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Review

Durable Plastic Goods: A Source of Microplastics and Chemical Additives in the Built and Natural Environments

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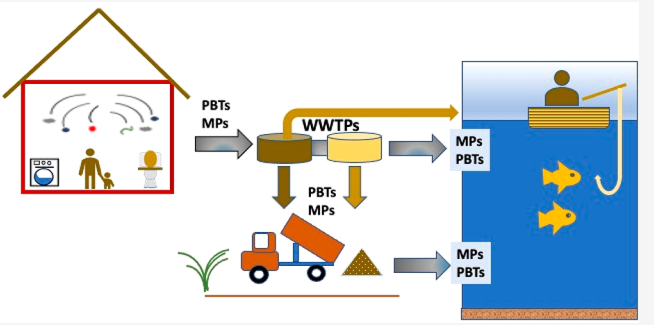
ABSTRACT: Marine plastic pollution by single-use packaging is an emerging concern. However, more than half of all plastics manufactured are designed and utilized for longer-term uses (e.g., as indoor furnishings, insulation, electrical devices, conduits, and textiles). Such durable plastics are more likely to contain persistent, bioaccumulative, and toxic chemical additives (PBTs). Considerable additives and polymer fragments are released into enclosed indoor spaces over the service lives of these plastic products, with resultant human exposure, and then pass to wastewater treatment plants. However, globally only approximately half of all wastewaters receive any treatment. For affluent nations, efficiencies of removal of microplastics and PBTs of ≥90% are commonly quoted for effluents, but some wastewaters therein receive primary or less treatment. Regardless, PBTs and microplastics largely survive even sophisticated treatment, and most are deposited into settled solids. Such "biosolids" may then be repurposed to enrich soils due to their nutrient content. Associated contaminants may affect soil communities and later be dispersed via hydrologic and aeolian processes. To date, regulatory efforts have been insufficient to stem microplastic and additive emissions to air, water, and soils. Upgrading wastewater treatment to tertiary and excluding floating or primary settled solids from land-applied biosolids would substantially reduce releases of these contaminants.



KEYWORDS: *emerging contaminants, biosolids, PBTs, wastewater treatment, residuals, multimedia, indoor dust, reuse*

INTRODUCTION

Greatest attention with regard to environmental contamination by plastics has focused on single-use packaging, as its use and disposal are most conspicuous. However, such applications constitute <50% of polymer production.[1] Most plastics are used in durable goods that are deployed for years in indoor applications. Plastics are also often portrayed as simple, homogeneous materials, leading to misconceptions regarding their environmental fate and toxicological potential. Plastic products are diverse in physical form, including molded and blown items, coatings, modified paper/plastic hybrids, and textiles. In addition to differences in base polymer composition (many consisting of composites of different polymers), plastic products commonly contain residual monomers, catalysts, processing aids, fillers, blowing agents, and additives.[2] Additives alone contribute on average 7% by weight of the product[1] and include plasticizers, pigments, surface modifiers, flame retardants, antimicrobial agents, and stabilizers.[3,4] Some additives are persistent, bioaccumulative, and toxic chemicals (PBTs). Over time, weathering and abrasion fragment products into microplastics (<5 mm) and nanoplastics (<1 μm). Small particle sizes facilitate their transport, overall chemical reactivity (due to larger surface areas), potential for biological uptake, and toxic effects.[5,6] Plastic macrodebris, microplastics (and nanoplastics), and associated chemical additives, especially PBTs, pose risks to environmental and human health.[7,8] While often discussed in a marine context, these are particularly abundant indoors[9] and in urbanized terrestrial areas, i.e., the built environment.[10] Although intensive commercial plastic production began in only the 1950s, microplastics have become ubiquitous, reaching even remote environments, e.g., glaciated landscapes, mid-ocean gyres, and deep trenches, via long-range atmospheric and aquatic transport processes.[11,12]

The manufacture and use of plastics are increasing. Jambeck et al. projected that contamination of the global ocean would increase 10-fold between 2015 and 2025.[13] While there are important aquatic sources (e.g., fishing gear, shipboard cargo and vessel losses, offshore waste discharges, etc.), most plastic

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debris and PBT chemicals originate from land-based sources. Nonetheless, to date such plastic pollution has received less attention.[14,15] Despite calls to reduce plastic use and waste, U.S. production of shale gas (a source of plastic precursors) has risen dramatically.[16] This, and increasing global demand for plastics, has fueled an expansion of plastic manufacturing infrastructure. The recent COVID pandemic further spurred the manufacture, use, and discard of plastic personal protection equipment (PPE).[17] Market conditions and concerns over virus exposure have impeded plastic recycling and reuse.[18] The U.S. plastic recycling rate dropped substantially from 8.7% in 2018 to <6% in 2021.[19]

Plastic product performance and safety dictate that additives remain within the host polymer. However, these do migrate from both in-service and discarded products, accelerating product embrittlement and failure. The time window for the release of additives from plastics used indoors (e.g., home furnishings) spans years, extending to centuries for discarded items. Events such as fires and extreme weather such as floods may also facilitate release of microplastics and additives.[20] The viscous nature of most polymeric matrices (including the extent and arrangement of rubbery and glassy regions) controls additive release. Additional factors that influence migration include ambient chemical (e.g., presence of solubilizing agents that may penetrate the polymer and enhance extraction) and physical conditions (e.g., temperature, which affects the glass transition state).[5,21] Fragmentation of plastics exposes fresh polymer surfaces and decreases the distance additives must traverse within the polymer to reach the particle surface, facilitating chemical additive release.[2,6] Additives released can contaminate surrounding media (including air, dust, water, soil, sediments, and food and beverages in the case of packaging). When ingested or inhaled, microplastics and fibers encounter conditions (e.g., higher temperature and greater fluid solvent strength) that expedite leaching of the additives therein, enhancing their bioaccessibility.[22] Plastic leachates are complex and may be toxic.[23]

Chemical safety and migration issues have been best studied in plastics used for food and beverage applications. Here, infiltration of additives, impurities, and reaction and degradation products (known as "non-intentionally added substances") into consumables has been examined.[24] Additives in indoor durable plastic goods (e.g., electronics, thermal insulation, and carpet underlayment) were long assumed to be immobilized within the host polymer. Hence, our understanding of the identities, levels, and release of such chemicals is limited. However, the long service lives of many of these products provide opportunities for microplastics and associated chemicals to be released into indoor spaces, even years after being banned from new applications. The United Nations Stockholm Convention regulates the manufacture, transport, use, disposal, and environmental release of select PBTs.[25] However, legacy contamination of built environments remains and represents an environmental justice issue. One example is Pb-containing paints, some of which were polymer-based. Deterioration of these paints transfers Pb to indoor dust, which may then be inhaled or ingested by residents.[26] Aging home furnishings, electronics, and insulation can release metallic and halogenated flame retardants into indoor air and suspended and settled dust.[27] Plastic debris also serves as a long-term repository of such chemicals following the discarding of products.[2] Additionally, plastics may sorb and concentrate contaminants orders of magnitude above ambient environmental levels,[28] a phenomenon exploited by the International Pellet-Watch Program to evaluate PBT burdens in shoreline-deposited, preproduction plastics and local waters.[29]

The 2015 U.S. Microbead-Free Waters Act curtailed the use and release of microplastic abrasives in select rinse-off personal care products.[30] While such microbeads received considerable attention, they constituted a small percentage of total microplastics. Microbeads continue to be used in some makeup, lotions, deodorants, and household and industrial abrasive formulations. Nylon and rayon microfibers are also common in cosmetics such as mascara. Additionally, per- and polyfluorinated alkyl substances (PFAS) are constituents in some "long-lasting" cosmetics.[31,32]

Affluent nations have long exported plastic waste (including electronic or "e-waste") under the guise of recycling to less affluent ones that lack adequate regulation and infrastructure to deal with even their own waste.[33] This has resulted in local contamination, an environmental justice concern, and eventually contamination of the world ocean.[34] The global hierarchy of waste plastic exports was disrupted in 2018 when the largest importer, China, banned their entry.[35] Much waste then was redirected elsewhere, especially to southeast Asian countries. In 2019, the Conference of the Parties to the Basel Convention on Hazardous Materials adopted amendments to control the international transport of plastic waste.[36] Under this convention, plastics containing PBTs (i.e., lead, mercury, and halogenated organic additives) were deemed hazardous waste and further controlled. However, not all nations (including the United States) ratified the convention. The complete chemical compositions of many newly synthesized plastic products are enigmatic, in part due to confidential business information protections.[4] The presence of PBT additives in plastics may compromise their safe recycling, placing downstream users at risk.[37,38] Compositional knowledge of recycled materials (derived from multiple sources) is particularly lacking.[39] For example, brominated flame retardants (BFRs) and metals originating from plastics derived from e-waste have been discovered in toys, food packaging, eyewear, and jewelry.[40,41]

TRENDS IN GLOBAL CHEMICAL PRODUCTION AND ENVIRONMENTAL CONTAMINATION

As the use of chemicals and plastics increases and humans spend more time indoors, our exposure and subsequent contaminant release to the outdoor environment follow suit. Between 2000 and 2017, the global chemical industry's production capacity nearly doubled.[42] Recently, the cumulative number of chemicals produced (i.e., >350000) was reported to exceed long-standing estimates by >3-fold.[43] More than 10000 chemicals have been associated with the manufacturing of plastics, 2400 of which exhibit PBT properties.[4] The geographic distribution of chemical production and use is also shifting, from the European Union and North America to developing nations. The U.N. Environment Programme predicted that by 2030 Asian chemical production would constitute 70% of global sales.[42] Production is also expanding in Africa, South America, and the Middle East. "Right to know" laws, environmental regulations, and wastewater treatment in these regions are often less rigorous than in Europe and North America.

As production and sales metrics do not reflect chemical toxicity or persistence, they must be interpreted with caution

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when assessing environmental and health implications. Differences in toxicological chemical potencies can outweigh this criterion. Unfortunately, environmental fate and toxicity data for many chemicals remain elusive. Small production volume chemical identities and data on impurities, residual intermediates, and degradation products are often not publicly accessible.[43] Nonetheless, some may be highly toxic. For example, an environmental transformation product of a phenyldiamine-based tire rubber antioxidant additive (6PPD quinone) was recently linked to mass mortalities of salmon returning to spawn in the U.S. Pacific Northwest.[44] It and precursors have also been detected in indoor dust, especially within vehicles.[45] Another example of unintentionally produced contaminants, PFAS residuals, arose from treatment of high-density polyethylene pesticide storage containers with fluorine gas (to reduce their permeability).[46]

In 1977, the U.S. Environmental Protection Agency (EPA) released its "Priority Pollutant List", which initially included 129 metals and organic contaminants [in some cases mixtures, e.g., polychlorinated biphenyls (PCBs)]. The list was anticipated to evolve over time.[47] However, despite the growing diversity and manufacture of chemicals and knowledge of their effects in the intervening >40 years, no additional chemicals have been added. The list encompasses <0.1% of the chemicals in commerce. It does not include emerging contaminants such as pharmaceuticals, many polymer additives (e.g., BFRs and PFAS), or microplastics. Nonetheless, it remains the "go-to" compendium for many wastewater, regulatory, and monitoring programs.

MICROPLASTICS AND POLYMER ADDITIVES IN THE INDOOR ENVIRONMENT AND SUBSEQUENT HUMAN EXPOSURE

Considerable attention regarding the exposure of humans to microplastics has focused on their possible ingestion via seafood.[48] Perhaps more important is the fact that residents in developed countries spend >90% of their lives indoors or inside vehicles, which exhibit limited air exchange and high plastic product densities.[49] This accentuates human exposure to microplastics and associated chemicals. Microplastic levels have been reported to be 10-fold higher indoors than outdoors.[9] While data for the indoor compartment remain limited, a recent report suggests that microplastics constitute percent contributions of some indoor dusts.[50] Particle size and shape also influence transport and toxicity. Small particles (<2.5 μm) can enter human lung aveoli, and <10 μm particles may penetrate cell membranes.[51] Shape will affect particle ingestion, elimination, and physical damage on delicate respiratory or digestive surfaces.[52] Polymer additives (e.g., phthalates, flame retardants, pigments, antioxidants, and surface modifiers such as PFAS) used in upholstered furniture, surface treatments, electronics, lubricants, insulation, and textiles may be at milligram per kilogram levels in indoor dust.[53] The human health implications of exposure to microplastics and associated chemicals warrant further investigation.[54]

Indoor dust ingestion and inhalation are pathways for human pollutant exposure.[55] Dermal contact with microplastics and additives is common,[9] especially with personal care products such as cosmetics. The migration of additives from plastics occurs as a function of their physical properties (e.g., volatility) and ambient environmental conditions [e.g., temperature or presence of sorptive surfaces (e.g., dust) or solvating agents such as digestive fluids]. Polymers that easily fragment (e.g., polyurethane foam in furniture vs high-impact polystyrene in electronics casings) will generate additive-rich microplastics, which then accumulate in indoor dust.[56,57] Using forensic microscopy, Rauert et al. illustrated the physical relationship between microplastics generated by fragmentation of textiles and chemical additives (i.e., the hydrophobic, low-volatility, additive hexabromocyclododecane) in indoor dust.[58] Laundering of clothing and other textiles may release microfibers and flame retardants [e.g., water-soluble tris(1-chloro-2-propyl)phosphate (TClPP)] to "gray" water.[59] PFAS applied to textiles and other indoor surfaces can also be transferred to wash water, indoor air, and dust.[60] Chemicals released may then enter wastewater treatment plants (WWTPs) or septic systems or be released directly to the environment if no treatment is imposed. Clothes dryers also generate and release microfibers to both indoor and outdoor environments.[61]

MISMATCH BETWEEN CONTAMINANTS IN THE ENVIRONMENT, BIOACCESSIBILITY, AND EFFECTS

The presence of contaminants in environmental media does not equate to organismal exposure or toxic consequences. Thus, research must progress from simply documenting levels in different compartments to evaluating critical exposures and resulting effects. Accordingly, Vianello et al. utilized a breathing thermal manikin to simulate human respiratory intake of airborne particulates.[62] While proteinaceous (likely from skin and hair) and cellulosic particles composed 95% of particles, these authors also observed polyester, polyethylene, and nylon microplastics. These tended to be smaller than natural particles and thus likely to be inhaled more deeply into respiratory tracts. The authors also expressed concern regarding the toxicological impact of additives. Microplastics in human lungs and human sputum, as a surrogate for their presence in the respiratory tract, have also been reported.[63,64] Here, the dominant polymer type observed was polyurethane, followed by polyester, chlorinated polyethylene, and alkyd varnish. These polymers differ from those commonly present in single-use plastics. Using personal air samplers positioned within the breathing zone of the wearer, Schreder et al. concluded that inhalation of flame retardant-laden airborne indoor particles was a more significant exposure route than ingestion of settled dust.[65] La Guardia et al. evaluated the probability of flame retardant-laden air particles to infiltrate the human lung.[66] They suggested that the inhalable fraction (>4 mm particles), which can be expelled or swallowed and then absorbed through the digestive tract, might contribute more polymer additive than the smaller respirable fraction.

Microplastics have also been detected in human feces. Therein, polyethylene terephthalate (PET) fragments (likely originating from single-use bottles) and nylon fibers (from textiles and carpeting) dominated.[67] Interestingly, they reported that microplastic levels in feces from individuals with inflammatory bowel disease were statistically higher than those from healthy persons. Zhang et al. observed 10-fold higher levels of microplastics (PET and polycarbonate) in stool from infants than adults.[68] The authors hypothesized this was related to the substantial use of plastic containers and utensils for feeding infants. They reported these microplastics in meconium of newborns, albeit at lower levels than in adults. Microplastics have also been detected in human placenta, the crucial conduit between mother and fetus.[69] Flame retardant

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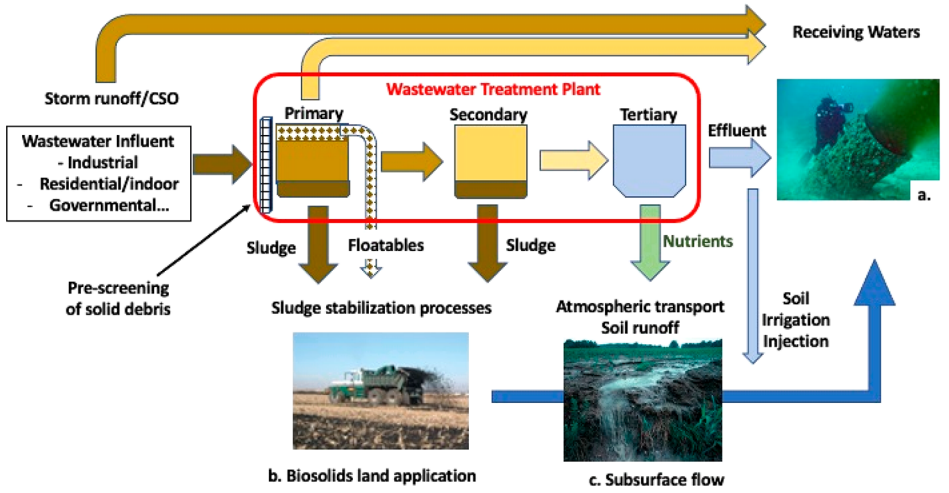

Figure 1. Flow through an advanced wastewater treatment plant and fate of the residuals generated. Influents contain diverse contaminants (plastic debris, chemicals, and other waste) from multiple sources and often receive a sequence of treatment steps. Primary-only treatment allows substantial amounts of contaminants to reach receiving waters. In some cases (e.g., for combined sewer overflows), untreated stormwater runoff may directly enter surface waters. Treated wastewaters are increasingly used to irrigate soils, injected into groundwater reservoirs to replenish supplies, or deployed as potable water. Wastewater solids separated by flotation/skimming and by settling during primary and secondary treatment may be landfilled or incinerated. Alternatively, they can be repurposed as soil amendments, i.e., biosolids. Image credits: (a) Brad Bedford, Florida Atlantic University-Harbor Branch Oceanographic Institute; (b) adapted with permission from ref 114; and (c) reprinted with permission under a Creative Commons license CC by 4.0. Copyright 1999. Lynn Betts, U.S. Natural Resources Conservation Service.

additives have also been observed in umbilical cord and baby's blood.[70] Organophosphate ester (OPE) flame retardants and plasticizers in indoor dust have been correlated with urine metabolite levels in pregnant women.[71] Greater BFR and OPE additive levels have been reported in blood of infants than in that of adults.[27] This may relate to their levels in baby products or behaviors such as mouthing of articles, close contact with surfaces, or dust ingestion.[72] OPE, BFR, and PFAS additives in indoor dust can elicit negative toxicological outcomes in exposed humans.[73] Vethaak and Leslie identified several means for plastics to affect health, including particle-, chemical-, and pathogen-mediated toxicity.[74] Indoor pets also may exhibit elevated burdens of BFRs, mediated by contaminated indoor environments or diet. For example, high levels of polymer additives in house cats have been reported and correlated with negative health outcomes such as hyperthyroidism.[75]

PLASTIC PRODUCTS AND CHEMICALS USED INDOORS CONTRIBUTE TO WASTEWATER AND SLUDGE CONTAMINANT COMPOSITION

Due to the growing recognition of health consequences, the amount of wastewater generated and treated globally has increased dramatically recently. Wastewater sources include industries, residential and institutional housing, and governmental and medical facilities. Jones et al.[76] estimated that 359 billion m^3 of wastewater is generated annually, but 48% remains untreated, comparable to a recent U.N. assessment.[77] The percent untreated varies widely globally. Approximately 80% of domestic wastewater in North America and Europe receives at least secondary treatment, but this level decreases to 26% for central and southern Asia. Approximately 60% of wastewater globally arises from domestic versus industrial sources.[77] Plastic personal care products are commonly flushed down toilets but may later enter surface waters via treatment system failures.[78] Discarded single-use plastics, tire wear, and road marking paint also enter WWTPs via stormwater or surface runoff.[5] Therein, chemical contaminants and microplastics co-mingle. The hydrophobic nature and exaggerated surface areas of microplastics facilitate sorption of chemical contaminants within wastewater streams.[78,79] In addition, weathering (fragmentation, biofilm formation, and surface oxidation) of plastics in WWTPs alters contaminant sorption potentials.[80]

Upon reaching WWTPs, >5 mm plastic debris (coincidentally the cutoff size for microplastics) is often removed by screens and discarded as solid waste (Figure 1). As a result of disruptions and combined sewer overflows, large debris may be obvious in releases.[78] While data are limited, microplastics in WWTPs appear enriched in polymers that are abundant indoors, such as alkyds, polyesters, polyurethanes, and polyamides, and fibers from textiles released during laundering.[81,82]

WWTPs apply a series of treatment steps (Figure 1), but most are not engineered to destroy microplastics or PBTs. After initial influent screening and grit settling, wastewater commonly enters quiescent basins, where surface skimming and solids settling reduce floating and suspended solids, respectively (primary treatment). Organic or inorganic coagulants and flocculants (some polymers themselves) may then be added to facilitate settling. Secondary treatment may follow, typically consisting of microbially mediated aerobic digestion to reduce biochemical oxygen demand (BOD) and additional settling to remove the solids generated. Advanced facilities often apply microbial denitrification, ultrafiltration, or reverse osmosis, aimed at removing nutrients, solids, and high-molecular weight organic contaminants (tertiary treatment). Final steps to reduce effluent pathogens (e.g., chlorination, ozonation, or ultraviolet irradiation) are common. High rates of removal of microplastics (90−99%) from effluents at tertiary treatment plants are oft-quoted.[3] However, substantial releases of untreated wastewaters occur due to design shortcomings (e.g., combined sewer overflows), aging infrastructure, and disruptions following increasingly frequent extreme weather events (Figures 1 and 2).[5]

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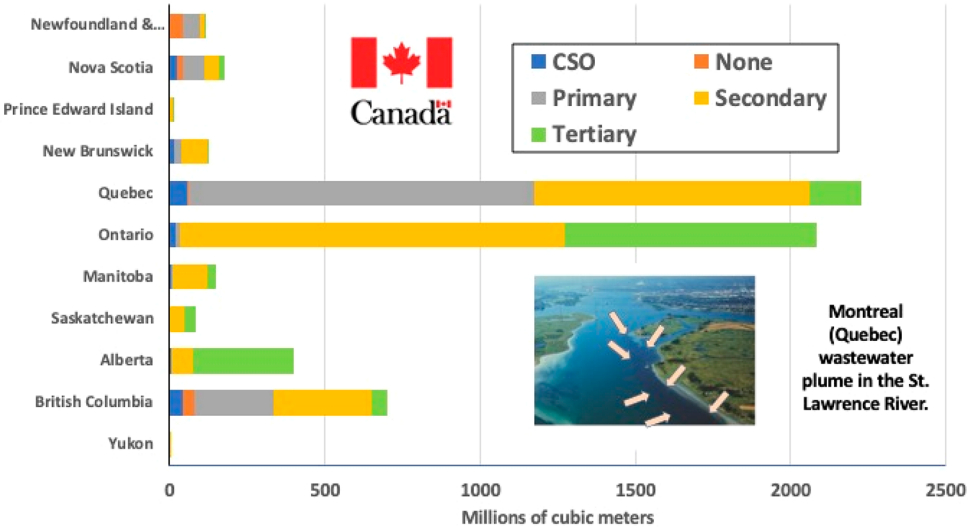

Figure 2. Canadian wastewaters discharged in 2017 by province and degree of treatment (combined sewer overflows, no treatment, primary, secondary, and tertiary). The substantial Quebec primary treatment effluent total (gray bar) derives predominantly from Montreal, the largest WWTP in North America. It discharges to the St. Lawrence River, not a marine outfall, as is common for the Canadian maritime provinces and for U.S. facilities with 301(h) discharge waivers. The Montreal effluent plume is delineated by arrows in the inset (photo credit, C. Hudon).

Even in affluent nations, some wastewaters receive only primary treatment, especially if destined for well-flushed, marine outfalls. In the United States, facilities can request a 301(h) waiver under 40 CFR § 125.57 of the Clean Water Act, allowing less stringent wastewater treatment. These have been premised on the supposition that subsequent dilution and degradation/assimilation of "conventional" contaminants (e.g., BOD, suspended solids, and nutrients) and release of a short list of "priority pollutants" would not compromise receiving water quality. For example, the large WWTPs serving San Diego and Honolulu in the United States and Vancouver and Victoria in Canada long released primary-only treated waste (and associated microplastics and PBTs) to adjacent marine waters. Treatment at some such facilities has recently been upgraded, or upgrades are in progress. But other U.S. (including those in Alaska, Maine, Massachusetts, American Samoa, and Puerto Rico) and Canadian WWTPs (Figure 2; Quebec, Newfoundland, Labrador, and British Columbia) still discharge effluents after only primary treatment. Waivers fail to recognize the ramifications of releasing PBTs and microplastics to the global ocean. Interestingly, the largest WWTP in North America (Montreal, QC) releases 3.5 million m^3 day^{-1} of primary treated wastewater (enhanced by chemical flocculation and ozonation) to the freshwater reach of the St. Laurence River[83] (Figure 2). Even modest sized outfalls may be important due to high levels of contaminants. Herzke et al. reported the releases of microplastic fibers from an untreated discharge to the Arctic Svalbard archipelago serving 2500 inhabitants were comparable to settlements of 1.3 million that employed advanced treatment.[84] They noted that ~1 million Norwegian homes either are not connected or are linked to WWTPs utilizing only primary treatment.

The U.S. McMurdo Research Station, the largest Antarctic base, further demonstrates the influence of indoor, non-industrial (i.e., domestic) wastewaters entering aquatic environments. Maintaining pristine environmental conditions is paramount in Antarctica. Nonetheless, until 2003 (when secondary treatment commenced) McMurdo wastewater was simply macerated and discharged into coastal marine waters.[85] Influent sources were limited to those presumed to be biodegradable (e.g., from restrooms, bathing, food preparation, and laundry), and effluent was rapidly dispersed upon discharge. However, substantial BFR polymer additive concentrations were later detected in the surrounding marine sediments and biota. BFRs were also observed in indoor dust, which likely served as a major source to the station's wastewater. Nonetheless, such treatment met the requirements of Annex III of the Protocol on Environmental Protection to the Antarctic Treaty. Limited wastewater treatment remains common at Antarctic research facilities.[86] Increased BFR levels, as well as PFAS, were also detected in samples associated with the Australian Casey Antarctic Station.[87] These were postulated to have arisen from indoor station activities. Casey applies secondary wastewater treatment, but capacity periodically is overwhelmed.

WASTEWATER TREATMENT SOLIDS

Wastewater treatment solids were historically discarded in landfills, dumped in the ocean, or incinerated. Over time, the financial and environmental costs of those practices spurred efforts to identify alternatives.[88] Today, more than half of the wastewater solids produced in the United States and Canada are repurposed as soil amendments (also known as biosolids), predicated on their high organic carbon and nutrient contents.[89] In the European Union, biosolid application varies widely by country from 0% to >90% of the total produced.[90] Land application is also substantial in Australia and China.[91] This practice represents a significant urban-to-rural transfer of associated contaminants. Land application regulations typically focus on pathogens and toxic metals. Metals have been identified as a concern due to their environmental persistence and accumulation in soils with repeated sludge applications.[92] In the United States, organic pollutants were originally deemed of negligible risk under the assumption that manufacture of persistent organic pollutants (POPs) had ceased and sludge levels of these were decreasing. The 40 Code of Federal Regulations Part 503 rule governing land application of biosolids in the United States, and the associated risk

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assessment, was published in the 1990s,[92] predating the discovery of PBT polymer additives such as BFRs[56] and PFAS.[93] Microplastics represent an additional emerging concern. Not surprisingly, a strong correlation between microplastics in archived Australian and U.K. biosolid samples and escalating global plastic manufacture between 1950 and 2016 has recently been observed.[94]

The U.S. National Academy of Sciences was asked in 2002 "to independently review the technical basis of the chemical and pathogen regulations for biosolids, focusing only on human health". In its report, the term "plastic" was mentioned once, but only in the context of polymer additives.[89] "PBT" was not mentioned at all, and "POPs" were mentioned only once. In 2018, the U.S. EPA Inspector General criticized the existing regulation of contaminants in biosolids, stating, "The EPA's controls over the land application of sewage sludge (biosolids) were incomplete or had weaknesses and may not fully protect human health and the environment".[95] Even in this recent assessment, the terms "microplastic" and "additive" were absent. Microplastics, polymer additives, and many PBT chemicals remain underscrutinized and largely unregulated in biosolids.

Consideration of the treatment processes used by WWTPs reveals opportunities to reduce releases of microplastics and PBTs. In most wastewater treatment schemes, fats, oils, and grease (FOG) and buoyant microplastics [e.g., some olefinic polymers and polystyrenes (PS)] concentrate at the surface of settling basins and are skimmed off.[82] These often are reintroduced into the treatment process or added to the biosolids. Denser and biofouled microplastics, as well as surface-active or hydrophobic chemicals,[79,96] are largely captured in primary and secondary treatment settled solids (Figure 1). In the case of PBTs, North estimated that 96% of hydrophobic BFR polymer additives were entrained in wastewater sludge.[97] Microplastics in these solids have also been revealed to be physically larger than those remaining in the treated effluent.[81] This is important from a mass balance basis, especially as the size of microplastics is ephemeral. Most studies of microplastics report particle numbers in a limited size range (often >100 μm), not actual masses. Thus, consideration of the larger numbers and particle sizes (masses) of microplastics suggests that their contributions in sludges are underestimated. Koutnik et al. postulated that 96% of the microplastics in wastewater sludge may go undetected due to methodological shortcomings.[98] Once therein, stabilization practices, land application, and later environmental processes will fragment these into ever greater numbers of smaller particles, including nanoplastics. These are likely more chemically and toxicologically active, but more difficult to detect.[2,5,99]

WWTP solid stabilization approaches (designed to reduce malodors, pathogens, vector attraction, and water content) prior to land application range from minimal to combinations of thermal hydrolysis, anaerobic digestion, liming, dewatering, thermal drying, and other techniques.[100] Impacts of these practices on the stability, properties, bioavailability, and toxicity of associated PBTs, additives, and microplastics merit scrutiny. Increased temperatures and levels of dissolved organic carbon, both common in wastewater/sludge treatment schemes, can increase the level of leaching of hydrophobic additives from microplastics.[5] Subsequent application of sludge-derived biosolids deposits incorporated contaminants into soils.[101] Surface runoff, groundwater infiltration, and aeolian processes will eventually redistribute these across environmental compartments,[3,101−105] including the waters that WWTPs aim to protect (Figure 1).

Nizzetto et al. estimated that land application of biosolids deposits 44000−300000 tons of microplastics in North America and 63000−430000 tons in Europe annually on agricultural soils.[90] The wide ranges reflect the crudeness of existing data. They speculated that this total exceeds that believed to be present in the surface waters of the world ocean. With respect to PBTs, Venkatesan and Halden postulated the land application of biosolids added ∼30000−45000 kg of BFRs to U.S. agricultural soils.[106] The potential for PBT and microplastic dilution is lower in terrestrial than marine environments, as on land most is constrained to a few centimeters of the surface.[107] In contrast, the global ocean, with its immense surface area and depths, provides a far larger dispersal volume. Much of the microplastic entering the ocean will eventually sink due to their innate densities or following biofouling or biological ingestion.[108]

The choices for disposal of WWTP solids control the extent of environmental introduction and subsequent fate of the associated contaminants. For example, FOG and primary treatment solids contain >50% of the microplastics and PBTs entering WWTPs.[81,101,102] Hence, their exclusion from biosolids destined for land application would substantially decrease the rate of introduction of microplastics into agricultural soils.

METHODS FOR SAMPLING AND QUANTIFYING MICROPLASTICS IN WWTP MATRICES REMAIN INADEQUATE

While efforts to investigate microplastics in wastewaters, sludges, and effluents are increasing, available analytical methods remain inadequate to detect the full range of sizes, shapes, and types (i.e., polymer composition) present.[99] Microplastics <100 μm in size are often unretained by sampling and preparation techniques.[109] The small size of microplastics and the complexity of wastewater-related matrices complicate accurate identification and quantification of microplastics.[98,99] Polymer types beyond the "usual suspects" (i.e., polyethylene, polypropylene, PET, PS, and polyvinyl chloride) often go unquantified. Understudied polymer classes include paint/coating-associated urethanes, acrylates, and butadiene-based rubbers common in tire wear.[81,82,99,109] These may be toxicologically relevant (e.g., the tire antioxidant additive degradant described previously).[44] Sample preparation techniques for removing analytical interferences, such as aggressive oxidation, may alter some polymers, e.g., those derived from polyamides, biopolymers, and tire wear.[99,109] Fibers from clothing and carpeting laundering are a dominant source of microplastics to municipal wastewaters.[79,110] Textiles and fabrics may also be treated with stain and water repellant (e.g., PFAS), flame retardant, colorizing compounds, plasticizers, and other additives.[31] Microfibers are particularly difficult to collect and detect due to their small cross-sectional areas and physical similarity to natural polymers.[99,111] Cellulose-derived polymers (e.g., rayon/viscose) are common in both natural and wastewaters but often are excluded from surveys of microplastics.[99,112] The mechanisms responsible for human and ecosystem health effects from microplastic exposure (be they physical such as the obstruction of, or injury to, digestive or respiratory surfaces or chemical toxicity-mediated) merit investigation.[5] Accord-

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ingly, it may be premature to ignore possible health consequences of natural and "naturally derived" polymers, especially because of their high abundances.[5,112] Ultimately, due to the limitations of sampling and analysis, existing microplastic levels in environmental media are underestimated.[98,99,109] This, and the increasing plastic volumes being produced and discarded, suggest that mandating studies evaluating toxicological effects be confined to currently available "environmentally relevant" concentrations is ill-advised.

CONCLUSIONS

Durable plastic products are abundant in built environments, especially indoors, and are major sources of microplastics and polymer additives. Humans in developed nations spend most of their lives indoors and are exposed to contaminated air and dust. Pollutants enter WWTPs, forming complex and variable mixtures. PBTs and microplastics in wastewater discharges concentrate in the resulting settled solids. An emerging area of inquiry involves the effects of biosolid contaminants on soil health, food safety, and ecosystem services.[31,90,101,113] The continuous supply and persistence of microplastics and polymer additives ensure that effects on human and environmental health will be long-term and increasing as plastics fragment and additives become more bioaccessible. This has been dubbed the "global plastic toxicity debt".[2] Once environmentally dispersed, these contaminants are increasingly difficult to remediate. Thus, gaining a better understanding of and preventing and mitigating releases are imperative. Analytical methods for detecting and characterizing residues in environmental matrices must improve to reveal the true extent of microplastic and PBT additive contamination. The manufacture and release of plastics/microplastics and additives are increasing, especially in developing nations. This, and associated health repercussions, should be factored into policies regarding WWTP discharges, effluent monitoring, and generation and reuse of wastewater residuals. As treatment and disposal practices in developing nations lag behind those in affluent countries, local receiving waters and soils there will be most immediately and seriously affected. However, microplastics and PBTs will eventually be disseminated by transport processes, resulting in regional and ultimately global environmental repercussions.

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Notes

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