



A recipe for plastic: Expert insights on plastic additives in the marine environment

Thomas Maes^{a,*}, Fiona Preston-Whyte^a, Stephanie Lavelle^a, Alessio Gomiero^b,
Andy M. Booth^c, Maria Jesus Belzunce-Segarra^d, Juan Bellas^e, Steven Brooks^f, Adil Bakir^g,
Lisa I. Devriese^h, Christopher Kim Phamⁱ, Bavo De Witte^j

^a GRID-Arendal, Teaterplassen 3, 4836 Arendal, Norway

^b NORCE Climate and Environment dep, Mekjarvik 12, 4072 Randaberg, Norway

^c SINTEF Ocean, Brattørkaia 17C, 7010 Trondheim, Norway

^d AZTI, Marine Research Division, Herrera kaia z/g, Portualdea, 20110 Pasai, Spain

^e Centro Oceanográfico de Vigo, Instituto Español de Oceanografía (IEO), CSIC, Subida a Radio Faro 50, Vigo 36390, Spain

^f Norwegian Institute for Water Research (NIVA), Økernveien 94, 0579 Oslo, Norway

^g Centre for Environment, Fisheries and Aquaculture Science (Cefas), Pakefield Road, Lowestoft, Suffolk NR33 0HT, UK

^h Flanders Marine Institute (VLIZ), InnovOcean Campus, Jacobsenstraat 1, 8400 Ostend, Belgium

ⁱ Instituto de Investigação em Ciências do Mar – OKEANOS, Universidade dos Açores, Horta, Portugal

^j Flanders Research Institute for Agriculture, Fisheries and Food (ILVO), Marine Research (ILVO-Marine), Jacobsenstraat 1, 8400 Ostend, Belgium

ARTICLE INFO

Keywords:

Plastic pollution
Chemical additives
Monitoring
Pathways
Ecotoxicity
Global Plastic Treaty

ABSTRACT

The production and consumption of plastic products had been steadily increasing over the years, leading to more plastic waste entering the environment. Plastic pollution is ubiquitous and comes in many types and forms. To enhance or modify their properties, chemical additives are added to plastic items during manufacturing. The presence and leakage of these additives, from managed and mismanaged plastic waste, into the environment are of growing concern. In this study, we gauged, via an online questionnaire, expert knowledge on the use, characteristics, monitoring and risks of plastic additives to the marine environment. We analysed the survey results against actual data to identify and prioritise risks and gaps. Participants also highlighted key factors for future consideration, including gaining a deeper understanding of the use and types of plastic additives, how they leach throughout the entire lifecycle, their toxicity, and the safety of alternative options. More extensive chemical regulation and an evaluation of the essentiality of their use should also be considered.

1. Introduction

1.1. Plastic pollution in the marine environment

Since the 1950s, a significant quantity of polymers has been produced, globally estimated at 9200 million metric tons in 2017 (UNEP, 2021). Inadequate waste management, and future projections show that even in the best-case scenario, large amounts of plastic will continue to leak into the environment (Borrelle et al., 2020; Lau et al., 2020; Nyberg et al., 2023). Plastic pollution poses a hazard to many organisms, including humans. The extent and mechanisms by which plastic pollution affects organisms are still poorly understood. The impacts are dependent on the organism and the size and shape of the debris, as well

as the composition of the plastic polymer and its additives (Werner et al., 2016). The leaching of additives is a process inherent to plastic pollution, by which plastics may exert additional harmful effects (Bridson et al., 2021).

1.2. Additives in plastic

Over 13,000 chemicals are associated with plastic production across a wide range of applications, of which over 3200 monomers, additives, processing aids and non-intentionally added substances are of potential concern due to their hazardous properties (United Nations Environment Programme, and Secretariat of the Basel, Rotterdam and Stockholm Conventions, 2023). On average, non-fibre plastics have been found to

* Corresponding author.

E-mail address: thomas.maes@grida.no (T. Maes).

<https://doi.org/10.1016/j.marpolbul.2023.115633>

Received 30 June 2023; Received in revised form 1 October 2023; Accepted 4 October 2023

Available online 19 October 2023

0025-326X/© 2023 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

contain 93 % polymer resin and 6–7 % additives by mass (Geyer et al., 2017), but the amount varies depending on the function of the materials; for example, additives can constitute up to 80 % of the total volume in polyvinyl chloride (PVC) (Hahladakis et al., 2018). The most widely used plastic additives being plasticisers, flame retardants, and fillers, which together make up 75 % of global annual additive production (Geyer, 2020; Geyer et al., 2017). Though data for plastic additives production and usage is limited, based on a plastic production volume of 438 million metric tonnes, 27 million tonnes of additives are estimated to have been produced in 2017 (Geyer, 2020). A key challenge in identifying and quantifying additives in plastic is the sheer number and diversity of substances utilised, as well as confidentiality and commercial sensitivity of obtaining information.

1.3. Additives in the environment

Many additives, although useful for the manufacturing and functionality of products, have widely documented risks of contaminating food, air, soil and water (Fauser et al., 2022; Rani et al., 2022; Wrona and Nerin, 2019). Generally, additives do not chemically bind to polymer chains, except reactive organic additives, e.g., some flame retardants (van Oers et al., 2012). Therefore, weakly bound additives can leach out throughout the entire life cycle of plastic items; through production, use, landfill, incineration, recycling and mismanaged waste streams (Wiesinger et al., 2021). Plastic fragmentation allows for migration far from the point of origin (Geyer et al., 2017), with significant proportions of plastic particles and associated additives found in soils and wastewater effluents (Lithner et al., 2011; Tun et al., 2022; Wagner and Schlummer, 2020). These hazardous substances may persist in the environment and/or bioaccumulate in organisms, ultimately adding to the chemical burden on our ocean and earth systems (Hansen et al., 2013). Once in the environment, additives and/or contaminants can react, degrade and become even more toxic when exposed to particular species (Hahladakis et al., 2018). Many have been classified as hazardous and are therefore of concern for human and environmental health (Groh et al., 2019; Rochman, 2015; Wiesinger et al., 2021). Though measuring additive composition in litter is developing (Corami et al., 2021, 2022; Rosso et al., 2022), the composition of additives in plastic products and debris is largely unknown.

1.4. Aim of this study

Given the gaps in knowledge acknowledged within literature regarding additives, the authors set out to measure the knowledge of additives from experts. In this study, we gauged expert knowledge through an online survey and evaluated the outcome against objective data. This study focuses on understanding knowledge in relation to (i) the use of additives globally, (ii) the characteristics of plastic additives (this refers to entry sources and pathways), and (iii) the risks associated with plastic additives in the environment.

2. Methods

This study employed an online survey to gauge how experts involved in plastic production or plastic litter research perceive their knowledge of plastic additive chemicals, with the aim of evaluating potential disparities between perception and actual knowledge.

2.1. Selection of compound groups/chemicals

Compound groups were selected following a literature review using “additives”, “chemicals” and “plastic” as keywords. Three publications that contained comprehensive lists of hazardous or high-risk additives and monomers (Groh et al., 2019; Lithner et al., 2011; Rodrigues et al., 2019) were used to define compound groups and select examples. The lists in each publication differed significantly, due to the difference in

research focus (packaging/daily life products; inclusion of monomers). The compound groups that appeared in at least one of the three publications were compiled, and cross-referenced against reviews by Andrade et al. (2021), Hermabessiere et al. (2017) and Hahladakis et al. (2018). If the compound groups/chemicals were classified as high risk in at least one publication (Groh et al., 2019; Lithner et al., 2011; Rodrigues et al., 2019) and mentioned in at least two publications out of these 6 publications, they were selected. Monomers were excluded from the final list to focus on additives. Solvents and initiators were also excluded as they are not intended to be inherently present in the final products. With the aim of including emerging compounds and based on the authors’ expert judgement, organotins, phenylenediamines and polybrominated diphenyl ethers were added to the list. The final list of additives (9) selected for inclusion in this research is as follows: phthalates, aromatic amines, organophosphates, metal acetates, PFASs/PFOS (perfluoroalkyl and polyfluoroalkyl substances; perfluorooctane sulfonate (PFOS) is a common example from the PFAS family of chemicals) metals, organotins, polybrominated diphenyl ethers and phenylenediamines. The complete list of additives extracted from the publications is provided in Supplementary Information (SI, Table S1).

An illustrative chemical from each group was provided to assist the respondents. Offering a representative chemical for each group made it easier for survey participants to frame the question but had the potential drawback of oversimplifying the actual situation. The examples given were as follows:

- Phthalates - e.g., benzyl butyl phthalate
- Aromatic amines - e.g., 4-4-methylenedianiline
- Organophosphates - e.g., tris(2-chloroethyl)phosphate
- Metal acetates - e.g., cobalt(II)diacetate
- PFAS/PFOS - e.g., perfluorooctanoic acid
- Metals - e.g., lead
- Organotins - e.g., fentin acetate
- Polybrominated diphenyl ethers - e.g., BDE-209
- Phenylenediamines - e.g., 6-PPD

2.2. Survey approach

A staggered survey approach was devised to measure the knowledge among experts who share a common interest in additives, chemical pollution, or the marine environment. The survey was initially released (14 February 2022–3 March 2022) to experts within the working groups of the International Council for the Exploration of the Sea (ICES) related to marine litter (WGML), marine chemistry (MCWG) and biological effects of contaminants (WGBEC). In the second phase, a broader range of relevant experts were invited to answer the survey (4 March 2022–6 May 2022). A complete list of organisations to which the survey was distributed can be found in SI (Table S2). The results from all surveys were combined for analysis.

A survey (provided in the SI, Table S3) was compiled to collect demographic information of each respondent, as well as their knowledge on the following topics for each compound group/chemical:

- if they were aware of the production volumes?
- Whether other sources and routes exist, aside from plastics, through which these compound groups enter the marine environment?
- whether there are standardised and established techniques for analysing this compound group, and if there are, to specify the method?
- if these compound groups are persistent, bioaccumulative and/or toxic?
- if these compound groups cause a risk to the lower marine trophic levels (plankton, algae etc.)?
- if these compound groups cause a risk to the higher marine trophic levels (fish, mammals, birds etc.)?

The respondent’s personal information was collected, processed, and managed within General Data Protection Regulation (GDPR) and other relevant international privacy laws.

2.3. Data analysis

2.3.1. Statistics

Descriptive statistics were applied to assess the expert responses.

- To begin, demographic information of the respondents was summarised using Tableau Public, an accessible online tool for data visualisation. This step allowed us to gain insights into the attributes of the survey participants, such as age, gender, and professional background.
- Microsoft Excel was used to organise and present the remaining survey data, ensuring that it was readily accessible for further analysis.
- For questions employing the Likert scale, median responses were considered (data provided in SI, Fig. S4). ‘I don’t know’ responses were treated and reported separately. The remaining responses were transformed into percentages of the total respondent count, excluding instances where ‘I do not know’ was chosen.
- To convey the intricate hierarchies within data concerning open-ended questions, tree-maps were employed. These tree-maps consist of nested rectangles, with each larger rectangle subdivided into smaller ones, proportionally representing distinct responses. This visualisation method helped capture the diversity of responses to open-ended questions.

2.3.2. Available lowest effect concentration (LOEC) data

A dedicated literature review was conducted to validate and substantiate the responses provided by the participants regarding the risk

posed by various additives to aquatic organisms at various positions in the food chain (see S.I. S7). Initially, representative chemicals were selected from each of the plastic additive chemical groups used in this study. Scientific databases such as Web of Science, Scopus, Google Scholar and PubMed were then explored to compile the reported lethal and sublethal toxicity levels of the selected plastic additives. Search terms comprised of specific keyword combinations, including the names of selected plastic additives, were used to identify scientific papers focusing on aquatic toxicity. Relevant articles and reports were selected and reviewed to establish the lethal and sublethal toxic effects of the selected substances, and the available toxicity data (lowest effect concentration, LOEC, median lethal concentration, LC50, median effective concentration, EC50) was collated (SI, Table S7). Finally, aquatic organisms were grouped into three trophic levels (namely microalgae, invertebrates, fish), and the toxicity threshold values for each chemical were estimated by calculating the average of the EC50/LC50/LOEC values obtained in the literature review within each trophic level.

3. Results

The survey was completed by 50 respondents, 17 ICES experts and 33 other specialist stakeholders. Of the 50 individuals (Fig. 1), the majority (82 %) were based in Europe, followed by Sub-Saharan Africa (10 %), Australia and Oceania (4 %), Asia (2 %) and North America (2 %). The majority of the respondents were employed in research and/or academia (76 %), the remaining fractions were employed in government (16 %), NGO/IGO sector (6 %) and civil society (2 %). In terms of gender, 44 % were female, 52 % were male, and 4 % preferred not to say. Most of them were in the age groups of 41–50 (32 %), 31–40 (28 %), and 51–60 (24 %). The vast majority (98 %) of the participants held a postgraduate degree.

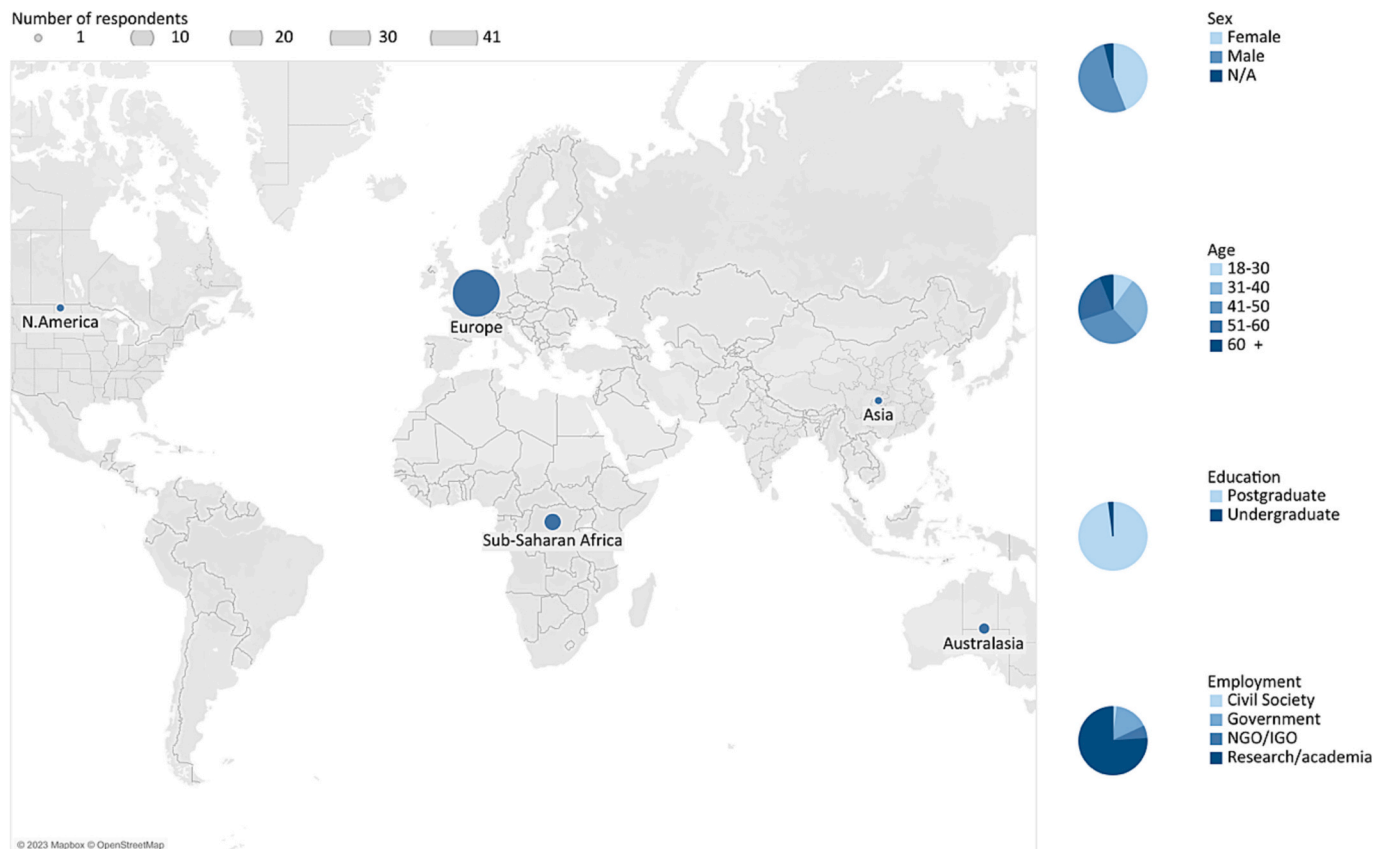


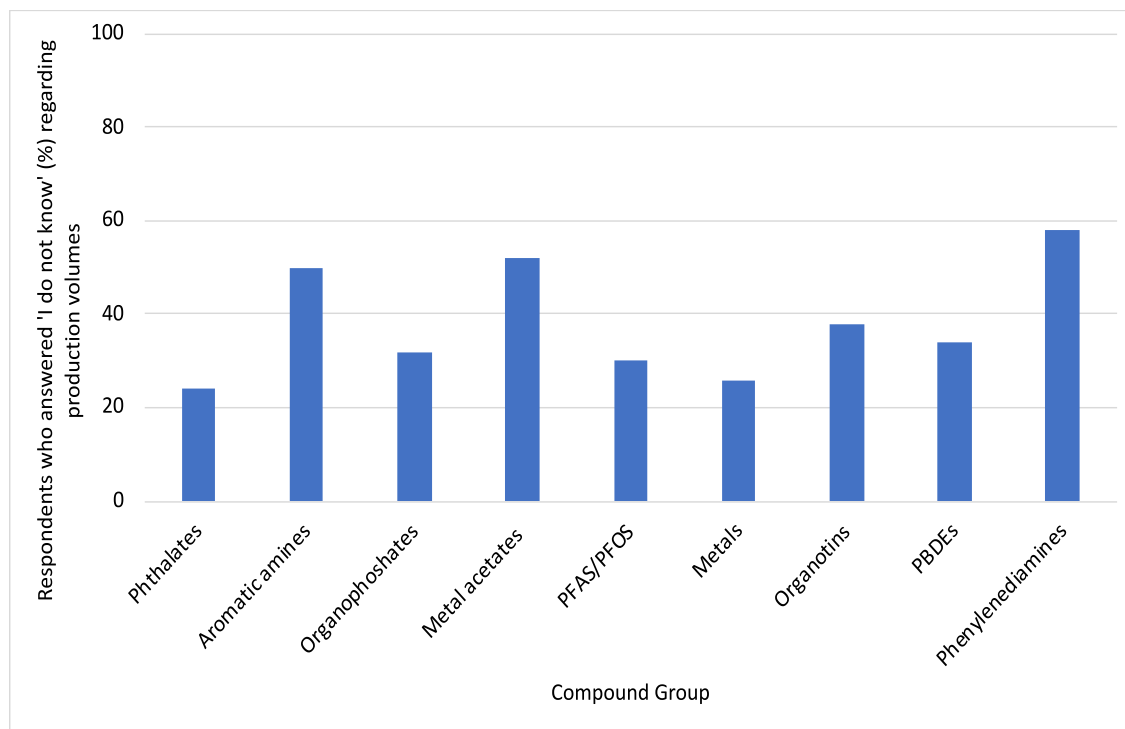
Fig. 1. Map showing the demographics of all respondents.

3.1. Use of additives globally

The participants exhibit a deficiency in their understanding of the use of additives globally (total production volumes) (Fig. 2a), especially

for metal acetates, aromatic amines and phenylenediamines (unknown production volume by at least 50 % of respondents). The highest level of awareness regarding production quantities was observed for phthalates and metals. More than 70 % of respondents indicated being able to

a)



b)

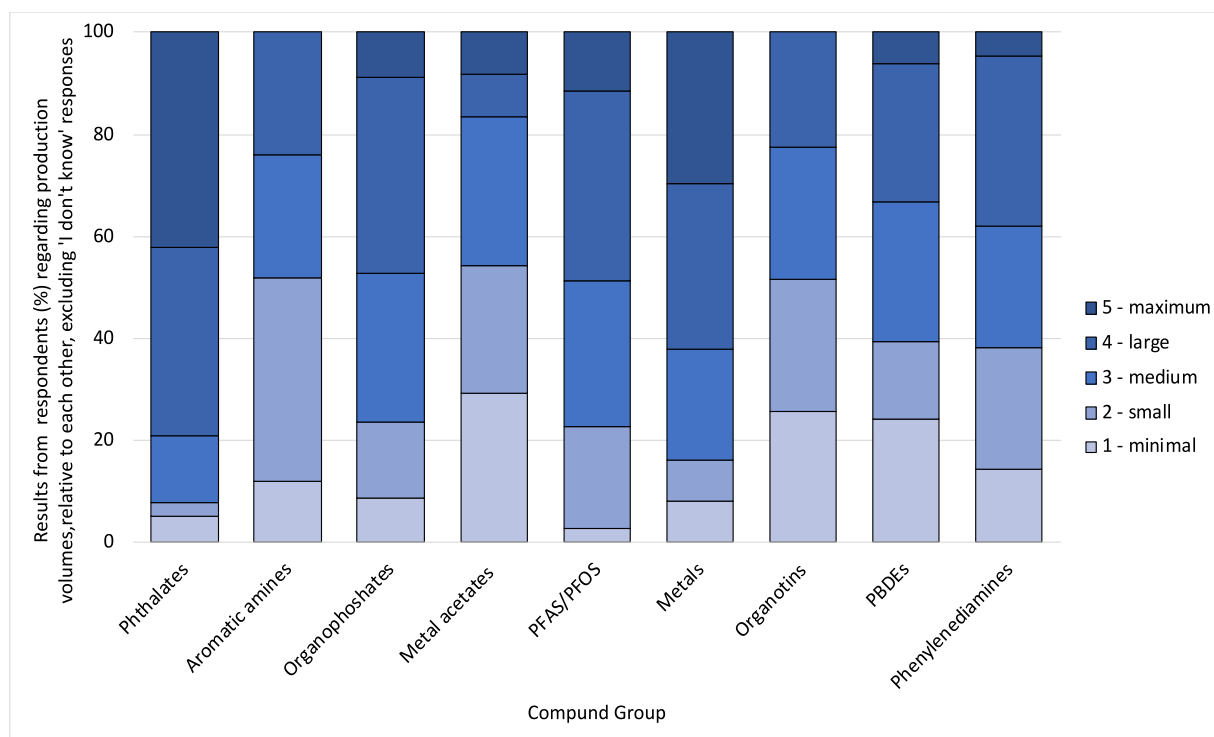


Fig. 2. Respondent's knowledge of the approximate volumes of compound groups produced globally, relative to each other with a) the respondents who answered 'I don't know' (%) and b) results from respondents with 'I don't know' responses removed (%).

estimate the relative production volumes for these compound groups, which also revealed the highest median scores (provided in SI Fig. S4). Phthalates and metals were recognised as being produced in large volumes by 37 and 32 % of respondents.

3.2. Characteristics of additives

For most compound groups, experts acknowledged routes leading into the marine environment that didn't involve plastics, with the exception of aromatic amines and phenylenediamines, whereby at least 50 % of respondents were unsure (Fig. 3a and b).

3.3. Monitoring of additives

For metal acetates and phenylenediamines, a relatively high number of respondents were unaware of the availability of analytical methods (62 % and 60 % of respondents, respectively) (Fig. 4a). For phthalates, organophosphates, PFAS/PFOS, metals, and PBDEs, >50 % of the experts mentioned the existence of established methods for analysing these compounds in the marine environment. Respondents were unsure of the other chemical groups or mainly indicate that methods are available but not yet standardised (Fig. 4b). Fig. 5 summarises the methods named by respondents.

3.4. The risks posed by additives

3.4.1. Persistent, bioaccumulative and/or toxic nature

The respondents' knowledge of the persistent, bioaccumulative and/or toxic (PBT) nature of the compound groups is presented in Fig. 6. Overall, the PBT nature is most known for metals, PFAS/PFOS and phthalates, while a larger group of respondents expressed being unsure about phenylenediamines, aromatic amines and methyl acetates. Phthalates, PFAS/PFOS, metals, organotins and PBDEs reach the highest PBT scores, with most respondents indicating that these compound groups possess each characteristic. A slight difference in response can be noted, with PFAS/PFOS and phthalates scoring highest on persistence (38 % and 34 % of respondents, respectively), metals, PBDE's and organotins scoring highest on bioaccumulation (33 %, 33 % and 33 % of respondents) and phenylenediamines, aromatic amines, metal acetates scoring high on toxicity (47 %, 47 % and 45 % of respondents).

3.4.2. Risk to lower marine trophic levels

The participants' understanding of the risk of compound groups to the lower levels of marine ecosystems appears to be largely lacking or unknown for all the compound groups (Fig. 7a), especially for phenylenediamines, metal acetates and aromatic amines. More respondents are aware of the risks of metals (Fig. 7a). Knowledge of the risk is dominated by average or extreme risk, whereas "no risk" was seldom selected (Fig. 7b). A conflicting response was given for phthalates with 16 % of respondents indicating no risk while 45 % indicating very high risk. Organotins, organophosphates and PBDEs were highlighted as an extreme risk to lower marine trophic levels by over 50 % of respondents. Respondents also recognise the risk of aromatic amines and metals, where "no risk" was not selected.

3.4.3. Risk to higher marine trophic levels

The survey participants exhibit a stronger awareness of the risks to higher marine trophic levels (as shown in Fig. 8a) compared to their understanding of risks at lower trophic levels (illustrated in Fig. 7a) across all the compound groups.

3.5. The way forward and knowledge gaps

The participants prioritise inquiring about the safety of alternative options when engaging with stakeholders (Fig. 9). At the same time, more than half of the respondents indicate that questions on further

regulation and the necessity of their use are also important. When considering other important questions (Fig. 10), questions around alternatives featured prominently (4 additional questions were listed). Other concerns include information around environmental/economic factors, gaps in knowledge on research and gaps in knowledge on recycling.

The prevailing response among the participants regarding which knowledge gaps should be tackled was the need for more research on alternative options (Fig. 11). Subsequently, in descending priority, there is a need for additional research on the impact, expanded risk assessments, further investigation into additives that leach under varying environmental conditions, and more comprehensive life cycle analyses. The respondents also submitted additional questions, which could be divided into the following topics (Fig. 12): research questions, alternatives and recycling, and environmental/economic.

4. Discussion

The leakage of additive chemicals into the environment from plastics is of growing concern. Even though a wide range of additives is used in plastic products, there is a significant lack of information regarding their use, characteristics, monitoring, and their potential risks to the marine environment.

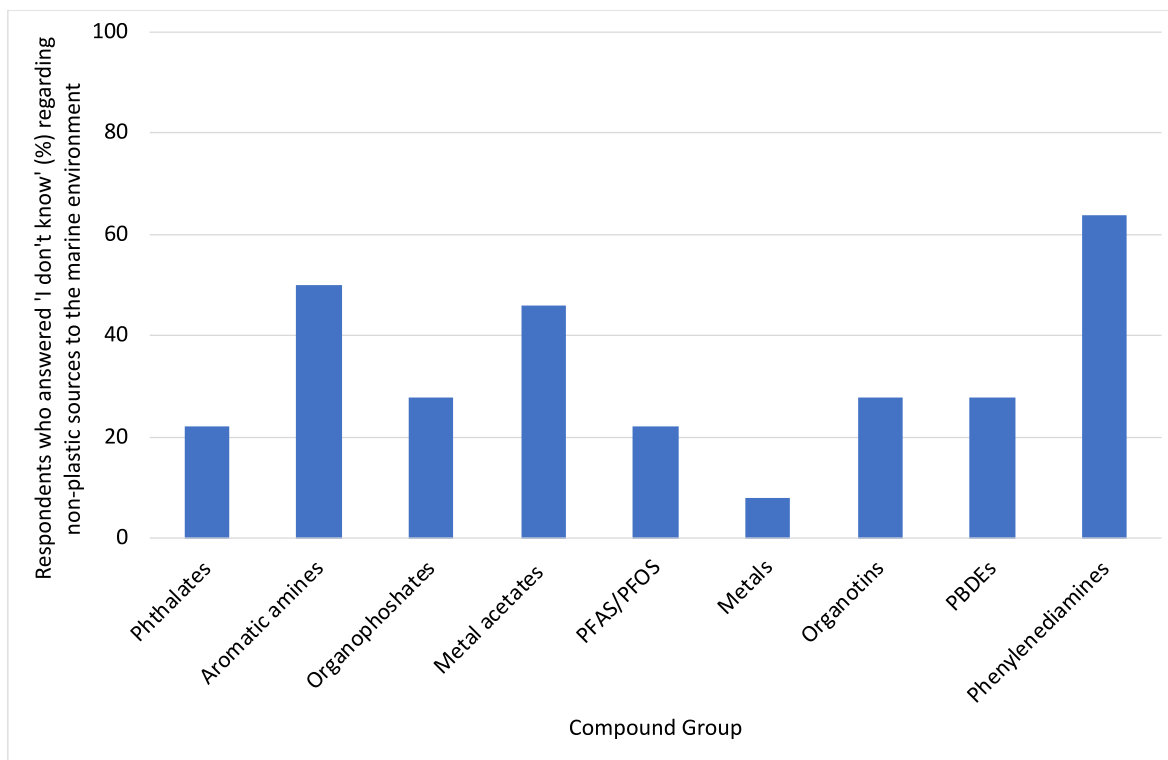
4.1. General considerations regarding gaps in knowledge

The respondent's knowledge on different chemical groups is highly variable, which partly reflects the historical research focus on specific groups. This duality is reflected in the survey results. For example, the risk of phthalates to lower marine trophic levels was estimated as an extreme risk (45 %) to no risk at all (16 %), with 38 % responding that they did not know (Fig. 7). The highest degree of knowledge was found for compounds that are known to have a high environmental persistence and/or bioaccumulation potential, such as metals, PFAS/PFOS and PBDEs. These compound groups are already classified as priority substances within European marine legislation, such as the Marine Strategy Framework Directive (MSFD) and/or in monitoring programmes of regional sea conventions in European waters (Tornero and Hanke, 2018). Ongoing marine environmental monitoring for many of these priority compounds has resulted in large datasets on their occurrence and toxicity in different media, fostering a good knowledge of the respondents. In contrast, phenylenediamines such as 6-PPD have only recently caught attention of the research community and regulators (Tian et al., 2021; Wagner et al., 2022), resulting in less knowledge of this compound group by respondents. A large group of respondent's mention being unaware of the production volumes (58 %), PBT characteristics (58 %) or risks to lower (70 %) and higher (56 %) aquatic organisms of phenylenediamines (Figs. 2a, 7a, 8a). Nevertheless, the 6-PPD oxidation product, 6-PPD-quinone, has recently been demonstrated to be ecotoxicological relevant and could be categorised as highly toxic for aquatic organisms (Tian et al., 2021, 2022) (see also Fig. S7).

4.2. What is known about the use of plastic additives?

The release of plastic additive chemicals and their impacts on the environment depends on the volumes produced, their use and their respective pathways into the marine environment. Estimating the volume of chemicals produced and used globally for plastic production is challenging for several reasons. First, the production of plastic involves a complex supply chain that spans multiple countries and companies, making it difficult to obtain accurate data on the volume of plastic additive chemicals used. Second, the vast array of plastic types requires different chemical inputs (types and amounts), depending on the desired properties of the final product. Finally, some of the chemicals used in plastic production are not specifically produced for that purpose and have multiple uses in various industries, making it difficult to

a)



b)

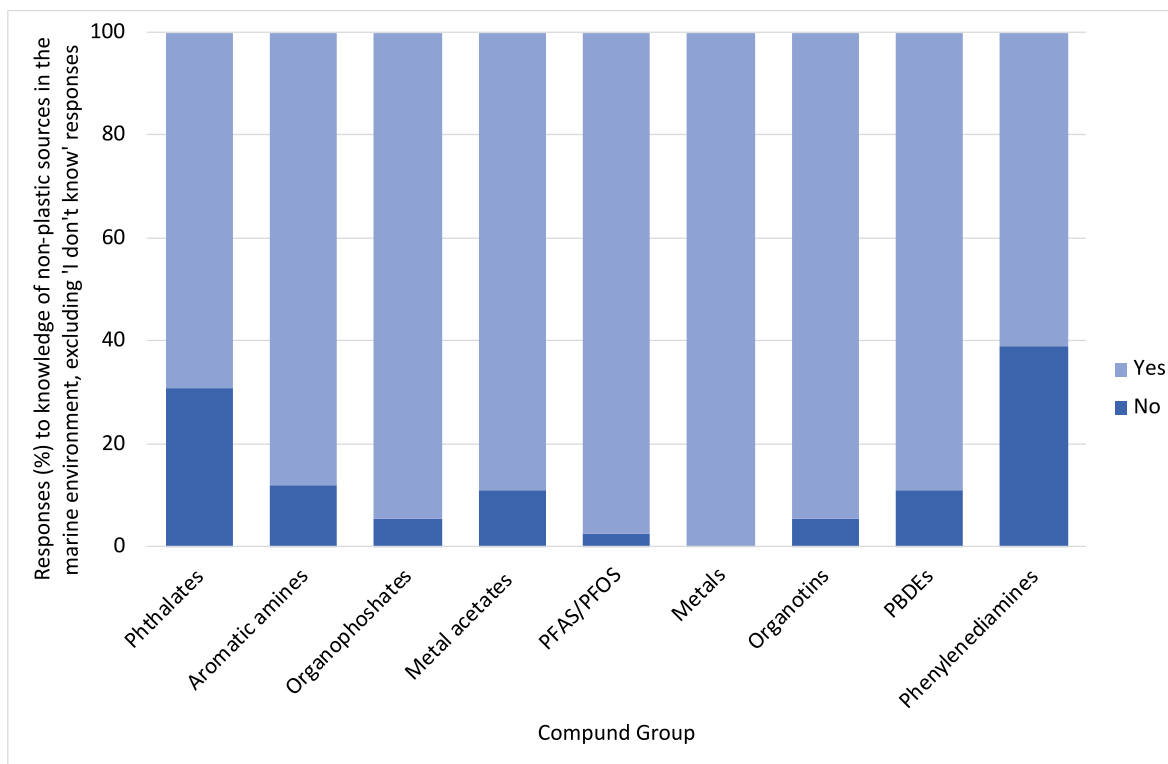
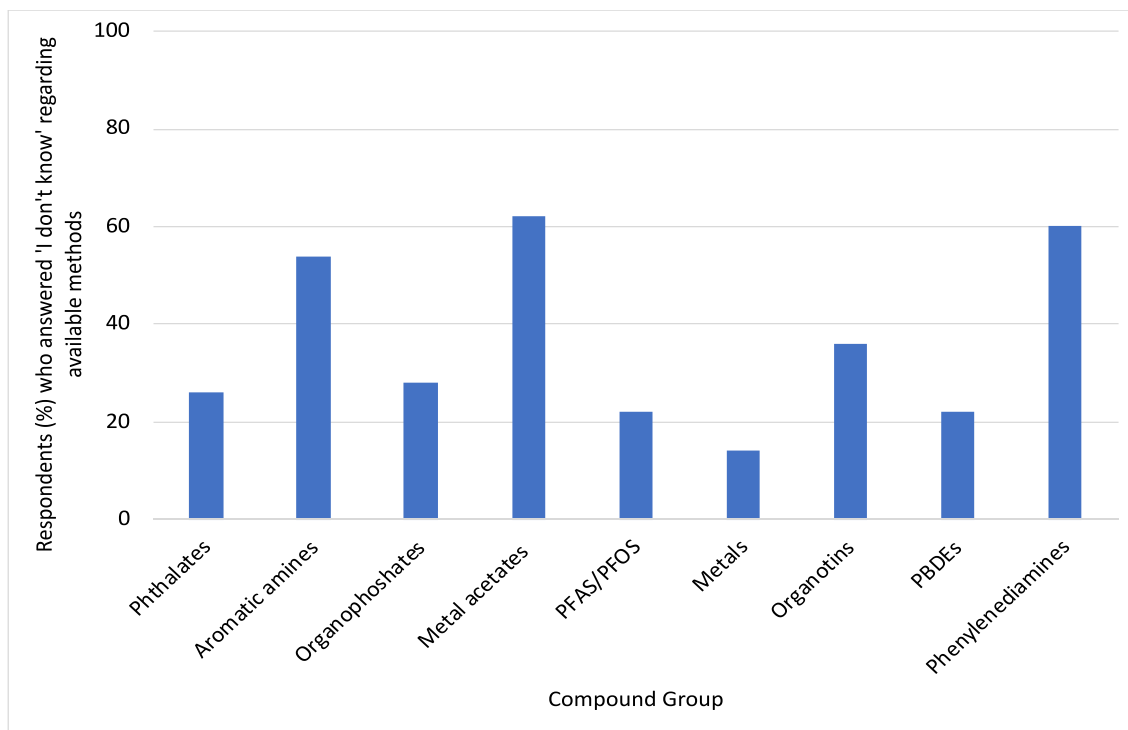


Fig. 3. Respondent's knowledge of existing sources and pathways, other than plastics, of these compound groups in the marine environment with a) the respondents who answered 'I don't know' (%) and b) results from respondents with 'I don't know' responses removed (%).

a)



b)

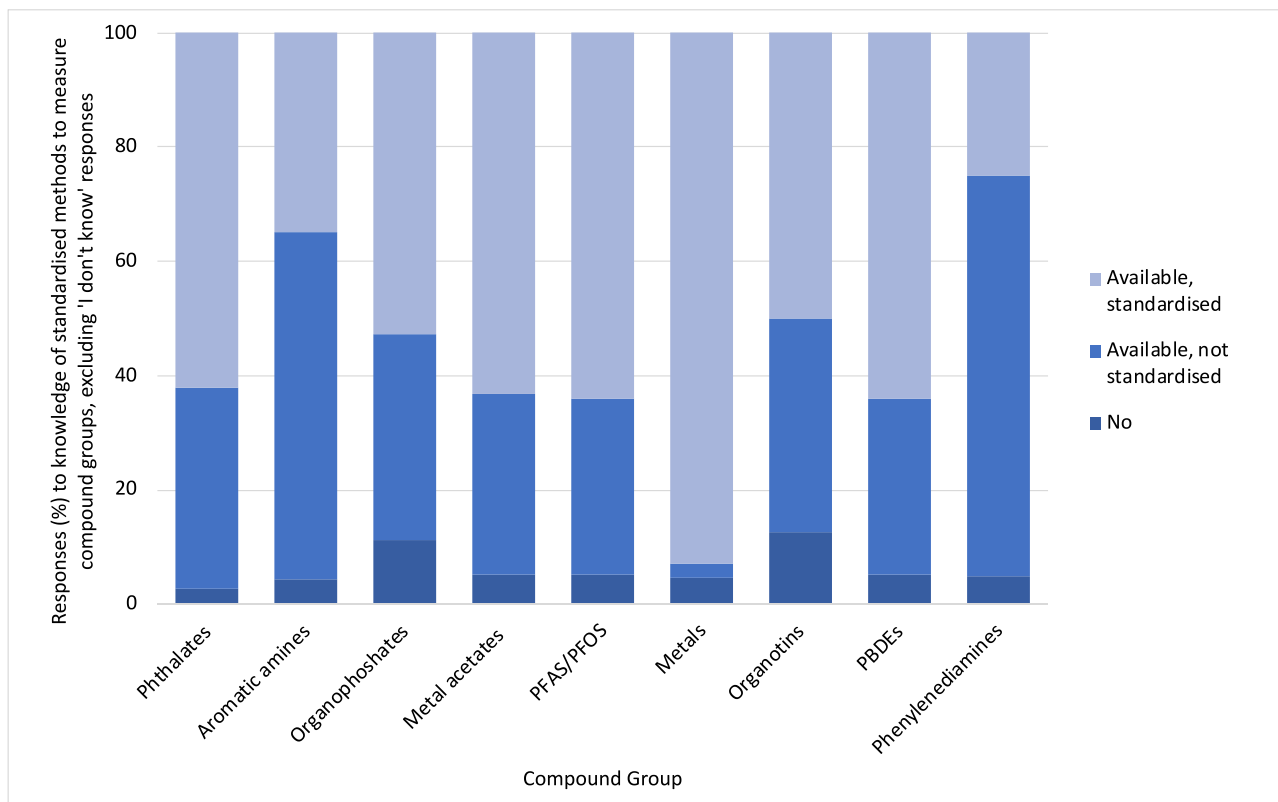


Fig. 4. The respondent's knowledge (%) of standardisation and developed methods to analyse these compound groups with a) the respondents who answered 'I don't know' (%) and b) results from respondents with 'I don't know' responses removed (%).

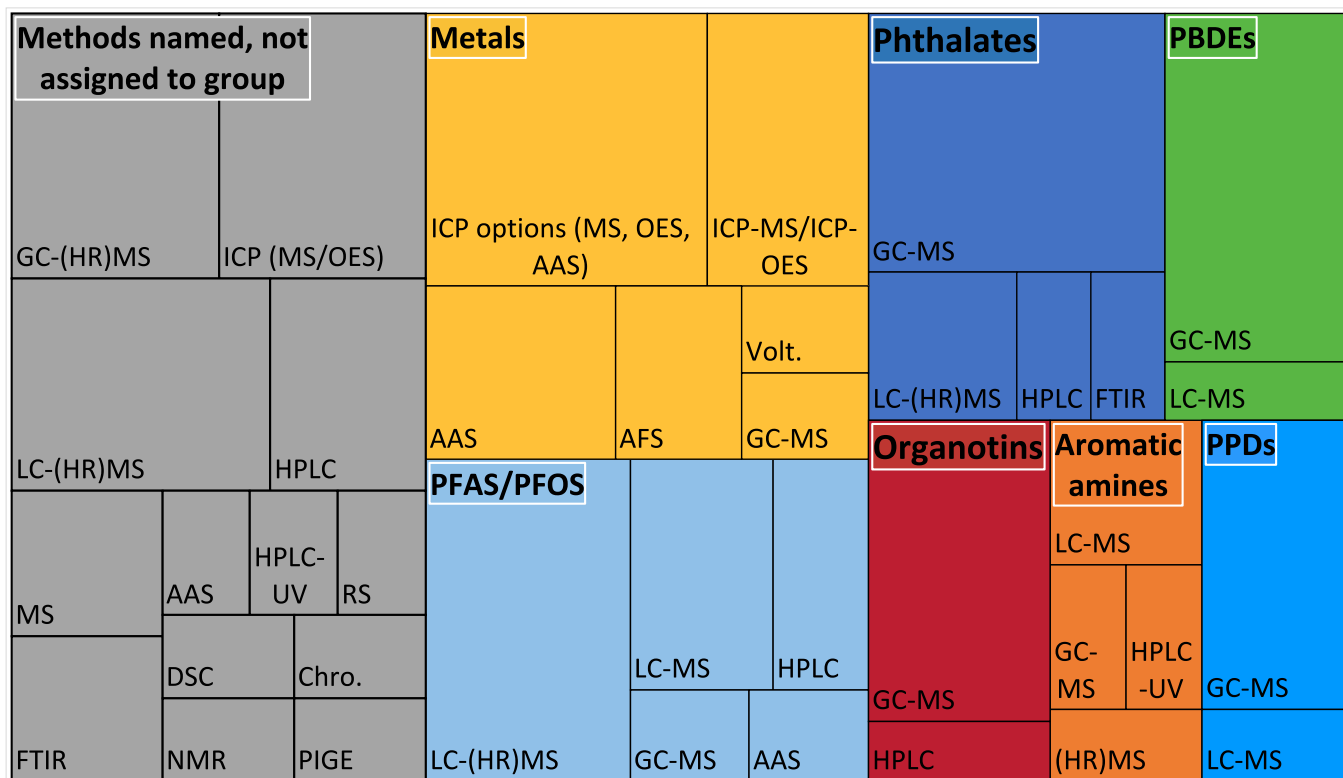


Fig. 5. Tree-map visualisation of methods that respondents are aware of to analyse the compound groups, in which the hierarchical data is displayed in nested triangles, with each larger rectangle subdivided into smaller ones proportionally representing different methods. Interpretation involves understanding the size and colour of the rectangles to grasp the most commonly suggested methods for each compound group. Data and abbreviations also presented in SI Table S5.

distinguish between the amount of chemicals produced for plastic production and those produced for other purposes. The lack of transparency and comprehensive management practices by chemical producers and plastic manufacturers, combined with fragmented policies and incomplete publicly available information, are significant barriers to increasing our knowledge of the volumes and types of additive chemicals used in plastic production.

4.2.1. Production and usage of plastic additives

Packaging, automotive, consumer goods, construction and electronics are some of the most important markets for plastic additives (Hahladakis et al., 2018). Regulations and reporting vary by country or region, but many plastic applications necessitate a safety or technical data sheet that mandates the disclosure of additives exceeding a certain percentage. This requirement applies to items such as building materials and household applications, but not to basic consumer products e.g., toys. A random plastic item can easily contain around 20 additives, with fillers and plasticisers used in the highest volumes (van Oers et al., 2012). Fillers (e.g., calcium carbonate, metal powder, clay) often constitute up to 50 % of the mass of plastics and represent around 28–50 % of the world's additive market (Geyer et al., 2017; Hansen et al., 2013; UNEP, 2021). Primarily, they're added to polymers to cut costs, enhance aesthetics, shape, durability (UV protection), and simplify polymer manufacturing (Groh et al., 2019; Wagner and Schlummer, 2020; Wiesinger et al., 2021). Reinforcement additives, such as glass fillers, can account for an additional 15–30 % of the volume of plastic materials (Hansen et al., 2013). Plasticisers are used to increase flexibility and can constitute 10–80 % of the mass of plastic materials, representing around 22–34 % of the world's additive market (>80 % of which are phthalates) (Geyer et al., 2017; UNEP, 2021; van Oers et al., 2012). Flame retardants (12–18 % of mass) are also used in relatively high volumes and, together with fillers and plasticisers, account for around 75 % of all additives (Geyer et al., 2017; Hansen et al., 2013). Colourants (0.25–5 % of mass)

and other functional additives (e.g., antioxidants, heat stabilisers, UV stabilisers and biocides) are used in lower volumes (0.001–3 % of mass). Non-intentionally added substances (NIAS) are chemicals added without a technical reason during the production process. They are often present in food packaging from which they can easily migrate into food. NIAS alone may account for more than half of the number of chemicals in plastics (Geueke et al., 2018).

4.2.2. Volumes of additives entering the marine environment

The current study highlighted an existing lack of knowledge among experts regarding the production volumes and usage of plastic additives, particularly metal acetates, aromatic amines, and phenylenediamines. This could indicate a lack of transparency on the use of specific groups of additives. Chemical inventories and databases available for 19 countries have revealed the registration of over 350,000 chemicals and chemical compounds (Wang et al., 2020). The European Chemicals Agency (ECHA), which is the administrative and technical support for the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) regulations, maintains one of the most comprehensive inventories. As of March 2023, ECHA's database contains over 26,000 REACH registrations. Around 80 % of these substances, however, are yet to be assessed, and data on production volumes and applications are still lacking (Persson et al., 2022). In this study, applicants' responses on plastic additive production volumes were cross-checked against the ECHA database (SI, Table S6). Commonly used metal and phthalate compounds represent some of the highest production volumes, agreeing with expert opinions gathered in this survey. However, it is difficult to estimate what proportions (particularly of metals) are used in plastic manufacturing. Phthalates are the most widely used plasticiser, with PVC accounting for ~80 % of total phthalate consumption (Holland, 2018). The global phthalate plasticiser market is estimated to be approximately 6–8 million tonnes per year, with China, the USA and Europe accounting for about 70 % of the market (Holland, 2018; United

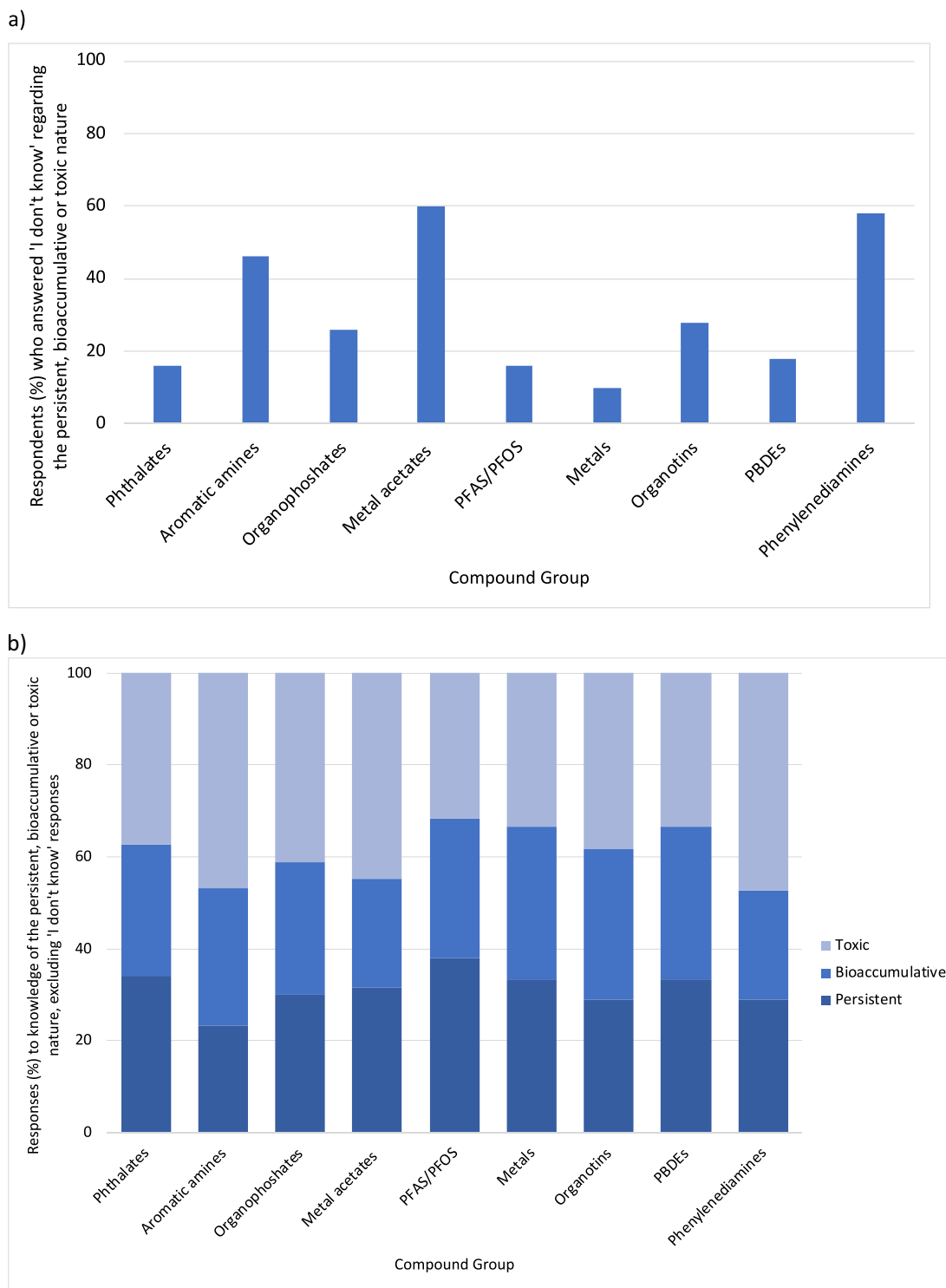


Fig. 6. Respondent's knowledge of the compound group's persistent, bioaccumulative and/or toxic nature with a) the respondents who answered 'I don't know' (%) and b) results from respondents with 'I don't know' responses removed (%).

Nations Environment Programme, and Secretariat of the Basel, Rotterdam and Stockholm Conventions, 2023).

4.3. Characteristics of additives

Plastic additives enter the environment via various sources and routes. To assess the risk of released additives in the environment, it's crucial to know global production volumes and understand how they

enter the environment. Identifying the routes of their release is essential for effective management, regardless of whether it's due to poor plastic waste handling or other factors. To reliably assess the risk to wildlife, we must determine the chemical identity, quantity, and exposure to these additives (Adlard, 2019). A current understanding of the pathways of each plastic additive chemical group into the marine environment, from both plastic and other sources, is presented below.

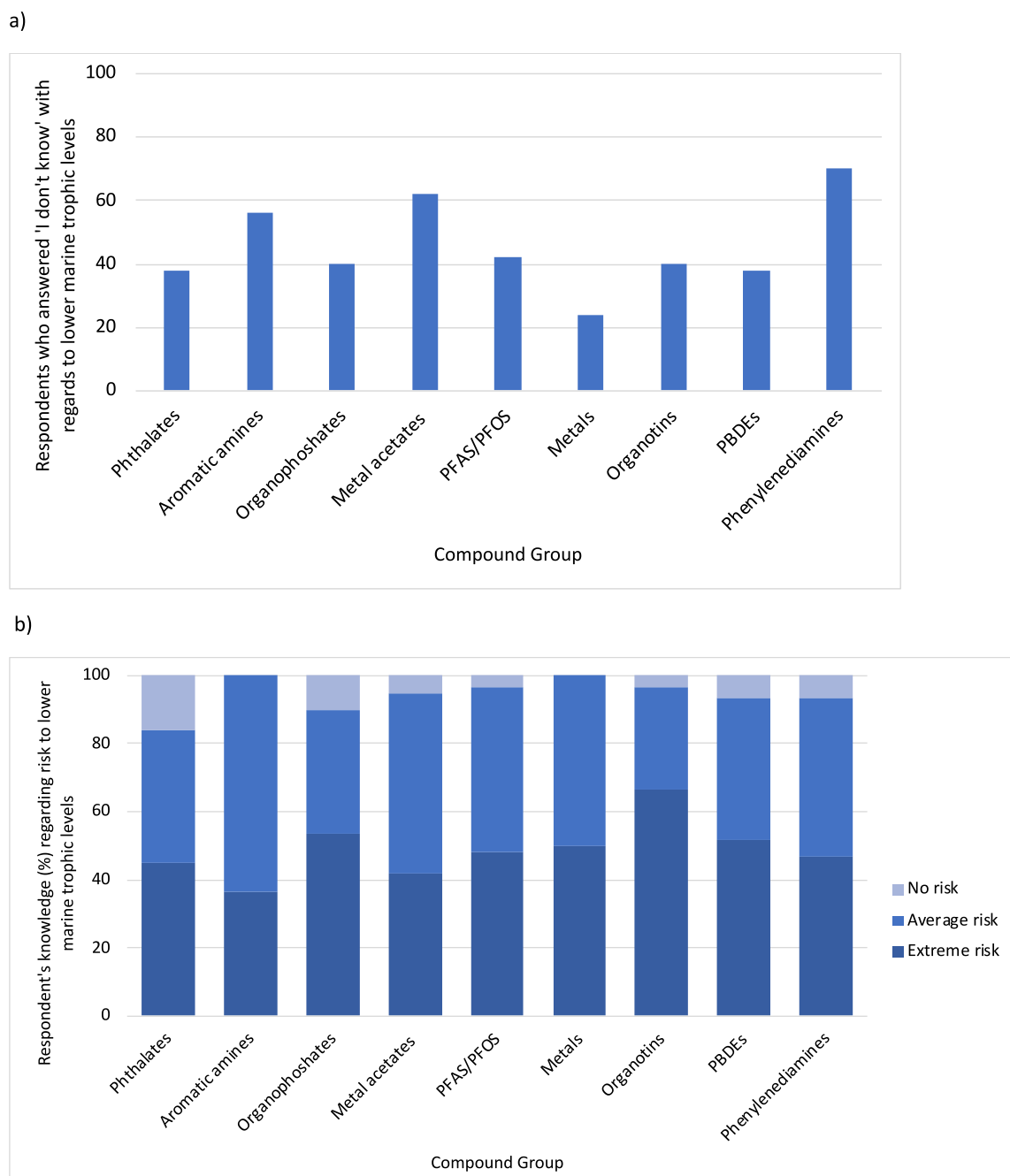


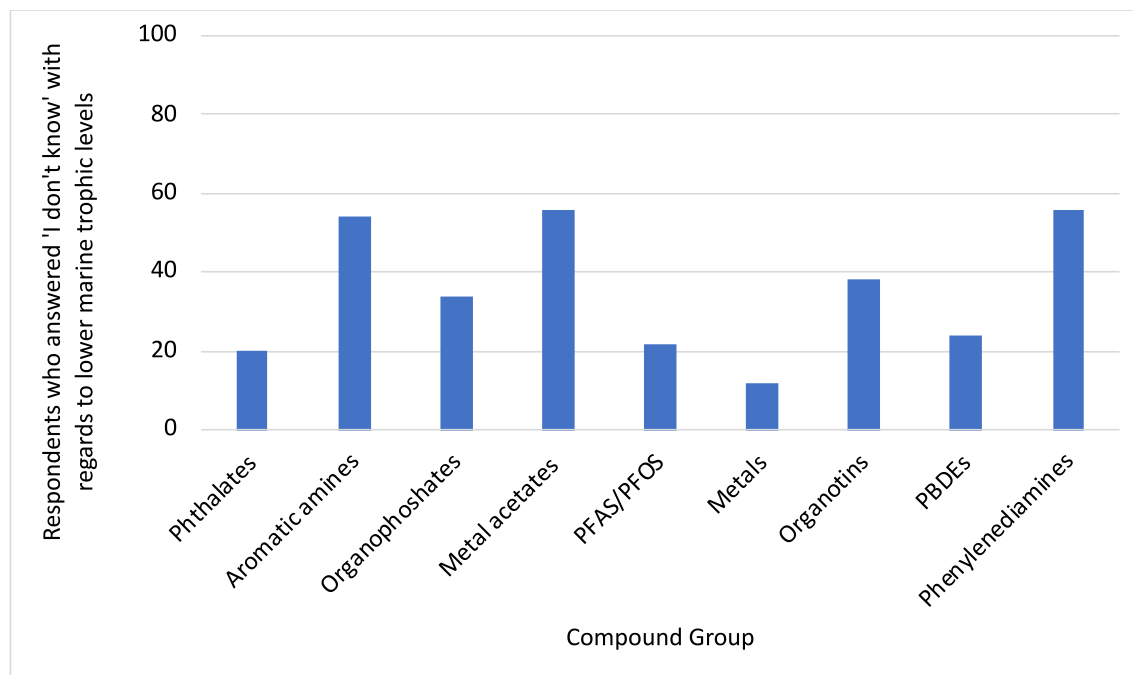
Fig. 7. The respondent's knowledge of the risk of the compound groups/chemicals to the lower marine trophic levels with a) the respondents who answered 'I don't know' (%) and b) results from respondents with 'I don't know' responses removed (%).

4.3.1. Phenylenediamines

Most respondents (64 %) were unsure about the pathways, other than plastics, of phenylenediamines into the marine environment (Fig. 3a), most likely reflecting that this group of chemicals is not very high profile from an environmental risk perspective. Large knowledge gaps still remain on their environmental occurrence and fate (Huang et al., 2021). The chemical *p*-phenylenediamine is primarily used as a dye intermediate and as a dye (National Center for Biotechnology Information, 2022a). Both *N*-(1,3-dimethylbutyl)-*N'*-phenyl-*p*-phenylenediamine (6-PPD) and *N*-(1,3-dimethylbutyl)-*N'*-phenyl-*p*-phenylenediamine-quinone (6-PPDQ) are chemicals associated with vehicle tyres, and therefore with tyre wear particles (TWP) and end of life tyres repurposed as crumb rubber that is commonly used in road asphalt and infill material for artificial turf sports fields. A number of

other phenylenediamines and phenylenediamine mixtures are used as antioxidant additive chemicals in vehicle tyres, as well as other rubber and latex products including shoe soles, conveyor belts, and floor coverings. TWPs and tyre crumb rubber have been identified as important land-based sources of microplastics in the environment (Kole et al., 2017; Magnusson et al., 2016; Parker-Jurd et al., 2020). A complex mixture of chemicals has been demonstrated to leach from TWPs and crumb rubber once they are released to the marine environment (Capolupo et al., 2020, 2021; Halsband et al., 2020). Zeng et al. (2023) reported the widespread occurrence and distribution of PPDs and their derived PPD-Qs in sediments on a large geographical scale, among which 6PPD and 6PPD-Q were the most prevalent in the aquatic environment. Total sedimentary concentrations of PPDs and PPD-Qs presented a clear spatial trend with decreasing levels from urban rivers

a)



b)

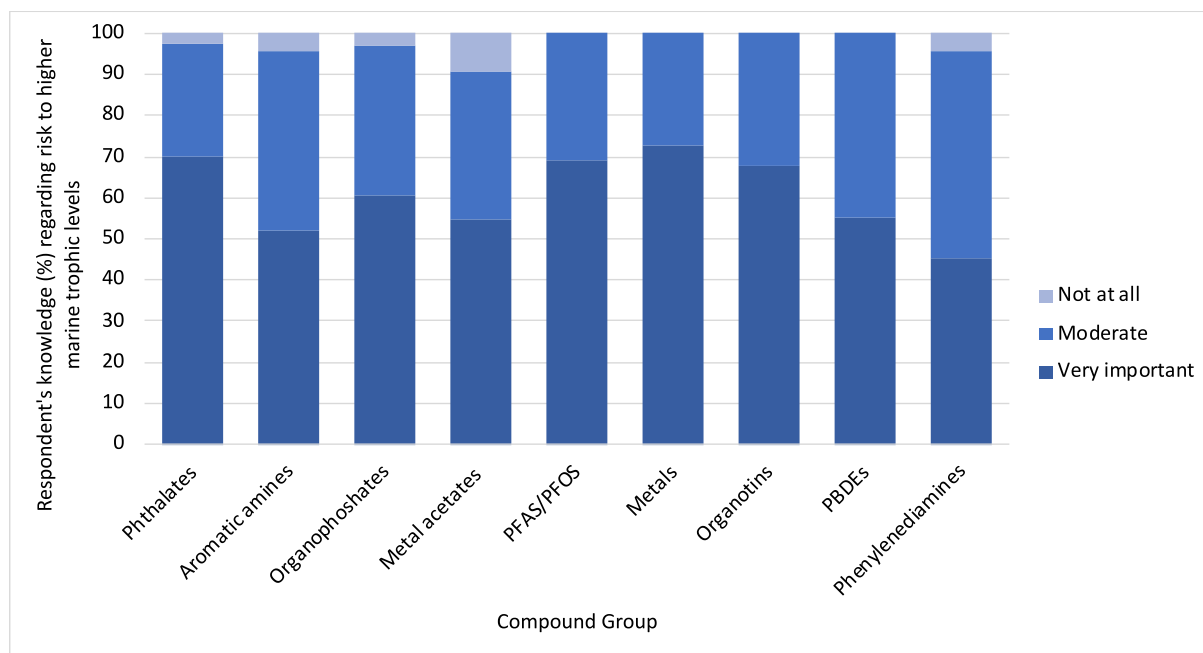


Fig. 8. The respondent's knowledge of the risk of the compound groups/chemicals to the higher marine trophic levels with a) the respondents who answered 'I don't know' (%) and b) results from respondents with 'I don't know' responses removed (%).

(medians: 39.7 and 15.2 ng/g) to estuaries (14.0 and 5.85 ng/g) and then toward coasts (9.47 and 2.97 ng/g) and deep-sea regions (5.24 and 3.96 ng/g) (Zeng et al., 2023). Data highlighted the importance of rivers as pathways for the entry of those compounds to the marine environment.

4.3.2. PBDEs

The majority of respondents (72 %) were aware of the pathways, in addition to plastics, of PBDEs into the marine environment (Fig. 3b). No

known natural sources of PBDEs have been identified except for a few marine organisms that produce forms of PBDEs that contain higher oxygen levels (US EPA, 2017), meaning these derive entirely from anthropogenic sources. PBDEs have been used widely since the 1970s as flame retardants in polyurethane foams in upholstery, polymer resins, and plastics used as components in electrical equipment (Environment Agency, 2019). PBDEs are not chemically bound to the plastic or foam in which they are incorporated, resulting in high potential for their leaching into the environment (Kwan and Takada, 2016; Tanaka et al.,

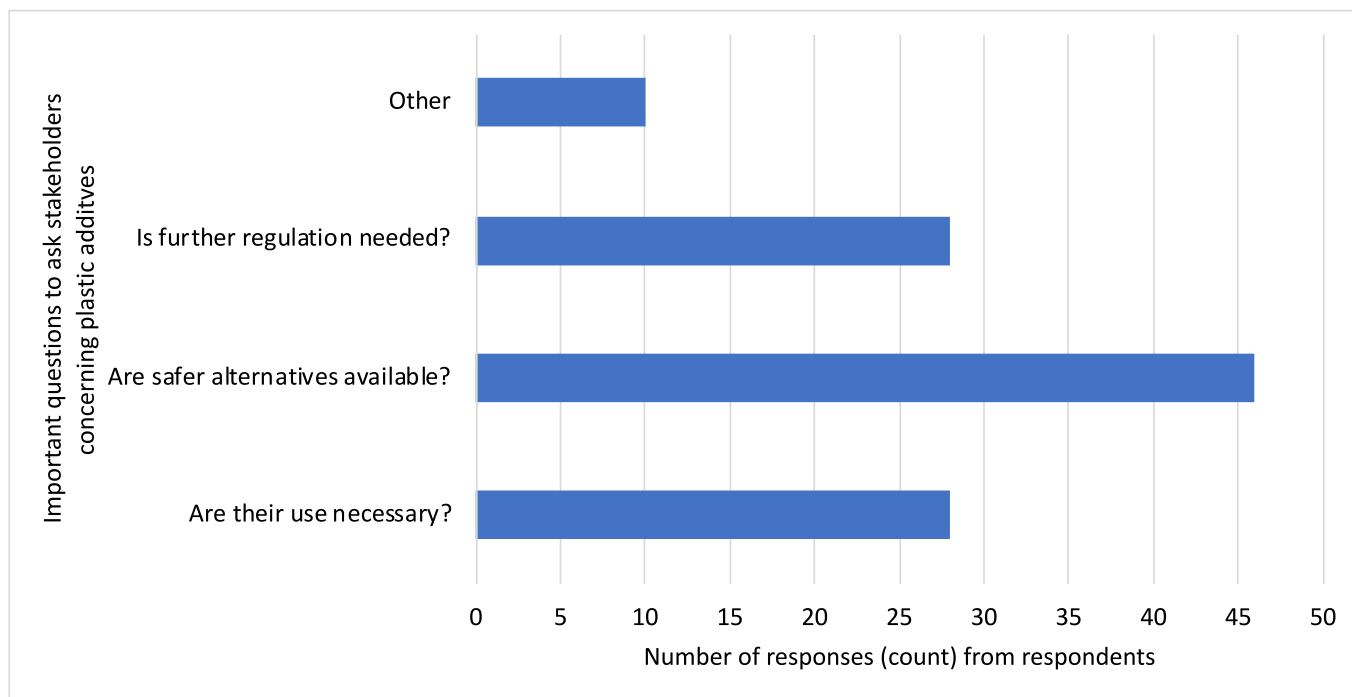


Fig. 9. Respondent's suggestions as to important questions to ask stakeholders (e.g., a policy maker or the plastic industry) concerning the use of plastic additives.

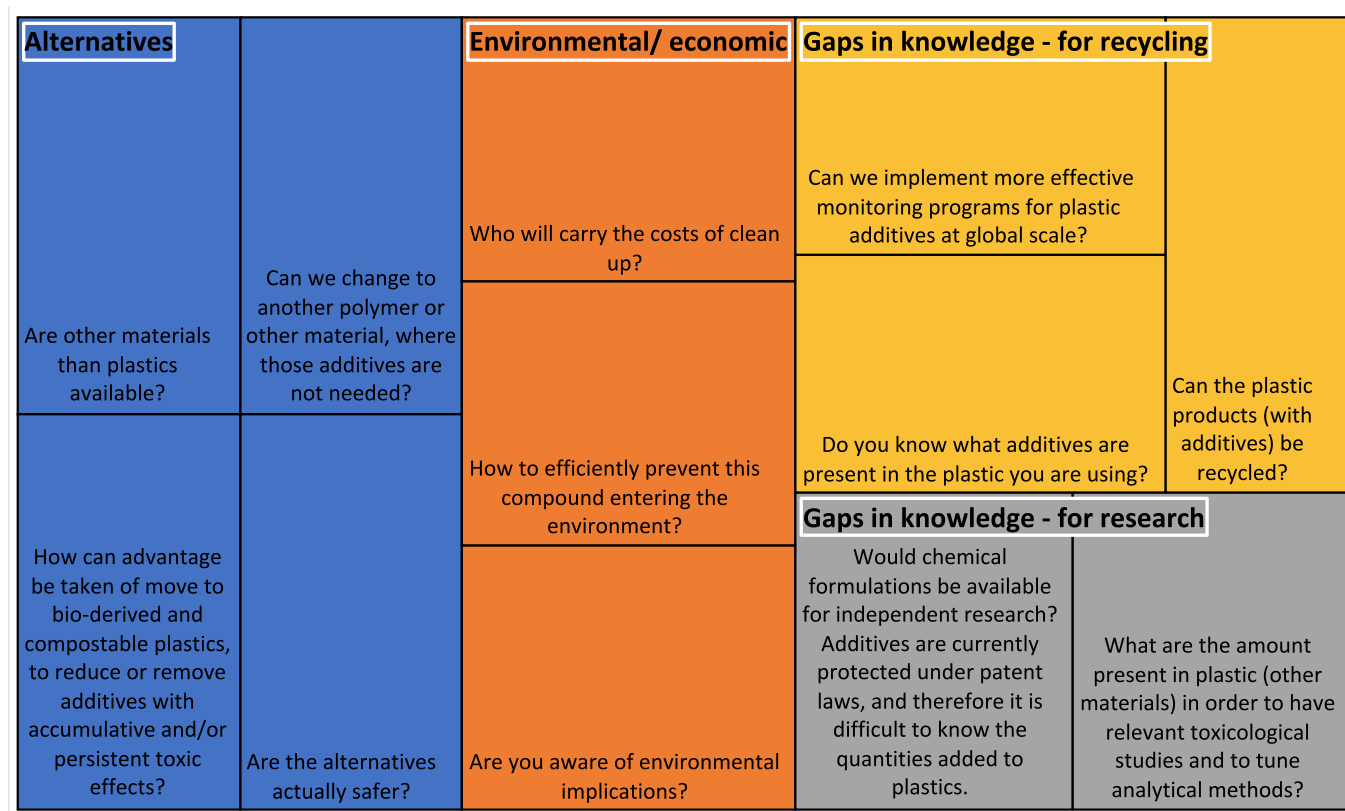


Fig. 10. Tree-map visualisation of the 'other' questions the respondents think are important to ask stakeholders (e.g. policy maker or plastic industry) concerning plastic additives. The hierarchal data is displayed in nested triangles, with each larger rectangle subdivided into smaller ones proportionally representing different methods. Interpretation involves understanding the size and colour of the rectangles to grasp the most commonly suggested questions for each factor considered.

2015). Although PBDEs are not considered to be very mobile due to their high sorptive properties and high hydrophobicity (Environment Agency, 2019), they have been detected in marine sediments across the globe

and in biota at all trophic levels (Barber et al., 2021; de Wit et al., 2010; Law et al., 2014; Yogui and Sericano, 2009). PBDEs can enter the environment from various sources. Wastewater treatment plants have

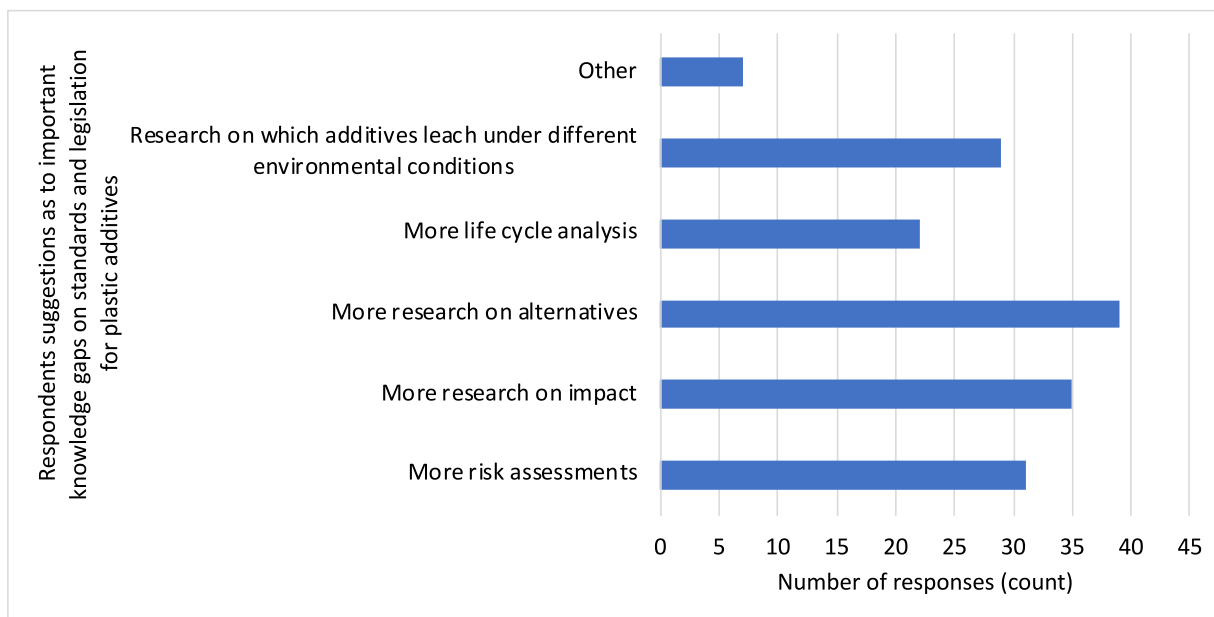


Fig. 11. Respondent’s suggestions as to important knowledge gaps that need to be addressed urgently to support policies on standards and legislation for plastic additives.

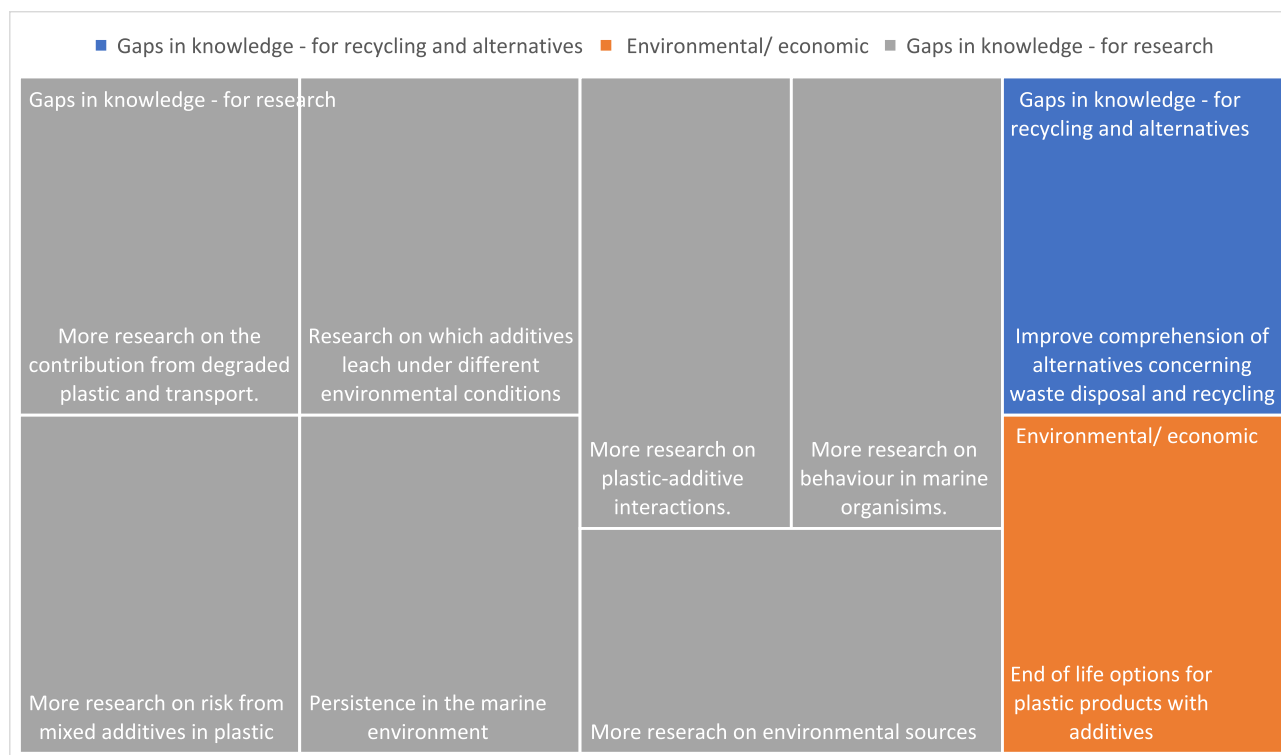


Fig. 12. Tree-map visualisation of the “other” inquiries raised by the respondents, which they believe are crucial knowledge gaps requiring immediate attention to bolster policies on standards and regulations for plastic additives. The hierarchal data is displayed in nested triangles, with each larger rectangle subdivided into smaller ones proportionally representing different methods. Interpretation involves understanding the size and colour of the rectangles to grasp the most commonly suggested questions for each factor considered.

been shown to be a substantial source of PBDEs in the environment. Peng et al. (2009) detected PBDEs in sewage treatment plants in the Pearl River Delta (South China) in concentrations ranging from 13.3 to 2496.4 ng L⁻¹ in the raw wastewater with 0.9 to 4.4 ng L⁻¹ in the treated effluent (Peng et al., 2009). Rocha-Gutierrez and Lee (2013) also investigated the occurrence of PBDEs in Wastewater Treatment Plants

(WWTPs) along the U.S. and Mexico Border with concentrations ranging from 30.2 to 342 ngL⁻¹ in wastewater influents, from not detected to 209 ngL⁻¹ in effluents, and from not detected to 1303 ng g⁻¹ in sludge. Both studies also suggested the incomplete removal of PBDEs from the WWTPs (in some cases) leading to continuous release of PBDEs to rivers and irrigation canals or via sewage sludge (Rocha-Gutierrez and Lee,

2013; Peng et al., 2009).

4.3.3. Organotins

The majority of respondents (72 %) were aware of the pathways, in addition to plastics, of organotins in the marine environment (Fig. 3b). Organotins are mainly used as biocides, polyvinyl chloride stabilisers and industrial catalysts to manufacture silicone and polyurethane foams (Sousa et al., 2014). In the marine environment, organotins were previously used extensively in antifouling solutions and paints, although several of the most toxic, such as tributyltin (TBT), have since been banned. They do, however, remain as a legacy pollutant in places where such paints are still used or where such paints remain on older ships. The environmental behaviour of organotin antifouling compounds in the marine environment is complex, as seawater pH strongly affects their speciation (Beyer et al., 2022). TBT is, however, expected to behave relatively similarly to other hydrophobic organic contaminants under typical seawater pH. TBT-contaminated sediments are likely to play a role as a major reservoir of potential pollution unless active sediment remediation measures, such as dredging and capping, are performed (Beyer et al., 2022; Langston and Pope, 1995).

4.3.4. Metals

Most respondents (92 %) were aware of the pathways, in addition to plastics, of metals into the marine environment (Fig. 3b). Heavy metals are natural constituents of the Earth's crust, generally being found in very low concentrations (Ansari et al., 2004). However, industrial and anthropogenic activities such as industrial waste discharges, agricultural practices, coastal construction and dredging have increased their concentrations in the marine environment (Ansari et al., 2004; Fu and Wang, 2011; Shah, 2021). While plastics do not represent an important source of metals into the marine environment, metals can be found in so-called 'oxo-biodegradable' plastics in which a prooxidative additive is added to initiate degradation. The most common additives on the market are d2w and TDPA (Totally Degradable Plastic Additives), usually produced from cobalt, manganese, and iron compounds (Markowicz and Szymańska-Pulikowska, 2019; Ojeda et al., 2009). Furthermore, a number of plastic additive chemicals and catalysts were based on compounds of toxic metals (and metalloids), like arsenic, cadmium, chromium (VI), and lead. Despite subsequent restrictions, hazardous metal additives remain in plastics in societal circulation, where they continue to leach into the natural environment (Turner and Filella, 2021).

4.3.5. PFAS/PFOS

The majority of the respondents (78 %) were aware of the pathways, in addition to plastics, of PFAS/PFOS into the marine environment (Fig. 3b). PFAS have unique physicochemical properties due to their combination of hydrophilic and lipophilic characteristics. Certain PFAS family chemicals (e.g., PFOS) and PFAS-related substances have been used historically to provide soil, oil and water resistance to textiles, clothes, home furnishings and upholstery, carpets and leather, paper and packaging, and coatings and coating additives. Some major industry sectors using PFAS chemicals include aerospace and defence, automotive, aviation, food contact materials, textiles, leather and apparel, construction and household products, electronics, firefighting, food processing, and medical articles (ECHA, 2022). Despite the recent restrictions on use, older consumer products such as carpets, textiles and upholstery treated with PFOS or PFAS-related substances will continue to act as a source of these chemicals. Emissions can occur during the use, washing and disposal of such items, entering the environment through wastewater treatment works or waste management facilities. The available scientific evidence suggests only a limited removal of PFAS/PFOS is likely to occur through adsorption to sludge. Academic studies indicate that PFOA and PFOS may also be formed during wastewater treatment from precursor compounds (Environment Agency, 2004, 2021; Xiao, 2022). Additional and potentially significant sources of

PFAS to the natural environment include landfill sites, industrial discharges and local historical contamination – especially around sites such as military bases and airports where there may have been significant use of aqueous film-forming foams during firefighting training. PFAS can be present in soil from such historical sources or from spreading sewage sludge to land. Because PFAS is moderately soluble in water, it can enter surface waters from contaminated soil and leach to groundwater. PFAS has a moderate potential to sorb to soil and a lower sorption potential in sediments. The degree of sorption can vary depending on the level of salinity, pH and total organic carbon (Environment Agency, 2004, 2021). PFAS is mainly transported in the dissolved phase in rivers rather than being adsorbed to suspended solids and has the potential for long-range transport in the water column. PFAS has been detected in UK freshwater, estuarine and coastal biota (Environment Agency, 2021) and sediments (Barber et al., 2021). PFAS were detected above quantitation limits in 85 of the 103 sediment samples analysed, at concentrations up to 4.93 µg/kg d.w. for \sum PFAS (or 4.54 µg/kg d.w. normalised to 1 % TOC) (Barber et al., 2021).

4.3.6. Metal acetates

Over half of respondents (54 %) were aware of the pathways, in addition to plastics, of metal acetates into the marine environment (Fig. 3b). For example, zinc acetate is used as the catalyst for the industrial production of vinyl acetate, a precursor to PVA and EVA. Other uses for zinc acetate include its use as a wood preservative, mordant in dyeing, antiseptic, feed additive, a cross-linking agent for polymers, catalyst, waterproofing agent, therapeutic styptic and astringent, and topical fungicide (National Center for Biotechnology Information, 2022b). Sodium acetate is used in the textile industry and synthetic rubber production. Sodium acetate is also used in the food industry as a preservative (E262) and buffer in the cosmetics industry in various personal care products. Metal acetates enter the marine environment mainly from land-based sources via sewage and riverine discharge (National Center for Biotechnology Information, 2022c).

4.3.7. Organophosphates

The majority of respondents (72 %) were aware of the pathways, in addition to plastics, of organophosphates into the marine environment (Fig. 3b). These chemicals are the main components of herbicides, pesticides and insecticides. Organophosphate esters have also been widely used as flame retardants and plasticisers (Castro-Jiménez et al., 2016). Organophosphate flame retardants are not chemically bound to the polymer with potential for leaching into the surrounding environment (Liao et al., 2020). Organophosphates are thought to enter the marine environment mainly from land-based sources via atmospheric transport and riverine discharge (Xie et al., 2022). These chemicals have also been detected in coastal waters and sediments, with the highest concentrations being found mainly near to populated and industrial areas (Liao et al., 2020; Xie et al., 2022).

4.3.8. Aromatic amines

Half of the respondents (50 %) were unsure about the pathways, in addition to plastics, of aromatic amines into the marine environment (Fig. 3a). These chemicals are mainly used in the creation of artificial dyes, as well as oil refining, synthetic polymers, adhesives, rubbers, perfume, pharmaceuticals, pesticides and explosives. Aromatic amines can also be generated from diesel fuel combustion, as well as the combustion of wood chips, rubber and tobacco. Some types of aromatic amines are also generated during cooking (Pereira et al., 2015). Aromatic amines are used as precursors of azo dyes, which represent about two-thirds of all synthetic dyes produced, making them the market's most widely used class of organic dyes (Freeman, 2013). Aromatic amines enter the marine environment mainly from land-based sources via the atmosphere, sewage and riverine discharge (Pereira et al., 2015).

4.3.9. Phthalates

A majority of respondents (78 %) were aware of the pathways, in addition to plastics, of phthalates into the marine environment (Fig. 3b). As phthalates are mainly used as plasticisers, they are found in a wide range of polymer- and plastic-based consumer products, such as adhesives, sealants, paints, rubber materials, wires and cables, flooring, packaging, food contact materials, medical devices, sports equipment and personal care products (ECHA, 2022). The primary sources of phthalate esters in the marine environment include leaching from plasticised plastic materials, industrial water, wastewater discharges and riverine discharge. As some phthalate esters have a low water solubility and a high octanol-water partition coefficient, they can become concentrated in suspended matter and sediment (Arfaeinia et al., 2019). Phthalates can be considered ubiquitous in the environment, with several studies reporting the occurrence and abundance of the commonly used di(2-ethylhexyl) phthalate (DEHP) in environmental samples, including marine waters, sediment and biota (Hidalgo-Serrano et al., 2022). Phthalates are also semi-volatile organic compounds. Their atmospheric transport has also been shown to be an important route of entry in the environment with DEHP and DBP being the main compounds in both indoor and outdoor air phthalates (Jia et al., 2019; Tran et al., 2022). Phthalates can also accumulate in settled dust (Bi et al., 2018; Hua et al., 2022). Urban road dust has also been suggested as an additional transport pathway of phthalates into surface waters (Tran et al., 2022). Their environmental fate, transport and transformation/degradation under natural conditions are, however, highly dependent on their physical and chemical properties (Prasad, 2021).

4.4. Monitoring additives in the environment

The analysis of extractable and leachable organic chemicals from plastic debris is typically performed using gas chromatography mass spectrometry (GC-MS) or liquid chromatography mass spectrometry (LC-MS) techniques. Owing to their sensitivity, specificity, reproducibility, and versatility, both analytical techniques have been widely utilised for the quantification and semi-quantitative screening of leaching compounds, including plastic additives (Adlard, 2019). Analysis of inorganics usually involves spectrometric or spectroscopic detection using inductively coupled plasma mass spectrometry (ICP-MS), ICP atomic/optical emission spectroscopy (ICP-AES/ICP-OES) or atomic absorption spectrometry (AAS) (Dehouck et al., 2016). Most respondents appeared to be relatively knowledgeable about the available instrumentation and the most commonly used analytical techniques for detecting and quantifying the different plastic additives (Fig. 5). This included GC-MS for more non-polar and semi-volatile organic substances such as phthalates, PBDEs and organotins, LC-MS for thermally unstable, polar or non-volatile compounds such as PFAS/PFOS and aromatic amines, and ICP-based approaches for metals.

A majority (at least 50 %) of respondents indicated that standardised methods are available for phthalates, organophosphates, PFAS/PFOS, metals, metal acetates, organotins and PBDE's (Fig. 4). Respondents frequently refer to available ISO standard methods specific to soil or freshwater samples for plastic additive analysis. However, it should be stressed that specific analytical needs might be required for marine samples due to the higher salt concentration in the respective matrices or due to lower contaminant concentrations in the marine environment. For aromatic amines and phenylenediamines in particular, the respondents were typically unable to suggest standardised methods, highlighting the need for efforts on method standardisation to correctly estimate the risks of these groups of plastic additives in the marine environment.

Additional focus needs to be given to providing information on the relative abundance of different additive chemical groups in plastics, as well as improved data regarding the relevant physicochemical properties (e.g., vapour pressure, boiling point, solubility and KOW) for predicting their migration into surrounding media. Furthermore, plastic

additive migration into relevant environmental media should be evaluated through the whole plastic life cycle to identify where such emissions are most likely to occur (Askham et al., 2023). There is also an urgent need for representative additive chemical-containing plastic reference materials to evaluate leaching more accurately and reproducibly, as well as for use in ecotoxicity testing for establishing clear regulatory guidelines. As the methods used to prepare homogenous plastic additive chemical exposure solutions can strongly impact bioavailability and toxicity, harmonised testing approaches need to be developed, which consider marine and freshwater media, also incorporating solid matrices with variable contents of organic matter.

4.5. Risks posed by additives

4.5.1. The risks posed by additives in the environment

The physicochemical properties of the plastic polymers (e.g., polymer type, crystallinity, porosity, degree of degradation), the physicochemical properties of the different additives (e.g., molecular weight, hydrophobicity) and the properties of the surrounding media (pH, temperature, salinity, UV levels) may significantly influence the degree and rate of the diffusion-controlled process of plastic additive migration from plastic items and particles to the environment. When present in aquatic compartments, aqueous leaching of plastic additives into natural waters can result in conventional chemical exposure to organisms. Under certain conditions, plastic additives may also leach directly into organisms following the ingestion of plastic particles (Kühn and van Franeker, 2020). Therefore, risk assessment of plastic additives must also consider their persistence in the natural environment and their potential for accumulation in specific matrices (e.g., sediments) and bioaccumulation across different trophic levels. Exposure to released plastic additives could potentially lead to multiple adverse effects in a range of organisms. In humans, exposure to additives have been linked with impacting childhood development, the nervous and endocrine system (Lithner et al., 2011), impairing reproductive functions and causing carcinogenic effects, even at very low doses (Aurisano et al., 2021; Meeker et al., 2009). However, only a limited number of additives have been widely studied, with even less considering low dose, long-term or mixed substance effects. Still, they may include harmful compounds such as bisphenol A (BPA) or metals, including cadmium, lead or arsenic (van Oers et al., 2012).

Knowing and dissociating the mechanisms of plastic toxicity that are driven by (i) the physical polymer material and (ii) plastic-associated chemicals (additives and NIAS) is of high importance for conducting robust risk assessment and for the development of effective mitigation strategies. The risks associated with any chemicals emitted into the environment can be divided into two assessment categories. First is the type of risk that a chemical presents; Is the chemical persistent, bioaccumulative or toxic to organisms, or a combination of all three. Second is the trophic level at which the chemical represents a hazard; Is the chemical a threat to lower or higher marine trophic levels, or both? In this study, we assessed experts' understanding regarding the risks linked to plastic additives, identified which trophic levels are most susceptible to these risks, and then compared this with existing threshold values for specific chemicals.

4.5.2. PBT assessment

The survey results show that the respondents are confident of their knowledge regarding the PBT properties of Figure 6 metal, PFAS/PFOS and phthalate plastic additives (Fig. 6b). This might indicate there is a comprehensive level of research and widespread dissemination of information available within the scientific community and beyond, particularly for these specific sets of plastic additives. Furthermore, some of the general PBT data available for these chemicals and metals derives from a wealth of research conducted over many decades that has not been directly focused on their use as plastic additives. In contrast, many respondents expressed a low level of knowledge regarding the PBT

properties of phenylenediamines, methyl acetates, organophosphates and aromatic amines (Fig. 6a). This result indicates a low level of knowledge regarding the risks associated with these plastic additives, including from individuals working directly in the field of plastics and additives. While some PBT data is available for these groups of chemicals, they are less studied and there has been inadequate knowledge transfer of the existing data to the broader scientific community and the public. This lack of knowledge and knowledge transfer has the potential to result in delayed scientific advice and preventive actions. It is also important to note that regulatory toxicity assessment of most chemicals is conducted in single compounds, generally using methods that employ high-dose, short-term responses of a specific indicator organism (often a freshwater organism). This approach limits our understanding of long-term, low-dose impacts and mixed chemical exposures throughout the food web (Leslie et al., 2022), typically only defining a quantitative boundary for safe exposure levels regarding a specific chemical or compound. Only a few chemical pollutants have been defined by quantitative boundaries with regards to causing ecosystem-level impacts (e.g., CO₂, CFCs).

4.5.3. Risks at different trophic levels

Understanding the PBT of additives within the marine environment is an important part in understanding the risks additives pose, across different trophic levels. The ecotoxicological consequences of plastic additives are gaining interest within the research community, however, there often remains a disconnect between persistent, bioaccumulative, and/or toxic data generated for individual chemicals within in each plastic additive chemical group included in the current study and the link to (micro)plastic toxicity interpretation.

In this study, respondents were asked to consider the perceived risk associated with the different plastic additives to higher and lower trophic level marine organisms in function of the persistent, bioaccumulative and/or toxic nature of each chemical group. For most additives, the respondents felt they had a greater knowledge of the risks to higher trophic marine level organisms than for those from lower trophic levels (Figs. 7 and 8). This difference in perceived knowledge presents a somewhat unusual situation given that bioaccumulation and toxicity data is typically much more widely available for lower trophic organisms, given the low cost and relative ease of producing it. It is important to note that the respondents may have a scientific background more strongly anchored in PBT and risk knowledge related to higher trophic-level organisms.

The literature review revealed plastic additives toxicity threshold levels for a range of aquatic organisms from different trophic levels, at both sublethal and lethal exposures (Table & Fig. S7, Supplementary Information). Fig. S7 shows the log toxicity threshold values for three different trophic levels that have been calculated for a selection of plastic additive chemicals representing the chemical groups included in the current study. The LOEC for each substance was: 0.1 µg/L for BDE-209, 0.5 µg/L for fentin acetate, 15 µg/L for lead, 18.2 µg/L for 4-4-methylenedianiline, 25 µg/L for 6-PPD, 100 µg/L for benzyl butyl phthalate, 2.2 mg/L for tris(2-chloroethyl) phosphate and 6.25 mg/L for perfluorooctanoic acid. This information was used to discuss the respondents answers in Section 3.4.

Data for different trophic levels is often missing, therefore, determining the risk of plastic additives to organisms at different trophic levels is thus important to address in the future, as this may vary significantly between individual chemicals. The literature review (see SI, Table and Fig. S7) suggests that benzyl butyl phthalate and BDE-209 pose a hazard to fish only, 6-PPD pose a hazard to invertebrates and fish (no microalgae data were available), while fentin acetate may impact all three trophic levels and could threaten aquatic ecosystems. The use of these four specific plastic additives, as well as others in the respective chemical groups, should be considered for urgent regulation.

4.5.3.1. Risks to lower trophic levels. The majority of respondents perceived the risk of organotins (66 %) and, to a slightly lesser extent, organophosphates (53 %) as being high for low marine trophic levels (Fig. 7b). This result is supported in part by the reasonable quantity of available toxicity data for these two plastic additive chemical groups. For example, fentin acetate (organotin group chemical) was the most toxic to microalgae (Walsh et al., 1985; Fargašová, 1996) and to invertebrates (Roessink et al., 2005; Novelli et al., 2002) among the selected compound groups (SI, Fig. S7). Despite a perceived high risk by respondents, the tris(2-chloroethyl) phosphate (organophosphate group chemical) has been reported to cause only moderate toxicity to aquatic invertebrates (Yoshioka, 1986) (SI, Fig. S7). In the case of aromatic amines, metal acetates and phenylenediamines, respondents typically perceived these compound groups to be a lower risk. In this case, the perceived low risk of phenylenediamines contrasts with the reported toxicity of these substances to marine invertebrates (SI, Fig. S7). Specifically, the phenylenediamine 6-PPD has been shown to affect immobility, population growth and mortality of aquatic invertebrates (Isanta-Navarro et al., 2022; Hiki and Yamamoto, 2022). The results suggest that the respondents had more accurate risk perceptions for the more common and widely discussed plastic additive chemical groups, especially those representing chemicals and chemical classes that have been studied for many years. In contrast, lesser known or emerging plastic additive chemical groups saw more discrepancies between perceived and experimentally determined toxicity and risks. However, it is important to highlight that >35 % of respondents provided a 'do not know' response to all plastic additive chemical groups, with the exception of metals (24 %). In the case of metal acetates, aromatic amines and phenylenediamines, >50 % of respondents gave an answer of 'do not know' (Fig. 7a).

4.5.3.2. Risks to higher trophic levels. The majority of respondents (60–75 %) perceived the risk of phthalates, organophosphates, PFAS/PFOS, metals and organotins as being high for high marine trophic levels (Fig. 8b). The perceived high risk for organotins and phthalates is supported by the available toxicity data for these compounds (SI, Fig. S7). Fentin acetate caused cytological alterations (Strmac and Braunbeck, 1999), and benzyl butyl phthalate caused effects on the feeding and shoaling behaviour of fish (Wibe et al., 2002; Kaplan et al., 2013). However, the available data for metals (Pb) indicate only moderate toxicity to fish (Reynolds et al., 2018; Karthikeyan et al., 2021), while organophosphates and PFAS/PFOS substances presented the lowest toxicity among the selected substances (Giesy et al., 2010) (SI, Fig. S7). Although the risk of PBDEs and phenylenediamines for high trophic levels was mostly perceived as moderate to high by the respondents (Fig. 8), the available toxicity data indicate that BDE-209 and 6-PPD are highly toxic to fish (SI, Fig. S7). BDE-209 has been shown to cause oxidative damage and genotoxicity in fish, affecting their swimming activity (Pérez-Iglesias et al., 2022), whereas 6-PPD affected the hatching and swimming behaviour and inhibited acetylcholinesterase activity (Hiki and Yamamoto, 2022; Ji et al., 2022; Varshney et al., 2022). Again, the results suggest that the respondents had more accurate risk perceptions for the more common and widely discussed plastic additive chemical groups (e.g., organotins, phthalates), especially those representing chemicals and chemical classes that have been studied for many years. Similar to the low trophic level risks, lesser known or emerging plastic additive chemical groups (e.g., phenylenediamines) saw more discrepancies between perceived and experimentally determined toxicity and risks for higher trophic levels. It is again worth noting that >30 % of respondents provided a 'do not know' response for the organophosphate, organotin, aromatic amine, metal acetate and phenylenediamines plastic additive chemical groups, with this rising to >50 % of respondents for the last three groups (Fig. 8a). Overall, the results from the low and high trophic levels questions indicate a specific lack of knowledge regarding the risk of the aromatic amine, metal

acetate and phenylenediamines plastic additive chemical groups. This finding is not necessarily unexpected for metal acetates, for which there is scarce aquatic toxicity data, and for phenylenediamines, for which there is fragmentary information. However, there is more information available on the toxicity of aromatic amines in aquatic organisms, indicating that the flow and dissemination of information for this specific group of plastic additive chemicals may not be sufficient.

4.5.4. Limitations in studying the risks of additives

The toxicity studies with plastic additives conducted to date demonstrate that the choice of organism, assay and biological response is an important consideration (Karami, 2017). For example, standard acute toxicity assays may lack realism and alternative long-term exposure experiments are needed (Barrick et al., 2021). Similarly, certain sublethal biological responses may be more relevant for some additive groups than others, depending on the mechanism of toxicity of each type of substance (Capolupo et al., 2021). The integration of sublethal stress indices covering a variety of toxicity mechanisms (e.g., neurotoxicity, endocrine disruption, genotoxicity, oxidative stress, immunotoxicity) into plastic additive toxicity assessment can assist in documenting a broader range of potential adverse outcome pathways (Jeong and Choi, 2022). Moreover, it may help in the development of toxicodynamic assessments for plastic additives, where integrating a high throughput assay that tests an extended selection of different biological endpoints over short timescales may assist in differentiating the toxic effect of plastic particles from that of additives, as well as between one group of additives and another (Gao et al., 2022). Differentiation of the toxicological effects of plastic particles from their associated chemical additives and NIAS can be achieved through the use of combined testing approaches using exposures comprising samples representing (i) plastic particles plus leachates, and (ii) leachates only. There is also potential to conduct long term leaching of chemicals from plastic particles followed by isolation and testing of the pre-leached particles in toxicity studies to identify the effects derived only from the physical particles. Measuring endpoints at lower levels of the biological organisation may become more relevant in the future, while assays that selectively follow up the different toxicity pathways may be necessary to characterise individual and ecosystem effects fully (Lemos, 2021).

A correct interpretation of additives toxicity is further advocated when considering chemical-physical processes such as microplastics weathering and ageing. This makes ecotoxicity assessment challenging as the number of hypothetically leachable additives can result in a cocktail of chemical toxicants and stressors (Hahladakis et al., 2018). Different plastic polymers display diverse degradation patterns within similar environmental conditions, which further confuses the hazard classification identification of both the plastic polymers as well as the related additives (Andrady and Rajapakse, 2016). Ecotoxicological assessment entails the capability to differentiate between the effects of microplastics from those related to the degradation phenomena.

4.6. Limitations of this study

The authors acknowledge that the number of respondents (50) and, specifically, their geographical and employment range is a limitation of this study. The number of experts within the field of additives and toxicology is relatively small. As such, the number of respondents itself is not a problem and reflects the limited number of experts in the field. However, the geographical range is dominated by Europe (82 %) followed by Sub Saharan Africa (10 %), with limited or no responses from other regions (Fig. 1). Thus, the present assessment does not represent the full range of experts at the global level. The respondents were dominated by academia (78 %, Fig. 1), but did include a small number of respondents in the categories of government, NGO/IGOs and civil society representatives. However, no industry representatives completed the survey. Insight information from industry would have been highly beneficial to this study – as this could have potentially highlighted any

reduced knowledge transfer between the plastic production industry and other stakeholder experts.

5. Conclusions

To fully understand and assess the impact of plastic additives on the (marine) environment, detailed information on production volumes, usage, pathways and risks is indispensable. This study represents the perspectives and current knowledge of highly educated, mainly research and academic, experts, predominantly in Europe, concerning chemical use for additives in plastics and other applications globally.

5.1.1. Use of additives

This survey demonstrated a lack of knowledge on production volumes and use of plastic-related additives, particularly metal acetates, aromatic amines and phenylenediamines. Survey participants expressed greater awareness regarding the production quantities of phthalates, metals, and PFAS, and they noted that these additives are manufactured in substantial amounts.

5.1.2. Characteristics of additives

In this study, most of the responses emphasized that there are broader sources and routes, beyond plastics, for most additives. There are substantial gaps in knowledge regarding comparative ecotoxicity studies of diverse polymer types and the effects of the related weathering phenomena. These can increase and decrease (micro)plastics' sorption capacity for environmental contaminants and alter their mobility through environmental compartments. Although the results of many studies investigating the effects of plastic leachates have been presented, there is a lack of comparative studies that differentiate between the impact of plastic degradation and the co-occurring release of additives. Other issues of concern among experts revolve around what occurs to these plastic additives as plastics break down or how they affect recycling procedures, which underscore the necessity for comprehensive Life Cycle Assessment (LCA) studies.

5.1.3. Monitoring of additives

Standardised analytical methods are available for phthalates, organophosphates, PFAS/PFOS, metals, and PBDE's. For the other chemical groups, methods are available but not yet standardised, hampering further assessment and our understanding of their presence and environmental impact. The survey highlighted that the availability of standardised analytical chemical methods is limited but critical for conducting robust environmental concentration determination and for supporting the toxicity testing necessary for assessing the potential risks different types of plastic additive chemicals present to the marine environment. In addition to improving and standardising methods to measure the presence of plastic additives, there is also a need to improve the current methods for characterising the leaching of additives from plastics into different environmental media, i.e., water, sediment and biota.

5.1.4. Risks of additives

This survey highlights a low knowledge by survey participants of any PBT nature of additives. The PBT nature is only known for metals, PFAS/PFOS and phthalates, while a larger group of respondents expressed to be unsure about the PBT aspects of phenylenediamines, aromatic amines and methyl acetates. Overall, the respondents show a greater knowledge of the risk to higher marine trophic levels than lower trophic levels for most additives. The knowledge gaps on specific chemical additive groups indicate that risk assessments, which rely on an understanding of the presence and toxicity of those chemicals, are lacking crucial data to complete. With more evidence indicating that plastic-associated

chemicals (additives and NIAS) may be one of the primary drivers for observed toxicological responses to plastic exposure, there is a need for holistic risk assessments that consider and separate plastics, additives and their associated contaminants.

In addition, there are many questions about the potential implications of using plastic additives, both environmental and public health-focused, highlighting the need for in-depth environmental and risk assessment studies by experts.

5.1.5. Way forward

In-depth environmental and risk assessment studies are also inextricably linked to strengthening transparency on the use of specific plastic additives in products and materials toward users. Survey participants highlight the importance of using and researching safer alternatives and closing knowledge gaps as the way forward. However, such transitions to other options must be dealt with caution, 99 % of plastic feedstocks are fossil fuel-based (Nielsen et al., 2020). Although more alternative, bio-based and biodegradable plastic materials are emerging to market, many still rely on the same additives to provide aesthetics and functionality (Gómez and Michel, 2013). Restrictions on some chemical use have led to similar chemicals being used, which may have other undesired consequences if not thoroughly evaluated, such as the shift in BPA to BPS (Qiu et al., 2021; Rochester and Bolden, 2015). Overall, collaboration is needed between environmental scientists, chemists, design engineers, manufacturers and other relevant stakeholders to transition to safe and sustainable chemical use.

5.1.6. A chemical group of concern - phenylenediamines e.g., 6-PPD

A chemical group of concern highlighted from this survey are phenylenediamines – e.g., 6-PPD. Phenylenediamines, particularly 6-PPD, come up repeatedly in this study with knowledge gaps across pathways into the environment, available analytical methods, nature (i.e., persistent, bioaccumulative, and toxic nature) and risk across tropic levels. It is noted that other additives (aromatic amines, metal acetates and methyl acetates) all show data gaps within these areas. However, only phenylenediamines are identified to have gaps in knowledge across all areas. Combined with what is known, these gaps indicate that 6-PPD is an emerging pollutant, making 6-PPD a growing concern. The additive 6-PPD has been used in tires since the mid-1960s as a tire rubber antioxidant additive. However, it was only in 2020 that its degradation was identified (Tian et al., 2021). This degradation turns 6-PPD into 6-PPD-quinone, which is toxic and has been determined to cause acute mortality in salmon populations in urbanised rivers (Tian et al., 2021). What happens to the additives and their degradation in the environment is a knowledge gap that highlights the need for more focused risk assessments, as additives can react with each other or contaminants or degrade to form (as with 6-PPD) and attract other toxic molecules (Hahladakis et al., 2018).

5.2. Implications for policy

Plastics may impact human and environmental health due to the physical effects caused as particles as well as the (toxicological) effects caused by plastic additive chemicals. A holistic definition of plastic encompassing chemical additives is needed to address the impact of plastic in legislation. Strategies such as the EU Green deal and the Zero Pollution Action Plan 2030 or environmental goals such as the SDG14 targets or MSFD criteria do not specifically address the evaluation of plastic additives in marine environments. In the lead-up to the UN Plastic Treaty, experts and scientists are pushing for the broader definition of plastic as a material made of chemicals, giving the use and disposal of plastic additives a legally binding framework.

The experts from this survey recognise the importance of regulation and legislation in mitigation. Policies and decision making have already been used to mitigate additives, with some additives banned by international and national regulations (Wagner and Schlummer, 2020).

Despite such legislation, some of these illegal additives are still in circulation due to a lack of consistent enforcement (Turner and Filella, 2021), improper waste or recycling management (Wagner and Schlummer, 2020), or exemptions (UNEP, 2021). In light of these fragmented efforts, the recent UNEA resolution 5/14 in March 2022 requesting the convening of an intergovernmental negotiating committee (INC) to develop an international legally binding agreement on plastic pollution, including in the marine environment, provides an opportunity for alignment and development of a comprehensive set of standards and methods that considers plastics and their additives. Global leaders and policy mechanisms such as this are needed to support the development of National Action Plans (NAPs) on marine litter and plastic pollution, promoting universal bans of harmful substances, aligning chemicals of concern lists, agreeing to threshold limits for substances of concern in use, as well as aligning testing methods to evaluate the safety of substances and products.

CRedit authorship contribution statement

Thomas Maes: Conceptualization, Methodology, Validation, Resources, Writing – original draft, Writing – review & editing, Supervision, Project administration, Funding acquisition. **Fiona Preston-Whyte:** Methodology, Validation, Investigation, Writing – original draft, Writing – review & editing, Project administration. **Stephanie Lavelle:** Methodology, Validation, Formal analysis, Data curation, Writing – original draft, Writing – review & editing, Visualization. **Alessio Gomiero:** Validation, Writing – review & editing. **Andy M. Booth:** Validation, Writing – review & editing. **Maria Jesus Belzunce-Segarra:** Validation, Writing – review & editing. **Juan Bellas:** Validation, Writing – review & editing. **Steven Brooks:** Validation, Writing – review & editing. **Adil Bakir:** Validation, Writing – review & editing. **Lisa I. Devriese:** Validation, Writing – review & editing. **Christopher Kim Pham:** Validation, Writing – review & editing. **Bavo De Witte:** Validation, Writing – review & editing, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The data that has been used is confidential.

Acknowledgements

The authors would like to thank all the respondents for giving their valuable time and technical expertise to complete the survey. The authors would like to thank the International Council for the Exploration of the Sea (ICES) facilitating cooperation between different ICES expert groups (working group on marine litter (WGML); marine chemistry working group (MCWG), working group on the biological effect of contamination (WGBEC)). Thomas Maes, Andy Booth and Lisa Devriese received funding from the European Union's Horizon Europe programme under grant agreement N0. 101060213 (SOS Zeropol2030 project). Alessio Gomiero and Andy Booth received funding from the North Atlantic Microplastics Centre –NAMC.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.marpolbul.2023.115633>.

References

- Adlard, E.R., 2019. Michele Suman (Ed): food contact materials analysis: mass spectrometry techniques. *Chromatographia* 82, 1585–1586. <https://doi.org/10.1007/s10337-019-03770-5>.
- Andrade, H., Glüge, J., Herzke, D., et al., 2021. Oceanic long-range transport of organic additives present in plastic products: an overview. *Environ. Sci. Eur.* 33, 85. <https://doi.org/10.1186/s12302-021-00522-x>.
- Andrady, A.L., Rajapakse, N., 2016. In: Takada, H., Karapanagioti, H. (Eds.), *Hazardous Chemicals Associated with Plastics in the Marine Environment, The Handbook of Environmental Chemistry*, 78. Springer, Cham. https://doi.org/10.1007/978_2016_124.
- Ansari, T.M., Marr, I.L., Tariq, N., 2004. Heavy metals in marine pollution perspective—a mini review. *J. Appl. Sci.* 4 (1), 1–20.
- Arfaeina, H., Fazlzadeh, M., Taghizadeh, F., Saeedi, R., Spitz, J., Dobaradaran, S., 2019. Phthalate acid esters (PAEs) accumulation in coastal sediments from regions with different land use configuration along the Persian Gulf. *Ecotoxicol. Environ. Saf.* 169, 496–506.
- Askham, C., Pauna, V.H., Boulay, A.M., Fantke, P., Jolliet, O., Lavoie, J., Booth, A.M., Coutiris, C., Verones, F., Weber, M., Vijver, M.G., Lusher, A., Hajjar, C., 2023. Generating environmental sampling and testing data for micro- and nanoplastics for use in life cycle impact assessment. *Sci. Total Environ.* 859 (Pt 2), 160038 <https://doi.org/10.1016/j.scitotenv.2022.160038>. Feb 10. Epub 2022 Nov 14. PMID: 36395847; PMCID: PMC9760571.
- Aurisano, N., Huang, L., Milà i Canals, L., Jolliet, O., Fantke, P., 2021. Chemicals of concern in plastic toys. *Environ. Int.* 146, 106194 <https://doi.org/10.1016/j.envint.2020.106194>.
- Barber, J.L., Miles, A., Losada, S., van Kesteren, J., Bergmann, T., 2021. Project C8115: Determining Levels of PFAS Chemicals in Estuarine and Coastal Sediments.
- Barrick, A., Champeau, O., Chatel, A., Manier, N., Northcott, G., Tremblay, L.A., 2021. Plastic additives: challenges in ecotoxic hazard assessment. *PeerJ* 9, e11300. <https://doi.org/10.7717/peerj.11300>.
- Beyer, J., Song, Y., Tollefsen, K.E., Berge, J.A., Tveiten, L., Helland, A., Øxnevad, S., Schøyen, M., 2022. The ecotoxicology of marine tributyltin (TBT) hotspots: a review. *Mar. Environ. Res.* 105689.
- Bi, C., Maestre, J.P., Li, H., Zhang, G., Givehchi, R., Mahdavi, A., Kinney, K.A., Siegel, J., Horner, S.D., Xu, Y., 2018. Phthalates and organophosphates in settled dust and HVAC filter dust of US low-income homes: association with season, building characteristics, and childhood asthma. *Environ. Int.* 121, 916–930.
- Borrelle, S.B., Ringma, J., Law, K.L., Monnahan, C.C., Lebreton, L., Mcgovern, A., Murphy, E., Jambeck, J., Leonard, G.H., Hilleary, M.A., Eriksen, M., Possingham, H. P., De Frond, H., Gerber, L.R., Polidoro, B., Tahir, A., Bernard, M., Mallos, N., Barnes, M., Rochman, C.M., 2020. Predicted growth in plastic waste exceeds efforts to mitigate plastic pollution. *Science* 1518 (September), 1515–1518. <https://doi.org/10.1126/science.aba3656>.
- Bridson, J.H., Gaugler, E.C., Smith, D.A., Northcott, G.L., Gaw, S., 2021. Leaching and extraction of additives from plastic pollution to inform environmental risk: a multidisciplinary review of analytical approaches. *J. Hazard. Mater.* 414, 125571 <https://doi.org/10.1016/j.jhazmat.2021.125571>.
- Capolupo, M., Sørensen, L., Jayasena, K.D.R., Booth, A.M., Fabbri, E., 2020. Chemical composition and ecotoxicity of plastic and car tire rubber leachates to aquatic organisms. *Water Res.* 169, 115270.
- Capolupo, M., Gunaalan, K., Booth, A.M., Sørensen, L., Valbonesi, P., Fabbri, E., 2021. The sub-lethal impact of plastic and tire rubber leachates on the Mediterranean mussel *Mytilus galloprovincialis*. *Environ. Pollut.* 283, 117081.
- Castro-Jiménez, J., González-Gaya, B., Pizarro, M., Casal, P., Pizarro-Álvarez, C., Dachs, J., 2016. Organophosphate ester flame retardants and plasticizers in the global oceanic atmosphere. *Environ. Sci. Technol.* 50 (23), 12831–12839.
- Corami, F., Rosso, B., Morabito, E., Rensi, V., Gambaro, A., Barbante, C., 2021. Small microplastics (<100 µm), plasticizers and additives in seawater and sediments: oleo-extraction, purification, quantification, and polymer characterization using Micro-FTIR. *Sci. Total Environ.* 797, 148937 <https://doi.org/10.1016/j.scitotenv.2021.148937>.
- Corami, F., Rosso, B., Sfriso, A.A., Gambaro, A., Mistri, M., Munari, C., Barbante, C., 2022. Additives, plasticizers, small microplastics (<100 µm), and other microlitter components in the gastrointestinal tract of commercial teleost fish: method of extraction, purification, quantification, and characterization using Micro-FTIR. *Mar. Pollut. Bull.* 177, 113477 <https://doi.org/10.1016/j.marpolbul.2022.113477>.
- Dehouck, P., Cordeiro, F., Snell, J., de la Calle, B., 2016. State of the art in the determination of trace elements in seawater: a worldwide proficiency test. *Anal. Bioanal. Chem.* 408 (12), 3223–3232. <https://doi.org/10.1007/s00216-016-9390-6>. May. Epub 2016 Feb 17. PMID: 26886745; PMCID: PMC4830889.
- de Wit, C.A., Herzke, D., Vorkamp, K., 2010. Brominated flame retardants in the Arctic environment—trends and new candidates. *Sci. Total Environ.* 408 (15), 2885–2918.
- ECHA, 2022. Phthalates. <https://echa.europa.eu/hot-topics/phthalates>.
- Environment Agency, 2004. *Environment Agency (2004) Environmental Risk Evaluation Report: Perfluorooctanesulphonate (PFOS)*.
- Environment Agency, 2019. *Polybrominated Diphenyl Ethers (PBDEs): Sources, Pathways and Environmental Data*.
- Environment Agency, 2021. *Perfluorooctane Sulphonate (PFOS) and Related Substances: Challenges for the Environment*.
- Fargašová, A., 1996. Inhibitive effect of organotin compounds on the chlorophyll content of the green freshwater alga *Scenedesmus quadricauda*. *Bull. Environ. Contam. Toxicol.* 57 (99), 106. <https://doi.org/10.1007/s001289900161>.
- Fausser, P., Vorkamp, K., Strand, J., 2022. Residual additives in marine microplastics and their risk assessment – a critical review. *Mar. Pollut. Bull.* 177, 113467 <https://doi.org/10.1016/j.marpolbul.2022.113467>.
- Freeman, H.S., 2013. Aromatic amines: use in azo dye chemistry. *Front. Biosci.-Landmark* 18 (1), 145–164.
- Fu, F., Wang, Q., 2011. Removal of heavy metal ions from wastewaters: a review. *J. Environ. Manag.* 92 (3), 407–418.
- Gao, Z., Yu, H., Li, M., Li, X., Lei, J., He, D., Wu, G., Fu, Y., Chen, Q., Shi, H., 2022. A battery of baseline toxicity bioassays directed evaluation of plastic leachates towards the establishment of bioanalytical monitoring tools for plastics. *Sci. Total Environ.* 1 (828), 154387 <https://doi.org/10.1016/j.scitotenv.2022.154387>. Jul. Epub 2022 Mar 8. PMID: 35276177.
- Geueke, B., Groh, K., Muncke, J., 2018. Food packaging in the circular economy: overview of chemical safety aspects for commonly used materials. *J. Clean. Prod.* 193, 491–505. <https://doi.org/10.1016/j.jclepro.2018.05.005>.
- Geyer, R., 2020. Chapter 2—production, use, and fate of synthetic polymers. In: Letcher, T.M. (Ed.), *Plastic Waste and Recycling*. Academic Press, pp. 13–32. <https://doi.org/10.1016/B978-0-12-817880-5.00002-5>.
- Geyer, R., Jambeck, J.R., Law, K.L., 2017. Production, use, and fate of all plastics ever made. *Sci. Adv.* 3 (7), 25–29. <https://doi.org/10.1126/sciadv.1700782>.
- Giesy, J.P., Naile, J.E., Khim, J.S., Jones, P.D., Newsted, J.L., 2010. Aquatic toxicology of perfluorinated chemicals. *Rev. Environ. Contam. Toxicol.* 202, 1–52. https://doi.org/10.1007/978-1-4419-1157-5_1. PMID: 19898760.
- Gómez, E.F., Michel, F., 2013. Biodegradability of conventional and bio-based plastics and natural fiber composites during composting, anaerobic digestion and long-term soil incubation. *Polym. Degrad. Stab.* 98, 2583–2591.
- Groh, K.J., Backhaus, T., Carney-Almroth, B., Geueke, B., Inostroza, P.A., Lennquist, A., Leslie, H.A., Maffini, M., Slunge, D., Trasande, L., Warhurst, A.M., Muncke, J., 2019. Overview of known plastic packaging-associated chemicals and their hazards. *Sci. Total Environ.* 651, 3253–3268. <https://doi.org/10.1016/j.scitotenv.2018.10.015>.
- Hahladakis, J.N., Velis, C.A., Weber, R., Iacovidou, E., Purnell, P., 2018. An overview of chemical additives present in plastics: migration, release, fate and environmental impact during their use, disposal and recycling. *J. Hazard. Mater.* 344, 179–199. <https://doi.org/10.1016/j.jhazmat.2017.10.014>. ISSN 0304-3894.
- Halsband, C., Sørensen, L., Booth, A.M., Herzke, D., 2020. Car tire crumb rubber: does leaching produce a toxic chemical cocktail in coastal marine systems? *Front. Environ. Sci.* 125.
- Hansen, E., Nilson, N.H., Lithner, L., Lassen, C., 2013. Hazardous substances in plastic materials. In: *COWI in Corporation With Danish Technological Institute*, vol. 11(12), p. 16.
- Hermabessiere, L., Dehaut, A., Paul-Pont, I., Lacroix, C., Jézequel, R., Soudant, P., Duflos, G., 2017. Occurrence and effects of plastic additives on marine organisms and organisms: a review. *Chemosphere* 182, 781–793. <https://doi.org/10.1016/j.chemosphere.2017.05.096>.
- Hidalgo-Serrano, M., Borrull, F., Marcé, R.M., Pocurull, E., 2022. Phthalate esters in marine ecosystems: analytical methods, occurrence and distribution. *TrAC Trends Anal. Chem.* 116598.
- Hiki, K., Yamamoto, H., 2022. Concentration and leachability of N-(1,3-dimethylbutyl)-N-phenyl-p-phenylenediamine (6PPD) and its quinone transformation product (6PPD-Q) in road dust collected in Tokyo, Japan. *Environ. Pollut.* 302, 119082 <https://doi.org/10.1016/j.envpol.2022.119082>.
- Holland, M., 2018. Socio-economic assessment of phthalates. In: *OECD Environment Working Papers*, No. 133. OECD Publishing, Paris. <https://doi.org/10.1787/a38a0e34-en>.
- Hua, L., Guo, S., Xu, J., Yang, X., Zhu, H., Yao, Y., Zhu, L., Li, Y., Zhang, J., Sun, H., 2022. Phthalates in dormitory dust and human urine: a study of exposure characteristics and risk assessments of university students. *Sci. Total Environ.* 845, 157251.
- Huang, W., Shi, Y., Huang, J., Deng, C., Tang, S., Liu, X., Chen, D., 2021. Occurrence of substituted p-phenylenediamine antioxidants in dusts. *Environ. Sci. Technol. Lett.* 8 (5), 381–385.
- Isanta-Navarro, J., Klauschie, T., Wacker, A., Martin-Creuzburg, D., 2022. A sterol-mediated gleaner-opportunist trade-off underlies the evolution of grazer resistance to cyanobacteria. *Proc. R. Soc. B* 289.
- Jeong, J., Choi, J., 2022. Artificial intelligence-based toxicity prediction of environmental chemicals: future directions for chemical management applications. *Environ. Sci. Technol.* 56 (12), 7532–7543. <https://doi.org/10.1021/acs.est.1c07413>. Jun 21. Epub 2022 Jun 6. PMID: 35666838.
- Ji, J., Huang, J., Cao, N., Hao, X., Wu, Y., Ma, Y., An, D., Pang, S., Li, X., 2022. Multiview behavior and neurotransmitter analysis of zebrafish dyskinesia induced by 6PPD and its metabolites. *Sci. Total Environ.* 838 (Pt 2), 156013 <https://doi.org/10.1016/j.scitotenv.2022.156013>. Sep 10. Epub 2022 May 16. PMID: 35588826.
- Jia, S., Sankaran, G., Wang, B., Shang, H., Tan, S.T., Yap, H.M., Shen, J., Gutiérrez, R.A., Fang, W., Liu, M., et al., 2019. Exposure and risk assessment of volatile organic compounds and airborne phthalates in Singapore's Child Care Centers. *Chemosphere* 224, 85–92. <https://doi.org/10.1016/j.chemosphere.2019.02.120>.
- Kaplan, L.A., Nabel, M., Van Cleef-Toedt, K., Proffitt, A.R., Pylpyiw Jr., H.M., 2013. Impact of benzyl butyl phthalate on shoaling behavior in *Fundulus heteroclitus* (mummichog) populations. *Mar. Environ. Res.* 86, 70–75. <https://doi.org/10.1016/j.marenvres.2013.02.014>. May. Epub 2013 Mar 14. PMID: 23541602.
- Karami, A., 2017. Gaps in aquatic toxicological studies of microplastics. *Chemosphere* 184, 841–848. <https://doi.org/10.1016/j.chemosphere.2017.06.048>.
- Karthikeyan, P., Marigoudar, S.R., Mohan, D., Sharma, K.V., Ramana Murthy, M.V., 2021. Prescribing sea water quality criteria for arsenic, cadmium and lead through species sensitivity distribution. *Ecotoxicol. Environ. Saf.* 15 (208), 111612 <https://doi.org/10.1016/j.ecoenv.2020.111612>. Jan. Epub 2020 Nov 14. PMID: 33396132.

- Kole, P.J., Löhr, A.J., Van Belleghem, F., Ragas, A., 2017. Wear and tear of tyres: a stealthy source of microplastics in the environment. *Int. J. Environ. Res. Public Health* 14 (10), 1265.
- Kühn, S., van Franeker, J.A., 2020. Quantitative overview of marine debris ingested by marine megafauna. *Mar. Pollut. Bull.* 151, 110858.
- Kwan, C.S., Takada, H., 2016. Release of additives and monomers from plastic wastes. In: *Hazardous Chemicals Associated With Plastics in the Marine Environment*, pp. 51–70.
- Langston, W.J., Pope, N.D., 1995. Determinants of TBT adsorption and desorption in estuarine sediments. *Mar. Pollut. Bull.* 31 (1–3), 32–43.
- Leslie, H.A., van Velzen, M.J.M., Brandsma, S.H., Vethaak, A.D., Garcia-Vallejo, J.J., Lamoree, M.H., 2022. Discovery and quantification of plastic particle pollution in human blood. *Environ. Int.* 163, 107199 <https://doi.org/10.1016/j.envint.2022.107199>. May, Epub 2022 Mar 24. PMID: 35367073.
- Lau, W.W.Y., Shiran, Y., Bailey, R.M., Cook, E., Stuchtey, M.R., Koskella, J., Velis, C.A., Godfrey, L., Boucher, J., Murphy, M.B., Thompson, R.C., Jankowska, E., Castillo, A. C., Pilditch, T.D., Dixon, B., Koerselman, L., Kosior, E., Favoino, E., Gutberlet, J., Palardy, J.E., 2020. Evaluating scenarios toward zero plastic pollution. *Science* 369 (6510), 1455–1461. <https://doi.org/10.1126/science.aba9475>.
- Law, R.J., Covaci, A., Harrad, S., Herzke, D., Abdallah, M.A.-E., Fernie, K., Toms, L.-M.L., Takigami, H., 2014. Levels and trends of PBDEs and HBCDs in the global environment: status at the end of 2012. *Environ. Int.* 65, 147–158.
- Lemos, M.F.L., 2021. Biomarker studies in stress biology: from the gene to population, from the organism to the application. *Biology* 10 (12), 1340. <https://doi.org/10.3390/biology10121340>.
- Liao, C., Kim, U.-J., Kannan, K., 2020. Occurrence and distribution of organophosphate esters in sediment from northern Chinese coastal waters. *Sci. Total Environ.* 704, 135328.
- Lithner, D., Larsson, Å., Dave, G., 2011. Environmental and health hazard ranking and assessment of plastic polymers based on chemical composition. *Sci. Total Environ.* 409 (18), 3309–3324. <https://doi.org/10.1016/j.scitotenv.2011.04.038>.
- Magnusson, K., Eliaeson, K., Fråne, A., Haikonen, K., Olshammar, M., Stadmark, J., Hultén, J., 2016. Swedish sources and pathways for microplastics to the marine environment. In: *IVL Svenska Miljöinstitutet*.
- Markowicz, F., Szymańska-Pulikowska, A., 2019. Analysis of the possibility of environmental pollution by composted biodegradable and oxo-biodegradable plastics. *Geosciences* 9 (11), 460.
- Meeker, J.D., Sathyanarayana, S., Swan, S.H., 2009. Phthalates and other additives in plastics: human exposure and associated health outcomes. *Philos. Trans. R. Soc. B* 364 (1526), 2097–2113.
- National Center for Biotechnology Information, 2022a. PubChem compound summary for CID 7814, p-phenylenediamine. <https://pubchem.ncbi.nlm.nih.gov/compound/p-Phenylenediamine>.
- National Center for Biotechnology Information, 2022b. PubChem compound summary for CID 11192. In: ZINC Acetate. <https://pubchem.ncbi.nlm.nih.gov/compound/ZINC-acetate#section=FDA-National-Drug-Code-Directory>.
- National Center for Biotechnology Information, 2022c. PubChem compound summary for CID 517045. In: Sodium Acetate. <https://pubchem.ncbi.nlm.nih.gov/compound/Sodium-acetate#section=Therapeutic-Uses>.
- Nielsen, T.D., Hasselbalch, J., Holmberg, K., Stripple, J., 2020. Politics and the plastic crisis: a review throughout the plastic life cycle. *Wiley Interdiscip. Rev.: Energy Environ.* 9 (1), e360.
- Novelli, A.A., Argese, E., Tagliapietra, D., Bettoli, C., Ghirardini, A.V., 2002. Toxicity of tributyltin and triphenyltin to early life-stages of *Paracentrotus lividus* (Echinodermata: Echinoidea). *Environ. Toxicol. Chem.* 21 (4), 859–864. Apr. PMID: 11951962.
- Nyberg, B., Harris, P.T., Kane, I., Maes, T., 2023. Leaving a plastic legacy: current and future scenarios for mismanaged plastic waste in rivers. *Sci. Total Environ.* 869, 161821 <https://doi.org/10.1016/j.scitotenv.2023.161821>.
- Ojeda, T.F.M., Dalmolin, E., Forte, M.M.C., Jacques, R.J.S., Bento, F.M., Camargo, F.A.O., 2009. Abiotic and biotic degradation of oxo-biodegradable polyethylenes. *Polym. Degrad. Stab.* 94 (6), 965–970.
- Parker-Jurd, F., Napper, I.E., Abbott, G.D., Hann, S., Wright, S.L., Thompson, R.C., 2020. Investigating the Sources and Pathways of Synthetic Fibre and Vehicle Tyre Wear Contamination Into the Marine Environment.
- Peng, X., Tang, C., Yu, Y., Tan, J., Huang, Q., Wu, J., Chen, S., Mai, B., 2009. Concentrations, transport, fate, and releases of polybrominated diphenyl ethers in sewage treatment plants in the Pearl River Delta, South China. *Environ. Int.* 35 (2), 303–309. <https://doi.org/10.1016/j.envint.2008.07.021>. Feb. Epub 2008 Sep 5. PMID: 18774173.
- Pereira, L., Mondal, P.K., Alves, M., 2015. Aromatic amines sources, environmental impact and remediation. In: *Pollutants in Buildings, Water and Living Organisms*. Springer, pp. 297–346.
- Pérez-Iglesias, J.M., González, P., Calderón, M.R., Natale, G.S., Almeida, C.A., 2022. Comprehensive evaluation of the toxicity of the flame retardant (decabromodiphenyl ether) in a bioindicator fish (*Gambusia affinis*). *Environ. Sci. Pollut. Res. Int.* 29 (33), 50845–50855. <https://doi.org/10.1007/s11356-022-19462-x>. Jul. Epub 2022 Mar 3. PMID: 35243576.
- Persson, L., Almroth, Carney, Collins, C.D., Cornell, S., de Wit, C., et al., 2022. Outside the safe operating space of the planetary boundary for novel entities. *Environ. Sci. Technol.* <https://doi.org/10.1021/acs.est.1c04158>.
- Prasad, B., 2021. Phthalate pollution: environmental fate and cumulative human exposure index using the multivariate analysis approach. *Environ. Sci. Process Impacts* 23 (3), 389–399.
- Qiu, W., Liu, S., Chen, H., Luo, S., Xiong, Y., Wang, X., Xu, B., Zheng, C., Wang, K.J., 2021. The comparative toxicities of BPA, BPB, BPS, BPF, and BPAF on the reproductive neuroendocrine system of zebrafish embryos and its mechanisms. *J. Hazard. Mater.* 406, 124303 <https://doi.org/10.1016/j.jhazmat.2020.124303>. Mar 15. Epub 2020 Oct 19. PMID: 33121856.
- Rani, M., Meenu, Shanker, U., 2022. Environmental risk assessment of plastics and its additives. In: Shanker, U., Hussain, C.M., Rani, M. (Eds.), *Handbook of Green and Sustainable Nanotechnology: Fundamentals, Developments and Applications*. Springer International Publishing, pp. 1–26. https://doi.org/10.1007/978-3-030-69023-6_33-1.
- Reynolds, E.J., Smith, D.S., Chowdhury, M.J., Hoang, T.C., 2018. Chronic effects of lead exposure on topmelt fish (*Atherinops affinis*): influence of salinity, organism age, and relative sensitivity to other marine species. *Environ. Toxicol. Chem.* 37 (10), 2705–2713. <https://doi.org/10.1002/etc.4241>. Oct. Epub 2018 Aug 31. PMID: 30044002.
- Rocha-Gutierrez, B., Lee, W.-Y., 2013. Investigation of polybrominated diphenyl ethers in wastewater treatment plants along the US and Mexico border: a trans-boundary study. *Water Air Soil Pollut.* 224, 1–13.
- Rochester, J.R., Bolden, A.L., 2015. Bisphenol S and F: a systematic review and comparison of the hormonal activity of bisphenol A substitutes. *Environ. Health Perspect.* 123 (7), 643–650. <https://doi.org/10.1289/ehp.1408989>. Jul. Epub 2015 Mar 16. PMID: 25775505; PMCID: PMC4492270.
- Rochman, C.M., 2015. In: Bergmann, M., Gutow, L., Klages, M. (Eds.), *The Complex Mixture, Fate and Toxicity of Chemicals Associated With Plastic Debris in the Marine Environment* BT - *Marine Anthropogenic Litter*. Springer International Publishing, pp. 117–140. https://doi.org/10.1007/978-3-319-16510-3_5.
- Rodrigues, J.P., Duarte, A.C., Santos-Echeandía, J., Rocha-Santos, T., 2019. Significance of interactions between microplastics and POPs in the marine environment: a critical overview. *TrAC Trends Anal. Chem.* 111 (February), 252–260. <https://doi.org/10.1016/j.trac.2018.11.038>.
- Roessink, I., Belgers, J.D.M., Crum, S.J.H., van den Brink, P.J., Brock, T.C.M., 2005. Comparison of species sensitivity distributions between laboratory and semi-field for the organotin triphenylacetate (TPT-AC). In: *Abstract book; SETAC Europe 15th Annual Meeting*. SETAC Europe, p. 391.
- Rosso, B., Corami, F., Vezzaro, L., Biondi, S., Bravo, B., Barbante, C., Gambaro, A., 2022. Quantification and characterization of additives, plasticizers, and small microplastics (5–100 µm) in highway stormwater runoff. *J. Environ. Manag.* 324, 116348 <https://doi.org/10.1016/j.jenvman.2022.116348>.
- Shah, S.B., 2021. Heavy metals in the marine environment—an overview. In: *Heavy Metals in Scleractinian Corals*, pp. 1–26.
- Sousa, A.C.A., Pastorinho, M.R., Takahashi, S., Tanabe, S., 2014. History on organotin compounds, from snails to humans. *Environ. Chem. Lett.* 12 (1), 117–137.
- Strmac, M., Braunbeck, T., 1999. Effects of triphenyltin acetate on survival, hatching success, and liver ultrastructure of early life stages of zebrafish (*Danio rerio*). *Ecotoxicol. Environ. Saf.* 44 (1), 25–39. <https://doi.org/10.1006/eesa.1999.1781>. Sep. PMID: 10499986.
- Tanaka, K., Takada, H., Yamashita, R., Mizukawa, K., Fukuwaka, M., Watanuki, Y., 2015. Facilitated leaching of additive-derived PBDEs from plastic by seabirds' stomach oil and accumulation in tissues. *Environ. Sci. Technol.* 49 (19), 11799–11807.
- Tian, Z., Zhao, H., Peter, K.T., Gonzalez, M., Wetzel, J., Wu, C., Hu, X., Prat, J., Mudrock, E., Hettinger, R., Cortina, A.E., Ghosh Biswas, B., Crizóstomo Kock, F.V., Soong, R., Jenne, A., Du, B., Hou, F., He, H., Lundeen, R., Kolodziej, E.P., 2021. A ubiquitous tire rubber-derived chemical induces acute mortality in coho salmon. *Science* 371 (6525). <https://doi.org/10.1126/science.abd6951>.
- Tian, Z., Gonzalez, M., Rideout, C.A., Zhao, H.N., Hu, X., Wetzel, J., Mudrock, E., James, C.A., McIntyre, J.K., Kolodziej, E.P., 2022. 6PPD-Quinone: revised toxicity assessment and quantification with a commercial standard. *Environ. Sci. Technol. Lett.* 9 (2), 140–146. <https://doi.org/10.1021/acs.estlett.1c00910>.
- Tornero, A.M.V., Hanke, G., 2018. Marine Chemical Contaminants – Support to Harmonized MSFD Reporting: Substances Considered for MSFD Descriptor 8. JRC Publications Repository. <https://doi.org/10.2760/804610> (July 17).
- Tran, H.T., Lin, C., Bui, X.-T., Nguyen, M.K., Cao, N.D.T., Mukhtar, H., Hoang, H.G., Varjani, S., Ngo, H.H., Nghiem, L.D., 2022. Phthalates in the environment: characteristics, fate and transport, and advanced wastewater treatment technologies. *Bioresour. Technol.* 344, 126249.
- Tun, T.Z., Kunisue, T., Tanabe, S., Prudente, M., Subramanian, A., Sudaryanto, A., Viet, P.H., Nakata, H., 2022. Microplastics in dumping site soils from six Asian countries as a source of plastic additives. *Sci. Total Environ.* 806 (Pt 4), 150912 <https://doi.org/10.1016/j.scitotenv.2021.150912>.
- Turner, A., Filella, M., 2021. Hazardous metal additives in plastics and their environmental impacts. *Environ. Int.* 156, 106622 <https://doi.org/10.1016/j.envint.2021.106622>. Nov. Epub 2021 May 21. PMID: 34030075.
- UNEP, 2021. Drowning in Plastics – Marine Litter and Plastic Waste Vital Graphics. United Nations Environment Programme & Secretariat of the Basel, Rotterdam and Stockholm Conventions, 2023. *Chemicals in Plastics: A Technical Report*.
- US EPA, 2017. *Technical Fact Sheet—Polybrominated Diphenyl Ethers (PBDEs)*.
- van Oers, L., van der Voet, E., Grundmann, V., 2012. Additives in the plastics industry. In: Bilitewski, B., Darbra, R.M., Barceló, D. (Eds.), *Global Risk-based Management of Chemical Additives I: Production, Usage and Environmental Occurrence*. Springer, pp. 133–149. <https://doi.org/10.1007/978-94-9001-112-1>.
- Varshney, S., Gora, A.H., Siriappagounder, P., Kiron, V., Olsvik, P.A., 2022. Toxicological effects of 6PPD and 6PPD quinone in zebrafish larvae. *J. Hazard. Mater.* 424 (Pt C), 127623 <https://doi.org/10.1016/j.jhazmat.2021.127623>. Feb 15. Epub 2021 Oct 30. PMID: 34742612.
- Wagner, S., Schlummer, M., 2020. Legacy additives in a circular economy of plastics: current dilemma, policy analysis, and emerging countermeasures. *Resour. Conserv. Recycl.* 158, 104800 <https://doi.org/10.1016/j.resconrec.2020.104800>.

- Wagner, S., Klöckner, P., Reemtsma, T., 2022. Aging of tire and road wear particles in terrestrial and freshwater environments – a review on processes, testing, analysis and impact. *Chemosphere* 288, 132467. <https://doi.org/10.1016/j.chemosphere.2021.132467>.
- Walsh, G.E., McLaughlan, L.L., Lores, E.M., Louie, M.K., Deans, C.H., 1985. Effects of organotin on growth and survival of two marine diatoms, *Skeletoneina costatum* and *Thalassiosira pseudonata*. *Chemosphere* 14, 383–392.
- Wang, Z., Walker, G.W., Muir, D.C.G., Nagatani-Yoshida, K., 2020. *Environ. Sci. Technol.* 54 (5), 2575–2584. <https://doi.org/10.1021/acs.est.9b06379>.
- Werner, S., Budziak, A., Van, J., Galgani, F., Hanke, G., Maes, T., Matiddi, M., Nilsson, P., Oosterbaan, L., Priestland, E., Veiga, J., Vlachogianni, T., 2016. Harm Caused by Marine Litter, pp. 1–92. <https://doi.org/10.2788/19937>.
- Wibe, A.E., Billing, A., Rosenqvist, G., Jenssen, B.M., 2002. Butyl benzyl phthalate affects shoaling behavior and bottom-dwelling behavior in threespine stickleback. *Environ. Res.* 89 (2), 180–187. <https://doi.org/10.1006/enrs.2002.4360>. Jun. PMID: 12123651.
- Wiesinger, H., Wang, Z., Hellweg, S., 2021. Deep dive into plastic monomers, additives, and processing aids. *Environ. Sci. Technol.* 55 (13), 9339–9351. <https://doi.org/10.1021/acs.est.1c00976>.
- Wrona, M., Nerin, C., 2019. Chapter 7. Risk Assessment of Plastic Packaging for Food Applications, pp. 163–191. <https://doi.org/10.1039/9781788012973-00163>.
- Xiao, F., 2022. An overview of the formation of PFOA and PFOS in drinking-water and wastewater treatment processes. *J. Environ. Eng.* 148 (4), 1822001.
- Xie, Z., Wang, P., Wang, X., Castro-Jiménez, J., Kallenborn, R., Liao, C., Mi, W., Lohmann, R., Vila-Costa, M., Dachs, J., 2022. Organophosphate ester pollution in the oceans. *Nat. Rev. Earth Environ.* 3 (5), 309–322.
- Yogui, G.T., Sericano, J.L., 2009. Polybrominated diphenyl ether flame retardants in the US marine environment: a review. *Environ. Int.* 35 (3), 655–666.
- Yoshioka, E., 1986. Histological studies on the gonad and gametogenesis of the chiton *Acanthopleura japonica* (Lischke) (In Japanese with English abstract). *Venus* 45, 109–115.
- Zeng, L., Li, Y., Sun, Y., Liu, L.-Y., Shen, M., Du, B., 2023. Widespread occurrence and transport of p-phenylenediamines and their quinones in sediments across urban rivers, estuaries, coasts, and deep-sea regions. *Environ. Sci. Technol.* 57 (6), 2393–2403.