A Synthesis of Microplastic Sources and Pathways to Urban Runoff



















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Executive Summary

California Senate Bill 1263 (2018) tasks the Ocean Protection Council (OPC) with leading statewide efforts to address microplastic pollution, and requires the OPC to adopt and implement a Statewide Microplastics Strategy related to microplastic materials that pose an emerging concern for ocean health. Key questions remain about the sources and pathways of microplastics, particularly to urban runoff, to inform an effective statewide microplastics management strategy. The OPC funded this work to inform these microplastics efforts. The purpose of this project was to build conceptual models that synthesize and integrate our current understanding of microplastic sources and pathways to urban runoff in order to provide future research priorities that will inform how best to mitigate microplastic pollution. Specifically, we developed conceptual models for cigarette butts and associated cellulose acetate fibers (Section 2), fibers other than cellulose acetate (Section 3), single-use plastic foodware and related microplastics (Section 4), and tire particles (Section 5), which were prioritized based on findings from the recent urban stormwater monitoring of microplastics in the San Francisco Bay region. Conceptual models specific to each of these particle types are valuable tools to refine source identification and elucidate potential source-specific data gaps and management options.

The conceptual models presented herein trace the pathways of macroplastics and their microplastic breakdown products on the urban landscape and their transport to urban runoff. We developed a systematic approach to identify the diverse array of potential sources for each microplastic focus and narrow down the most likely dominant sources. We then highlight the broad range of potential microplastics mitigation opportunities from prevention strategies like reducing plastic use and re-designing plastic products, to reducing the release of microplastics and associated chemicals, to downstream approaches to capture or collect microplastics.

Cigarette Butts and Cellulose Acetate Fibers

Cellulose acetate was one of the dominant fiber polymers identified in San Francisco Bay Area urban runoff, and cigarette butts are likely the main source of these cellulose acetate fibers. Cigarette butts are one of the most commonly littered items, despite public campaigns against butt litter and municipal ordinances to curb smoking in public spaces. An important reason is that the public still misunderstands their toxicity and persistence because they seem like they are made of organic materials and should be readily biodegradable, but they are not. There are many available approaches to mitigating cigarette butt and cellulose acetate fiber pollution, from ongoing public health efforts to reduce tobacco use, to preventative measures requiring tobacco manufacturers to remove or re-formulate filters, to further downstream approaches to reducing cigarette butt litter. Various policies and programs have been piloted or implemented to reduce cigarette butt litter; therefore, a priority research need to inform management is improved understanding of the relative efficacy of various cigarette butt litter prevention measures. Improved methods to distinguish cellulose acetate fibers from other cellulosic fibers in the field are needed to monitor and collect the data needed for these evaluations.

Fibers

Fibers composed nearly half of the microplastics identified in recent urban runoff monitoring in the San Francisco Bay region, and are one of the most ubiquitous forms of microplastic pollution reported globally. Due in part to their high surface area to volume ratio, fibers emitted to the outdoor air can transport through the air for long distances, as evidenced by detections in remote locations worldwide. There are many potential sources of fibers from products used indoors and outdoors. Indoor fibers can be transported to the outdoors through various activities. While there are significant data gaps to understand whether other outdoor products may be an important source of fibers, available information suggests that dryer vents may be an important source. In the US and Canada, use of mechanical air clothing dryers with outdoor ventilation is significantly higher than in other parts of the world, where unvented condenser dryers and hanging items to dry are more common. These geographic differences may explain why drver vents are an under-studied source of microplastic pollution. We outlined several approaches to mitigating fiber pollution from textiles, from preventative measures requiring the textile industry to redesign products to reduce the level of fibers shed, to changing consumer behavior to reduce use of traditional dryers, to removing fibers from dryer exhaust, to further downstream approaches to capture fibers after release into the environment.

Despite lack of attention to dryers, there is widespread awareness of apparel and other textiles as a source of microplastic pollution (although the focus has been on laundry wash water and the wastewater pathway). The apparel and textile industries are facing increasing pressure from state and federal legislatures and environmental groups to address fiber pollution. A priority science question to inform management of fibers is to understand the major sources of fibers, placing the contributions from textiles and dryers to urban runoff in the context of other potential sources. Additionally, understanding whether natural fibers (with and without chemical modification) are less toxic than synthetic fibers is important to inform whether natural fibers are a safer alternative to synthetic fibers.

Single-Use Plastic Foodware (SUPF) and Related Microplastics

Single-use plastic foodware, broadly defined as disposable plastic items designed for single-use to serve, package, transport, or consume food and beverages, are used ubiquitously in urban environments. SUPF use continues to increase and SUPF litter is widely observed on land and in aquatic environments. SUPF litter in both terrestrial and aquatic environments can deteriorate and form microplastics over time through complex interactions of various chemical and physical processes. The ubiquitous presence of SUPF in litter suggests that SUPF may be an important contributor to microplastics. However, partial degradation processes are very complex and, therefore, there is limited ability to translate this information to understand the generation of microplastics from SUPF and link microplastics to their potential SUPF sources.

We summarized the wide variety of approaches to mitigating SUPF and related microplastics in the environment, from preventative measures to reduce SUPF use and

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waste, to downstream approaches to improve removal of trash and associated microplastics. The priority science needed to address this data gap is the establishment of standardized frameworks for monitoring, analyzing, characterizing, and reporting trash and microplastics; this will support integrating work and solutions for both trash and microplastics in land and water environments. In addition, development of analytical methods to link secondary microplastics to their original sources will help identify the key sources to target for mitigation of SUPF and related microplastics in the environment.

Tire Particles

Modeling studies indicate tire wear may be one of the top sources of microplastic releases to the environment globally. US tire wear particle environmental releases have been estimated to be between 3.0 and 5.5 kg/y per capita. Tire particles have the potential to harm organisms due to both their physical characteristics and the chemicals they contain. Aquatic toxicologists have found that tire particle leachates can be toxic to a variety of aquatic organisms. One of the chemicals identified in tire particle leachates, 6PPD-quinone (an environmental transformation product of a tire preservative), causes pre-spawn mortality in coho salmon. While there are other potential sources of rubber to urban runoff, these are minor compared to tire wear from vehicles.

Tire wear particles have a relatively unique size distribution, spanning a range that includes tiny particles <2.5 μ m in diameter that transport long distances in air, and larger particles >10 μ m that typically deposit close to their near-ground point of emission. Tire wear particles also have a higher density than many other types of microplastics detected in the environment. Sample collection and microplastic separation methods likely have underestimated the presence of tire particles in aquatic environments, due to their small (<100 μ m) size distribution and density.

Motor transportation is a long-recognized climate change, air quality, and infrastructure challenge in California; mitigating microplastics generation from vehicles is likely to be a similarly formidable challenge. But our systematic approach to understanding the generation and pathways of tire wear highlights many available mitigation approaches. These range from preventative measures to reduce the volume and toxicity of tire wear particles (such as shifting to rail transportation and removing toxic ingredients), to reducing tire wear debris formation by reducing vehicle miles traveled or shifting to low-wear tires, to using innovative new technology to collect tire wear particles on vehicles, or downstream approaches like collecting dispersed tire wear particles in porous road pavement, or removing tire particles and tire-related chemicals from urban runoff before it flows into creeks, rivers, or the ocean. Priority data gaps (in addition to improved monitoring methods and additional monitoring data) are those that will help focus management strategies by informing what particle sizes from tire wear are important to capture to mitigate water quality impacts, as well as identifying vehicle classes that have the highest total tire particle emissions in California.

Definitions

Vocabulary	Definition as Used in this Report
Aerodynamic diameter	The diameter of an idealized spherical particle used to express the behavior of airborne particles (which actually have irregular shapes).
Apparel industry	Economic activity concerned with the production of clothing. This report's use is inclusive of activity in the textile industry.
Aspect ratio	Ratio of length to width (i.e., length : width).
Biodegradation	The action of microorganisms to chemically alter plastic material.
Biofouling	The colonization of a particle by (micro)organisms, leading to accumulation of biological material on a particle surface.
Circular economy	An economic system that employs reuse, sharing, repair, refurbishment, remanufacturing, and recycling to create a closed-loop system, minimising the use of resource inputs and the creation of waste, pollution, and carbon emissions. A circular economy stands in contrast to a linear "make-use-dispose" economy.
Complete degradation	Combination of abiotic and biotic processes leading to complete breakdown and mineralization of plastics into inorganic small molecules (e.g., water, carbon dioxide, methane).
Conceptual model	A pictorial representation used to express relationships and highlight key or dominant processes.
Directly connected impervious area	Dense urban areas with high proportions of impervious surfaces directly connected to storm drains.
Downcycling	The recycling of waste where the resulting recycled material is of lower quality and functionality than the original material.
Fate	The degradation processes microplastics are subject to and where they end up after environmental release.
Fibers	Microplastics that have a long, narrow thread-like shape, significantly longer in one dimension than in the other two dimensions. This term is used instead of "microfibers" to avoid inconsistency with the terminology used by the textiles industry.
Green stormwater infrastructure	Systems designed to slow, infiltrate, use, and/or treat urban runoff using vegetation, soils, and natural processes.
Greenwashing	Public communications that provide misleading information about the sustainability of products that are meant to deceive customers to believe products or practices are more environmentally friendly than they really are.
Hydrolysis	The chemical breakdown of plastic due to reaction with water.
Macroplastics	Plastic particles greater than 5 mm in size.

Vocabulary	Definition as Used in this Report
Microfibers	Synonymous with fibers. Some members of the textile community use this term slightly differently, to refer to textiles made of very fine synthetic material threads.
Microplastics	Plastic particles smaller than 5 mm in at least one external dimension. This report's use is inclusive of nanoplastics. This definition is consistent with the California State Water Resources Control Board Resolution 2020-0021 defining microplastics in drinking water, which defines plastic as solid polymeric materials to which chemical additives or other substances may have been added; excludes polymers that are derived in nature that have not been chemically modified (other than by hydrolysis) such as cellulose; and includes synthetic polymer composites, co-polymers, modified natural polymers (i.e., synthetic polymer-encapsulated natural polymers or natural polymers with synthetic polymer content greater than or equal to 1% by mass).
Nanoplastics	Plastic particles smaller than 1 μm. Included within this report's definition of microplastics.
Oxidation	The abiotic process that deteriorates plastic through contact with a chemical oxidizer (usually oxygen).
Partial degradation	Intermediary steps in the complete degradation process where plastic characteristics are changed (e.g., discoloration, surface cracking, fragmentation), but complete mineralization is not yet achieved.
Pathway	The environmental compartment through which microplastics are transported from sources. Examples include stormwater and wastewater.
Photooxidation	The abiotic process that deteriorates plastic with an initial exposure of UV rays (i.e., sunlight) that primes the material for subsequent oxidation reactions (see Oxidation).
Primary microplastics	Microplastics that are designed and manufactured to be small for a variety of uses, including pellets for plastic production (e.g., "nurdles"), abrasive blasting, paints and adhesives, agricultural applications, and for use in personal care products. Primary microplastics are released to the environment as a consequence of the use of products that intentionally contain or release them during maintenance and use.
Secondary microplastics	Microplastics that originate from the partial degradation of larger plastic items, regardless of when this breakdown occurs.
Single-use plastic foodware	Disposable plastic items designed for single-use to serve, package, transport, and consume prepared food and beverages, including bags, bottles, bowls, caps, cups, cutlery, plates, straws, stirrers, takeout containers, trays, and wrappers.
Source	The product(s) from which microplastics originated.

Vocabulary	Definition as Used in this Report
Stormwater	Runoff that is generated from storm-driven rain events.
Textile industry	Economic activity associated with production of fibers, yarns, and fabrics, which are used for various products and applications. This report's use is inclusive of activity in the apparel industry.
Textiles	Material and finished product made from fibers, can be woven or nonwoven.
Tire particles	All types of microplastics containing tire material, including both tire wear particles and microplastics created from end of life tires.
Tire wear particles	Refers only to microplastics generated by tire wear on pavement.
Transport	The movement processes microplastics are subject to after environmental release.
Urban runoff	Surface water runoff (includes storm-driven rain and dry weather flows) from urban (mostly impervious surface) landscapes that can pick up contaminants as the water flows towards receiving waters.
Washoff fraction	The portion of particles washed away in runoff instead of remaining sequestered on land.

Acronym	Definition
6PPD	N-(1,3-dimethylbutyl)-N'-phenyl-p-phenylenediamine
CalRecycle	California Department of Resources Recycling and Recovery
Caltrans	California Department of Transportation
DCIA	Directly connected impervious area
DTSC	California Department of Toxic Substances Control
ECHA	European Chemicals Agency
EPR	Extended Producer Responsibility
EPS	Expanded polystyrene
EVALI	E-cigarette or vaping product use-associated lung injury
FEMA	United States Federal Emergency Management Agency
GADSL	Global Automotive Declarable Substance List
GC-MS	Gas chromatography-mass spectrometry
HDPE	High-density polyethylene
IMDS	International Material Data System
LDPE	Low-density polyethylene
mm	Millimeter
MPE products	Molded, poured, and extruded products
MRSLs	Manufacturing restricted substance lists
OPC	Ocean Protection Council
PAH	Polyaromatic hydrocarbon
PE	Polyethylene
PET	Polyethylene terephthalate
PP	Polypropylene
PS	Polystyrene
RSLs	Restricted substance lists
SBR	Styrene-butadiene rubber
SUPF	Single-use plastic foodware
TED-GC-MS	Thermoextraction desorption gas chromatography mass spectrometry

Acronym	Definition
TMDL	Total Maximum Daily Load
US	United States of America
USEPA	United States Environmental Protection Agency
VMT	Vehicle miles traveled
μm	Micrometer

1. Introduction

1.1. Background

"Just one word...Plastics. There is a great future in plastics." This memorable line of dialogue from the classic 1967 film "The Graduate" encapsulated the optimistic vision of a generation of Americans: the notion that plastics could be key to transforming everyday living and to creating a hugely profitable industry in the process. What began as a mid-century promise to address the problems of a generation has also produced its own multigenerational legacy of intractable challenges, as plastics today are ubiquitous in everyday consumer and industrial products, ranging from product packaging and shopping bags to food containers and medical supplies. The use of plastics has increased on a worldwide scale over the past century, which has resulted in an increase of plastic debris in our oceans (Andrady, 2011; Eriksen et al., 2014), caused endangerment to wildlife via entanglement and ingestion (Gall & Thompson, 2015; Wilcox et al., 2018), and led to habitat damage (Lusher et al., 2015; Rochman et al., 2013) and significant economic losses (Leggett et al., 2018; Newman et al., 2015). While the increase of macroplastics (defined herein as particles greater than 5 mm in size) in the environment has long recieved management attention, microplastics (herein defined as particles smaller than 5 mm in at least one external dimension) and nanoplastics (the subset of microplastics smaller than 1 µm) are a more recent focus of environmental research. Environmental occurrence and effects of micro- and nanoplastics are a growing concern, only beginning to be addressed by management actions.

Microplastics appear in virtually every environment on Earth. Microplastics occur in water and sediment in rivers, estuaries, beaches, and coastal oceans receiving urban runoff and municipal wastewater discharges (Browne et al., 2011; Sutton et al., 2019). Microplastics also appear in the open ocean (Barrows et al. 2018), Arctic (Athey, Adams, et al., 2020), deep ocean trenches (Jamieson et al., 2019), and remote mountain wilderness (Allen et al., 2019; Brahney et al., 2020; Napper et al., 2020).

While macroplastic pollution has long been a concern for its hazards to wildlife, mounting evidence suggests microplastics are also a concern for wildlife and human health. Aquatic organisms at every trophic level are exposed to microplastics (Besseling et al., 2019; de Sá et al., 2018; Wright et al., 2013). However, the specific health risks posed remain uncertain due to variations based on the plastic type, shape, species, and exposure. Ingested microplastics can impact the biochemical and physiological processes of many different types of animals (Burns & Boxall, 2018; Foley et al., 2018; Prokić et al., 2019; Wright et al., 2013). Microplastics can also expose organisms to potentially harmful chemicals, especially plastic-associated contaminants and additives such as flame retardants, plasticizers, or dyes (Fries et al., 2013; Rochman et al., 2019). Micro- and nanoplastics and harmful plastic chemical ingredients and additives can also be transferred up food chains (Athey et al., 2020; Carbery et al., 2018; Chae et al., 2018; Chagnon et al., 2018; Farrell and Nelson, 2013; Mattsson et al., 2017; Setälä et al., 2014; Tosetto et al., 2017). Increasing evidence suggests that people are also exposed to microplastics via inhalation and consumption of contaminated food and

beverages (Mohamed Nor et al., 2021) and smoking (Pauly et al., 2002), and that human health may be adversely affected by microplastic exposure (Campanale et al., 2020; Smith et al., 2018).

The term 'microplastics' broadly includes a wide range of small plastic materials; environmental contamination occurs through the release of primary or secondary microplastics. Primary microplastics are plastics designed to be small for a variety of uses, including pellets for plastic production (e.g., "nurdles"), abrasive blasting, paints and adhesives, and for use in personal care products. Secondary microplastics originate from the partial degradation of larger plastic items, regardless of when this breakdown occurs during the product life cycle. Tiny tire wear particles generated during vehicle use as well as tiny plastic fragments from the breakdown of plastic litter long after its release would both be considered secondary microplastics. Currently there is no single established definition of microplastics, although a few scientific and regulatory definitions of microplastics have been proposed and adopted for specific contexts (California State Water Resources Control Board Resolution 2020-0021, 2020; European Chemicals Agency, 2019; Hartmann et al., 2019). The California State Water Resources Control Board has adopted a broad definition of microplastics in drinking water, which includes all solid polymeric particles ranging from 1 nm - 5 mm, excluding certain natural materials (California State Water Resources Control Board Resolution 2020-0021, 2020). Materials such as rubber and cellulose acetate are considered plastic under this definition. This is the definition we use in this report, though our focus is on a different matrix, urban runoff. Definitions may continue to change in the future, reflecting the rapidly evolving understanding of microplastic occurrence, hazards, and regulatory context.

One of the major challenges to addressing microplastic pollution is identifying the product source of microplastics, as well as the pathways by which they are transported into the environment. We consider sources to be the products from which microplastics originate, whereas environmental compartments through which microplastics are transported (e.g., urban runoff or wastewater) are considered pathways. Understanding the sources of microplastics, as well as their dominant transport pathways to the environment, is crucial to informing microplastics management strategies and policies to direct actions to reduce pollution. Data on microplastic characteristics such as polymer composition, color, size, and morphology are commonly collected to provide clues as to their potential sources. Further refinement of these characteristics, as well as characterizing surface topology (Cowger et al., 2020), tensile strength (e.g., hard or soft), and texture (e.g., elastic or brittle or compressible)¹ have been suggested to further characterize polymers and help link microplastics to their potential sources.

Recent investigations in San Francisco Bay identified urban runoff as a major pathway for microplastics to enter receiving waters (Sutton et al., 2019; Werbowski et al., 2021; Zhu et al., 2021), with average concentrations of microplastics in urban stormwater

¹ Discussed during Joint Meeting of Regional Monitoring Program for the San Francisco Bay microplastics Workgroup and Ocean Protection Council Stakeholders on April 21, 2021.

(runoff generated from rain events) approximately two orders of magnitude higher than those in treated wastewater effluent. Nearly half of the particles observed in urban stormwater were suspected to be tire wear particles due to their morphology, distinctive black color, and rubbery texture. Fibers were the second most common class of microplastics observed in stormwater. The majority of fibers were identified as 'anthropogenic unknown' (indicating they had been dyed with a dye or coloring agent, but the underlying fiber composition could not be identified), polyester, or cellulose acetate. Other than the suspected tire wear particles and fibers, the most abundant materials that could be identified in stormwater samples were fragments of polypropylene (PP) and polyethylene (PE), common plastics used in a variety of products. Other pathways of potential interest, including atmospheric deposition and agricultural runoff, were not examined in this study.

In most urban environments in California and the western US, rainfall and runoff wash particles into stormwater collection systems that discharge directly to receiving waters without treatment. The separate storm drain system design partially explains the high microplastic concentrations observed in San Francisco Bay stormwater relative to treated wastewater effluent. Many other studies of microplastics around the world have characterized microplastic transport and/or fate in urban settings with combined sewer systems, an older type of drainage infrastructure design where urban runoff drains into the municipal wastewater collection system and (if the system does not overflow) the combined wastewater and runoff flows through a wastewater treatment plant prior to its release to the environment. To our knowledge, there has not yet been monitoring of microplastics in California urban runoff outside of the San Francisco Bay Area, though the relatively high microplastic concentrations measured in San Francisco Bay stormwater are generally consistent with limited observations of microplastics in stormwater in other locations, such as Toronto, Canada (Grbić et al., 2020).

We hypothesize that the findings about the importance of microplastics in urban runoff in the San Francisco Bay can be extrapolated to other dense urban areas in California. The concentrations observed suggest that urban runoff may be a more important pathway for microplastics to enter the coastal ocean compared to the wastewater pathway, where treatment would be expected to remove a significant portion of microplastics from final effluent. Other studies are needed to assess the hypotheses that urban runoff throughout California has similarly high concentrations of microplastics relative to treated municipal wastewater effluent, and that similar distributions of particle types (i.e., tire particles, cellulose acetate and other plastic fibers, PP and PE fragments) are present. These findings indicate a need to better understand microplastics in urban runoff, an under-studied transport pathway for these contaminants.

1.2. Purpose and Scope of Report

The purpose of this project was to build conceptual models that synthesize and integrate our current understanding of microplastic sources and pathways to urban runoff to provide future research and management recommendations. The Ocean Protection Council (OPC) funded this work to inform development and implementation

of a Statewide Microplastics Strategy related to microplastic materials that pose an emerging concern for ocean health, as mandated in California Senate Bill 1263 (2018). In addition to threats to ocean health, urban runoff and stormwater conveyance often occurs through important freshwater habitat areas in streams, creeks, rivers, and wetlands, which can also be threatened by microplastic pollution. Key questions remain about the sources and pathways of microplastics, particularly to urban runoff, to inform an effective statewide management strategy for microplastics (Wyer et al., 2020).

This report focuses on urban runoff, primarily due to the prevalence of microplastics observed in this pathway in the San Francisco Bay Area. As noted previously, the scope is further guided by the fact that most of California has municipal storm drain systems that are fully separated from municipal wastewater collection systems. Municipal wastewater collection systems flow to municipal wastewater treatment plants that discharge directly to surface water (and not into urban runoff). The San Francisco Bay data indicate that urban runoff is a significant pathway for microplastics to enter surface waters, yet most microplastics investigations and many management actions focus on wastewater. Except in unusual circumstances (e.g., sewer line overflows), municipal wastewater is not a source of microplastics to California urban runoff. This drainage design, which is common in the western US, stands in contrast to Europe and Asia, where much microplastics research has been conducted in urban watersheds served by combined sewer systems that receive and treat both indoor wastewater and urban runoff.

Although it is currently difficult to link many microplastics back to their specific sources, the San Francisco Bay results indicate a few specific likely sources for certain material/morphology combinations in urban runoff (Sutton et al., 2019; Werbowski et al., 2021.; Zhu et al., 2021), which we prioritized for further study. The cellulose acetate fibers observed in urban runoff are likely from cigarette filters because cigarette butts are commonly littered and are by far the most common use of cellulose acetate that would break down into the fiber shape. The sources of other fiber materials in urban runoff are less clear because the other commonly observed plastics are used in a wider variety of applications. However, apparel is likely to be one major source. The black rubbery fragments are suspected tire wear particles, a linkage further supported by independent modeling studies. The other commonly observed fragment materials may be tentatively linked with single-use plastic foodware (SUPF) because of their material and status as commonly littered items in urban settings; however, at this time it is not possible to distinguish microplastics from SUPF and those from other possible sources made of the same plastic polymers.

Therefore, for this report, we specifically developed conceptual models for cigarette butts and associated cellulose acetate fibers (Section 2), fibers other than cellulose acetate (Section 3), single-use plastic foodware (Section 4), and tire particles (Section 5) in urban runoff. Conceptual models specific to each of these sources and/or particle types are valuable tools to refine source identification and elucidate potential source-specific management options.

The conceptual models presented herein trace the pathways of macroplastics and their microplastic breakdown products on the urban landscape and their transport to urban

runoff, as shown in Figure 1.1. Microplastics in urban runoff will flow further downstream to receiving waters, including streams, rivers, coastal embayments, the ocean, and sediment beds. Small microplastics may be transported in the air compartment before being transported to urban runoff via wet or dry deposition on the urban landscape. Other major pathways for microplastics to enter receiving waters are generally not included in the conceptual models; these include direct littering of macro- and microplastics to receiving surface waters, direct microplastic deposition from the air to receiving surface waters, microplastics in wastewater, microplastics in agricultural runoff, and aquatic shore activities that generate microplastics. Our objective was to link the dominant microplastics identified in San Francisco Bay Area stormwater to sources and pathways in the urban environment to inform management actions, including source control. Each section contains a review of the literature linking the observed microplastic types to major sources and transport pathways relevant to urban runoff. We further briefly discuss the uncertainties and priority data gaps that limit our ability to identify the major sources and pathways.

To inform the development of a microplastics management strategy, we also briefly outlined potential microplastics control strategies that we identified through development of each conceptual model. There are many available approaches for mitigating microplastic and microplastic-related chemical environmental releases, from preventive measures to avoid emissions and remove toxic ingredients, to measures for capturing microplastics and microplastic-related chemicals in urban runoff (Figure 1.2). These summaries highlight the broad range of potential mitigation opportunities and identify the actors (e.g., industry, government, or consumers) that would implement each measure, but do not address implementation mechanisms (e.g., voluntary/mandatory or financial incentive/penalty) or public policy questions (e.g., economic or social costs).

Our framework for presenting control strategies, using generic categories of management actions, is shown in Figure 1.2. We frame the management actions (discussed below) on a scale from preventative (reducing use and release of microplastics) to remediative (collecting and removing microplastics). Each urban runoff conceptual model section of this report includes a more detailed and specific control strategy diagram. These control strategies are not comprehensive, in that they do not address all impacts of relevant macroplastic products. Related management concerns beyond microplastic pollution, such as climate change/greenhouse gas emissions, chemical release, water use, non-renewable resources, soil depletion, loss of ecosystem services, etc., are not addressed.

We have categorized management actions based on the party responsible for implementing each measure. It is important to note that actions implemented by industry or consumers may be triggered and overseen by government agencies as a result of legislation or regulation. Currently, most industries do not consider the whole life cycle of their products. Contrary to the current system operating on a linear "make-use-dispose" process and concentrated on waste management at the products' end-of-life (usually not by the product producers), the circular economy would function as a closed loop to vastly improve reuse and recycling of plastic materials. Overall, this reframing of responsibility could refocus waste management on elimination of waste

throughout the life cycle of products to maximize use and lifetime of materials and resulting products. Since most microplastics also contain potentially toxic chemicals (the effects of which often cannot readily be separated from the effects of the microplastics that carry them), industries can reduce microplastic impacts through product reformulation to eliminate or minimize persistent or toxic ingredients and to eliminate generation of microplastics during product use. Government-led management actions include legislative and local actions to incentivize or enforce product design, changed behaviors (of both industry and individuals), optimize waste management systems and processes, and support research and infrastructure changes for improved capture and removal of macro- and microplastics from land and water. Community-wide actions outside of governmental actions are also important. These include reduced use, consumption, and environmental release of plastic products and secondary microplastics, as well as various environmental cleanup efforts.

Each chapter of this report includes identification of general data gaps in the literature, and highlights specific priority data needs to inform management. The priority data needs outline specific next studies we believe are essential to inform management decisions regarding each type of microplastic. These individual priority data needs are synthesized in the Conclusions.

1.3. General Concepts in the Formation of Secondary Microplastics

Various breakdown processes are important in the formation of secondary microplastics from macroplastic sources, and these processes play a role in the urban runoff conceptual models presented in this report. Both macroplastics and microplastics may make their way through many of the pathways outlined in the conceptual models. Cigarette butts (Section 2) and single-use plastic foodware (Section 4) are commonly littered items that can be partially degraded to microplastics. Fibers (Section 3) are released from clothing and other textiles during their production, use, maintenance, and disposal. Tire particles (Section 5) wear off tire tread when vehicles are driven and are produced during waste tire recycling.

Plastics are designed specifically for their stability and durability, and their degradation times are very long compared to most natural organic materials. The half-lives of microplastics are not known with certainty due to the broad variety of polymers in use and varying environmental conditions (Chamas et al., 2020); however, it is widely believed that these particles resist environmental degradation and will persist in the environment long after their release (European Chemicals Agency, 2019; SAPEA, 2019). The varying characteristics of plastics and conditions in the environment (geography, sunlight availability, temperature, mechanical forces, microbial community, anthropogenic impacts, etc.) create a complex series of interactions that lead to generation and transformation of microplastics. Once in the environment, microplastics can continue to break down into smaller microplastics and chemically transform for decades to centuries before their complete degradation and mineralization to small inorganic molecules (e.g., carbon dioxide or methane).

The term 'degradation' is used inconsistently in common language and the scientific literature to refer to both partial breakdown and complete mineralization. In this report, we use the term 'complete degradation' as the combination of abiotic (physical and chemical) and biotic processes leading to complete breakdown and mineralization of plastics into small inorganic molecules like water and carbon dioxide. We explicitly use the term 'partial degradation' when referring to intermediary steps in this process where plastic characteristics are changed (e.g., discoloration, surface cracking, and fragmentation into micro- and nanoplastics), but complete mineralization is not yet achieved.

Photooxidation is the abiotic process that deteriorates plastic with an initial exposure of ultraviolet (UV) light (i.e., sunlight) that primes the material for oxidation reactions. Photolysis is particularly efficient when plastics on land surfaces are exposed to the air, especially where high temperatures further increase the rate of deterioration, such as on beach surfaces (Andrady, 2017). Plastic is also deteriorated by mechanical forces in the environment, including abrasion by foot and vehicles, wave action, and turbulence from wind and water. This is especially relevant for urban runoff, as a combination of mechanical forces including wind, water, and anthropogenic forces (humans, vehicles, etc.) can fragment plastics. Across all environments, photooxidation and mechanical action are considered the most important processes of deterioration and degradation of plastic products and related microplastics, though the contributions of biodegradation and hydrolysis are also relevant.

Biodegradation, the action of microorganisms to chemically alter plastic material, can occur throughout the environment. Biodegradation may be enhanced by the formation of biofilms on plastic surfaces (biofouling), though this may contribute to less surface availability for the action of abiotic processes (Andrady, 2017; Rummel et al., 2017). Plastics with hydrolyzable covalent bonds may also break down via hydrolysis when exposed to water. Though biodegradation and hydrolysis in the environment are generally considered too slow to meaningfully degrade plastics, the synergistic effects of these diverse processes contribute to the fragmentation and deterioration of plastic macro-debris into microplastics and may also contribute to microplastic breakdown (Andrady, 2017).

There are several published methodologies to examine the deterioration and degradation of plastics, but few of these can be translated to generation of microplastics. Within the literature, analyses can be categorized into assessments of mass loss, chemical changes, and physical changes to the original polymer structure. Mass loss simply uses changes in mass to determine breakdown of a plastic product. This can include complete degradation, as well as partial degradation and fragmentation to microplastics. Assessments of chemical changes often examine the concentrations of specific functional groups in polymers to determine the extent of deterioration. The composition of the plastics is thus important to deterioration rates, though the complexity of plastic mixtures can make understanding these rates more difficult (Andrady, 2017; Li, 2018). Partial degradation generally takes place at the plastic surface, so surface area is an important consideration (Chamas et al., 2020). Microplastics in the environment may also leach plastic additives and/or absorb other

contaminants (Coffin et al., 2018), which further complicate their deterioration and identification. Methods looking at changes in physical properties typically examine the strength of the polymer through thermal and surface analysis.

Although some studies have tried to develop quantitative measures for understanding these phenomena, most provide wide ranging values for the breakdown of various plastic materials that are difficult to compare to each other because metrics and experimental conditions have not yet been standardized across studies (Andrady, 2017; Chamas et al., 2020). Further, the broad characterization of partial and complete degradation and numerous metrics used make it challenging to extrapolate results to understand the generation of microplastics. Further discussion of the partial degradation processes that generate microplastics are described in Section 4 because partial degradation may be more relevant to the generation of secondary microplastics from SUPF, compared to the other sources discussed in this report. The challenges highlighted are an indication of the overall limitations of current science to definitively link many microplastics to potential sources, a priority data gap that must be addressed to inform identification and management of additional microplastic sources.



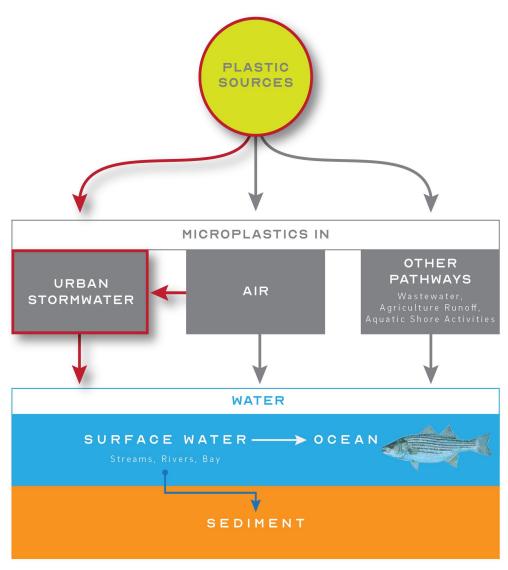


Figure 1.1. General pathways of macroplastics and their microplastic breakdown products and their transport to downstream receiving waters (streams, rivers, coastal embayments, and the ocean) and sediment beds. The red circle indicates the scope of the conceptual models presented in this report. Air transport includes both short and long-range transport to the urban stormwater pathway via wet or dry deposition. Other major pathways for microplastics to enter receiving waters, including direct deposition from the air to receiving waters, wastewater, agricultural runoff, and aquatic shore activities that generate microplastics, are generally not included in the conceptual models.

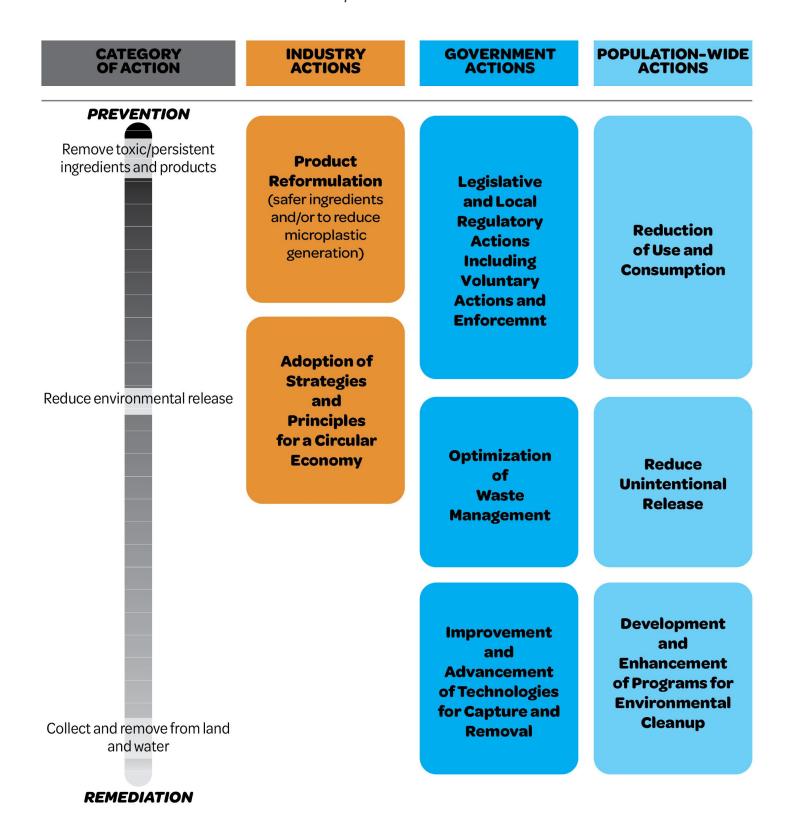


Figure 1.2. General microplastics management options, framed on a scale from preventative (reducing use and release of microplastics) to remediative (collecting and removing microplastics) measures. Options are color-coded by who would be responsible for implementation, with industry in orange, government in dark blue, and community-wide (non-governmental organizations) in light blue.

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2. Cigarette Butts and Associated Cellulose Acetate Fibers in Urban Runoff

2.1. Background

The purpose of this conceptual model (Figure 2.1) is to synthesize and integrate our understanding of the sources and pathways of cigarette butts and associated fiber degradates entering urban runoff. This document uses the word "fibers" instead of "microfibers" to avoid inconsistency with the terminology used by some members of the textiles industry. All other fibers are discussed in Section 3.

Cigarette butts, fibers, and associated contaminants pose toxicological risk to wildlife species. Cigarette butt leachate contains many toxic contaminants, including heavy metals, and can be acutely toxic to aquatic life (Lee & Lee, 2015; Moerman & Potts, 2011; Montalvão et al., 2019; Slaughter et al., 2011; Xu et al., 2019). Nicotine and cotinine (the primary metabolite of nicotine) are frequently detected in surface waters and are difficult to remove via drinking water treatment (Huerta-Fontela et al., 2008). Fibers from cigarette filters have received little toxicological study, but may also be a concern (Belzagui et al., 2021), especially because studies on other types of microplastic fibers frequently report toxic effects (Bucci et al., 2020; Jacob et al., 2020). The conceptual model is meant to inform future research and management recommendations for managing microplastic pollution.

2.2. Sources of Cellulose Acetate Fibers in Urban Runoff

Cigarette butts are one of the most commonly littered items, despite public campaigns against butt litter and municipal ordinances to curb smoking in public spaces. Cigarette butts are the most frequent form of litter found on beaches (Bergmann et al., 2015; California Coastal Commission, 2019). High numbers of cigarette butts are found in urban outdoor public places (Mock & Hendlin, 2019; Valiente et al., 2020) and on urban roadsides (Moriwaki et al., 2009), with higher population densities corresponding to more cigarette butt pollution (Valiente et al., 2020; Weideman et al., 2020). Trash surveys indicate cigarette butts are one of the most frequently littered items that collect in trash capture devices in California (EOA, Inc., 2014, 2016; Lippner et al., 2020).

Because they seem like they are made of organic materials and should be readily biodegradable, the public still misunderstands cigarette butt toxicity and persistence (Allen et al., 2017; Epperson et al., 2021; M. Patel et al., 2021). Butt littering behavior appears to be the norm among smokers in urban settings, with a clear majority of observed smokers littering their cigarette butts even when bins are readily available (V. Patel et al., 2013). In the US, public health concerns have led to restrictions on indoor smoking and movement of smoking to outdoor locations, likely increasing the number of cigarette butts littered. Out of the approximately 6 trillion cigarettes smoked per year worldwide, an estimated 4.5 trillion are littered in the environment (Araújo & Costa, 2019). An estimated 13.13 billion cigarettes were sold in 2018 in California, equating to approximately 3 million kg of cigarette butt waste (T. Novotny, personal communication,

June 26, 2021; Vuong et al., 2019). In a 2012 survey, the majority (74%) of smokers reported having littered cigarette butts at least once in their life, over half (56%) in the month leading up to the time of survey (Rath et al., 2012).

The majority of cigarette filters (~90%) are made of cellulose acetate¹ fibers (Abdul Kadir & Sarani, 2015); alternative filter materials such as charcoal have had little acceptance by US consumers (Granda-Orive et al., 2020; Hoffmann & Hoffmann, 1997). A single filter is composed of greater than 12,000 fibers of cellulose acetate, and when released to the environment, filters break down and release fibers (Belzagui et al., 2020). Acetylated cellulose is persistent in the environment. It is not readily biodegraded by organisms that utilize cellulase enzymes due to its additional acetyl groups that require esterases to break down, and it has limited photodegradability in sunlight (Puls et al., 2011; Yadav & Hakkarainen, 2020). Mechanical forces such as being stepped on or driven over can help cigarette butts break down and release fibers. Even without mechanical means, littered cigarette butts initially decompose fairly quickly, losing about 15% of their mass within the first 30 days, but decompose slowly after that time, with rates depending on environmental factors such as nitrogen availability and microbiome composition (Bonanomi et al., 2020). Without mechanical disturbance or exposure to soil microbiota, cigarette butts show only minor chemical and morphological changes after several years, while cigarette butts may be more quickly transformed after de-acetylation when they are on grassland soil (Bonanomi et al., 2020).

Due to their ubiquity, cigarette butts are likely the main source of cellulose acetate fibers entering urban runoff. However, cellulose acetate is not only used in cigarette filters; it also has been and continues to be used in other textile applications such as in diapers, medical gauze, ribbons, apparel linings, and home furnishings, often blended with other fiber materials (Law, 2004). Therefore, the use and improper disposal of these types of textiles may also be a (likely minor) source of cellulose acetate fibers entering urban runoff. Cellulose acetate is also used in products such as photo films, eyeglasses, and toys (e.g., Lego bricks were made of cellulose acetate until 1963) (Brunning, 2018; Wells, 2015), but these products are not frequently littered and furthermore are not likely to break down into fibers.

2.3. Cigarette Butts and Cellulose Acetate Fibers Urban Runoff Conceptual Model

Figure 2.1 illustrates our conceptual model of cigarette butts and cellulose acetate fibers sources and pathways to urban runoff. The information below provides additional descriptive information.

of substitution of ~2.5.

¹ Cellulose acetate can be produced with a range of degrees of substitution, depending on the desired properties. The degree of substitution refers to the number of substituents per anhydroglucose unit; for cellulose acetate, the maximum degree of substitution is 3, and the cellulose acetate used in cigarette filters usually has a degree

Fate and Transport

In urban settings, cigarette butts are frequently littered onto impervious surfaces such as pavement streets, sidewalks, and parking lots, where they may be more likely to be subjected to mechanical breakdown forces. For example, outdoor public places such as hospitality venues, transportation stops, and entrances to educational venues and playgrounds have high concentrations of cigarette butts (Valiente et al., 2020). In the San Francisco Bay Area, cigarette butts are a frequent litter item in school parking lots (Mock & Hendlin, 2019). Smokers also frequently step on butts to extinguish them, hastening mechanical breakdown. Street sweeping may remove cigarette butts and some cellulose acetate fibers, as street sweeping, when operated slowly enough for optimum performance, can efficiently remove particles above ~125 µm in size (Selbig & Bannerman, 2007). However, most cigarette butts are littered on sidewalks—not roads—where they will not be picked up by street sweeping, and both butts and fibers may be blown or washed off impervious surfaces prior to street sweeping. The ubiquity of cigarette butts found in trash collection surveys conducted on urban roadsides (Moriwaki et al., 2009) and beaches (Allen et al., 2017; T. Novotny et al., 2009) indicates street sweeping is not a sufficient removal mechanism. Urban runoff can transport cigarette butts and cellulose acetate fibers from land into creeks and the ocean.

The distribution of cigarette butt waste in the urban environment is linked to patterns of cigarette sales and consumption (Marah & Novotny, 2011). Urban mapping efforts have identified areas with a broad range of leisure, commercial, business, and tourist activities, and the highest population densities to have correspondingly more cigarette butt pollution (Valiente et al., 2020). Stormwater outlets in industrial and mixed commercial/residential areas have been observed to have higher cigarette butt loads than those in residential areas (Weideman et al., 2020). Greened areas also often form sinks for cigarette butts, due to transport via urban runoff and wind, as well as from smokers clandestinely disposing of their butts in areas "off the beaten [impervious] path" because of negative stigma associated with smoking.

Once in the environment, cigarette butts may be consumed by aquatic animals such as turtles (Macedo et al., 2011) and by terrestrial animals such as dogs and cats (Novotny et al., 2011) and birds (Suárez-Rodríguez et al., 2013) before they have the chance to degrade. Cellulose acetate fibers may also be ingested by a variety of aquatic animals (Miller et al., 2019; Sutton et al., 2019; Wright et al., 2015). If not consumed, cellulose acetate fibers may be entrapped in sediment or be transported to the open ocean (Sutton et al., 2019).

Once cigarette butts enter urban runoff, they may be removed by trash full capture devices, although only items littered upstream of these devices are successfully captured. Multiple California trash surveys, though limited in extent, confirm that cigarette butts are collected in trash capture devices (EOA, Inc., 2014, 2016; Lippner et al., 2020). Of the material collected, cigarette butts consistently represent a large number by count. However, the degree to which captured butts degrade over time and eventually bypass the filters/screens in the trash capture devices designed to allow smaller material and stormwater to pass remains unknown. In California, trash management—via full capture systems and other actions—only addresses macrotrash

larger than 5 mm (State Water Resources Control Board, 2015), and therefore plastics smaller than this size, including cellulose acetate fibers, can readily be transported to water bodies via urban runoff. However, other green stormwater infrastructure, such as bioretention rain gardens, can successfully trap microplastics, including fibers (Gilbreath et al., 2019; Smyth et al., 2021).

Although transport from the ocean via air is an important pathway for other types of fibers (see Chapter 3), air transport of cellulose acetate fibers is expected to be minimal. Cigarette butts can float, but the density of cellulose acetate is 1.28 – 1.32 g/cm³, meaning individual cellulose acetate fibers should sink in non-turbulent water, as they are denser than fresh or estuarine water (which has density of 1.0 – 1.025 g/cm³). Stormwater conveyance systems typically carry turbulent water by design, so settling is unlikely to occur in stormwater channels. In less turbulent waters, higher density particles will settle out relatively quickly, slowing transport and increasing potential for these particles to become sequestered on land or in aquatic sediment (where possible toxicity effects remain unknown). Cellulose acetate fibers are therefore unlikely to be released from the ocean surface to the air.

Waste Management and Reuse

Cigarette butts disposed of properly by smokers via garbage or ash catchers, as well as littered butts that have been removed from the environment, are treated as regular solid waste in California and are generally landfilled or incinerated.

Alternative solutions for the recovery and reuse of materials in cigarette butts are beginning to be explored, such as use in asphalt concrete and fired clay bricks, as a carbon source, sound absorbing material, corrosion inhibitor, and more (Kurmus & Mohajerani, 2020), although these are still generally pilot-scale efforts (Marinello et al., 2020). Companies such as TerraCycle say they are developing ways to recycle cigarette butts into plastic pellets for reuse in other plastic products (Lohan, 2019). However, these efforts result in lower quality and less functional materials than the original products, and the number of butts collected and downcycled is only a tiny fraction of the cigarettes produced and sold, meaning these programs currently have minimal effects in terms of reducing the waste burden of cigarettes. There is also concern that a focus on recycling efforts promotes and greenwashes the tobacco industry (Houghton et al., 2018; Novotny, 2019) by providing misleading information or false impressions that their marketed products are beneficial to the environment.

Depending on their use and disposal, the products produced from cigarette butt recycling efforts may also eventually become a source of cellulose acetate particles entering the environment. Furthermore, depending on the recycling process, toxic chemicals found in cigarette butts may pose additional concerns, as they may remain in recycled and downcycled products or may be released to the environment during production.

2.4. Assumptions

The conceptual model focuses on the most significant urban runoff cigarette butt and cellulose acetate fiber sources and pathways. Assumptions include:

<u>Cigarette butts are the main source of cellulose acetate fibers.</u> The model acknowledges that a small fraction of cellulose acetate fibers are likely from other textile sources, but assumes cigarette butts are the primary source. This assumption is consistent with litter frequency of different items, but does not account for other possible pathways of cellulose acetate fibers from textiles, such as air deposition (e.g., from dryer lint).

<u>California follows global trends</u> in the proportion of smoked cigarette butts that are littered vs. disposed properly. The model assumes that California smokers hold similar beliefs about cigarette pollution and act similarly to smokers in other places. This is a conservative assumption; recent survey data indicate 90% of California adults agree that cigarette butts damage the environment and are poisonous to children, pets, and wildlife (California Tobacco Control Program, 2021), whereas surveys in other places (e.g., Kotz & Kastaun, 2021; M. Patel et al., 2021; Philip Morris International, 2020; Vultaggio, 2021) indicate a larger fraction of the global population do not know about the potential harms associated with cigarette litter.

<u>Use and litter patterns will be relatively similar in all urban areas</u>, such that the results of studies of cigarette butt distribution in urban areas outside of California will be applicable to California urban cigarette butt pollution.

2.5. Data Gaps

Cigarette butt litter is well established as an environmental pollution problem, yet concerns regarding the aquatic toxicity of cigarette butt leachate and microplastic fibers have only recently begun to be explored. While a great deal of information is available regarding litter of cigarette butts, comparatively little is known about cellulose acetate fibers, and development of effective mitigation strategies still requires additional data.

Sources

- How do different environmental factors such as sunlight (see Section 1.3: General Concepts in the Formation of Secondary Microplastics) affect the release of cellulose acetate fibers from littered cigarette butts over time? How long does it take for cellulose acetate fibers to completely degrade? Cigarette butt degradation is generally measured only as loss of mass, which does not distinguish between physical breakdown (resulting in fiber release) and complete degradation.
- How do the littering habits of Californian smokers compare to those of smokers in other urban areas? Is the assumption that different land use categories have different levels of cigarette butt generation onto the watershed valid? Which places are most likely to be a source of cigarette butt litter in California cities?
- What are the litter rates and fiber release potential of other cellulose acetate products? This information is necessary to determine whether cigarette butts are truly the main source of cellulose acetate fibers, and the relative importance of other cellulose acetate sources.

Toxicity

 What are the aquatic toxicity mechanisms and thresholds of concern for cigarette butt leachate and cellulose acetate fibers? Several studies have begun to address these complex questions (Belzagui et al., 2021; Lee & Lee, 2015; Montalvão et al., 2019; Parker & Rayburn, 2017; Slaughter et al., 2011; Wright et al., 2015), but there are not yet clear thresholds for risk assessment.

Environmental Monitoring

 Identification of cellulose acetate using Fourier Transformed Infrared (FTIR) or Raman spectroscopy is currently not very reliable, making it difficult to accurately identify and quantify cellulose acetate fibers in stormwater compared to other similar materials such as cotton (Cai et al., 2019). Can the 2.5 substitution be used to better identify cellulose acetate in microplastics compared to other cellulosic-based fibers? What about tobacco-associated contaminants that may be sorbed to the fibers?

Mitigation

- What are the most effective and economical existing interventions for preventing cigarette butt litter? Additional data on the effects of mitigation measures designed to change human behaviors, such as public space smoking restrictions, litter fees, public information campaigns, expanded waste receptacles, fines for butt littering, and bans on filtered cigarette sales are needed in order to identify and prioritize interventions.
- What are macrotrash littering and microplastic generation rates from vaping (an increasingly common alternative to cigarette smoking)? How do these compare with cigarette butts and what challenges does that present for future trends?
- What are the most effective and economical ways to remove cigarette butts and cellulose acetate fibers from the environment?
- How do environmental cleanup and recycling compare to source reduction (i.e., prevention of cigarette butts being littered) in terms of cost and benefit to the environment/public health?
- Are current cigarette butt recycling efforts environmentally safe (i.e., no release of toxic compounds or microplastics)? Additional information on the hazards of cigarette butt waste is needed in order to predict recycling program costs and environmental and human health risks.
- What regulatory approaches to tobacco products and their marketing might reduce the environmental impact of cigarette butt litter while supporting California's long term public health goal to achieve a smoke-free society by 2035

2.6. Identification of Theoretical Mitigation Measures

There are many available approaches to mitigating cigarette butt and cellulose acetate fiber release, from preventative measures to reduce tobacco use and/or remove toxicand persistent ingredients from cigarettes to downstream approaches to collect and remove cigarette debris on land and water (Figure 2.2). The purpose of this section is tohighlight the broad range of potential mitigation opportunities to effect meaningful change. We identify the actors (e.g., industry, government, non-governmental organizations, and consumers) that would implement each measure, but do not addressimplementation mechanisms (e.g., regulation and oversight), public policy questions (e.g., economic or social costs), or other environmental implications (e.g., greenhouse gas emissions). An in-depth review of the effectiveness of these mitigation measures and trade-offs with other environmental, social, and economic considerations is outside of the scope of this effort.

Prevention

While many people think of smokers as both the cause of the cigarette butt pollution problem and the source of its solution, tobacco companies are responsible for the design and marketing of their products and therefore, under an extended producer responsibility perspective, the full life cycle costs of tobacco product waste (Hoek et al., 2019). Recommendation #5 from the set of actions to address plastic pollution endorsed by the Ocean Protection Council (OPC) is to encourage a statewide prohibition of cigarette filters, pending final conclusions from research commissioned by the Department of Public Health indicating that cigarette filters do not reduce the harm caused by smoking (Ocean Protection Council, 2021). There is general consensus among experts that cigarette filters do not reduce harm from smoking and are falsely advertised (National Cancer Institute, 2005; Song et al., 2017). Removing filters or redesigning them to be biodegradable would likely reduce the cellulose acetate plastic litter into the environment, although this action may encourage smokers to continue to litter cigarette butts (Smith & Novotny, 2011). Cigarette redesign such that they can be extinguished without mechanical force could also reduce fiber release, although this would not reduce release of other toxic contaminants from cigarette butts. While these actions could be legislated, they could also be voluntary on the part of tobacco product manufacturers.

State and local government regulation of cigarette manufacture, sales, and disposal play an important role in current and future cigarette butt pollution mitigation. California's tobacco control strategies to date, including increased cigarette taxes, programs focused on disrupting the social norms that support cigarette smoking, and increased protections against second-hand smoke exposure, have been successful in reducing cigarette sales and per capita consumption, especially when compared with the rest of the US (Pierce et al., 2018). However, these strategies have certainly not eliminated smoking in California. The recently published Online California Adult Tobacco Survey results indicate that while cigarette smoking and vaping has decreased since the outbreak of e-cigarette or vaping product use-associated lung injury (EVALI outbreak) and the COVID-19 pandemic, approximately 10% of adults aged 18 to 64 smoked cigarettes or used other tobacco products in the 30 days prior to taking the survey (California Tobacco Control Program, 2021). Data from the most recent wave of the survey also indicate that 56% of adults were supportive of gradually banning the sale of

cigarettes and 50% agreed that smoking in public places should be illegal. In lieu of bans, policy strategies to make tobacco products less affordable to consumers or less economical to retailers/distributors/manufacturers could help decrease consumption. These strategies include additional taxes or fees on cigarette sales, setting minimum prices for cigarettes, prohibiting redemption of coupons or other discounts for cheaper packs of cigarettes, establishing hazardous waste standards for retailers/distributors/manufacturers of tobacco products, and designating tobacco waste as a public nuisance for which businesses are responsible for the generated waste. It is important to note that any regulatory bans and monetary actions must be enforced consistently to be effective. Fines and fees may disproportionately affect lower-income people and communities, which are often people and communities of color. On the other hand, the moneys collected from these types of control actions may be used to support social programs that most help those at highest risk for smoking-related illness: the poor and minority populations. This complex and nuanced social issue is outside the scope of this review

Reduction of Environmental Release

Surveys of smokers and non-smokers indicate most people understand cigarette butt litter is harmful to the environment, but fewer people know that cigarette filters are made of plastic and are not biodegradable (Epperson et al., 2021; Hoek et al., 2019; Patel et al., 2021), indicating a need for public education. People who know that filters do not make smoking less harmful and have environmental impacts are more likely to support fees or bans on cigarette sales (Patel et al., 2021). Smokers who think cigarette butts are biodegradable and don't "count" as litter are more likely to litter (Rath et al., 2012). Other factors associated with littering behavior include age, the presence of existing litter, and the availability of trash receptacles (Schultz et al., 2013). Fines for violating smoking and littering bans may also help decrease the incidence of butt littering. When asked what mitigation strategies they thought would be most successful at reducing tobacco product waste litter, most people chose changes to product design, fines for littering, and expanded smoke-free spaces (Hoek et al., 2019). However, smokers and non-smokers held different views, with smokers favoring more educational approaches and non-smokers favoring more restrictive policies.

Public education increasing awareness of environmental harms caused by cigarette butts and motivating smokers to properly dispose of butts could take the form of additional labelling on tobacco products (either voluntarily or as a federal requirement by the FDA), or as efforts spearheaded by local governments or grassroots organizations. Local groups may be more effective in inspiring change due to their familiarity with the local community and culture (language, values, etc.). Public education alone is only effective when costs, benefits, and social norms also support changed behaviors. Local efforts to increase the availability of proper disposal receptacles such as public bins and portable ashtrays to make proper butt disposal easier can have a substantial effect on reducing littering (Castaldi et al., 2020). However, the presence of public bins may also unintentionally send the message that smoking is acceptable in that location, and therefore increase the frequency of butt litter (e.g., when people step on their butts to put them out). Additional societal changes to decrease the acceptability of smoking in outdoor public places, such as increasing the

availability of affordable and accessible addiction support/treatment and urban beautification may be required to truly change smoker behavior.

Tobacco companies or local governments could also implement filter take-back or deposit/return programs similar to how "bottle bills" have been used successfully around the world as a way to reduce the hazards, clean-up costs, and waste of discarded glass beverage containers (Novotny et al., 2009). For example, cigarettes could be sold with a "butt deposit" to be refunded when the pack is returned to the vendor with the butts, providing an additional cost incentive for smokers not to litter, and for others to pick up previously littered butts. This type of program could also be paired with butt recycling projects, such as using cellulose acetate from butts as a fiber modifier in concrete (Rahman et al., 2020) to offset costs. The City of San Rafael's "Bounty for Butts" pilot cigarette buyback program, wherein participants received \$1 per ounce of discarded cigarette litter, spurred the collection of over 230,000 cigarette butts over the course of the two-month pilot (San Rafael Volunteers, 2020), indicating this type of program can be an effective tool for reducing butt littering and supporting environmental cleanup.

Remove Litter from Roads and Runoff

Given the high amount of cigarette butt litter, their small size, and their widespread dispersion, collection of cigarette butt litter is costly and often ineffective at reducing pollution when not also paired with preventative mitigation (Torkashvand et al., 2020). Trash collection campaigns are infrequent and participants often focus on larger trash items and leave cigarette butts behind. Street sweeping and trash capture devices may capture some cigarette butts. Bioretention and similar soil media-based filtration-type runoff treatment and infiltration systems have been shown to remove microplastics from urban runoff (Gilbreath et al., 2019; Smyth et al., 2021). However, the feasibility of applying this approach widely may be limited by the high cost of installing highly engineered green stormwater infrastructure. For infiltration, particularly into potential future sources of drinking water, there are also unknowns about the safety implications, considering the many toxic chemicals associated with fibers from cigarette butts.

2.7. Priority Data Needs to Inform Management

Among the many data gaps identified in the conceptual model development process, several have significant implications for future decisions around management of fibers from cigarette butts:

- Improve understanding of the relative efficacy of cigarette butt litter prevention measures: Additional data comparing mitigation measures designed to change human behaviors, such as public space smoking restrictions, litter fees, public information campaigns, expanded waste receptacles, fines for butt littering, and bans on filtered cigarette sales is needed in order to support and prioritize interventions.
- Improve identification of cellulose acetate in the field: Current spectroscopic methods of polymer identification for microplastics do not clearly distinguish between cellulose acetate fibers and natural cellulosic fibers such as cotton. This hinders accurate monitoring of cellulose acetate fibers in the environment,

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decreasing our ability to measure the effectiveness of management actions. Improved methods for differentiating between cellulose and cellulose acetate using infrared or Raman spectroscopy, pyrolysis, or other techniques are needed.



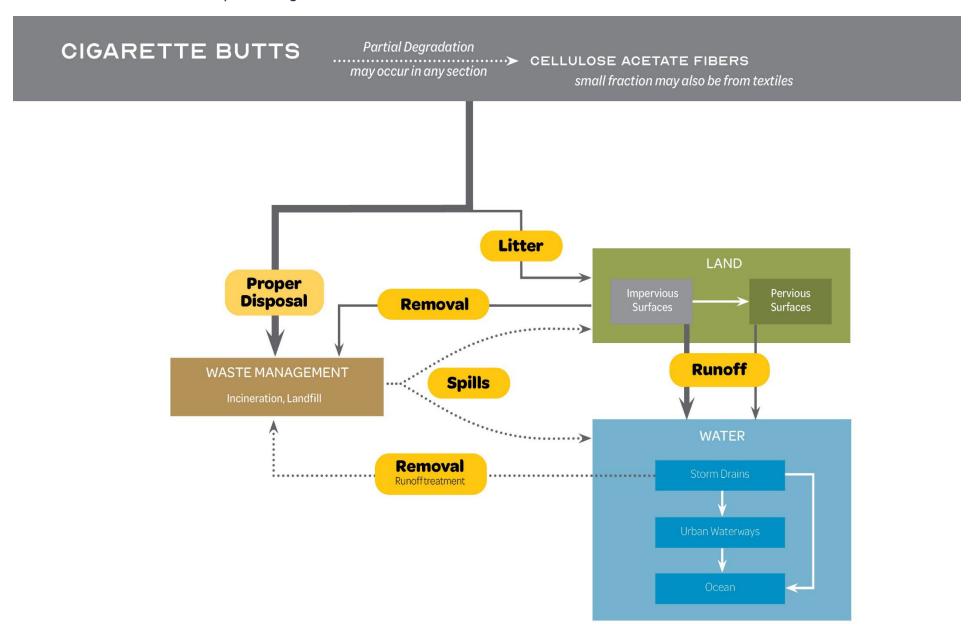


Figure 2.1. Conceptual model of the sources and pathways of cellulose acetate fibers to urban stormwater. The major source of cellulose acetate is most likely cigarette butts, which can themselves be transported via the same pathways. Major pathways are indicated by larger arrows, and dotted arrows represent minor pathways.

MANAGEMENT OPTIONS: CIGARETTE BUTTS **TOBACCO GOVERNMENT POPULATION-WIDE CATEGORY MANUFACTURER OF ACTION ACTIONS ACTIONS ACTION PREVENTION** Legislative and local **Decrease smoking** Cigarette Redesign Remove toxic/persistent bans on sales of (filtered) incidence: Remove filters ingredients or products cigarettes Decrease social Re-formulate filters acceptability Implement filter Legislative and local Provide addiction take-back programs smoking bans (e.g., on beaches or in public places) treatment and support · Change so can be extinguished without Make cigarettes mechanical force less affordable/ economical Taxes and/or fees **Additional labeling** Increase proper butt on cigarette Set minimum prices disposal: Reduce environmental release packaging Prohibit redemption of • Educate smokers about (voluntary or legislatively coupons or discounts butt toxicity/persistence required) Establish hazmat Make proper disposal standards for sellers easier by increasing Set businesses availability of bins and responsible for waste providing smokers with Fines for violating bans portable ashtrays • Reduce other urban litter Implement deposit/ return programs **Street sweeping Increase of** Trash capture cleanup frequency devices and efficacy of trash collection Collect and remove debris Improved runoff campaigns from roads and runoff treatment REMEDIATION Dashed line around indicates All others (no lines) indicate Novel/Not Widely Implemented Already widely implemented

Figure 2.2. Management options for cigarette butt and cellulose acetate fiber pollution, framed on a scale from preventative (reducing use and release) to remediative (collection and removal) measures. Options are color-coded by who would be responsible for implementation, with industry in orange, government in dark blue, and community-wide (non-governmental organizations) in light blue. The current frequency of implementation of each management option is indicated by the box borders, with dashed lines indicating novel or not widely implemented actions and no lines indicating actions that have already been widely implemented.

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3. Fibers in Urban Runoff

3.1. Background

Fibers are a category of microplastics that are distinguished from other microplastics based on the particle morphology and dimensions. Fibers have a long, narrow thread-like shape, significantly longer in one dimension than in the other two dimensions. A standard definition of fibers in the environment has not been adopted widely yet. A major challenge for defining fibers is that scientists and water quality regulators do not sufficiently understand how the size, dimensions, and type of polymer influence the toxicity of fibers. This document uses the word "fibers" instead of "microfibers" to avoid inconsistency with the terminology used by some members of the textiles industry to refer to very fine synthetic material threads used to achieve desired material characteristics.

Various definitions of microplastic fibers have been proposed. Human-made or human-modified polymer fibers less than 5 mm in length would be considered microplastics according to the California definition of microplastics developed for drinking water (California State Water Resources Control Board Resolution 2020-0021, 2020). The European Chemicals Agency (ECHA) has proposed to include longer fibers (up to 15 mm) in its microplastics definition (European Chemicals Agency, 2019). The California Code of Regulations recognizes that particle size and dimensions are important factors in human health risks due to inhalation risk, and that fibers have long aspect ratios compared to particles. With the stated goal of protecting human health from inhalation risks to fibers, California regulations define fibers (not specific to microplastic fibers) as particles having an aspect ratio (length to width) of 3:1 and a width less than or equal to 3 micrometers (California Code of Regulations Title 22, Section 69405.71). Textile industry and environmental scientists have proposed a general 100:1 length to width aspect ratio for defining fibers (J. Liu et al., 2019) in the context of textile fibers and microplastics monitoring in the environment. Both State Water Resources Control Board and ECHA definitions of microplastics exclude fibers from unmodified (except by hydrolysis) natural fibers (e.g., unprocessed wool fibers), but include chemically modified natural fibers (with greater than 1% synthetic polymer content) because their toxicological properties are likely altered (California State Water Resources Control Board Resolution 2020-0021, 2020; Hartmann et al., 2019). In 2020, Congress passed the Save Our Seas Act 2.0, which specifically requires a report on fibers that provides a standard definition for fibers, and requires federal agencies to develop a five-year plan on how they can collaborate to reduce fiber pollution. This report is currently under development by Materevolve², the U.S. Environmental Protection Agency, and the Interagency Marine Debris Coordinating Committee, and is due to Congress in 2022.

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¹ https://www.law.cornell.edu/regulations/california/22-CCR-Sec-69405-7

² https://www.materevolve.com

As noted previously, an investigation of urban stormwater runoff in the San Francisco Bay region found urban runoff microparticle concentrations (1 – 30 microparticles/L) to be significantly higher than wastewater microparticle concentrations (0.008 – 0.2 microparticles/L) (Sutton et al., 2019). Fibers were the second most common particle type in urban runoff, composing 39% of all particle counts in sampled urban runoff. Prior to Sutton et al.'s (2019) finding that urban runoff appears to be the major source of fibers in San Francisco Bay, fibers had generally been assumed to reach surface waters primarily via municipal wastewater effluent (e.g., from washing clothing) (e.g., Gavigan et al. 2020; Browne et al. 2011; Hartline et al. 2016).

Spectroscopic techniques applied to a subset of the fibers in urban stormwater revealed the two most common fiber polymers identified were polyester (polyethylene terephthalate) and cellulose acetate (discussed in Section 2) (Sutton et al. 2019). However, because only a subset of all collected fibers could be assessed, and the majority of fibers were not readily identifiable via spectroscopy (i.e., > 50% of fibers were classified as "anthropogenic unknown," primarily due to the presence of dyes that masked the identifying characteristics of the underlying material), the distribution of specific plastic polymers may not be representative of overall presence in stormwater samples.

We hypothesize that these findings about the importance of fibers in urban runoff can be extrapolated to other densely urban areas in California. The San Francisco Bay findings suggest that outdoor sources and emissions of fibers may be more significant sources of fibers to the coastal ocean environment compared to fibers from laundering and the wastewater pathway. Fibers can also contain many chemical additives, including dyes, flame retardants, plasticizers, antimicrobials, surfactants, and poly- and perfluoroalkyl substances, which may be toxic to humans and aquatic and terrestrial wildlife.

The purpose of the fibers conceptual model (Figure 3.1) is to synthesize and integrate our understanding of the sources and pathways of fibers entering urban runoff. This model identifies many possible sources of fibers. Because there is limited information about the emission rates of fibers from fiber sources, we cannot yet identify the dominant sources of fibers. This model does not address cellulose acetate from cigarette butts, which is discussed in a separate conceptual model (Section 2). The general fibers conceptual model is meant to inform future research and management recommendations for managing microplastic fiber pollution.

3.2. Sources of Fibers in Urban Runoff

Fibers have multiple uses in urban environments. In addition to use in household and industrial textiles (clothing, carpets, upholstery, tarps, awnings, tents, storm drain filter fabric, erosion control blankets, and other geotextiles), fibers fill furniture, toys, diapers, and pillows; compose artificial hair; form cordage (rope, cords, and twine); insulate buildings; and provide structural support for a plethora of outdoor products, such as composite building materials, construction and landscape materials and their transport bags, as well as belts beneath the rubber surface of vehicle tires.

Fibers occur in diverse urban outdoor surface coverings such as carpet, artificial turf, and landscape fabrics. Cordage has outdoor uses in construction, gardening, and surveying. Nonwoven textiles, often found in industrial materials, are also used in surgical masks, an all-too-common element of outdoor trash during the current COVID-19 pandemic. Large plastic items made of nylon and high-density polyethylene (HPDE) that are not textiles may also form fibers (and microplastic fragments) through environmental degradation (Naik et al., 2020). The material characteristics of the plastic polymer are important to determining the formation of fiber versus fragment microplastics (Naik et al., 2020).

At present, it is not possible to ascertain which sources are the predominant contributors of fibers to urban runoff. As noted previously, prior investigations of microplastic fibers have focused primarily on the release of fibers to the wastewater pathway via laundering of clothing and indoor textiles. Very little work has been done to characterize microplastics in urban runoff, particularly in locations where stormwater is channeled directly to surface receiving waters without treatment. Below, we discuss in detail a comprehensive list of potential contributors of microplastic fibers to urban runoff.

Non-Industrial Fibers

Wear, washing, drying, and disposal of clothing and other non-industrial goods release fibers into the environment. For textiles, fiber release rates depend on the nature of the textile product (e.g., filament type; whether it has a mechanically processed surface), manufacturing processes used in textile and garment construction (particularly the textile cutting process), washing conditions, and textile age (Y. Cai et al. 2020; De Falco et al. 2020; Almroth et al. 2018; Palacios-Mateo et al. 2021). For other types of common fiber-containing goods that are infrequently washed, such as carpets and fiber fill (e.g., furniture, pillows), fiber releases are likely primarily due to physical contact with the item (abrasion, mechanical degradation, cleaning).

A more limited range of fiber-containing non-industrial goods commonly appear in outdoor urban environments. These include carpet, artificial turf, outdoor furniture, netting, swimming pool and outdoor furniture covers, tarps, tents, and boat and vehicle covers. In those rare urban locations where fishing occurs, fishing gear (ropes, nets, and line) may be used. In areas with unsheltered populations, additional fiber containing non-industrial goods may be located outdoors, such as items that sheltered populations normally use or store indoors (e.g., clothing) and items that are not normally used long-term in urban areas (e.g., tarps, tents). These items can release fibers directly into the outdoor environment, facilitating transfer into urban runoff. Outdoor fiber releases may occur as a result of degradation as well as physically contacting or moving the item.

Because most fiber-containing goods are located indoors, most fiber releases likely occur indoors. While most fibers used or released to air indoors will remain indoors, they can be tracked or blown outdoors, or released during material relocation or disposal. Fiber concentrations in air are lower outdoors than indoors (Dris et al., 2017; C. Liu et al., 2019; Y. Zhang et al., 2020). Ventilation, such as through open windows and exhaust fans, from homes and commercial buildings can be a significant source of particles in outdoor air (Björklund et al., 2012; Sundt et al., 2014)

Textiles—and specifically clothing washing—have been a focus of research on environmental fiber releases. In the US, clothing wash water drains into municipal sewer systems or septic tanks, making it unlikely to transfer fibers into urban runoff. Fiber losses into the air while wearing garments may rival the losses during washing (De Falco et al., 2020); however, since people spend most of their time indoors (< 10% outdoors according to Klepeis et al. 2001), despite examples of clothing and camping fabric-related fiber emissions at remote locations (e.g., Napper et al. 2020), most of the fiber air emissions from wearing clothing likely remain indoors.

In contrast to clothing washing, clothing drying is a potentially significant source of fibers in urban runoff. In the US and Canada, dryer use is significantly higher than in other parts of the world (Kapp & Miller, 2020). The use of mechanical air dryers with outside ventilation in the US contrasts with practices in many other parts of the world, where unvented condenser dryers and hanging items to dry are more common (Energy Star, 2011). Much of the scientific literature around fibers in the outdoor environment is from areas where outdoor-vented mechanical air dryers are less common than in the US. Most US residential and commercial clothing dryers vent directly to the outdoors without treatment, dispersing fibers not collected in lint traps to the outdoors. Significant quantities of fibers are known to pass through dryer lint filters, as evidenced by the US Federal Emergency Management Agency finding that lint buildup in dryer lint traps and exhaust ducting are the primary cause of residential dryer fires (Federal Emergency Management Agency (FEMA), 2012). This outdoor fiber emission source has received surprisingly little investigation. Based on fibers collected in a condensing dryer lint trap, Pirc et al. (2016) estimated that fiber releases during tumble drying of clothing were 3.5 times higher than releases during washing. This study did not measure fibers passing through the lint trap and exhausted to the outdoors. In a small study, Kapp and Miller (2020) measured emissions from tumble drying pink polyester fleece blanks in typical North American mechanical dryers with outdoor exhausts and found that the mass of fibers collected outdoors rivaled the amount collected in the two tested dryers' lint traps.

Industrial Fibers

The diverse industrial uses of fibers may release fibers into the outdoor environment (e.g., air emissions, ground deposition) from manufacturing facilities, product use, weathering, and disposal. Unlike clothing, many industrial textiles are nonwoven, with different fiber characteristics (Martínez Silva & Nanny, 2020). Examples of the long list of industrial and construction uses of fibers (Adanur, 2017; Paul, 2019) that are notable from the urban runoff perspective include:

- Geotextiles Filtering and other textiles used for construction runoff treatment, silt fences, erosion control, weed control, bank and coastal stabilization, and drainage systems may release fibers when cut, mechanically abraded, or through degradation. Geotextile fibers can contain persistent and toxic additives (Wiewel & Lamoree, 2016).
- <u>Construction and landscaping material packaging</u> Fiber-reinforced bags, particularly those used for construction and landscape material and for flood protection (plastic-fiber sand bags), may release fibers when cut open, mechanically abraded, or left outdoors to decay.

- Ropes, string, and twine Construction, landscaping, agriculture, and fishing may release fibers into the environment when cut or during use. Some items may be left to degrade (e.g., in landscaping), from which fibers may be released into runoff. Plastic string used in vegetable production, which is similar to string used in landscaping, might be a significant source of fibers in soil (G. S. Zhang & Liu, 2018).
- Outdoor architectural fabrics Roofing, awnings, and temporary fence covers may release fibers through degradation.
- Vehicle parts Parts such as brake pads and mechanical equipment belts may release fibers to the air during vehicle operation.
- Concrete Fibers may be added to concrete to address cracking and to improve structural properties, particularly with the newer "foam concrete" technology (Amran et al., 2020; Shafei et al., 2021). In addition to natural fibers, metal, glass, and polymer fibers (e.g., polyester, acrylic, aramid, polyvinyl alcohol, polyethylene, polypropylene, nylon) may be added to concrete. Due to its desirable properties and low cost, polypropylene is reportedly the most commonly used fiber type (Shafei et al., 2021). Construction operations such as concrete cutting, building demolition, and concrete recycling from building demolition debris could release fibers into outdoor air.
- <u>Building wraps and insulation</u> Modern wood building construction often includes a permeable textile membrane on the outside of the wood below the building siding. Building insulation may contain various fibers, including recycled textiles (Islam & Bhat, 2019). On-site cutting of materials (e.g., cutting window and door openings in building wraps) may release fibers to outdoor air.

Litter

Examples of litter items that can release fibers into urban runoff include:

- <u>Macroplastics that degrade into fibers</u> Depending on the manufacturing processes, macroplastics in the outdoor environment could potentially degrade into fibers. Naik et al. (2020) discovered that nylon and high density polyethylene pellets, which are used in plastic product manufacturing, degraded into microfibers under UV light. This study did not examine end-use products produced with the pellets (Naik et al., 2020).
- Fiber-containing items abandoned outdoors Dumping of upholstered furniture, mattresses, carpet, tarps, and other textiles outdoors may contribute fibers to runoff, particularly if abandoned items are not collected and managed prior to their degradation. Fiber-containing items are often abandoned in areas where unsheltered populations camp (these areas typically lack waste collection services) (Bay Area Stormwater Management Agencies Association, 2020). Disposable diapers containing fibers are rare in California litter (Bay Area Stormwater Management Agencies Association, 2020; Miller-Cassman et al., 2016; Moore, 2016).

Personal Protective Equipment - Fibers provide structural support for many medical goods, most of which typically remain indoors. Due to the current COVID-19 pandemic, medical masks (e.g., surgical masks) and cleaning wipes composed of nonwoven fibers have become common outdoor litter items (Ammendolia et al., 2021; Prata et al., 2020; Wilson, 2020). Synthetic polymers, such as polyethylene terephthalate (PET) and polypropylene (PP), have been identified as components of wet wipes (Ó Briain et al., 2020).

3.3. Fibers in Urban Runoff Conceptual Model

Figure 3.1 illustrates our conceptual model of indoor and outdoor fibers sources and pathways to urban runoff. The information below provides additional descriptive information.

As illustrated in the top two boxes in the conceptual model, fibers can be released into the outdoor environment from both indoor and outdoor uses:

- (1) <u>Indoors Transport from indoors to outdoors</u>. For example, emissions from personal, laundromat, or industrial clothing dryers; industrial facility emissions; carpet fibers tracked outdoors; shaking out rugs outdoors to clean them; fibers in indoor air transferring to the outdoors through open windows and building ventilation exhausts.
- (2) Outdoors Loss of fibers during installation, use, and storage. For example, fibers lost from clothing while outdoors; wear of tents and tarps; runoff from awnings; wear from walking on outdoor carpet or playing sports on artificial turf; failure of fiber-containing building materials; driving over fiber-based bags; weather damage to fabric fence coverings; cutting fiber-containing building wraps and insulation; cutting roofing fabric; cutting and installing twine landscaping supports; using storm drain filter fabric.

Fate and Transport

After release to outdoor environments, fibers may deposit on impervious or pervious surfaces, or be washed out of the air by rainfall. Deposited particles—particularly those deposited on impervious surfaces—may be resuspended, redistributed, and modified by vehicles or other human activity, or simply by the motion of the air. Street sweeping, which can efficiently remove particles above ~125 µm in size (Selbig & Bannerman, 2007), may remove larger fibers from gutters and streets. Some fibers may be sequestered in soils and landscape covers.

While in the outdoor urban environment, processes such as photodegradation, hydrolysis, biological degradation, and mechanical degradation can modify fibers (Sait et al., 2021; Sørensen et al., 2021; Zambrano et al., 2019). Degradation rates depend on both the environmental exposure situation and the fiber material (Sait et al., 2021; Zambrano et al., 2019). In addition to releasing fibers and reducing fiber sizes, degradation in the urban environment has potential to release chemical additives within fibers to the environment (Sait et al., 2021; Sørensen et al., 2021; Wiewel & Lamoree, 2016).

Due in part to their high surface area to volume ratio, fibers emitted to the outdoor air can transport through the air for long distances, as evidenced by detections in remote global locations (e.g., the Arctic, mountain wilderness) (Allen et al., 2019; Athey et al., 2020; Brahney et al., 2020). Re-emission of previously deposited particles from the ocean surface can be a significant source of deposition on land (Allen et al., 2020). Fibers typically (but not always) dominate microplastic air deposition measurements made using traditional air quality research methods (e.g., elevated passive collection platforms away from known local emissions sources) (Allen et al. 2019; L. Cai et al. 2017; S. L. Wright et al. 2020; Brahney et al. 2020; Dris et al. 2018; Y. Zhang et al. 2020). Available information is insufficient to determine the source of these fibers, though exhaust from clothing dryers seems likely to contribute (Kapp & Miller, 2020).

In urban environments, rainfall and runoff wash particles into stormwater collection systems. Washoff from impervious surfaces (streets, sidewalks, roofs) is far more efficient than from pervious surfaces (lawns, gardens, agricultural fields) (Field et al., 2000; Water Environment Federation & American Society of Civil Engineers, 1998). Biofouling, which is the growth and accumulation of microorganisms on surfaces (which modifies density, aids agglomeration, and facilitates loss of chemicals contained in microplastics) likely plays an important role in the transport and fate of fibers in the environment, in addition to physical factors like wind and water flows (Barrows et al., 2018; Rummel et al., 2017). In the limited locations where urban runoff receives treatment, fibers may be removed, particularly in systems that filter runoff like bioretention treatment systems (Gilbreath et al., 2019; Smyth et al., 2021). While denser fibers may be temporarily retained in low points in the stormwater collection system under low flow conditions, turbulent flows during larger storm events will likely mobilize these particles and carry them into surface waters (Hoellein et al., 2019).

Waste Management and Reuse

Unwanted fiber-containing products may be disposed with solid waste or reused, such as through clothing resellers and textile recyclers. Some handling and processing of waste and materials for reuse may occur outdoors, but in California, most operations are indoors (e.g., solid waste transfer stations, clothing sorting facilities, clothing resellers) to protect environmental quality or to protect the quality of the textiles for potential reuse. If landfilled, waste fiber-containing products can degrade to produce fibers.

Fibers may be released into the environment during the waste management process, such as via dispersal of fiber fill material from furniture while awaiting transport to a waste management facility, losses in waste collection (e.g., dumpsters) and transport if not properly covered, emptying vacuum cleaning equipment, building demolition, crushing fiber-containing concrete for recycling, and vehicle crushing. As discussed above, improper disposal (litter) also releases fibers into the environment.

A few reuse options for waste fiber products could release fibers into the environment. For example, use of waste carpet to line drainage systems has been proposed and simulated in laboratory experiments implemented in pilot projects (J. Wright, 2019), but has not been implemented.

Within California, once fiber-containing wastes (including biosolids) reach landfills, opportunities for transport of disposed fibers into urban runoff are relatively limited due to California requirements for daily cover of landfilled waste, secondary containment, and leachate collection and treatment systems (California Code of Regulations Titles 14 and 27). Activities at landfills, such as depositing waste in the landfill and compacting waste prior to application of daily cover, could potentially release fibers into the air. Outside of California, particularly in areas with fewer environmental protection regulations, disposal activities can be far less regulated and could be a potentially significant source of fiber emissions to air and to runoff (e.g., Palacios-Mateo et al. 2021).

3.4. Assumptions

The model development process assumes that differences in sampling and analyzing fibers from different studies do not generally affect the identification of sources and pathways for fiber transport in urban runoff. Researchers use differing methods for collecting, identifying, and even defining microplastics, which create inconsistencies in data. While fiber contamination is common in environmental samples (Scopetani et al. 2020), this limits the comparability of data from different locations and studies.

A major hypothesis in this conceptual model is that mechanical air clothing dryers with outdoor ventilation may be a significant source of fibers in urban runoff. As discussed above, few studies have investigated this fiber source and pathway to urban runoff. This is a priority data gap as discussed below in Section 3.7: Priority Data Needs to Inform Management.

The conceptual model focuses on potentially significant urban runoff fiber sources and pathways. Examples of excluded potential sources and pathways are described below.

Indoor fiber uses/wastewater discharges are excluded from the conceptual model. The conceptual model assumes that municipal urban runoff does not contain meaningful quantities of water that was originally discharged from indoor drains to the municipal wastewater collection (sewer) system. In modern urban construction (which occurs in all portions of California except San Francisco and part of Sacramento), outdoor drains flow through drainage structures like curbs and gutters to separate storm drains that discharge directly to surface water, usually without any treatment. While some cross connections exist—and exfiltration from sewer lines can enter storm drains—these sewage flows typically compose a negligible portion of urban runoff flow in most of California.

<u>Sewer overflows are excluded from the conceptual model.</u> Sewer overflows do occur and likely compose an irregular source of fiber discharges to surface water. Sampling locations in the San Francisco Bay regional study avoided areas where sewer overflows commonly occur. Given the extensive separate efforts to control sewer overflows, this source is excluded from the conceptual model.

Agricultural runoff is excluded from the conceptual model. This conceptual model focuses on urban runoff and, therefore, agricultural runoff is outside the model scope. Some agricultural land use may be present in urban settings, but is assumed to not be a significant portion of urban land use and flows to the storm drain system. Agricultural fiber sources, such as textile drainage systems, crop and erosion prevention geotextiles, shade fabric, bird netting, plastic string, and weed prevention, may release fibers when cut, mechanically abraded, or through degradation. Runoff from biosolids treated fields may contain fibers removed from wastewater during treatment (Nizzetto et al. 2016; Gavigan et al. 2020; L. Zhang et al. 2020; G. S. Zhang and Liu 2018).

<u>Fiberglass</u>, carbon fiber, glass, metal, and ceramic sources are excluded from the conceptual model. These materials are not included within the definition of microplastics.

<u>Vehicle tires are excluded from the conceptual model</u>. Fibers in belts are covered by tread. While "bald" tires occur on the road, these are uncommon due to failure risks. Therefore, vehicle tires are not expected to contribute significantly to fiber pollution. (However, tires are a significant contributor to microplastic pollution - see Section 5: Tires in Urban Runoff)

<u>Asphalt is excluded from the conceptual model</u>. While fibers (like polypropylene and polyester fibers) can be used in asphalt mixes, this use was not reported for California (McDaniel et al., 2015).

3.5. Data Gaps

Sources

- What are outdoor emission rates of fibers from clothing dryers in California?
- Are outdoor industrial materials a major source of microplastic fibers in urban runoff?
- Compared to other urban runoff fiber sources, what is the relative importance of deposition of fibers from long-range transport via air and from the ocean into coastal air?
- Can physical or chemical characteristics of fibers be used to more specifically identify their sources?
- Are methods available that can improve and facilitate identification of fiber sources in urban runoff?

Aquatic Toxicity/Chemistry

- Do chemicals incorporated into fibers contribute to their aquatic toxicity?
- Are natural fibers (with and without chemical modification) less toxic than synthetic fibers?

Environmental Monitoring

- How can scientists and water quality managers develop a consistent terminology and framework for measuring and reporting microplastic fibers so that results can be compared across studies? Should natural fibers (with chemical modifications or additives) be included in environmental microplastic monitoring studies?
- What are the environmental concentrations of fibers and fiber-related chemicals and particles?
- How do runoff and surface water fiber concentrations relate to watershed characteristics?

Fate and Transport

- How does the fate (particularly the persistence) of natural fibers (with and without chemical modification) compare to that of synthetic fibers?
- What is the wash-off fraction of fibers deposited on impervious and pervious surfaces that gets transported to receiving waters by urban runoff?

3.6. Identification of Theoretical Mitigation Measures

A significant challenge with addressing fiber pollution is that, apart from cellulose acetate fibers derived from cigarette butts (Section 2), the major sources and pathways of fibers are not understood, and there is a long list of potential sources. Fibers from clothing and other textiles were identified in the general fibers conceptual model (Figure 3.1) as a potentially important source of fiber emissions to urban runoff; therefore, we chose to briefly outline theoretical mitigation approaches addressing these sources (Figure 3.2). The focus of this section is specifically on fibers and associated chemicals released while consumers are using and maintaining apparel and other textiles; however, some measures address industrial textiles and non-textile fibers as well. Fibers and chemicals released during manufacture are excluded. Further investigations to identify other major sources of fibers, as mentioned in Section 3.5: Data Gaps, can guide next steps in developing mitigation measures for other sources.

There are many available approaches for mitigating fibers and fiber-related chemical environmental releases, from preventive measures to reduce shedding rates from clothing and remove toxic ingredients, to reducing fiber released to the environment, to capturing fibers and fiber-related chemicals in runoff (Figure 3.2). The purpose of this section is to highlight the broad range of potential mitigation opportunities. It identifies the actors (e.g., industry, government, and consumers) that would implement each measure, but does not address the implementation mechanisms (e.g., voluntary/mandatory or financial incentive/penalty) or public policy questions (e.g., economic or social costs), nor does it consider other environmental implications (e.g., greenhouse gas emissions). For example, reducing the demand and supply of new apparel could help mitigate the environmental impacts from manufacturing and disposal, but this approach is not included in Figure 3.2 because there is not sufficient evidence as to whether this will reduce fiber emission during the typical use of the product.

Public awareness about textiles as a major source of microplastic pollution, as well as state and federal legislation on microplastics, has already brought representatives from the apparel and textile industry, government agencies, and environmental organizations together to discuss control strategies for fiber pollution (e.g., Save Our Seas Act 2.0¹, California Assembly Bill 2379 Bloom (2018)², California Assembly Bill 129 Bloom (2020)³).

Prevention - Remove Toxic Ingredients

Chemicals are added to textiles to impart desired characteristics in the finished product or are used in the manufacturing process. Many of these chemicals are toxic to humans and wildlife. These chemicals can be released from the clothing during use and washing. For example, per- and polyfluoroalkyl substances (PFAS) used to make clothing water and stain resistant may leach into laundry water (CEC, 2017). There is growing evidence that PFAS are persistent, bioaccumulative, and toxic in the environment. Previous examples of hazardous chemicals being replaced or removed in textiles and apparel exist. For example, the use of phthalates in child apparel is restricted by regulations in the US, European Union, and South Korea.⁴ While the textile and apparel industries must be the primary actors in removing toxic ingredients, government regulations and the threat of government regulations are significant drivers for industry changes.

Currently, apparel brands utilize restricted substance lists (RSLs) and manufacturing restricted substance lists (MRSLs) maintained by industry collaborations^{5,6} to manage chemicals used in their supply chain and final products. RSLs and MRSLs comprehensively list the materials, chemicals, and substances that are restricted or banned in finished home textiles, apparel, and footwear products due to regulation or law. Managing and implementing MSRLs and RSLs is complex and challenging because of the many businesses and operations in the supply chain; this includes textile manufacturers and processors, textile chemistry experts, dye and finish manufacturers, fiber manufacturers, materials innovation companies, and apparel manufacturers, as well as brand leaders.

Prevention - Reduce Shedding Rates

The apparel and textile industry can design and manufacture textile products to shed fewer fibers during their use. Most scientific (Y. Cai et al., 2020; De Falco et al., 2019; Hernandez et al., 2017; Pirc et al., 2016) and industry research⁷ on fiber shedding has been focused on measuring shedding rates during washing. The apparel industry is developing a globally-recognized standardized method to measure fiber release during washing, which is expected to support innovations in product design to reduce shedding rates.⁸ There are currently no standardized industry methods for measuring fibers

¹ https://www.congress.gov/bill/116th-congress/senate-bill/1982/text

² https://leginfo.legislature.ca.gov/faces/billTextClient.xhtml?bill_id=201720180AB2379

³ https://leginfo.legislature.ca.gov/faces/billTextClient.xhtml?bill_id=201920200AB129

⁴ https://www.aafaglobal.org/AAFA/Solutions_Pages/Restricted_Substance_List.aspx

⁵ https://www.aafaglobal.org/AAFA/Solutions_Pages/Restricted_Substance_List.aspx

⁶ https://www.roadmaptozero.com/about

⁷ https://www.patagonia.com/stories/an-update-on-microfiber-pollution/story-31370.html

⁸ https://www.microfibreconsortium.com/work

released during drying and wearing. Once standardized methods are established for measuring shedding rates from washing, drying, and wearing, it may be reasonable for government agencies and industry players to develop quality standards that specify a maximum allowable shedding rate for apparel and other textiles sold in the market.

Reduce Fiber Releases

Reducing textile washing frequency and modifying textile drying practices, such as by hanging textiles to dry or using non-vented condenser dryers, can reduce fiber release from textiles (and may increase textile lifetime). Encouraging consumers to reduce laundering and drying should weigh impacts of potential alternatives to avoid regrettable substitutions, such as increased use of antimicrobial-treated textiles, which can release more chemicals to the environment.

Public education increasing awareness of environmental harms caused by fiber pollution and individual strategies to reduce emissions could take the form of additional labelling on clothing or education programs. Education programs could be led by the apparel industry, local government, or environmental organizations. Public education alone is only effective when costs, benefits, and social norms also support changed behaviors.

Government has a role to support these changes in the public's behavior, such as by providing economic incentives to switch to condenser dryers and removing legal barriers to hang-drying clothing. California law generally allows people to dry their clothes outdoors⁹, but there may be restrictions from landlords or homeowners' associations, or this practice may not be socially acceptable.

Extended producer responsibility of textiles and reduction in supply and demand of new clothing may also reduce total fibers emissions when considering the whole life cycle of the product; it is unclear whether this would reduce fiber emissions during the use of the product.

Reduce Outdoor Emissions

Capturing lint fibers generated during apparel drying before they are released to the outdoors could also reduce outdoor emissions. Dryer emissions could be reduced by improving lint removal by dryers (e.g., improved lint screens) or by capturing fibers at the indoor dryer exhaust connection or outdoor building exhaust point. All lint capture systems would need to be carefully installed and maintained (e.g., cleaned) to maintain sufficient air flow for drying operations and avoid build-up of lint that can present a fire hazard. Captured fibers would require proper disposal.

In the commercial sector, multiple vendors offer lint emissions control equipment designed for large facilities (e.g., industrial or commercial laundries), including some with high lint removal efficiency (>95% removal). 10 This equipment may be at the

 $^{^9\} https://leginfo.legislature.ca.gov/faces/billTextClient.xhtml?bill_id=201520160AB1448$ ¹⁰ E.g., Clean Cycle (http://www.cleancyclesystems.com/) and Energenics Corporation (https://www.energenics.com/) advertises exhaust lint filters for commercial applications.

building exhaust point (where it functions like other air pollution emissions control equipment) or within the building ducting. This type of equipment may be installed for air pollution compliance purposes, to prevent lint build-up in exhaust or recirculated air, or for aesthetic purposes. This type of equipment is not suitable for residences due to its design and because it requires ongoing professional management to ensure adequate air flow and prevent potentially flammable lint accumulation. Government regulations or incentives may be important to support adoption at centralized laundry facilities that are not currently subject to air pollution control regulation (e.g., laundromats and hotels).

While various new filters and products are being sold to reduce fiber emissions from residential washing machines (McIlwraith et al., 2019), to our knowledge, such products have not been developed for residential dryers. In residential settings, scientists measuring fiber emissions have installed simple filter fabric bags over exhaust vents. However, capturing lint more effectively within a dryer or in connection with residential building exhaust systems has the potential to restrict dryer exhaust air flow and/or build up flammable lint, which could pose a fire hazard. These issues would need to be evaluated to determine the feasibility of these approaches to capturing fibers from dryers in residential settings.

Remove Fibers from Runoff

Bioretention and similar soil media-based filtration-type runoff treatment and infiltration systems have been shown to remove small particles like fibers from runoff (Gilbreath et al., 2019; Smyth et al., 2021). However, feasibility of applying this approach widely may be limited by the high cost of installing highly engineered green stormwater infrastructure. For infiltration, particularly into potential future sources of drinking water, there may be questions about the safety implications of chemicals in fibers. Street sweeping may have limited efficacy in removing smaller fibers (Selbig & Bannerman, 2007).

3.7. Priority Data Needs to Inform Management

Among the many data gaps identified as part of the conceptual model development process, several have significant implications for future decisions around management of fibers:

- Quantify clothing dryer exhaust fiber emission and deposition: Quantifying emissions will determine whether fibers from clothing dryers are a priority for mitigation. Dryer exhausts propel fibers into the air, potentially facilitating both short-range and long-range transport. Priority data needs include: (1) quantifying fiber emission rates from clothing dryers (household, laundromat, industrial laundry), (2) estimating the fraction of dryer fiber emissions deposited locally (i.e., within watershed and potentially contributing to local runoff), and (3) estimating the fraction of dryer fiber emissions subject to long-range transport (i.e., outside of watershed and potentially contributing to runoff in downwind watersheds).
- Measure fiber air deposition from local and long-range sources: Air deposition measurements in remote locations can clarify the relative importance of

long-range fiber transport (e.g., carried in air currents from Asia and re-emitted from the ocean into coastal air) as compared to local sources of fibers in urban runoff. Air deposition measurements in near-source locations can reveal the potential significance of air-emitted local fiber sources (like outdoor industrial fibers used in building construction).

- Improved understanding of toxicity and fate of natural fibers compared to synthetic fibers: Some industry and environmental organizations already advocate switching to natural textiles to reduce microplastic fiber pollution. However, there is not yet sufficient scientific understanding as to whether natural fibers are less toxic compared to synthetic fibers. Research to understand how fibers from natural materials (e.g., cotton and wool) compare to other synthetic materials in persistence and toxicity to wildlife would clarify the effectiveness of mitigation measures substituting natural fibers for synthetic ones.
- Standardized terminology and framework for monitoring microplastic fibers: The
 current lack of standardized approaches to measure and report fibers in the
 environment make it challenging to compare studies. What sizes, polymers, and
 materials should be included and how should results be reported? Should natural
 fibers with chemical modifications or additives be included? The definition should
 be informed by improved understanding of the toxicity and fate of natural fibers
 compared to synthetic fibers.



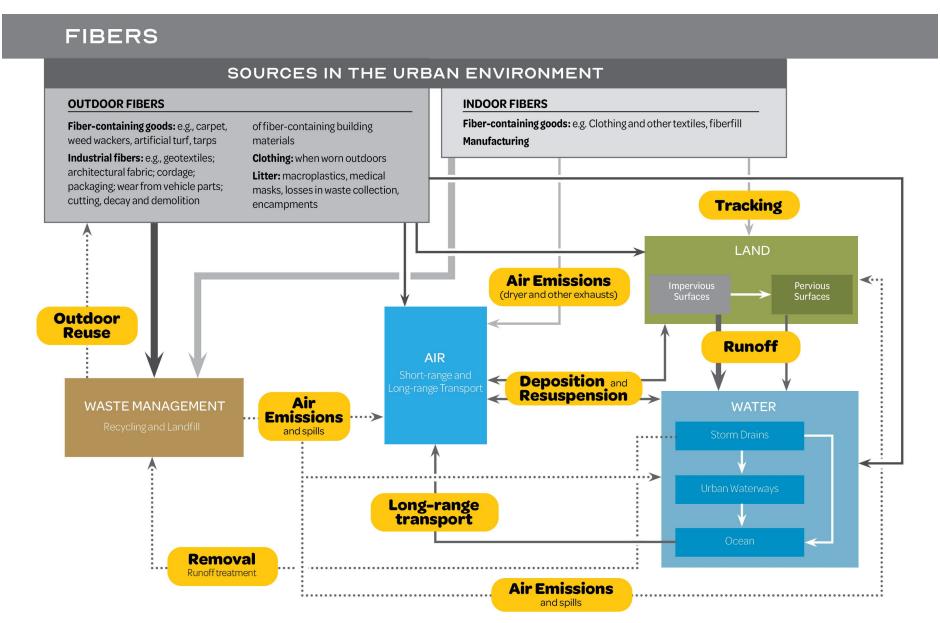


Figure 3.1. Conceptual model of the sources and pathways of fibers (excluding cellulose acetate) to urban stormwater. Indoor and outdoor fiber sources are distinguished because these sources have different transport pathways. Major pathways are indicated by larger arrows, and dotted arrows represent minor pathways.

MANAGEMENT OPTIONS: FIBERS **TEXTILE AND APPAREL INDUSTRY ACTIONS POPULATION-WIDE GOVERNMENT CATEGORY ACTIONS ACTIONS OF ACTION APPLIANCE & BUILDING** INDUSTRY ACTIONS **PREVENTION** Regulate and restrict **Product Redesign** Remove toxic ingredients/ the use of toxic Remove toxic Reduce shedding rates chemicals ingredients in manufacturing and Establish maximum final products allowable fibers loss Change product for textiles construction to reduce ("shed standards") fibers shed during use **Develop methods** and standards to Reduce use of measure shedding Provide economic traditional dryers incentives to switch vented outdoors: Labeling and public to condenser dryers Reduce fibers released Reduce overall washing education about fiber frequency pollution and how to Remove legal • Switch to unvented minimize through proper barriers to using material care condenser dryers outdoor clothes lines Hang textiles to dry **Extended producer** responsibility Improve lint traps in dryers (with consideration for fire safety) Reduce outdoor emissions **Capture lint at** entry or exit point in dryer building exhaust system (with consideration for fire safety) Street sweeping Remove fibers from runoff Improved runoff treatment

Figure 3.2. Management options for fiber pollution, framed on a scale from preventative (reducing use and release) to remediative (collection and removal) measures. Options are color-coded by who would be responsible for implementation, with the textile and apparel industry in light orange, the appliance/filtration industry in dark orange, government in dark blue, and community-wide/consumers in light blue.

REMEDIATION

3.8. References

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4. Single-Use Plastic Foodware (SUPF) and Related Microplastics in Urban Runoff

4.1. Background

The focus for this conceptual model is SUPF, which consists of disposable plastic items designed for single-use to serve, package, transport, and consume prepared food and beverages, including any bags, bottles, bowls, caps, cups, cutlery, plates, straws, stirrers, takeout containers, trays, and wrappers. Our definition includes the proposed revised food packaging definition in the California Department of Toxic Substances Control (DTSC) Draft Three Year Priority Product Work Plan (DTSC, 2021); DTSC proposes to define food packaging as "any food contact article used to package hot, cold, frozen, or room-temperature food or beverage items and that is available for wholesale to restaurants and grocery stores or retail sale to consumers." Food packaging items, including any product designed to facilitate food transport by the consumer, are used widely for food preservation, transportation, and delivery to retail sale points (DTSC, 2021). Our definition for this model expands on DTSC's definition of food packaging to include items used for food consumption, such as utensils, straws, and stirrers. This definition is broad to ensure the inclusion of the variety of plastic items from typical sources relating to food.

Single-use plastic foodware products, intended for short-term use and disposal, are low-cost and versatile, which has led to increased usage throughout the world. In 2017, nearly half of all virgin plastic was converted to products with a lifetime under three years (Geyer, 2020). Plastic packaging, with a typical lifespan of fewer than six months, constitutes the largest segment of virgin plastic, consuming roughly 36% of the total virgin plastic used in 2017 (Geyer, 2020). The expectation is that plastic production will rise by 250% by the year 2050, relative to 2017 levels (Geyer, 2020).

Widespread plastic use has led to increased plastic waste in the environment. The US is considered the largest plastic waste producer globally, with roughly 1.13 to 2.24 million metric tons (Mt) of mismanaged plastic per year (Law et al., 2020). A significant amount of this waste is inadequately managed (mismanaged) through open dumping, littering, or uncontrolled disposal (i.e., spills from trash cans). Estimates are that 42 Mt of plastics were openly dumped and littered into the environment worldwide (Geyer et al., 2017; WWF & Dalberg Advisors, 2019). In 2016, roughly three-quarters of the primary plastic produced ended up as waste, with more than 40% from plastic packaging waste alone (Geyer et al., 2017; WWF & Dalberg Advisors, 2019).

Littered plastic, composed predominantly of SUPF, is a significant portion of mismanaged waste worldwide. Recent studies in international coastal cleanups have consistently shown SUPF, such as food wrappers, grocery bags, and plastic bottles, among its most collected litter, taking nine of the top ten spots (Morales-Caselles et al., 2021; Munari et al., 2016; Nelms et al., 2017; Ocean Conservancy, 2020). Similar studies of urban pollution have also found varying degrees of plastic pollution, with most consisting of SUPF (Becherucci & Seco Pon, 2014; Magnusson et al., 2016). A study of litter in urban stormwater in South Africa over twenty years found high levels of plastics

Chapter 4: SUPF and Related Microplastics in Urban Runoff

by count and mass, especially of SUPF including foamed plastics and clamshell containers (Ryan et al., 2020; Verster & Bouwman, 2020; Weideman et al., 2020).

Regionwide studies in California have shown that plastics are the most common litter items found in rivers and streams, on land, and in the ocean (BASMAA, 2020; Moore et al., 2016). The amount of area on the ocean floor found to have plastic has increased since 1994 (Moore et al., 2016). A major subcategory of SUPF, takeout containers, had not been specifically counted in any of these environments and were not included on datasheets. However, in 2018, they were requested to be included on the datasheets for the large-scale trawl surveys done in southern California, as they were being found more consistently in benthic trawls.

Recent investigations in the San Francisco Bay identified urban runoff as a major pathway for microplastics to enter receiving waters. Average concentrations of microplastics in urban runoff from the San Francisco Bay were approximately two orders of magnitude higher than wastewater (Sutton et al., 2019). Fragments were the most common particle type in urban runoff, with the majority of these particles identified as tire wear particles. Other than tire wear particles, the most abundant types of plastic polymers that could be identified were polypropylene (PP) and polyethylene (PE) (Sutton et al., 2019). These common plastics are used in SUPF and a variety of different products; there may be a linkage to SUPF due to the materials, as well as their status as commonly littered items. Still, it is not possible to definitively distinguish microplastics from SUPF and those from other sources made of the same plastic polymers. To date, there are few studies that are able to directly link microplastics to SUPF. One study in China has inferred the source of microplastics to be SUPF by examining a relatively remote lake, where they found PE and polystyrene (PS) microplastics were predominant, implicating improper disposal (littering) of food packaging and containers from tourism as the likely culprit (Xiong et al., 2018). These limited findings on litter and secondary microplastics suggest a potential connection that needs further exploration, especially in understanding their significance in urban runoff.

The conceptual model presented here (Figure 4.1) illustrates the current knowledge of the sources and pathways of SUPF and related microplastics infiltrating the environment from urban runoff. One of the greatest challenges is linking both macro and microplastics in the environment to product sources and geographic locations to inform management policies that could be directed at reducing pollution. Attempts to determine the pathways of macro trash (stormwater, littering, illegal dumping, wind-blown, encampments, or unknown origin) to rivers and streams have occurred, but this is a difficult assessment to make with confidence (BASMAA, 2020; Moore et al., 2020). It is even more challenging to determine the original plastic sources of diverse secondary microplastics recovered from environmental samples. Despite these current uncertainties, our synthesis of available information indicates the current scientific and management focus on SUPF and related microplastics is warranted. This model is intended to inform future research needs and management actions to regulate microplastic pollution.

4.2. Sources of SUPF in Urban Runoff

There are several different polymer types used to create SUPF; the most common are high-density polyethylene (HDPE), low-density polyethylene (LDPE), polyethylene terephthalate (PET), polypropylene (PP), polystyrene (PS), and expanded polystyrene (EPS) (Singh & Devi, 2019; UNEP, 2018; Verster & Bouwman, 2020). Notably, HDPE, LDPE, PET, and PP constitute the vast majority of packaging plastics, the largest single-use plastics sector (Geyer, 2020). In addition, PS and EPS are polymers commonly used for utensils and take-out containers, respectively (Turner, 2020; UNEP, 2018; Wagner, 2020). These synthetic polymers are thermoplastics, which are melt processed and theoretically can be recycled, as opposed to thermoset polymers, which undergo a chemical change and cannot be recycled.

Table 4.1. Summary of important characteristics of the polymers most commonly found in single-use plastic foodware (SUPF).

Plastic Polymers	Example Uses	Properties
High Density Polyethylene (HDPE)	Milk bottles, freezer bags	Typical Density (g/cm³): 0.94-0.97 UV/Oxidation Resistance: Low % Crystallinity: 80-90
Low Density Polyethylene (LDPE)	Bags, containers, plastic film, bottles	Typical Density (g/cm³): 0.89-0.94 UV/Oxidation Resistance: Low % Crystallinity: 30-50
Polyethylene Terephthalate (PET)	Beverage bottles, plastic film, microwaveable packaging	Typical Density (g/cm³): 1.29-1.4 UV/Oxidation Resistance: High % Crystallinity: 10-30
Polypropylene (PP)	Microwave dishes, chip bags, bottle caps	Typical Density (g/cm³): 0.85-0.94 UV/Oxidation Resistance: Low % Crystallinity: 30-50
Polystyrene (PS)	Cutlery, plates, cups, lids, bowls, takeout containers, straws	Typical Density (g/cm³): 0.96 -1.08 UV/Oxidation Resistance: Moderate % Crystallinity: 0
Expanded Polystyrene (EPS)	Insulated food and beverage packaging such as cups, plates, bowls, trays, and containers	Typical Density (g/cm³): 0.015-0.04 % Crystallinity: 0

Sources: (Andrady, 2017; Gomiero et al., 2019; UNEP, 2018; Verster & Bouwman, 2020; WWF & Dalberg Advisors, 2019)

Table 4.1 highlights some important uses and properties of these polymers. It is essential to note that these polymers are not unique to SUPF; at present, the proportions of various plastic polymers used to produce SUPF relative to other products

is unknown. Additionally, SUPF and other plastic products may be composed of mixtures of polymers and include plastic additives, such as phthalates, bisphenols, flame retardants, and alkylphenol ethoxylates, all of which can modify their properties. The characteristics of each plastic polymer type featured in Table 1 are important indicators of the potential for degradation in the environment. The density, thickness, and morphology of litter, especially in marine environments, are critical factors in understanding the surface area's availability for degradation processes. Most of these polymers have lower densities compared to fresh and marine waters (1 and 1.025 g/cm³, respectively), meaning they are likely to float. However, this is likely a transient phase, as foulants form on the plastic and increase density (Cózar et al., 2014). The specific chemical composition of each polymer also influences the rate of degradability, with certain functional groups imparting higher resistance to the photooxidative process (UV/Oxidation). Another factor is the degree of chemical structural order, or crystallinity, with higher levels generally indicating a greater potential for oxidative degradation in the environment (Andrady, 2017).

The pervasiveness of SUPF, particularly as packaging, indicates use across a wide swath of commercial and institutional entities. These items are also present in most households, with specific SUPF designed for home use. There may also be unique sources of SUPF depending on the urban area, including warehouses of online vendors for food products and encampments, where disposable use items are common. Overall, likely areas of use and release of SUPF in the urban environment have been highlighted below:

Food Providers

- Cafes
- Cafeterias
- Fast Food
- Food Trucks
- Food Delivery Services
- Grocery Stores
- Restaurants

Places Used

- Convenience Stores
- Gas Stations
- Hospitals
- Public Areas (incl. Park, Events, Encampments)
- Pharmacies
- Residential Areas
- Schools

4.3. SUPF and Related Microplastics Urban Runoff Conceptual Model

Fate and Transport

Figure 4.1 illustrates our conceptual model of the sources and pathways of SUPF and related microplastics to urban stormwater. The information below provides additional descriptive information.

Designed to be readily transported and easily used outdoors (where proper disposal is less convenient), SUPF products are likely to be improperly disposed of directly onto the land and into aquatic environments after their brief usage. The release of SUPF is relatively common in urban areas with higher population densities (Moore et al., 2016). Littering is considered the primary pathway for SUPF entering the environment (Law et al., 2020).

SUPF litter in both terrestrial and aquatic environments can partially degrade and form microplastics over time through complex interactions of various chemical and physical processes, including mechanical forces, photooxidation, biodegradation, and hydrolysis. SUPF litter on land includes impervious surfaces (i.e., concrete, asphalt) and pervious surfaces (i.e., lawns, porous construction materials). Rainfall and runoff in urban areas allow SUPF, microplastics, and pollutants to enter stormwater collection systems.

Particle washoff from impervious surfaces (streets, sidewalks, roofs) is far more efficient than from pervious surfaces (lawns, gardens, agricultural fields) (Field et al., 2000; Water Environment Federation & American Society of Civil Engineers, 1998). Urban runoff in California is typically channeled through impervious (piped) storm drain systems into urban creeks and larger urban waterways (such as channelized rivers). With a few exceptions (i.e., locations with combined sewer systems and those limited areas where urban stormwater flows through land-based treatment systems), stormwater is directly discharged without treatment to receiving water bodies like creeks, rivers, estuaries, and the ocean. This discharge can lead to faster rates of SUPF and related microplastics entering the environment via this pathway. Aquatic compartments, especially urban waterways and coastal embayments, are also vulnerable to receiving litter directly.

To understand the impacts of different environmental factors on partial degradation of SUPF, their relative importance is qualitatively shown in Table 4.2. Solar and mechanical forces are the most significant mechanisms with the potential to partially degrade SUPF and other plastic trash to microplastics; complete degradation via these mechanisms is expected to be extremely slow. The impacts of exposure to UV light are amplified by increasing oxygen availability, leading to subsequent oxidation, and increasing temperature of the plastic item (sample temperature) across environments and conditions. Biofouling of the surface, usually through bacterial growth and biofilm development, dampens the ability of UV light to degrade materials. Several environmental compartments are highlighted here with ratings from very low to very high for each important environmental factor, relevant to solar and mechanical forces, that when aggregated together could lead to partial degradation. The environmental compartments are generalized representations of terrestrial and aquatic environments.

For surface water and urban runoff, there are ranges represented for certain characteristics due to the complexity of conditions within those compartments (i.e., ambient, storm conditions, piped, open channel, etc.).

Overall, the land and beach/coastal environmental compartments are the zones most likely to accelerate degradation in the environment. Theoretically, these compartments present the greatest likelihood for the generation of secondary microplastics through direct photooxidation and various mechanical forces. Although microplastics can also form in the marine environment, generally lower temperatures cause much slower rates of abiotic and biotic factors leading to partial degradation. Additionally, the lower availability of plastic surface due to foulants (i.e., bacteria and biofilms) and placement in the water column make pathways of partial and complete degradation significantly slower (Andrady, 2017; ter Halle et al., 2016). Urban runoff presents a unique interface between both land and marine environments, where partial degradation may occur at a moderate level, while simultaneously transporting SUPF and related microplastics to the environment.

Table 4.2. Qualitative summary of important environmental characteristics for partial and complete degradation of plastics and microplastics (adapted from Andrady 2017).

Environmental Compartment	Mechanical Forces	Solar (UV)	Sample Temperature	Oxygen Availability	Biofouling
Land (Urban)	Very High	High	Very High	Very High	Low
Beach (Coastal)	Very High	High	Very High	Very High	Low
Surface Water	Low - High	High	Low	Low - High	High
Urban Runoff	Low - Very High	High	Moderate	Low - High	High
Midwater-deepwater	Low	Very Low	Low	Low	Low
Marine Sediment	Low	Very Low	Very Low	Very Low	High

Waste Management and Reuse

Even within proper waste management, there still remain gaps that allow for litter to "spill" back into the environment through transportation of plastic waste, domestically or internationally, and overall mismanagement of waste intended for recycling. In the US, properly managed plastic waste is estimated to be largely landfilled (81%), where it may take several centuries to completely degrade (US EPA, 2020). California has particularly robust management of landfills that limits the potential for losses, exposure, and pollution to the environment. Still, trash can escape from managed bins during trash collection and further contribute to improper disposal in the environment. There are some mechanisms for removing litter for proper waste management, including street sweeping, trash capture devices, and organized trash clean-ups. Currently, there is an insufficient amount of data to quantify mismanaged waste in California.

Recycling accounts for 9% of plastic waste disposal in the US, though there are no assurances that these materials will ultimately be recycled. While some plastics can be recycled to new products (e.g., PET bottles), certain plastic waste cannot be recycled due to multiple factors, including: chemical contamination, complex polymer mixtures, technical challenges, economic feasibility, and/or lack of market/financial incentive. In addition, plastic waste collected after recycling can be exported internationally, with the US trading nearly 2 million tons in 2016 to countries with lower standards of waste management (Law et al., 2020). When recycled, some of the plastic can be remade into similar products; however, most is typically downcycled into a product of a lower quality, and is unable to displace products made from virgin plastics (Schell et al., 2020).

4.4. Assumptions

Possible urban runoff sources and pathways of SUPF and related microplastics are illustrated in this conceptual model. The model highlights litter as a significant source of SUPF entering the environment, extrapolating that use and litter disposal patterns across urban regions are generally comparable. In addition, this model presumes California is consistent with patterns of properly disposed (including landfill and recycling) and littered SUPF observed in regions with comparable waste-related infrastructure. Examples of excluded potential sources and pathways are described below.

<u>Wastewater discharges are excluded from the conceptual model</u>. These discharges are relatively infrequent contributors to urban runoff, and are limited to areas with combined sewer overflow; as a result, they are assumed to constitute an insignificant portion of urban runoff and therefore contribute little to no SUPF microplastics to the environment.

Non-food related packaging is excluded from the conceptual model.

Long-range transport of microplastics via the atmosphere or oceans is another set of pathways with limited data. Few studies have examined the transmission of microplastics in air, especially in urban areas (Allen et al., 2019; Bergmann et al., 2019; Brahney et al., 2020, 2021; Cai et al., 2017; Enyoh et al., 2019; Klein & Fischer, 2019). The differences in sampling methodologies and analysis have further constrained understanding of the air pathway. There are no studies of the long-range movement within the ocean and ultimately ending up on land.

4.5. Data Gaps

Sources

- What are the usage rates of SUPF overall and by type in California?
- What are the litter rates of SUPF overall and by type, particularly in the urban environment?
- Can physical or chemical characteristics of SUPF be used to identify sources (such as product, manufacturer, and where it came from)? What are the product formulations for SUPF?
- How can methods be developed and improved to link secondary microplastics to their original products/sources?

Aquatic Toxicity/Chemistry

- Do chemicals incorporated into SUPF products contribute to their aquatic toxicity?
- What is the toxicity of partial degradation products and byproducts? What is the toxicity of polymer monomers?

Environmental Monitoring

- What are the concentrations of SUPF and related chemical ingredients within the environment?
- What are methodologies for collection and how comparable are they across studies for both macro trash and associated microplastics? What are the best sample collection and separation procedures?

Fate and Transport

- What are the rates of degradation of SUPF (including partial and complete degradation) from various environmental processes? What factors and processes affect degradation pathways? What are the byproducts of partial degradation?
- What are the generation rates of secondary microplastics from SUPF? What is the relative contribution of SUPF litter to microplastics in urban runoff?
- How important is the role of long-range transport of SUPF secondary microplastics via air and from the ocean back to land compared to other pathways?

Mitigation

- What are the alternatives available to uses of plastic in SUPF? Are identified alternatives sustainable, safer, and economical compared to the original materials (i.e., not regrettable substitutions)?
- How can recycling of SUPF become more economically sound and effective (i.e., reduce use of virgin plastic)?
- What are the social and behavioral attitudes towards SUPF and related littering? How can these be leveraged to create effective control measures of releases?

4.6. Identification of Theoretical Mitigation Measures

There are many available approaches to mitigating SUPF and associated secondary microplastic environmental releases, from preventative measures to reduce the use of SUPF through product re-design or replacements, to downstream approaches to capture SUPF trash and litter, to further downstream approaches to capturing microplastics in urban runoff (Figure 4.2). The purpose of this section is to highlight the broad range of potential mitigation opportunities. It identifies actors (e.g., industry, government, and consumers) that would implement each measure, but does not address implementation mechanisms (e.g., voluntary/mandatory or financial incentive/penalty) or public policy questions (e.g., economic or social costs), nor does it address other environmental implications (e.g., greenhouse gas emissions). Many policy approaches have been developed to address plastic waste, and while some are

briefly outlined here, a more thorough list of these policies is discussed elsewhere (e.g., De Silva et al., 2021).

Prevention

Given the huge challenge of managing growing amounts of plastic waste, a wide variety of approaches and policy tools have been deployed to reduce the use of SUPF.

Phase-outs or bans have been implemented to reduce some types of SUPF. The Break Free from Plastic Pollution Act (*H.R.5845 - 116th Congress (2019-2020)*, 2020) calls for phasing out or banning a variety of single-use products by January 2022. Local, regional, and statewide bans on single-use plastics, including foodware and bags, have been passed in many states throughout the US, and particularly in California. Bans on specific items include the statewide ban on plastic bags from retail stores (SB 270), and local bans throughout the state focused on specific items such as expanded polystyrene, drinking straws, and cigarettes. Cities such as Richmond, Berkeley, Los Angeles, and others have passed foodware ordinances to limit the use of SUPF to reduce litter and waste. Another example is California Assembly Bill 1884 (2018), which prohibits full-service restaurants from providing single-use plastic straws unless requested by the customer.

Most recently, the Sustainable Packaging for the State of California Act of 2018 (Senate Bill 1335) prohibits food service facilities located in, functioning as, or contracted by state-owned entities from dispensing prepared food unless using a type of food service packaging on a list the department publishes of approved packaging that is reusable, recyclable, or compostable. As more of these foodware ordinances are passed, the prevalence of SUPF in the environment is expected to diminish. A number of studies have shown that bans, such as plastic bag bans, have been effective in reducing the number of bags in the environment (EOA, Inc., 2014, 2016; Moore et al., 2016).

Further, it is important for industries that use SUPF, especially food and beverage, to minimize their use to limit their demand and motivate development of safer alternatives. Currently, alternatives include reusable (i.e., metal, glass, etc.) and paper products, though it is important to conduct due diligence on replacements to ensure they do not contain another hazard.

Individuals can reduce the use of SUPF by purchasing reusable glass food containers, using personal cups for water and coffee, and cooking at home. A recent publication by Upstream, a non-profit organization focused on "upstream" source solutions, states that "trying to solve the problem by targeting plastics alone misses the point. The problem isn't just plastic. It's our throw-away culture that's driving plastic pollution and a host of other environmental problems. Solving this requires a paradigm shift from single-use to reuse" (Gordon, 2021).

Economic instruments including taxes, credits, and subsidies are policy tools that can be used to nudge actors to change their behaviors. Several economic instruments are highlighted in The Break Free from Plastic Pollution Act to encourage the reduction of single-use products, including by establishing a tax or fee on carryout bags. Alternatively, incentives (such as grants, credits, and rebates, deposit and refund schemes) could also be offered to help reduce the costs of transitioning to alternative (reusable/non-plastic) items as well as to motivate preferred consumer actions.

Together with the government, plastic firms, the food and beverage industry, and others can develop and contribute to these programs. A recent ordinance banning expanded polystyrene use in Sebastopol, California also advocated for merchants to offer a potential credit of up to \$0.25 for consumers using their own reusable containers for takeout (Wagner 2020).

The ban or phase-out of a material or product can lead to regrettable substitution, in which the replacement, thought to be less harmful, is actually hazardous. Some substitutes for SUPF include food contact papers and paperboard containers made of molded fibers or other plant-based materials with the potential to be coated with perand polyfluoroalkyl substances (PFAS) (Schaider et al., 2017), a class of compounds that are generally considered both persistent and toxic (Kwiatkowski et al., 2020). Continued testing has revealed that several molded fiber products consistently contain PFAS (CEH, 2018). A recent study also found PFAS in some paper and other plant-based drinking straws used as replacements for plastic straws (Timshina et al., 2021). In response to these findings, third-party certifiers of foodware compostability have begun to exclude products with intentionally added PFAS, ¹⁵ and some foodware ordinances now require use of PFAS-free single-use or reusable products.

Reduce Environmental Release

Federal, state, and local government regulation of SUPF throughout its lifecycle from production to disposal are critical in the success of current and future mitigation efforts. There are several control strategies for SUPF with many important pieces of legislation and policies at all levels of government, highlighting the important role it plays in motivating action of producers and consumers to ultimately reduce SUPF pollution.

Within this context, it is important to recognize the critical role plastic producers and the food and beverage industry play in the success of reducing SUPF waste. Extended Producer Responsibility (EPR), as defined by CalRecycle, is a framework to place a shared responsibility for end-of-life product management on industry, instead of the general public (CalRecycle, 2019). EPR also encourages review of the full life cycle of the product to make conscious design choices that minimize negative impacts on human and environmental health. Overall, this places the primary responsibility on producers and other industry players who make design and marketing decisions, while allowing the costs of processing and disposal to be incorporated into the total cost of theproduct (CalRecycle, 2019). Already in California, there are several EPR programs in place for products such as paint, carpet, mattresses, and pesticide containers. Althoughthe government is important in these efforts, it is also critical for industry groups to develop EPR programs to take direct responsibility for the environmentals costs of their products. To further understand these efforts and their applicability to plastics, Recommendation #6 of the set of actions to address plastic pollution supported by the OPC is to compile existing research on the feasibility of EPR for recycling or composting of plastic packaging and SUPF (Ocean Protection Council, 2021).

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¹⁵ For example, the Biodegradable Products Institute (BPI): https://bpiworld.org/Fluorinated-Chemicals

Reformulation of SUPF is a method for the plastics industry to tackle the problem directly, which can often be motivated by EPR schemes (voluntary or mandated). Options for redesigning SUPF include the sourcing of sustainable alternative materials, minimization of additives, and use of standardized and simpler product formulations to make it easier to recycle.

In addition, application of standards and labeling schemes would be useful to fully understand the positives, hazards, and risks of each product and its source materials. Standards would help to establish criteria on quality and safety as well as eco-design principles (such as recyclability and reusability). Labels could note meeting of these standards as well as full disclosure of ingredients, especially polymers and additives, and the amount of recyclable material within products. Under The Break Free from Plastic Pollution Act, the federal Environmental Protection Agency must develop and publish guidelines for a nationally standardized labeling system for recycling and composting receptacles. Producers will need to include labels on their products, which are easy to read and indicate whether the products are recyclable, compostable, or reusable. In California, there are also several efforts to develop markets of downcycled materials, ensuring they are converted to high-quality, value-added products (CalRecycle, 2020). CalRecycle also provides funding and technical expertise for market development through programs across the state.

Public education efforts are another important action to raise awareness of the impacts of SUPF and littering as a whole. Local government and grassroot organizations can play a vital role in education to the public and help individuals make a difference in the amounts of SUPF by using best practices in refusing, reducing and reusing SUPF items as well as properly disposing of SUPF. Public education efforts in Australia have led to significant reductions of waste into the environment, noting their potential effectiveness in application in California and the US (Willis et al., 2018). However, public education alone is only effective when costs, benefits, and social norms also support changed behaviors. Further efforts to increase the availability of trash services and recycling bins, especially in areas of encampments, could also help decrease littering.

Remove SUPF Trash and Microplastics from Runoff

Trash in the environment has become a renewed focus of policy throughout the state of California over the last decade. In California, two main types of policies have been passed, including: 1) total maximum daily loads (TMDLs); and 2) the Statewide Trash Amendments. Regional Water Quality Control Boards have passed TMDLs for many water bodies, including those specific to trash for at least 15 water bodies. The most well-known TMDL for the Los Angeles River, established in 2001, was one of the nation's first trash TMDLs (LACRWQCB, 2007, 2015). The goal for this TMDL required 100% trash load reduction accomplished by September 2016. Many affected local jurisdictions have achieved this using full trash capture devices or alternative institutional controls such as street sweeping and education. The Statewide Trash Amendments take such TMDLs to a broader level, as jurisdictions throughout the state now must either install full trash capture devices (Track 1) or partial capture devices and institutional controls (Track 2; SWRCB, 2015). These devices are designed to keep larger trash from flowing into receiving waters from conveyances used to collect or convey stormwater (e.g., storm drains, pipes, ditches), which are owned by a state, city,

or other public entity, and are most commonly known as Municipal Separate Storm Sewer Systems or MS4s.

Bioretention and similar soil media-based filtration-type runoff treatment and infiltration systems have been shown to remove small particles like microplastics from runoff (Gilbreath et al., 2019; Smyth et al., 2021). However, the feasibility of applying this approach widely may be limited by the high cost of installing highly engineered green stormwater infrastructure. For infiltration, particularly into potential future sources of drinking water, there may be questions about the safety implications of chemicals in fibers. Street sweeping may have limited efficacy in removing small microplastics (Selbig & Bannerman, 2007).

4.7. Priority Data Needs to Inform Management

Of the many data gaps identified in this conceptual model, several have significant implications for future decisions regarding the management of SUPF:

- <u>Establish standardized frameworks</u> for monitoring, laboratory analysis, and datacollection of trash and microplastics to fully understand the sources, transport, and fate of SUPF and related microplastics. This includes integration of studies and solutions across both land and water environments, which are often considered separately.
- Improve understanding of the rates of partial and complete degradation of SUPF from various noted processes, especially in real environmental conditions. This is critical to understanding the prevalence of secondary microplastics and other potential byproducts as well as their adverse effects on the environment.
- <u>Develop analytical methods to link secondary microplastics to their original sources.</u> This helps us understand the key sources to target for mitigation of SUPF and related microplastics in the environment.



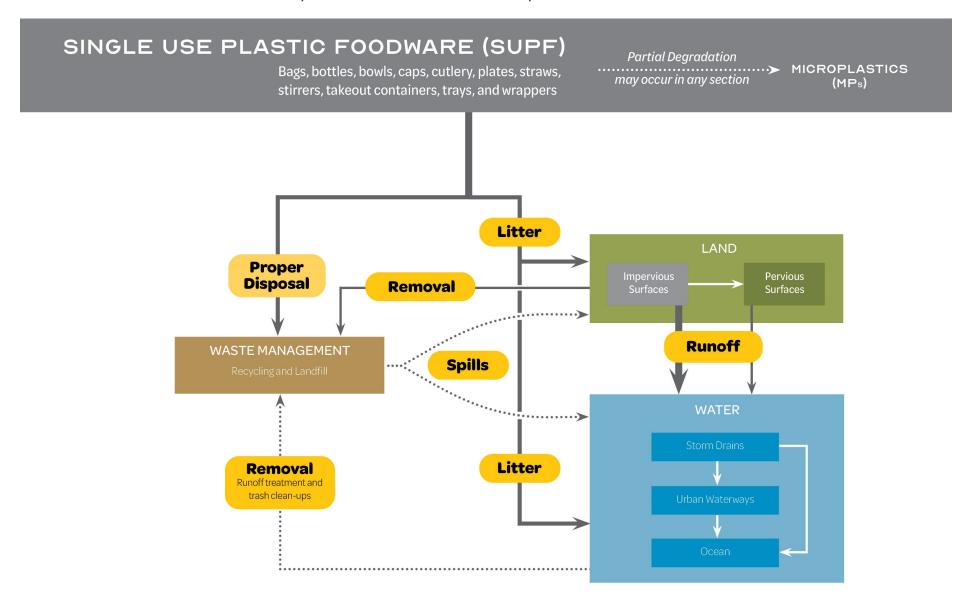


Figure 4.1. Conceptual model of the sources and pathways of single-use plastic foodware (SUPF) and associated microplastic degradates to urban stormwater. Sources of SUPF litter to the environment are places where SUPF is frequently sold (food providers) or used. Major pathways are indicated by larger arrows, and dotted arrows represent minor pathways.

MANAGEMENT OPTIONS: SINGLE USE PLASTIC FOODWARE (SUPF) **PLASTICS INDUSTRY ACTIONS CATEGORY GOVERNMENT POPULATION-WIDE OFACTION ACTIONS ACTIONS INDUSTRY ACTIONS PREVENTION** Remove toxic/persistent Minimize use of Reduce use **Bans on SUPF** ingredients and products SUPF and switch to alternative products, where **Product** available Reformulation Economic • Develop sustainable Instruments materials Taxes/Fees Grants/Credits · Ensure recyclability Deposit/Refund Abate additives Scheme Use of simpler Reduce environmental release and standardized formulations Invest in waste Improve proper collection and disposal of SUPF recycling systems Extended Educate impacts of including Producer best available SUPF and related Responsibility technologies littering Increase availability Standards and of trash and recycling **Labeling Schemes** bans **Increased Local** Removal Street Sweeping **Increase** • Trash Capture Devices of cleanup frequency and Trash Clean-Ups efficacy of

Figure 4.2. Management options for SUPF and associated secondary microplastic pollution, framed on a scale from preventative (reducing use and release) to remediative (collection and removal) measures. Options are color-coded by who would be responsible for implementation, with the plastics industry in dark orange, food and beverage industry in light orange, government in dark blue, and community-wide (non-governmental organizations) in light blue. The current frequency of implementation of each management option is indicated by the box borders, with solid lines indicating novel or not widely implemented actions and dotted lines indicating actions that have already been widely implemented.

Improved runoff

treatment

trash collection

campaigns

Collect and remove from land

and water

REMEDIATION

4.8. References

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5. Tire Particles in Urban Runoff

5.1. Background

Every vehicle on the road sheds tiny particles from its rubber tires into the environment. As they disperse into the environment, these microplastics convey tire tread ingredients into the air, runoff, and eventually into California estuaries and coastal ocean. Tire particles are among the least studied types of microplastics (Halle et al., 2020), but the body of research is growing quickly.

In this discussion, "tire particles" refer to all types of microplastics containing tire material, including both tire wear particles and microplastics created from end of life tires. "Tire wear particles" refers only to particles generated by tire wear on pavement.

Environmental monitoring has revealed the presence of tire particles in air, aquatic environments, and organisms (Baensch-Baltruschat et al., 2020; Leads & Weinstein, 2019; Siegfried et al., 2017; Tian et al., 2017; Werbowski et al., 2021; Wik & Dave, 2009). Modeling studies indicate tire wear may be one of the top sources of microplastic releases to the environment globally (Boucher & Friot, 2017; Hann et al., 2018; Kole et al., 2017; Sieber et al., 2020). On a mass basis, total environmental emissions from tires likely exceed emissions of other well-known pollutant classes like pharmaceuticals and pesticides (Wagner et al., 2018).

Sample collection and microplastic separation methods likely have underestimated the presence of tire particles in aquatic environments. Almost all tire wear particles are less than 100 μ m (Kreider et al., 2010), which is smaller than the smallest sieve size fraction analyzed in most microplastics studies (125 – 355 μ m). Rubber particles are denser than freshwater and seawater, which (in addition to sieve size) explains why tire particles have not commonly been identified in surface water. Common density-based methods for separating microplastics from environmental samples may exclude denser tire wear particles (Klöckner et al., 2019).

Due to the tires' black color, confirmation of the identity of environmental tire particles requires different analytical methods than used for other microplastics, which are usually identified with Raman or infrared spectroscopy. Typically, researchers screen particles for color (i.e., black) and elasticity (i.e., is it rubbery?). For chemical confirmation of tire particles, most researchers rely on pyrolysis-gas chromatography-mass spectrometry (pyrolysis GC-MS), which can be used to identify the presence of major tire polymers, including synthetic (styrene-butadiene) rubber (SBR). Recently, Rauert et al. (2021) confirmed that tires' SBR content differs greatly among individual tires (<0.05 - 28%), which raises the potential that this chemical confirmation method can underestimate the presence of tire particles without more careful interpretation that considers the real-world range of tire formulations. Due to the costs associated with using multiple secondary confirmation techniques, pyrolysis GC-MS typically is only conducted on a small subset of individual particles in microplastics studies. Other studies have successfully identified rubber particles in environmental samples using thermoextraction desorption gas chromatography mass spectrometry (TED-GC-MS) and pyrolysis-GC-MS methods (Eisentraut et al., 2018; Unice et al., 2013).

Very limited urban runoff microplastic monitoring data have been collected. Tire wear particles, in particular, have rarely been detected due to the use of microplastic sampling methods that do not capture tire wear particles (see above). The San Francisco Bay monitoring study found black, rubbery tire particles in urban stormwater (Sutton et al., 2019; Werbowski et al., 2021). The particles were identified as tire particles based on color, elasticity, and—for a subset of particles—identity confirmation with pyrolysis GC-MS. These were the most common microplastics in urban stormwater runoff and were present in all sampled urban runoff discharges to San Francisco Bay. The San Francisco Bay investigation was relatively unique in measuring microplastics in urban runoff that flows directly to surface waters without any treatment. Most other microplastics studies—particularly those examining tire particles—have involved surface waters or effluents from combined sewer systems where urban runoff flows to a wastewater treatment plant prior to its release to the environment. Werbowski et al. (2021) also collected particle sizes smaller than most other microplastics studies, with a smallest sieve size of 125 µm, as compared to a more common size cut-off in the range of 300 µm. Even this smaller sieve size is larger than most tire wear particles.

Tire particles have the potential to harm organisms due to both their physical characteristics and the chemicals they contain. Exposures can occur via organism uptake or by leaching of tire chemicals into environmental water (e.g., in runoff, surface water, or sediment pore water). The inability to separate tire particles from the chemicals they contain poses a challenge for designing tire particle toxicity experiments. It is impossible to obtain tire particles without their additive chemicals, making it impossible to conduct laboratory experiments that examine the effects of tire particle size/shape while excluding effects of the chemicals within the tire particle. Studies examining the toxicity of tire particles have found mixed evidence of toxicity, with differences among the individual or small groups of tires tested (Baensch-Baltruschat et al., 2020; Camponelli et al., 2009; Chibwe et al., 2021; Halle et al., 2020; Khan et al., 2019; Panko et al., 2013; Redondo Hasselerharm et al., 2018; Wagner et al., 2018; Wik & Dave, 2009). Aquatic toxicity thresholds for tire particles cannot currently be estimated because available tire wear particle studies have experimental design limitations, largely due to the inability of researchers to obtain samples accurately representing the diverse formulations and particle sizes of tire wear particles emitted to the environment (Baensch-Baltruschat et al., 2020; Wagner et al., 2018).

Tires can contain more than 100 different materials, including hundreds of chemicals (Tire Industry Project, 2021). Tire rubber composition varies among brands and products. Table 5.1 outlines typical tire rubber ingredients. Each part of a tire (e.g., tread, sidewall) may have a different proprietary formulation

Table 5.1. Generalized tire rubber composition (adapted from Wagner et al. 2018).

Compounds	Content (%)	Ingredients	
Rubber/elastomer	40–60	Poly-butadiene, styrene-butadiene, neoprene, isoprene, polysulfide	
Fillers/reinforcing agents	20–35	Carbon black, silica, silanes	
Process oils/extender oils	12–15	Mineral oils (often aromatic hydrocarbons)	
Vulcanization agents	1–2	ZnO, S, Se, Te, thiazoles, organic peroxides, nitro compounds (e.g., diphenylguanidine)	
Additives	5–10	Preservatives, anti-oxidants, desiccants, processing aids (peptizers, plasticizers, softeners)	
Metal mesh or textiles (for structure)		Steel, polyester, nylon and other metals and textiles	

Some tire chemical ingredients and their transformation products can leach from environmentally distributed tire particles and appear in urban runoff (Johannessen et al., 2021; Peter et al., 2018, 2020; Tian et al., 2021). Aquatic toxicologists have found that tire particle leachates can be toxic to a variety of aquatic organisms (Capolupo et al., 2020; Gualtieri et al., 2005; Halle et al., 2020; Halsband et al., 2020; Kolomijeca et al., 2020; Wik & Dave, 2009). One of the chemicals identified in tire particle leachates. 6PPD-quinone (an environmental transformation product of a tire preservative), causes pre-spawn mortality to coho salmon (Tian et al., 2021). Non-targeted chemical analysis has identified other potentially toxic tire ingredients and transformation products in leachates and environmental media (Overdahl et al., 2021; Peter et al., 2018, 2020; Seiwert et al., 2020). Examples of chemicals contained in tire particles that are of potential interest from the aquatic life perspective include nitrosamines, zinc and other metals, benzothiazoles, 1,3-diphenylquanidine and other bicyclic amines, hexa(methoxymethyl)melamine and other methoxymethyl melamines, chlorinated paraffins, polyaromatic hydrocarbons (PAHs), bisphenols, phthalates, glycols and glycol ethers, and alkylphenol ethoxylates and their transformation products (Beita-Sandí et al., 2019; Brandsma et al., 2019; Celeiro et al., 2021; European Chemicals Agency (ECHA), 2021; Halsband et al., 2020; Llompart et al., 2013; Peter et al., 2018; Sadiktsis et al., 2012; Skoczyńska et al., 2021; Wik & Dave, 2009).

5.2. Sources of Rubber Particles in Urban Runoff

Only a few products emit rubber into the outdoor urban environment. In the US, rubber's main outdoor use is in vehicle tires. Non-tire rubber uses include shoe soles, non-slip and waterproof coatings, and molded, poured, and extruded (MPE) products. Examples of MPE products include non-tire automotive and machine parts (hoses, belts, seals, gaskets, inner tubes); vehicle traffic safety and control devices; mats and pavers for walkways; playgrounds; weather stripping and seals; vibration damping; rubber bands; balloons; hoses; gloves; wet suits; gas masks; carpet backing; flooring; toys; wire insulation; and roofing sheets (International Labour Office (United Nations), 2021; US Environmental Protection Agency Office of Enforcement and Compliance Assurance, 2005).

After their use, tires may be converted into a variety of products, some of which also have outdoor uses with potential to disperse tire particles into the environment. California's Department of Resource Recycling and Recovery (CalRecycle) has a mandate to build a sustainable statewide market infrastructure for tire-derived products. Examples of products created from used tires include retreaded tires, tire-derived fuel, artificial turf infill, rubberized asphalt, rubber-containing pavement sealcoats, and products made from pieces of tire rubber (see Section 5.3). Virgin and non-tire recycled rubber also has some (unknown quantity) market presence in products similar to recycled tire products, such as the small program to recycle running shoe soles for artificial turf infill. No government or other widespread program exists to facilitate recycling of non-tire rubber.

5.3. Tire and Other Rubber Particles Conceptual Model

In the last five years, multiple authors have published conceptual models, numeric models, and literature reviews on tire particles, estimating environmental emissions, examining transport and fate, and summarizing data regarding their detections in environmental media (Baensch-Baltruschat et al., 2020, 2021; Brahney et al., 2021; Evangeliou et al., 2020; Kole et al., 2017; Piscitello et al., 2021; Sieber et al., 2020; Unice et al., 2019a, 2019b; Wagner et al., 2018). Our conceptual model (Figure 5.1) builds on these prior publications, with a focus on elements relevant to urban runoff in the context of California's land uses, geography, vehicles, and end-of-life tire management systems. Figure 5.1 illustrates tire and non-tire rubber sources and pathways to urban runoff.

Tire Rubber

The conceptual model focuses on tires because they are the dominant source of rubber microplastic releases to California's environment. Among all outdoor rubber uses, only tires (and rubber shoe soles, a small rubber market) are designed to wear off during the use of the product. Whenever a vehicle is driven, abrasion of tires by pavement creates small rubber particles known as "tire wear particles," microplastics with diameters ranging from <0.1 µm to 220 µm (Cadle & Williams, 1978; Kreider et al., 2010).

While multiple factors affect tire wear particle generation rates (e.g., characteristics of tires, vehicles, road surfaces, and vehicle operation), from a watershed perspective averaging over many thousands of vehicles, the most important factor is vehicle miles traveled. Due to their high weight and large tires, among all vehicle classes, buses and trucks are suspected to have the highest tire wear particle generation rates (Baensch-Baltruschat et al., 2020; Councell et al., 2004; Kole et al., 2017; Wagner et al., 2018). Using estimated wear rates and vehicle miles traveled (VMT) or estimated tread wear quantities, US tire wear particle environmental releases have been estimated to be between 3.0 – 5.5 kg/y per capita (Baensch-Baltruschat et al., 2020; Councell et al., 2004; Kole et al., 2017; Wagner et al., 2018). These emissions estimates do not include the relatively minor emissions from airplanes, bicycles, and non-road (non-licensed) vehicles like construction equipment, recreational off-road vehicles, and forklifts. No California-specific tire wear emissions estimates were identified in the literature.

Due to lower per capita vehicle miles traveled elsewhere in the world, release estimates for other countries are much lower than for the US. Kole et al. (2017) estimated environmental releases from tire tread wear in various countries (in Europe, and Asia, Australia and Brazil) range between 0.23 – 1.9 kg/y per capita (Kole et al., 2017).

<u>Products Composed of Used Tires</u>

After their original use, tires are often downcycled into microplastic-containing products—like tire crumb and buffings—that have some uses with potential to distribute tire microplastics into the environment. Used tire processors separate tire rubber from tire structural components (e.g., steel belts) to produce various sized tire rubber pieces classified as buffings, ground, crumb, or aggregate, some of which (crumb rubber, buffings, and tire-derived aggregate) contain or are entirely composed of microplastics (see Table 5.2).

Table 5.2. End of life tire particle size marketing classifications (microplastic-containing products in green).

Tire Particle Type	Size	
Ground rubber	Typically 6 – 19 mm and larger	
Crumb rubber	≤6 mm	
Buffings	Typically a few mm thick; sold in size categorized based on length (up to 50 mm long)	
Tire-derived aggregate (shredded whole tires)	Type A: <75 mm (<5% smaller than 5mm)* Type B: <300 mm (<1% smaller than 5mm)**	
Tire-derived mulch (may be dyed)	10 – 30 mm pieces (sometimes composed of buffings)	

Source: (CalRecycle, 2016a, 2016b, 2020)

While more than half of California used tires are reused on vehicles or burned for fuel (Table 5.3), once separated from structural components, tire pieces can replace or supplement virgin rubber in molded, poured, and extruded products and non-slip and waterproof coatings, or be downcycled for uses like mulch, solid playground surfacing, tire-derived construction aggregate (gravel alternative), road surfacing (primarily "rubberized asphalt"), artificial turf infill, stormwater management system infill, and landfill "alternative daily cover" (CalRecycle, 2021).

^{*}Type A tire-derived aggregate products in the California market contain an average of 0.55% (by mass) particles smaller than 5 mm.

^{**}Type B tire-derived aggregate products in the California market contain an average of 0.24% (by mass) particles smaller than 5 mm.

Table 5.3. California waste tire use summary 2018.*

Use	Examples	Quantity (Metric Tons)
Combustion (export)	Burned at non-California facilities	130,000
Combustion	"Tire-derived fuel" burned at four California facilities	82,000
Landfill	Disposal, alternative daily cover	98,000
Reuse on vehicles	Used tires and retreads	82,000
Crumb/ground rubber**	Rubberized asphalt pavement (60 – 67%)*** Artificial turf infill (11 – 14%) Mulch and ground covers (3 – 5%) Molded & extruded products (19 – 20%)	81,000
Civil engineering applications	Landfill structures, construction fill, vibration damping, and stormwater capture and treatment systems	4,600
Other recycling	Unspecified	3,100

Source: (CalRecycle, 2019)

Activities at used tire processing facilities, and storage at production facilities, combustion facilities, construction sites, and other end-use locations might contribute particles to runoff if the storage piles and air emissions are not properly controlled in the manners typically required by construction and industrial facility clean water permits and air quality permits. Improper management could lead to measurable releases into urban runoff in local areas.

Tire shred and crumb products may be locally important sources of tire microplastics in runoff if they are installed in manners exposed to urban runoff. For example, landscape mulch may contain microplastics, or (if composed entirely of "buffings") may be primarily microplastics.

Three uses notable for their current or future potential to release tire microplastics into urban runoff are artificial turf infill, rubber-containing asphalt pavement treatments, and stormwater capture and treatment systems:

^{*}Includes material imported from out of state.

^{**}Excludes "buffings" for which no data are available. According to CalRecycle, "Buffings are used extensively in certain market segments, especially pour-in-place

[&]quot;Buπings are used extensively in certain market segments, especially pour-in-plac playground surfacing, molded products, and landscape mulch products."

^{***}In US locations outside of California, the tire crumb/ground rubber market share for athletic fields and landscape mulch appears to be greater than in California and rubberized asphalt use may be lower. According to the US Tire Manufacturers Association (2018), about 610,000 metric tons of tire rubber were reused in the US 2017 for sports surfaces (23%); playground mulch (24%), rubberized asphalt (12%), formulation into molded/extruded products (28%), and other automotive and export uses (2%) (US Tire Manufacturers Association, 2018).

- Artificial turf infill. Artificial turf typically relies on a particulate "infill" material to support the turf blades. Many installations use infill composed of crumb rubber microplastics created by cryomilling or granulation of whole used tires into microplastics (typically 0.25 4 mm) (US EPA & CDC/ATSDR, 2019). CalRecycle (2019) estimated that about 90 110 new and 25 35 replacement synthetic turf fields were installed in California in 2018. Of these, 60 80% used crumb rubber infill, down from nearly 100% in prior years due to health and safety questions and reuse of existing fill material when replacing existing fields (CalRecycle, 2019). Total estimated crumb rubber use for turf infill was 2 4 million kg (CalRecycle, 2019), about 0.05 0.1 kg/capita. While loss estimates vary, only a small portion of this amount should be released to the environment (Hann et al., 2018; Kole et al., 2017). Although such releases could be measurable in local areas, overall, due to the relatively smaller use in California as compared to other geographic areas, it appears releases from artificial turf infill are small (<<1%) as compared to tire wear emissions.</p>
- Rubberized asphalt and rubber-containing sealcoats. Road surfaces may contain tire crumb, either as a component of asphalt pavement ("rubberized asphalt") or as a component of a surface coating ("chip seal" or "slurry seal"). California governments typically follow Caltrans standards or similar standards in a public works standards compendium (the "Greenbook") used primarily by Southern California local governments (Caltrans, 2019; Public Works Standards, Inc., 2021). Caltrans and the Greenbook have similar standards for rubberized asphalt and chip seal. The Greenbook also allows rubberized slurry seal.
 - Rubberized asphalt. Per CalRecycle (2019), Caltrans is required by statute (AB 338, Levine, Chapter 709, Statutes of 2005) to use tire rubber in 35% of its asphalt paving projects. This is the primary use of tire particles on roads. Since this mandate is relatively new and addresses less than half of all paving, currently less than half of California roads have "rubberized asphalt" pavement, which is not actually rubbery at all. Tire crumb rubber composes only a small fraction—about 1.60%—of the final pavement by weight, because, according to Caltrans pavement standards, on average, each metric ton of pavement contains only 35.3 pounds of crumb rubber (Caltrans, 2020). When making rubberized asphalt for use in California, crumb rubber is mixed in liquid asphalt ("bitumen") before the thick asphalt/rubber hot liquid is combined with aggregate (rock) in an approximately 1:10 mixture (ten times more aggregate than asphalt/rubber mix). Preparing the hot asphalt-crumb rubber mixture requires heating until the tire crumb rubber swells, which integrates it into the asphalt but does not completely dissolve it (Caltrans, 2019). (Caltrans standards do not include an alternative "dry-mix" process used elsewhere that mixes (but does not integrate) the tire crumb into the hot asphalt). After this hot mixing process, the tire crumb maintains integrity within the mixture (Caltrans, 2006). As a whole, the resulting pavement has such a low rubber content that it is not elastic (i.e., it is not a rubbery material).

- Rubberized chip seal. Caltrans and California local governments coat roads with "chip seal" (composed of layers of liquid asphalt and aggregate or sand) to extend pavement lifetime. Rubberized chip seal consists of the same hot asphalt/rubber mixture used to create rubberized asphalt pavement, but instead of being mixed with aggregate to form a thick pavement, it is applied as a surface coating (Caltrans, 2006). To chip seal road surfaces, the hot asphalt/rubber mixture is applied to a road, followed by a layer of rock aggregate that is pressed into the asphalt mixture. A final thin layer of asphalt (without rubber) and sand may subsequently be applied (Caltrans, 2006). Rubberized chip seal also may be placed beneath an asphalt top layer on a road (as an "interlayer"), to seal the old pavement layer (Caltrans, 2019).
- Other rubberized pavement sealants. Like chip seal, slurry seal and other types of pavement sealants protect pavement surfaces. Local governments may apply slurry seal (a mix of asphalt and sand) or other types of seal coatings to roads (Public Works Standards, Inc., 2021). Private property owners may apply pavement sealant on driveways and parking lots. Proprietary product formulations vary. Some of these products contain rubber; a few may contain whole tire crumb. Some formulations are consistent with the Caltrans hot/asphalt rubber mixture standard, but some use different processes that provide less integration of the tire crumb into the asphalt liquid.

While rubberized asphalt only partially wears away during its lifetime, in contrast, the majority of pavement sealant materials wear away due to mechanical abrasion and weathering because the sealant is entirely on the pavement surface. Particles released when rubberized asphalt, chip seal, and rubberized seal coats wear away are primarily rock and non-rubbery asphalt/tire crumb blends. Even at Caltrans' standard concentration of 21% tire crumb (somewhat higher than allowable under other standards), the asphalt/rubber chip mixture used in rubberized asphalt, rubberized chip seal, and most sealants does not contain sufficient rubber to be elastic or "rubbery." Heat softening on hot days or in strong sunlight sometimes makes these materials—as well as asphalt—appear rubbery, but this disappears at laboratory temperatures, ensuring that particles of these materials would not be identified as rubbery microplastics in monitoring studies.

If tire crumb particles within the hot asphalt/rubber mixture can separate from their asphalt binder, pavement wear could release tire crumb microparticles into the environment. For Caltrans standard rubberized asphalt, this potential microplastic source is unlikely to rival tire wear due to long pavement lifetime and lower tire particle content. For pavement surface coatings, the potential importance is unclear, particularly for coatings that may contain whole tire crumb (i.e., coatings not conforming to the Caltrans hot asphalt/rubber standard). This theoretical (but unproven) possibility merits investigation.

Rubberized asphalt, chip seal, and other pavement sealcoats could release tire-related chemicals into the environment as they wear (pavement surface) or when exposed to water (surfaces and interlayers). Again, this potential is greatest for surface coatings, as wear processes will disperse the coating material into the environment.

Stormwater capture and treatment. Tire rubber pieces (tire-derived aggregate, possibly crumb rubber) may be used in stormwater capture and treatment systems (e.g., bioretention) or as bottom fill for roadside drains (CalRecycle, 2016b; Deng et al., 2016). Crumb rubber may remove some pollutants (e.g., metals) from runoff (e.g., Deng et al., 2016). Such use is only at the pilot phase (CalRecycle, 2016b, 2019). Since these materials contain tire microplastics (Table 5.2), unless the design standards and installation practices that are in development prevent microplastics or tire-related chemical releases, there is a potential for these uses to release tire microplastics and tire-related chemicals into urban runoff or groundwater.

Non-Tire Rubber

A few outdoor virgin rubber uses can release microplastics to the outdoor environment, but these uses are believed to make negligible contributions to the quantities of rubber in urban runoff:

- <u>Shoes</u>. While rubber shoe soles may create rubbery microplastics as they wear, they compose only about 3% of the U.S. rubber market (US Environmental Protection Agency Office of Enforcement and Compliance Assurance, 2005) and annual emissions, which are estimated at 0.002 kg/year per capita (Ingre-Khans et al., 2010), are orders of magnitude smaller than tire microplastic emissions (Wagner et al., 2018).
- Outdoor mats and pavers. Like shoes, outdoor mats and pavers should have low wear rates due to their light duty use, long lifetimes, and limited presence outdoors.
- <u>Coatings</u>. Rubbery non-skid/waterproof coatings are used as vehicle undercoatings; truck bed coatings; boat, pool, walkway, stairway, curb ramp, and roof deck surfacing; roofing; leak repair sealants; and to seal drainage systems. These coatings, which compose only a small fraction of the rubber market (US Environmental Protection Agency Office of Enforcement and Compliance Assurance, 2005), may age and decay to form microplastics. Actions like power washing to prepare a surface for re-coating could disperse such microplastics into the local environment.
- Recycled virgin rubber. Most other virgin rubber items are designed as durable
 products intended to maintain their structural form throughout their lifetimes, thus
 limiting microplastic release. The market for recycled virgin and non-tire recycled
 rubber is assumed to be small due to the smaller market share of such products
 and the minimal infrastructure to support their recycling.

Fate and Transport

After their initial release to the air, tire wear particles may travel short (less than a few meters) or long (kilometers) distances prior to depositing on the ground, depending on the size of the particle. Tire particles may deposit on impervious or pervious surfaces, or be washed out of the air by rainfall. Deposited particles—particularly those deposited on pavement—may be resuspended, redistributed, and modified by vehicles. The action of vehicle tires driving over previously deposited particles grinds up tire wear particles and mixes the larger particles that fall directly onto road pavement with other road-related materials ("road dust"), like pavement debris and soil. This grinding process may reduce particle size, encrust particles with other road debris, and modify the particle shape (Kreider et al., 2010; Park et al., 2018).

Like other particles emitted into outdoor environments, most tire wear particles that fall to the earth's surface wind up sequestered on land (e.g., in soils, landscaping, or by grinding into pores in pavement); the remainder is washed off into runoff (Field et al., 2000; Water Environment Federation & American Society of Civil Engineers, 1998). Sequestered particles might release tire-related chemicals into the environment post-sequestration (Peter et al., 2020).

Some tire chemical ingredients (such as benzothiazoles, styrene-butadiene rubber, and zinc) have been used as markers for the presence of tires and occasionally as the basis for quantification of tire wear particles in environmental samples (Adachi & Tainosho, 2004; Cadle & Williams, 1978; Fauser et al., 1999; Klöckner et al., 2019; Kumata et al., 2002). For example, tire markers showed that tire particles are retained at the surface after deposition on roadside soils (Baensch-Baltruschat et al., 2021). While the markers have provided valuable insights on tire wear particle transport, due to other environmental sources and to the high variability among tire formulations, recent analyses suggest such methods are unreliable for quantification of tire wear particles in environmental samples (Rauert et al., 2021; Wagner et al., 2018).

Particle size and transport of tires wear particles and tire-related pollutants. Tire wear particles have a relatively unique size distribution, spanning a range that includes tiny particles <2.5 μm in diameter that transport long distances in air, and larger particles >10 μm that typically deposit close to their near-ground point of emission (Cadle & Williams, 1978; Fauser et al., 2002; Kreider et al., 2010; Wagner et al., 2018). For purposes of understanding particle transport, tire particles can be divided into three groups based on diameter: coarse (>2.5 μm), fine (<2.5 μm and >0.1 μm), and ultrafine (<0.1 μm) (Wagner et al., 2018). The full particle size distribution (by number of particles) is bimodal with peaks in the ultrafine and coarse particle size groups (Cadle & Williams, 1978; Dahl et al., 2006; Fauser et al., 2002; Gustafsson et al., 2008; Kreider et al., 2010). This unusual particle size distribution cannot readily be measured by a single method (Wagner et al., 2018). Air quality research typically involves methods that best measure particle sizes <10 μm ("PM10") or <2.5 μm ("PM2.5") (e.g., Cadle & Williams, 1978; Dahl et al., 2006; Fauser et al., 2002), while scientists outside of the air quality field rely on methods that perform best with particles >10 μm (e.g., Kreider et al., 2010).

Available particle size distribution data indicate that most tire wear particle volume (and therefore mass) is in the coarse fraction. Figure 5.2 (adapted from Kreider et al., 2010)

shows a bimodal tire wear particle size distribution (number of particles) measured using transmission optical microscopy in a road simulator laboratory from tires abraded by a rotating asphalt surface. This measurement has an apparent detection limit of a few µm (as indicated by the steep drop below 4 µm), so it excludes fine and ultrafine particles. Consequently, the figure shows only a portion of the peak occurring in the size range below the detection limit that is reported from measurements by others using methods only detecting particles smaller than about 10 µm (e.g., the aerodynamic particle sizer used by (Alves et al., 2020) or the optical particle sizer used by (Park et al., 2018). Although there is no single particle size distribution in the literature that shows the full range of particle sizes, when the particles below the detection limit in Figure 5.2 are taken into consideration, it appears that most individual tire wear particles (by number of particles) are fine and ultrafine particles (Cadle & Williams, 1978; Fauser et al., 2002; Kreider et al., 2010). Recognizing that the mass of the smallest particles is very small, these data suggest that 90% of the particle volume consists of particles >50 um, a size that will typically deposit quickly, landing on or close to the pavement. The smaller coarse and larger fine particles, particularly those between 1 µm and 10 µm, can be entrained into the atmosphere through mechanical processes, such as from the intense turbulence in the wakes of vehicles (Brahney et al., 2021) and have atmospheric residence times of 8 days (PM10) to 28 days (PM 2.5) (Evangeliou et al., 2020).

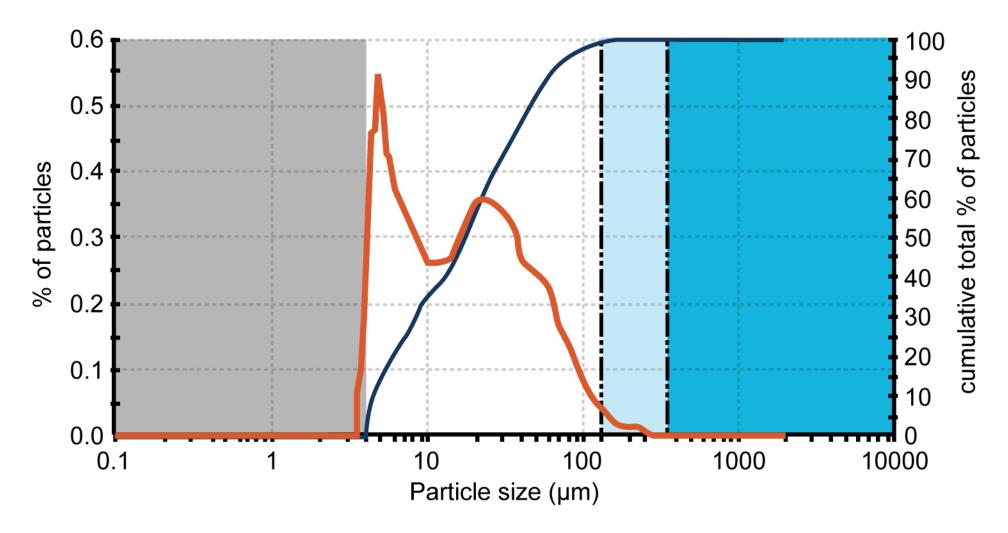


Figure 5.2. Tire wear particle size distribution (by number of particles) adapted from Kreider et al., 2010. The orange line shows particle count, expressed as a percentage of total particles collected (left side y-axis). Dashed black lines indicate sieve sizes used in San Francisco Bay Area microplastic monitoring: 125 μm (stormwater) and 355 μm (surface water). The blue shading identifies which particles are captured when using the associated sieve sizes. The dark blue line indicates the cumulative total particle number in percent (right side y-axis). The gray shading denotes the exclusion of fine and ultrafine particles due to the instrument detection limit at 3 - 4 μm.

<u>Fate</u>. While in the outdoor urban environment, tire particles, which are relatively persistent, are subject to environmental processes that can partially degrade them, such as photodegradation, oxidation, hydrolysis, biological degradation, and mechanical degradation. Small microplastics like tire particles could potentially agglomerate with other particles in runoff during transport. Given tire rubber persistence, physical modification other than mechanical degradation should be relatively limited (Wagner et al., 2018).

Despite their physical persistence, aging of tire particle surfaces in the urban environment releases tire chemical additives and their transformation products to the environment (Huang et al., 2021; Johannessen et al., 2021; Peter et al., 2018, 2020; Tian et al., 2021). Chemical release can occur within the watershed, causing tire-related chemicals to appear in urban runoff (Johannessen et al., 2021; Peter et al., 2018, 2020; Tian et al., 2021). Particles transported to surface water may continue to release chemicals into aquatic environments and organisms that take up the particles.

Density and tire particle transport. Density affects particle transport. While tire rubber has a density of about 1.1 – 1.2 g/cm³ (Degaffe & Turner, 2011; Tang et al., 2006), tire wear particles collected near roads and generated by road simulators typically contain particle encrustations that increase the particle's density. Encrustations consist of "road dust," a variable material reflecting local conditions that is composed of many particle types (local soil, pavement material, brake pad wear debris, road marking materials, and other particles from sources other than road traffic) (Penkała et al., 2018; Thorpe & Harrison, 2008). The density increase depends on the nature and quantity of encrustations. Recent measurements suggest that tire wear particles have a range of density, primarily between 1.3 – 1.7 g/cm³ (Klöckner et al., 2021), similar to prior estimates of 1.25 – 1.8 g/cm³ (Wagner et al., 2018).

Fate and transport processes in urban runoff might reduce encrustations, decreasing particle density and increasing mobility. Using visual microscopy, which might not have sufficient detail to observe many encrustations, no encrusted material was observable in the tire microplastics collected in San Francisco Bay Area urban runoff (Werbowski et al., 2021).

Both tire particles and tire wear particles should sink in non-turbulent water, as they are denser than fresh or estuarine water (which has density of 1.0 - 1.025 g/cm³). Higher density, more encrusted particles will settle out more quickly, slowing transport and increasing potential for these particles to become sequestered on land or in aquatic sediment. Non-encrusted particles have sufficiently low density to mobilize relatively readily in moving water (Tang et al., 2006).

<u>Specific Surface Area</u>. Specific surface area (total surface area per unit mass) is a key indicator of potential for release and/or transformation of chemicals contained in environmental particles like tire wear particles. The greater the surface area, the greater the potential for transformation product formation and for chemical release from the particle into the environment. No specific surface area measurements for tire wear particles were identified in the literature.

Scanning electron micrograph photos and focused ion beam images of tire wear particles reveal rough, irregular surfaces, which suggest that tire wear particles may have high surface areas (Kreider et al., 2010; Milani et al., 2004). Surface area has great practical implications for environmental chemical exposures, as evidenced by the discovery that copper leached from vehicle brake pad wear particles at a high rate compared to some high-copper reference materials, a finding that was attributed to the wear particle surface area being more than 150 times greater than the powdered reference materials (Hur et al., 2003).

Specific surface area typically has an inverse correlation with particle size, i.e., smaller particles typically have greater total surface area per unit mass. Formation mechanisms likely differ among tire particle sizes (Piscitello et al., 2021), suggesting that different tire particle sizes may have different morphologies, which may give them relative surface areas that cannot be predicted solely based on particle size. Due to the lack of surface area data, the portion of the particle size distribution (and associated transport pathway) with the greatest potential to release tire-related chemicals into the environment is unknown. Modeling in the Seattle area found that the correlation between the presence of one tire-related chemical (6PPD-quinone) in stormwater and vehicle miles traveled in the area's subwatersheds was slightly stronger than the correlation with subwatershed impervious area (Feist et al., 2017). If this finding is repeated in other locations, it would suggest that coarse tire particles, which deposit near roads, might release more chemicals into urban runoff than fine and ultrafine particles, which deposit throughout watersheds.

<u>Air Transport</u>. Most of the mass of tire wear particles is in the >50 μm size range that typically deposits relatively quickly, i.e., in seconds (Hinds & Hinds, 1999). Based on the concentrations of tire rubber markers (zinc, benzothiazoles, and styrene-butadiene rubber), tire wear particle concentrations in roadside soils appear to be highest within about 6 meters of the pavement edge (Blok, 2005; Cadle & Williams, 1978; Saito, 1989; Spies et al., 1987; Wik & Dave, 2009). Exponentially decreasing levels of deposition occur up to about 30 meters from the pavement edge (Blok, 2005; Saito, 1989; Wik & Dave, 2009).

In contrast, fine and ultrafine particles can transport through the air for long distances, even to remote global locations (e.g., the Arctic, mountain wilderness) (Brahney et al., 2021; Evangeliou et al., 2020). Road particles (including tires) have been estimated to compose more than 80% of all microplastic air deposition (Brahney et al., 2021). Although these fine and ultrafine particles compose only a small fraction of total mass of tire wear particles, because they compose a large fraction of the total number of emitted particles, their surface area might make them important vectors for tire chemical transport beyond the immediate roadside area. Due to limited data addressing tire particles across the full particle size distribution, the role and importance of air deposition in tire particle transport within and between watersheds remains largely unknown.

For some microplastics, re-emission of previously deposited particles from the ocean surface can be a significant source of deposition on land (Allen et al., 2020). Because

tire wear particles are denser than seawater, making them likely to sink to the sea floor, the ocean to land pathway is unlikely for tire particles.

<u>Washoff.</u> In urban environments, rainfall and runoff wash particles and pollutants into stormwater collection systems. Particle washoff from impervious surfaces (streets, sidewalks, roofs) is far more efficient than from pervious surfaces (lawns, gardens, agricultural fields) (Field et al., 2000; Water Environment Federation & American Society of Civil Engineers, 1998). The "washoff fraction" is the portion of particles (on a mass basis) washed away in runoff instead of remaining sequestered on land. For tires, this application of the "washoff fraction" concept has the shortcoming of not addressing the number of particles, nor particle surface area (i.e., chemical release potential). Nonetheless, the concept provides useful insights into the transport of tire particles.

Tire wear particle washoff fractions are highest in dense urban areas with high proportions of impervious surfaces directly connected to storm drains (directly connected impervious area or "DCIA"). Due to urban design features like sidewalks, driveways, and short setbacks from the road, in highly urban areas, surfaces where tire wear particles are most likely to deposit (i.e., within 6 meters from roads) are likely to be DCIA. Due to compaction (e.g., from foot traffic), pervious surfaces near roads likely have higher washoff fractions than ideal pervious surfaces like a well-maintained lawn (Pitt et al., 2008). Higher DCIA and compacted pervious surfaces in the near-road area increase transport of tire wear particles and other road-related pollutants from the land surface into surface water. For example, Donigian et al. (2009) estimated that accounting for the near-roadside imperviousness increased estimated transport of roadside copper deposition from vehicle brake pads by as much as 20 – 30% if the near-roadway area is 100% impervious (Donigian et al., 2009).

The fraction of tire wear particles transported to surface water in urban runoff via the piped storm drain systems typical in California is currently unknown. Typically, published estimates of the amount of tire wear particle transfer to surface water rely on rough estimates of environmental transfer fraction (backed by very limited data), integrate both washoff and any losses in the stormwater collection system (e.g., deposition in pipes and any runoff treatment systems), and exclude the contribution of air deposition from the long-range air transport pathway. Most published estimates addressed geographic areas where most urban runoff flows to municipal wastewater treatment plants (which remove a substantial fraction of tire particles) rather than directly to surface water via a separate storm drain system of the type common in California urban areas (e.g., Kole et al., 2017; Lassen et al., 2015; Sieber et al., 2020; Siegfried et al., 2017; Sundt et al., 2014; A. Verschoor et al., 2016; Wagner et al., 2018). A few studies included separate—but very different—tire wear debris washoff estimates, e.g., . Wagner et al. (2018) (15 - 50%), Blok (2005) (35%) and (Emission Factors for Contaminants Released by Motor Vehicles in New Zealand, 2002) (80%). Unice et al. (2019) used standard build up/washoff modeling (accounting for particle mobilization based on site-specific rainfall intensity) to estimate tire wear debris transport to surface water; however, like most other studies, the estimate that 18% of tire wear particles were transported to surface water in the study area integrated reductions provided by a wastewater treatment plant (Unice et al., 2019a). As Laermanns et al. (2021) noted in a

preliminary examination of microplastic washoff from rough surfaces, additional data are needed to support development of accurate washoff estimates for microplastics, including tire wear particles (Laermanns et al., 2021).

Street Sweeping. Street sweeping appears unlikely to provide substantial reduction of tire particles in runoff. Street sweepers collect debris from local streets and periodically from highways. Street sweepers are known to collect both tire particles and tire-related chemicals, but the portion of all tire wear particles collected cannot be reliably estimated from available data (Klöckner et al., 2020; Polukarova et al., 2020; Sieber et al., 2020). Tire wear particles deposit on many urban impervious surfaces not subject to street sweeping, such as sidewalks, driveways, and rooftops. Street sweepers are only moderately efficient in collecting particles in the size range of tire wear debris, particularly when considering realistic sweeping frequencies and equipment types (Amato et al., 2010; Residential Street-Dirt Accumulation Rates and Chemical Composition, and Removal Efficiencies by Mechanical- and Vacuum-Type Sweepers, New Bedford, Massachusetts, 2003–04, 2005; Piscitello et al., 2021; Selbig & Bannerman, 2007).

Collection systems and runoff treatment. In the limited locations where urban runoff receives treatment, tire particles may be removed, particularly in systems that filter runoff like bioretention treatment systems ("green stormwater infrastructure") (Gilbreath et al., 2019; Smyth et al., 2021). While tire particles may be temporarily retained in low points in the stormwater collection system under low flow conditions due to their density, turbulent flows during larger storm events will likely mobilize these particles and carry them into surface waters (Hoellein et al., 2019).

Waste Management

According to CalRecycle (Table 5.3), most waste tires enter the waste management system, where they are combusted or landfilled. Within California, once tire material reaches landfills, opportunities for transport of disposed tire particles into urban runoff are relatively limited due to California requirements for daily cover of landfilled waste, secondary containment, and leachate collection and treatment systems (Title 14, Natural Resources--Division 7, 2021; Title 27, Environmental Protection--Division 2, Solid Waste, 2021). Activities at landfills, such as application of tire-derived aggregate (which contains microplastics) as daily cover, could potentially release tire particles into the air, but these would likely deposit on the ground nearby due to their size and density.

Outside of California—particularly in areas with fewer environmental protection regulations—disposal activities, such as at facilities storing tire pieces for future combustion, can be far less regulated and could be a potentially significant source of tire particle and/or chemical emissions to runoff.

5.4. Assumptions

The conceptual model focuses on potentially significant urban runoff tire and non-tire rubber sources and pathways. Examples of excluded potential sources and pathways are described below.

Indoor uses/wastewater discharges/sewer overflows are excluded. Other than shoes and flooring (discussed above in the context of their outdoor uses), most indoor rubber uses would not be subject to wear, making them unlikely to be significant rubber microplastic sources. The conceptual model assumes that municipal urban runoff does not contain meaningful quantities of water that was originally discharged from indoor drains to the municipal wastewater collection (sewer) system.

<u>Agricultural runoff is excluded</u>. This conceptual model focuses on urban runoff and therefore, agricultural runoff is outside the model scope. Additionally, agricultural tire and other miscellaneous rubber uses are relatively limited and generally occur in locations where runoff flows first into pervious areas (e.g., soils) where tire particles would likely be sequestered, limiting their washoff.

Molded, extruded, and poured rubber products are excluded. These include non-tire automotive and machine parts (hoses, belts, seals, inner tubes), vehicle traffic safety and control devices, weather stripping and seals, vibration damping, rubber bands, balloons, hoses, gloves, wet suits, gas masks, carpet backing, flooring, toys, wire insulation, and roofing sheets. Except for outdoor mats and pavers, these products were excluded due to their size (which is large relative to microplastics) and low wear potentials.

<u>Landfilled materials are excluded.</u> Landfill disposal, civil engineering applications, and alternative daily cover were excluded due to the runoff containment provided by the landfill.

<u>Whole tire storage is excluded</u>. Due to tire rubber persistence, microplastics would not be expected to be released to the environment from whole tires that are not subject to mechanical forces (Wagner et al., 2018).

<u>Civil engineering applications of tire-derived aggregate, crumb, and ground rubber with little or no urban runoff contact are excluded</u>. These include uses of tire material in contained manners protected from runoff, such as retaining wall backfill, landfill construction, and road base (structural material below pavement).

5.5. Data Gaps

Emerging concerns around exposures to tire particles and tire tread chemical ingredients have fueled intensifying investigations by researchers around the world, who are studying their toxicity, chemistry, and occurrence in organisms and environmental compartments. To date, tire particle fate and transport in watershed and aquatic ecosystems have been receiving less attention, with a bifurcation between studies of fine and ultrafine particles (traditional air quality topics often focused on human exposures) and studies of coarse particles, rather than a holistic approach addressing

surface waters. While a few organizations have explored mitigation measures, key data needs to support identification and design of effective mitigation strategies have not yet begun to be addressed. We list data gaps we have identified below:

Sources

• Which vehicle classes and usage types have the highest total statewide tire wear particle emissions in California?

Aquatic Toxicity/Chemistry

- Are tire wear particles themselves toxic, or just the leached contaminants?
- What tire-related chemicals (other than zinc and 6PPD-quinone) may cause or contribute to harm of aquatic organisms or ecosystems?
- What are the aquatic toxicity thresholds for tire-related chemicals?
- Do some tires still contain PAHs and/or dioxins at concentrations sufficient to cause or contribute to harm of aquatic organisms or ecosystems?
- Does tire wear particle aging form potentially harmful chemicals other than 6PPD-quinone?
- How can representative tire particles be generated for toxicity, chemistry, and particle characterization studies?

Environmental Monitoring

- What are the best sample collection and separation methods for tire particles?
- What are the environmental concentrations of tire-related chemicals and particles?
- How do tire-related chemical and particle concentrations in runoff and surface water correlate with watershed characteristics?
- What scientific method(s) can be used to accurately quantify the total mass of tire particles in environmental samples?

Fate and Transport

- What are the characteristics of tire particles (i.e., density, surface area, surface charge) and how do these affect tire wear particle fate and transport?
- What particle characteristics and fate processes facilitate transfer of tire-related chemicals into urban runoff?
- Which tire wear particles—the fine and ultrafine particles that travel primarily through the air or the coarse particles that fall on or near the road—have the greatest overall surface area, and thus the greatest potential to support formation and release of tire-related pollutants like 6PPD-quinone into the environment?
- Are particles modified during transport between on-road emission and surface water?
- What role does air transport (short, medium, and long-range) and deposition play in tire wear particle transport to runoff?

Mitigation

- Which particle sizes and transport pathways should be prioritized for tire-related mitigation strategies?
- In the long term, can tire-related chemicals be released from tire particles collected in stormwater treatment systems and flow into runoff and/or groundwater?
- What is the potential (realistic) effectiveness of various theoretical mitigation options?

5.6. Identification of Theoretical Mitigation Measures

There are many available approaches for mitigating tire wear particle and tire-related chemical environmental releases, from preventative measures to avoid emissions and remove toxic ingredients, to reducing tire wear debris formation and emissions, to capturing tire particles or tire-related chemicals in runoff (Figure 5.3). The purpose of this section is to highlight the broad range of potential mitigation opportunities. It identifies the actors (e.g., industry, government, and consumers) that would implement each measure, but does not address implementation mechanisms (e.g., voluntary/mandatory or financial incentive/penalty) or public policy questions (e.g., economic or social costs), nor does it address other environmental implications (e.g., greenhouse gas emissions).

Addressing tire wear particles and tire-related chemicals will require a holistic approach, because it is not possible to separate the tire microplastics from the chemicals that they contain, and because it is necessary to address topics typically managed by different types of agencies and scientists (air, water, toxic chemicals). Consequently, the mitigation options figure addresses tire wear particles and tire-related chemicals together.

Potential tire wear particle and tire-related chemical mitigation options were identified from the literature, government reports, industry sustainability plans, and interviews with industry representatives (Andersson-Sköld et al., 2020; California DTSC, 2021; CARB, 2019; DTSC, 2021; European TRWP Platform, 2019; Gilbreath et al., 2019; Hann et al., 2018; Kole et al., 2017; A. Verschoor et al., 2016; McIntyre et al., 2015; Vogelsang et al., 2019; Piscitello et al., 2021; Smyth et al., 2021; The Tyre Collective, 2021; Tire Industry Project, 2021; Gelbko Environmental Vehicle Solutions, 2021). The many potential mitigation strategies identified run the gamut from prevention to treatment. Actions by four sectors could potentially contribute to mitigation: tire manufacturers, vehicle manufacturers, government, and the general population.

Prevention

Prevention strategies address the toxicity and volume of tire wear particles. Modifying tire tread formulations to remove chemicals associated with aquatic toxicity could occur through voluntary or mandatory tire ingredient assessments, including the alternatives analysis process that may be required by the California Department of Toxic Substances Control (California DTSC, 2021; DTSC, 2021). Existing chemical hazard evaluation tools like Green Screen for Safer Chemicals might provide a framework for such evaluations (Clean Production Action, 2021). Vehicle manufacturers could

potentially restrict chemical ingredients in tires on new vehicles (but not replacement tires sold by others) through their existing International Material Data System (IMDS) system, which is based on the Global Automotive Declarable Substance List (GADSL), a list of chemicals of interest due to existing or anticipated regulation, human or environmental hazard, or association with functional problems in vehicle parts (DXC Technology, 2021; Global Automotive Stakeholders Group, 2021).

While today almost all motor vehicles ride on rubber tires, other mobility technologies exist that do not require rubber tires, such as walking, rail transport, and hovercraft vehicles. Rail freight transport has long been an alternative to heavy-duty trucks. Similarly, urban commutes often involve rail and walking in lieu of driving a vehicle. While hovercraft and other non-rubber or non-tire on-road vehicle technologies do not appear reasonably foreseeable, for completeness, this possibility is included among potential mitigation options.

Reduced Wear Debris Formation

Multiple organizations have suggested redesigning tires to reduce wear debris formation (potentially involving regulatory standards) (e.g., European TRWP Platform, 2019; Hann et al., 2018; Piscitello et al., 2021; Tire Industry Project, 2021; A. Verschoor et al., 2016). Maintaining tire pressure, either through airless tires (a technology already entering the market) or tire pressure monitors (starting to appear on some new vehicles), can reduce wear rates. Tire pressure monitors could potentially reduce tire wear particle emissions by 14% (A. J. Verschoor & de Valk, 2018).

While improving pavement quality with smoother surfaces can reduce tire wear rates, this costly approach would require resurfacing all busy roads. Similarly, changing driver behavior to reduce wear debris formation (e.g., reduce speed, hard cornering and braking) seems unlikely to succeed on a practical basis because public education alone is only effective when costs, benefits, and social norms also support changed behaviors.

Since tire wear relates directly to vehicle miles traveled, California's climate-change-motivated VMT reduction efforts could reduce tire wear debris emissions (CARB, 2019). For example, transportation mode shifts away from driving (e.g., bus transit, bicycling) and trip-reducing lifestyle changes (e.g., working from home) reduce annual per-capita tire wear.

Reduced Wear Debris Emissions

Capturing pollutant emissions at the source typically is more cost-effective than removing it from the environment after dispersal. Two companies are patenting systems to collect tire wear debris on vehicles. Upcoming on-road testing will begin to provide insights into the potential emissions reductions available, should such systems be installed on motor vehicles. The effectiveness of such systems from the water quality perspective will depend, in part, on their ability to capture particle sizes with the greatest potential to release chemicals or to harm aquatic organisms. Understanding which

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¹ The Tyre Collective and GelbKo Environmental Solutions.

vehicle classes (e.g., cars, heavy duty trucks) emit the greatest total quantity of tire wear debris in California would inform priorities for installation of such systems.

Collect Dispersed Tire Wear Particles

After their emission to the environment, collecting tire wear particles poses significant challenges. Existing widely implemented street sweeping programs are episodic (not continuous), do not address all of the directly connected impervious surfaces where tire particles deposit, and typically rely on equipment that inefficiently captures particles in the size range of tire wear particles (Amato et al., 2010; Piscitello et al., 2021).

Although it increases tire wear, porous pavement may reduce tire particle washoff by retaining a significant fraction (as much as 40%) of emitted tire wear particles (Stanard et al., 2007; A. Verschoor et al., 2016). It is unknown if retained particles eventually wash away. Retained tire particles could continue to release tire-related chemicals into the environment. The Netherlands makes extensive use of porous pavement (A. Verschoor et al., 2016). In California, Caltrans allows use of two categories of porous pavement. True pervious pavement is uncommon, as it is not used on highways or city streets (Caltrans, 2021b). Less porous "open-graded" asphalt has common use due to the safety benefits of its rapid drainage (Caltrans, 2021a). While porous pavement could have multiple benefits, this costly approach would require rebuilding all busy roads.

Remove Tire Wear Particles and Tire-related Chemicals from Runoff

Once tire particles contact runoff, they may not only be mobilized as particles, they may also release chemicals directly into runoff (Peter et al., 2020; Tian et al., 2021). This doubles the challenge for urban runoff treatment, requiring systems that capture both small particles and chemicals of interest (not all of which are currently known). Both bioretention and similar soil-based pollutant capture-type runoff treatment systems and infiltration systems are well known to have this type of capacity (e.g., Gilbreath et al., 2019; Laws et al., 2011; McIntyre et al., 2015; Smyth et al., 2021). Other green and gray urban runoff treatment infrastructure types have not been tested for tire particle and chemical removal. Significant tire particle and chemical removal would not be expected of urban runoff treatment systems that do not provide significant removal of fine solids (solids that pass through a 75 μ m sieve) or dissolved pollutants, e.g., ponds and hydrodynamic separators (Clary et al., 2020; Wagner et al., 2018). Infiltration-based runoff treatment systems could allow soluble tire-related chemicals to pass into groundwater, raising questions about the safety implications for groundwater aquifers that are current or potential future drinking water sources.

Though not widely implemented in the US, some municipalities direct "first flush" runoff from select road segments into the municipal wastewater collection system that flows to municipal wastewater treatment plants, which likely remove a significant portion of tire particles (Wagner et al., 2018). Deliberate, pre-rain event washing has also been suggested (Kreiger, Jr., 1994). Because both chemicals and particles mobilize from urban surfaces throughout storm events, these approaches would reduce, but not eliminate, tire particles and chemicals in runoff. Engineering feasibility, risks of wastewater overflows, and regulatory compliance challenges have limited implementation of these measures in California.

5.7. Priority Data Needs to Inform Management

Among the many data gaps identified in the conceptual model development process, several have significant implications for future decisions around management of tire wear debris:

- Measure tire wear particle surface area, determining the size fraction of tire wear debris that accounts for the majority of the surface area, to inform design and location of on-vehicle and environmental systems to collect tire particles.
- <u>Develop accurate sample collection, separation, and quantification methods</u> for tire particles spanning the full particle size distribution.
- Identify the vehicle classes that have the highest total tire wear particle emissions in California using California vehicle miles traveled and tire wear debris generation rates matched to California vehicle classes. Due to their high weight and large tires, buses and trucks are suspected to have the highest particle generation rates, but their overall emissions relative to other high-mileage vehicle types (e.g., taxis, shuttles, delivery vans) remains highly uncertain.
- Conduct additional measurements of environmental concentrations of tire-related chemicals and particles, particularly in urban runoff and wet and dry air deposition. For tire-related chemicals, source-specific data (e.g., tire particle leaching studies) can clarify whether tires are a significant source.



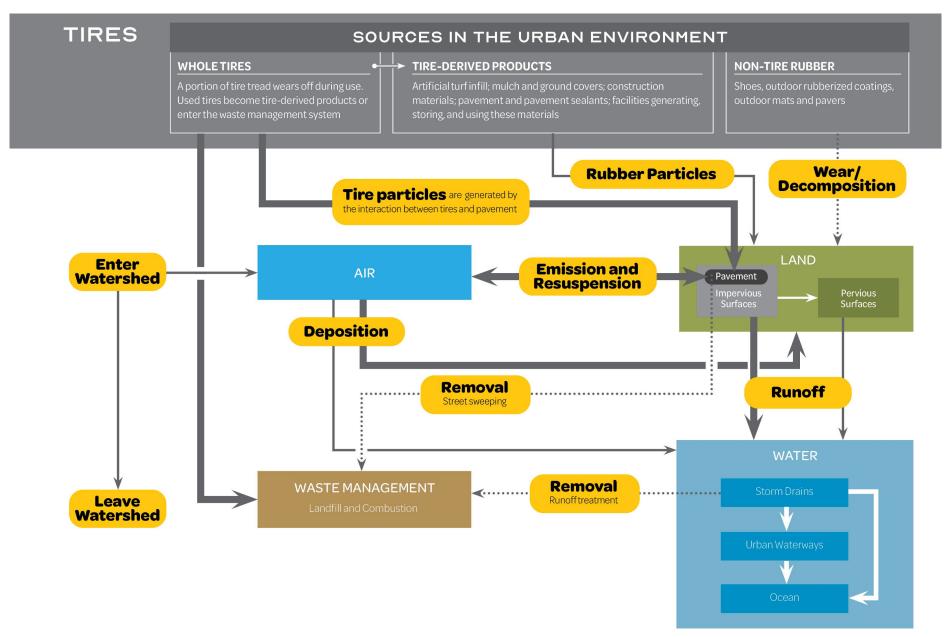


Figure 5.1. Conceptual model of the sources and pathways of rubber particles to urban stormwater. Major pathways are indicated by larger arrows, and dotted arrows represent minor pathways. The major source of rubber particles is tire wear, with tire-derived and non-tire rubber products representing smaller sources.

MANAGEMENT OPTIONS: TIRE WEAR

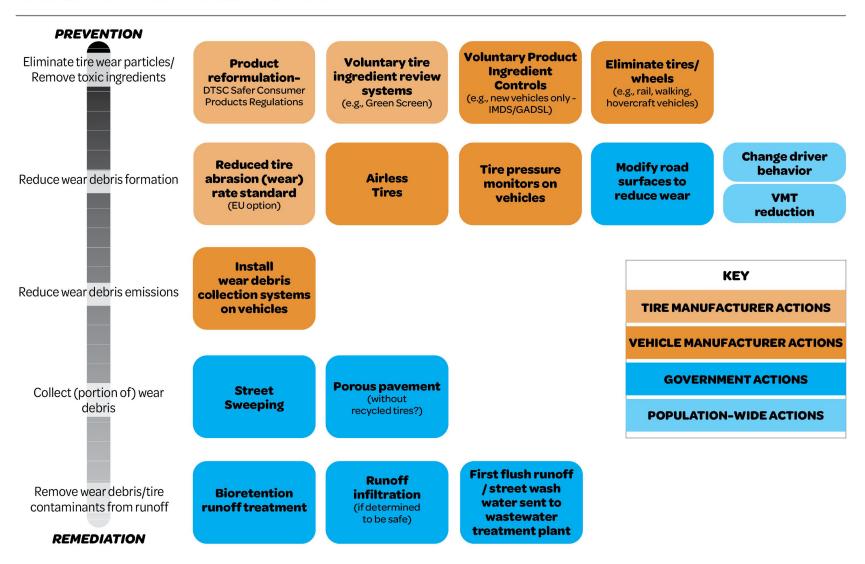


Figure 5.3. Management options for tire wear particle pollution, framed on a scale from preventative (reducing use and release) to remediative (collection and removal) measures. Options are color-coded by who would be responsible for implementation, with tire manufacturers in light orange, vehicle manufacturers in dark orange, government in dark blue, and community-wide (non-governmental organizations) in light blue.

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6. Conclusions

California's Ocean Protection Council has the unique opportunity—and challenge—to develop the first state microplastics management strategy in the US. Microplastics are widely recognized as a growing environmental challenge, and the synthesis of key scientific information can inform selection of effective management strategies. Recent investigations identified urban runoff as a major pathway for microplastics to enter California coastal waters, broadening the focus of the state's microplastics management efforts to address a set of previously unrecognized sources and pathways of microplastics in the environment. For the first time, this report identifies major sources of microplastics in urban runoff, provides a broad menu of potential management strategies for California, and highlights priority data needs to inform California's future management decisions.

Effective management of microplastics will require different approaches depending on the specific source. The conceptual models helped us identify the diverse approaches available for mitigating microplastics, which include multiple innovative options that go well beyond frequently discussed strategies, such as bans of specific plastic items or treatment of urban runoff. Science must inform state priorities for future action in order to be effective.

Fibers are a priority for management because fibers are one of the most ubiquitous forms of microplastic pollution reported globally, and one of the most common types of microplastics in urban runoff. We identified an urgent need to improve understanding of the major sources of fibers in urban runoff. Our synthesis suggests the novel finding that dryer emissions are likely to be a major source of fibers in California coastal waters. This can be confirmed by monitoring fiber emissions from dryers and comparing these to other currently unknown sources and pathways (e.g., long-range transport and emission rates from outdoor sources like construction materials). Cellulose acetate fibers from cigarette butts are an important source of fibers. Developing improved methods to identify cellulose acetate in the field is a science priority, which would also support monitoring the effectiveness of management actions aimed at reducing associated microplastics by reducing tobacco use and littering.

Single-use plastic foodware (SUPF) items are among the top litter items observed in California and worldwide, and a major focus of management actions. Based on current science, it is still challenging to link microplastics to potential SUPF sources. We recommend prioritizing the establishment of standardized frameworks for monitoring, analyzing, characterizing, and reporting trash and microplastics, which will support integrating science and solutions for both trash and microplastics in land and water environments. California is already taking major steps to reduce plastic use and capture trash, which are also valuable as approaches to managing microplastics. However, these efforts alone will be insufficient to address all microplastics in the environment, because fibers and tire wear particles are major types of microplastics identified in urban runoff.

Tires are a top priority for management because tire wear may be the most important source of microplastics globally, and recent scientific findings are highlighting potential

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toxic ingredients in tires. Research on tire particles is rapidly growing. The wide range of tire particle sizes generated by driving requires scientists to work across air quality and water quality disciplines. The priority science questions we identified involve pinpointing the particle size range that will cause the greatest impacts to water quality, to inform mitigation options. Another priority is identifying the vehicle classes that produce the highest total tire particle emissions in California; this may inform which vehicle classes to target for implementing innovative technologies that can prevent, collect (on vehicles), or reduce tire wear emissions.

While there are still many important data gaps about microplastics, there is sufficient information to begin to take actions to mitigate microplastic pollution. We outlined many approaches available to manage microplastics from each source examined, ranging from prevention approaches like re-designing products to reduce or eliminate chemical and particle emissions, to practices that can reduce or capture emissions at the source, to downstream approaches to capture microplastics in urban runoff. Identified control strategies involve multiple actors (industry, government, and consumers). Some promising opportunities, like prevention strategies that address microplastics sources, will likely require collaboration among multiple government agencies to avoid regrettable substitutions that cause unintended harm, including downstream water quality concerns. While addressing microplastic pollution is a formidable global challenge, the energy from the community and government to address plastics and microplastics gives us optimism that we can use science to protect the beautiful state we call home.



Appendix A: Additional Analysis of Cellulose Acetate Fibers Monitoring Data from Sutton et al., 2019

A.1. Exploration of San Francisco Bay Microplastic Data

This analysis includes all particles we identified specifically as cellulose acetate in both the main Bay Area microplastics study (Sutton et al., 2019; Werbowski et al., 2021; Zhu et al., 2021) and the bivalves side-project (Klasios et al., 2021; Miller et al., 2020). Of the total 7351 particles analysed for polymer composition during these projects, we identified 141 particles as cellulose acetate; 125 particles were found in environmental samples and the remaining 16 were found in blanks.

Figure A.1.A shows the shapes of the cellulose acetate particles found in all samples. Of the 125 cellulose acetate particles we found in environmental samples, the vast majority were fibers (107) or fiber bundles (3); these categories are grouped as "fibers" for subsequent analysis. There were only a few cellulose acetate films (5), fragments (9), and spheres (1).

The cellulose acetate fibers found in environmental samples were a variety of colors (Figure A.1.B). We observed black, blue, brown, clear, gold, gray, green, orange, pink, purple, red, and white. We expect cellulose acetate fibers from cigarette filters would be clear or white, although they may also be stained yellow, gray, or brown from smoking. These colors made up only 39% (43 out of 110) of the cellulose acetate fibers, with the other most common colors being blue and red (both 20%, or 22 out of 110 particles). The ratios of confirmed cellulose acetate fiber colors is probably at least partially an artifact of the Rochman lab's process for choosing which particles to spectroscopically analyze (discussed below). The diversity of colors we observed indicates there may be cellulose acetate fiber sources other than cigarette filters. However, this may also be an artifact of imperfect identification methods. It is difficult to clearly distinguish between semi-synthetic and natural celluloses using current spectroscopic methods of polymer identification for microplastics (Cai et al., 2019), so some of the colored fibers identified as cellulose acetate may actually be natural cellulosic fibers such as cotton (and some fibers identified as natural may actually be cellulose acetate).

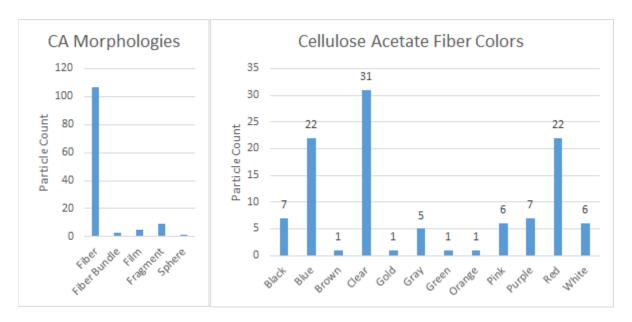


Figure A.1. A) Morphologies of cellulose acetate (CA) particles found in San Francisco Bay. B) Colors of CA fibers.

The cellulose acetate fibers were not split evenly among the different environmental matrices (Figure A.2). Only one (gray) cellulose acetate fiber was found in surface water manta trawl samples ("manta" in the graph). Bivalves and sediment ("sed") had mostly clear cellulose acetate fibers, while prey fish ("fish") had mostly blue and red cellulose acetate fibers. However, the fish sample analysis excluded clear and white fibers due to laboratory contamination with this category of microparticles. Wastewater effluent ("eff") may be a significant source of many of the colored cellulose acetate fibers, but also contributed some clear cellulose acetate fibers. Stormwater ("sw") had the most cellulose acetate fiber color variation, but was dominated by clear, blue, and red.

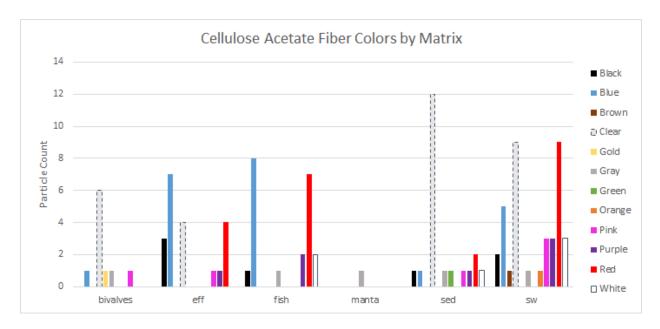


Figure A.2. Colors of cellulose acetate fibers found in each environmental matrix: bivalves, surface water manta trawl samples ("manta"), sediment ("sed"), prey fish ("fish"), wastewater effluent ("eff"), and stormwater ("sw").

The Rochman lab subsampled microplastics for spectroscopy by choosing the first 10 of each color/morphology combination. This system for choosing which particles to analyze was designed to capture the breadth of microplastic diversity rather than to be representative of the proportions of each material. The high percentage of colored cellulose acetate fibers is a likely artifact of this subsampling scheme, as we would expect the majority of cellulose acetate fibers to be from cigarettes, and therefore not be dyed. While some of the colored fibers identified as cellulose acetate may actually be other materials (see above), colored cellulose acetate fibers in stormwater may be from sources such as:

- Ribbons not suggested to be common as litter
- Apparel linings may be in laundry effluent or air deposition (dryer lint) or litter to stormwater
- Home furnishings may be in laundry effluent or air deposition (dryer lint) or litter to stormwater

Because they are all fairly rare, none of these potential colored cellulose acetate sources are expected to release as many fibers into stormwater as littered cigarette butts.

To demonstrate the method bias likely leading to over-counting of colored cellulose acetate fibers compared to those from cigarette butts, we also assessed what percentage of white and clear fibers were confirmed to be cellulose acetate compared

to other materials (Figure A.3). Across all matrices (excluding blanks), there were 5724 white and clear fibers. Of these, 5301 were not spectroscopically analyzed. Of the 423 white and clear fibers that were characterized, 38 (9%) were cellulose acetate.

Using microscopes to count and identify fibers has a bias to overestimate brightly coloured fibres (e.g., blue, red) in comparison with other particles because they are more easily recognized. Uncoloured plastic particles are harder to identify and therefore may be underestimated (Dris et al., 2015). A total of 4909 fibers and fiber bundles were found in stormwater samples, 1998 of which were white or clear fibers. It is unclear whether this ratio of uncolored to colored particles is skewed to underestimate white and clear fibers. Of the white and clear fibers, 1916 were not characterized. Of the 82 spectroscopically analyzed white and clear fibers, 12 (15%) were confirmed as cellulose acetate (yellow piece of the pie). This was only a smaller fraction than polyester and cellulosic (cotton).

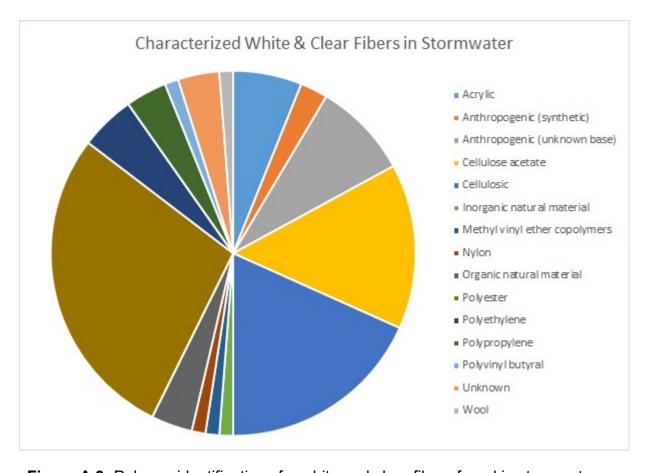


Figure A.3. Polymer identifications for white and clear fibers found in stormwater.

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Appendix B: Additional Analysis of Tire Particle Monitoring Data from Sutton et al., 2019

B.1. Background

Multiple authors have published sometimes differing descriptions of tire wear particle morphology. Tire wear particles were long believed to be elongated (Dannis, 1974; Kreider et al., 2010); however, more recent data indicate that tire wear particles may also be kidney-shaped or round (Kovochich et al., 2021; Park et al., 2018; Sommer et al., 2018). Recent papers suggest that tire wear particles in different size ranges may have different morphologies due to having different formation processes (Park et al., 2018; Yan et al., 2021). Most studies examined morphology using scanning electron micrograph (SEM) images.

B.2. Exploration of San Francisco Bay Microplastic Data

The recent San Francisco Bay Area microplastics study (Sutton et al., 2019; Werbowski et al., 2021; Zhu et al., 2021) included visual microscope photographs of 683 black, rubbery particles collected in stormwater, but not SEM images of these particles. Figure B.1 shows example black rubbery particles collected from San Francisco Bay Area stormwater. These visual microscopy images do show some particle morphology and allow measurement of dimensions, but (unlike SEMs) do not provide sufficient detail to examine detailed features or encrustations in the black rubbery particles.

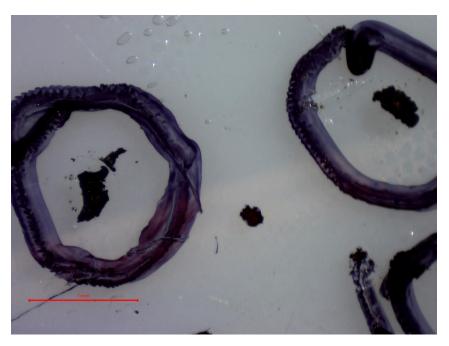


Figure B.1. Photo of suspected tire particles collected from San Francisco Bay urban runoff, circled in black sharpie. Scale bar represents 1 mm.

Appendix B

We reexamined the photographs of the black, rubbery particles collected in stormwater. They had diverse dimensions (Table B.1.) and morphologies.

Table B.1. San Francisco Bay Area black rubbery particle dimensions.

		<u> </u>
Summary Statistics	Length (µm)	Width (µm)
Min	96	7
Max	8550	1621
Median	460	185
Mode	397	140

Qualitatively examining the San Francisco Bay Area stormwater particles, while some particles had elongated shapes or elongated extensions ("tails"), many were kidney shaped or otherwise irregularly shaped. One possible reason for the visual morphological differences between the black rubbery stormwater particles and the tire wear particles is that much of the tire wear particle literature has focused on particles <100 µm (Kovochich et al., 2021; Kreider et al., 2010). These laboratory-generated tire wear particles are smaller than most of the particles that collected from stormwater in the recent San Francisco Bay Area microplastics study (Table B.1), which collected only larger particles due to use of a 125 µm filter to separate microplastics from stormwater (Sutton et al., 2019; Werbowski et al., 2021; Zhu et al., 2021).

Particle aspect ratios provide a means to compare the black rubbery San Francisco Bay Area stormwater particles to laboratory generated tire wear particles quantitatively. The stormwater particle aspect ratios were relatively similar to the tire wear particle aspect ratios recently published by Kovochich et al. (2021) (Figure B.2). San Francisco Bay area stormwater particles had a slightly greater fraction of the more linear high aspect ratio particles (>2.5) as compared to the tire wear particles assessed by Kovochich et al. The difference in observed morphology between the two studies might potentially relate to either the relatively large size of the San Francisco Bay area particles compared to the full tire wear particle size distribution or to particle fate between the point of release on the road and the stormwater sampling location.

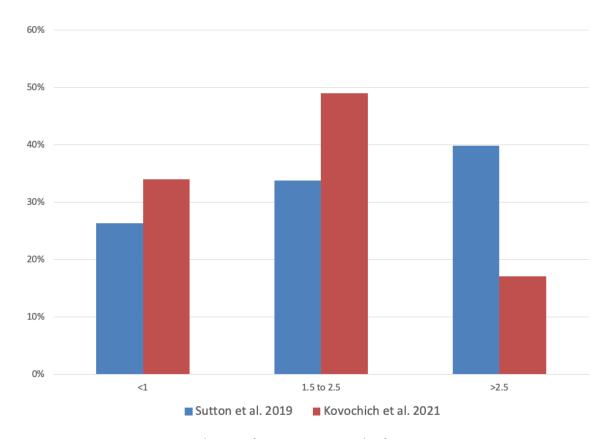


Figure B.2. Aspect ratios (ratio of length to width) of black, rubber microplastic particles from Bay Area stormwater (n=683, Sutton et al. 2019) and lab-generated tire and road wear particles (N=164, Kovochich et al. 2021).

Sommer et al. (2018) measured tire wear particle sizes from material collected by an elevated passive air deposition sampler near highways. They reported a mean aspect ratio of 2.8 (convert from axial = 0.36), somewhat higher than the SFEI and Kovochich et al. (2021) data (Figure B.2). The Sommer et al. (2018) aspect ratio may be biased by their visual tire particle identification method, which assumed tire wear particles must have an elongated shape.

Particles associated with waste (whole) tire recycling, such as those used in artificial turf infill and asphalt, can be generated by cryomilling or by ambient temperature granulation. Granulated particles have unknown shape. Cryomilled particles are typically a more jagged, sharp-edged shape as compared to the rounded edged tire wear particles (Kovochich et al., 2021; Kreider et al., 2010). While it is possible that sharp edges could potentially be removed during transport, these sharp edges were not common among the stormwater particles. Examining morphology of these other particles as compared to stormwater particles could provide another line of evidence (beyond the recycled tire product volume data from CalRecycle) to confirm that turf infill, tire recycling facilities, or users of recycled tire products are unlikely to be important sources of tire particles in runoff, except in unusual local circumstances (e.g., improper storage).

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