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Persistent, mobile, and toxic plastic additives in Canada: properties and prioritization†

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The hazards of many plastic additives on human and environmental health are well documented. However, little emphasis has been put on plastic additives that are persistent, mobile, and toxic (PMT) rather than persistent, bioaccumulative, and toxic. Due to their high mobility and stability, it is unlikely that wastewater treatment plants will effectively remove PMT plastic additives. Herein, an *in silico* analysis was performed to (1) assess the retention of PMT plastic additives registered for use in Canada in wastewater treatment plants; and (2) determine whether their physical–chemical properties and structural features can be used as identifiers for PMT plastic additives with particularly low retention. We identified 124 PMT plastic additives of which 52% had less than 20% removal from wastewater treatment based on predictions using the model SimpleTreat. Log K_{aw}, log K_{ow}/D_{ow}, and log K_{oc}/D_{oc} ranges were defined that are indicative of low retention PMT plastic additives. Furthermore, it was found that non-halogenated PMT plastic additives that contain nitrogen are most likely to be poorly retained in wastewater treatment plants. The results of this study provide screening and prioritization criteria, as well as a suspect list for PMT plastic additives.

Environmental significance

Persistent, Mobile and Toxic (PMT) substances have been recognised as having the potential to impact environmental and human health. However, there are significant challenges associated with their identification, quantification, and assessment in the environment given the amount of potential PMT substances and the diversity of chemistries and properties they encompass. Here we present a method to identify and prioritize PMT substances based on their physical–chemical property space, structural identifiers, use category, and *in silico* retention in wastewater treatment plants – with a focus on PMT plastic additives registered for use in Canada. The results can be used by the scientific community, regulators, and industry alike to screen chemicals for potential PMT characteristics.

1. Introduction

With the rise in global plastic pollution, plastic-associated contaminants have become an increasing threat to the environment and human health.[1–7] Plastics consist of two main components: an organic polymer matrix serving as the main plastic structure, and additives used to impart specific physical–chemical properties.[8] The most common types of these additives are functional additives (*e.g.*, antioxidants, flame retardants, plasticizers), colorants, fillers, and reinforcement agents.[9,10] Their contribution to the plastic (per mass) can range from less than one percent to fifty percent or more of the total plastic composition.[10,11]

Many functional plastic additives are not covalently bound to the polymer they are added to.[8] This means that these compounds can be released from the plastic polymer over time and enter the surrounding environment.[8,10] This leaching into the environment is concerning as many functional organic plastic additives have been associated with health effects such as different cancers, endocrine disruption, adverse effects to reproduction and development, as well as general toxicity in a variety of organisms.[1,2,12,13]

Given their potential toxicity and detection in the environment, some plastic additives have been identified as potential contaminants of concern or even as persistent organic pollutants (POPs).[13] Nevertheless, many hazardous plastic additives are not restricted.[4,13] The current regulations in Canada under the Canadian Environmental Protection Act (CEPA) control the emissions of many hydrophobic plastic additives, if they meet the regulatory criteria for persistence, bioaccumulation potential, and toxicity (PBT).[14] Moreover, plastic pollution has become a regulatory priority for Canada leading to the creation of a comprehensive "Plastic Science Agenda"[15] as well as the "Zero Plastic Waste Initiative".[16] There is a regulatory safeguard in

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Canada against the environmental risk of many plastic additives that are bioaccumulative but not for plastic additives that are persistent, *mobile in water*, and toxic (PMT).[17,18]

PMT substances are toxic polar organic compounds that are persistent in water and can therefore pose a long-term risk to the aquatic environment.[17] Due to their polarity, generally low volatility, and low potential to sorb to organic matter or particles, emitted PMT substances remain in water or can penetrate groundwater aquifers and threaten the quality of drinking water sources.[17,19,20] Moreover, the high environmental stability of PMT substances can enable them to undergo long-range water-based transport into remote environments such as the Arctic Ocean.[18] The German Environmental Protection Agency (UBA) has released the following criteria for the identification of PMT substances:[19]

A substance is considered persistent (P) if (a) the degradation half-life in marine water at 9 °C is higher than 60 days, (b) the degradation half-life in fresh or estuarine water at 12 °C is higher than 40 days, (c) the degradation half-life in marine sediment at 9 °C is higher than 180 days, (d) the degradation half-life in fresh or estuarine water sediment at 12 °C is higher than 120 days, or (e) the degradation half-life in soil at 12 °C is higher than 120 days.[19]

A substance is considered mobile (M) if the lowest experimental organic carbon–water coefficient log K_{oc} over the pH range of 4–9 is less than 4.0.[19]

A substance is considered toxic (T) if it meets any of the criteria for toxicity set out in Annex XIII, 1.1.3 of the European Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) regulatory framework. These criteria are (a) the long-term no-observed effect concentration (NOEC) or EC_{10} for marine or freshwater organisms is less than 0.01 mg L^{-1}, (b) the substance meets the criteria for classification as carcinogenic (category 1A or 1B), germ cell mutagenic (category 1A or 1B), or toxic for reproduction (category 1A, 1B, or 2), or (c) there is other evidence of chronic toxicity.[19]

Because PMT substances are highly mobile in water, wastewater treatment plants (WWTPs) are not expected to retain PMT substances and thus release them into the aquatic environment.[20] PMT substances entering closed and semi-closed water cycles may even concentrate over time, with dilution being the only process able to reduce concentrations.[17] However, the identities and potential retention of different PMT substances in WWTPs are currently unknown. In this research, we aimed to (A) identify PMT substances used in Canada, (B) identify which of those were plastic additives, (C) model their retention in WWTPs, and (D) outline physical–chemical properties that can be used as screening criteria to identify them.

2. Methods

2.1 Developing a list of PMT plastic additives registered for use in Canada

A comprehensive list of PMT plastic additives registered for use in Canada was developed based on the PMT criteria and PMT suspect list published by UBA.[19] This "UBA PMT list" (available on the Norman Network, **https://www.norman-network.net**) was combined and compared to the Canadian Domestic Substances List (DSL) using CAS numbers to create a list of PMT substances registered for use in Canada (Table S1†). This list was then compared to the PlasticMAP database published by Wiesinger *et al.*[8] that contains over 10 000 plastic monomers, additives, and processing aids to create a list of PMT plastic additives registered for use in Canada (Table S2†). Physical–chemical property data for PMT substances was collected from Arp *et al.*[21] as well as the CompTox Chemistry Dashboard by the US Environmental Protection Agency[22] for property data not available in the Arp *et al.* publication.

2.2 Prioritizing PMT plastic additives based on their predicted removal efficiencies from WWTPs

The open access wastewater treatment model SimpleTreat 4.0 (ref. 23) was used to model the removal efficiencies of the PMT plastic additives from wastewater effluent to assess their potential for human and environmental exposure. SimpleTreat is a fugacity mass-balance model that predicts the removal of a chemical in a WWTP with conventional primary and secondary treatment as well as its partitioning into different media (air, sludge, effluent) during the treatment process using physical–chemical property data and information on biodegradation.[23] For ionizable substances, SimpleTreat determines the fraction of the substance that is in acid/neutral/base form using the pK_a of the substance and the pH that are input.[23] The model only considers neutral molecules for partitioning through the air–water boundary, and as such, the ionic fraction of a substance is not considered for any type of volatilization. Consequently, the ionic fraction is only modeled based on the partitioning between the aqueous and sludge phases.[23] For each substance, the following physical–chemical properties were retrieved from Arp *et al.*,[21] CompTox Chemicals Dashboard, or the European Chemical Agency (ECHA) database (**https://comptox.epa.gov/dashboard/dsstoxdb/batch_search**, **https://echa.europa.eu**) and were inputted into SimpleTreat 4.0: molecular weight (g mol^{-1}), octanol–water partition coefficient (K_{ow}/D_{ow}), organic carbon–water partition coefficient (K_{oc}/D_{oc}), water solubility (mg L^{-1}), vapor pressure (Pa), Henry's law constant (Pa m^3 mol^{-1}), biodegradation half-life (days), and pK_a for ionizable compounds. D_{ow}, D_{oc}, and water solubility were the minimum measured or predicted values at a pH between 4 and 10, while vapor pressure and K_{aw} values were measured or predicted at 298 K.[28] K_{ow}/D_{ow} values were only used whenever K_{oc}/D_{oc} values were not available. This was done in accordance with Arp *et al.*,[21] who estimated log K_{oc} from log K_{ow} using the linear relationship between the two when an experimental log K_{oc} was unavailable. The default settings in SimpleTreat 4.0 were used for the 'Mode of operation' and 'Emission scenario'.

The removal of PMT substances in wastewater treatment plants was modelled using two scenarios. For the first scenario, all PMT substances were classed as non-biodegradable based on their regulatory classification as "persistent". This constitutes a 'worst case scenario' as a number of PMT substances on the UBA list are expected to be at least moderately degradable in

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wastewater treatment plants[19] (Table S2†). For the second scenario, predicted or, where available, measured biodegradation half-lives (retrieved from CompTox) were used to model the removal efficiency of the PMT compounds (Table S3†).

Once the removal efficiencies were acquired (Table S3†) the PMT substances were ranked based on their estimated emissions *via* effluent (Table S4†). The 'emission *via* effluent' was used to group the PMT substances into four emission groups: <10%, 10–50%, 50–80%, and 80–100% emission. The 95% percentile ranges were calculated for each group to identify any overlapping groups.

Additionally, the partitioning of PMT plastic additives into sludge and air was investigated. The amount of PMT substances that partitioned into sludge was determined by taking the sum of the SimpleTreat output for percent elimination of the substance *via* primary and surplus sludge. The amount of PMT substances that partitioned into air was determined by taking the sum of the SimpleTreat output for percent elimination by volatilization and stripping (Table S3†).

2.3 Identifying the physical–chemical property space and use of PMT plastic additives

The physical–chemical property space of PMT plastic additives with high emission *via* effluent (80–100% emission) was investigated to identify the combination of $\log K_{oc}/D_{oc}/K_{ow}/D_{ow}$, $\log K_{aw}$, and vapor pressure that is indicative of PMT substances with low retention in WWTPs. Wherever available $\log K_{oc}/D_{oc}$ were used. If multiple values were available for $\log K_{oc}/D_{oc}/K_{ow}/D_{ow}$ (at pH 4 to 10), minimum values were used as a "worst-case scenario" with regards to potential mobility.[21] Since $\log K_{oc}/D_{oc}/K_{ow}/D_{ow}$ and water solubility are negatively correlated, only $\log K_{oc}/D_{oc}/K_{ow}/D_{ow}$ was used in the analysis as there was more experimental data for it compared to water solubility. The resulting physical–chemical property space for the partitioning into air, sludge and effluent were combined to determine the dominant fate of the individual PMT plastic additives in WWTPs.

Additionally, the individual use functions of the identified PMT plastic additives were collected from the PlasticMAP published by Wiesinger *et al.*[8] (Table S5†).

2.4 Statistical analysis and sensitivity analysis

Comparison and filtering of all data as well as the calculation of statistics and chemical space plots were conducted using Microsoft 356 excel for enterprise. Further data analysis and graphics were created using R Studio version 1.3.1093.

A sensitivity analysis based on the method by Morgan and Henrion[24] was performed to assess the sensitivity of the total modelled emissions through effluent to the variability in input data. The sensitivity to a specific input parameter was tested by individually changing the median of the respective input parameter one at a time by 10% and calculating the impact on the total emission *via* the effluent. The sensitivity (*S*) of the emissions *via* effluent to an individual parameter was quantified using:

$$S_{EE} = \frac{\Delta EE}{EE} \frac{I}{\Delta I} \qquad (1)$$

with S_{EE} defined as sensitivity of the emission *via* the effluent; ΔEE defined as changes in the emission *via* the effluent, I as the initial input parameter value and ΔI as the change in input parameter value.

2.5 Quality assurance

Given the high variability of physical–chemical property data amongst different sources and databases, collecting data from a single source was important to ensure consistency of the data. For this reason, the database ranking REACH-registered substances by their known or suspected persistency and mobility, published by Arp *et al.*,[21] was used to collect nearly all the physical–chemical property data for this study (Table S2†). While this database does contain both experimental and modelled data, the aim was to transfer the consistency by which those data were collected to this study. Only two of the identified PMT plastic additives (CAS 156-60-0 and CAS 57-41-0) were not present in the Arp *et al.*[21] database. For these substances, physical–chemical property data was collected from the CompTox Chemistry Dashboard[22] or ECHA (https://echa.europa.eu). Biodegradation half-lives were collected from the CompTox Chemistry Dashboard[22] for all substances included in this study.

3. Results

3.1 Estimated removal of PMT plastic additives from wastewater in relation to their properties

In total 208 of the 260 PMT substances identified by UBA are currently registered for use in Canada (Table S1†). One hundred and twenty-four (124) of these compounds (60%) were classified as plastic additives (Tables 1 and S2†). Among these, 50% contained nitrogen, 33% were halogenated, 13% contained sulphur, 4% contained phosphorus, and 3% contained silicon (Table S3†). The physical–chemical property ranges of the identified PMT plastic additives are presented in Table 1.

The modelled emission *via* effluent for the 124 PMT plastic additives ranged from 0% to 100% with the majority of PMT plastic additives having median emissions *via* the effluent of over 75% (Fig. 1).

There were 64 compounds (52% of the PMT plastic additives) with an 80–100% emission *via* effluent (Table 1). Of these, 26 (42%) were ionizable, 24 (38%) were neutral, and 14 were amphiprotic/polyprotic (Table S2†); 69% contained nitrogen, 19% contained sulphur, 23% were halogenated, and 3% contained phosphorus, and none contained silicon (Table S3†). There were 24 compounds with a 50–80% emission *via* effluent, 18 compounds with emissions *via* effluent between 10–50%, and 18 compounds with emissions *via* effluent of <10% (Table 1 and Fig. 2). Similar ranges for $\log K_{oc}/D_{oc}/K_{ow}/D_{ow}$ and $\log K_{aw}$ were observed for both experimental and predicted values (Fig. S2†).

In addition to the 64 compounds with ≥80% emission in the effluent, there were 21 compounds with ≥80% emission in the

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Table 1 Physical–chemical property range of PMT plastic additives registered for use in Canada, and 5th to 95th percentile property space for with PMT plastic additives ≥80% effluent emission, 50–80% effluent emission, 10–50% effluent emission, or ≤10% effluent emission from a modeled WWTP

| Property range	Total PMT plastic additives	≥80% effluent emission	50–80% effluent emission	10–50% effluent emission	≤10% effluent emission
Log K_{ow}/D_{ow} (median)	−11 to 9.9 (1.9)	−3.9 to 4.1 (0.87)	−7.1 to 4.4 (1.1)	0.33 to 5.7 (2.8)	1.4 to 4.7 (2.5)
Log K_{oc}/D_{oc} (median)	−8.3 to 4.0 (1.7)	−3.7 to 3.1 (1.3)	−0.32 to 3.7 (2.2)	0.42 to 3.6 (2.4)	0.60 to 3.2 (1.8)
Log K_{aw} (median)	−32 to 2.4 (−4.3)	−16 to −2.3 (−6.3)	−13 to −2.0 (−5.2)	−6.2 to −0.54 (−1.3)	−2.4 to 1.3 (−0.27)
Water solubility (median) [mg L^{-1}]	4.2×10^{-5} to 3.3×10^{12} (3.3×10^3)	0.043 to 1.9×10^{10} (5.8×10^3)	3.7 to 6.7×10^8 (4.1×10^3)	15 to 1.1×10^{10} (1.1×10^3)	41 to 3.5×10^5 (1.3×10^3)
Vapor pressure (median) [Pa]	1.2×10^{-23} to 6.5×10^5 (3.0)	8.1×10^{-20} to 2.2×10^3 (6.0×10^{-2})	1.7×10^{-8} to 1.9×10^3 (0.20)	1.4×10^{-3} to 2.2×10^4 (2.3×10^2)	2.0×10^2 to 5.9×10^5 (1.3×10^4)
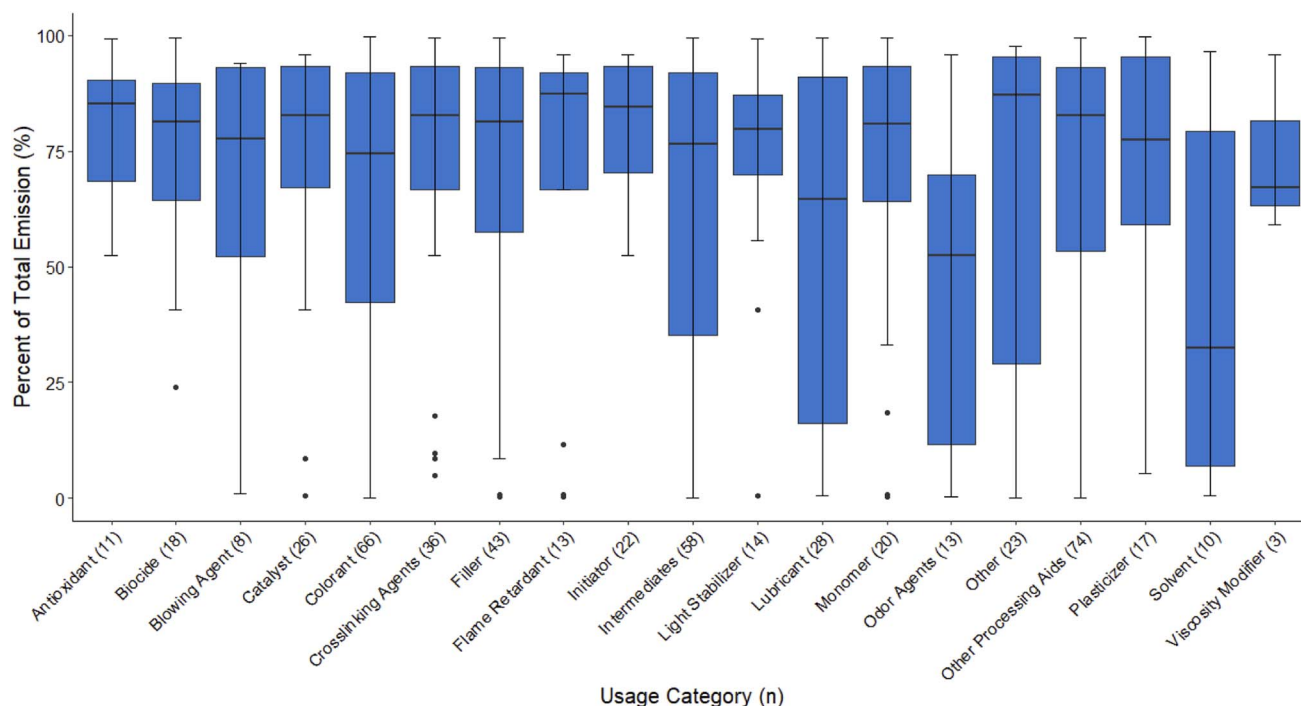
Fig. 1 The distribution of effluent emission (as a percentage of total emission) of 124 PMT plastic additives, based on their use category.
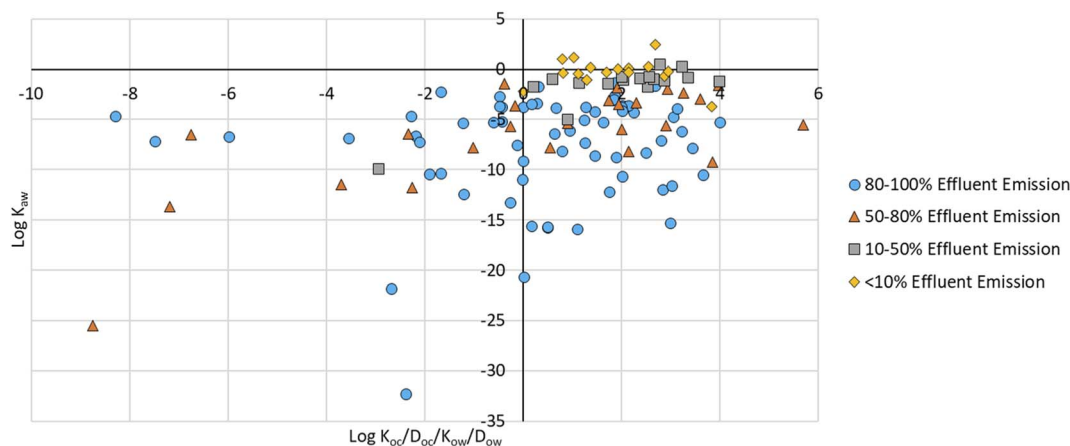

Fig. 2 Log $K_{oc}/D_{oc}/K_{ow}/D_{ow}$ versus log K_{aw} graph of 124 PMT plastic additives plotted according to their predicted % emission via effluent in a WWTP simulation.

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Table 2 Physical–chemical property space (5th to 95th percentile) of PMT plastic additives registered for use in Canada with ≥80% emission through effluent or air from WWTP, or ≥40% partitioning into sludge

Property range	≥80% emission *via* effluent	≥80% emission *via* air	≥40% partitioning into sludge
Log K_{ow}/D_{ow}	−3.9 to 4.1	1.4 to 4.4	5.7 to 6.0
Log K_{oc}/D_{oc}	−3.7 to 3.1	0.51 to 3.1	3.9 to 4.0
Log K_{aw}	−16 to −2.4	−2.2 to 1.1	−8.9 to −2.0
Water solubility [mg L^{-1}]	0.043 to 1.9×10^{10}	47 to 6.7×10^{4}	0.65 to 1.8×10^{4}
Vapor pressure [Pa]	8.1×10^{-20} to 2.2×10^{3}	56 to 5.8×10^{5}	1.7×10^{-4} to 3.7×10^{-3}

air, and three compounds with ≥40% partitioning into the sludge (Table 2).

3.2 Estimated removal of PMT plastic additives based on their use category

Use functions for the PMT plastic additives were taken from the PlasticMAP database published by Wiesinger *et al.*[8] This database is composed of over 10 000 plastic monomers, additives, and processing aids, and was created by compiling and filtering information from 63 data sources. Wiesinger *et al.* assessed the quality of the information in this database based their origin, and entries were assigned an appropriate confidence level (low, medium, high). For the use function information gathered from the PlasticMAP database for the 124 PMT plastic additives in this study, 113 compounds (91%) had a high confidence score, 10 compounds had a medium confidence score, and one compound (*N*-methyl-benzenamine) had a low confidence score. Additionally, the 10 compounds with a medium confidence score had no defined use function, and as such, fell under the "other" category.

These use categories covered 101 of the 124 PMT plastic additives, with the remaining 23 compounds falling under the "other" category. Of these 101 PMT plastic additives with a known use, 89 fell into more than one category, with a total of 504 different entries when separated by use category (Table S5†). Most of the 101 PMT plastic additives fell into the 'other processing aids' (60%), 'colorants' (53%), and 'intermediates' (47%) use categories (Fig. S3†). This was true for PMT plastic additives in general as well as for PMT plastic additives with a high predicted emission through wastewater effluent (Fig. S3†). The use categories with the fewest PMT substances were 'heat stabilizers' and 'antistatic agents' with one substance each (Table S5†).

A majority of the use categories showed the greatest amount of PMT emissions to the effluent, followed by air, and lastly, partitioning into the sludge (Fig. 3). Odor agents and solvents had notably lower effluent emissions compared to the other categories (Fig. 3). While effluent emission was lower for odor agents compared to most other use categories, the median (53%) emission into effluent was still greater than that of air (32%) and sludge (0.14%), meaning effluent was the primary emission pathway. Solvents were the only use category in which air, rather than effluent, was the primary emission pathway (median 59%) (Fig. 3).

For all other use categories, air was the second-most prevalent emission pathway, with emissions between 0 and 95% for 9 of the 19 use categories (Fig. S4†). These categories were blowing agents, colorants, intermediates, lubricants, monomers, odor agents, other processing aids, viscosity modifiers, and other. PMT plastic additive partitioning into sludge was low for PMT plastic additives from all use categories (Fig. S4†). Antioxidants had the highest median partitioning into sludge with 10% (Fig. 3).

Of the 124 PMT plastic additives, 23 fell under the "other" use category, as a usage function could not be identified for these substances. Despite this, they followed the same general pattern observed in the other use categories (effluent emission > air emission > sludge partitioning).

4. Discussion

In this study, we identified 124 PMT plastic additives registered for use in Canada with 64 substances with less than 20% retention in WWTP. Some of these low retention PMT plastic additives were known organic water contaminants, such as sulphanilic acid and dapsone,[25,26] while others are new and emerging contaminants that have been scarcely studied, such as 1,3-di-*o*-tolylguanidine and trifluoromethanesulfonic acid (Table S2†). In previous research, Schulze *et al.*[25] used an environmental emission score (E-score) calculation based on the annual tonnage and use characteristics of the substances to rank the likelihood of a persistent, mobile organic contaminant (PMOC) to be emitted into the environment. 61 of the 124 PMT plastic additives in this study were also ranked previously by Schulze *et al.*[25] There were similarities between the relative ranks of the common substances in the two lists, such as sulphanilic acid, 1,4-diazabicyclo[2.2.2]octane and pigment yellow 83 ranking within the top 20% of both lists. With that being said, the relative ranks of many of the other substances did not align. This is likely due to the disparity in the methods by which the rankings of the two lists were performed. Since E-score focusses on annual tonnage, use characteristics, and the physical–chemical properties of a substance, the rankings done by Schulze *et al.* provide a more thorough assessment of emission potential. In contrast, the analysis performed in this study focusses on the partitioning and removal from a typical WWTP. As such, these rankings are complementary to one another and combining such ranking or scoring systems in the future may provide a more comprehensive list of substances for environmental screening and regulation.

Most of the top 20 ranked PMT plastic additives in this study had high PMOC and mobility scores (>3) based on the
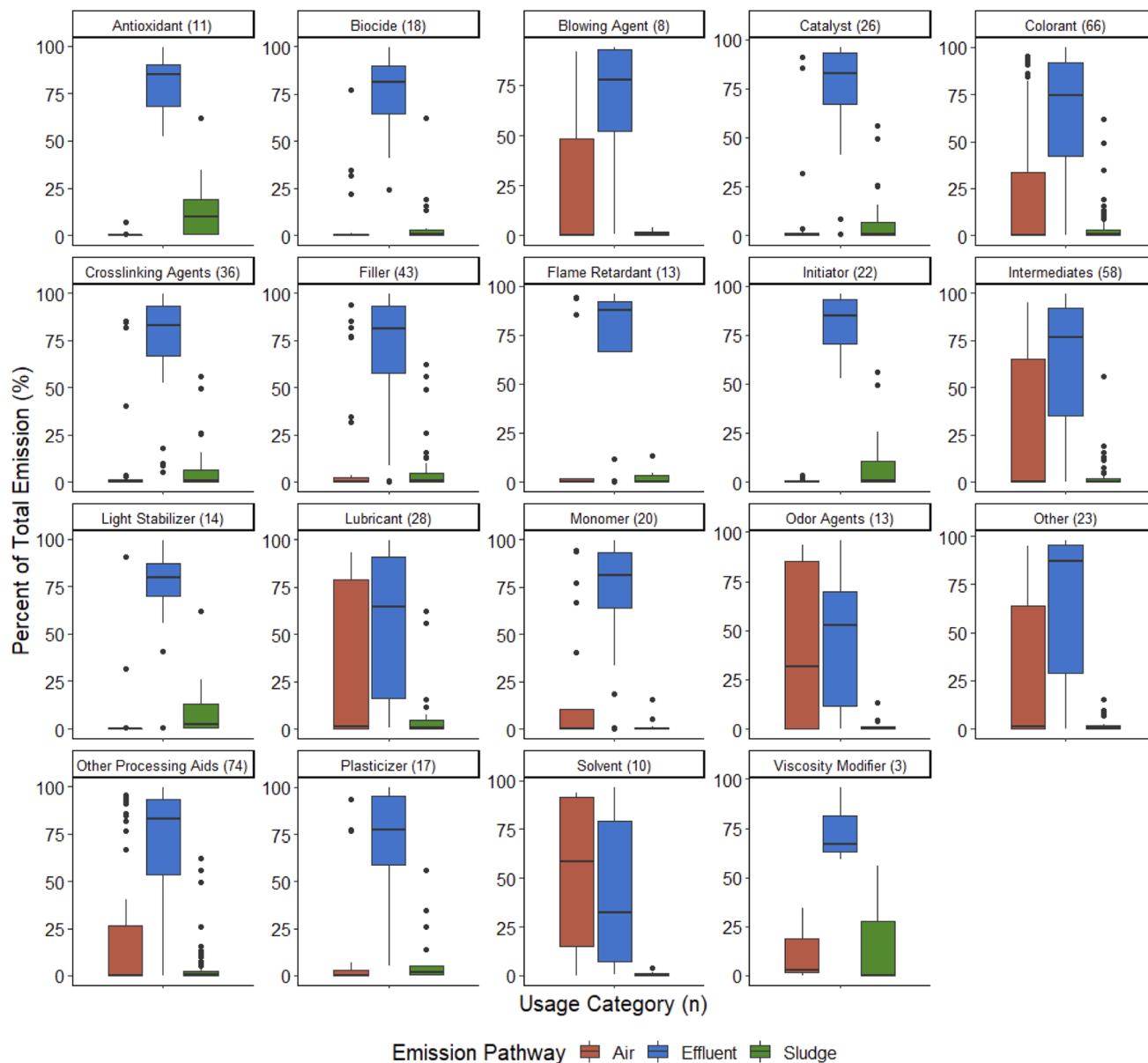
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Fig. 3 The distribution of emission data for the 124 PMT substances, based on the three emission pathways (air, effluent, sludge) from a theoretical WWTP and the known use categories of each substance.

classification proposed by Arp *et al.*[21] (Table S2†). However, there were some notable discrepancies, such as for organophosphate esters that were ranked as "unstable mobile organic compounds" in Arp *et al.*[21] whereas many of them were ranked highly among the PMT substances in this analysis. The degradability of organophosphate esters has recently been questioned due to their prevalence in Arctic environments.[18,27] Based on these findings, it has been suggested that organophosphate esters are PMOC substances rather than unstable mobile compounds.[28] The differences in the respective rankings and the omission of many of the top 20 ranked PMT plastic additives in the environmental emission score calculation by Schulze *et al.*[25] highlights the importance for combinations of prioritization mechanisms as well as specific local foci to

understand and manage potential risks of emerging contaminants such as PMT plastic additives.

4.1 Prioritization of PMT plastic additives

The results of our research showed that PMT substances are used in Canada and 60% of them can be used as plastic additives. In an assessment of over 10 000 plastic additives, monomers, and processing aids used on the global market, Wiesinger *et al.*[8] showed that only 57 were considered either PBT or very persistent and very bioaccumulative (vPvB). Conversely, we identified 124 plastic additives registered for use in Canada that meet or potentially meet the criteria for PMT. Considering that PMT substances can pose a risk to drinking water security[17,29] and that PMT substances can be considered an equivalent level

of concern to substances of very high concern (SVHCs) under REACH[20] the use of PMT substances in Canada clearly merits further investigation. This is particularly important for the 64 PMT plastic additives with predicted emissions through wastewater effluent of >80%. These poorly retained that are persistent and toxic could pose a poorly reversible risk to drinking water security and the quality of receiving aquatic environments.

Considering the number of identified PMT plastic additives and range of uses, prioritization is key to address potential risks for human health and the environment from compounds that are most susceptible to pass through WWTPs and into surface and drinking waters. In Section 73 (1)[30] under 'Priority Substances and Other Substances', the Canadian Environmental Protection Act (CEPA) states that substances should be categorized that "(a) may present, to individuals in Canada, the greatest potential for exposure; or (b) are persistent or bioaccumulative in accordance with the regulations, and inherently toxic to human beings or to non-human organisms, as determined by laboratory or other studies". While mobility is not currently a parameter for prioritization, many PMT substances meet one or both criteria to some extent. Classifications like this in the CEPA, along with prioritization mechanisms based on the expected retention in WWTPs enable prioritization of PMTs for water quality monitoring and alterations to their use in industry.

4.1.1 Using the physical–chemical space to identify and prioritize future PMT plastic additives. PMT plastic additives share common physicochemical properties ($\log K_{ow}/D_{ow}/K_{oc}/D_{oc}$, $\log K_{aw}$, and vapor pressure) that can be used to identify them. As expected, substances that were polar and non-volatile

($\log K_{ow}/D_{ow}/K_{oc}/D_{oc}$ between -3.7 and 4.1, and $\log K_{aw}$ between -16 and -2.3) were primarily emitted in the effluent, while those that were polar and volatile ($\log K_{ow}/D_{ow}/K_{oc}/D_{oc}$ between 0.51 and 4.4, and $\log K_{aw}$ between -2.2 and 1.1) were primarily emitted in the air, and those that were hydrophobic ($\log K_{ow}/D_{ow}/K_{oc}/D_{oc}$ between 3.9 and 6.0) primarily partitioned into the sludge (Table S3†). Interestingly, substances primarily emitted in the effluent not only covered the low $\log K_{ow}/D_{ow}/K_{oc}/D_{oc}$ values, but also contained several substances with higher $\log K_{ow}/D_{ow}$ values up to a maximum of 4.4. This was likely due to the lack of $\log K_{oc}/D_{oc}$ values for a 48 of the 124 PMT plastic additives, where $\log K_{ow}/D_{ow}$ values had to be used, extending the property range above what would be expected (<4.0) (Fig. 4).

The observed property ranges can be used in the future for the initial screening of PMT substances entering WWTPs. The property ranges (Fig. 4) indicated that as a compound's percent emission *via* effluent increased, its water solubility increased (lower $\log K_{oc}/D_{oc}$) and volatility decreased (lower $\log K_{aw}$). Specifically, PMTs with a $\log K_{oc}/D_{oc}$ between -3.7 and 3.1, and a $\log K_{aw}$ between -16 and -2.4 at a 90th percentile were highly emitted in the effluent (80–100%). However, it is important to note that several PMT plastic additives were also modelled to be emitted to a considerable extent *via* air (Fig. 4).

While emission *via* air was expected for PMT plastic additives in use categories that have typically volatile compounds, such as solvents or odor agents, emission into air also occurred for PMT plastic additives in the lubricants, colorants, and intermediates use categories (Fig. S4†). Overall, 69% of the PMT plastic additives had ≥80% emissions from WWTP through either effluent or air (Table S3†) further highlighting the low retention of these compounds. The physical chemical property
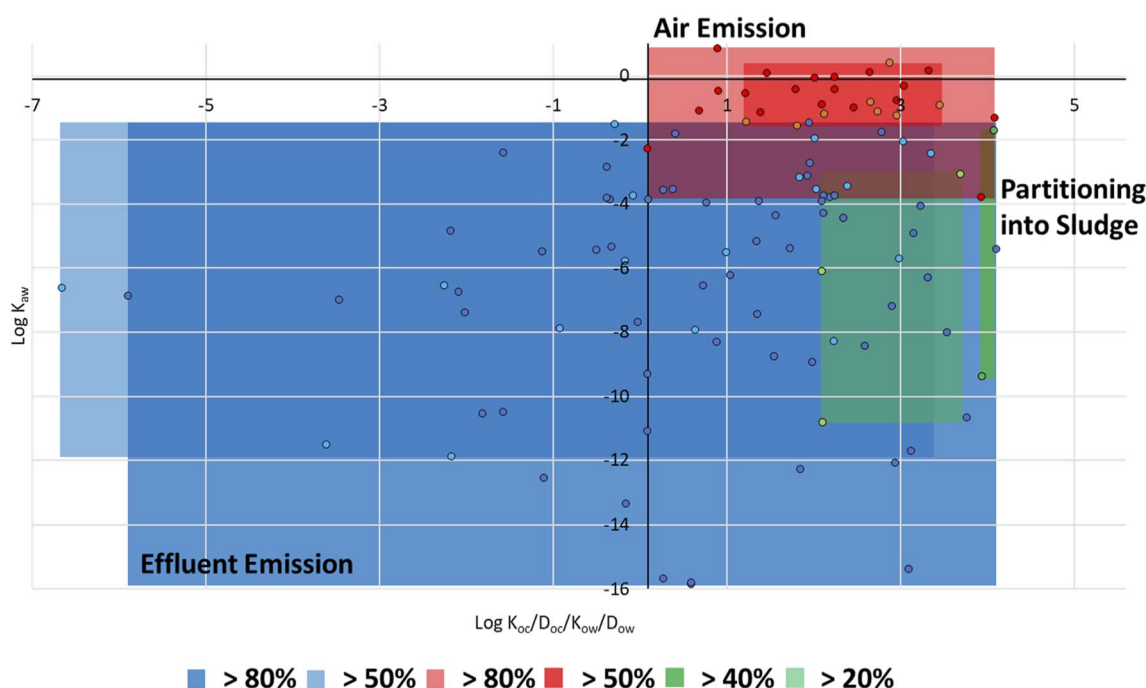

Fig. 4 The partitioning space plot showing the dominant emission pathways (effluent, air, sludge) of 124 PMT plastic additives at a 90th percentile.

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ranges that were identified as indicative for high emission PMT plastic additives through air, effluent, or 40% partitioning into sludge are presented in Table 2.

4.1.2 Prioritization based on structural identifiers and use categories. In total, 62 of the PMT plastic additives contained nitrogen, 40 were halogenated, 16 contained sulphur, five contained phosphorus, and four contained silicon (Table S3†). There was a clear difference between common structural components in high emission PMT plastic additives compared to the overall PMT plastic additives. 69% of the high emission PMT plastic additives (80–100% emission) contained nitrogen, indicating that PMT plastic additives that contain nitrogen have a high potential to be released in the effluent from WWTPs (Table S3†). Halogenated compounds, on the other hand, only made up 23% of the high emission PMT substances, while they constituted 42% of the PMT plastic additives with <80% emissions through the effluent (Table S3†). The proportion of sulphur and phosphorus containing PMT plastic additives was similar between the high emission PMT plastic additives and the overall PMT plastic additives with <80% emissions *via* the effluent (Table S3†), indicating that the presence of these atoms does not have a strong impact on the ability of a PMT plastic additive to move through wastewater treatment plants. Silicon was only present in PMT plastic additives with <10% emission *via* the effluent. However, all of the silicon containing compounds had >80% predicted emissions *via* air, highlighting the importance of this emission pathway for certain chemicals (Table S3†).

The majority (88%) of PMT plastic additives fell under more than one use category (Table S5†). This is due to the diverse physical–chemical properties that PMT substances possess and speaks to the versatility and ubiquitous use of PMT plastic additives. All use categories apart from odor agents and viscosity modifiers had median modelled emission through effluent of >60% showing the widespread use of potential high-emission PMT plastic additives (Fig. 1). However, while some categories showed a wide spread of modelled emission, others contained substances where the modelled emissions *via* effluent exceeded 50% for all compounds apart from a few individual outliers (Fig. 1). These product categories with exclusively high-emission PMT plastic additives are antioxidants, crosslinking agents, flame retardants, initiators, light stabilizers, and viscosity modifiers (Fig. 1). Consistent with the overall PMT plastic additive results, the use categories with high-emission PMT plastic additives were dominated by nitrogen-containing compounds (Table S5†). Flame retardants and crosslinking agents were the only categories where halogenated compounds exceeded the amount nitrogen-containing compounds (Table S5†). The use categories provide a sense of what applications and industries the substances originate from, and are therefore an important measure for potential emission reduction efforts. Moreover, the combination of information on physical–chemical properties and use category can provide a valuable indication on whether a PMT plastic additive can be expected to consistently fall into the high-emission PMT plastic additive category.

4.2 Comparison with observational data on PMT plastic additives in WWTP

To assess the accuracy of the SimpleTreat model in predicting the removal of PMT plastic additives from a WWTP, the removal efficiencies of substances from this study were compared to removal efficiencies reported in the literature (Table S7†). The removal efficiencies for 18 substances in this study that were also reported in the literature were recorded and compared. Removal efficiencies reported in the literature were generally higher than the model results from the SimpleTreat analysis, but also highly variable with standard deviations ranging from 0.7 to 23% (Table S7†). There were six substances that had removal efficiencies within 25% of observational data reported in the literature (1*H*-benzotriazole, TCEP, TDCP, atrazine, diuron, and naphthalene), six within 50% the data reported in the literature (anthracene, TCPP, triclosan, TMDD, EDTA, and melamine), and six with a difference greater than 50% from the literature values (TBEP, benzothiazole, benzophenone-3, paracetamol, saccharin, and acenaphthene). The substances in each of these groups had no distinct differences in their physical chemical properties with similar median $\log K_{ow}/D_{ow}$ ($\sim$3), $\log K_{oc}/D_{oc}$ ($\sim$2), $\log K_{aw}$ ($\sim$−6), log vapor pressure ($\sim$−3.5), and no common structural features.

While these results indicate that the SimpleTreat model might underestimate the removal of PMT substances in WWTPs, it is important to note that the literature values originate from a variety of studies, and thus, a variety of different WWTPs and regions. The removal mechanisms of many of these WWTPs may have been more advanced (*e.g.* including tertiary treatment) than the 'typical' primary and secondary WWTP modeled by SimpleTreat, which would explain some of the disparity between the literature values and those found in this study. Additionally, the WWTP influent and effluent collection methods varied greatly amongst the different literature studies, with some taking 24 hour composite samples over a single day or several days, and some taking grab samples (ref. S4–S5, S9, S14, S16 and S25†). Regardless, many of these studies still offer a valuable reference for the removal efficiencies of PMT plastic additives from WWTPs, especially in those studies that surveyed more than one WWTP or region (ref. S4, S5 and S9†). While the comparability of the SimpleTreat results was quite variable for the 18 substances, this is only a small number of the total PMT plastic additives investigated here, and as such, does not reflect the overall accuracy of the study. Further research should consider analyzing PMT substances under different WWTP scenarios/technologies in the model to determine whether the removal efficiencies are more reflective of their literature counterparts.

4.3 Uncertainty of the modelled removal and limitations

4.3.1 Uncertainty. A major source of uncertainty in this study was the use of predicted physical–chemical property data where experimental data was not available. Given that many of the PMT plastic additives investigated here remain largely unreported or under-studied, experimental data for several of the physical–chemical properties was unavailable, and

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predicted values had to be used instead. This led to uncertainty in the modelled removal – particularly with regards to the potential removal through biodegradation. The sensitivity analysis showed that altering $K_{oc}/D_{oc}/K_{ow}/D_{ow}$, K_{aw}, vapor pressure, or water solubility by 10% led to less than 10% change in the overall modeled emissions. Conversely, a 10% change in biodegradation half-live values led to changes in the modeled emissions of over 10% (Table S8†).

To evaluate the uncertainty based on biodegradation, two scenarios were investigated: one where no biodegradation was input for each substance, representing a 'worst case scenario' where no biodegradation occurs at all, and a second scenario where the experimental or predicted biodegradation half-lives for each substance were used. The differences in modeled effluent emission between these two scenarios ranged between 0.1 and 57% (median 4.9%), with 19 of the 124 PMTs having a difference greater than 20% between the two scenarios (Table S4†). While a median 4.9% difference does not represent a large difference between the two scenarios, it does lead to uncertainty about the magnitude of the physical–chemical parameters, including biodegradative half-lives, and how a large difference between the experimental and predicted values may have influenced the results. The sensitivity of the model's results to the highly variable and uncertain environmental half-lives explains some of the discrepancy between observed and modelled emissions for the analysed PMT substances and highlights the need for more experimental biodegradation data in different environments and for different wastewater treatment technologies.[21,31]

Despite this, the two scenarios and comparison with the literature showed a general agreement for the relative ranking of PMT substances with regards to their emissions. This shows that the relevant physical–chemical property ranges offer a valuable tool for PMT plastic additive screening and prioritization. In addition to the uncertainty introduced by the lack of available experimental property and monitoring data, there is an ongoing discussion on how to evaluate the mobility of chemicals, which means that the definition of what a "mobile" substance is might still change in the future.[32] Log K_{oc} is generally considered a suitable measure for assessing the mobility of neutral organic compounds, as well as for some ionic compounds, because soil and sediment organic carbon (and its cation exchange properties) will rule the sorption for these compounds.[33–35] With that said, log K_{oc} can be highly variable, especially for many ionic compounds, based on pH, ion exchange interactions with soil minerals, and weathering effects, to name a few.[36–38] While a log K_{oc} value <4.0 over a pH range of 4–9 is the current metric for mobility assessment by UBA, discussions are underway to reassess the mobility criteria, namely reducing this log K_{oc} value from <4 to <3, as a log K_{oc} value <4 can include several substances which can be highly adsorbed, and thus unlikely to be mobile.[32,33] Nevertheless, log K_{oc} serves as the best measure of mobility for a wide range of compounds. Given the variability and complexity in its measurement, experimental log K_{oc} values are often not available, and instead log K_{ow} or log D_{ow} (for ionizable substances), may be used as an analog to log K_{oc}. Although log K_{ow} and log D_{ow} do not account for the soil sorption properties that log K_{oc} does, they do provide a measure of the hydrophilicity of the compound and thereby its sorption properties.[21,37]

4.3.2 Limitations. An important limitation of this study was that the substances evaluated were all known plastic additives registered for use on the Canadian Domestic Substance list that have been evaluated as persistent and toxic. This means that the study did not cover intermediates, precursors, or transformation products that have PMT properties. Likewise, substances that were not assessed as persistent were not included in the evaluation.

It is also important to note that SimpleTreat models the removal from a conventional WWTP with primary and secondary biological treatment. While this is representative for most conventional WWTPs in Europe and North America,[23] the PMT removal in WWTP with tertiary treatment processes could be underestimated in the current analysis. Likewise, the PMT emissions are likely to be higher than what was estimated in this study if only primary treatment is applied. The limited availability of observational data also significantly limited our ability to validate the modelled removal of the analysed PMT substances. To enhance the overall assessment of PMT substances as potential environmental pollutants it is paramount to increase the availability of observational and experimental data on PMT properties and occurrence. The current shortness of experimental and monitoring data is due to a lack of previous regulatory and scientific focus on PMT substances as a group as well as substantial analytical challenges with regards to the detection and quantification of PMT substances. As emphasized in Reemtsma et al.,[17] there is a significant gap between the typical physical–chemical property ranges amenable to common analysis techniques like reverse phase liquid chromatography (RPLC) and gas chromatography (GC), and the physical–chemical property ranges of PMT substances. Techniques like mixed-mode liquid chromatography (MMLC) and hydrophilic interaction liquid chromatography (HILIC) are beginning to show promise in analyzing these very polar substances, but many studies only apply this to a small library of PMT substances.[39,40] This problem is extended further when considering the number of PMT transformation products that can arise from typical WWTP processes like ozonation, or natural transformations that can occur in the environment.[26,29] These transformation products have the potential to be just as, if not more hazardous than their predecessors, further emphasizing the need for robust analytical methods that can identify and characterize such compounds in addition to the growing number of PMT substances.

5. Recommendations

Based on the findings of this study, the use, and emissions of PMT plastic additives and PMT substances in general merit further investigation and environmental monitoring. To understand the fate and potential risk of PMT substances for the environment, water quality, and consequently human health we consider research and development in the following areas as crucial:

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5.1 Development of analytical methods for the identification of PMT substances

Sensitive and robust analytical methods play a key role in the future of researching and monitoring PMT substances. Additional research and analytical techniques, as well as standards are needed to enable the investigation of PMT substances as potential environmental contaminants.

5.2 Investigate the occurrence and dominant sources of PMT substances in the Canadian aquatic environment

The current study resulted in a ranked list of PMT plastic additives registered for use in Canada that are expected to have little to no retention in WWTPs. However, it is currently unknown to what extent these compounds are used and emitted. To ground-truth the *in silico* analysis presented here, the Canadian PMT plastic additive suspect list should be used as a target list for the screening of surface water, stormwater, and influent as well as effluent samples across different watersheds, including the Great Lakes. Such investigation from different geographic locations (from within and outside of Canada) would allow for further refinement of prioritization criteria for PMT plastic additives to be then included in regular monitoring efforts. In the future, the Canadian PMT plastic additive suspect list could be extended to include PMT plastic additives used outside of Canada, further PMT substances (that are not plastic additives) as well as PMT transformation products or more broadly PMOC substances.

5.3 Tools and data to conduct fate and transport and/or risk assessments of priority PMT substances

The lack of experimental physical–chemical property data as well as information on tonnages are substantial barriers to an effective fate and risk analysis of PMT plastic additives and PMT substances in general. More experimental data on physical–chemical properties should be collected and made available for further scientific and regulatory assessment of these compounds. Annual tonnage and specific use characteristics of a substance are especially important parameters that, if made available, would aid in further prioritization of PMT plastic additives, especially when implemented in a comprehensive ranking such as the E-score presented Schulze *et al.*[25] In addition, *in silico* tools need to be developed or updated to include PMT substances in their applicability domain.

There is a clear need for greater emphasis on the development of experiments, models, and regulations for PMT plastic additives given their abundance and lack of current regulatory mechanisms. This is true in Canada as well as globally as persistent and mobile chemicals pose transboundary environmental and health risks that can only be addressed through integrated regulatory approaches and information exchange.[41,42]

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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