplastics decrease adhesive interaction

The results showed that an increase in exposure to UV light significantly alters PP's adhesion force (**Figure 7-5**). The adhesion force decreased from 34.5 nN (control, no UV exposure) to 33.9, 10.8, 5.8, and 8.1 nN for 1, 5, 15 and 30 minutes, respectively. The adhesion force decreased significantly ($p < 0.05$) after 5 minutes of UV exposure and stayed relatively stagnant between 0-1 minute and 15-30 minutes. It only required 5 minutes of intense UV weathering for adhesion to drop by around 67.6%, where it then plateaued despite increasing weathering time. This trend is parallel to the significant decrease in contact angle that happens around similar UV exposure time. The experimental results verify our expectations based on the theoretical model (**Equation 7-1**), as adhesion force is proportional to contact angle; therefore, a decrease in contact angle with UV weathering should have resulted in a decrease in adhesion force. This study is the first to use AFM to examine how UV weathering, and the resulting decreasing hydrophobicity, can quantitatively affect the microscale interaction between microplastics and quartz. Comparing this trend to another study which compared adhesion trends between

hydrophilic (5-51º) and hydrophobic (110º) substrates and controlling for RH at 50%, hydrophilic surfaces had an adhesion force 1.2 times that of hydrophobic surfaces (Fukunishi & Mori, 2006). This result is opposite to the results in this study, which found decreasing adhesion force with hydrophobicity. However, this study was not looking at interactions between plastic and quartz, and adhesion force is highly dependent on the geometry and composition of tips which differs between these studies. Furthermore, UV weathering can also alter surface roughness, charge, and other surface properties alongside hydrophobicity- all of which can play a role in adhesion forces. For example, UV weathering is known to increase the surface area of a material, which can weaken the intermolecular forces as they are spread over a larger surface area. Overall, because of environmental weathering, the surface properties of microplastics are altered, which in turns decreases the adhesive strength at which plastic binds to sand. Due to this decrease in adhesive strength, microplastics have reduced resistance to the shear stress from wind. Thus, the emission potential of weathered microplastics could be significantly higher than non-weathered microplastics, and this effect happens quite quickly in the environment.

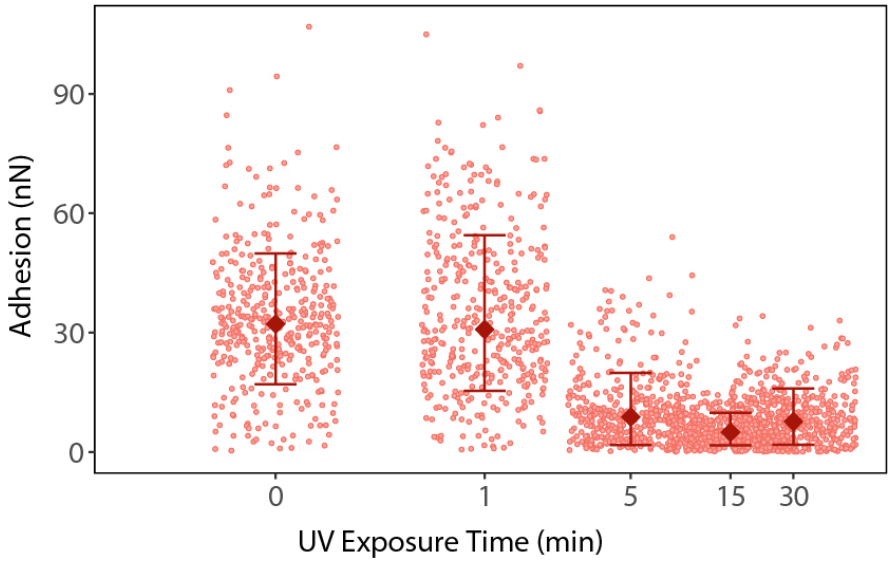

Figure 7-5: Adhesion (nN) decreases significantly as UV exposure time (0, 1, 5, 15 ,30 minutes) increases. Boxplots visualize the median and IQR, while the background points represent the n=2000 force curves analyzed across all exposure times.

7.3.3. Adhesion decreases with increased relative humidity

The results revealed that an increase in relative humidity decreased adhesion on PP surface and glass, though the effect is more prominent for PP (**Figure 7-6**). For PP, the adhesion force decreased by 84.0 %, from 378.5 nN to 60.5 nN, after RH increased just 10%. After which, RH continues to significantly decrease the adhesion force, though not as drastically. Adhesion forces dropped to 36.0 nN (-41%), 22.3 nN (-38.0%), and 17.8 nN (-20.1%) for 70, 80, and 90% RH. This trend demonstrates that most changes in the magnitude of adhesion force due to relative humidity (RH) occur almost immediately. Any subsequent increase in RH results in a comparatively smaller decrease in adhesion force. The adhesion force for silica, taken as a control, decreased more steadily from 365.8 nN at 50% (resting RH) to 365.5 nN (-0.1%), 336.6 nN (-8.0%), 151.9 nN (-55.0%), and 141. 9 nN (-6.6%) at 60, 70, 80, and 90% RH. The decrease in adhesion alongside increasing RH is both more extreme and more rapid for plastic than the control. Across the experimental range of 40% RH, plastic showed a 95% net decrease in adhesion force while silica had only a 62% decrease. The difference in adhesion interactions may be attributed to the difference in surface properties between the two samples: PP and silica. Silica- used to represent sand which is primarily quartz- is a highly porous and hydrophilic substance (Xi et al., 1995). Its porous surface allows for high surface area and mechanical interlocking between two sand particles (modelled by our Si-SiO_2 interaction) which could increase the intermolecular force strength between the surfaces making it more resistant to humidity effects. Furthermore, silica is already a hydrophilic substance, compared to plastic with is hydrophobic, so the hydrophilic surface function groups like hydroxyl (-OH) may form additional hydrogen bonds and this hydration controls its wettability and may contribute to its prevalent adhesive forces even at high humidity (Laskowski & Kitchener, 1969).

The experimental results are unexpected when compared to the theoretical model (**Equation 7-1**), as adhesion force is proportional to RH; therefore, an increase in RH should have resulted in a decrease in adhesion force. However, this relationship is complex. Other experimental studies have also confirmed the counterintuitive effect of increasing RH actually decreasing fluid threshold- which is reliant on adhesion force (Ravi et al., 2004, 2006). While these effects typically occur at middle-range humidity (50-65% RH) before liquid bridge formation for normal soil particles; for microplastics, this range could be much higher (maybe 80-90%) perhaps due to plastic's hydrophobicity. Other studies looking at adhesion as a result of RH, have seen trends which decrease (Ando, 2000; Farshchi-Tabrizia et al., 2008; Fisher & Israelachvili, 1981b; R. Jones et al., 2002; Kim et al., 2016), stay constant (Fukunishi & Mori, 2006), or increase (Ata, 2008) depending on the sample type and AFM tip. Thus, my results are critical to specifically analyze the adhesion force between plastic and sand since trends in adhesion cannot necessarily be applied across wide ranges of materials and rely heavily on the surface properties of the sample. Overall, this result indicates that as humidity rises, plastic is less tightly bound and preferentially eroded compared to other traditional pollutants which are primarily silica.

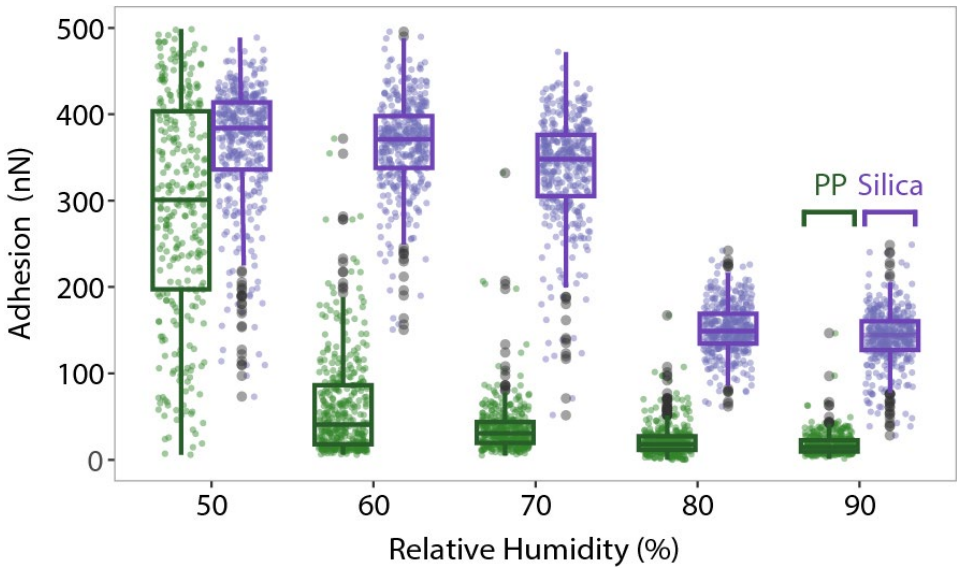

Figure 7-6: Adhesion (nN) decreases significantly as relative humidity increases (50-90%) for both PP-silica and silica-silica interaction. The decrease in adhesion is more extreme and more rapid for plastic (green), then for silica (purple). Boxplots visualize the median and IQR, while the background points represent the n = 4000 force curves analyzed across both samples and all humidities.

7.4. Environmental Implications

This study uses experimental and theoretical results to prove that UV weathering and increased RH both decrease the adhesion force between PP and sand, which makes microplastics more susceptible to wind erosion even in wet conditions (high humidity). Furthermore, this adverse effect is stronger for PP than for silica. This means plastics may experience lower binding forces to terrestrial surfaces than predominantly silica substances, like sand, likely due to their hydrophobicity, which may increase their atmospheric emission potential. Additionally, most plastics in the environment are weathered and the results imply these weathered microplastics are even more likely to be preferentially loaded into the atmosphere than the pristine one used in the laboratory. Thus, laboratory results may have underestimated the emission potential of microplastics. These microplastics, once suspended by wind, can travel long distances and may expose populations far from known sources or typical risk factors; future studies should evaluate this risk.

Furthermore, this paper provides surprising evidence that an increase in humidity decreases the interaction of microplastics with terrestrial sand surfaces. Contrary to previous assumptions, an increase in humidity does not enhance the adhesion of microplastics to the soil surface. Similar to the case for traditional soil particles at middle-range RH (50-65%), adhesion force actually falls with an increase humidity as the liquid bridge effect is not yet dominant. This trend expands to include a higher RH range (50-90%) for microplastics- perhaps due to their hydrophobicity-driven resistance to the formation of liquid bridges. For microplastics in high humidity conditions, the adhesion force falls, thereby facilitating the emission of microplastics- a finding that challenges the conventional understanding. Thus, future studies should examine microplastic emission in wind tunnel experiment by changing humidity and confirm whether increased humidity will indeed result in increased emissions. The implication is that in more humid seasons and climate zones, there is an exacerbated effect of decreasing adhesion strength and increasing emission potential for microplastics. As climate change is known to shift climate zones, which are associated with changes in average humidity, this information could be used to predict future geographical regions that should prioritize microplastic research and modeling which could better assess inhalation exposure risk to their populations. Overall, the fundamental results in this study improve our mechanistic understanding of why microplastics are preferentially loaded into the atmosphere and should be used to improve atmospheric dispersion and public health exposure models.

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8. CHAPTER 8 – CONCLUSIONS AND RECOMMENDATIONS

8.1. Conclusions

Impact of climate and land use on exposure to airborne microplastics: The analysis of deposited airborne microplastic concentrations reported in 24 viable studies from 15 countries yields the following conclusions: Deposition rate (n m^{-2} day^{-1}) can vary by 5 orders of magnitude, though variability stabilizes when sampling duration exceeds 10 days. Higher deposition rates were observed in the arid and tropical climates, whereas deposition rates in temperate and continental climates were similar. Deposition rates did not vary significantly with land use, potentially due to the long-range transport of airborne microplastics across geographical boundaries. A region's climate classification could play a greater role than land use in predicting the deposition rate of airborne microplastics. To improve monitoring, efforts should be focused on standardizing sampling duration and increasing the availability of data in the global South.

Historical contamination of microplastics in deep-sea sediment ocean cores: The vertical distribution of microplastic concentrations in sediment revealed most microplastics are trapped within the top 6 cm of the ocean, implying rapid environmental contamination within the last two decades. The observed concentration in seawater in this study was at least 3 orders of magnitude more than that reported in other studies from the same region, and the detection limit was proven to have a high impact on the reported concentrations of microplastics. Thus, the extent of microplastic pollution in the environment could be much higher than predicted based on previous studies that used a larger size cut-off.

Lack of methods standardization for biomonitoring of airborne microplastics: Among the three tested exposure factors—the height of the leaf above ground, land-use type, and leaf surface hydrophobicity—height contributes most to variability in the concentration of

microplastics on leaves. Leaf hydrophilicity, based on contact measurement, is weakly correlated with microplastic concentrations on the leaves. Land use types do not influence microplastic concentration on leaves, potentially due to homogenous atmospheric deposition of airborne microplastics originating from distant sources. SEM analysis confirmed that leaf surfaces are contaminated with larger microplastics (> 100 μm) which could be transported by the wind, as well as smaller (< 10 μm) microplastics, which can potentially enter leaf stomata. Overall, high variability and lack of predictive strength indicates that leaves may not serve as a reliable passive sampler for atmospheric microplastics.

Impact of socioeconomic factors and proximity to sources on exposure to airborne microplastics: Microplastic concentrations in deposited dust in urban regions have little to no correlation with land use and socioeconomic factors of the regions, which is inconsistent with environmental justice literature that has shown that these factors are typically associated with higher levels of particulate matter in the air. Only poverty exhibited a significant relationship with microplastic concentrations over 30 n g^{-1}, albeit weak and not linearly proportional. Regardless of socioeconomic status and irrespective of where people live, they are all exposed to similar concentrations of microplastics in the air, likely due to wind-driven dispersion of suspended microplastics.

Cause of preferential emission of microplastics: Microplastics are more likely to be transported by wind below the characteristic fluid threshold of soil, due to their lower density and weaker liquid bridge bonding potential compared to traditional soil particles. Polypropylene (PP) was estimated to have a fluid threshold of 1.64 m s^{-1} at 2 m above the ground compared to the experimentally determined threshold velocity of soil particles at this site, 5.84 m s^{-1} (Dukes et al., 2018). The preferential emission of microplastics due to their lower fluid threshold results in a

269% increase in the number of events that would resuspend microplastics within a three-month period. The theoretical framework proposed in this study may help predict microplastic emissions more accurately by accounting for microplastic fluxes typically underrepresented which can improve human inhalation exposure assessments.

Impact of UV weathering and relative humidity on emission potential of microplastics: UV weathering and increased relative humidity (RH) both decrease the adhesion force between PP and sand. The emission potential of weathered microplastics is significantly higher than non-weathered microplastics, with most of the adhesive strength lost after just 5 minutes of intense UV weathering. The impact of UV weathering and increased RH is stronger for PP than for silica, meaning plastics may experience lower binding forces to terrestrial surfaces than predominantly silica substances, like sand, likely due to their hydrophobicity. The results imply that microplastic's emission potential into the atmosphere could be increased as residence time in the environment increases, or in more humid environments.

8.2. Recommendations for future studies

Monitoring of Airborne Microplastics: Identifying inhalation risks of microplastics requires estimating microplastic concentration in air. The research described in Chapters 2, 4, and 5 confirmed the need for better sampling protocols to assess airborne microplastic concentrations. Although the results from these chapters inform how to develop effective models for the transport of atmospheric microplastics, they also highlighted flaws in the experimental methods used to assess microplastic concentration in the environment. Most studies cited in these chapters rarely followed a uniform protocol such as a unit describing microplastic concentrations (number vs mass), detection limit, and sampling duration. These inconsistencies led to variations in sample collection methods, detection of microplastics, and reporting of data. Thus, future studies should

focus on improving those limitations particularly related to sampling duration, frequency and measurement protocols as described below.

Sampling Duration and Conditions: Most studies rarely reported sampling duration, which can affect the concentration of microplastics in deposited dust. Without duration, it is difficult to compare the exposure risks between locations. For example, samples collected for months may accumulate more microplastics than those collected within a few days or hours. Similarly, antecedent conditions such as the drying period or last rainfall time could affect the air quality. Thus, future studies should record sampling duration and antecedent weather conditions when reporting concentrations of deposited airborne microplastics (n m^{-2} day^{-1}). A sampling duration of at least 10 days should be used to rule out any temporal variability in the sample, particularly overestimation of the microplastic concentration when sampled for a few hours.

Detection Limit: Many studies reported microplastic contaminations in the environment, but most of them do not report the detection limit of their measurement methods. The detection limit for a study has been shown to greatly influence the reported concentration, on the order of multiple magnitudes. Thus, it is critical for future projects to report their detection limit, so that its effect can be better understood, and steps be taken to standardize microplastic quantification methods to reduce this variability.

Long-Term, Varied Monitoring of Airborne Microplastics: Future sampling protocols should prioritize collecting long-term passive or active sampler data in multiple locations to establish a baseline contamination level for urban atmospheres. Furthermore, high-frequency data could reveal links between microplastic exposure and localized conditions contributing to microplastic exposure. For example, results from Chapters 1 and 5 showed that local climate, UV levels, relative humidity, and poverty in a region could play a role in exposure to airborne

microplastics. Thus, a database of high-frequency temporal data will help identify any patterns correlating these conditions with the concentration of airborne microplastics.

Wind Tunnel Experiments to Verify Preferential Emission of Microplastics: Results of Chapter 6 showed that the low density and high hydrophobicity of microplastics lowered their fluid threshold, leading to higher emissions into the atmosphere. However, the results are not verified in large-scale experiments. Thus, wind tunnel experiments should be conducted to verify the fluid threshold for a range of microplastics with varying density and hydrophobicity. The results from these controlled experiments could inform the fugitive dust emission model tailored for microplastics and improve the mechanistic understanding of microplastic transport by wind.

Mechanistic Understanding of the Factors Influencing Microplastic Adhesion Forces with Terrestrial Surfaces: Chapter 7 offers preliminary evidence that environmental factors such as UV exposure and relative humidity (RH) have a direct influence on the strength of adhesive forces between plastics and sand. Future studies should analyze the combined effect of these environmental factors in both AFM studies and wind tunnel experiments. Furthermore, UV exposure weathers microplastics, reducing their hydrophobicity, as well as affecting their material surface toughness, elasticity, rigidity, and uniformity (Iniguez et al., 2018; Moezzi & Ghane, 2013; ter Halle et al., 2017; Weinstein et al., 2016). The connection between UV exposure, changes in surface characteristics, and changes in the strength of adhesion forces should be confirmed in future AFM studies. Additionally, climate change will shift the distribution of regional climate and humidity patterns, making humid climates more humid and drier climates drier. Thus, future work can analyze adhesion forces between microplastic and sand under the full range of humidity from very dry (10%) to very humid (90%). This can be used to predict microplastic emissions under

changing climates and inform risk assessments for urban regions with increased vulnerability to emissions.

Influence of co-pollutants and biofilm on the emission potential of microplastics: Future research should investigate the influence of co-pollutants and biofilms on the emission potential of microplastics. For example, microplastics used in these Chapter were commercial and pristine, and biological presence on the surface was not characterized. Biofilms may alter the surface properties of microplastics, potentially affecting their microscale interactions, transport, and breakdown. Microplastics could instead be left in culture and later analyzed using AFM, SEM, FTIR, and PCR to understand how the presence of a biofilm on microplastic's surface could alter their ability to be stained fluorescent, contact angle (hydrophobicity), surface area, and adhesion forces between sand and plastic. Furthermore, understanding how co-pollutants such as: heavy metals, persistent organic pollutants (POPs), polycyclic aromatic hydrocarbons (PAHs), pesticides, endocrine disruptors, pharmaceuticals and personal care products (PPCPs), or industrial chemicals (nonylphenol), alter these same microscale interactions could reveal synergistic or antagonistic effects on their degradation and emission potential into the environment. Studies focusing on these interactions will provide a more comprehensive understanding of the emission potential and risks associated with microplastic pollution.

8.3. References

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