2022 Doctoral Thesis

Integrating Potential Environmental Impacts of Plastic Pollution into Life Cycle Assessment of Plastic and Plastic Related Products

(製品ライフサイクルを思考した海洋プラスチック汚染の影響評価手法の開発)

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EXECUTIVE SUMMARY

The present study focused on the integration of plastic litter impacts into plastic and plastic-related product life cycle assessment (LCA). This included both life cycle inventory (LCI) development and life cycle impact assessment (LCIA) methods development. Considering the scientific maturity (which is at the primary stage) of the plastic litter related LCI and LCIA, especially the ecological and human impacts estimation methods for the plastic litter, the scope of the study has been chosen for the floating and suspended microplastics fate to water environments and roadside emissions (i.e. tire-wear particle) to the environmental compartments. Apart from that, the development of the method for potentially toxic element identification in microplastics was also conducted to estimate the potentially toxic element intake of humans with microplastics.

First, the identification of plastic pollution-related impacts was done by rewiring the relevant peer-reviewed literature. Then the LCA perspectives were studied through refereeing to standard literature and the other publications on LCA methods and LCIA models such as ReCePi and LIME. Using the findings of the literature review on plastic pollution and LCA, the development of an overall LCA (LCI and LCIA) framework for the integration of impacts of plastic litter into LCA was done.

Then the development of primary life cycle inventory (LCI) data for tire-wear particle micro plastics in Japan was conducted. For this, data collection of annual different class vehicle transport distances for Japan (secondary sources) and Meta data through literature review on tire-wear particle emissions for unit distance traveling of different classes of vehicles was conducted. Then using the Meta-analysis of data and identifying the tire-wear emission factors for different classes of vehicles (there were six classes of vehicles considered in the study). Finally, the tire-wear particle emission for Japan was estimated. The average value obtained was 33,796 metric tons (MT)/year. The obtained value was compared with the available literature and found it is within the range of UNEP, 2018 information (21,150-35,109).

Then the two main different types of microplastics were considered for the LCIA model developments. Here the fate factor development was the main focus since the exposure and effects require further information such as human toxicity of microplastics.

The microplastic litter LCIA, the first section was focused on floating and suspended microplastics larger than 300 μm (suing river floating and suspended microplastic measurement data). First the microplastic sampling method standardization, limitations identification, and development methods to overcome the limitations. Relationships between microplastic surface area and the mass for different categories of microplastics were conducted. The correlations were found to be good indicating R^2 values larger than 80 in most of the microplastic categories. Then the development of a GIS-based model to estimate the riverine microplastic flow at 10 km grid level and prefectural level. Development of methods to generate microplastic data suitable for the GIS model input (such as particle concentration conversion into mass concentration). Then the estimate of the emissions factors to water environment of microplastics at 10 km grid level and prefectural levels was conducted. The per capita emissions were also calculated. The prefectures such as Tokyo (37 MT/year), and Kanagawa (23 MT/year) with high population densities indicated a higher degree of microplastic releases.

Then a wastewater treatment plant model (2114 plants) has been developed for the microplastic fate estimations since down the drain microplastics such as synthetic textile fiber and roadside microplastic emissions such as tire-wear abrasion particles. The reason for this in Japan is a considerable fraction of wastewater passes through the wastewater treatment plants and street runoff in some cases passes through the wastewater treatment plants due to the presence of combined sewers. The model was developed for Japan (10 km scale and prefectural level), including the treatment level, treatment capacity, location, etc. information. Application of the wastewater treatment plant model and developed inventory to estimate the microplastic fate at the grid level (10 km) and prefectural level. Even though the high emission of tier-wear abrasion particles from the major cities such as Tokyo, Yokohama, and Osaka, due to the presence of combined sewers, a considerable fraction of the microplastics passes through the wastewater treatment plants. The total airborne (as PM 10) was estimated as 676 MT/year. The total leakage of tier-wear particles into the water was estimated for two scenarios. The long-term scenario which considers the complete wash of the street provided 27,300 MT/year emission to the water environment. The short-term runoff scenario (which considers 21% particle goes with street runoff) that 5,678 MT/year fate in water. The annual release of

synthetic microfiber from domestic textile washing is 17,612 MT/year and the fate going to water environments is 5246 MT/year. The largest quantity transfers into wastewater treatment plant sludge (12,348 MT/year). This provides a countrywide fate factor of 0.30 to water, 0.70 to wastewater treatment plant sludge. However, 79.8% of the sludge in Japan is incinerated. Hence the fate in sludge which is potentially re-contaminates the environment is 2,494 MT/year. At the prefectural level, Tokyo has the lowest fate factor to the water environment (0.11) due to higher centralized coverage of wastewater and the higher degree of treatment. In the prefectures with comparatively lower coverage of sewerage the fate factor to water increases over 0.50. However, interestingly, the Aichi prefecture is the highest annual emissions to the water environment (fate factor to water, 0.33), due to the combination of medium fate factor and comparatively higher emission at the source due to higher population.

The potentially toxic element (PTE) analysis in microplastic method development using x-ray fluorescence spectroscopy (XRF) was conducted. The detection of several toxic heavy metals and other elements was possible.

The LCA integration of impacts of plastic pollution is presently developing. The LCI and LCIA development on a national and global scale will make this possible in near future. The scientific evidence on estimating toxic effects on ecosystems and humans needs to be addressed. The findings of the present study provide recommendations such as, to address the microplastic pollution such as tire-were particles, the counter measures such as public transportation with non-rubber tire based (i.e. trains, tram-cars) and increasing the combined sewer coverage in hotspot areas (such as cities where higher breaking and sudden accelerations of vehicles happen).

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Contents

INTRODUCTION

1.1 Background (Plastic litter and importance of assessing the impact of plastic litter)

Global Situation

Plastic litter is accumulating in different environmental compartments and is associated with impacts on ecology and creates a potential human health threat. Between 1997 and 2015 the number of species that are known to have become entangled in or ingested marine anthropogenic litter doubled from 267 to 557 species. In addition, such overview estimates likely underrepresent the prevalence of entanglement and ingestion due to underreporting in the scientific literature. Whilst entanglement and ingestion impact mechanisms operate at the level of individual organisms, effects may scale up to the level of species populations and ecosystems (Browne et al., 2015). Consequently, the provision of marine ecosystem services may be compromised (Beaumont et al., 2019). Furthermore, even in the absence of biotic damage, ecosystem services, particularly cultural services, can be affected due to the unsightliness of anthropogenic litter that distracts from natural beauty (Rangel-Buitrago et al., 2018). For example, Jang et al. (2014) observed reduced ecotourism associated with increased coastal litter pollution. Additionally, the potential for marine litter, specifically plastics, to end up in the human food supply may have consequences for human health (Wright and Kelly, 2017).

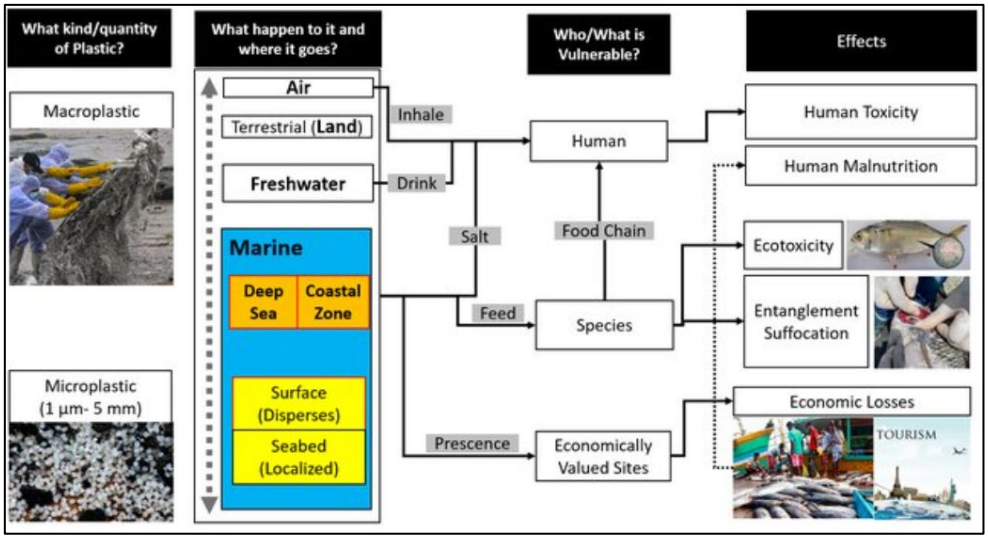


Fig. 1.1 Simplified impact pathways of plastic litter (Abeynayaka and Itsubo, 2019, Woods et al., 2021)

Globally, more than 98% of micro-litter in aquatic environments are plastic (Tekman et al., 2019). Plastic originates from the technosphere, i.e. the human modification of the environment, and its production continues to increase at a rate of approximately 4% annually (Plastics Europe, 2017), with plastics being used widely throughout a large range of product value chains (Andrady and Neal, 2009). Such an increase in plastic use is, in part, due to its versatility as a material, e.g. lightweight, mouldability, and chemical- and light-resistance (Andrady and Neal, 2009), and the importance of such material properties for some applications e.g. in the health sector. Efforts to seek substitutive materials for fossil-based plastic have been recent and, in most cases, still lack competitiveness at a market level (Lettner et al., 2017). Moreover, many innovative biobased polymers do not necessarily show significant environmental improvements as compared to fossil-based polymers in many planetary boundaries (Escobar et al., 2018, Ita-Nagy et al., 2020). The current plastic lock-in for many economic sectors, coupled with the potential for inappropriate disposal of plastic waste, which can occur both before or after use, ultimately leads to plastic litter in the environment and environmental damage (Jambeck et al., 2015, Jambeck et al., 2018).

Attempts to reduce the generation of potentially environmentally damaging marine plastic litter have started recently, including in the EU, which has new rules on single-use plastics by 2021 (European Commission, 2019), and Canada (Environment and Climate Change Canada, 2019). The EU rules on single-use plastics have already started to be implemented by, for example, Portugal (DRE, 2020). The rules exemplify an application of both upstream interventions that reduce supply, e.g. a ban on single-use products made of plastic for which alternatives exist on the market, and downstream interventions, such as a 90% collection target for plastic bottles by 2029, and extended producer responsibility e.g. covering costs of clean-up (European Commission, 2019).

Poznan workshop 30-31 August 2019	Environmental relevance	Scientific validity & maturity	Potential for consensus	Stakeholder needs	Applicability	Overall priority	Comments
Integration of HH						****	
Integration of EQ						****	
Integration of natural resources and ecosystem service endpoints						****	
Normalisation and weighting		**		***	***	****	
Nutritional impacts	***	**	?	*	?	***	For specific food LCA
Ocean acidification	**	**	?	*	**	***	
POCP effects on ecosystem quality	**	**	**	*	**	***	
Biotic resources	**	*	?	**	?	**	Start exploring/ include if consensus reached
Noise impacts	**	*	*	*	?	**	
Impacts caused by plastic pollution/litter	?	*	-	***	?	*	check results of Marilca
Invasive species	***	*	?	*	-	*	Explore existing work
Light pollution	*	*					
Antibiotic resistance	***	?					
Hormones & Endocrine	**						
Heat effects in water	*						
Urban heat island effects	*	*	?	**	?		
Non ionising radiation	-	?	?	**			

Life Cycle Initiative UN environment programme

Fig. 1.2 The status of plastic pollution according to UNEP-LCI, 2019

Figure 1.2. Indicates the situation of plastic litter in LCA. The environmental relevance is to be confirmed in a quantifiable way. Scientific validity and maturity as lower to quantify the environmental impacts. However, there are high stakeholder needs and demands for the integration into LCA. In 2019, the applicability was also to be confirmed stage.

IMPACT INDICATORS	Glass bottle	Aluminium can	PET bottle (0.5 l)	PET bottle (2 l)
Abiotic depletion potential	Highest Impact	Medium Impact	Medium Impact	Least Impact
Acidification	Highest Impact	Medium Impact	Medium Impact	Least Impact
Eutrophication	Medium Impact	Least Impact	Highest Impact	Medium Impact
Freshwater aquatic ecotoxicity potential	Medium Impact	Medium Impact	Highest Impact	Least Impact
Climate change	Highest Impact	Medium Impact	Medium Impact	Least Impact
Human toxicity potential	Medium Impact	Highest Impact	Medium Impact	Least Impact
Marine aquatic ecotoxicity potential	Medium Impact	Highest Impact	Medium Impact	Least Impact
Ozone layer depletion potential	Highest Impact	Least Impact	Medium Impact	Medium Impact
Photochemical oxidant creation potential	Highest Impact	Medium Impact	Medium Impact	Least Impact
Terrestrial ecotoxicity potential	Highest Impact	Least Impact	Medium Impact	Medium Impact
Primary energy demand	Highest Impact	Medium Impact	Medium Impact	Least Impact

Figure 1.3 Present-day LCA comparing PET bottles and alternatives. The PET indicated the least impact in most of the cases compared to the alternatives (UNEP-LCI, 2020).

Situation in Japan

Japan is the second in the world (after the USA) in terms of plastic packaging waste per capita. In 2015, plastic packaging waste accounted for 47% of the plastic waste generated globally, with half of that appearing to come from Asia. While China remains the largest worldwide generator of plastic packaging waste, the USA is the largest generator of plastic packaging waste on a per-capita basis, followed by Japan and the EU. According to the Plastic Waste Management Institute (PWMI), resin production and domestic plastics products consumption was 10.7 million and 10.3 million tons respectively in 2018. The amount of domestic waste discharged in Japan was 42.9 million tons/year, which equates to 920g of discharged waste per person. On examining the composition of garbage, FY2018 of a survey conducted by the Ministry of the Environment found that kitchen waste and paper made up 32% and 30%, respectively, followed by plastics at 12% and wood/bamboo/grass, metal, glass and fiber at 7-4% in terms of percentage wet weight. The average person in Japan uses up to 450 plastic bags per year. That's a national total of some 30 billion plastic bags, every year. The Tokyo Metropolitan Government announced its goal of getting all retailers to charge a small fee for plastic shopping bags by 2020, whereas places like Kyoto have already implemented legislation requiring large retailers to charge for plastic bags. Tokyo's Suginami ward established the first ordinance in Japan promoting a charge on plastic bags. Within 7 years, over 30% of consumers in the area refrained from using plastic bags and that percentage continues to grow, suggesting that the hand of government can make a major impact.

The average person in Japan buys 183 PET bottles per year. Put another way, 740 PET bottles are bought nationwide every second. As of fiscal 2017, the recycling rate for PET bottles was 84.8%, among the highest in the world. The flow chart analysis of PET bottles compiled by the council of PET bottle recycling shows that around 62% of PET bottles are collected by municipalities. 38% are collected by the private sector voluntarily. From municipalities, around 65% of collected bottles are recycled through designated body routes. 35% are recycled through independent routes. In late years over 95% of municipalities have sorted collection systems of PET bottles, which is much higher than the other countries. This has been achieved by the combination of the voluntary collection

whose rate is between 30% and 40% and municipal collection. The official "recycling rate" of end-of-life plastics in Japan is 84%. However, it's important to note that this figure includes incineration of plastic as a form of energy, so it may be a little misleading… Over half of the plastic that is "recycled" is incinerated. Here's how it works. Plastic recycling is generally divided into 3 categories, under current legislation: Thermal recycling: burning plastic for energy (56%). Material recycling: Reusing plastic (23%). Chemical recycling: Using raw materials for industrial purposes (4%). So, not all of the plastic we meticulously divide and put into the rubbish bin is reborn into new materials. Forbes Magazine claims that as much as 70% of the plastic collected for recycling in Japan is incinerated. Companies like Adidas, Lush, and Patagonia are also taking the lead, showing that the private sector will not stand idle as we witness the destruction of the environment through irresponsible consumption habits.

As exports of plastic waste decline, reports suggest that domestic facilities are struggling to cope with the spiked demand. Recent reports suggest that the Ministry of Environment will ask municipalities to accept and dispose of industrial plastic waste as an emergency measure. Some companies are quickly making strategic investments into plastic recycling facilities to meet this rising demand. For example, Daiei Kankyo Holdings, a recycling company based in Kobe, is set to open a 1.5 billion yen ($13.8 million) plant in Osaka in 2020. In 2017, Japan generated 9 million tons of plastic waste. About 10% of that was shipped to China (70% of Japan's total exports of plastic waste). That same year (2017), China banned plastic waste imports to reduce pollution from the recycling process. This has left the government in a major pickle. As a result, Japan has had to shift its plastic waste export strategy, by increasing exports to countries such as Indonesia and Vietnam and by increasing plastic recycling capacity in Japan as alternatives. However, because there is no efficient waste treatment facility readily available in these countries, this waste is mostly unrecyclable. Also, underpaid workers do their best to salvage the small amounts of still-use plastic to create cheap and "downcycled" products instead.

In 2018, Japan's government unveiled a proposal to reduce the country's 9.4 million tonnes of plastic waste a year by 25% by 2030. It is also looking to make reusability and recyclability mandatory for all plastic containers and packaging by 2025, to hit a recycling

rate for these items of 40% by 2030, and a "100% effective utilization of used plastics by 2035." Some steps have already been made, with retailers now banned from giving out free plastic shopping bags unless they are composed of at least 15% biomass The government has also decided to make it mandatory for retail shops to charge for plastic shopping bags in a bid to reduce waste, although this has yet to be enforced at the national level. To the dismay of many environmentalists, however, both Japan and the USA declined to sign the G7 Pact to reduce the use of single-use plastics and prevent plastic pollution in 2018.

There's also a growing awareness of the plastics problem on the consumer side, with a rising number of grassroots movements (e.g. 530 weeks), social ventures using their voice to speak up about the issue. So, there is some reason for hope. But, like in many other countries, the question remains as to how to create real, system-wide change around plastic production and consumption (an integral part of UN Sustainable Development Goal 12). The future is looking bleak; by 2050, there are predicted to be more plastics in the ocean than fish, and there is mounting evidence that plastics pose serious health risks to humans on a global scale. In response, many countries are moving towards outright bans of single-use plastics around the world.

Sustainable development goals (SDGs) and life cycle assessment (LCA) perspectives of plastic litter

Sustainable development goals (SDGs) 11,1, 12, and 14 "Conserve and sustainably use the oceans, seas and marine resources for sustainable development" further motivates the requirement for actions to stem the marine litter problem. For example, goal 14.1 targets prevention and a significant reduction in marine pollution of all kinds by 2025, including marine debris and nutrient pollution arising from land-based activities (e.g. van Puijenbroek et al. (2019)). For further discussion of SDGs concerning plastic pollution, see, for example, Plastic Soup Foundation (2018). Whilst it is becoming clear that marine plastic litter is a mainstream environmental problem, current strategies to reduce marine plastic litter may only reduce the rate of plastic litter accumulation in the ocean (Borrelle et al., 2020, Lau et al., 2020). Additionally, such strategies potentially result in trade-offs with other environmental problems, and the magnitude of the impact of marine plastic litter on humans and ecosystems needs to be compared to other environmental impacts.

Life Cycle Assessment (LCA) is a tool for supporting environmental decision making that is specifically designed to allow for the identification of these potential trade-offs between product alternatives e.g. a single-use plastic cup versus a paper one, looking at a broad set of impacts on human health and ecosystems such as climate change, human and eco-toxicity, eutrophication and acidification. However, whilst LCA is widely used in support of decision making, approaches for considering several existing marine impacts in LCA methods remain generally lacking (Woods et al., 2016, Ziegler et al., 2016). This absence of assessment methods has become particularly apparent for marine plastic litter, e.g. Schweitzer et al. (2018), and is recognized by the LCA research community as an area in need of urgent developments (Sonnemann and Valdivia, 2017). As such, whilst trade-offs between different plastic reduction options can already be assessed using LCA, an impact assessment approach for indicating the potential impact of residual plastic litter is still lacking. However, research on the sources, fate, and consequences of plastic litter is still maturing. This presents both challenges and opportunities for extending life cycle impact assessment (LCIA) methods to account for marine littering.

1.2 Objectives and Scope of the Study

The overall objective of this study is to initiate a method to integrate plastic litter into LCA. To achieve the overall objective following objectives were set considering the methodological process and the maturity of the scientific knowledge at present. Considering the limitations such as the present scientific maturity of the plastic pollution quantification and effect estimation, the study focused on the following objectives and scope.

Chapter 2:

- Develop a consistent and comprehensive framework for modeling plastic litter impact pathways in life cycle impact assessment (LCIA) models to ensure the development of characterization factor models, which allows for a more holistic picture of plastic impacts in LCIA methods.

Chapter 3:

- Life Cycle Inventory data development for the tire wear particle emissions in Japan
- Compare with the available data in the literature to verify the data.

Chapter 4:

- Develop a wastewater treatment plant-based model for Japan to estimate the down the drain microplastics (such as textile washing microfiber) and road runoff microplastics (such as tire wear particles) fate estimation.
- Using the developed model, estimation of microplastic emissions into waste, sludge, and other compartments at the prefectural level.

Chapter 5:

- Development of sampling and analytical methods for floating and suspended microplastics in japan.
- Improve the data quality to match estimation model input
- Development and GIS-based model for Japan to estimate the microplastic emissions to water environments at prefectural level.
- Find the microplastic fate into the water at prefectural levels.

Chapter 6:

- Development of potentially toxic metal detection method for microplastics using x-ray florescence spectroscopy

Chapter 7:

- Conclusions and future recommendations for microplastic and LCA work.

1.3 Chapters and Flow of the Study

Figure 1.4 illustrates the chapters, chapter contents, and the flow of the study.

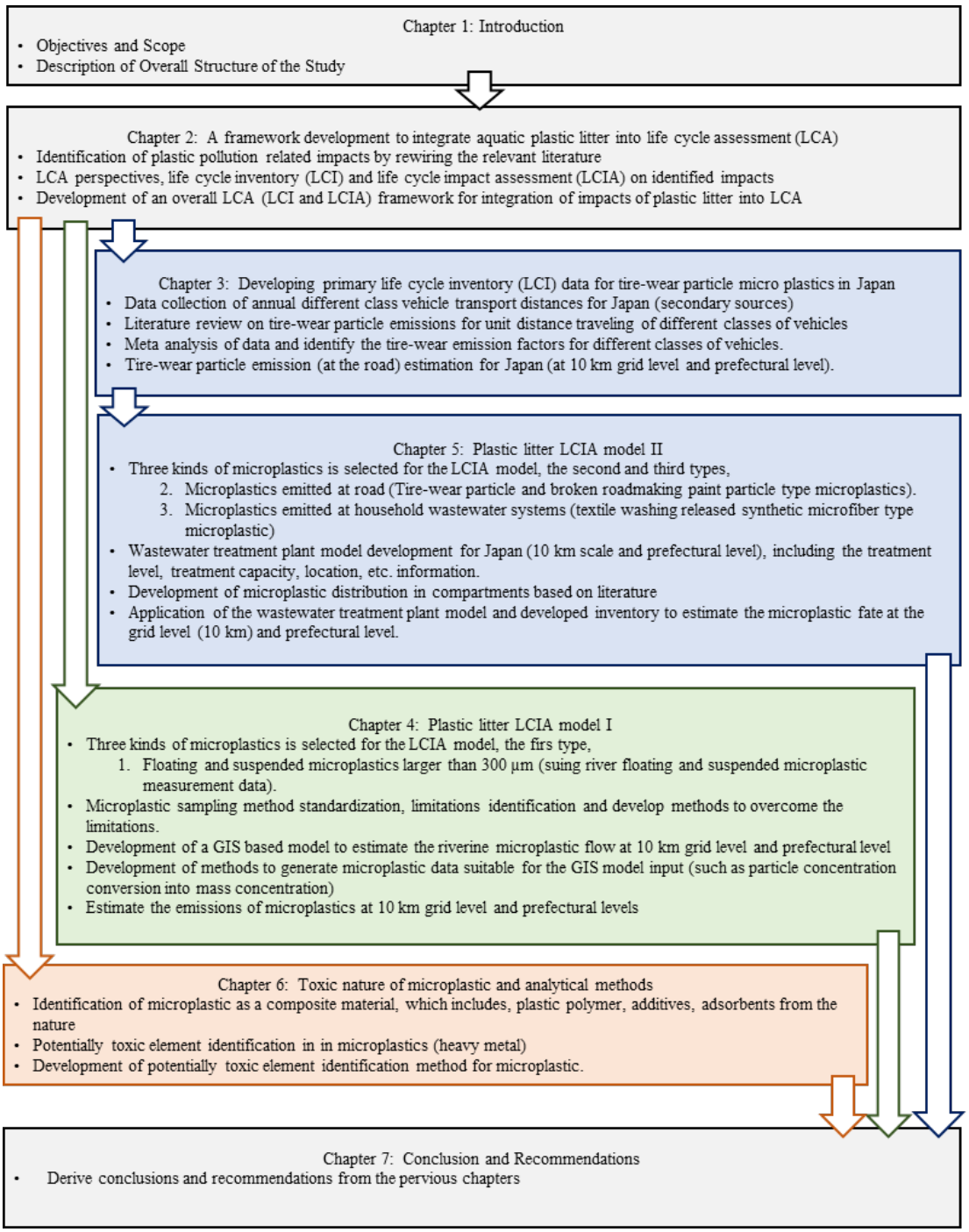


Fig. 1.4 Chapters and the flow of the study.

CHAPTER 2. FRAMEWORK TO INTEGRATE AQUATIC PLASTIC LITTER INTO LCA

2.1 Introduction and Literature Review

The global production of plastics has increased by about 9% per year since 1950. The revenue of the plastic industry is over 800 billion USD in 2018 (GreenView Research (2019). The issue of plastics contaminating freshwater resources and oceans is widely discussed. Research focusing on the impacts associated with the exposure of organisms to aquatic plastics has been ongoing for years. However, studies linking the processes in the plastic value chain to plastics being released to the oceans are only starting to emerge (Ryberg et al., 2018). Lack of a solid framework and having plenty of knowledge gaps limit the incorporation of ecosystem damage due to plastics. There is substantial work to be done to incorporate the damage due to aquatic plastic pollutants into product life cycle assessments (LCAs).

International Standard Organization (ISO), describes the LCA methodology as illustrated in Fig. 1 (a). Efforts to establish life cycle inventory (LCI) for aquatic plastics are being undertaken. Approaches based on polymers (Delphine & Bernd, 2019), selected products (Kočí et al., 2019), and sectors (Ryberg et al., 2018) are found in the literature. Life cycle impact assessment (LCIA) supports the interpretation of LCA studies by translating emissions and resource extractions into a limited number of environmental impact scores (Hauschild, 2017). This is done using characterization factors (CF), which indicate the environmental impact per unit of the stressor.

A substantial number of studies have been carried out regarding the plastics released into environments. There are several approaches to develop inventories of such kinds of plastic releases to environments. These include plastic polymer-based inventory (Delphine & Bernd, 2019), product-based inventory (Ryberg et al., 2018), origin-based inventory (Hauschild, 2017).

Impacts of aquatic plastics such as ecotoxicity (Ostle et al., 2019; Alexiadou et al., 2019; Lavers et al., 2019), entanglement (Ostle et al., 2019, Jepsen et al., 2019) were also being discussed in the literature. However, plastic-related product LCA studies have not incorporated aquatic plastics into the impact analysis phase. The lack of information at various steps of LCA limits such kinds of incorporation. To establish both LCI and

LCIA for aquatic plastics, it is important to have a framework to understand the entire process and the areas to be improved. Hence the objective of this research paper is to develop and discuss a framework to incorporate aquatic plastic into product lifecycle assessment.

In the LCIA stage of an LCA of a product or service, an inventory of environmental interventions, i.e. a list of resources used and emissions produced in association with the life cycle of the product or service, is converted into indicators of environmental impact (Verones et al., 2017). This conversion is termed impact characterization and is conducted using characterization factors (CFs). CFs are resource- or emission- substance-specific and link to specific environmental problems (impact categories). CFs are constructed through impact pathway modeling that links e.g. a mass of emission (kg) to a metric of damage to an Area of Protection (AoP) and as such include consideration of e.g. the dispersion and lifetime of a substance in the environment and the potential for exposure and effects on sensitive receptors e.g. species that may suffer deleterious effects.

This study describes plastic litter impact pathways leading to damage to different Areas of Protection (AoPs), and, more specifically, identifies the predominant impact pathways from plastic leakage to environmental damage. We first discuss key considerations related to plastic litter impact pathways before structuring these within a framework for the modeling of relevant pathways within LCIA. Our approach to identifying the main impact pathways is rooted in, primarily, a qualitative assessment of which impact pathways contribute most to 'leaked' plastic waste. Where possible, we discuss the potential contribution of different impact pathways to overall damage on each of the apps recommended by the UNEP-SETAC life cycle initiative (Verones et al., 2017): ecosystem quality, human health, natural heritage, cultural heritage, socio-economic assets, and ecosystem services. Consideration of potential damage on the natural resources AoP is mostly covered by the assessment of upstream plastic manufacturing processes (Schulze et al., 2020) and is beyond the scope of the present framework focusing on marine litter.

Within the framework, we comment on the strength of scientific evidence supporting different stages of plastic litter impact pathways. In addition, we identify

initial data requirements for the proposed modeling steps in the framework and relevant required levels of spatial differentiation and of litter/waste categorization in terms of e.g. fragment size and material type.

2.2 Methods

2.2.1. Life Cycle Assessment (LCA)

LCA framework is recommended by the ISO14040 series methodology. Figure 2.1. indicates the basic steps in ISO. Intending to incorporate aquatic plastic into plastic product LCA, this study focused on the ISO LCA methods in general.

2.2.2. Life Cycle Inventory (LCI) for Plastic

Plastic-related Life Cycle Inventory (LCI) development is discussed using the previous studies. A literature survey was conducted according to Scopus keyword searches. Various methods found in the literature were analyzed and summarized. A comparison of methods was conducted to find appropriate methods to formulate inventories.

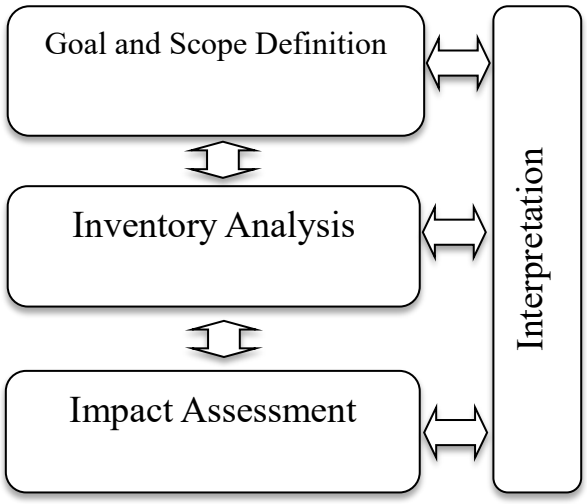


Fig. 2.1: Life cycle assessment framework as per ISO 14040 and 14044, 2006 (ISO, 2006)

2.2.3. Life Cycle Impact Assessment (LCIA) for Plastic

A review of the existing literature within and outside the field of LCIA was carried out to select environmental models and databases per midpoint and endpoint impact categories. Moreover, the selected models that we're able to quantify damage pathways

relevant for the mid to endpoints. Four LICA models are assessed and integrated with the developments of plastic-related impact publications to develop the impact frameworks for the aquatic plastic products. The development framework incorporating both LCI and LCIA phases is indicated in Fig. 2. 2. The development of Fig. 2. 2 was conducted after analyzing the LCIA development models available and the plastic-related literature.

2.2.4. Development of a Framework to Integrate Plastic Litter into LCA

The generic formula to relate interventions of the plastic product (IP) to the damage indicator (DI) can be expressed in equation 1 below.

- Damage Indicator (DI) = IP *CF
- CF= Fate Factor* Exposure Factor *Effect Factor
- FF= Fate Factor (Distribution and longevity)
- EF= Exposure Factor (Interaction with sensitive receptors)
- EFF= Effect Factor (Consequence of interaction on sensitive receptor and contribution to Areas of Protection)

The development of the frameworks was initiated by considering the damaged sectors due to aquatic plastic pollution. Literature published in indexed journals was referred and the damage sectors were selected. Receptors were linked to the environmental compartments through exposure pathways.

2.3 Results and Discussion

2.3.1. Plastic LCI

There are several approaches found in the literature as described previously. As the objectives of LCA vary the orientation of inventories and approaches to develop the inventories can be different. Moreover, still, a clear understanding of the impact model could be one reason for different approaches to developing LCI. However, considering the future LCIA models data input sources, the geo-data-based approaches.

2.3.2. Environmental Sampling

Several studies have focused on environmental sampling from terrestrial, freshwater, marine environments. Environmental sampling encounters practical

difficulties and application in LCA is limited due to the plastic size in the environment, the incompleteness of the available analytical methods, the natural degradation and transportation mechanisms in the different environmental compartments, etc.

2.3.3. Environmental Compartments

Plastic pollutants are present in terrestrial, water, and air. Some compartments shall be divided into sub-compartments. For example, freshwater bodies consist of sediment and water. Fate analyses shall be considered the inter-and intra- transportation of plastics. This made the difficulty of selecting only a single compartment (in this case water). Hence the studies shall focus on all the compartments during the framework stage. Hence the spatial data formats need to be considered.

2.3.4. Global-regionalized Impacts.

Especially the aquatic phase of plastic pollution is mainly associated with the marine environment. Considering the longevity, the plastics circulate in the compartment. Hence oceanic models shall express concentration and time. Moreover, data must include spatial properties. Currently, the available formats may not support the commonly used spreadsheets formats, etc. Hence the spatial data formats need to be considered.

2.3.5. Distribution and longevity

There are two types of distributions, distribution within the environmental compartment and distribution across the environmental compartments. This consideration is much more important for plastic which takes longer periods to disappear from the environment. Moreover, plastics have several different density groups, especially both less than water and higher than water, their behavior in the water environment is quite complex. Distribution through the air, water, soil, the associated matter is reported in the literature. Movements between compartments can be natural or human intervention.

2.3.6. Damage Sectors

Table 2.1 indicates the damage sectors discussed in widely used LCIA models. Human health, socio-economic assessments, ecosystem quality, biodiversity, cultural heritage, human heritage, and primary production are the damage sectors focused on the four models. In the context of plastic pollution human health, ecosystem quality (or biodiversity), socio-economic assets (or social assets), primary production, natural and cultural heritage can be affected.

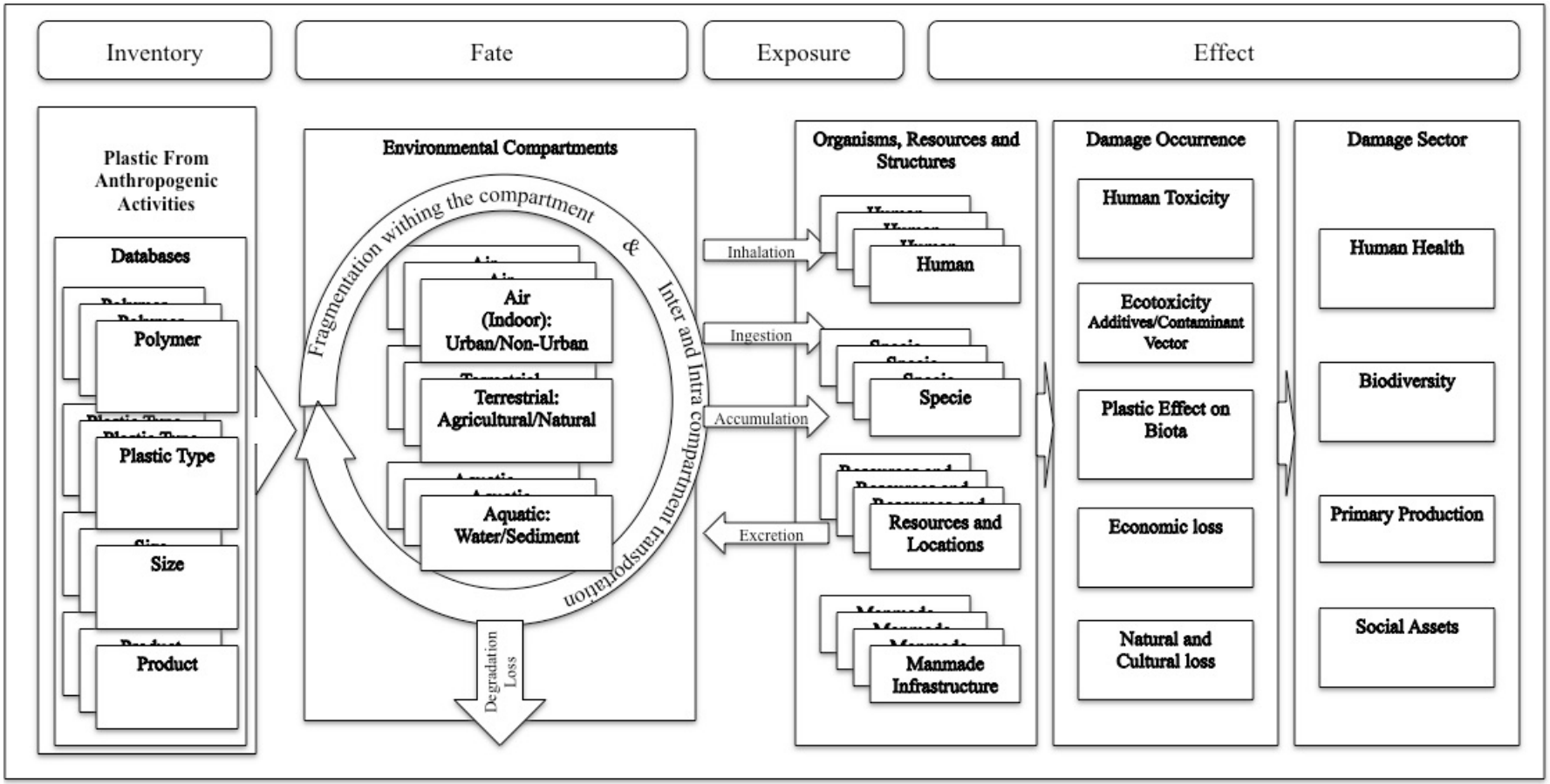


Fig. 2.2: LCA framework for aquatic plastic (Modified from the LCIA models: LIME3, ReCiPe, Impact World+, and IC-IMPACT). (Modified from Abeynayaka and Itsubo, 2019)

Table 2.1: Damage sectors in widely used LCIA models (Bulle et al., 2019; Huijbregts et al., 2017; Verones et al., 2017; Inaba & Itsubo, 20)

LCIA Model	Damage Sectors
LIME3	Human health, Biodiversity Social assets Primary production
ReCiPe	Human health, Ecosystem quality Socio-economic assets Natural heritage Cultural heritage
Impact World+	Human Health, Ecosystem quality Resources & ecosystem services
LC-IMPACT	Human Health, Ecosystem quality Resources

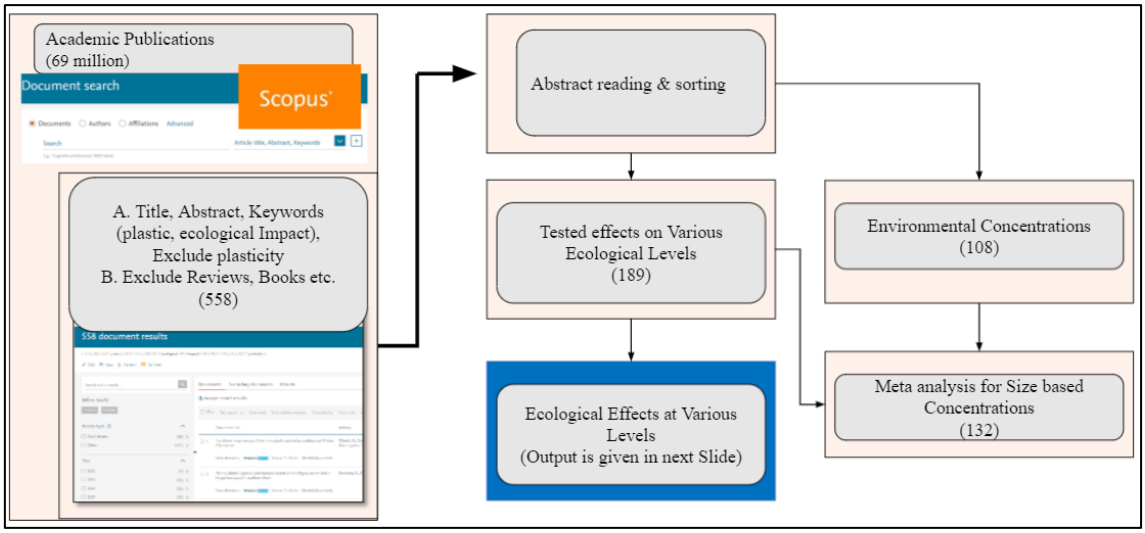


Fig. 2.3: Systematic literature review to identify the impacts of plastic litter

The Meta-analysis of the reported literature dictates, the major two regions of plastic-related effects.

A. Effects due to microplastics (10 nm -1 cm) reported. Often related to oral exposure such as ingestion and associated toxicity.

B. Entanglement-related ecological effects are reported with plastics ranging from cm-100 m range.

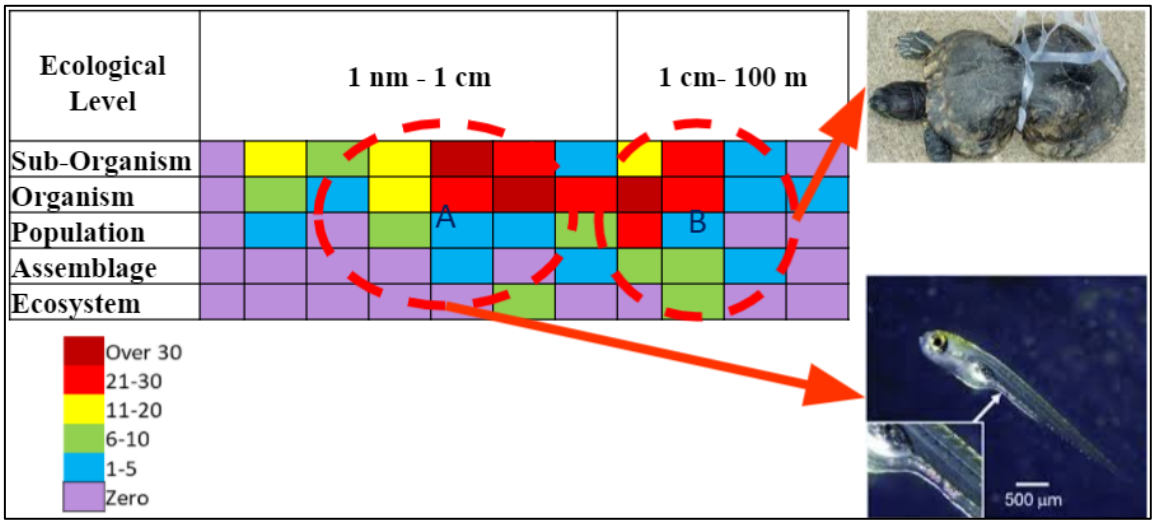


Fig. 2.4: The two major effects associated with plastic litter.

2.3.7. Fate Analysis and Microplastic as a Vector of Toxic pollutants

In LCA, fate analyses and exposure pathways are two significant steps to assess the ecological risk or human toxicity. Plastic product life cycles are globally associated. Consequently, global models are useful tools to evaluate the LCA of products. Research groups are focusing on the potential environmental and human health impacts of microplastics in the aquatic environment. Aquatic ecotoxicity is a midpoint impact indicator associated with microplastics in the aquatic environment. USEPA practice ecological risk characterization as the final phase of ecological risk assessment. Toxic pollutant concertations are an essential component of ecotoxicity assessment. Ecotoxicity is associated with plastics due to plastic particles and adsorbed toxic compounds. Persistent bioaccumulative and toxic substances (PBTs) are hydrophobic. Hence, tend to attach to the plastic phase in an aqua-plastic mixture. Therefore, risk quotient calculations should account for this consideration.

Moreover, global marine toxicity studies are often considered rough globalized toxic compound concentrations due to data unavailability. However, marine plastics concentrations show temporal and spatial variations. Hence, a mechanism to provide concentration variations would enhance the meaningfulness of the ecological risk descriptions.

Previous researchers have developed models for terrestrial water circulation and implemented them on LCA studies. Integration of terrestrial water circulation and oceanic

currents associated models would provide a useful aid to LCA studies on impact evaluations. A grid-cell-based global hydrological model providing terrestrial pollution loads into oceans and oceanic simulation models would provide a global overview of temporal and spatial plastic concentration variations.

A combination of such kind of microplastic concentrations data with PTBs concentrations will provide the exposure potential. Ingestion, inhalation-related information (specie population densities) would provide exposure to both plastic and PTBs. These kinds of observations could enhance the meaningfulness of impact studies. However, the incorporation of fate modeling must be done to integrate ecotoxicity into plastic product LCA.

LCIA Models

- The selection criteria for the environmental models:
- The models should refer to the global scale since the aquatic plastic-associated impacts are global.
- The models should be able to reflect the current state of the art in science and predict the future.
- The models should maintain consistency between the modeling of different impact categories, particularly relevant for toxicity.
- In the case of multiple suitable global models, the preference goes to the models that can be easily integrated with globally accepted LCIA models (such as LIME3, ReCePi, Impact World+, LC-IMPACT).

Review of the existing literature within and outside the field of LCIA to select a preferred combination of environmental models and databases per midpoint impact category. We also selected models that we're able to quantify damage pathways relevant to the mid-to-endpoint factors. For several impact categories, notably fine particulate matter formation, photochemical ozone formation, land use, and water use, there is a fast increasing number of global models published in the literature.

2.4 Conclusions and Recommendations

Aquatic plastic is a global issue. Available LCIA models need to be improved to assess the impact of aquatic plastic pollution. Hence global collaboration and sharing platforms are necessary. Plastic LCI needs to be improved and globally available. Fate modeling needs to be materialized considering the environmental conditions. The impacts are mainly in two sections. A. Macroplastic-related entanglement and B. Microplastic-related toxicity (ingestion). The impacts can be Human, Ecological (biodiversity), primary production, and social assets (such as a detreated view).

CHAPTER 3. LIFE CYCLE INVENTORY (LCI) FOR SELECTED MICROPLASTIC LITTER IN JAPAN

3.1 Introduction and Literature review

Identifying Processes for the Life Cycle Inventory (LCI) Model

This first step of the LCI details the coarse initial system diagram made under the scope item System boundaries and draws upon the related completeness requirements. The outcome of the step is a detailed depiction of the foreground system, i.e., all the processes it is composed of and their links, and the processes of the background system 'neighboring' the foreground system, i.e., where links to LCI database processes will be established.

Level 0: Identification of the unit process/processes.
(In a study where a window is the reference flow, the level 0 process is the assembly of the window.)

Level 1: Identification of the processes required to deliver flows that will be physically embodied in the reference flow
In the window example, examples of level 1 processes are the production of glass and the window frame.

Level 2: The processes required to deliver flows that perform a supporting function to the level 0 process should then be identified. In the window example, examples of level 2 processes are the supply of electricity used in the assembly of the window or the transportation needed to deliver the flows of the level 1 processes to the level 0 process.

Level 3: The processes required to deliver services to the level 0 processes should then be identified. In the window example,
examples of level 3 processes are administration and marketing.

Level 4: The processes required to produce and maintain the infrastructure that enables the level 0 process should then be identified. In the window example, examples of level 4 processes are production and maintenance (oiling, replacing, and repairing parts) of the assembly machines.

Fig. 3.1 Detailing the Physical Value Chain

Processes downstream, i.e. in the use and waste management stages, should be identified similarly. The procedure is, in principle, repeated until the foreground system is completed and can be linked to LCI database processes of the background system, as described later in this chapter. When carrying out this procedure, the LCA practitioner should identify all multifunctional processes, because they have to be handled next. Note that the step of identifying processes for the LCI model and the step of planning and collection of data are somewhat interrelated. For example, data collected for a given process may lead to the realization that one or more upstream processes are different than the ones previously (assumed) identified. During data collection, the LCA practitioner may, for example, realize that a plastic component is produced from biomaterials rather than petrochemicals, as was initially assumed. The identified processes in this first inventory step should therefore be considered preliminary.

In practice, many processes belonging to levels 3 and 4 will end up being entirely omitted from an LCI model, because their contribution to the indicator score is expected to be insignificant and because data can be hard to find, at least when using the 'bottom-up' (=process-based) approach to constructing inventories. In such cases, the environmental impacts of product systems are systematically underestimated by various degrees. It is an important task of the inventory analysis and consecutive impact assessment to ensure that this underestimation does not violate the completeness requirements for the study.

The life-cycle inventories compiled for all products under consideration are fully based on Ecoinvent v3.4 for both foreground and background data (Wernet et al., 2016). This was necessitated due to the breadth of the study but also to ensure comparability between all modeled systems. All stages from raw materials extraction up to and including the use phase are considered. End-of-life treatment is excluded from the LCI scope due to end-of-life fates being considered separately in the Strategy on Plastics Impact Assessment model. For dealing with co-product allocation, the system expansion method is used via the Ecoinvent "consequential" model, as is generally recommended for studies with decision support in mind (Ekvall and Weidema, 2004). Via system expansion, the consequences of changes in demand for products from unconstrained suppliers (such that can respond to changes in demand, i.e. those that are expected to change) are modeled. Under system expansion, the products modeled receive the full

burdens of the impacts of their inputs and emissions but also receive benefits ("credits", i.e. impacts that are subtracted) for any by-products produced that can substitute other products (such as waste heat used for energy generation). For a fuller discussion on consequential modeling, refer to Ekvall and Weidema (2004) and Wernet et al. (2016).

Japanese Database, IDEA v2 is developed by JEMAI and AIST which are both top organizations that have led LCA movements in Japan. IDEA is a Japanese database that mainly uses national statistics as its data source and aims to model the environmental impacts of all Japanese businesses comprehensively with a high resolution. The former version of IDEA v1.1 was used as the background database to generate the GHG emission factors for the national Carbon Footprint Trial project. It was also used in the Japanese LCA software MiLCA that has been widely used in Japan by many users. The former versions of IDEA supported impact areas such as climate change, acidification, ozone depletion, urban air pollution, mineral, and fossil resources depletion. Since version 2, water resources and land use were added and IDEA now covers a wider range of environmental impact areas used in LCA.

This study focused on the user phase microplastic emissions of the tire. Figure 3.2 illustrates the life cycle of tires.

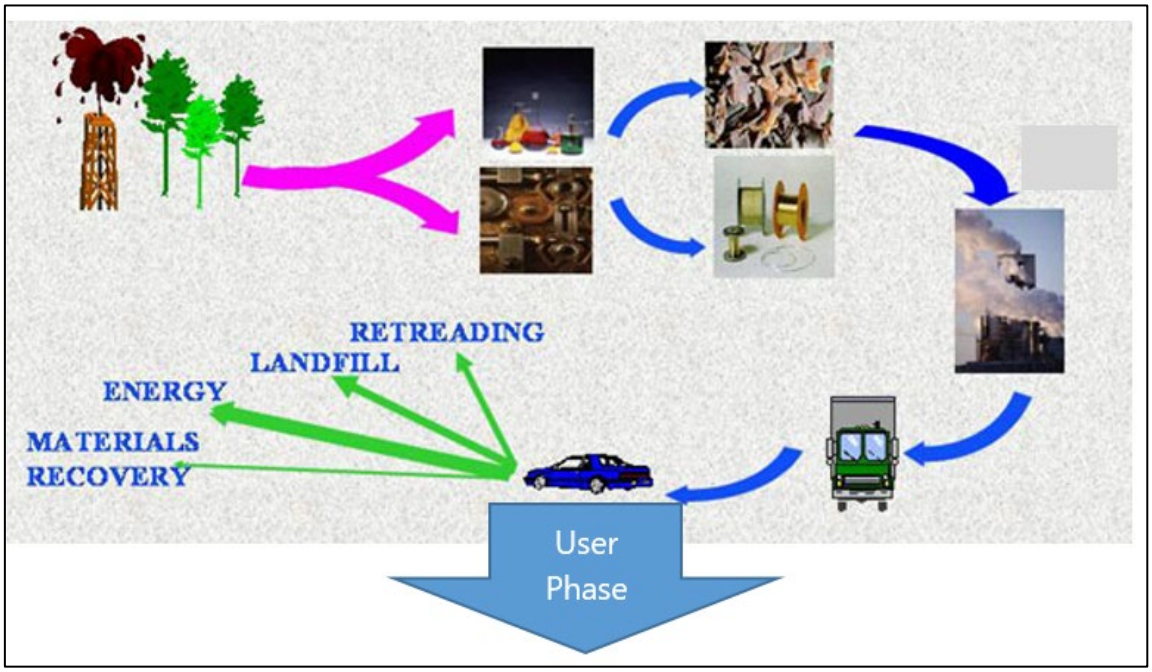


Figure 3.2 Life cycle of tires and the relevance to this study: the user phase tire-wear particle emissions.

In the case of Japan, rubber, and tire imports/exports exists. During production, transportation, usage, and end of life, there are microplastic emissions into the environment. However, this study considered the user phase emissions, which are the wear particles emitted at the road. Hence the considered emissions occur in the geographical area of Japan and the inventories are expressed on that basis.

3.2 Methods

In the present study, tire wear abrasion particle emissions by different vehicle classes have been estimated for Japan. To achieve this, the tire wear emissions per unit distance of traveling have been collected for Japan. The overall process of tire-wear particle inventory development is given in figure 3.3 The summary of data for the vehicle distance traveling is given in annexure 1 table A1.1.

Vehicle tire-wear particle emissions were extracted from peer-reviewed academic publications. The Metadata was summarized according to six vehicle classes namely motorcycle, light vehicle, passenger car, medium vehicle, bus, and heavy vehicle. The summarized data table is given in Annexure 1 Table A1.2.

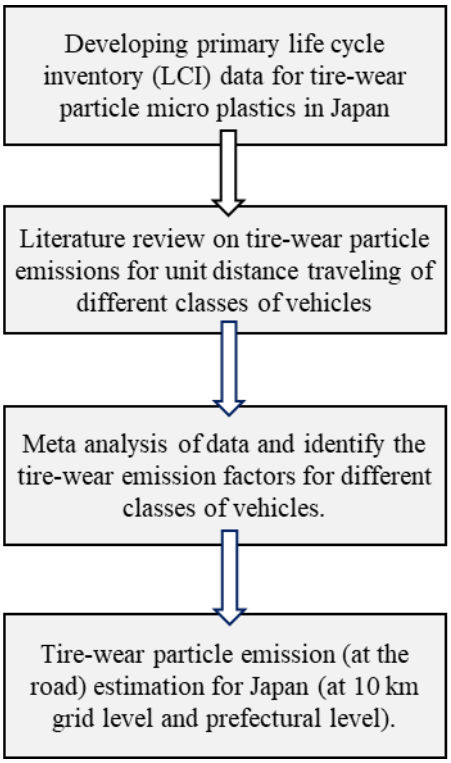


Fig. 3.3 LCI development process for tire-wear particles

Equation 3.1 provides the average annual tier-wear particle emissions calculation formula.

$$\mathbf{ATEi = AUEi * ATDi} \quad \text{---------- Equation 3.1}$$

Annual average tier wear particle per each vehicle class (i): ATEi
Average tire wear particle emission per unit distance vehicle class (i): AUEi
Annual vehicle travel distance: ATDi

3.3 Results and Discussion

3.3.1. Inventory data for tire-wear particles emitted at roads

Table 3.1 summarizes the Meta-analysis of tire-wear particle emissions per unit travel distance data for the six-vehicle classes considered in the study. The values vary in a larger range. The data period is about 25 years. The USEPA data the oldest considered, are having a comparatively lower range, while the recent studies indicate comparatively larger values. Figure 3.4 illustrates the Meta-analysis information in mg of tire-wear particle released per 1 km of vehicle travel. The averages with standard deviations and the first quartile (Q1) and third quartile (Q3) values are also given.

Table 3.2 summarizes the vehicle travel distances for Japan year 2019 (detailed table is given in annexure 1, Table A1.1) and the annual estimated emissions for the average.

Table3.1: Average vehicle tire wear particle releases in mg per km travel distance

Vehicle Class	Motorcycle	Light Vehicle	Passenger Car	Medium Vehicle	Bus	Heavy Vehicle
Average Tire wear particle emissions	7.42 ±5.76	13.86 ±16.26	38.95 ±47.5	47.25 ±47.54	304.08 ±266.1	332.79 ±391.98
Q1	4.1	4.6125	2.71	7.25	99.5	39.5
Q3	8.5	16.25	70	89.25	472	481

Table 3.2: Japan Annual vehicle travel distance (2019) and tire-wear particle emissions for different class vehicles

Vehicle class	Distance (km)	Tire-wear particle emissions (Metric Ton/year)		
		For average	For Q1	For Q3
Motorcycle	1.2757.E+11	946.6	523.0	1084.3
Light Vehicle	2.5514.E+11	3536.9	1176.8	4146.0
Passenger Car	4.0601.E+10	1581.3	110.0	2842.1
Medium vehicle	4.1193.E+11	19463.7	2986.5	36764.8
Bus	5.6973.E+09	1732.4	566.9	2689.1
Heavy Vehicle	1.9637.E+10	6535.1	775.7	9445.4
Total		**33796.0**	**6138.9**	**56971.7**

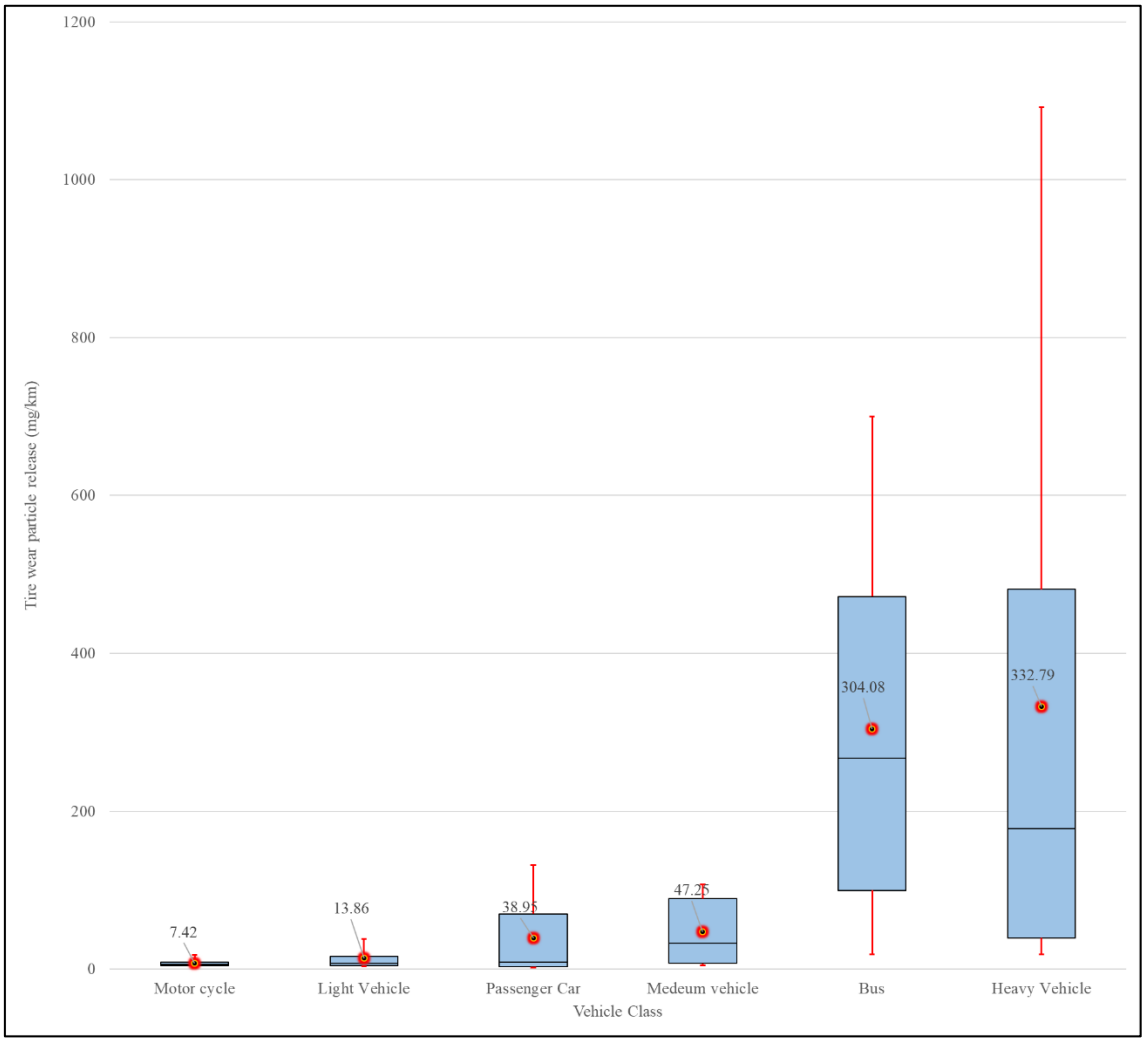


Fig. 3.4. Meta-analysis information in mg of tire-wear particle released per 1 km of vehicle travel

The average tire-wear particle emission at the road estimation is 33,796.0 metric tons (MT)/year for Japan. The estimation of the UNEP report (Ryberg et al., 2018; Ryberg et al., 2019) global tire wear emissions is given as 1,410,000 MT/year. The same report mention that the contribution from Japan is 2% (the value is given to the nearest integer). This provides an approximate value of 28,200 MT/year for Japan. Considering the possible values for two decimal places, 1.50%, and 2.49%, the final answer provides a range starting from 21150 to 35,109. The average value estimated in this study is within the potential range of the UNEP report (Ryberg et al., 2018; Ryberg et al., 2019).

For the estimation of the life cycle impact of tires, the life cycle emissions of tire-wear particles are important. Hence, considering the average life of a tire is 30,000 km,

the tire wears particle emissions per life of attire has also been estimated. Table 3.3 provides the tire-wear particle emissions per life of each vehicle class. Table 3.4 provides the values for a tire in each vehicle class.

Table 3.3: Tire-wear particle emissions per life of a tire of each vehicle class (considering all the tires of a vehicle)

Vehicle class	Life travel Distance (km)	Tire-wear particle emissions (g/life)		
		For average	For Q1	For Q3
Motorcycle	30,000	222.6	123.0	255.0
Light Vehicle	30,000	415.9	138.4	487.5
Passenger Car	30,000	1168.4	81.3	2100.0
Medium vehicle	30,000	1417.5	217.5	2677.5
Bus	30,000	9122.4	2985.0	14160.0
Heavy Vehicle	30,000	9983.8	1185.0	14430.0

Table 3.4: Tire-wear particle emissions per life of tires of each vehicle class (considering the average number of tires per each vehicle class and average life travel distance 30,000 km)

Vehicle class	The average number of tires of the vehicle class	Tire-wear particle emissions (g/life)		
		For average	For Q1	For Q3
Motorcycle	2	111.3	61.5	127.5
Light Vehicle	4	104.0	34.6	121.9
Passenger Car	4	292.1	20.3	525.0
Medium vehicle	4	354.4	54.4	669.4
Bus	6	1520.4	497.5	2360.0
Heavy Vehicle	8	1664.0	148.1	1803.8

3.4 Conclusions and Recommendations

The average tire-wear particle emission at the road estimation is 33,796.0 metric tons (MT)/year for Japan. The average value estimated in this study is within the range of the UNEP report (Ryberg et al., 2018; Ryberg et al., 2019) even though the methods are different. Further improvements can be conducted considering the geographical specific values, for example, the tire wear depends on the road conditions, the traffic density, and other parameters. Hence the tire wear particle emissions for unit travel distance can be vary based on the geographical locations, hence specific data is required.

CHAPTER 4. PLASTIC LITTER LIFE CYCLE IMPACT ASSESSMENT (LCIA) MODEL II: MICROPLASTICS EMITTED AT ROAD AND MICROPLASTICS EMITTED AT HOUSEHOLD WASTEWATER SYSTEMS

4.1 Introduction and Literature Review

4.1.1. Microplastics in biological wastewater treatment

Microplastics that enter centralized wastewater treatment systems can be traced to multiple origins. For simplicity of understanding, the entrance pathway can be divided into two origins: items that go "down the drain", which includes microplastics from kitchen sink wastewater, bathroom wastewater, and toilet flushes. The second origin concerns "street wash" (the street runoff) reaching the WWTPs through combined sewers (Figure 4.1).

The "down the drain" category includes primary microplastics such as microbeads from personal-care products (microbeads are tiny pieces of plastics added as exfoliants, to health and beauty products, such as some cleansers and toothpaste), and textile microfiber from washing machines or other clothes washing activities (such as hand washing). These types of microplastics enter WWTPs through either combined sewers or separate sewer systems. There can also be smaller fractions of secondary microplastics that originate from plastics in WWT systems or from household items. "Street wash", or stormwater runoff, carries primary microplastics such as tire abrasion particles, broken road-marking paint, decomposed plastic household materials due to prolonged exposure to the sun, and secondary microplastics that originate from macroplastic debris.

Through either combined or separate sewers, WWTPs act as major receivers of microplastics. The diversity of plastic polymers and plastic-associated chemicals (Rochman et al., 2019) can vary depending on the sewer network catchment characteristics. Knowledge of the origin of microplastics is useful for WWT system designers, practitioners, policymakers, product designers, and other stakeholders (including educators, who can advise people on how to recycle plastics). System design approaches can be top-down, bottom-up, or hybrid. For example, textile microfibers that are trapped at the household level can be addressed with technological interventions such

as redesigning washing machines or improving the removal efficiency at WWTPs through advanced processes. An additional example concerns broken tire abrasion particles: environmental contamination can be minimized through speed control, promotion of non-tire-based vehicle transport modes (i.e., trains), or expanding combined sewer networks, especially in high-traffic-density areas (coupled with effective wastewater treatment technologies and sludge management approaches in WWT systems).

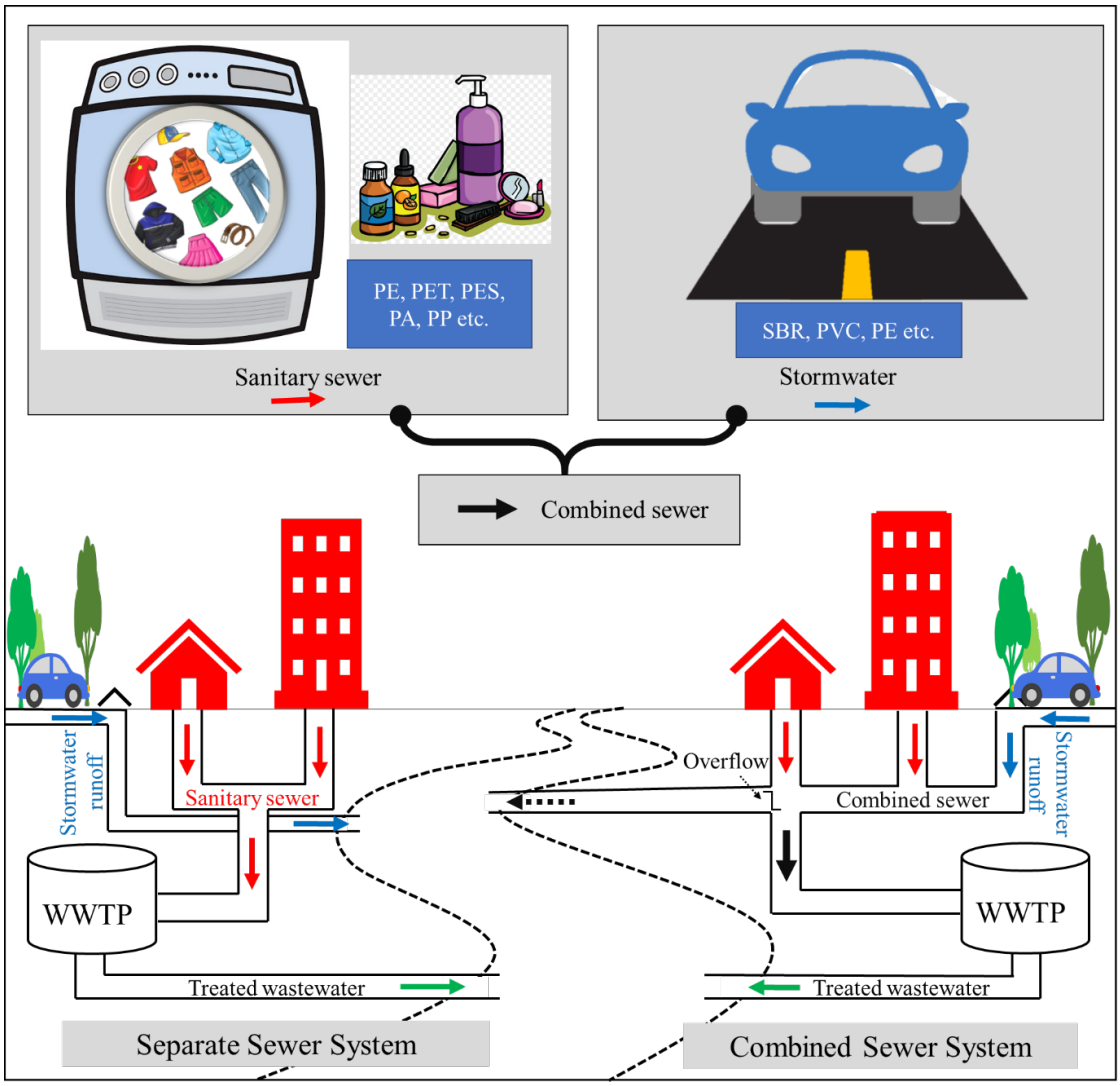


Fig. 4.1 Microplastic inflow to WWTPs through separate and combined sewer systems. [Polyethylene (PE); polyethylene terephthalate (PET); Polyethersulfone (PES); Polyamide (PA); Polypropylene (PP); Styrene-butadiene rubber (SBR); Polyvinyl chloride (PVC)].

4.1.2. Down-the-drain microplastics

A major fraction of "down-the-drain" microplastics are primary microplastics from the textile washing process. Addressing fibrous microplastics derived from textiles is challenging not only due to the microplastic content but also associated chemicals such as perfluoroalkyl acids (PFAAs) (Henry et al., 2019). Research conducted using domestic washing simulations in Italy suggests that significant amounts of microfibers are released from polyester and polypropylene fabrics (De Falco et al., 2020). Microfiber released during washing with detergent can vary from 650 to 3500 fibers per gram of cloth. Even though many studies focus on cases that involve washing-machine-based clothes washing, hand-washing is also a common method practiced in developing countries. Hence, it is also interesting to investigate potential microfiber leakage into the water during the process of washing clothes by hand. Tian et al. (2021) identified that the average length of microfiber from either hand or machine washing is approximately 600 μm. While microfiber lengths fall in a similar range, the quantity of microfiber released was found to be significantly different. The median values of the number of microfibers released were 10,500 fibers per item from hand-washed new clothes and 75,200 fibers per item for machine-washed new clothes (Tian et al., 2021). Considering these differences, the inflow of textile microfibers into WWTPs can vary in a large range based on the "wastewater catchment" characteristics, such as the methods of washing clothes.

Microbeads, an ingredient added to personal care and cosmetic products (PCCPs), which are also discharged into WWTPs through daily human activities, are another major "down-the-drain" microplastic material (Ding et al., 2020). In China, it is estimated that more than 80% of microbeads emitted into the aquatic environment originate from incomplete removal in WWTPs (Cheung and Fok, 2017). Despite the lower mass, due to the smaller size range of microbeads, their estimated quantity is projected to be enormous (Gouin et al. 2015). Microbeads used in the cosmetics industry are often made of polyethylene (PE) or polypropylene (PP). It is estimated that 10,000 tons of microbeads per year are released into the environment through the use of personal care and cosmetic products (Ryberg et al., 2019). Hence, microbeads can also be considered as major microplastics entering WWTPs. With regard to "down-the-drain" microplastics (mainly

coming from washrooms), WWTPs can be considered as potential barriers to microbeads. Although WWTPs may potentially be effective in preventing environmental contamination, direct human contact while using PCCPs is unavoidable. Hence, upstream measures such as regulating the use of microbeads are widely discussed at the policymaking level. Many developed countries (Netherlands, Australia, Canada, Italy, South Korea, New Zealand, Sweden, United Kingdom, and United States) have regulated the use of microbeads in personal care products (OECD, 2020).

4.1.3. Microplastics from street wash runoff

Tire abrasion was found to be the largest source of microplastics contaminating the environment. Globally, such microplastics are estimated to make up 1.4 million tons per year (Ryberg et al., 2019). Life cycle analyses suggest that roughly 20% of synthetic rubber in a tire is displaced its lifetime (Boucher and Friot, 2017). For urban areas, where half of the world population lives, tire abrasion particles are likely to enter sewage systems. This can either reach WWTPs through combined sewers or be released directly to aquatic environments with stormwater runoff. While the specific gravity is 0.94 for tire elastomers, such as SBR, the vulcanized SBR used in tires has a specific gravity of about 1.2 (Bondan, 2019), which makes these rubber particles non-buoyant in water environments. Moreover, the size of microplastics from tire abrasion might be below the detection limit in water environments, as the typical opening size of sampling nets is about 300 μm. In addition, there is also the possibility that microplastics associated with tires are captured WWTPs before reaching water environments or being released into terrestrial environments.

Fragments of broken road markings are another common form of microplastic associated with stormwater runoff originating from streets. These particles can enter WWTPs through combined sewers. Fragmented road markings were reported at high concentrations in cities where there were large amounts of road markings and high traffic density. At the same time, combined sewers are often connected to city sewer networks. There were reported cases of fragmented road markings detected in city dust (e.g., in Japan, Nepal, and Vietnam; (Yukioka et al. 2020) and Norway (Vogelsang et al. 2018)). Various acrylic resins, including methyl 2-methyl propanoate (PMMA), were detected in

road dust samples (Kitahara & Nakata 2020; Yukioka et al. 2020). Properties such as good weatherproofing and paintability of acrylic resins allow for wide application of such resins for various road markings such as lane markings, road crossings on general roads, urban roads, highways, etc. Toxic chemicals associated with some resins are substantial, and their effects on biological WWTPs need to be considered (see Section 5 for further discussion).

4.1.4. Microplastic detection methods, presence, and removal efficiency in WWTPs

Discusses the microplastic detection methods, especially focusing on WWTPs, the reported information on microplastics in various steps of the WWT process, and the removal efficiency of microplastics in WWTPs. Several studies conducted over the past decade have focused on developing analytical methods for sampling, extraction, and identification of microplastics in complex environmental media. However, at present no standardized sampling or analytical protocols have been agreed upon for international use. Sampling methods can be divided into two categories: grab sampling of smaller volume and on-site filtration sampling (using nets or sieves) of comparatively large volume. Grab sampling mostly focuses on sediment, sludge, or (comparatively) smaller-sized microplastic analysis. On-site filtration sampling using nets or sieves for floating and suspended microplastics can be conducted by dragging a net (often a plankton net) or pumping water through a net (or a sieve). Nets (commonly referred to as plankton or manta nets) with mesh opening sizes of about 100 and 300 µm are conventionally used for sampling microplastics in marine or freshwater environments (Lima et al., 2015; Zhang et al., 2020; Maes et al., 2017a). However, dragging such nets is impractical in WWTPs. To overcome this, motorized units coupled with nets and flow measuring devices (Figure 4.1B) have been introduced. Nevertheless, large volume sampling with nets is impeded by the higher amounts of suspended solids in WWTPs, which often results in clogging. Increasing the opening size of the net to minimize clogging would adversely affect the capture of microplastics. This is especially important when considering the size range and the shape of microfibers derived from textiles, as such nets have been found to be ineffective at capturing small to medium-sized fibers (discussed further in Figure 3.2).

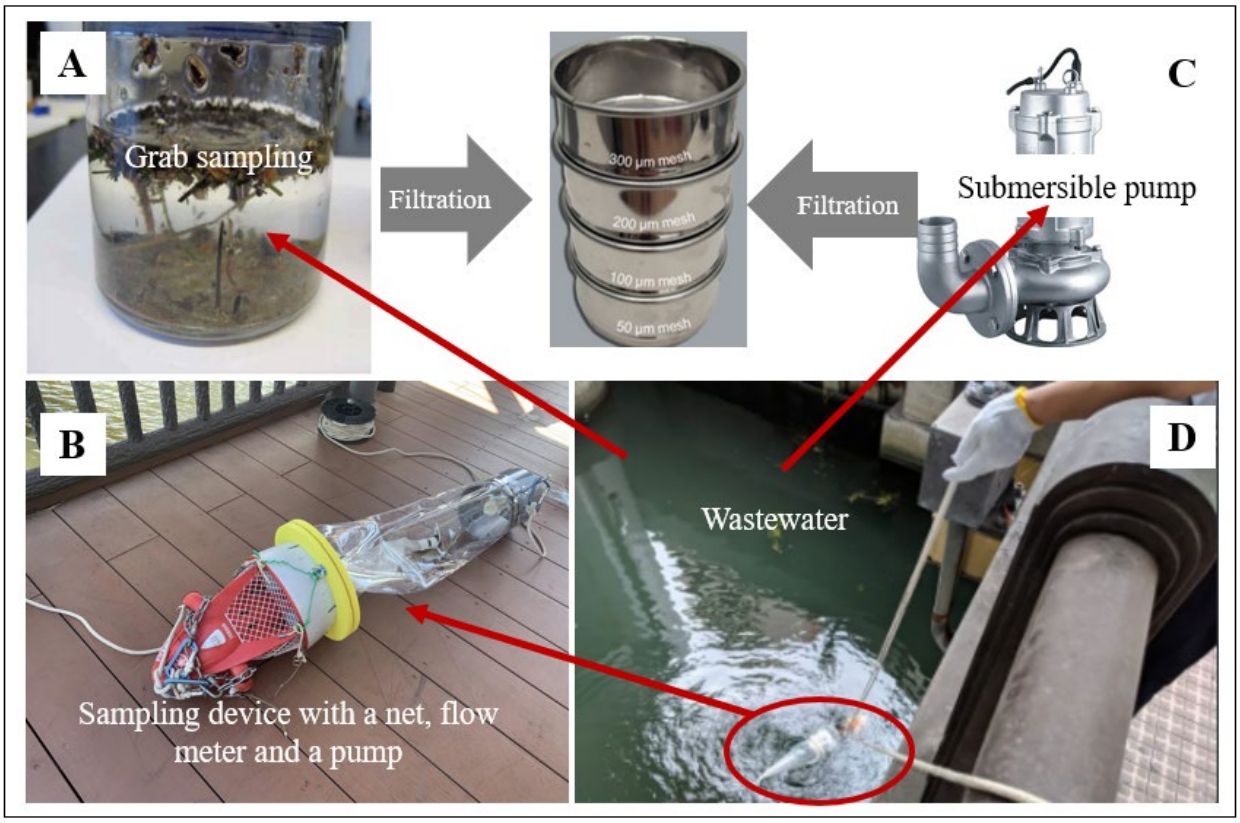


Fig. 4.2. (**A**) Grab sampling followed by filtration, (**B**) large water volume microplastic sampling device (Pirika Inc.), (**C**) submersible pump coupled with sieving system for filtration, and (**D**) sampling at a biological WWTP in Yokohama, Japan.

The best practices and methods for collection, preparation, and analysis recommend that sampling should be straightforward, simple to follow, reproducible, and designed to prevent contamination (Cook & Allen, 2020). In order to avoid contamination, materials such as stainless steel are recommended for sampling devices and equipment. This is an alternative to PA (polyamide or nylon) net-based sampling devices. It is recommended to use a submersible pump (Figure 4.1C) or peristaltic pump with the sampling apparatus. The collection of sludge samples is often conducted using waste-activated sludge in the location illustrated in Figure 4.5G (Li et al, 2018; Lars et al., 2018), sludge from digesters (Bayo et al., 2016), or at sludge post-treatment facilities (Jiang et al., 2020). Composite and simple grab sampling is often practiced in the case of WWTP sludge sampling.

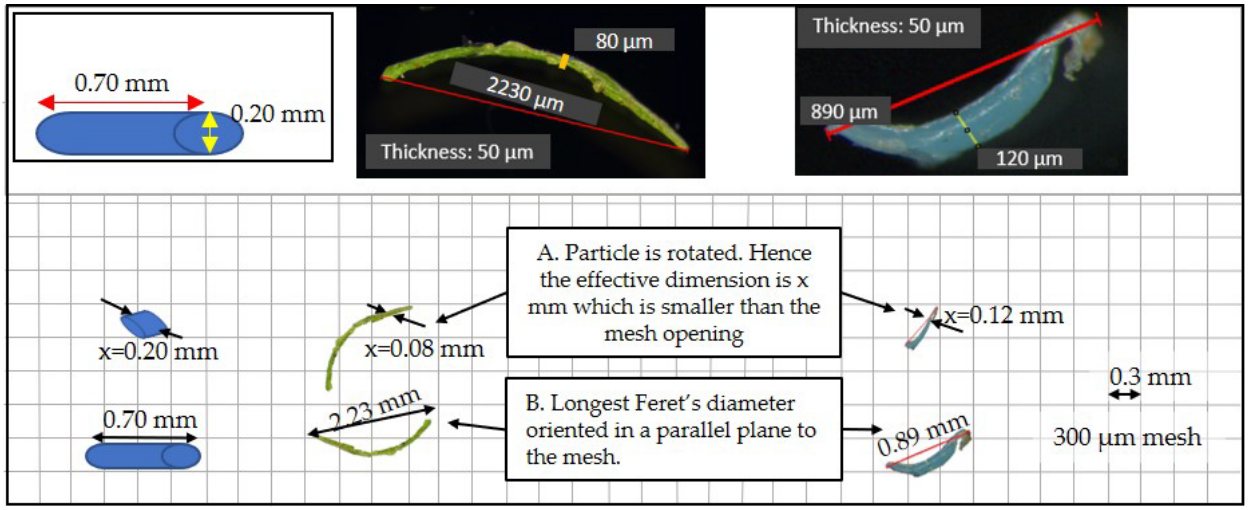


Fig. 4.3. Effect of size and shape of microplastics on retention with net-based sampling. (Figure modified from Abeynayaka et al., 2020.)

As mentioned above, Figure 3.2 depicts the influence of size and shape on the retention of microplastics by nets. In this case, fibers have a higher potential to escape through the nets even though the length of the fiber exceeds the net opening. Hence, microplastic sampling activities at WWTPs may indicate less microfiber recovery from wastewater than is actually the case. Grab sampling presents another option, but may limit the sample volume. Ultimately, grab sampling is also associated with filtration through sieves (or nets) at the lab to separate the suspended solids from the wastewater (Figure 4.1A). In the case of lower volume, the application of nets or filter papers with smaller openings is possible with vacuum filtration if needed (Schmiedgruber et al., 2019; Sabbah et al, 2019; Ben-Davis et al., 2021). In studies focusing on the lower range of microplastics or fibrous microplastics, the preferred method is grab sampling followed by highly sensitive analytical methods. Nevertheless, it is possible to apply large volume methods in secondary or tertiary treatment effluents where the suspended solid concentrations are comparatively low. However, due to the lower concentrations of large microplastics in such effluents, net-based sampling (Figure 4.1B) has been found to be less productive. Large volume sampling with sieves (as described in US-EPA, 2020; extensively discussed by Sun et al., 2019) is the recommended method (Figure 4.1C) to detect low concentrations of small-sized (and/or fibrous) microplastics (Dyachenko et al., 2016).

Due to the challenges outlined above, the application of a uniform method at various levels of WWTPs and sludge treatment makes observations difficult, especially when comparing the removal efficiency and fate fractions of microplastics in field situations. In order to address these challenges, several studies have been conducted with metal-doped microplastics to improve removal efficiency and fate analyses (Keller et al, 2019; Schmiedgruber et al., 2019; Frehland et al., 2020).

Microplastic extraction from collected samples (i.e., samples retained on nets, sieves, etc.) and separation from other debris (such as biosolids) represents another important sampling step. In the case of environmental samples, the often-used protocol for separating microplastics from other debris is density-based separation (using salt solutions of NaCl, $ZnCl_2$, KI, etc.) and degradation of organics (Fenton's degradation) (Tagg et al., 2017). The protocols are described extensively in environmental microplastic-related publications (Ben-Davis et al., 2021). Figure 4.3 (modified from Ben-Davis et al., 2021) illustrates the experimental flow of sampling, extraction, and analysis.

Table 4.1 provides the specific gravity values of typical plastic polymers and solutions used for density-based separation of microplastics. Common low-density polymer types can be readily separated using saturated NaCl solutions. However, for high-density polymer types such as PVC and PET, a saturated NaCl solution may not be effective for purposes of separation. In such situations, $ZnCl_2$ or KI solutions are effective. However, the high cost (more than tenfold more expensive) and the special disposal measures required are among the drawbacks of KI and $ZnCl_2$ solutions. In view of these issues, it is suggested that researchers carefully select the appropriate solution for the density separation based on the objective of the study (i.e., the target polymers to be detected).

Table 4.1. Density of common plastic polymers and solutions commonly used for density separation.

Plastic or Solution	Specific Gravity	Reference
Polypropylene (PP)	0.85-0.94	Lambert and Wanger, 2018
Polyethylene (PE)	0.93-0.97	Alaerts et al., 2018
Polystyrene (PS)	0.96-1.05	Lambert and Wanger, 2018
Polyethylene terephthalate (PET)	1.35-1.39	Alaerts et al., 2018
Polyvinyl chloride (PVC)	1.10-1.45	Alaerts et al., 2018
Polymethyl methacrylate (PMMA)	1.09-1.18	Grigorescu et al., 2019
Styrene-butadiene rubber (SBR)	0.94-1.295	Bondan 2019
Polyamide (PA)	1.12-1.14	Lambert and Wanger, 2018
NaCl solution (saturated)	1.20	Moffitt and Myerson, 2019
$ZnCl_2$ solution (60-65% saturation)	1.60-1.80	Rodrigues et al., 2020
KI solution (saturated)	1.67	Moffitt and Myerson, 2019
Bulk sludge	1.4	Tang and Sillanpaa, 2018

Microplastic separation/extraction from sludge matrices is challenging, considering that the bulk density of sludge is approximately 1400 mg/L (Tang and Sillanpaa, 2018). On the other hand, microplastics associated with biofilms may have adverse effects on simple physicochemical separation activities. In this context, degradation of organics should be conducted as a first step (as illustrated in Figure 4.3), after which, if required, density separation is recommended to separate the plastic polymers from the inert material.

Thereafter, polymer identification can be conducted. Li et al. (2018) reported 22,700 MPP/kg sludge weight by density separation using NaCl solution followed by organic degradation with 30% H_2O_2 method with sampling. The authors reported steering the NaCl–sludge solution for the separation of microplastics from biomass.

Several researchers have used tweezers to examine and pick microplastics from sludge matrices (Lars et al., 2018). In the case of dried sludge, researchers have added small amounts of distilled water to break up the sludge material, and then identified microplastics using an optical microscope. The authors have used additional steps such as changing the background color to enhance the recovery of microplastics during the analysis. This background color variation helps to distinguish microplastics from sludge based on color. However, the full recovery can be affected, and it depends on the skills of the analyst. In this context, total macroplastic concentrations reported for sludge samples may be underestimated.

As outlined above, according to the published literature and organizational best practices, sampling methodologies are diverse and continuing to evolve. Researchers need to assess their research objectives and identify the scope on a case-by-case basis and select the appropriate methods in order to obtain clear results.

Once the sampling has been done, certain properties of microplastics can be analyzed. Rochman et al. (2019) reviewed the physical and chemical properties of microplastics: the common physical properties are mass, shape, and color and the chemical properties are polymer-type and associated chemicals. In the context of WWTPs, both the physical and chemical properties are useful information. Table 4.2 summarizes the important features of the analytical equipment used in microplastic-related research. μRaman and FT-IR-based analytical methods have often been used by researchers for polymer identification. For polymer identification using microscopy, often fluorescence staining (such as Nile Red) is used (Erni-Cassola et al. 2017; Maes et al. 2017b). Sierra et al. (2019) reported microplastics detection in wastewater samples with a polarized optical microscope. Apart from polymer identification, the detectable size range, affordability, and time needed for the analysis are important considerations in the selection/use of equipment.

The detection limit (the smallest particles to be detected) is also an important parameter to consider. The detection limit depends not only on the equipment but also on the analytical skills of the operators. Research related to a smaller range of microplastics (1-100 μm) is hindered due to the unavailability of analytical equipment and a robust method. Moreover, there is no acceptable method of detecting plastic polymers in the nano-plastic range. There were reported studies of nano plastics detected in marine environments with pyrolysis GC-MS, however, the sample quantity is an influential factor (Ter-Halle et al., 2017). Confirmation of the particles extracted from wastewater and sludge matrices in the range of nano plastics has not yet been done. However, in special situations such as polymer degradation (with known polymers), the size reduction observation type of analysis is possible with SEM (von der Esch et al., 2020). Another challenge is analyzing plastic-related chemicals, such as toxic metal analysis. Common metal analysis methods such as inductively coupled plasma mass spectroscopy (ICP-MS) require a sample weight of several grams. However, the weight of a microplastic is less than a milligram (mg). This limits the analysis of toxic metals in microplastics. However, there have been several efforts in toxic metal analysis of larger-sized microplastics using X-ray fluorescence spectroscopy (Turner, 2017; Abeynayaka et al., 2022).

The selection of analytical equipment needs to be done based on various factors. The research objectives should be in line with the available facilities to provide a meaningful outcome. For example, if the available analytical equipment is ATR-FT-IR, studying microplastic removal efficiency by a membrane bioreactor (MBR) or analyzing grab samples with small volumes of tertiary treated effluent would not provide a meaningful scientific output.

4.2 Methods

The basic approach is as listed below

- Wastewater treatment plant information gathering
- Wastewater treatment plant microplastic removal metadata preparation
- Wastewater treatment plant location extraction for Japan (2114 wastewater treatment plants) including the combined sewer information.

- Use the chapter 3 LCI data for tire-wear particle emissions for Japan and estimate the fate of microplastics (considered types: Tire-wear particle and textile microfiber), fate into water environments.

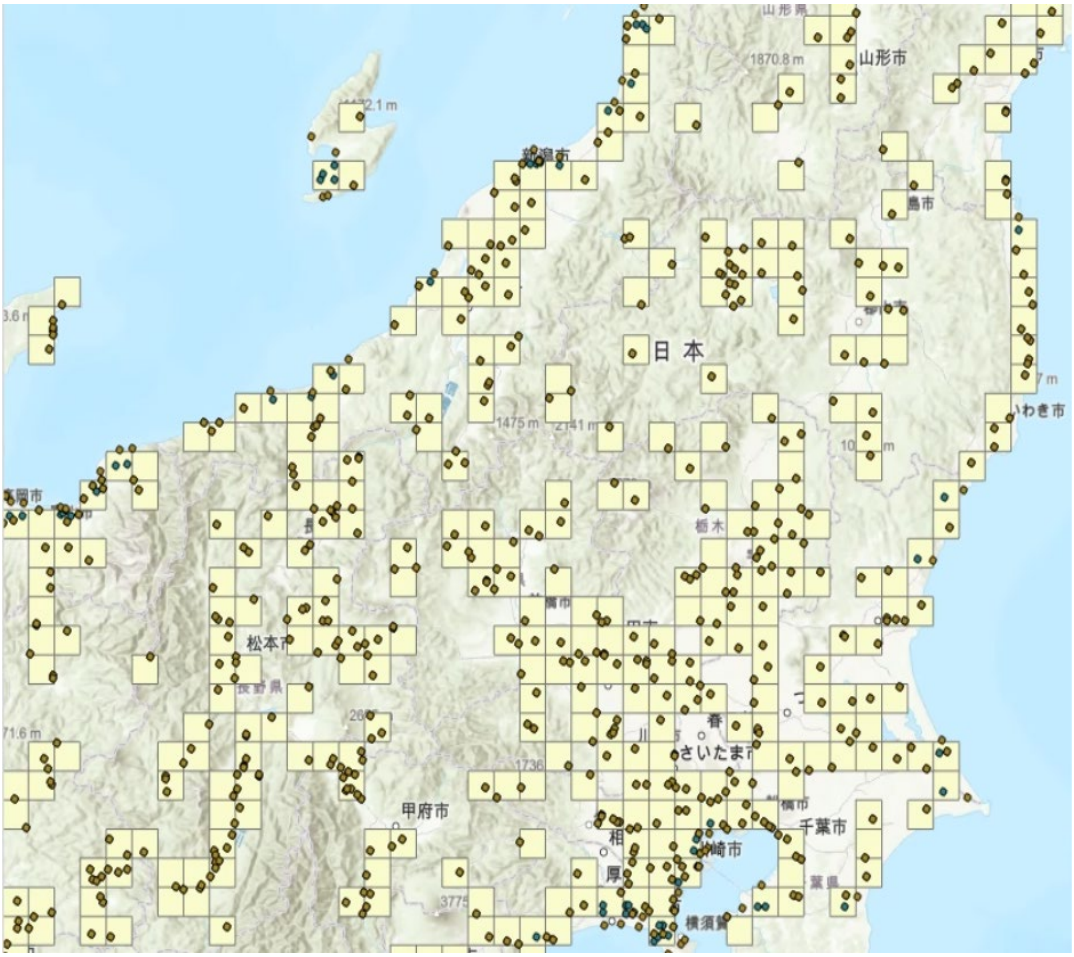


Fig. 4.4 WWTP 10 km grid-level model for the tire abrasion particle emissions and textile fiber emissions fate factor to water estimation

Microplastics emitted at the road (Tire-wear particle and broken roadmaking paint particle type microplastics). Microplastics emitted at household wastewater systems (textile washing released synthetic microfiber type microplastic)

Table 4.2 First distribution of roadside emitted microplastic

Compartment	Component	%	Reference
Air	PM10	2% of initial generation	(Panko et al., 2013; Sjödin et al., 2010)
Drain	>10 µm	21%	POLMIT (2002)
Roadside Soil	>10 µm	67%	(Amundsen and Roseth, 2004; Sundt et al., 2014).

4.3 Results and Discussion

4.3.1. Microplastic concentrations in WWTPs

The reported microplastic concentrations in WWTPs should be analyzed with care to avoid misunderstandings about their magnitude. For example, as discussed in Sections 4.1 and 4.2, there are different sampling and analytical methods. Based on the sampling method, the range of macroplastics capture can vary. Moreover, the lower limit of analytical equipment can affect the measured concentrations and values reported. Figure 4.3 compares microplastic concentrations in WWTP effluent based on analytical limitations and the level of treatment. The analytical limitations affect the reported effluent concentrations. Concentrations measured with methods incompatible with smaller particle detection (limit over 45 µm) had a mean concentration of 0.2 microplastic particles/L, while samples analyzed with methods capable of detecting a smaller range of microplastics (such as µRaman and µFTIR; Table 4.2) indicated a mean concentration of 9.5 microplastic particles/L. WWTP effluents have a lower content of microplastics than larger-sized microplastics.

Considering the level of treatment, with both analytical conditions the tertiary treatment effluent indicates a lower microplastics concentration. However, a comparison between tertiary treatment effluent measured by smaller-size microplastic detection methods and secondary treatment effluent with larger-sized microplastic detection gave misleading results (SE detection limit of >45 (0.4) vs. TE detection limit of <45 (5.3); Figure 4.4). Hence, the importance of having detection limitations when comparing microplastic concentrations from different studies is highlighted.

A study by Talvitie et al. (2017) compares the removal efficiency of different treatment options (Table 4.3). Compared to RSF and DAF, MBR provides an effective barrier to microplastics due to the membrane cutoff. The higher percentages of microplastics in wastewater are transferred to the sludge phase in the three treatment options given in Table 4.3. However, a considerable portion remains in the water phase in the case of RSF and DAF. In all three cases, sludge management and disposal play important roles in counteracting environmental contamination by microplastics.

Table 4.3. Microplastic removal efficiency according to treatment option followed by activated sludge process in WWTPs (adapted and modified from Talvitie et al., 2017).

Treatment	**Influent type**	**Influent microplastics (particles/L)**	**Effluent microplastics (particles/L)**	**Removal (%)**
Rapid sand filtration (RSF)	Secondary	0.7	0.02	97.1
Dissolved air flotation (DAF)	Secondary	2.0	0.1	95
Membrane bioreactor (MBR)	Primary	6.9	0.005	99.9

Table 4.4 provides information related to the shape of microplastics. A shape-based comparison of microplastics in WWTPs indicates higher amounts of fiber-shaped microplastics in wastewater. As microfiber derived from textile washing is a major source of microplastics in WWTPs (Schellenberger et al. 2019), the higher presence of fibers can be understood. The primary clarifier is effective in the removal of both shapes of microplastics (fragments and fibers).

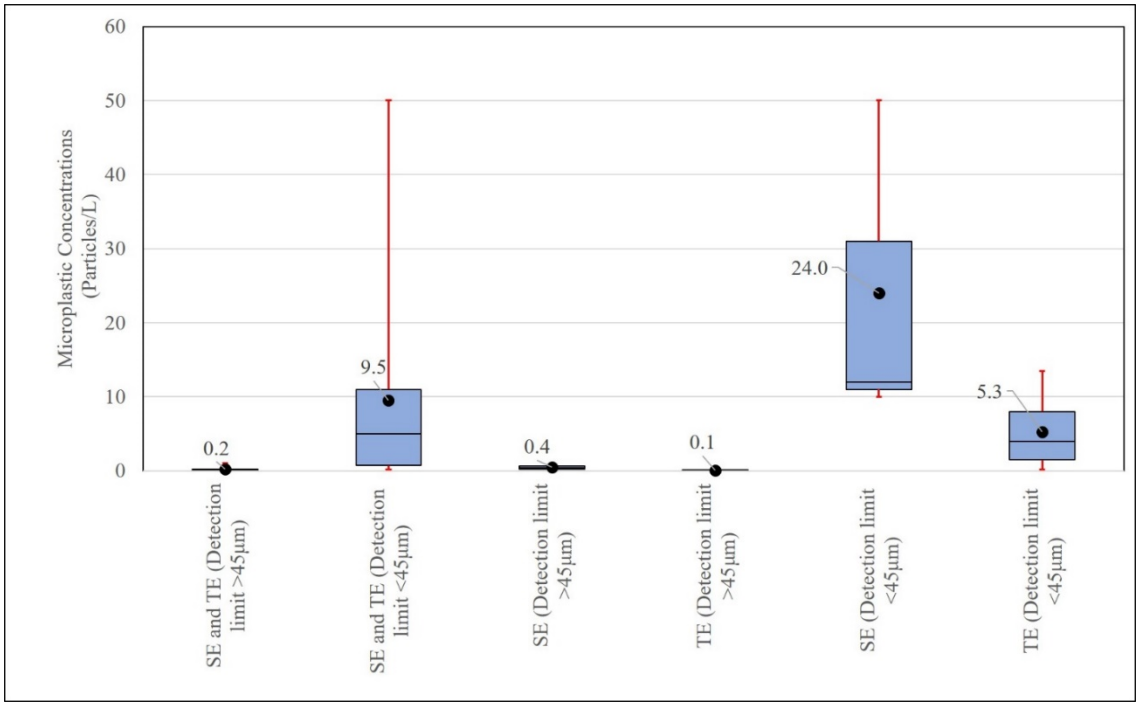


Fig. 4.5 Microplastic concentrations in wastewater treatment plant effluents (meta-analysis of values from reported literature). Detection limit comparison is based on sampling and detection methods; secondary treatment effluent (SE) and tertiary treatment effluent (TE) comparisons assess treatment level effects. The top edge of the box and bottom of the box are upper and lower quartile, respectively (box spans the interquartile range). The horizontal line inside the box and the point marker with a number represent median and mean value, respectively. Whiskers represent the highest and lowest observations.

Mean effluent microplastic concentrations from SE and TE (with smaller microplastic detection) are 24 and 5.3 particles/L respectively. It is estimated that 85 km^3 of wastewater is generated in North America annually (UNU, 2013). A simple calculation would show emissions of more than 1000 billion microplastic particles from WWTPs annually. Considering the amount of untreated wastewater and the level of treatment, the actual environmental contamination is even greater.

Table 4.4. Average concentrations of microplastic particles and fibers in wastewater (shape-based comparison) (adapted and modified from Lares et al., 2018).

Sampling point	**Average concentration of microplastic fragment (particles/L)**	**Average concentration of microplastic fiber (particles/L)**	**Average concentration of microplastic (particles/L)**
Influent	5 (±1.3)	52.6 (±11.3)	57.6 (±12.4)
Effluent from primary clarifier	0.2 (±0.1)	0.3 (±0.1)	0.6 (±0.2)
Final effluent	0.5 (±0.2)	0.5 (±0.3)	1.0 (±0.4)
MBR permeate	0.1 (±0.1)	0.2 (±0.1)	0.4 (±0.1)

This section discusses microplastics in biological wastewater treatment plant sludge and the environmental relevance concerning sludge treatment/disposal options. As previously discussed, a larger fraction of microplastics is removed from the water phase at WWTPs. This removed fraction is transferred to the biomass and withdrawn from wastewater treatment units with the sludge. Hence, the higher removal efficiency of microplastics at WWTPs means that the macroplastics are highly associated with the sludge. Therefore, sludge treatment and disposal options play important roles in reducing the environmental contaminants of macroplastics. Lars et al. (2018) reported that dry sludge from the activated sludge process, digested sludge, and MBR contained 23, 171, and 27 microplastic particles per gram of sludge, respectively. These values are significantly higher compared to the values reported in WTTP microplastic effluent. However, considering the balance of microplastic particle numbers in a WWTP, the numbers do not add up (assuming 3 grams of mixed liquor suspended solids (MLSS) per liter of wastewater and one liter of wastewater in an aeration tank consisting of 8-10

microplastics). The sludge of this aeration tank should produce more than 1000 microplastic particles per one gram of sludge. This difference can be due to the practical difficulties of detecting microplastics in sludge samples (Corradini et al. 2019) and the breaking of macroplastics into smaller pieces due to mechanical forces in WWTPs (where there is minimal potential biological degradation), resulting in the limited detectability.

Table 4.5 indicates the shape-based concentrations of microplastics in wastewater treatment plant sludge, showing higher amounts of fiber-shaped microplastics. MBR sludge indicates slightly higher microplastic concentration compared to conventional ASP sludge. This can be due to the membrane retention and hence accumulation of microplastics within the MBR reactor transferring it to the biosolids.

Table 4.5. Average concentrations of microplastic fragments and fibers in wastewater treatment plant sludge samples (adapted from Lars et al., 2018).

Sampling point	**Concentration of microplastic particles (particles/g dw*)**	**Concentration of microplastic fibers (particles/g dw*)**	**Total concentration of microplastic (particles/g dw*)**
Activated sludge	1.3 (±1.3)	21.7 (±4.6)	23.0 (±4.2)
MBR sludge	3.3 (±2.4)	24.1 (±6.1)	27.3 (±4.7)

*Particles/g dw: Microplastic particles per gram of dewatered sludge.

Figure 4.6 summarizes the presence of different microplastics in the sludge of different treatment systems. The mean values are in the range of 18.0-24.6 particles per gram of dry sludge. Considering the quantities of sludge produced in a year (Japan, 2000 t/year; USA, 6500 t/year; EU, 8900 t/year) (Mateo-Sagasta et al., 2015), the number of microplastics associated with sludge is in the millions (e.g., Japan, 40,000,000,000 particles/year). Hence sludge treatment and disposal play important roles, as mentioned previously. The common practices for sludge disposal are land disposal and incineration.

In Japan, over 80% of sludge is incinerated, restricting a major portion of microplastics from entering the environment. However, the other common disposal method, applying sludge on agricultural soils as a fertilizer (Corradini et al. 2019), provides a pathway for microplastics to re-enter the environment.

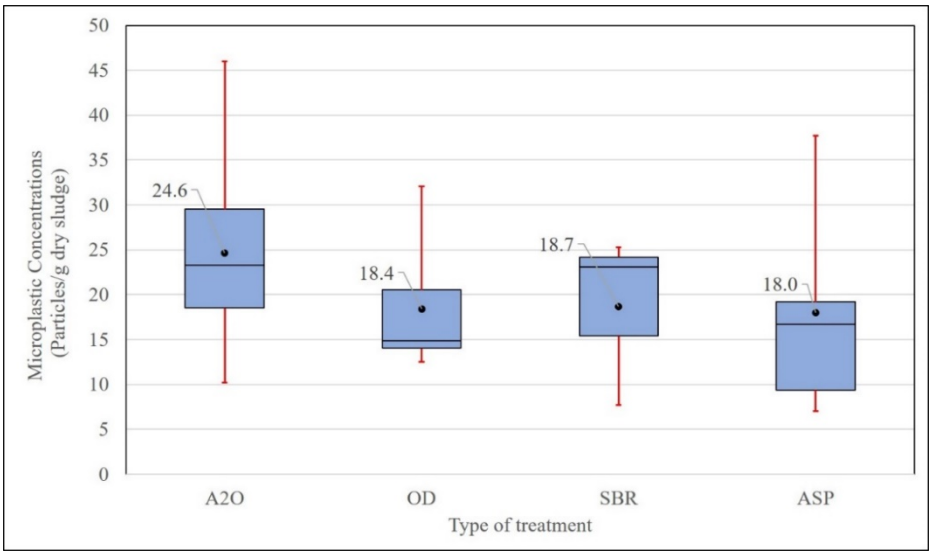


Fig. 4.6 Microplastics in sludge of wastewater treatment systems. Top edge of box and bottom of box are upper and lower quartile, respectively (box spans the interquartile range). Horizontal line inside box and point marker with number represent median and mean value, respectively. Whiskers represent highest and lowest observations. [Types of treatment are as follows: anaerobic, anoxic and oxic (A2O), oxidation ditch (OD), sequencing bioreactor (SBR), and activated sludge process (ASP)].

4.3.2. The tire-wear particle and synthetic textile washing microfiber emissions to the environment

Figure 4.7 and 4.8 provides the tire-wear fate factors into the water at the prefectural level for the long-term complete wash off from the roads and for the short-term 21% wash off from the roads to drain respectively. The fate factor to the water of Tokyo is the smallest, and compared to the rest of the prefectures, the value of 0.23 for the long-term complete wash-off is smaller. The presence of combined sewers is a main governing factor in this situation.

Tokyo: Wards area 10 combined sewer regions and Tama area 8 combined sewer regions considered exists. The annual CSO data collection (2018).

Due to the release of the 12% of expected runoff which assumed to be gone through the CSO network of Tokyo is actually overflowed due to exceeding the capacity of the wastewater treatment plants during high intensity rainfall events.

Assumed the tire-wear emissions are equally distributed in the rainfall runoff despite the rainfall intensity variations. Further studying on this targeting journal publication. (The variation of particulate matter in road runoff depending on rainfall intensity could be one recommendation for future studies to improve the accuracy of the fate factor estimation).

Fig. 4.10 provides the fate factors for textile microfiber into the water at the prefectural level. The consideration of CSO in Tokyo, have increased the emissions into water environments from 0.23 to 0.4. However this 0.4 could be further increased in the case where high intensity rainfalls wash more tire-wear particle from the streets than the common rainfalls where no CSO occur and the runoff passes through wastewater treatment plants.

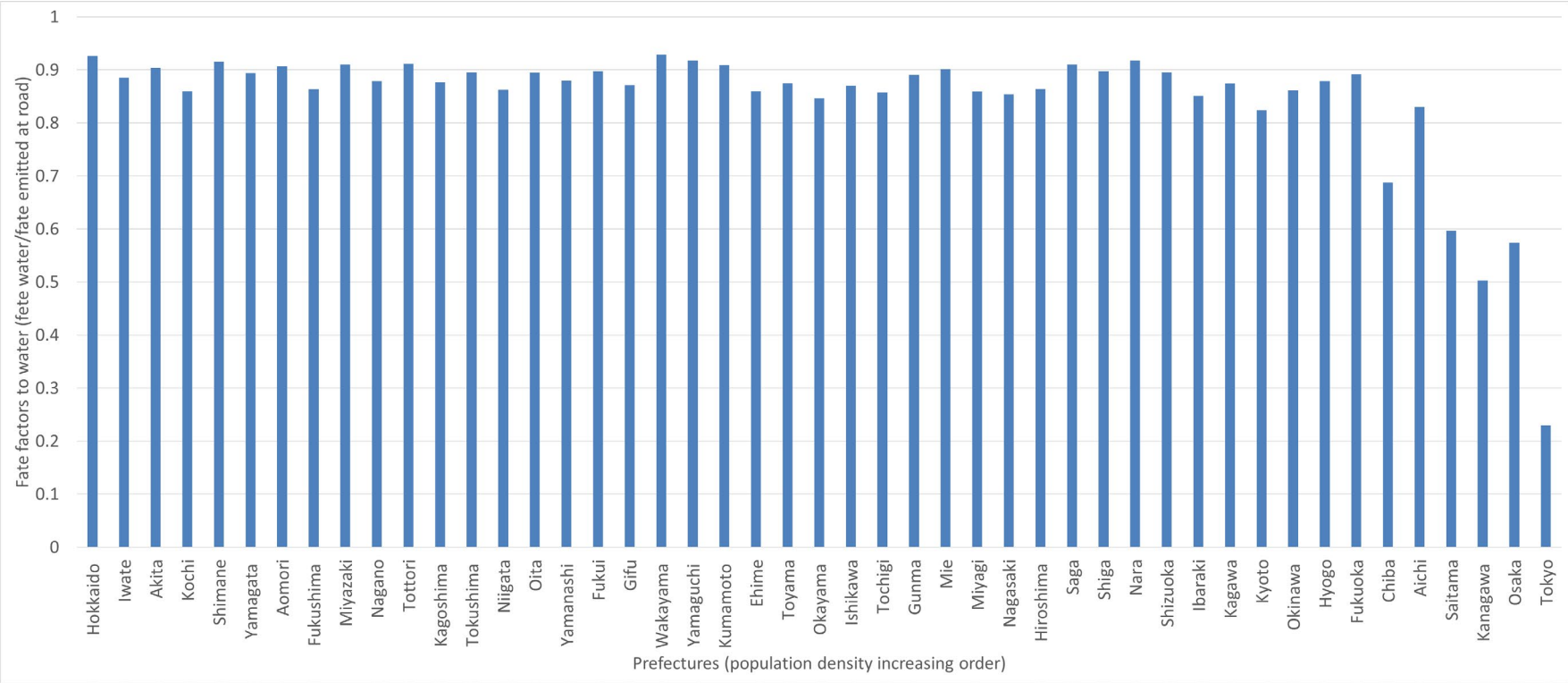


Fig. 4.7 Tire wear fate factors into the water at prefectural level (for the long-term complete wash off from the roads).

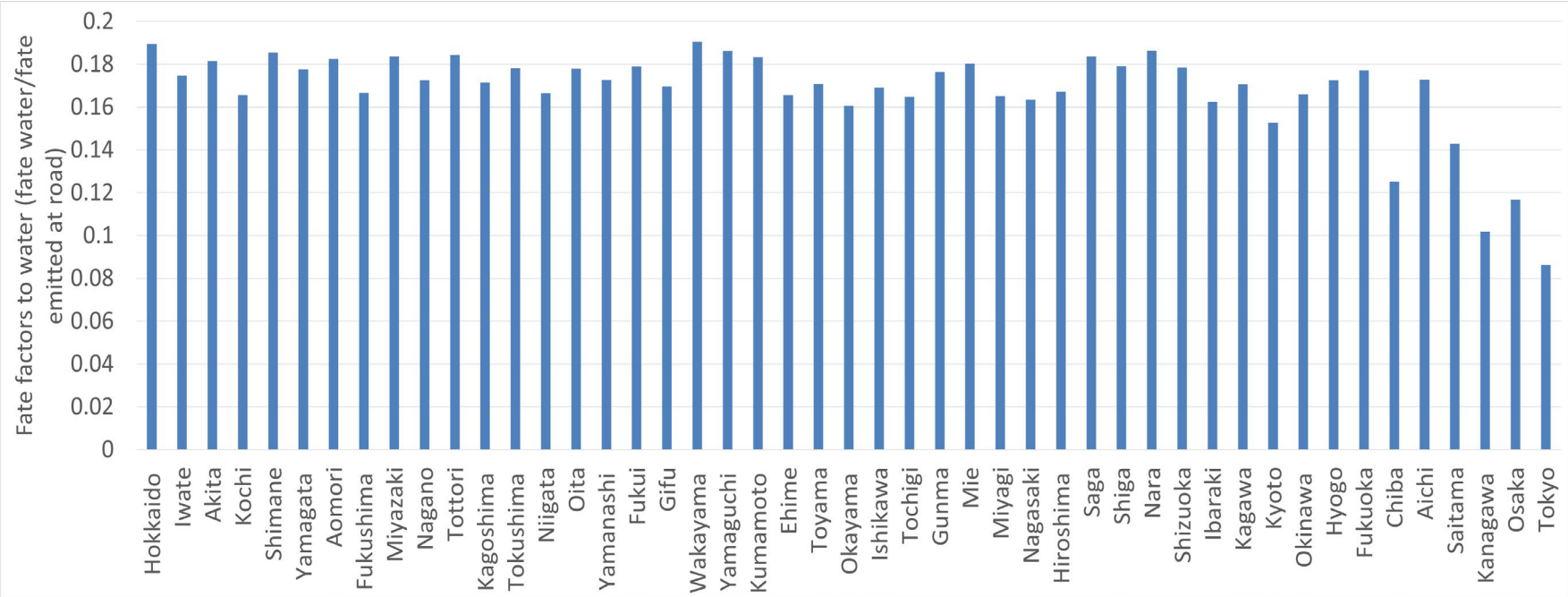


Fig. 4.8 Tire wear fate factors into the water at prefectural level (for the short-term 21% wash off from the roads to drain).

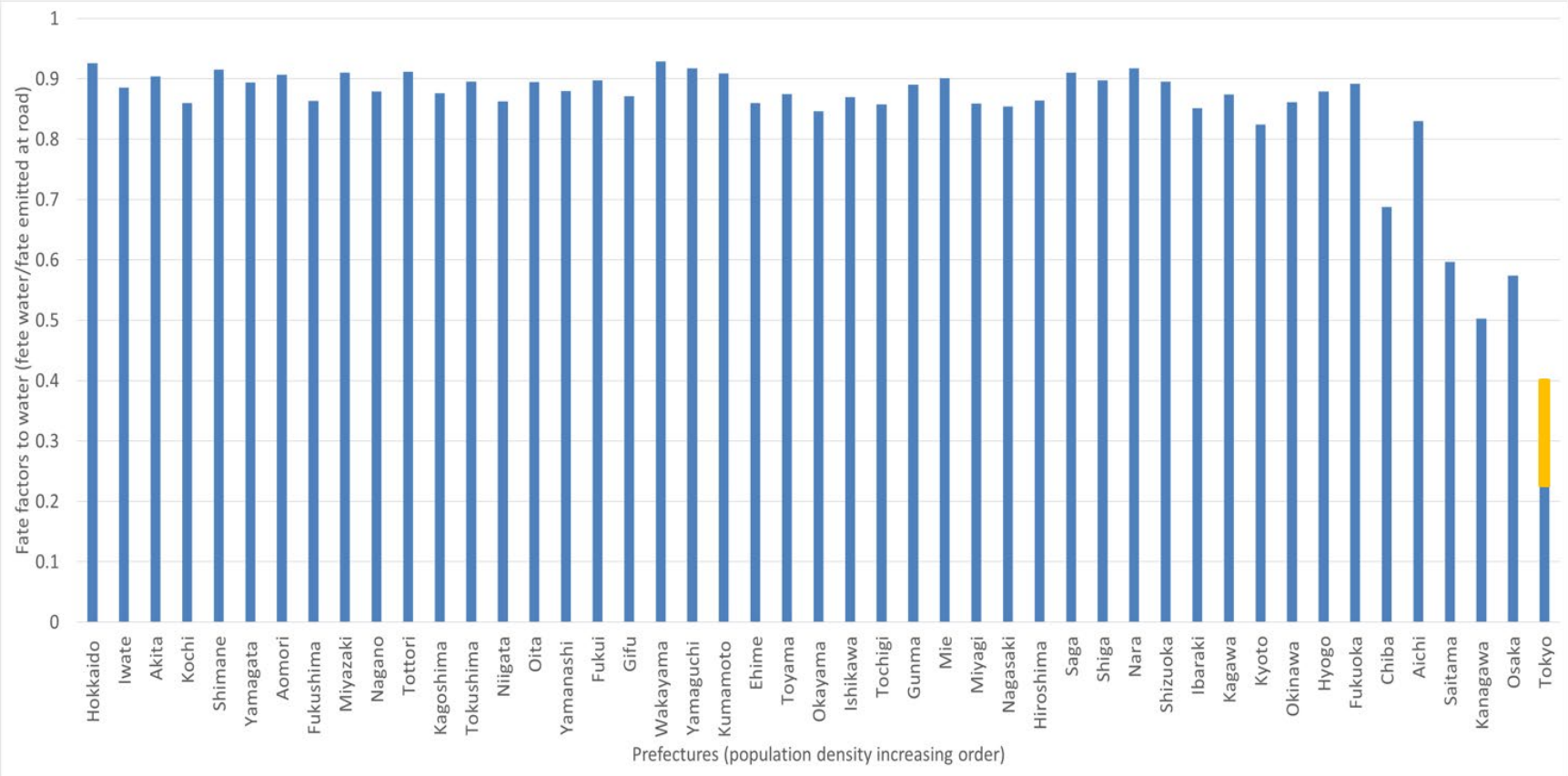


Fig. 4.9 Tire wear fate factors into the water at prefectural level (Long-term scenario considering CSO for Tokyo: Orange portion of the bar).

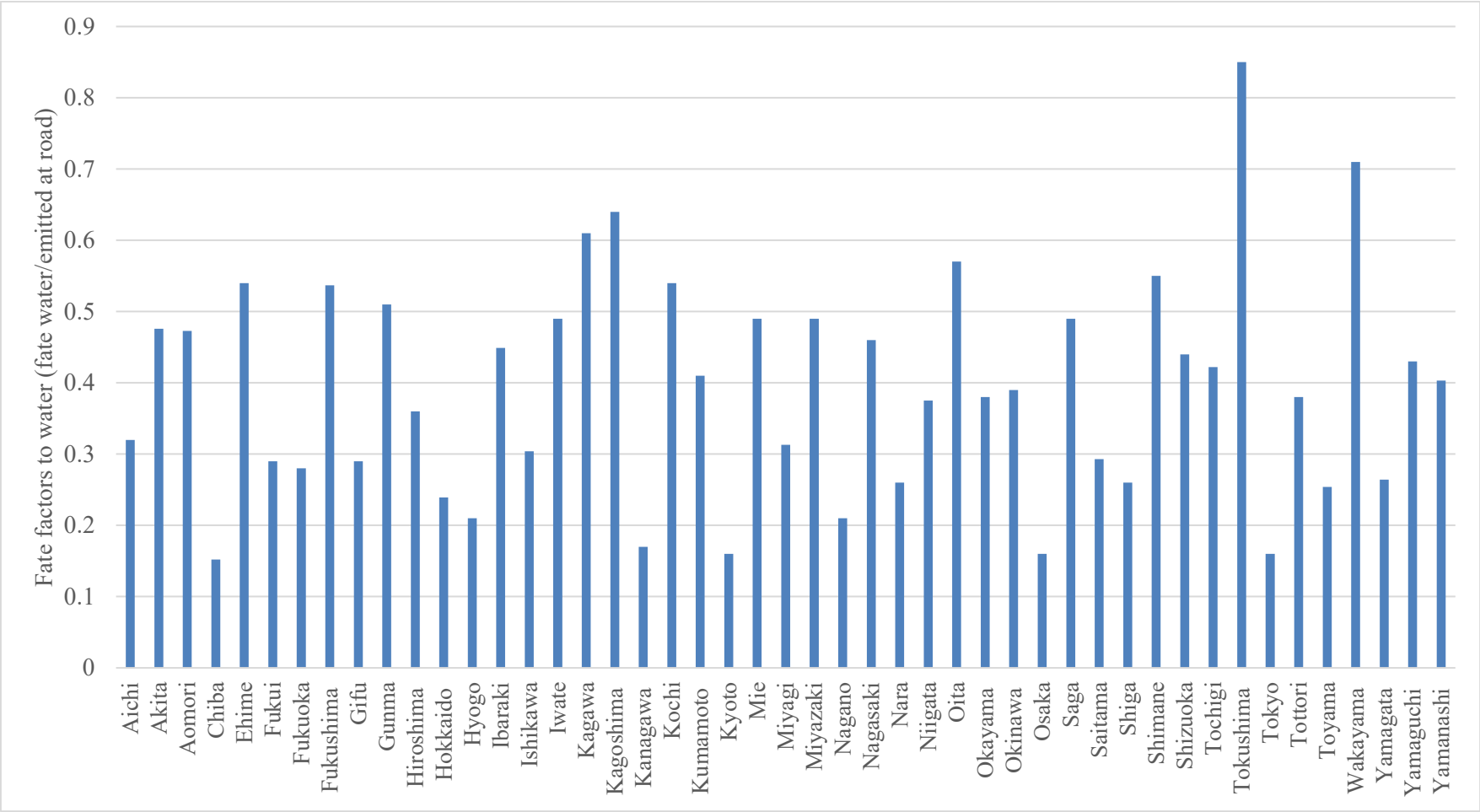


Fig. 4.10 Textile microfiber fate factors into water at prefectural level (down the drain from the washing)

4.4 Conclusions and Recommendations

Tire wear particle emissions are roads has the fate in few compartments (i.e. air, freshwater, soil and wastewater treatment plant sludge). The quantity goes to air is 2% (the smaller sized fraction such as PM 10, PM 2.5), which needs to be assessed with air pollutant transport models and estimate the human health and other impacts.

Rainwater carries the deposited particles with rainwater runoff. If combined sewers exist, then this reach the WWTPs. In the situation of textile microfiber, the major fraction goes into sludge (more than 99%).At the WWTPs, main transfer is into sludge phase while less than 1% goes with treated wastewater. Sludge management is the important step to avoid the recontamination. The main method used in Japan, the incineration eliminates this fraction and convert into CO_2.

For Japan, the sewer coverage (network coverage and combined sewer presence) and treatment level make the difference of fate into water environments. Combined sewer overflow (CSO) could be significant for the cities. Especially the high intensity rain could carry more road deposited particles with the runoff. Then, if CSO occur, the higher portion of particles could enter the water environments.

CHAPTER 5. PLASTIC LITTER LIFE CYCLE IMPACT ASSESSMENT (LCIA) MODEL I: FLOATING AND SUSPENDED MICROPLASTICS

5.1 Introduction and Literature review

This chapter discuss the development of fate factors for the floating and suspended microplastics larger than 300 µm suing river floating and suspended microplastic measurement data. The GIS based model development process is illustrated in the figure 5.1

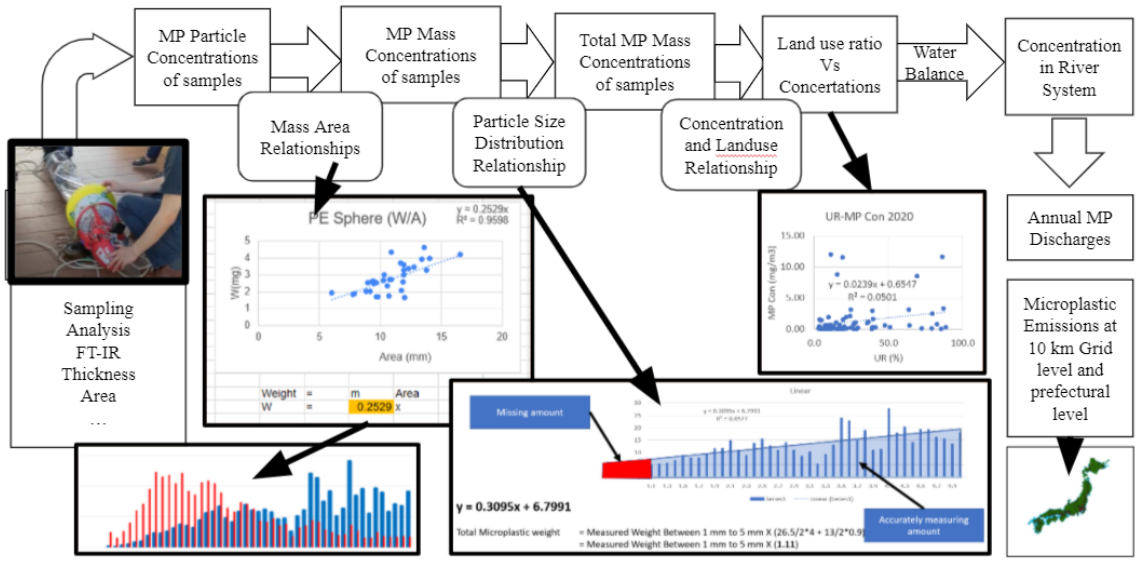


Fig. 5.1. The GIS-based model for development process floating and suspended microplastics larger than 300 µm using river floating and suspended microplastic measurement data

Aquatic plastic pollution is considered to be one of the most challenging global environmental issues. The longevity of plastic is estimated to be hundreds to thousands of years. Hence, the plastics released into the environment will remain for hundreds of years (Thompson et al., 2009) The adverse effects of plastic litter in the environment have been widely discussed in the literature (Horton et al., 2018; Bellasi et al., 2020). Plastic contaminants in freshwater are a threat to the ecosystem and a potential health hazard to humans (Jemec et al., 2016; Su et al., 2018; Redondo-Hasselerharm et al., 2018). Apart from that, freshwater resources (i.e., rivers) carry plastic litter into marine environments. Then, local and regional plastic pollution becomes a global concern. Besides, rivers are considered to be a primary contamination pathway of microplastics to reach the oceans.

It is estimated that between 1.15 and 2.41 million tons of plastic waste currently enter the ocean every year from rivers (Lebreton et al., 2017). According to Schmidt et al (2017), 80–90% of the plastic that reaches oceans comes from rivers. Major sources of the marine plastic litter that is transported through rivers originate from improper waste management practices, wastewater treatment plant effluent, and inland transportation-related activities (Ryberg et al., 2018). Due to the long half-life of plastic polymers, the plastic concentration in oceans is increasing. It is predicted that, within the next 50 years, in some oceanic regions, the microplastic concentrations will exceed 1 g/m^3 (Isobe et al., 2019). Thus, further research on plastic litter in riverine environments is important to mitigate aquatic (both freshwater and marine) plastic pollution.

Based on size, plastics are categorized into macro- (>25 mm), meso- (25–5 mm), micro- (5 mm–0.1 μm), and nano-plastics (<0.1 μm) (Cozzolino et al., 2020). Microplastics are divided into two categories, primary and secondary, based on their origin. Primary microplastics enter the environment directly as microplastics. Secondary microplastics derive from the breakdown of larger plastics in the environment (Microlastics, 2020). The environmental degradation of plastic is governed by a synergic effect of photo- and thermo-oxidative degradation, abrasion, and biological activities (Thompson et al., 2009; Hakkarainen et al., 2004).

Microplastics in a riverine environment can be sub-categorized into several groups, such as floating plastic, suspended plastic, plastic on the riverbed, plastic in the sediment, and plastics in biota (Van Emmerik and Schwarz, 2020). This depends on the hydraulics of the waterbody and properties of plastic, such as composition, specific gravity (SG), and shape. Plastic particles change in size and SG by degradation, aggregation, or by the growth of biofilms, and flow turbulences move these particles through inter-and intra-river sub-compartments.

Considering the SG range of various plastic polymers, floating and suspended plastics are dominated by polyethylene (PE) and polypropylene (PP) due to the lower specific gravities. Moreover, PE and PP are the highest released plastic polymers from land-based sources to the environment (Ryberg et al., 2018). However, as mentioned previously, due to river flow currents and the association with air bubbles, biofilms, etc., higher SG polymer microplastics could also exist in riverine environments as suspended

or floating particles. Quantitative information on microplastics in freshwater and coastal water environments is useful for several applications, including plastic litter fate assessments (Isobe et al., 2019) and risk assessment studies (Koelmans et al., 2019). Environmental sampling plays an important role in assessing the plastic concentration in aquatic environments. Several studies have been conducted to quantify microplastic contamination in riverine environments (Lebreton et al., 2017; Van Emmerik et al., 2018).

5.2 Methods

5.2.1. Microplastic sampling and the data set preparation

Essentially, there are two types of sampling for marine and freshwater environments, grab samples of smaller volumes and on-site filtration sampling with comparatively large volumes. Grab sampling is mostly focused on sediment sampling; however, some studies focused on floating and suspended microplastics sampling (Scircle et al., 2020; Alam et al., 2019). The onsite filtration sampling of floating and suspended microplastics can be subdivided into several methods based on the kind of devices used. Simple plankton nets (also referred to as manta nets) with mesh opening sizes about 100 and 300 µm comprised the basic method used by many researchers (Lima et al., 2015; Maes et al., 2017; Zhang et al., 2020). Often, plankton net sampling requires supporting infrastructures, such as a trawl/boat or, in the case of river sampling, a bridge or a solid riverbank. Sometimes, the water body is not deep enough to be sampled using a boat, and the filtered quantity cannot be measured accurately (Bordos et al., 2019). Furthermore, the filtered volume estimation in microplastic sampling with trawling net method often uses the boat speed and the opening area of the net (Lima et al., 2015; Maes et al., 2017; Zhang et al., 2020). The lack of accuracy of this sample volume calculation affects the expressed concentration. Additionally, sampling with nets (suspended from a bridge) using the natural river flow takes long durations (Kataoka et al., 2019). Hence, the lack of standard methods for sampling, concentration calculation, and reporting makes comparison difficult. For example, if coastal and riverine sampling methods are different and affect the accuracy of observations differently, the comparison of observations is less

meaningful. Therefore, the development of a method suitable for multiple environmental conditions is required.

Several studies focused on pumping followed by a filtration method for sampling in rivers. In this case, water is pumped out of the river, and then the water is filtered through a series of filters (Bordos et al., 2019). The complexity of submerged pumps, lower pump flow rates, the difficulties of using the bridges, and a large amount of equipment to carry are restrictions for wide applications of this method. However, Li et al (2020) reported a suction pump coupled with a submerged net acting as an in-situ filtration tray, which is different from the structure of the previously reported studies. The sampling methods and the devices are evolving, as riverine and coastal environments are diverse. There are challenging field situations and various objectives of sampling. As highlighted previously, riverine environments are more diversified, and applications of simple trawling-based methods are limited. Whereas, in marine environments, both trawling and pumping-based methods are applicable. However, in coastal environments including ports and bays, there are limitations of trawling. Additionally, microplastics have various compositions [polymer, color, shape, etc. (Rochman et al., 2019)]. Hence, to obtain representative environmental samples, a larger volume needs to be filtered. However, the bridge suspended nets with natural environmental flow require longer durations (especially when the river flow velocities are low), and in the tidal backflow influential zones, the mounted nets are inapplicable for proper functioning (Kataoka et al., 2019). Hence, considering the applicability in the field and the uniformity of the observations, we would like to emphasize the studies focusing on the development of microplastic sampling devices.

Microplastic sampling in many freshwater locations faces challenging conditions, such as access limitations, restricted budget, a lack of human resources, and time limitations. Riverine and coastal samplings are subjected to time limitations due to several reasons. For instance, urban waterways with boat/freight traffic sampling devices suspended from the bridges cannot remain unattained. There is a need for a reliable, easy, time-saving, and affordable sample collection method to expand the microplastic-related information in water environments. With the objectives of developing a pragmatic rapid sampling process applicable in both freshwater and coastal areas, we compared a

developed sampling device with a conventional sampling plankton net. This developed method was tested under several conditions to determine the suitability for environmental sampling. Then, the collected samples were purified and analyzed to determine the particle size distribution and the polymer composition of the collected microplastics.

To collect microplastics in challenging riverine and coastal water environments, the microplastic sampling device "Albatross" was developed. The development of the "Albatross" sampling device was conducted under a project initiated in 2015. A series of testing and device modifications were conducted in Japanese riverine and coastal water environments. This history and additional information can be accessed at https://en.plastic.research.pirika.org/. The performances of the two latest versions of this sampling device (Albatross Mark 5 (AM-5) and Albatross Mark 6 (AM-6)) were compared to a conventional plankton net (or manta net). Previous studies reported both terms, manta net and plankton net, which refer to a similar device in terms of microplastic sampling studies. Hereafter, in this manuscript, the term plankton net is used for uniformity. The initial step of the study focused on the comparison of these three devices in riverine environments to collect floating and suspended microplastics (mesh sizes 100 and 300 µm with all three sampling devices). The second step focused on microplastic sampling in Japanese riverine and coastal water environments with the AM-6 (mesh size 300 µm). In a considerable amount of the sampling locations, it was challenging to collect samples using conventional sampling methods (i.e., a bridge suspended nets and trawling nets). These challenging field conditions included limited riverbank access due to constructions, height from the bridge to the river surface over 10 m, tidal flow influenced zones, limited access from boats due to shallowness or security reasons, etc.

5.2.2. Sampling Process

AM-5 and AM-6 are microplastic sampling devices developed by Pirika Inc (Tokyo, Japan). Fig 5.2 b,c illustrate the basic components of AM-5 and AM-6. AM-5 consists of a plankton net with a mouth diameter of 24 cm (length 750 cm).

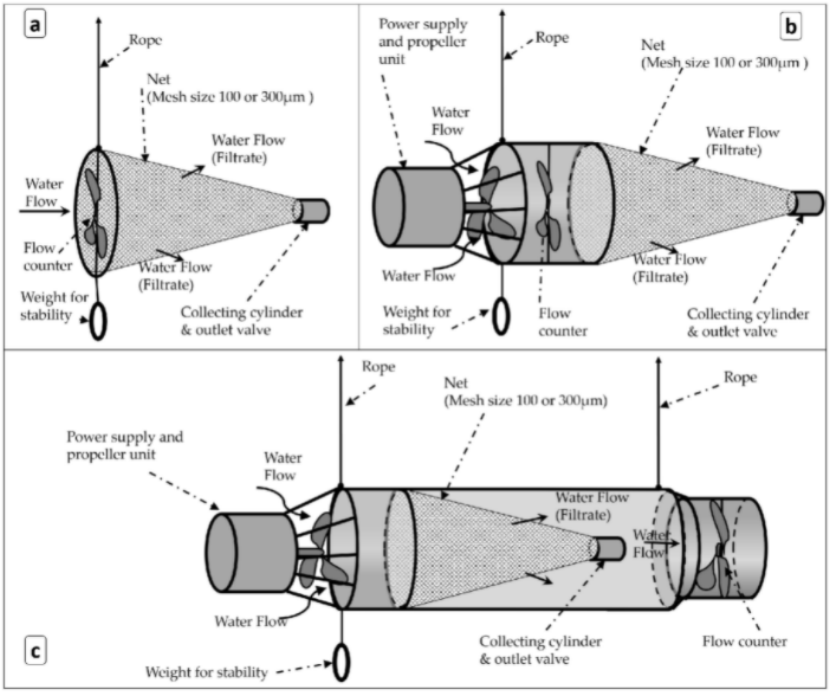


Fig. 5.2. Sampling devices were used at the initial stage for the verification of the performance of Albatross, which was used for the microplastic sampling for the model.

A submersible waterproof power supply unit with a propeller (RDS200, Yamaha, Japan) pumped water into the net. A flow counter (GO-2030 R6, General Oceanics, Miami, FL, US) recorded the number of rotations. The filtered volume was calculated using Equation (1). The plankton net filtered the water and retained the particles larger than the mesh openings

In AM-6, the flow counter was moved to the back of the plankton net. An additional tube was introduced to collect the water passing through the net. This tube governed the water through the flow counter. The impeller diameter of the flow counter (GO-2030 R6, General Oceanics, Miami, FL, US) was set up to be the same as the diameter of the end of the tube (18 cm). Hence, the total flow in the tube was influenced by the counter reading in AM-6, whereas in AM-5, the central flow was mainly counted at the front end of the net. The filtered volume of the AM-6 was calculated using Equation (1). The bridge suspended plankton net (BS-PN) components are illustrated in Fig 4.2 a. In the case of BS-PN, the plankton net was hung on the bridge over the water stream to fully submerge near the surface of the river, as described in detail in Kataoka et al. (2019). The weights hung in each device were to provide stability during the operation (i.e., to keep the device submerged and to reduce unintentional movements with high flow

currents). Both AM-5 and AM-6 were operated for approximately 3 min during this study. However, during the initial optimization period, AM-5 and AM-6 were operated for variable durations from 2 min to 8 min. Yet, considering sample collection duration, battery life, and mesh clogging factors, the suitable operational duration was selected as 3 min. Thus, a 3 min sampling time was applied during this study. The sampling duration of BS-PN was kept between 5 and 90 min. First, the river flow rate was measured, and then the sampling duration was decided to collect an approximate volume of 10 m^3.

The sampling locations and the details of the samples are given in annexure 4. Sampling was conducted in 2018 and 2019 at a total number of 95 (61 riverine and 34 coastal) sampling locations in Japan (Figure 2). All the samples were single replicates. The sampling locations fell into two categories: 1. riverine sampling, rivers where there was no tidal effect, and inland canals connecting rivers and lakes; 2. coastal sampling, with river mouths including bay areas with tidal effects and ports. For the initial comparison study of the three units (AM-6, AM-5, and BS-PN), 11 riverine sampling locations were selected. The selection of 11 locations was based on the availability of facilities such as bridges and access to the location from Tokyo during the initial project stage. No coastal sampling locations were used for comparison, as the BS-PN sampling in coastal zones was unfeasible due to the lack of river flow. All three units were used with 100 and 300 µm meshes separately during the comparison. In the second stage of the study, the sampling locations were selected in both riverine and coastal areas to compare particle size distribution, polymer composition, and morphology of microplastics. Moreover, the sampling locations were selected in different topographical areas in Japan with a wide range of land-use patterns. Application of a single sampling method (AM-6 with 300 µm net) at different locations including challenging conditions to conventional methods was conducted for proper comparison of the above-mentioned parameters. The second stage observations include the 83 sampling locations conducted after the first stage and the initial 11 locations (total 95 locations).

5.2.3. Analytical Process

Microplastics collection from the nets was conducted by rinsing the net with prefiltered water. However, the complete collection of small-sized particles from the 100

µm net was not achieved by rinsing the net (hence, 100–300 µm range was not considered within the scope of the study). The collected samples were pre-purified in the laboratory using a filtered (0.45 µm, glass fiber filter) sodium chloride solution (by dissolving 35.9 g NaCl per 100 mL of water: SG = 1.2) to separate the plastics from the heavier non-polymeric material particles (i.e., suspended sand and clay) collected due to water turbulences. This process was expected to extract only particles with an SG of less than 1.2. This group includes microplastics (i.e., polyethylene (PE; SG 0.92–0.97), polypropylene (PP; SG 0.90), and polystyrene (PS; SG 1.05)) and non-plastic low SG particles (i.e., humus). Microplastics with a higher SG (e.g., polyethylene terephthalate (PET; SG 1.40) and polyvinyl chloride PVC; SG 1.40) are not typically recovered using this process. Yet, environmental plastic samples may present as composites, air trapped, or in weathered conditions. Hence, the SG of the sampled plastic pieces could be altered compared to the SG of the virgin polymer. Therefore, during the polymer analysis process, the attention was not limited to the low SG polymers but included the high SG polymers as well. Then, the floating microplastic pieces were extracted using a vacuum suction respirator (Supplementary Document Figure S6 illustrates this separation process). Next, the organic matter in the suspension was degraded by heating at 60 °C for 8 h with a 2 M HCL solution. Thereafter, the samples were washed with prefiltered water on a 2 µm glass fiber filter to remove any salt precipitations (Supplementary Document Figure S8 illustrates the setup), which would interfere in the next analytical steps. After that, the filtered samples were transferred into glass Petri-dishes with lids. Then, the samples were dried at 105 °C in an oven for 2 h. Finally, the particles were picked up with stainless-steel forceps by visual observation based on their color and shape (Kataoka et al., 2019; Hidalgo-Ruz et al., 2012)

5.2.4. Contamination Control

Contamination control procedures were conducted with great care to minimize the probability of microplastic contamination from outside during sample collection, preparation, and laboratory analysis. Potential contaminations due to the plastic components of the AM-6 and the AM-5 were minimized. Reusing the plankton nets was avoided within a single sample collection visit. When more than one sample was collected within a visit, the net was replaced with a clean net to avoid cross-contamination at

different sampling locations. Nets were cleaned at the laboratory with prefiltered tap water (2 µm glass fiber filter). In AM-6, the pre-exposure to the flow counter impeller was avoided since the counter was placed behind the plankton net. Furthermore, the color, stability, and surface appearance of the three devices used in the study were closely investigated before and after the sampling. There were no missing plastic pieces from the devices identified by visual inspection. Moreover, collected samples were compared with the plastic components of the devices (AM-5, AM-6, and BS-PN) based on color and polymer. The color and polymer type of plastic components of the devices are given in Table S1 (Supplementary Material). The polymer types of the parts of the devices were polyvinyl chloride (PVC), polycarbonate, polyoxymethylene, and nylon. Plastic pieces matching those colors and polymer types were not observed in the collected samples. There were no blank control samples taken during the sampling process.

Glass jars (cleaned with dilute HNO_3 and rinsed thoroughly with deionized water) were used to contain the samples collected for further processing in the laboratory. Glassware and stainless-steel apparatuses were used during the sample processing, and aluminum foil was used to cover the samples to minimize contamination from airborne particles (Rose et al., 2019). In the present study, the considered sizes of microplastic particles were larger than 300 µm. Hence, the probability of airborne contaminations of the same scale particles was very low. A laboratory coat and nitrile gloves were always worn while the work was carried out in an enclosed laboratory. Microscopic examination and Fourier transform infrared spectrophotometry (FT-IR) analysis were carried out in a laboratory designed with sealed windows, and passage through the lab was restricted to minimize airborne contamination, as described by Rose and Webber (2019).

5.2.5. Size Measurement and Polymer Confirmation

Each particle was then processed following three steps. First, the size of each particle was measured by analyzing a picture of the particle taken by a stereoscopic microscope (M165C, Leica microsystems) installed with a USB camera (Leica DFC290, Wetzlar, Germany) using image processing software (ImageJ, downloaded from http://imagej.nih.gov) (Kataoka et al., 2019). The Feret's diameter, the longest distance between any two points along the selection boundary, also known as maximum caliper

and shortest caliper, was measured for each particle (number of particles (N):1811). Second, the shape of the particles was recorded (fiber, fragment, pellet, sheet, or sphere). Third, the compositions of the particles were identified using FT-IR (Nicolet iN10, Thermo Scientific, Waltham, MA, US). The particles were analyzed in transmission mode at a speed of 5 Hz, within the wavenumber range of 4000–600 cm−1. The resulting spectra were processed in the accompanying "OMNIC" spectra software and were compared with the pristine FT-IR spectra of common polymers (PE, PP, PS, nylon, PVC, PET, etc.) as specified in Rose and Webber (2019). Finally, the particle thicknesses were measured using a digital Vernier caliper (SHINWA 19974 digital Vernier caliper with Hold Function; least count 10 µm; Shinwam Niigata, Japan).

5.2.6. Calculations of microplastic concretions

Calculation of the Flow-Volume through the Devices is given in the following equation.

$$V = ab\frac{D^2}{4}\pi \quad (1)$$

V: volume passed through the net,
D: net mouth or tube diameter (for BS-PN: *D* = 24 cm, AM-5: *D* = 22 cm and for AM-6: *D* = 18 cm),
a: flow counter reading,
b: conversion coefficient (manufacturers value for the corresponding impellers; in this case for the impellers used for BS-PN, AM-5, and AM-6: *b* = 5.12),
π = 3.14.

The flow volumes through the devices were calculated using the following equation. Operated average flow rates were calculated by dividing the sampling volume by sampling duration.

5.2.7. Microplastic Particle Size Distribution

Microplastic particle size (Feret's diameters) distribution studies were conducted at 95 sampling locations using AM-6(300). The particle sizes considered for this study were ranged from 0.3 to 5 mm. Particle sizes were divided into size classes with 100 µm

class intervals. The relative frequency of each size class was calculated according to the Equation

$$f_x = \frac{n_x}{N}$$

f_x: relative frequency of microplastic particles in size class x
n_x: number of microplastic particles in size class x
N: total number of microplastic particles

The total number of microplastic particles (N) considered in this study was 1811. For the comparison between riverine and coastal particle size distributions, the particles were divided into two data sets. The riverine data set included 817 particles (Nr = 817), and the coastal data set included 994 particles (Nc = 994). The calculations of relative frequencies of microplastic particles in each data set were done with the modified Equation

5.2.8. Statistical Analyses

Statistical analyses were performed with SPSS (IBM SPSS Statistics, IBM Corp., Endicott, NY, US). A one-way ANOVA test was performed to compare the measured concentrations with different sampling devices. The significant difference between the measured riverine microplastic particle sizes and coastal microplastic particle sizes was tested using an independent samples t-test. The t value calculation is given in Equation.

$$t = \frac{M_r - M_c}{\sqrt{\frac{s_r^2}{N_r} + \frac{s_c^2}{N_c}}} \quad (3)$$

t: the ratio of the departure of the estimated value of mean particle size from its hypothesized value to its standard error
M_r: mean particle size of riverine microplastic
M_c: mean particle size of coastal microplastic
s_r: standard deviation for riverine microplastics
s_c: standard deviation for coastal microplastics
N_r: total number of riverine microplastic particles
N_c: total number of coastal microplastic particles

5.3 Results and Discussion

5.3.1. Operational Conditions of the Sampling Devices

Table 5.1 compares the operated flowrate range, sampling duration, human resource requirement, and filtrate volume measuring accuracy of the three devices. Both AM-5 and AM-6 were operated for approximately 3 min during the comparison period. Based on the water quality, the net clogging rate varied. The operated filtration rate found for AM-5 and AM-6 was between 2.5 and 7.0 m^3/min. Operation at this flowrate for 3 min filtered water volumes ranged from 7.5 to 21 m^3. However, during AM-5 and AM-6 development stages, the devices were operated at variable durations to observe the suitable operating conditions. The operation of shorter to longer durations than 3 min (2 min to 8 min) was possible with both devices. Yet, when the duration became longer, net clogging occurred, and the effective filtration through the net decreased. In the field conditions, for multiple samplings, battery power and time were also limiting factors. With battery power discharge, after several samplings, the pump speed decreased. The operation was limited to five samplings (with 3 min sampling duration) for one fully charged battery.

Table 5.1 Comparison of the operational conditions of the three sampling devices during the comparison stage of the study.

Scenario	BS-PN		AM-5		AM-6	
Parameter	**100 µm**	**300 µm**	**100 µm**	**300 µm**	**100 µm**	**300 µm**
Operated flowrate range (m^3/min)	0.11 to 0.67 [a]	0.17 to 1.00 [a]	2.5 to 7.0 [b]	2.5 to 7.0 [b]	2.5 to 7.0 [b]	2.5 to 7.0 [b]
Sampling duration (min)	15–90	10–60	3	3	3	3
Minimum human resources required (# people)	1–2	1–2	1–2	1–2	2–3	2–3
Effect of net clogging on flow volume measurement	The difference between river flowrate and filtration rate increases. Partial tilt-up the net. Volume measurement errors.		Flow counter at the center moves along with the forced inflow. Volume measurement errors.		Flow counter is located at the filtrate side. It measures only the filtered volume. Minimum volume measurement errors.	

[a]: Operated flow rates in this wide range were due to the river flow velocity variation from sampling location to location, and the observed range of river flow velocities was from 0.25 to 1.5 m/s. [b]: Flowrate through the net was independent of the river flow velocity (depended on the pump flowrate and flow resistance due to net clogging). BS-PN: bridge suspended plankton net.

5.3.2. Microplastic Concentrations Measured with Different Sampling Devices

Microplastic concentrations measured at 11 different sampling locations for comparison of the three sampling devices are presented in Figure 5.3. Concentrations of the 100 µm mesh samples were calculated using only the particle sizes greater than 300 µm for comparison with 300 µm mesh samples. The mean concentration values for all six methods were in the same range (minimum 1.62 and maximum 1.85 pieces/m^3).

ANOVA results indicated no significant difference of measured concentrations with different devices (Supplementary Material Figure S10 provides the ANOVA test output). Yet, there was a slight reduction of the mean values from 100 µm mesh to 300 µm mesh for AM-5 and AM-6. Those differences were 0.04 and 0.03 pieces/m3 for AM-6 and AM-5, respectively. Yet, for BS-PN, the mean value was reduced by 0.03 pieces/m3. From AM-5 to AM-6, the mean concentrations increased by 0.07 and 0.06 pieces/m3 for the 100 µm and the 300 µm mesh sizes. From the BS-PN to AM-6 devices, the mean values increased by 0.23 and 0.16 pieces/m3 for the 100 µm and the 300 µm mesh openings, respectively (12.8% and 9% increments, respectively).

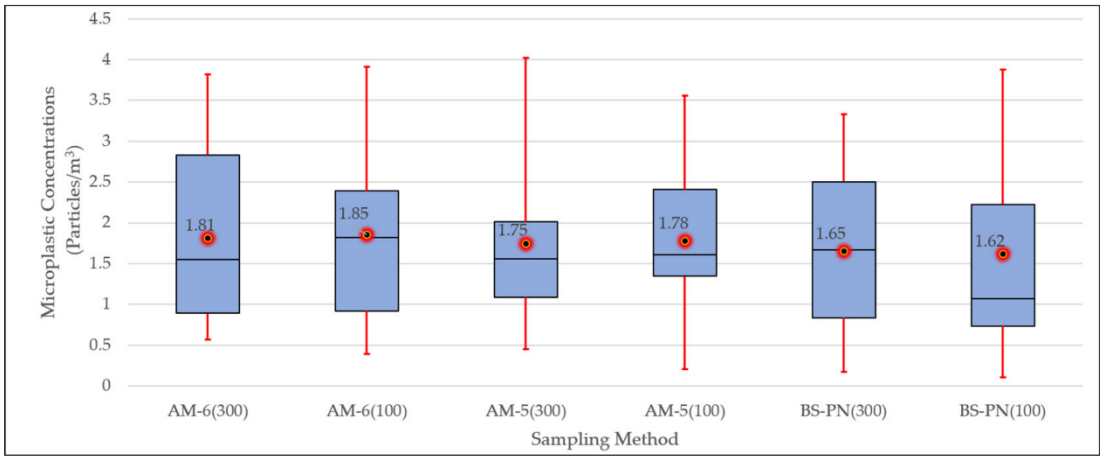


Fig. 5.3. The microplastic concentrations were measured at 11 sampling locations for comparison of the devices (for the nets with 100 µm mesh, the concentrations presented are particles larger than 300 µm size). The top edge of the box is the upper quartile. The bottom edge of the box is the lower quartile. Hence, the box spans the interquartile range. The median is marked by a horizontal line inside the box. The whiskers represent the highest and the lowest observations. The point marker with the number is the mean value.

5.3.3. Microplastic Particle Size Distributions for the Sampling in Japanese Riverine and Coastal Environments Using AM-6(300)

Sampling was conducted at 95 sampling locations using AM-6(300) for the microplastic particle size distribution studies. The distribution of relative frequency of microplastic particles in each size class is illustrated in Figure 5.4 a. Figure 44. b compares riverine and coastal microplastic particle size distributions. Apart from the

distribution per size class, the cumulative size variations are illustrated in the same graphs (secondary axis). The relative frequency of particle size class increased monotonically as the sizes decreased up to the range of 1–2 mm. Thereafter, a rapid reduction in the frequency of particles per size class occurred. Generally, larger-sized plastic litter gradually degrades and breaks into smaller pieces in water environments. Therefore, the monotonic increment up to a 1–2 mm range is acceptable. The particle frequency reduction with a further reduction of size class shall be understood with the sampling procedure and the particle shape. The sizes measured and expressed in this study were the longest length (longest Feret's diameter) of the particle (selected microplastic pieces observed in this study are given in Figure 4.5).

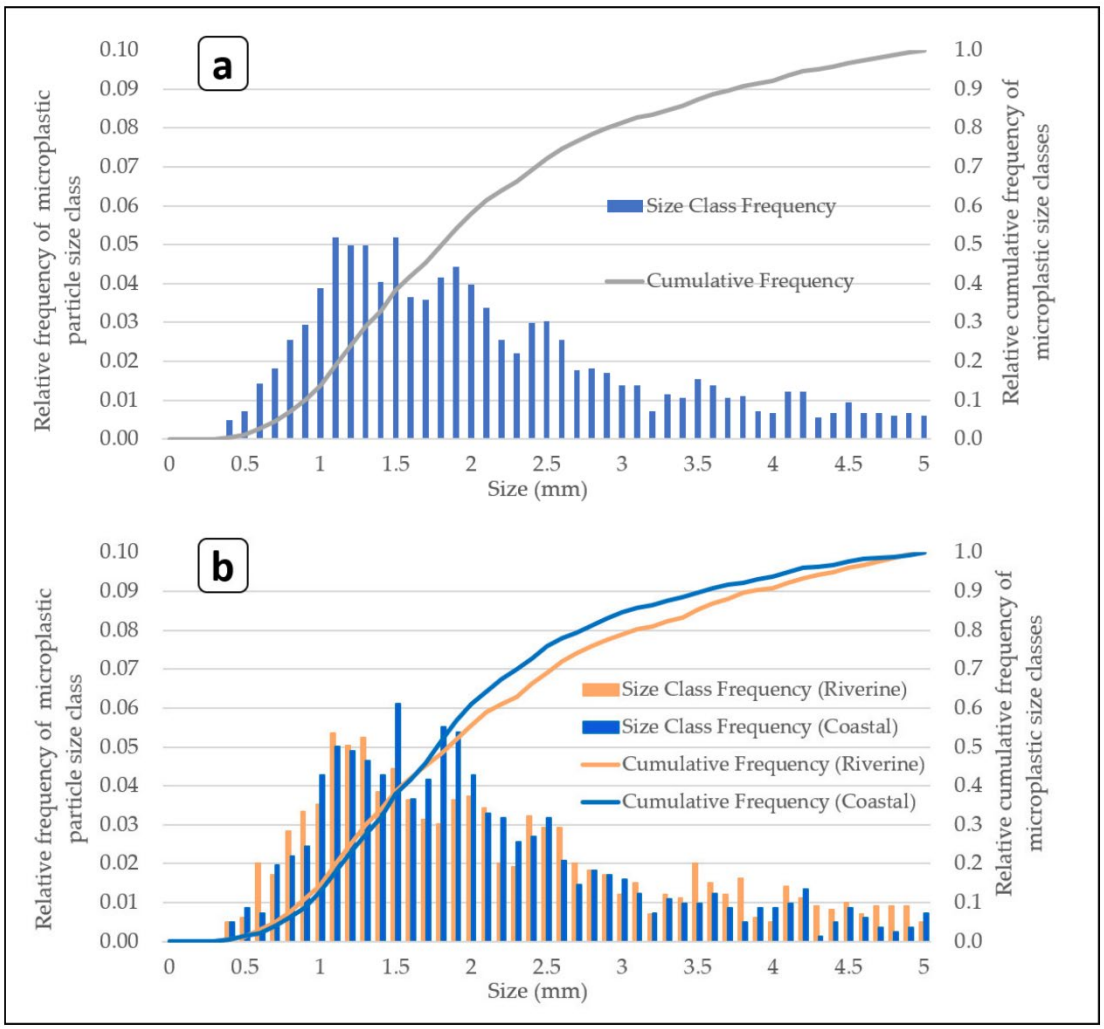


Fig. 5.4. The relative frequency of particle size class distribution (N = 1811) of the microplastics was collected at all survey sites using AM-6(300). (a): The distribution of

total sampling locations. (b): The distribution separately plotted for riverine environments and coastal environments (a comparison of percent study and East Asian seas surrounding Japan (Isobe et al., 2015) is given in Annexure 4 Fig. A4.2).

The t-test revealed a statistically significant difference between the mean diameter of riverine microplastic particles (Mr = 2.103 mm, sr = 1.133) and the mean diameter of coastal microplastic particles (Mc = 1.981 mm, sc = 0.999). The observed t = 2.389, p = 0.017, with the significance level (α) = 0.05. Riverine and coastal microplastic particle size distribution (Figure 5.4b) indicated a statistically significant reduction of the particle sizes from riverine environments to coastal environments.

5.3.4. Plastic Polymer Compositions in River and Coastal Areas of Japan

Fig. 4.5a indicates the polymer composition distribution of the microplastics collected. The dominant polymer types observed in the floating and the suspended microplastics collected in this study were PE (48%) and PP (40%). Yet, PS, PET, and other kinds of polymers were also detected in small amounts. The PE percentage decreased from riverine environments to coastal environments. However, the PP percentage increased slightly from riverine environments to coastal environments. The SG values of PE, PP, PS, and PET were 0.92–0.97, 0.90, 1.05, and 1.40, respectively. The sampling was carried out near the surface of the water body. The rotation of the propeller pumped the water into the net from the surroundings. This included the water from the surface (floating) and from inside (suspended in) the river. Hence, the sampled microplastics were likely to consist more of lower SG polymers.

Compared to the conventional BS-PN sampling device, the developed devices used in this study (AM-5 and AM-6) could collect samples in smaller canals, places without suitable structures to fix the nets, locations with lower river flowrates, locations with rapid water level variations, etc. (Supplementary Material Figure S1 indicates several locations where conventional sampling methods are hardly applicable. We describe these types of conditions as challenging conditions for conventional sampling). Both sampling devices, AM-5 and AM-6 were capable of sampling microplastics in challenging aquatic environments, such as rivers, bays areas, canals, and coastal areas, despite the flow rate of the water body or the space availability for a boat traveling. During the comparison stage of the study, sampling at these kinds of challenging locations was

not possible due to the inapplicability of BS-PN. However, with AM-6 during the second stage of the study, sample collection was conducted. This was one basic advantage of the developed sampling devices.

A sampling of larger volumes (from 7.5 to 21 m3) within a short period (3 min) is an advantage compared to the studies reported with smaller sample volumes (Bordos et al., 2019; Kapp and Yeatman, 2018). There were several advantages of sampling with AM-5 and AM-6 in terms of the sampling duration. Water level variations create errors in sampling due to partially submerged nets. Depending on the water quality, the net clogging rate varies. Furthermore, there are operational difficulties with BS-PN, such as the decision regarding the sampling duration. When maintaining a longer time, if the net becomes clogged, the flowrate measurements could be inaccurate. The effect of net clogging due to longer sampling durations was reported in riverine environments. Sampling volume calculation errors due to partially submerged BS-PNs were reported (Kataoka et al., 2019).

If the BS-PNs lifts within a short period, the filtered water volume might be not enough to obtain a representative sample. On the other hand, if the BS-PNs are kept for a longer time in the river, the net might become clogged, and a back-flow situation may occur. For increased sampling durations such as 5 min, this net clogging, and back-flow situation occurs with the developed devices (AM-5 and AM-6) as well. The reduction of mesh opening size from 300 to 100 µm makes this net clogging and back-flow impact higher (numerical data not given; authors observed this during the sampling). This back-flow creates a situation where the water passes through the flow counter, which is located at the center or next to the net. Yet, part of the water passing the flow counter returns without passing through the mesh. Observations during the study indicated several occasions where the water level of the stream varied within 10–20 min due to the upstream conditions.

Moreover, with the progressive clogging of the BS-PN, the resistance to the filtration flow increased. Hence, the upper side of the net frame tended to lift above the river surface. This net lifting overestimated the filtered water volume because the water flow was not completely across the mouth of the net. This was not observed with the developed sampling devices. We assume the pump operation kept the device submerged

(the pump is continuously sucking the water). Moreover, even with the BS-PN, by providing enough counterweight, the net lifting can be controlled. This includes the improvement of the strength of the net mouth to withstand the forces. This may provide an improved BS-PN with fewer potential volume measurement errors due to net lifting.

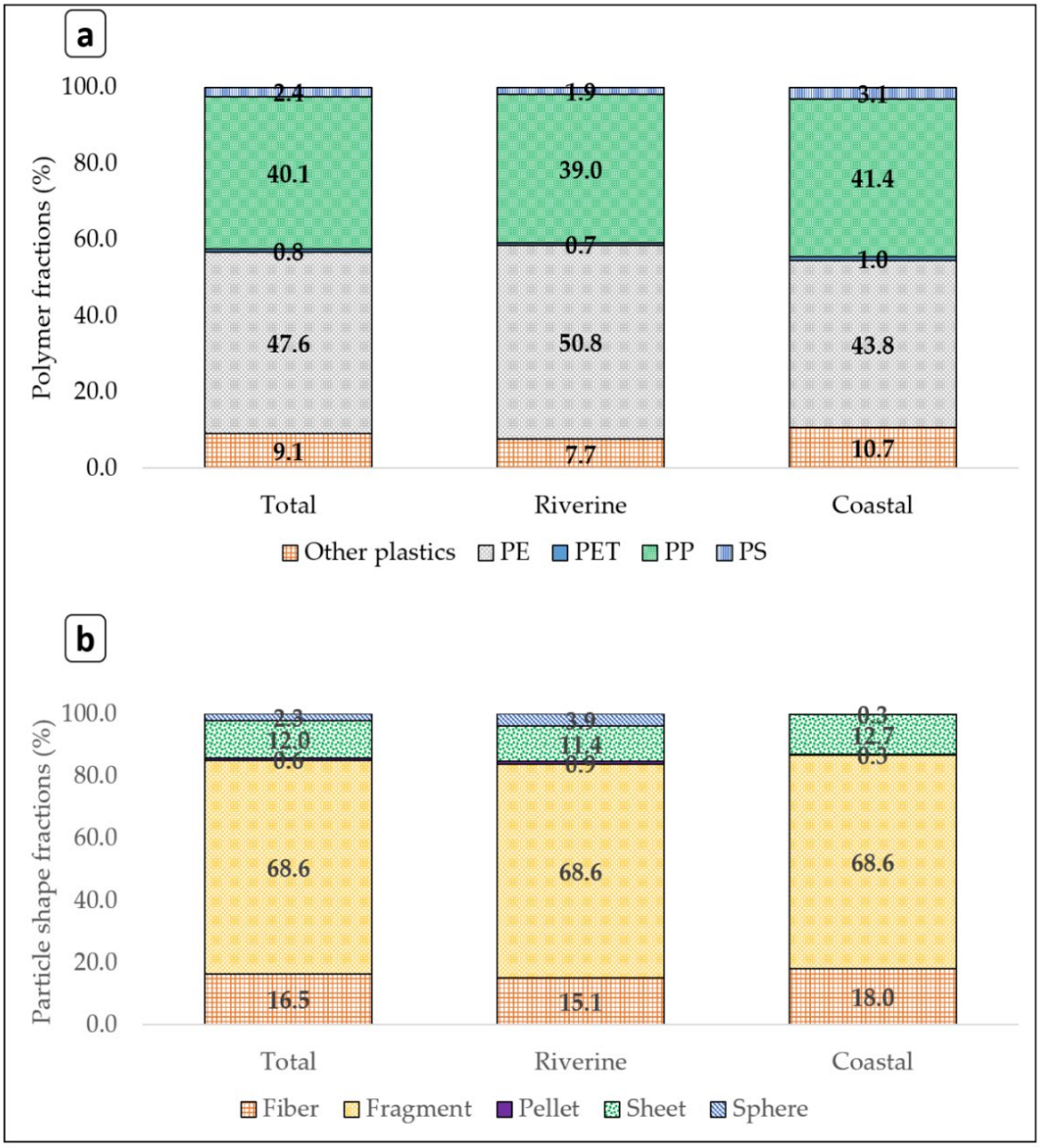


Fig. 5.5 The morphology and composition of microplastic samples from 95 sampling locations by AM-6(300). (a): Polymer composition comparison. (b): Fractional particle shape comparison (percentile values of each component are labeled).

There are special research objectives where the sampling must be done during floods, rainfall events, etc., where water level variations occur. AM-5 and AM-6 are applicable during these kinds of situations, as the method is applicable under varying water levels. As the sampling time is 3 min (or less), the collection of samples at a variable river within a short period is possible, and the evaluation of such kinds of variations is also possible. With AM-5 and AM-6, the depth of the sample collection location is adjustable during the sampling process. Hence, making the samples represent a larger section of the water body can be achieved by changing the sampling location in horizontal and vertical directions.

Certain methods, such as suction pump-based methods, are inoperable where the access from riverbanks and trawls/boats is challenging. When the overhead bridges are too tall (such as move than 10 m), suction pumps are inapplicable. Among the sample collection locations, the sample location (Fig. A4.1) located at the bridge over the river had a height of more than 12 m. These places are highly challenging for methods such as suction pumping due to the limitations of suction pressure.

Considering the human resources for the sample collection, BS-PN, and AM-5 can be operated even with only one person. Yet, AM-6 requires a minimum of two people to operate. However, usually during water sample collection research activities, due to safety issues, more than one person is participating. Hence, the available human resources for the operation of all three devices are comparable. However, the sampling equipment handling could be slightly negative from the researchers' perspectives with the AM-6 compared to AM-5 and BS-PN. Yet, sample collection duration was reduced with both the AM-5 and the AM-6 compared to the BS-PN. Compared to the previously established methods such as trawling a manta net (Zhang et al., 2020;), which requires both humans and support with boats, the devices discussed in this study can be handled with fewer human resources and supportive services.

5.3.5. Microplastic Concentrations Measured with Different Sampling Devices

Smaller-sized mesh openings (100 µm) were expected to capture a higher number of microplastics than the larger (300 µm) mesh. This higher capture was expected in the considered range of particles from 300 µm to 5 mm. Fig. 5.6 illustrates the higher

potential of particle retention with 100 µm compared to 300 µm mesh within the considered range of particles. Comparison of particle size distributions of the 100 and the 300 µm meshes show that there is a higher frequency of particles in the smaller sized range for 100 µm mesh compared to 300 µm mesh. This supports the retention mechanism expressed in fig 4.6. However, the observations did not indicate such a tendency. The microplastic concentrations measured at 11 locations did not indicate an advantage of using a 100 µm mesh-sized net. A possible reason for this could be associated with progressive net clogging and back-flow. This clogging is faster in smaller mesh openings since smaller meshes retain a larger range of suspended matter in water. This situation creates a faster back-flow situation. With back-flow, some of the microplastics could escape from the net. Moreover, the retention probability could reduce. However, the present study did not confirm the reason. Further studies are to be done to confirm this phenomenon.

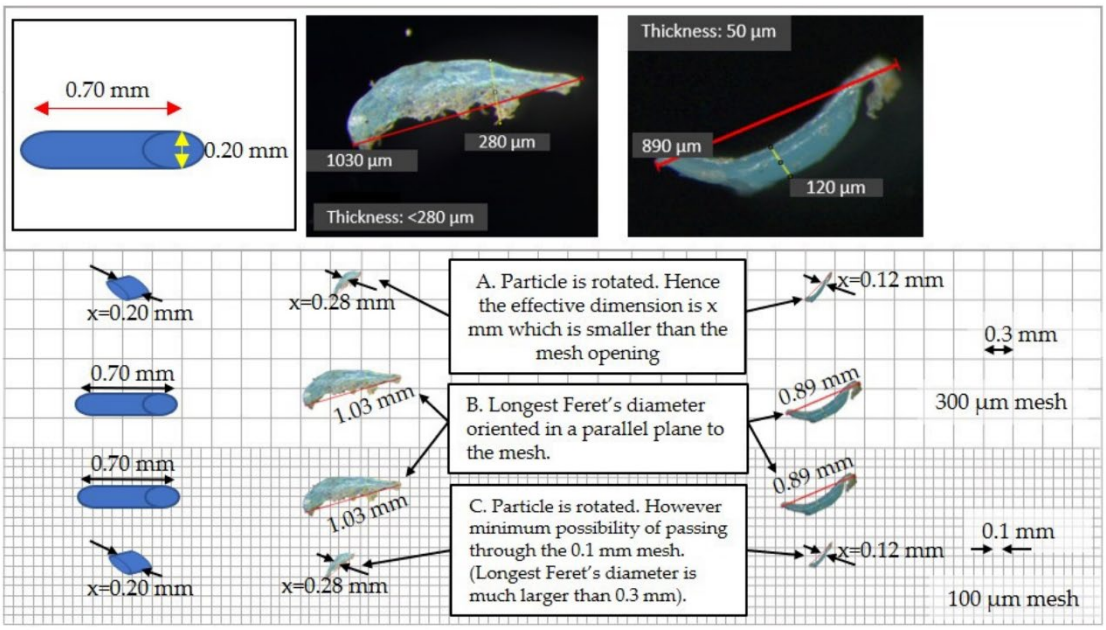


Fig. 5.6 Effect of particle orientation on the retention by 100 and 300 µm meshes.

A study conducted with BS-PN (Kataoka et al., 2019) reported volume measurement errors of 5%–10% due to the partial submerged net frame during the sampling. Partial submergence occurs with the increased force from river flow due to net clogging. Hence, the filtered water volume in the present study may also have been overestimated due to

the same reason. This could be a reason for the lower estimation of microplastic concentrations with the BS-PN sampling. Therefore, the reported lower microplastic concentrations with the 100 μm net could be due to net clogging. There were practical difficulties of 100% recovery of samples from the 100 μm net due to the attachment of the plastic particles to the net. However, the optimum size of the mesh depends on the objectives of the research. For example, there is capable analytical equipment for smaller microplastics, and risk assessment-related studies require smaller-sized particle sampling. Hence, there are limitations of applications of the devices developed in this study for the range of 1–100 μm sampling. In such cases, the common practice is to use grab sampling methods and laboratory sifting with a series of sieves.

Microplastic concentrations in marine environments vary by three orders of magnitude, whereas in freshwater resources, including riverine environments, the variation is five orders of magnitude (Besseling et al., 2019). The review study of Koelmans et al (2019) reported a variation of microplastic concentrations in riverine environments over a large range. These orders of magnitude differences in the two environments might be due to better mixing and/or representativeness of the sampling in the marine environment with less mixing and a larger variability of litter sources and flow conditions for freshwater environments. Besseling et al (2019) described the positive impact of sampling with a trawling net over several hundreds of meters in open water over the pointwise grab sampling with a bucket or flask. This is an advantage of trawling net sampling due to the improved representativeness over pointwise grab sampling. This advantage is applicable where trawling is possible. However, the accuracy of the sample volume and other practical situations need to be considered.

The concentration values reported in this study are within the values reported in the previous studies. However, the comparison of concentrations in different environments requires a comprehensive analysis of various aspects. Hence, correlating the observed concentrations in different environments to the sampling devices' efficiency is inappropriate. There was only one study found in the literature for Japanese riverine environments (Kataoka et al., 2019). This study indicated 1.6 ± 2.3 pieces/m^3, which is within the same range of the values observed in this study.

The particle distribution indicated a similar variation pattern with the reported microplastic particle size distributions in freshwater (Kataoka et al., 2019) and oceanic environments (Isobe et al., 2015; Jung et al., 2018; enders et al., 2015).

The frequency of size class was getting smaller from the river to the ocean. This is the first reporting of the comparison of the size distribution of microplastics in both riverine and coastal environments. Plastic litter degradation occurs in riverine environments as well. Hence, the plastics could break into smaller particles from the river to bay areas. Even though rivers are not the only microplastic source to the oceans, a potential reason for the significant particle size reduction reported in this study could be due to degradation during the river flow and the additional degradation in the coastal areas. Comparison of the size distribution values of the East Asian seas surrounding Japan (Isobe et al., 2015) with the values reported in this study indicated a progressive size reduction from rivers to the ocean. This could be considered as supportive evidence of the size reduction (fragmentation) of microplastics in water environments (from rivers to oceans).

The study of Isobe et al (2017) reported that the dominant polymer types of microplastics reported floating in East Asian seas (seas around Japan) were PE, PP, and PS. Kataoka et al (2019) also reported PE, PP, and PS as the dominant microplastic polymer types identified in Japanese riverine environments. The above two studies and the present study were focused on floating and suspended microplastic samples. Considering the SGs of the plastic polymers, it is understandable that the plastics with lower SG than water were abundant in water suspensions. However, the litter sources were also a governing factor to decide the polymer composition. The highest percentage of PE and PP are used in the packaging industry (over 60% and 49%, respectively), while PS is commonly used both in packaging and building construction (30% and 29%, respectively [9]). Materials used in items that last several decades, such as in buildings and construction with alternative disposal methods, unlike PE and PP, have a lower probability of entering water environments. That could be the reason for detecting less PS compared to PE and PP in this study.

The PE fraction decreased from riverine to coastal environments, while the PP percentages increased slightly (numerical percentage decreased from 50.8 to 43.8%). This

could have been due to several reasons, such as degradation rate variation or litter input variations in riverine and coastal environments. A weight-based concentration (mg/m^3) analysis would provide a better understanding of the litter input characteristics rather than a particle number-based concretion (particles/m^3) analysis.

Even though the SG of PET is about 1.4 (higher than the SG of freshwater), some samples included PET particles. In moving water, microplastics are subject to turbulent transport, settling, aggregation, biofouling, resuspension, and burial (Lambert et al., 2018). This results in particle movement within and between the water for different compartments of riverine environments (Lambert et al., 2018). This explains the potential reason for a PET presence in suspension, as the river flow turbulences could bring the higher SG particles into the suspension. In a water environment, the particles are associated with air bubbles and biofilms. Hence, the SGs could be altered from the SG of pure polymers. Hence, the presence of PET floating and in suspension could be understood.

Considering the structure of the sampling device (net with a 300 µm mesh opening), fragments and spheres had the least possibility of escape. However, the shapes including fibers and sheets could escape from a square-shaped opening even if the maximum dimension exceeded 300 µm. The morphology of microplastics is important in the recognition of the pollutant source (Rochman et al., 2019). The microplastic shape distribution in freshwater in Italy showed the dominating occurrence of fragments [73.7%; (Sighicelli et al., 2018; Campanale et al., 2020)], which agrees with the present study. As previously mentioned, shape influences the retained particles within the net. Fiber-like microplastics and sheet-like microplastics tend to escape more compared to pellets, spheres, and fragments. Considering the morphology, the study of Dris, et al (2018) discussed that targeting water environmental sampling of fibers required a smaller mesh opening than for other shapes. The use of an 80 µm mesh had a 250-times higher probability of sampling fibers compared to a 330 µm mesh. Hence, the detected fiber composition could be less than the actual relative presence in the water in this study.

5.3.6. Area mass relationships

Area mass relationships were developed for ten major categories of microplastics. The table provides the conversion equations and the coefficient of determination for each

case. The correlation between measured mass and calculated masses of microplastics could be affected due to several reasons. The volume estimation is conducted by using the measured surface area and thickness. The surface area is a two-dimensional projection and the thickness is an in most cases the largest thickness. Separation of the microplastics according to the shapes has increased the correlation. Moreover, the shapes such as spheres (identified as controlled release fertilized capsules), the similarity within the category are high. Hence the coefficient of determination (R2) is closer to 1.0 indicating a higher fitting. Figure 5.6 indicates the relationships of calculated mass and measured mass for different polymer and shape categories.

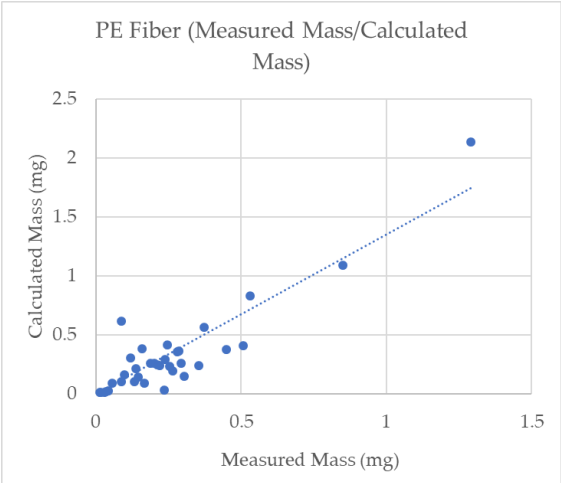


Fig. 5.6a measured mass vs calculated mass relationship for PE fiber

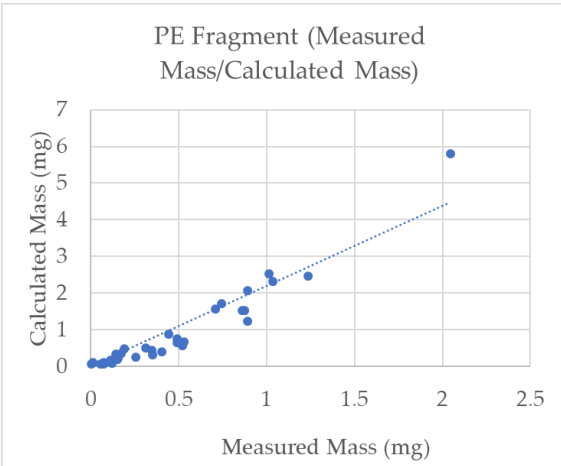


Fig. 5.6b measured mass vs calculated mass relationship for PE fragment

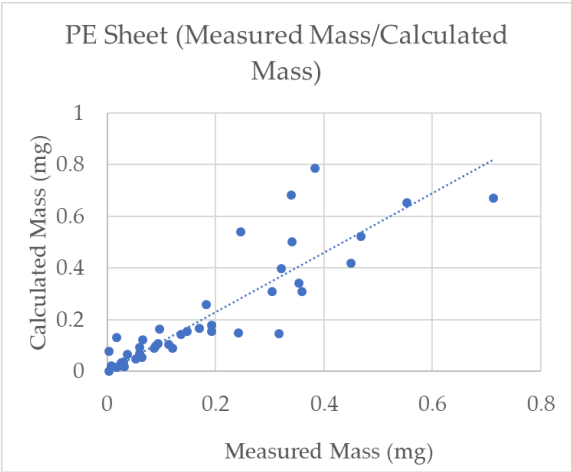


Fig. 5.6c measured mass vs calculated mass relationship for PE sheet

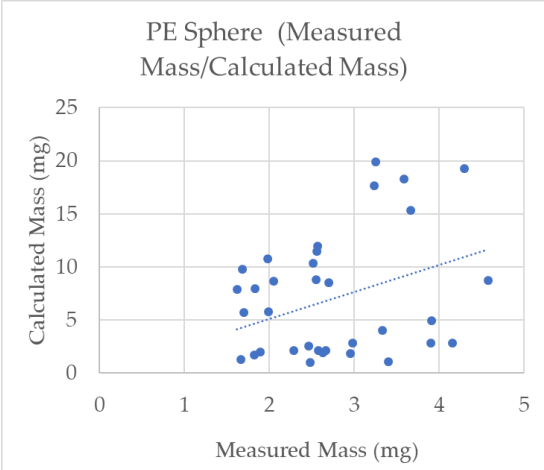


Fig. 5.6d measured mass vs calculated mass relationship for PE sphere

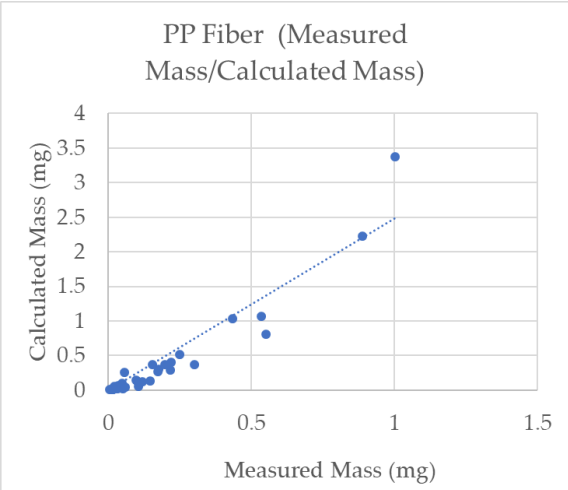


Fig. 5.6e measured mass vs calculated mass relationship for PP fiber

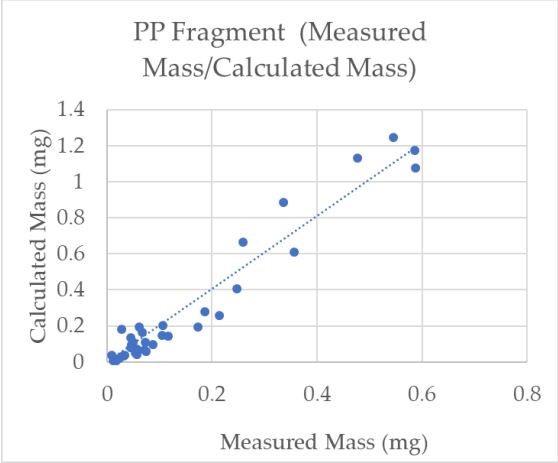


Fig. 5.6e measured mass vs calculated mass relationship for PP fragment

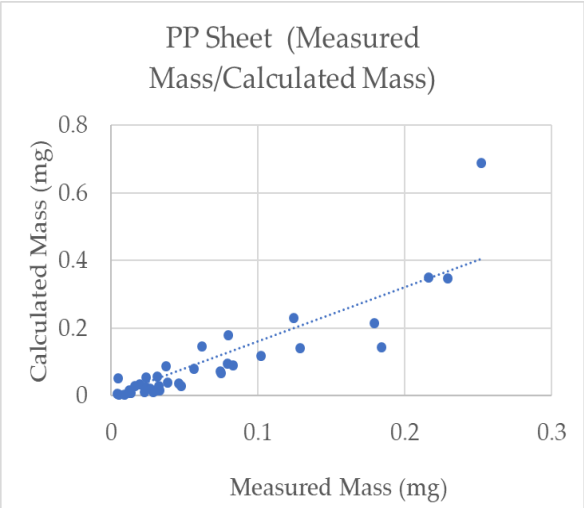


Fig. 5.6g measured mass vs calculated mass relationship for PP sheet

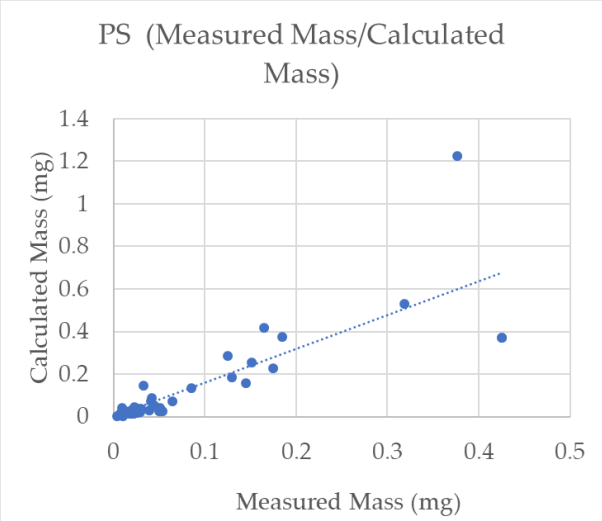


Fig. 5.6h measured mass vs calculated mass relationship for PS Sheet

Figure 5.9 illustrates the relationships of measures mass and projected surface area of the microplastic different polymer and shape categories. The projected area has been measured by image-j. Figure 5.7 illustrates the simplified process of measuring the projected area of microplastics. Apart from manual area demarcation, software-based (programed).

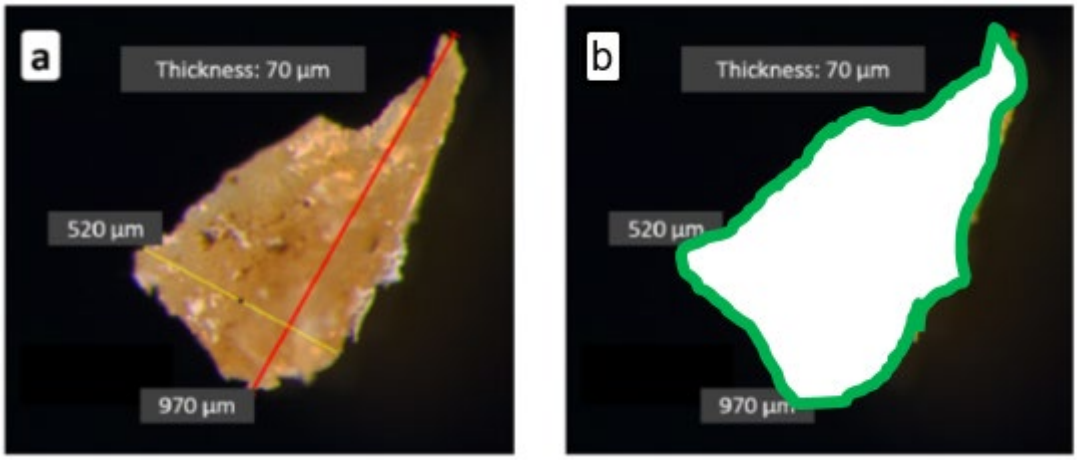


Fig. 5.7 Micrpasltic projected surface area measurement using Image-j

The program has been tested with the manually measured areas. Figure 5.8 provides the relationship between manually measured area and program measured area. The accuracy of the programmed area measurement has been verified with this observation and the observations indicate an acceptable relationship indicating the accuracy of the program is acceptable for continue.

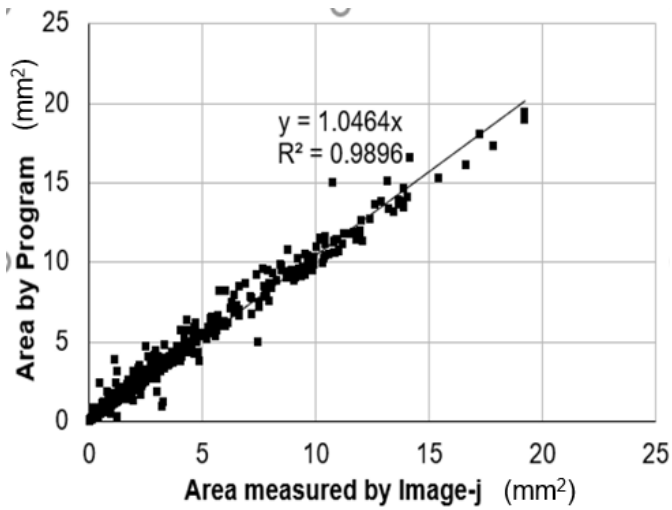


Fig. 5.8 Area by program and area by Image-j: Comparison

Figure 5.9 indicates the relationships between the measured projected areas and measured masses for different polymer categories and shapes. The objective is to overcome the mass measurement challenges and increase the data set quantity by using the un-mass measured data of microplastics.

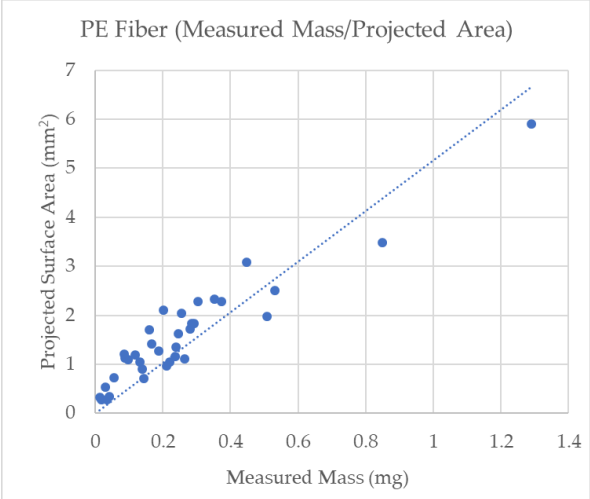


Fig. 5.9a Measured area vs measured mass relationships for PE fiber

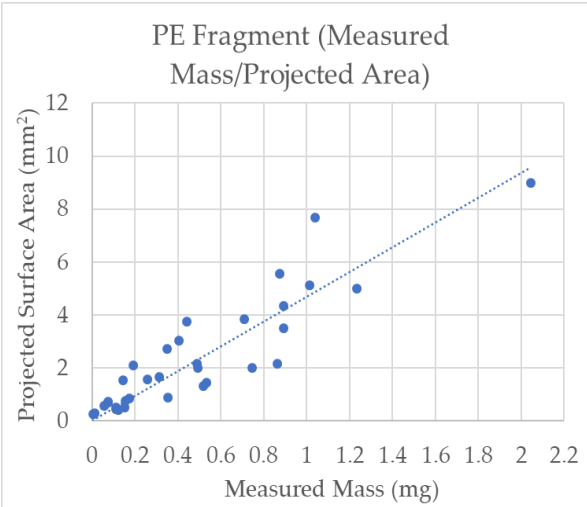


Fig. 5.9b Measured area vs measured mass relationships for PE fragment

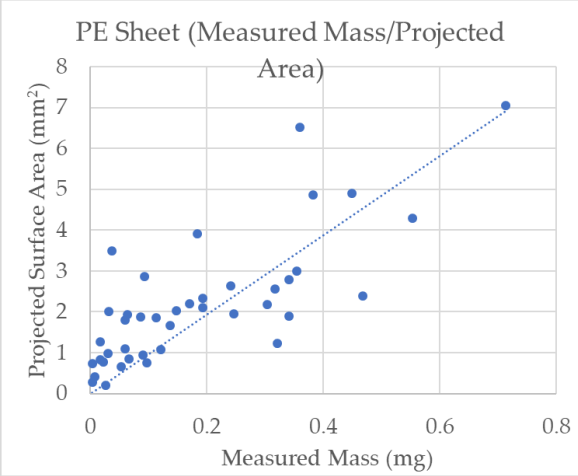


Fig. 5.9c Measured area vs measured mass relationships for PE sheet

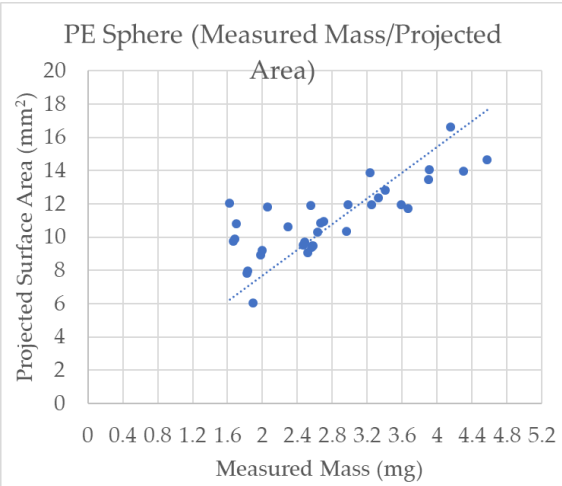


Fig. 5.9d Measured area vs measured mass relationships for PE sphere

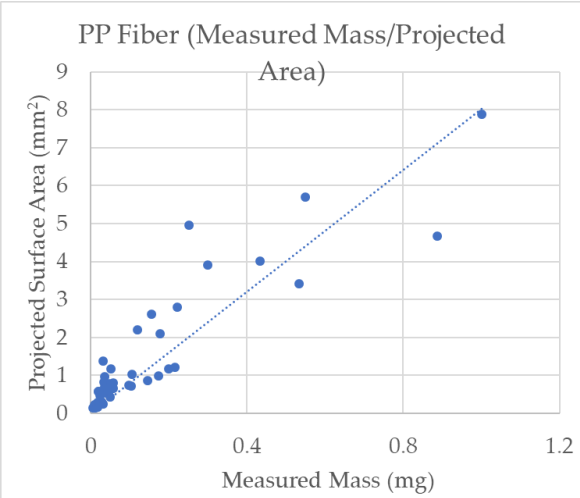


Fig. 5.9e Measured area vs measured mass relationships for PP fiber

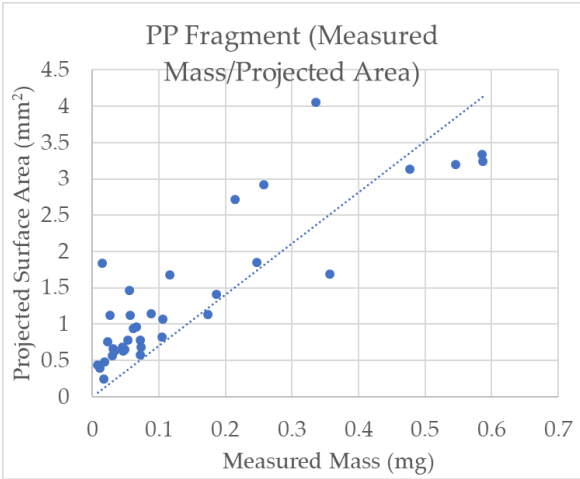


Fig. 5.9f Measured area vs measured mass relationships for PP fragment

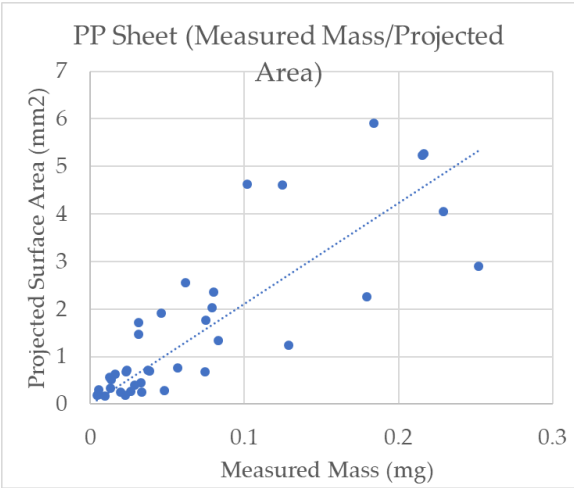


Fig. 5.9g Measured area vs measured mass relationships for PP sheet

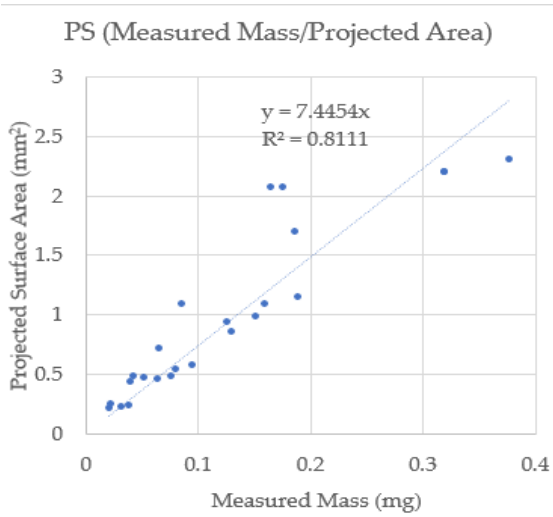


Fig. 5.9h Measured area vs measured mass relationships for PS (SPS)

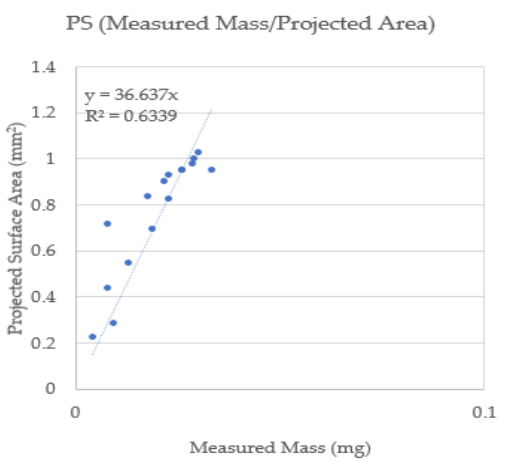


Fig. 5.9i Measured area vs measured mass relationships for PS (EPS)

5.3.7. Validation of estimated microplastic mass

Nine sampling locations were selected for the validation of the estimated masses. Fig. 5.10 illustrates the relationship between the measured masses of each sampling location and the corresponding mass estimations. Correlation analysis for measured sample masses and estimated sample masses indicates a high coefficient of determinations. This indicates that the obtained mass area relationships provide reasonably accurate overall mass conversions which ultimately gives accurate sample mass estimations.

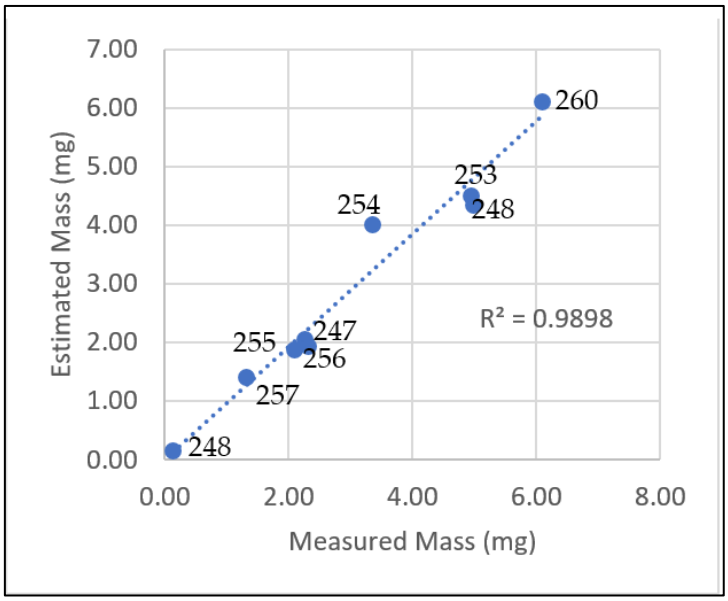


Fig. 5.10 The validation of estimated masses using 9 sampling locations

5.3.8. Water balance analysis

Annual precipitation was equal to the sum of the annual evapotranspiration, surface runoff, and underground infiltration, and the water volume was balanced within each grid. Only a water balance analysis, in which each grid was considered closed, was performed. Surface runoff and underground infiltration required for material transport could be calculated

Table 5.2 The rainwater infiltrations according to land use patterns (adapted from Nihei, 2021)

Land Use			E	Q_s	Q_i
Major	**Details**			**Coefficient f**	
Forest	Forests		Equation (1)	0.5 (Quaternary volcanic rock) 0.8(Other)	Equation (6)
	Bushes		Equation (2)	0.3	Equation (6)
	Mountainous bushes		$P - Q_s$	0.95	0
Agriculture area	Paddy fields	Irrigation	Equation (3)	0.8	Equation (6)
		No irrigation	Equation (2)	0.3	Equation (6)
	Other		Equation (2)	0.3	Equation (6)
Urban area	Building sites	Infiltration area	Equation (2)	0.3	Equation (6)
		No-infiltration area	$P - Q_s$	0.95	0
	Road, railways, and others		$P - Q_s$	0.95	0
Other	Golf courses		Equation (2)	0.3	Equation (6)
	Rivers and lakes		Equation (4)	$P - E$	0

Surface Runoff and Underground Infiltration

$$Q_s = f \times P$$

f is the runoff coefficient and P is precipitation

$$Q_i = P - E - Q_s$$

5.3.9. Floating and suspended microplastic emissions into the water from land

Table 5.3 provides the equations obtained and microplastic emissions (floating and suspended microplastics) from the land to water environments. The 2020 data provides the annual emission of 157 MT microplastic from Japan. The differences of 2019 could be due to the COVID-19 impact. However, the sampling locations have differences from 2019 to 2020. Hence deriving a conclusion on this is difficult due to several variables in the two datasets of 2019 and 2020.

Table 5.3 Results of the microplastic emissions according to the output of the GIS-based model

MP type	Equation		Mass (MT/Year)
Total	Linear 2020 data	MP = 0.0239UR + 0.6547	157
	Linear 2019-2020	MP = 0.0245 UR	103.4
Artificial Turf (AT)	Linear 2000	AT = 0.0058UR	24.5

UR: Urban ratio (Urban land area/total land area)

Fig. 5.11 and 5.12 provide the annual floating and suspended microplastic emissions at the prefectural level, and per-capita emissions. Highly urbanized prefectures such as Tokyo, Kanagawa, and Osaka emissions are comparatively higher than the other prefectures. The per-capita microplastic emissions provide the perspective of the microplastic leakage into water environments from land. The observations are listed according to the population density increasing order.

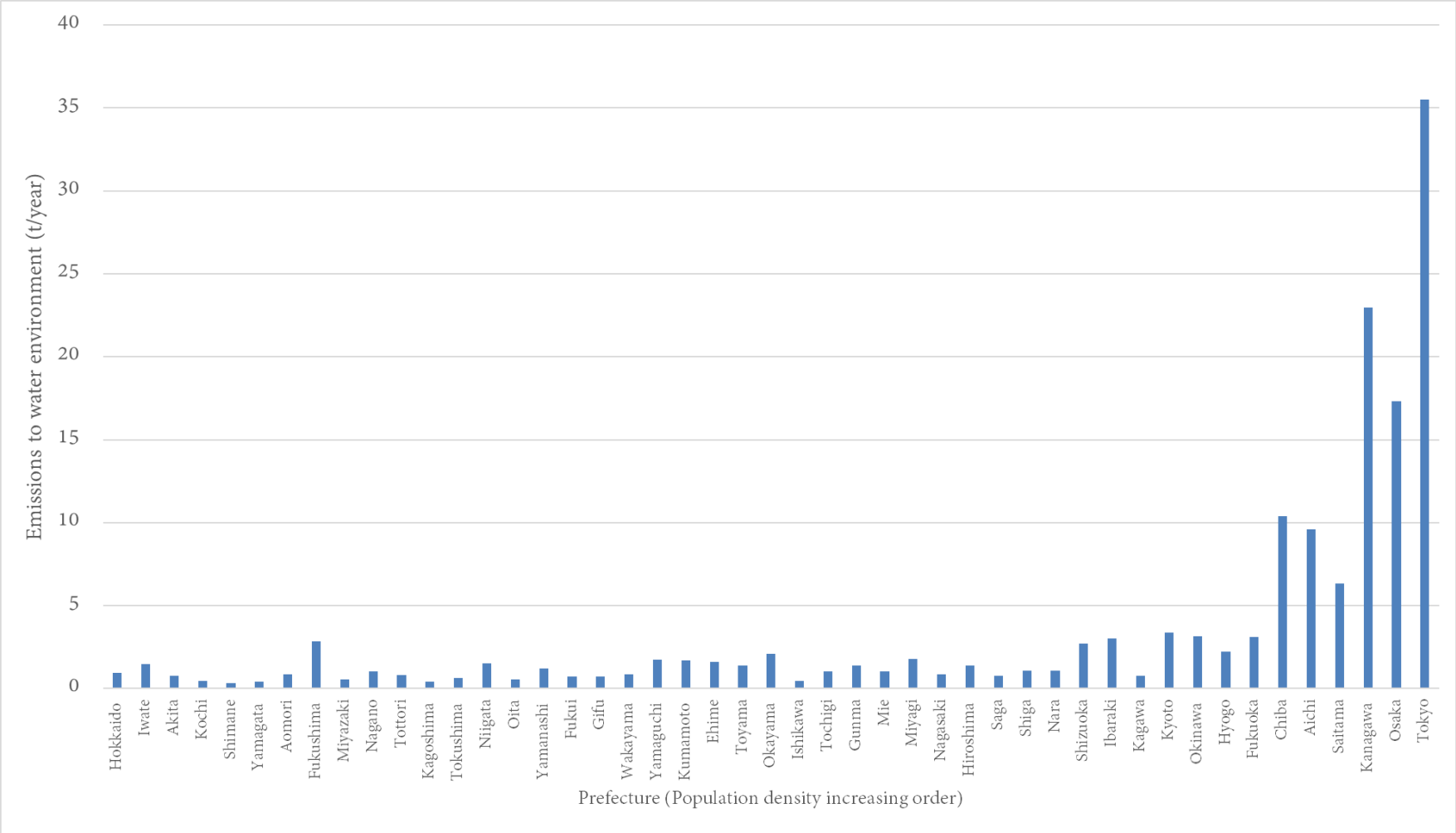


Fig. 5. 11 Annual floating and suspended microplastic emissions at prefectural level (2020 data)

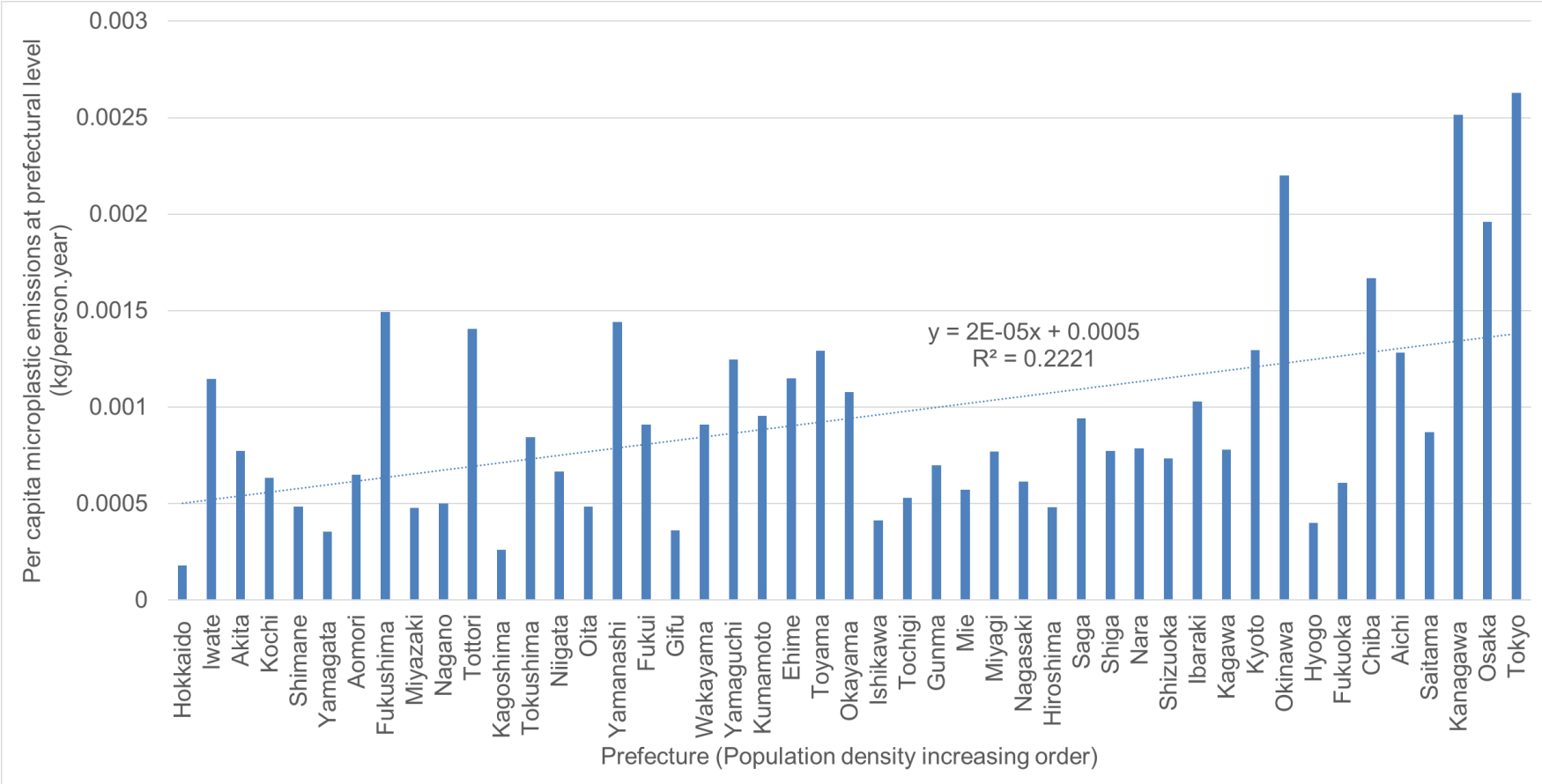


Fig. 5. 12 Annual floating and suspended microplastic emissions per person according to prefectural level (2020 data)

5.4 Conclusions and Recommendations

The microplastic concentrations and compositions found were in line with the conventional samples where comparison was possible. The observations supported a significant particle size reduction from rivers to coastal areas. PE and PP were the dominant microplastic polymer types floating and in suspension in the riverine and coastal water environments of Japan. The area mass relationship for the microplastic mass estimation was developed and tested. The results confirm that the developed relationships provide higher accurate sample mass estimations. The GIS-based model developed, provided 157 t/year microplastic emissions from land to water environments in 2020 year. The prefectural level estimations indicated higher emissions from Tokyo, Kanagawa, and Osaka. The per-person observations indicate up to 25 g/year microplastic emissions in Japan. However, this model is valid only for the floating and suspended microplastics (and mostly the non-fiber type microplastics) due to the sampling methods limitations such as 300 µm mesh size and the mesh-based sampling limitations allowing more fibers to pass through the nets.

Further improvements can be done with the accumulation of more microplastic sampling data (which is ongoing by various groups) and diversifying the sampling methods to expand the types of microplastics representing the model inputs.

CHAPTER 6. TOXIC NATURE OF MICROPLASTIC AND ANALYTICAL METHODS

6.1 Introduction and Literature Review

The process of the sampling and the scenario of the study are illustrated in figure 6.1.

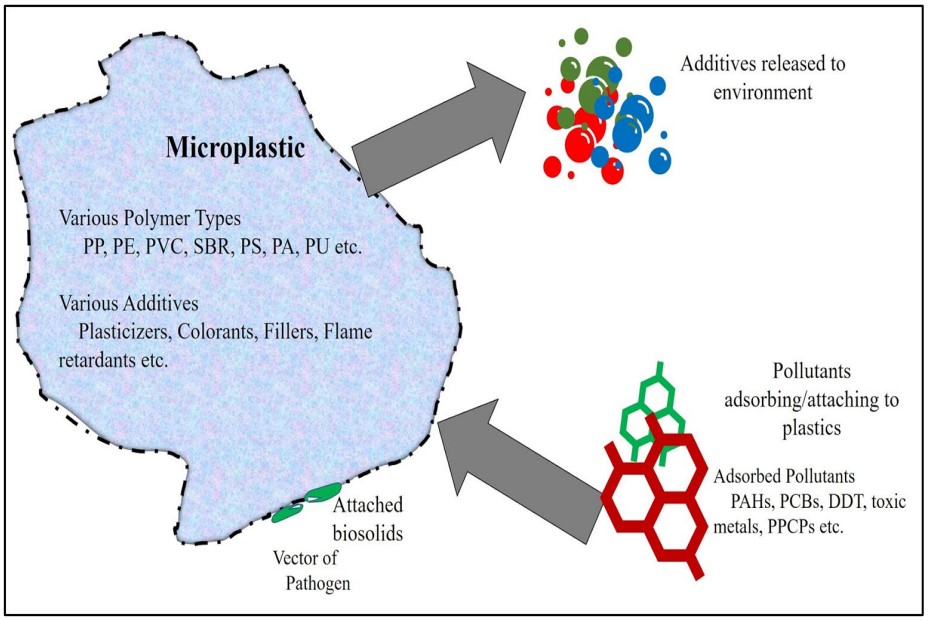


Fig. 6.1 Microplastic as a complex suite of chemicals

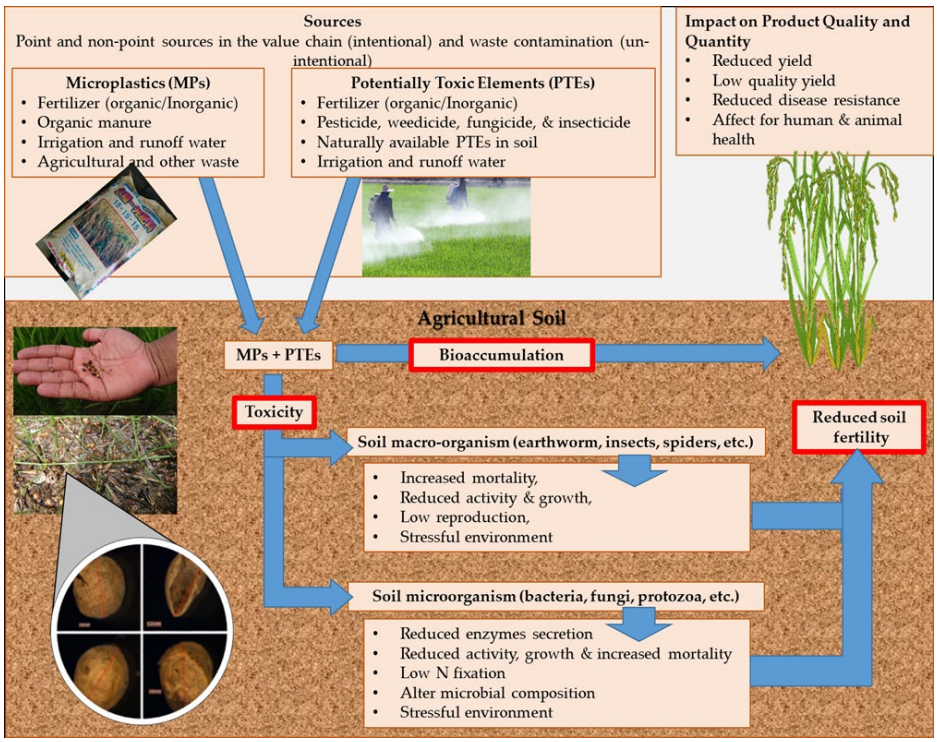


Fig. 6.2 Potentially toxic element (PTE) effects and potential contaminations

Table 6.1. Features of analytical equipment.

Features	***µ-Raman***	***µFT-IR***	***ATR-FT-IR***	***Microscope-based***	***SEM***	***Pyrolysis GC-MS***
Possible equipment price range*	3		2	1	5	3
	Prices vary from USD 20,000 to 1,000,000. The ranking is done from 1 to 5 based on relative price (lower number = cheaper equipment).					
Type of polymer	Yes				No	Yes
Detectable additives	Pigments	No				Yes
Particle surface chemical	Yes	No	Yes	No		
State of degradation	Surface oxidation	No	Surface oxidation	No		
Suitable sampling sites	Wastewater, water environment, drinking water		Wastewater, water for larger microplastics	Wastewater, water environment	Nanoplastics and small microplastics with known polymers	Wastewater, water environment
Dimension of specimen mass	ng-µg		µg-mg	ng-µg	ng	mg
Number of measurable particles per sample	10^2-10^5		One at a time	Microscopic visibility	Visibility	Depends on sample mass
Preparation and measuring time	hours to days		minutes	hours		minutes to hours
Detection level	>5 µm		>80 µm	>5 µm	>1 nm	Depends on sample mass
Example reference*	Wolff et al., 2019	Mintenig et al., 2017	Simon et al., 2018	Sierra et al., 2020	Nguyen et al., 2021	Hermabessiere et al., 2018

* Equipment prices were obtained through personal communication with the leading manufacturers (as of 2021) and available information on manufacturers' homepages. Reference is given for further reading as a case study of equipment usage. Tabulated information does not necessarily represent example reference content.

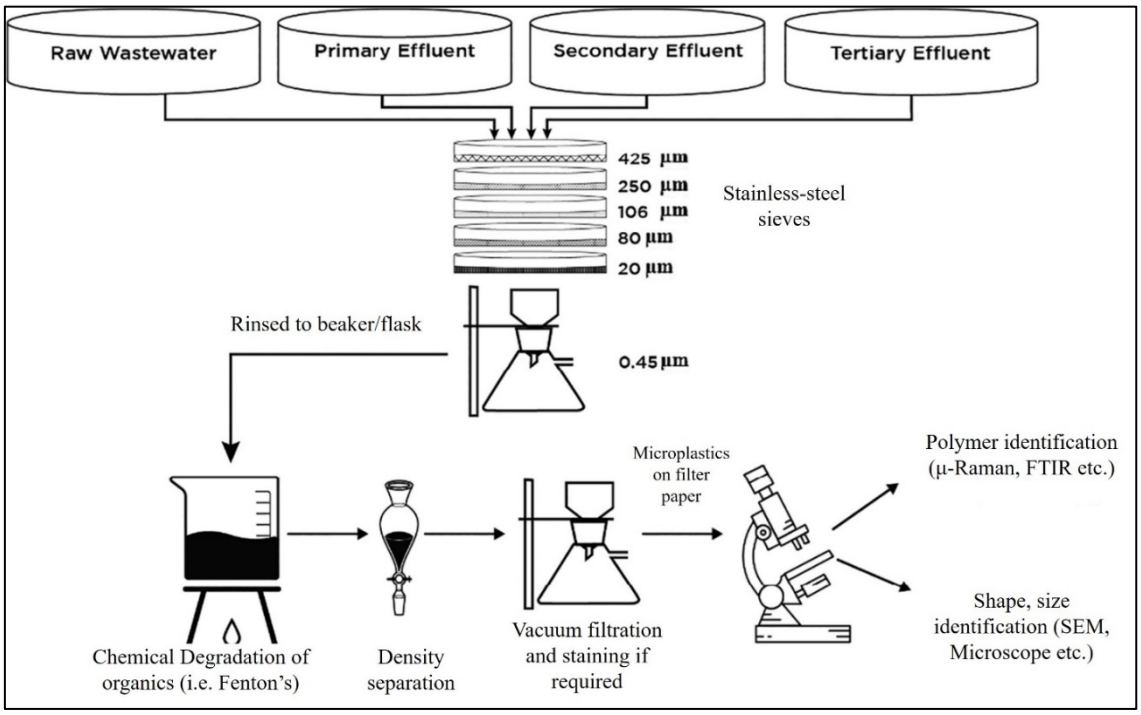
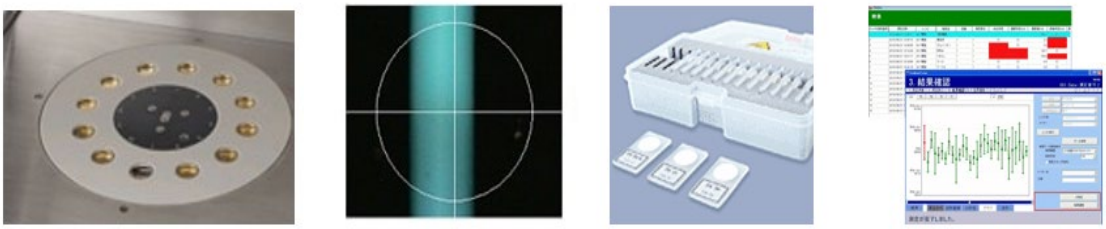


Figure 6. 3 Micropalstic sampling and pretreatment (Ben-Davis et al., 2021).

6.2 Methods



Sample loading X-ray fluorescence Satandadards Data processing

Fig. 6.4 potentially toxic element (PTE) effects and potential contaminations

And the samples were pre-treated with HCL/nitric acid. This process is expected to remove the heavy metals on the surface of the microplastics.

Fig. 6.5 Experimental flow

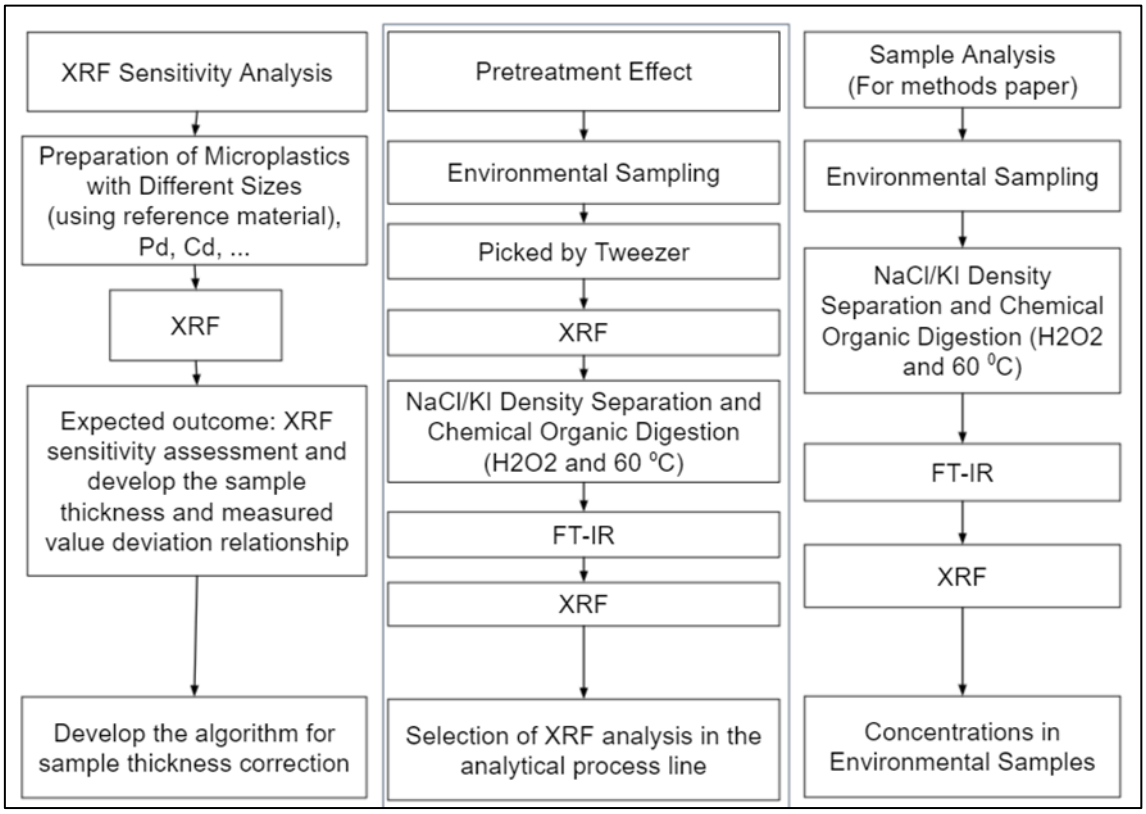


Fig. 6.6 the experimental flow of the element analysis in microplastics using the XRF equipment

At percent, pretreatment effect assessment is conducted. The sensitivity analysis and large quantity of sample analysis are yet to be conducted.

6.3 Results and Discussion

Heavy metal and other element compositions

- Barium sulfate serves as filler in the plastics and the paint and varnish industries.
- Titanium dioxide ensures UV rays do not penetrate and damage the product and its appearance. Titanium dioxide possesses non-flammable, insoluble, and durable qualities which makes it the obvious choice for use in outdoor plastics.
- Bromine present in brominated flame retardant (BFR) content
- Antimony (Sb) is a technology critical element whose presence is ubiquitous in manufactured products, and in particular in plastics where it is used extensively as a flame retardant synergist for brominated compounds, as a catalyst for polyethylene terephthalate production, and as a pigment for color
- Hexavalent chromium, Cr(VI), has traditionally been used as a pigment for paints and plastics, with both Chromium Yellow (PbCrO4) and Chromium Red
- Cr_2O_3 is known for its increased pigment opacity, high UV attenuation, poor deterioration

The main two types of microplastics identified as heavy metal contaminated areas, controlled-release fertilizer capsules, and broken artificial turf particles.

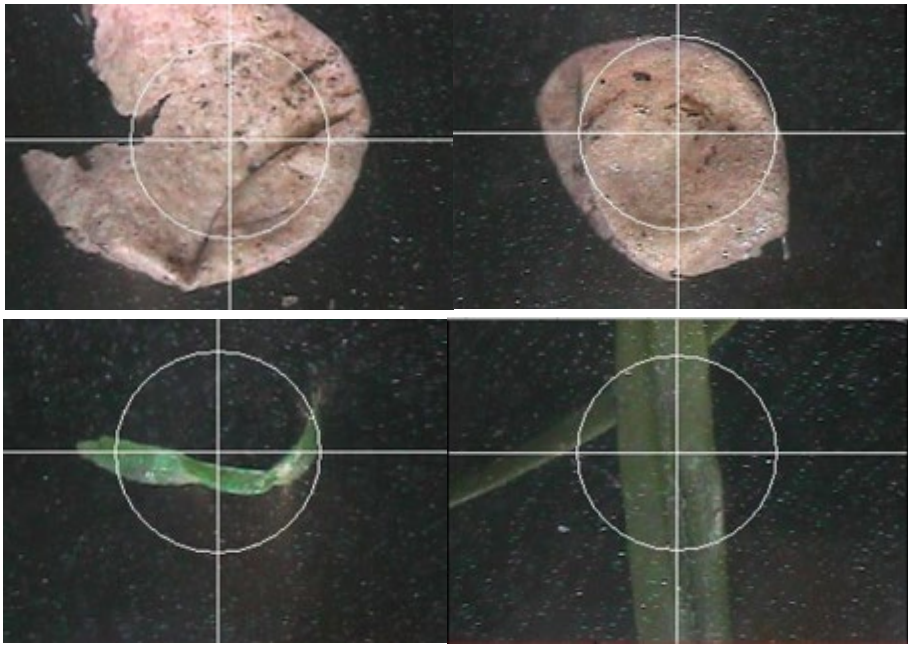


Fig. 6.7 Controlled release fertilizer capsule and artificial turf broken particle

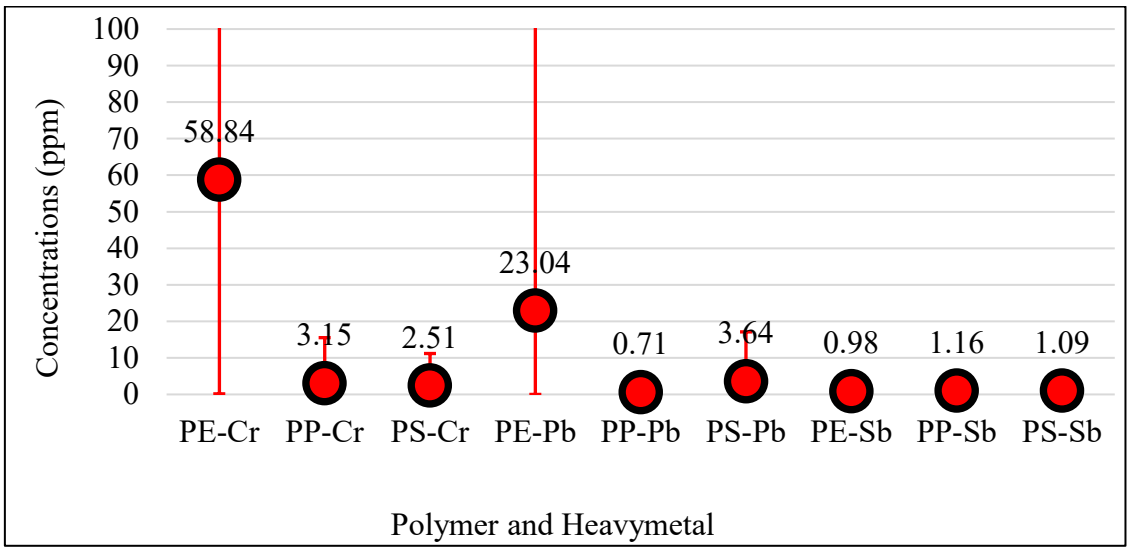


Fig. 6.8 provides some of the detected heavy metals in different polymers

The observations confirm that the XRF methods tested are capable of detecting heavy metals in the microplastics. However, there is a need of detecting assess the accuracy and the range of the XRF for detecting the potentially toxic elements in the microplastics. Further steps to be conducted to confirm the thickness effect on the element concentrations detected need to be done. Figure 6.9 indicates the preliminary observations supporting that the thickness of the microplastic affects the concentration.

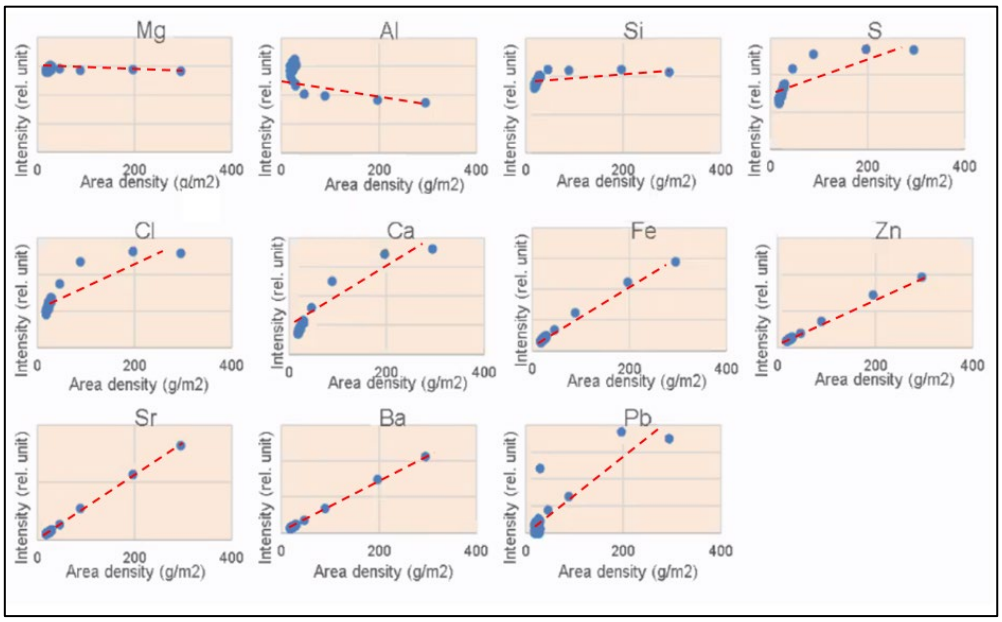


Fig. 6.9 Potentially toxic elements XRF signal intensities (without considering the thicknesses).

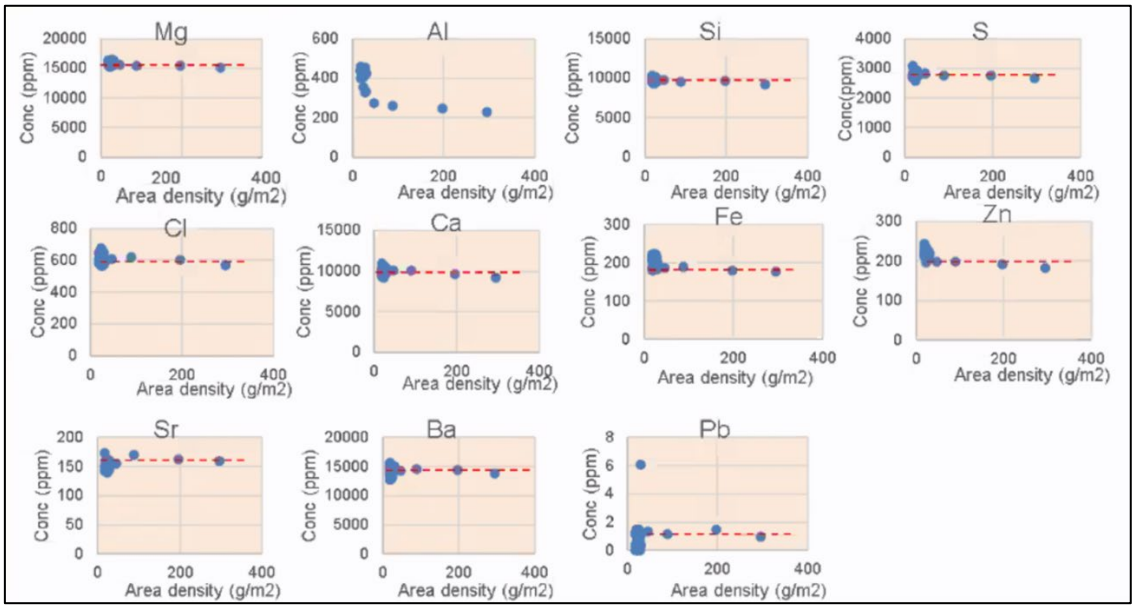


Fig. 6.10 Potentially toxic elements in microplastics detected by XRF.

6.4 Conclusions and Recommendations

XRF is capable of detecting PTEs, there is a variation of PTEs in different polymers and within the same polymer products. Additives can be the reason. The observations need to be verified further with standard plastic materials with PTEs. The thickness of microplastics affects the intensities. Identification of the thickness effect on the concentrations has been done during the study. Further testing with microplastics with different PTE concentrations is recommended for the confirmation of the thickness effect on different PTEs.

CHAPTER 7. CONCLUSION AND RECOMMENDATIONS

7.1 Policymaking process

Factors limiting leakage to the environment are one aspect to be considered to minimize the microplastic emissions into the environment. In the case of textile microfiber, wastewater treatment plant coverage, level of treatment, and sludge management practices are significant. Since a major component of the microplastics moves into the sludge, the practices such as land application of slude, recontaminate the environment. Apart from that, the circularity approach, the washing machine design improvement, and alternative materials such as the organic cotton introduction can be considered.

Table 7.1 Approaches to limit the textile microfiber and tire-wear particle into the environment.

Primary Microplastic	Factors Limiting Leakage to Environment	Circularity
Textile Microfiber from Washing	Wastewater treatment coverage Level of the wastewater treatment plant coverage Sludge management practices (i.e. landfill vs incineration)	Washing process improvement to intervene at the source. New material 
Tire-wear Particles	Combine sewer coverage Sludge management practices (i.e. landfill vs incineration)	Appropriate public transport where no rubber tire is used (i.e. train) Innovations to capture tire-abrasion particles at the source Combined sewer converge at hotspots 

Similarly for tire-wear particles, the combined sewer coverage-based approach for the high-density traffic areas to capture these microplastics and the alternative transportation modes introduction can be considered. Policy and governance-based solutions need to be introduced.

Necessary policies and institutionalized governance measures could be a key driver to create appropriate barriers to MPs entering humans through food chains (McDevitt et al, 2017; Rochman et al, 2016) and thus the PTEs associated with MPs as well.

Further, the integration of 3R and circular economy approach while restricting the MPs and PTEs release into the environment, recontamination, and spread of in the environment could support the mitigation process. Present plastic waste management policies mainly focused on waste disposal and regulation on land while the policy

regarding MPs has largely been focused on the prevention of aquatic MPs pollution (Harris et al, 2021). International and national agencies working on environmental protection and conservation intervened in the policies and regulations to protect aquatic environments from harmful impacts of microplastics (Guerranti et al., 2019; Xanthos and Walker, 2017). However, attention is growing on the detrimental impacts of MPs in terrestrial systems and the mitigative policy measure dialog (Harris et al, 2021; Galarpe et al, 2021). This would support the mitigation measures of the MPs and PTEs contaminations of agricultural lands and the human exposure through food chains originating at land-based agriculture. As elaborated in section 4.1, the major contamination pathways such as irrigation water, fertilizers, and soil conditioners including sewage sludge are to be considered in the counter measuring policy and institutionalization process. For example, the Basel convention plastic waste amendments, 2019 for the international movement of plastic waste (Liang et al, 2021), can be further strengthened for the products related to agriculture such as soil amendments and compost.

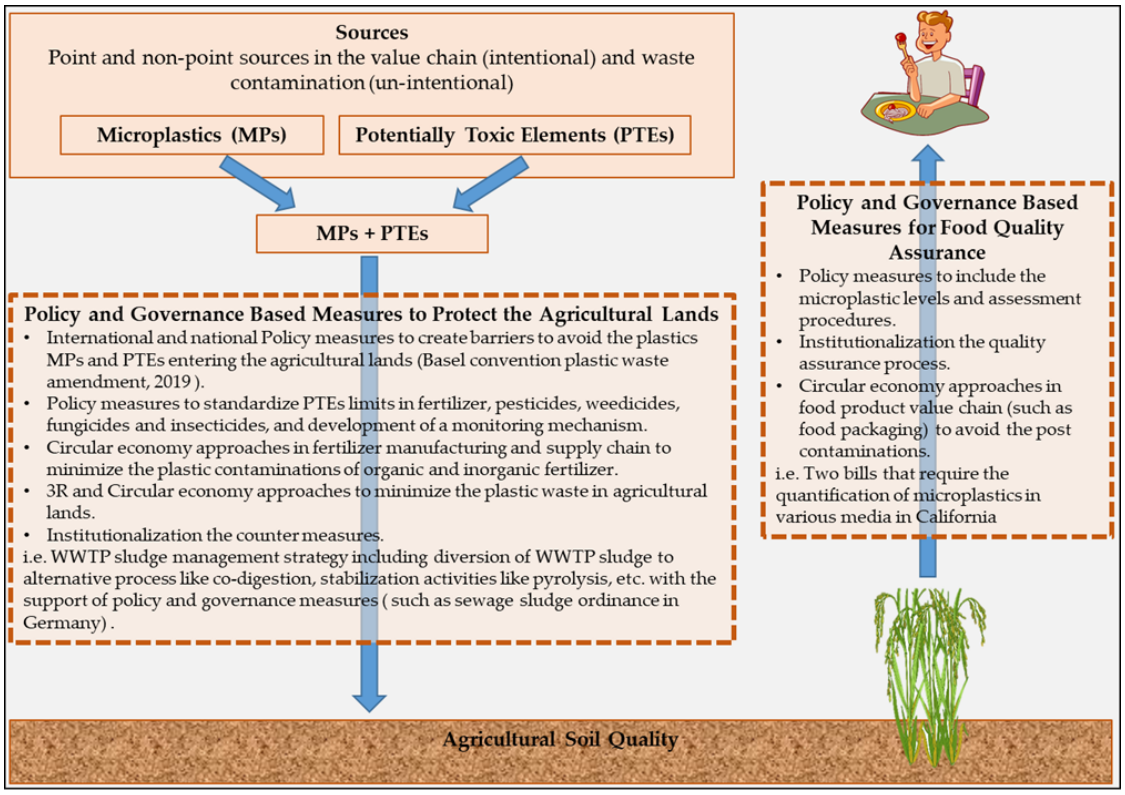


Fig. 7.2 Microplastic and PTE example policy-related approach for mitigation of human health threat.

Figure 7.2 illustrates the policy and governance-based countermeasures for MPs and PTEs reaching humans through ingestion. The California legislature enacted two bills that require the quantification of microplastics in various media and the development of new management strategies to address microplastic pollution targeting the mitigation of human exposure through ingestion (Rochman et al., 2020). The case of California is a classic example of recent policy developments to tackle the human contamination of MPs by ingestion.

7.2 Conclusions

A framework to integrate plastic litter into the LCA of plastics and plastic-related products has been conducted. Then the development of primary life cycle inventory (LCI) data for tire-wear particle microplastics in Japan was conducted. The tire-wear particle emission for Japan was estimated. The average value obtained was 33,796 metric tons (MT)/year. The obtained value was compared with the available literature and found it is within the range of UNEP, 2018 information (21,150-35,109).

Relationships between microplastic surface area and the mass for different categories of microplastics were conducted. The correlations were found to be good indicating R^2 values larger than 80 in most of the microplastic categories. Then the development of a GIS-based model to estimate the riverine microplastic flow at 10 km grid level and prefectural level. Development of methods to generate microplastic data suitable for the GIS model input (such as particle concentration conversion into mass concentration). Then the estimate of the emissions factors to the water environment of microplastics at 10 km grid level and prefectural levels was conducted. The per capita emissions were also calculated. The prefectures such as Tokyo (37 MT/year), and Kanagawa (23 MT/year) with high population densities indicated a higher degree of microplastic releases.

A wastewater treatment plant model (2114 plants) has been developed for the microplastic fate estimations since down the drain microplastics such as synthetic textile fiber and roadside microplastic emissions such as tire-wear abrasion particles. Even though the high emission of tier-wear abrasion particles from the major cities such as Tokyo, Yokohama, and Osaka, due to the presence of combined sewers, a considerable

fraction of the microplastics passes through the wastewater treatment plants. The total airborne (as PM 10) was estimated as 676 MT/year. The total leakage of tier-wear particles into the water was estimated for two scenarios. The long-term scenario which considers the complete wash of the street provided 27,300 MT/year emission to the water environment. The short-term runoff scenario (which considers 21% particle goes with street runoff) that 5,678 MT/year fate in water. The annual release of synthetic microfiber from domestic textile washing is 17,612 MT/year and the fate going to water environments is 5246 MT/year. The largest quantity transfers into wastewater treatment plant sludge (12,348 MT/year). This provides a countrywide fate factor of 0.30 to water, 0.70 to wastewater treatment plant sludge. However, 79.8% of the sludge in Japan is incinerated. Hence the fate in sludge which is potentially re-contaminates the environment is 2,494 MT/year. At the prefectural level, Tokyo has the lowest fate factor to the water environment (0.11) due to higher centralized coverage of wastewater and the higher degree of treatment. In the prefectures with comparatively lower coverage of sewerage the fate factor to water increases over 0.50. However, interestingly, the Aichi prefecture is the highest annual emissions to the water environment (fate factor to water, 0.33), due to the combination of medium fate factor and comparatively higher emission at the source due to higher population.

The potentially toxic element (PTE) analysis in microplastic method development using x-ray fluorescence spectroscopy (XRF) was conducted. The detection of several toxic heavy metals and other elements is possible. The confirmation of the XRF results is in progress.

The LCA integration of impacts of plastic pollution is presently developing. The LCI and LCIA development on a national and global scale will make this possible in near future. The scientific evidence on estimating toxic effects on ecosystems and humans needs to be addressed. The findings of the present study provide recommendations such as, to address the microplastic pollution such as tire-were particles, the countermeasures such as public transportation with non-rubber tire based (i.e. trains, tram-cars) and increasing the combined sewer coverage in hotspot areas (such as cities where higher breaking and sudden accelerations of vehicles happen). Microplastics in wastewater treatment systems are receiving increasing attention from researchers and professionals

in the field. With the development of sampling and analytical methods, the potential of microplastic-related studies in wastewater systems is also increasing. Wastewater treatment plants are a major point source of microplastic emissions into the environment. In these plants, the transfer of microplastics from the water phase to the sludge phase is the dominant (or presently only considered) removal mechanism due to low polymer biodegradability. This "removal" efficiency for secondary and tertiary treatment levels exceeds 99%. Since the majority of microplastics are transferred into sludge, sludge treatment and disposal are critical to protecting the environment from microplastic (re)contamination. Plastic-related chemicals and attached pathogens are other factors to consider. As the release of toxic chemicals from microplastics has not been studied in great detail, reconciling the limitations of analytical capabilities requires continued work. The increased introduction of biodegradable polymers worldwide may open new opportunities for biodegradability-related research and development studies shortly.

7.3 Recommendations and Future Studies

The continuation of XRF method development to estimate the PTE in microplastics. PTEs adsorb into MPs through physicochemical interactions and increase their mobility in soils. Hence, MPs act as vectors for PTEs in soil. MPs and PTEs complexes can be easily absorbed by plants. Hence, the accumulation of PTEs in plants can be enhanced in both MPs and PTEs contaminated soils. Furthermore, there is a high risk of food chain contamination with PTEs by crops grown in both MP and PTEs contaminated soils. Certain intentional (controlled-release fertilizer) and unintentional (WWTP sludge application), agricultural applications are identified sources of contaminations of agricultural lands. The policy and governance-based countermeasures shall be focusing on both agricultural land quality assurance and food quality assurance to minimize human exposure through ingestion of PTEs related to MPs.

The air emissions of roadside microplastics (as PM10) need to be considered as an air pollutant and using the available air transport models the fate of these microplastics needs to be estimated. Then the health impacts associated with PM 10 and PM 2.5 needs to be assessed.

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ANNEXURE 1

Table A1.1: Japan vehicle travel distances 2019 year

Month	Item	Unit	Total	business use					private or home use					Total	business use				private or home use			
				Total	registered vehicle (i.e. compact car or larger)			motor vehicle (up to 660cc and 64bhp)	Total	registered vehicle (i.e. compact car or larger)			light motor vehicle (up to 660cc and 64bhp)		Total	Bus (capacity of 11 or more passengers)		"Passenger car (Capacity: 10 passengers	Total	registered vehicle (i.e. compact car or larger)		light motor vehicle (up to 660cc and 64bhp)
					Ordinary	small car	special-			Ordinary	small car	special-				joint partner	chartering			Bus	passenger	
1	mileage	1000 km	16 379 631	5 552 339	3 686 397	122 927	1 231 796	511 219	10 827 293	1 450 780	3 377 348	409 288	5 589 876	47 369 812	1 089 437	266 592	81 737	741 108	46 280 375	122 202	30 236 007	15 922 166
		Change from previous month %	97.6	97.0	97.3	96.4	97.0	94.7	98.0	96.2	98.2	97.5	98.3	103.4	96.9	99.0	97.2	96.1	103.5	95.9	101.4	107.9
		Year-on-year change (%)	98.8	100.7	100.2	101.4	100.5	105.2	97.8	101.3	98.9	95.2	96.5	103.7	95.2	98.9	98.9	93.5	103.9	99.5	104.3	103.3
2	mileage	1000 km	14 676 069	5 050 404	3 329 890	114 851	1 144 053	461 610	9 625 665	1 305 114	2 913 723	389 981	5 016 846	41 285 315	994 691	239 313	89 539	665 839	40 290 624	111 673	26 790 626	13 388 324
		Change from previous month %	89.6	91.0	90.3	93.4	92.9	90.3	88.9	90.0	86.3	95.3	89.7	87.2	91.3	89.8	109.5	89.8	87.1	91.4	88.6	84.1

		Year-on-year change (%)	98.7	101.8	100.8	104.1	103.5	105.0	97.2	100.8	96.0	100.7	96.7	101.3	96.5	103.2	101.7	93.7	101.4	99.8	100.8	102.6
3	mileage	1000 km	16 706 326	5 710 801	3 824 913	129 179	1 234 242	522 467	10 995 525	1 529 554	3 362 237	436 201	5 667 534	46 722 778	1 090 826	257 307	92 585	740 934	45 631 952	124 843	30 573 691	14 933 418
		Change from previous month %	113.8	113.1	114.9	112.5	107.9	113.2	114.2	117.2	115.4	111.9	113.0	113.2	109.7	107.5	103.4	111.3	113.3	111.8	114.1	111.5
		Year-on-year change (%)	100.3	100.8	100.3	101.6	99.6	107.5	100.1	101.7	101.2	100.9	98.9	101.9	94.9	100.5	100.0	92.5	102.1	99.4	101.8	102.7
4	mileage	1000 km	16 125 994	5 548 619	3 689 477	120 131	1 228 086	510 925	10 577 375	1 460 805	3 279 803	397 939	5 438 829	45 683 307	1 058 485	255 098	98 519	704 868	44 624 822	117 628	29 270 002	15 237 192
		Change from previous month %	96.5	97.2	96.5	93.0	99.5	97.8	96.2	95.5	97.5	91.2	96.0	97.8	97.0	99.1	106.4	95.1	97.8	94.2	95.7	102.0
		Year-on-year change (%)	101.0	103.2	103.7	96.3	101.7	105.6	99.9	101.0	104.1	97.4	97.4	100.7	96.9	103.0	101.0	94.4	100.8	94.4	99.0	104.3
5	mileage	1000 km	16 423 096	5 575 451	3 696 090	126 722	1 243 430	509 209	10 847 645	1 454 012	3 304 642	414 329	5 674 661	47 028 958	1 098 831	260 661	111 675	726 494	45 930 128	122 102	30 597 393	15 210 633

		Change from previous month %	101.8	100.5	100.2	105.5	101.2	99.7	102.6	99.5	100.8	104.1	104.3	102.9	103.8	102.2	113.4	103.1	102.9	103.8	104.5	99.8
		Year-on-year change (%)	99.9	101.7	101.1	105.3	101.6	105.8	98.9	100.7	97.5	100.5	99.2	99.5	97.0	103.3	103.1	94.1	99.6	96.1	100.3	98.3
6	mileage	1000 km	16 051 349	5 438 642	3 610 410	119 751	1 208 734	499 748	10 612 706	1 461 861	3 272 984	391 557	5 486 305	45 085 149	1 061 216	246 324	113 710	701 182	44 023 933	125 527	29 450 240	14 448 166
		Change from previous month %	97.7	97.5	97.7	94.5	97.2	98.1	97.8	100.5	99.0	94.5	96.7	95.9	96.6	94.0	102.0	96.5	95.8	102.8	96.3	95.0
		Year-on-year change (%)	97.6	99.2	98.7	99.5	98.8	103.8	96.8	101.3	96.6	94.9	95.9	95.4	93.7	97.0	99.0	90.9	95.5	98.8	96.5	93.4
7	mileage	1000 km	16 473 322	5 609 880	3 694 206	125 658	1 244 937	545 080	10 863 441	1 473 325	3 347 821	402 962	5 639 333	47 223 302	1 087 750	254 036	105 353	728 360	46 135 552	121 935	30 697 322	15 316 295
		Change from previous month %	102.6	103.1	102.3	104.9	103.0	109.1	102.4	100.8	102.3	102.9	102.8	104.7	102.5	103.1	92.7	103.9	104.8	97.1	104.2	106.0
		Year-on-year change (%)	100.6	100.8	100.9	101.6	97.4	108.5	100.4	103.3	102.1	98.3	98.9	102.1	95.1	101.4	101.6	92.2	102.3	97.3	104.9	97.6

8	mileage	1000 km	16 566 067	5 598 325	3 701 578	124 260	1 245 184	527 304	10 967 742	1 467 585	3 254 506	403 624	5 842 027	48 604 302	1 074 752	256 787	98 330	719 635	47 529 550	120 741	31 278 550	16 130 259
		Change from previous month %	103.2	102.9	102.5	103.8	103.0	105.5	103.3	100.4	99.4	103.1	106.5	107.8	101.3	101.1	93.3	102.6	108.0	96.2	106.2	111.6
		Year-on-year change (%)	101.1	100.6	101.1	100.5	97.4	104.9	101.4	102.9	99.2	98.4	102.5	105.1	93.9	97.4	96.2	91.1	105.4	96.3	106.9	102.8
9	mileage	1000 km	15 906 742	5 442 445	3 620 232	119 850	1 197 543	504 820	10 464 297	1 474 640	3 214 460	398 147	5 377 050	46 264 499	1 044 891	251 269	102 218	691 404	45 219 608	123 545	29 544 716	15 551 346
		Change from previous month %	96.0	97.2	97.8	96.5	96.2	95.7	95.4	100.5	98.8	98.6	92.0	95.2	97.2	97.9	104.0	96.1	95.1	102.3	94.5	96.4
		Year-on-year change (%)	99.3	99.5	98.4	101.0	100.6	105.0	99.2	100.3	99.6	101.1	98.6	99.9	96.1	103.4	104.6	92.6	100.0	98.4	98.1	103.8
10	mileage	1000 km	16 554 202	5 629 603	3 722 812	123 836	1 265 408	517 547	10 924 599	1 532 770	3 389 597	414 648	5 587 583	47 996 450	1 059 167	239 915	115 071	704 182	46 937 283	124 946	30 496 552	16 315 785
		Change from previous month %	104.1	103.4	102.8	103.3	105.7	102.5	104.4	103.9	105.4	104.1	103.9	103.7	101.4	95.5	112.6	101.8	103.8	101.1	103.2	104.9

		Year-on-year change (%)	100.2	100.4	100.6	100.6	98.6	103.7	100.0	100.7	102.6	98.2	98.5	99.8	93.5	97.5	93.9	92.2	99.9	95.7	97.9	104.1
11	mileage	1000 km	15 951 803	5 448 977	3 598 338	121 507	1 228 515	500 616	10 502 827	1 482 774	3 203 135	410 556	5 406 362	45 262 623	1 036 268	236 044	117 194	683 030	44 226 355	121 287	29 206 740	14 898 329
		Change from previous month %	96.4	96.8	96.7	98.1	97.1	96.7	96.1	96.7	94.5	99.0	96.8	94.3	97.8	98.4	101.8	97.0	94.2	97.1	95.8	91.3
		Year-on-year change (%)	98.5	99.4	98.1	96.9	102.0	103.0	98.0	101.9	97.7	99.9	97.1	96.8	94.9	99.0	98.3	93.1	96.9	96.3	96.9	96.9
12	mileage	1000 km	16 412 298	5 648 861	3 725 783	125 037	1 267 680	530 361	10 763 437	1 536 974	3 207 216	428 069	5 591 179	47 883 404	1 056 090	267 354	81 789	706 947	46 827 314	122 480	31 375 948	15 328 886
		Change from previous month %	102.9	103.7	103.5	102.9	103.2	105.9	102.5	103.7	100.1	104.3	103.4	105.8	101.9	113.3	69.8	103.5	105.9	101.0	107.4	102.9
		Year-on-year change (%)	97.8	98.6	98.3	98.1	99.8	98.3	97.4	102.0	93.2	101.9	98.4	104.5	93.9	99.3	97.2	91.7	104.7	96.1	105.2	103.9

Table A1.2 Tire-wear particle emissions for different vehicle types (mg/vehicle km), as reported in literature.

Vehicle type	Urban roads	Rural roads	Highways	Reference
Light duty vehicles	5			EPA (1995)
Heavy duty vehicle	7.5			
Motorcycle	1.72			Reference year 1995 CEPMEIP (2020)
Passenger car	3.45			
Light duty vehicle	4.5			
Heavy duty vehicle	18.56			
Passenger car	53			Gebbe and Hartung (1997)
Van	107			
Bus	344			
Lorry	539			
Truck	1092			
Passenger car	6.1			Rauterberg-Wulff (1998)
Lorry	≤32			
Passenger car	Mean: 100; range: 40–360			Luhana et al. (2004)

Vehicle type	Urban roads	Rural roads	Highways	Reference
Passenger car	Mean: 90; range: 53–200			Hillenbrand et al. (2005)
Van/Lorry	Mean: 700; range: 107–1500			
Bus	700 (like van and lorry)			
Truck	Mean: 1200; range: 1000–1500			
Not specified	9			Kupiainen et al. (2005)
Car	50			Gustafsson et al. (2008)
Bus	700			
Motorized 2-wheeler	7			Aatmeeyata et al. (2009)
Not specified	2.2			Sjödin et al. (2010)
Passenger car	33			Anonymous (2012)
Light commercial	51			
Heavy commercial	178			
Not specified	2.4–7			Panko et al. (2013a)
Passenger car	8.8	6.8	5.8	NAEI (2017)
Motorcycle	3.8	2.9	2.5	
Moped	3.8	–	–	

Vehicle type	Urban roads	Rural roads	Highways	Reference
Light duty vehicle	14	11	9.1	
Heavy duty vehicle	47	27	31	
Bus/coach	21	17	14	
Passenger car	132	85	104	DELTARES and TNO (2016)
Motorcycle	60	39	47	
Moped	13	9	10	
Van	159	102	125	
Lorry	850	546	668	
Truck	658	423	517	
Bus	415	267	326	
Light special vehicle	159	102	125	
Heavy special vehicle	850	546	668	

ANNEXURE 2

Figure A2: Impacts of plastics (present LCA does not take into account).

ANNEXURE 3

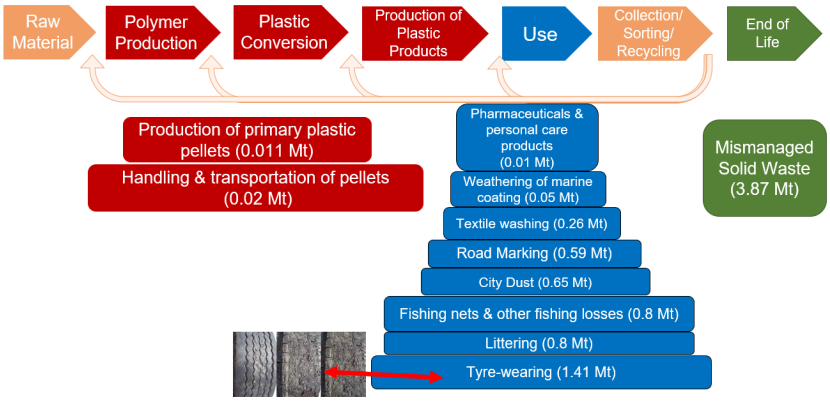


Figure A3: Plastic leakage into environment at various locations of the value chain.

ANNEXURE 4

Fig A4.1. The sampling device and some sampling locations

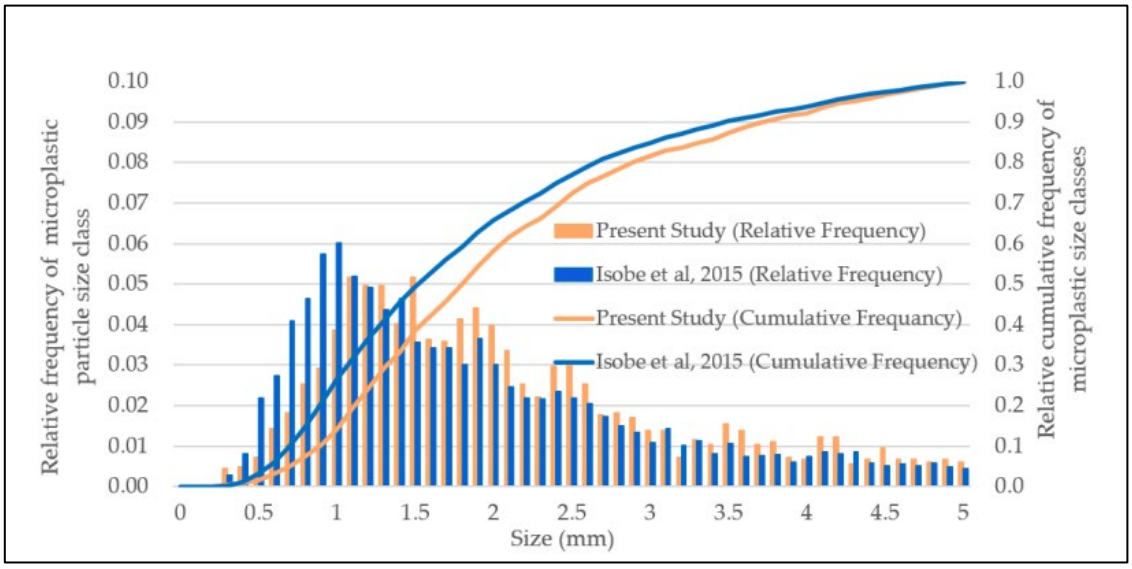


Fig A4.2 Comparison between this study and Isobe et al, 2015 [1] (East Aisan Seas around Japan). The cocentration values reported by Isobe et al, 2015 [1] were converted to fractions for the comparison with the present study.

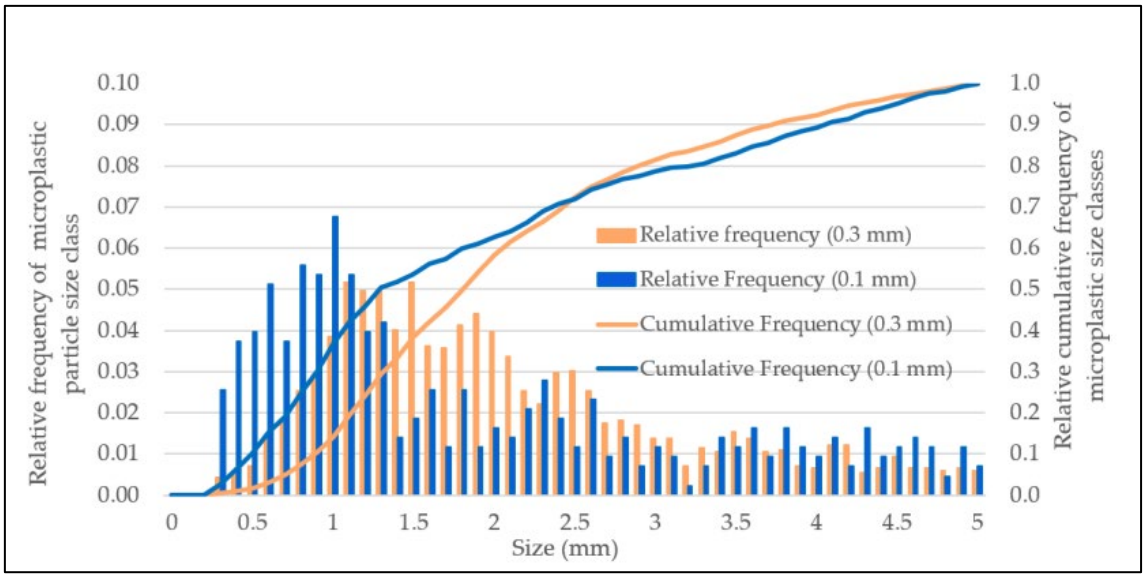


Fig A4.3 300 µm Vs 100 µm mesh (particles larger than 300 µm only)

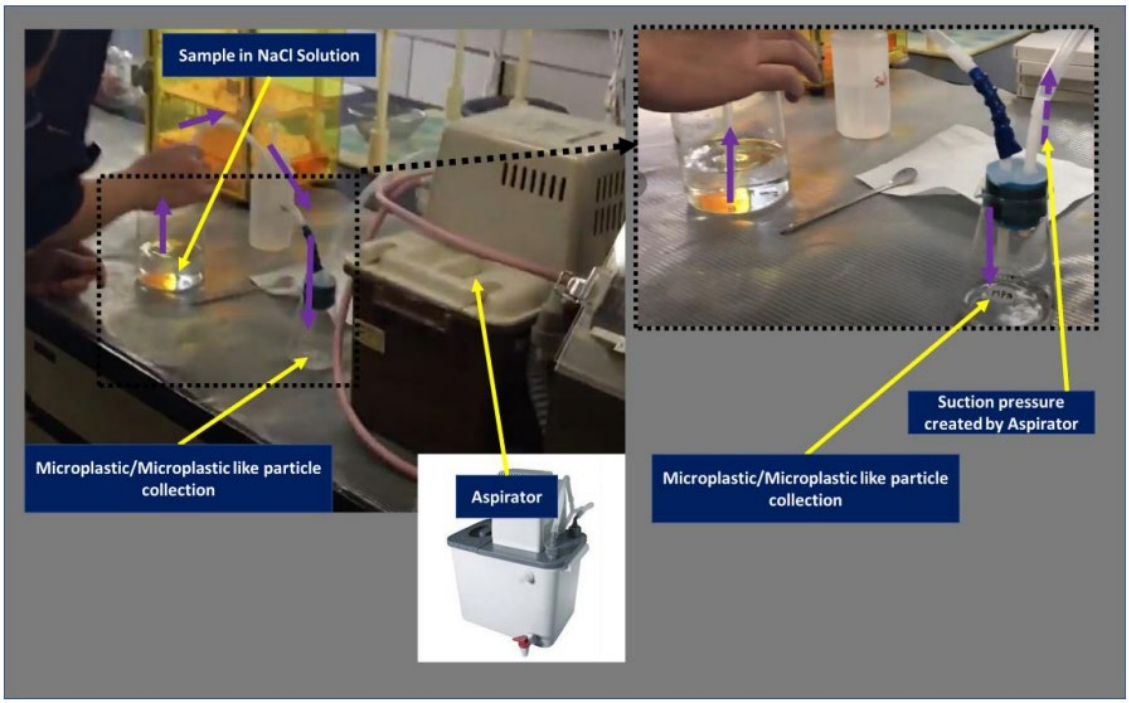


Fig A4.4. Sample extraction

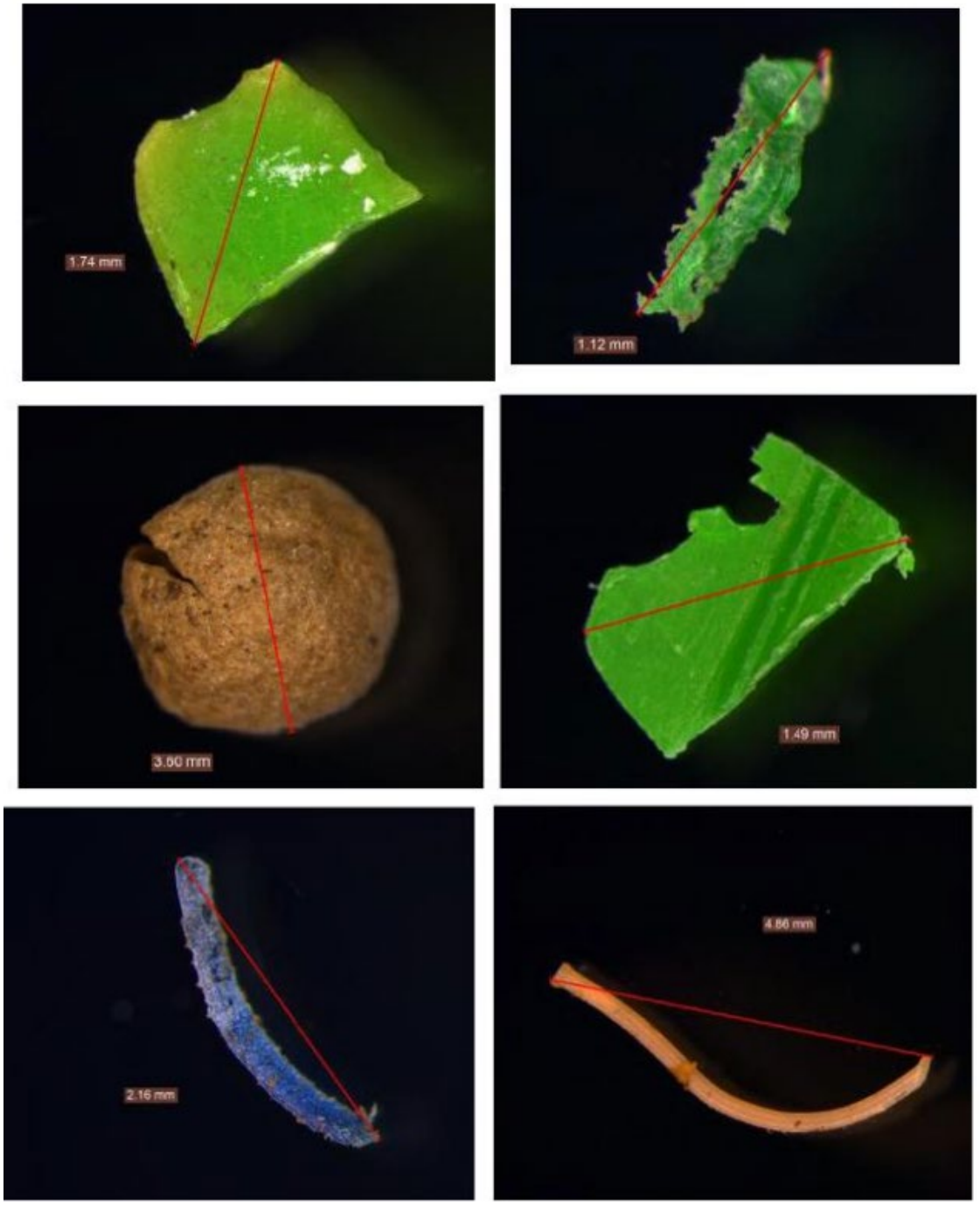


Fig. A4.5 Some microplastics sampled

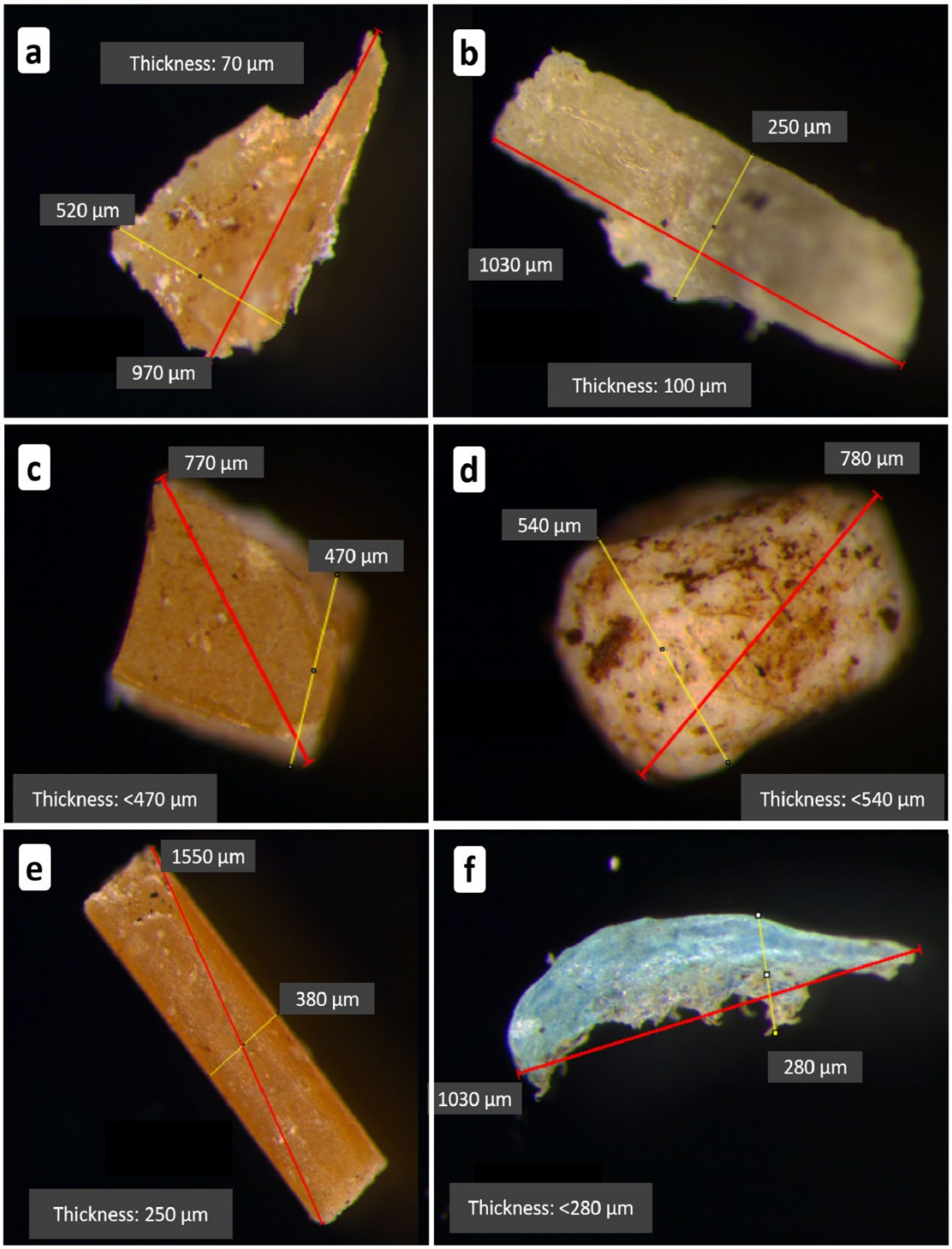


Fig. A4.6 Some microplastics sampled (Dimensions of selected microplastic pieces. (a): Other plastic; (b,e): polypropylene (PP); (c): polyethylene (PE); (d): polystyrene (PS); (f): combination of PE and PS (listed under another polymer). Red lines indicate the Feret's diameters while the yellow lines indicate a length, which would be effective for retaining the microplastic particle if the yellow line was oriented parallel direction to the mesh.)

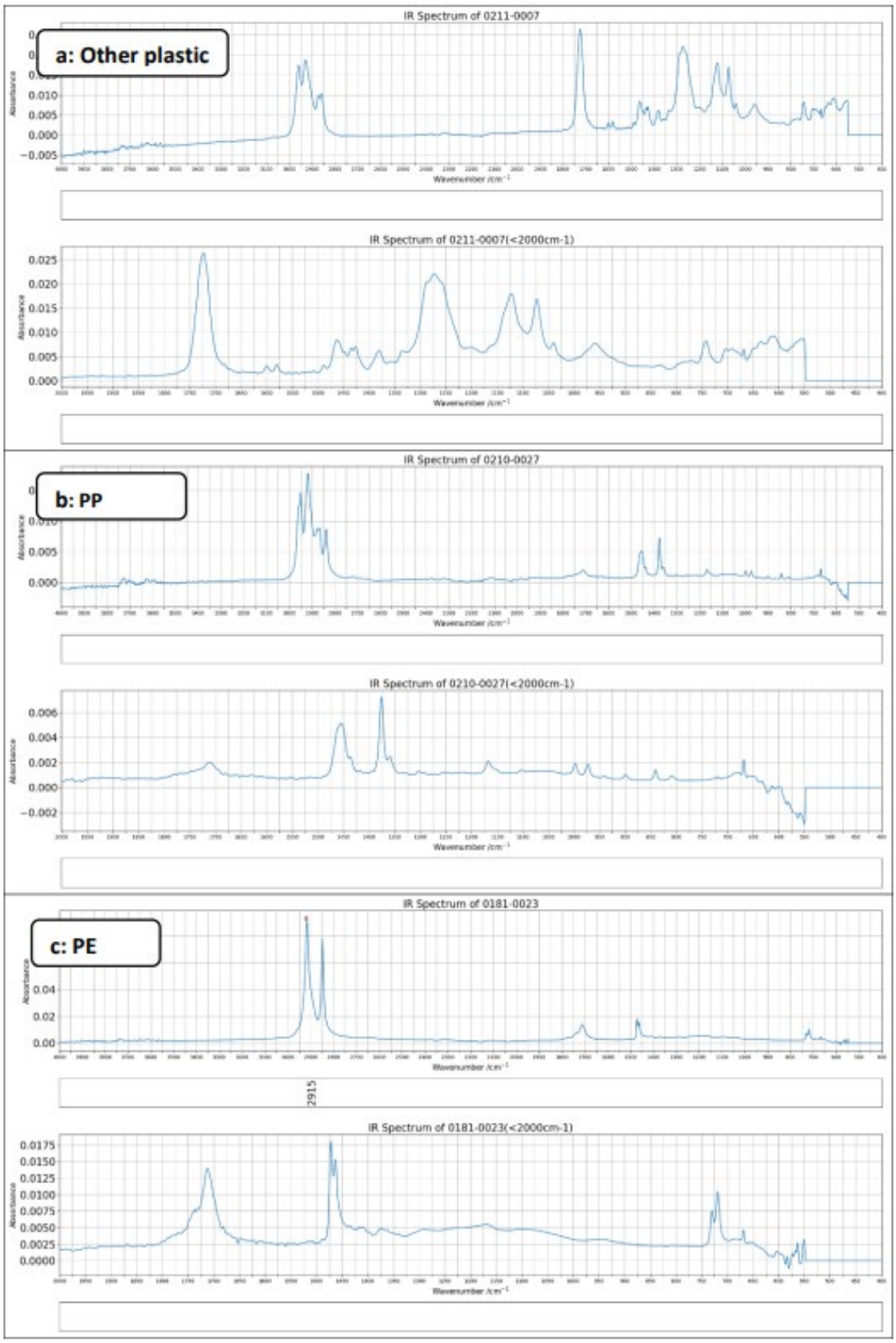


Fig. A4.7 Some IR Spectra of microplastics sampled (Fig A4.6)

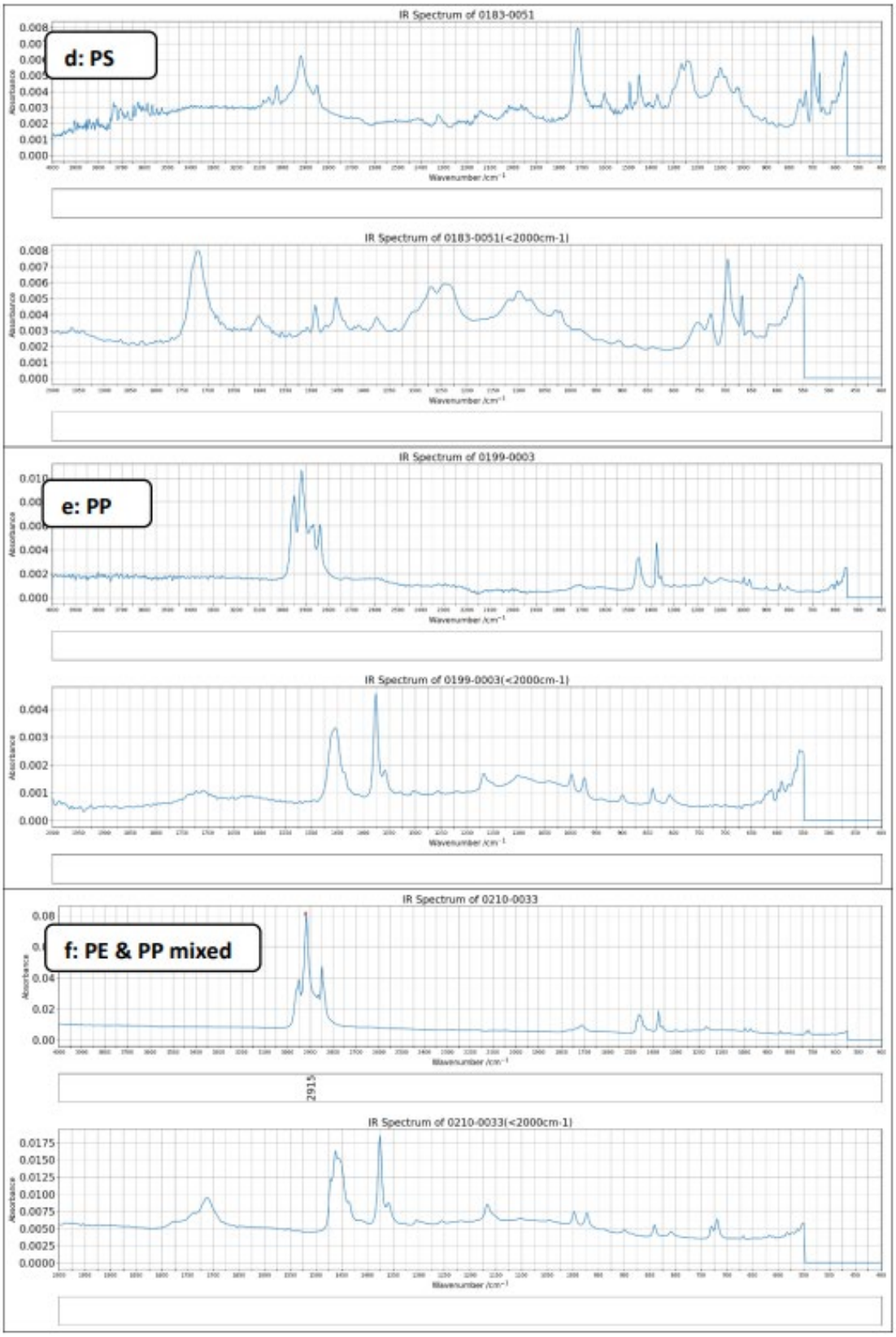


Fig. A4.8 Some IR Spectra of microplastics sampled (Fig A4.6)

Table A4.1Microplastic sampling locations (need to modify)

No.	River	Survey site	Number [particles/m^3]	Mass [mg/m^3]	Basin area [km^2]	Population density [persons/ km^2]	Urban area ratio [%]
1	Koetoi R.	Komatsu	0.1907	0.0013	84	4.13	1.15
2	Shimoebekorobetsu R.	Toyotomi	1.8139	0.1882	130	6.04	0.93
3	Ishikari R.	Tachihu-oohashi	4.1051	0.6850	13,637	38.05	2.39
4	Toyohira R.	Nijunijo-oohashi	1.2445	0.0566	932	125.89	3.34
5	Kitakami R.	Meiji	0.1443	0.0016	2,221	140.56	5.00
6a	Mogami R.	Shonai-oohashi	0.3600	0.0838	6,537	129.68	5.75
6b	Mogami R.	Kurotaki	0.4887	0.1242	4,118	182.10	7.72
6d	Mogami R.	Konoki	1.4821	0.0164	2,648	94.13	5.78
7	Su R.	Ochiai	8.1175	1.5248	884	362.17	14.70
8	Abukuma R.	Tenjin	0.3866	0.0073	4,103	215.97	10.04
9	Kuji R.	Tomioka	0.0295	0.0001	959	59.40	3.08
10	Naka R.	Nakagawa	0.6955	0.0347	2,242	144.76	8.11
11	Sakura R.	Sakaeri	2.4584	0.7412	311	264.98	17.19
12	Kinu R.	Toyomizu	0.3970	0.0059	1,797	54.11	11.14
13	Watarase R.	Nowatari	1.5333	0.0731	2,443	429.08	15.43
14a	Tone R.	Sakae	0.3652	0.0743	13,019	475.30	17.13
14b	Tone R.	Tonegawa	8.6760	2.3640	8,804	329.39	13.81
14c	Tone R.	Bando	0.1706	0.0302	1,920	413.50	13.65
15a	Ohori R.	Kisaki	4.4025	3.3136	23	7161.28	85.44
15b	Ohori R.	Kachi	12.8790	1.0765	14	6066.07	82.19

16	Edo R.	Noda	3.3203	0.5754	29	2365.69	56.62
17a	Naka R.	Yoshikoshi	2.3124	1.7801	697	1784.03	45.46
17b	Naka R.	Shinkai	5.9783	1.7416	250	999.81	37.26
18a	Ara R.	Hanekura	4.5686	0.9651	2,017	636.12	17.01
18b	Ara R.	Kaihei	7.4001	1.3671	1,245	403.43	12.41
18c	Ara R.	Onari	8.3522	0.3235	1,048	218.52	8.01
18d	Ara R.	Kumagaya	4.5910	0.0512	973	157.38	6.58
18e	Ara R.	Tamayodo	0.4371	0.0212	923	128.14	5.12
18f	Ara R.	Kyu-titibu	1.1494	0.1607	458	78.08	2.66
19	Ichino R.	Matsunaga	2.0912	0.4270	56	1002.50	42.32
20	Musashi Channel	Gese	1.3121	0.0433	5,521	329.84	11.52
21	Yoshino R.	Mannen	17.2694	0.5871	13	445.23	25.60
22	Yoro R.	Kasumi	0.7112	0.0031	251	208.20	9.68
23	Obitsu R.	Nakagawa	3.2922	0.1545	227	110.13	6.04
24	Koito R.	Rokusan	1.4345	0.1210	107	115.74	5.16
25	Tama R.	Maruko	1.1077	0.2431	1,198	2931.00	31.45
26a	Tsurumi R.	Shinyokohama	14.2359	3.3283	136	6619.04	72.10
26b	Tsurumi R.	Kamoike	13.8096	3.6232	118	6876.65	72.52
26c	Tsurumi R.	Kawawakitahassaku	30.6685	1.5228	66	5759.22	67.19
26d	Tsurumi R.	Ochiai	6.1514	1.1635	115	6752.30	71.85
26e	Tsurumi R.	Onmawari	10.5224	2.1093	44	5230.27	65.68
26f	Tsurumi R.	Sumiyoshi	2.5939	0.3160	10	5768.33	22.79
27	Sagami R.	Sagami-oohashi	0.3035	0.0431	1,522	446.06	12.09
28	Toneunga R.	Fureai	12.6587	2.8074	22	1333.00	49.50
29	Hayakido R.	Shibasawa	3.5119	0.1349	40	35.16	1.78

30	Saka R.	Midori	0.5960	0.4571	32	599.70	26.07
31	Shonai R.	Shin-meisei	63.8889	16.1510	1,303	2044.84	44.05
32	Kiso R.	Kawashima-oohashi	0.5515	0.0418	5,632	79.47	2.02
33	Nagara R.	Nagara-oohashi	1.7857	0.0442	1,971	110.03	6.14
34	Ibi R.	Ibi-oohashi	1.0067	0.0116	1,475	71.89	2.58
35	Kuzuryu R.	Nakakado	2.0086	0.0609	1,318	72.08	3.56
36	Asuwa R.	Kujuku	7.3453	1.6966	402	144.46	5.26
37	Kamo R.	Kyoukawa	4.9270	0.7746	246	2378.00	29.87
38	Katsura R.	Miyamae	9.5745	3.6134	1,351	924.22	14.21
39	Uji R.	Gokou	1.8327	1.2019	5,313	333.28	11.14
40	Yodo R.	Hijikata	2.0073	0.1107	8,595	490.56	12.16
41	Ina R.	Minamizono	6.3857	0.6773	480	1260.87	22.08
42a	Yamato R.	Taisho	6.9358	0.3726	1,179	1265.80	30.11
42b	Yamato R.	Gokou-oohashi	11.0924	2.5164	634	1192.00	30.76
43	Toga R.	Shimokawara	1.3847	0.0271	10	4276.28	27.58
44	Ikuta R.	Nunohiki	0.2201	0.0131	12	302.89	2.59
45	Sendai R.	Sendai-oohashi	0.9851	0.0104	1,308	83.00	3.92
46	Tenjin R.	Tenjin	1.9489	0.0423	578	86.13	4.25
47	Hino R.	Shin-hino	0.4491	0.0365	914	25.06	2.21
48	Hii R.	Mizuho-oohashi	0.2775	0.0063	1,153	53.47	3.75
49	Goemon R.	Hinode	3.9817	0.4549	20	643.18	29.32
50	Asahi R.	Okakita-oohashi	0.8997	0.0437	1,625	70.12	4.07
51	Nishiki R.	Gosho-oohashi	0.1107	0.0017	1,033	21.47	1.64
52	Saba R.	Okinohara	0.1221	0.0007	340	18.77	1.22
53	Fushino R.	Takada	0.6464	0.0169	254	339.22	12.99

54a	Mononobe R.	Mononobe	1.0695	0.1240	465	26.11	1.22
54b	Mononobe R.	Matchida	1.4793	0.1769	461	20.56	1.07
55	Niyodo R.	Niyodo-oohashi	3.7644	0.0262	1,451	42.74	1.79
56a	Shimanto R.	Rivermouth	1.3456	0.0432	2,198	37.14	1.82
56b	Shimanto R.	Downstream	0.3902	0.0048	2,181	36.79	1.80
57	Shigenobu R.	Deai	0.6354	0.0554	458	658.08	12.33
58	Yaoshi.R	Seisei	0.2626	0.0029	72	217.80	7.56
59	Hiji R.	Hatanomae	0.4169	0.0261	26	92.96	7.54
60	Onga R.	Kanroku	1.2664	0.0696	383	507.88	19.47
61	Hikosan R.	Okamori	5.2449	3.0385	307	405.66	16.71
62	Kagetsu R.	Kagetsugawa	1.3749	0.0486	128	100.12	4.02
63	Kikuchi R.	Yamagaseibu-oohashi	2.2815	3.1059	743	181.40	10.94
64	Kuro R.	Kurumagaeri	0.2097	0.0064	186	124.87	9.16
65	Shira R.	Yotsugi	5.5146	0.0109	455	334.19	12.25
66	Midori R.	Medomachi	8.2489	0.4267	705	66.89	4.94
67	Kuma R.	Seibu-oohashi	0.8391	0.1062	1,882	50.07	2.98
68	Sendai R.	Miyanojo	1.2062	0.6815	1,032	67.82	6.40
69	Fukido R.	South side	0.2310	0.0157	1	0.00	0.00
70a	Miyara R.	Kainan	12.7658	0.6213	23	11.86	1.80
70b	Miyara R.	Kawara	0.9746	0.3062	23	11.36	1.68

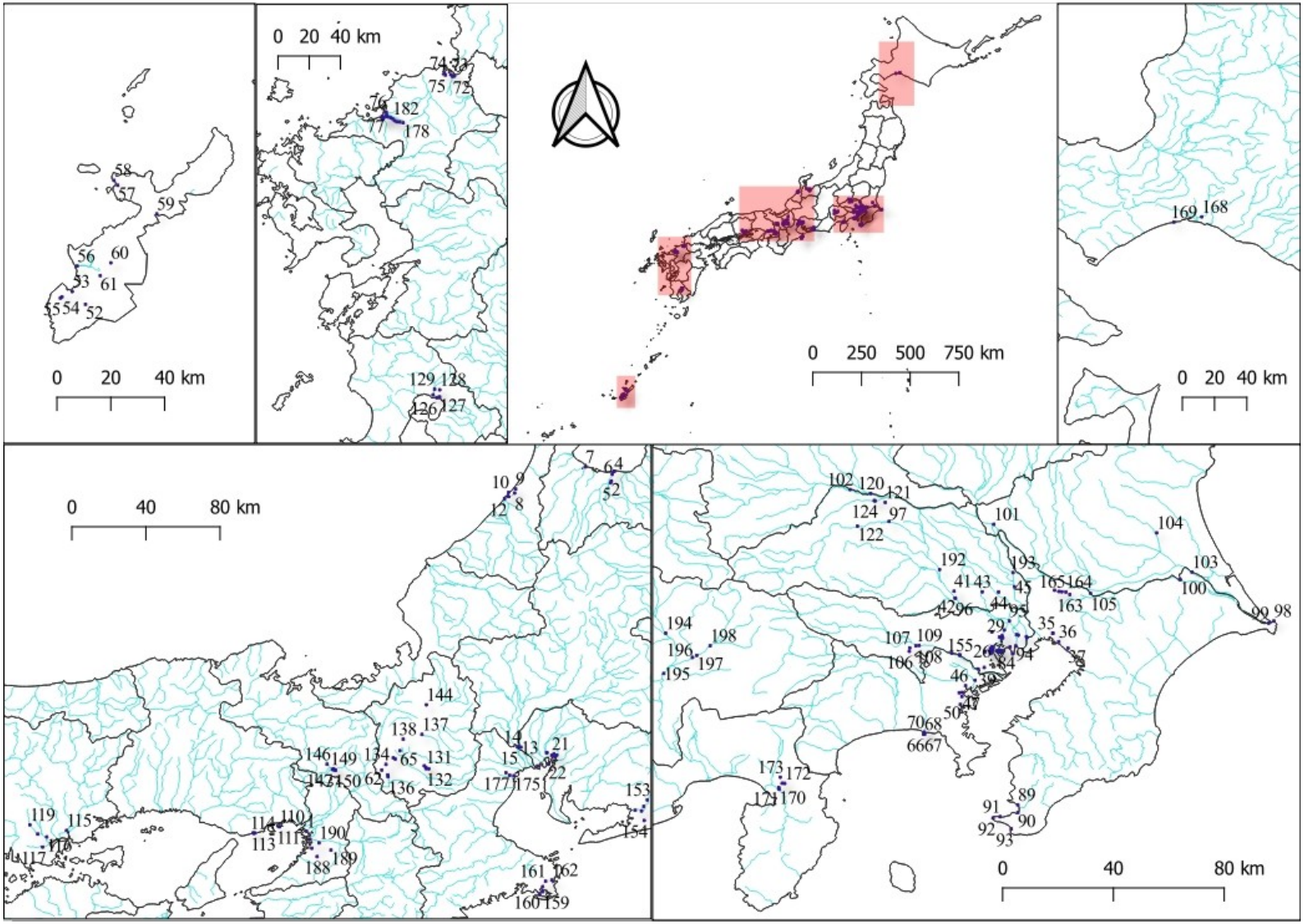

0 20 40 km
0 20 40 km
0 250 500 750 km
0 20 40 km
0 40 80 km
0 40 80 km

Table A4.2 Comparison of microplastic sampling devices

Device and Method Facts	Sampling Environment	Concentration (Particles/m^3)	Reference
Trawling a 300 µm net. (Sampling volume = 100 m^3).	Estuary, Brazil	0.1	[21]
Trawling 333 µm net.	River, China	0.1–5.6	[22]
Trawling a 333 µm net.	Channel, UK	0–1.5	[23]
Jet pump and external power supply unit (sampling volume ≈ 1.5 m^3). Complex system due to the requirement of the crew to carry equipment, boat travel, pumping out, and filtration through the sieves.	Riverine, Hungary	3.52–32.05	[24]
Bridge suspended net (300 µm); 5 to 30 min sampling durations. Capable only with rivers with high flow velocity and overhead bridges.	Riverine, Japan	1.6 ± 2.3	[25]
Vertically immersed filter with suction pump form the bottom 300 µm; 8 to 13 min taken for volume 0.1 m^3 sampling.	Estuary, China	67.5	[26]
Trawling a 300 µm net.	Riverine, Nigeria	0–0.2	[32]
100 µm net kept across the river flow (1–2 min duration). (V = 3.2 m^3). Longer sampling led to issues of clogging with organic matter. Capable only with rivers with high flow velocity.	Riverine, Wayni, USA	0–13.7	[31]
Trawling a 300 µm net Sampling volume = 10 m^3.	Estuary, South Africa	1–7	[33]
Trawling a 330 µm net.	Bay, USA	4.5 ± 2.3	[34]

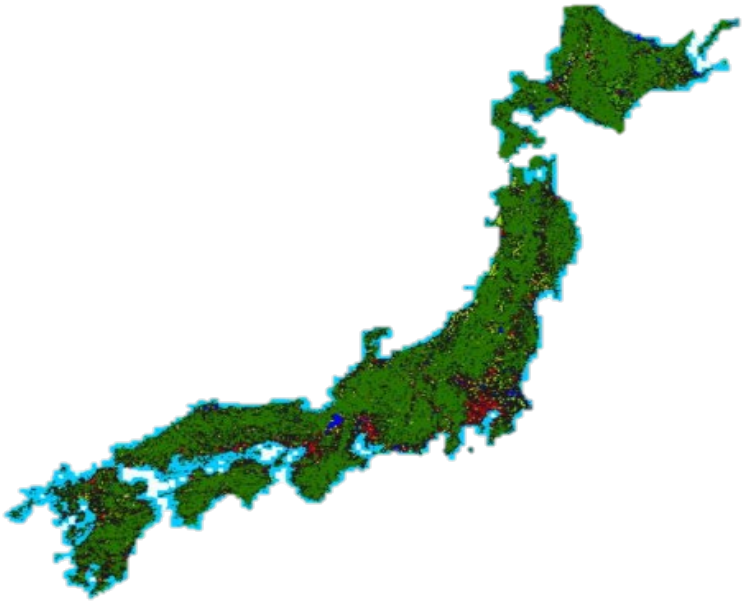


Fig. Grid level microplastic emissions (floating and suspended)

ANNEXURE 5

Research publications on microplastics in wastewater treatment plants (WWTPs)

Microplastic-related research has grown over the past decade. Research reported in peer-reviewed journals indicates a monotonically increasing trend in microplastics-associated research, especially in the last 10 years (Figure 1.1, blue bars). Compared to the general research topic of microplastics, there is less research focused on microplastics in wastewater (Figure 1.1, red bars). Until 2020, wastewater- and microplastics-related research comprised less than 10% of the total published research discussing the issue of microplastics. At present, however, examinations of microplastics in WWTPs, targeting of sampling and analytical methods of microplastics in wastewater matrices, and the removal efficiency of microplastics at various levels/configurations in relation to different types of WWTPs are attracting greater research attention. The growing focus on the development of analytical methods is understandable, given that microplastics in wastewater remains a relatively novel topic, and the complexity of wastewater matrices (Parrish and Fahrenfeld 2019) has led to complications in sampling, recovery, and detection of microplastics. Apart from the peer-reviewed publications, in recent years, the European Union [EU: Sabbah et al. (2019)] and the United States Environmental Protection Agency (USEPA) funded projects (Cook & Allen, 2020) have issued best practice recommendations for detecting and analyzing microplastic concentrations in wastewater. Recent developments in terms of sampling/analytical methodologies and identification of best practices for collection, preparation, and analysis are expected to incentivize continued research on microplastics in wastewater. The recent increment in the share of publications on microplastics in early 2021 (for the first time reaching over

10% for microplastics in wastewater among relevant publications) (Figure 1.2) is probably associated with these developments.

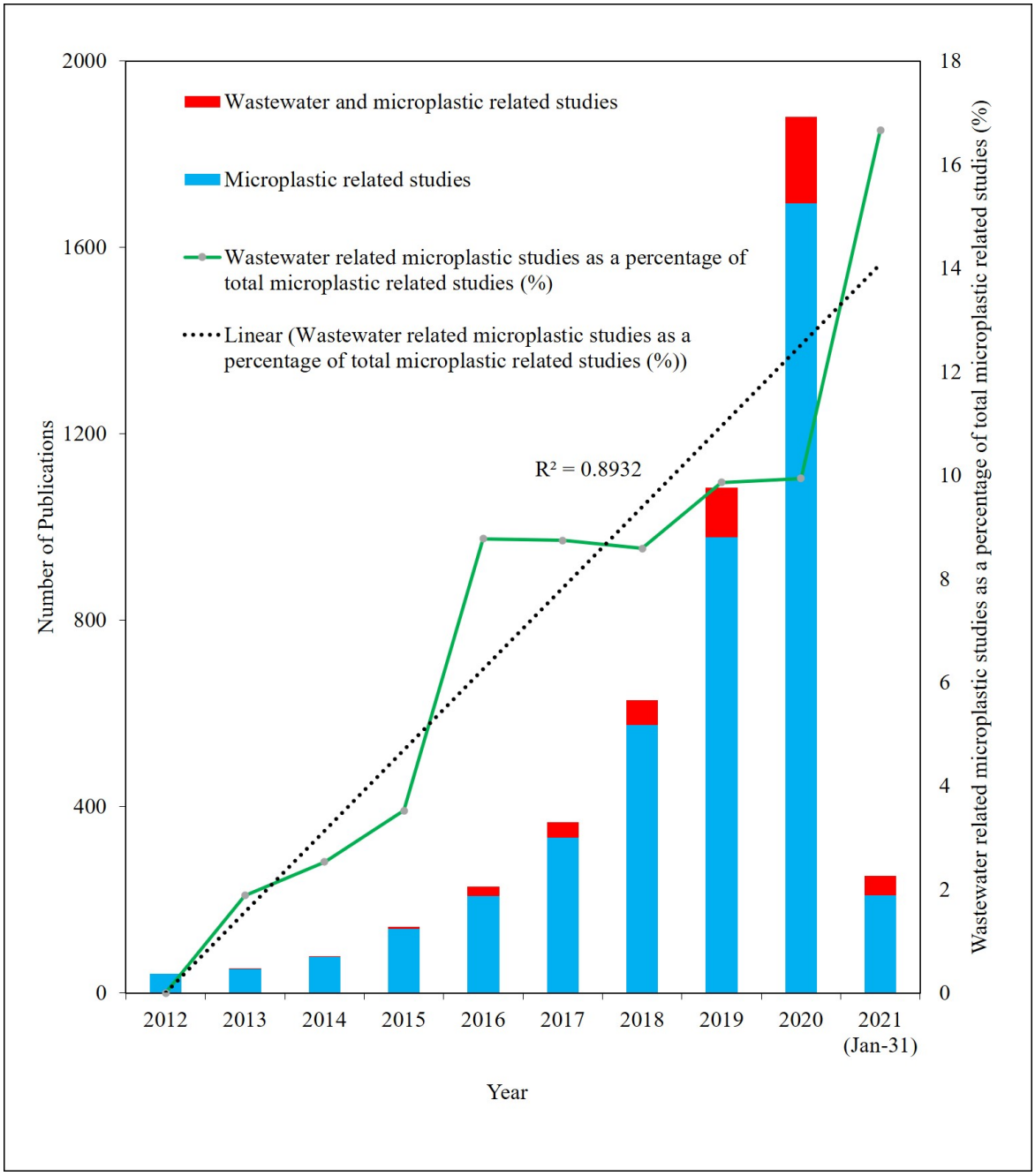


Figure A5.1. Research report***ing*** in peer-reviewed journals ***indicating the*** increasing trend in microplastics-associated research and microplastic-related research targeting wastewater treatment plants. (Scopus keyword search, January 31, 2021.)

Nevertheless, at present, a limited number of studies are focused on the biodegradability of microplastics in wastewater treatment plants and chemical leakage of microplastics in wastewater treatment systems. As of January 31, 2021, only three publications (out of a total of 4858 publications on microplastics) discussed the biodegradability of microplastics in WWTPs. Despite this low number, the growing attention on biobased and biodegradable plastics (Lakhawat et al, 2020) in the plastic industry may potentially lead to further examination of biodegradability in wastewater treatment plants and sludge disposal options, including land applications (Keller et al, 2019). Such a trend may generate additional research funds for exploring possible countermeasures at WWTPs to address microplastic leakages into the environment.

In sum, recent developments of solid protocols and testing methods, fine-tuning of analytical equipment, and increased attention on biodegradable plastics, *inter alia,* microplastics, and wastewater-related research can be expected to increase in the near future.

To date. more than 50% of the microplastic-related research has been published in three journals: [Marine Pollution Bulletin (MPB), *Science of the Total Environment* (STE), and *Environmental Pollution* (EP), respectively (Figure 1.3A)]. Microplastic-related pollution studies were initially focused on marine environments where plastic accumulation and degradation mostly occurs (Min et al., 2020). Sampling by a vessel dragging manta-nets in marine environments provided evidence for the presence of small-sized anthropogenic particles (microplastics). Moreover, land-based primary or secondary microplastics have also been found to contaminate ocean water (Lebreton et al., 2017; Abeynayaka et al., 2020). In this context, various studies on land-based

microplastics emissions have been published in journals such as MPB, where the scope has been limited to marine pollution. The scope of STE recommends research focusing on more than one environmental compartment, such as terrestrial and freshwater or atmospheric and marine. Therefore, as microplastic-related research is often multi-compartmental (due to properties such as high mobility and longevity), such research matches the scope of STE. Moreover, the development of sampling and analytical methods to detect microplastics in soil, freshwater, and other biota are also relevant to microplastic-related research in line with journals such as *Environmental Science and Technology* (EST) and *Water Research* (WR).

With regard to microplastics and wastewater-related research, journals such as STE, WR, MPB, and *Environmental Pollution* (EP) are some of the leading publications (Figure 1.3B). Wastewater-related studies often focus on the origins of microplastics (mostly land-based), wastewater treatment plant operations (in view of microplastic discharge into water environments), and microplastic transfer from wastewater to sludge (inter-compartmental movement) in WWTPs. As mentioned previously, this is in line with the scope of STE, with research focusing on more than one environmental medium. Research focusing extensively on WWTP performance and microplastics contamination of freshwater environments are more aligned with the scope of journals such as WR. Interested readers can review the top listed journals for microplastics- and wastewater-related research publications outlined Figure 1.3. A detailed analysis of research publications on the topic of microplastics and wastewater indicates that the top-cited studies are mostly published in WR (data not shown here).

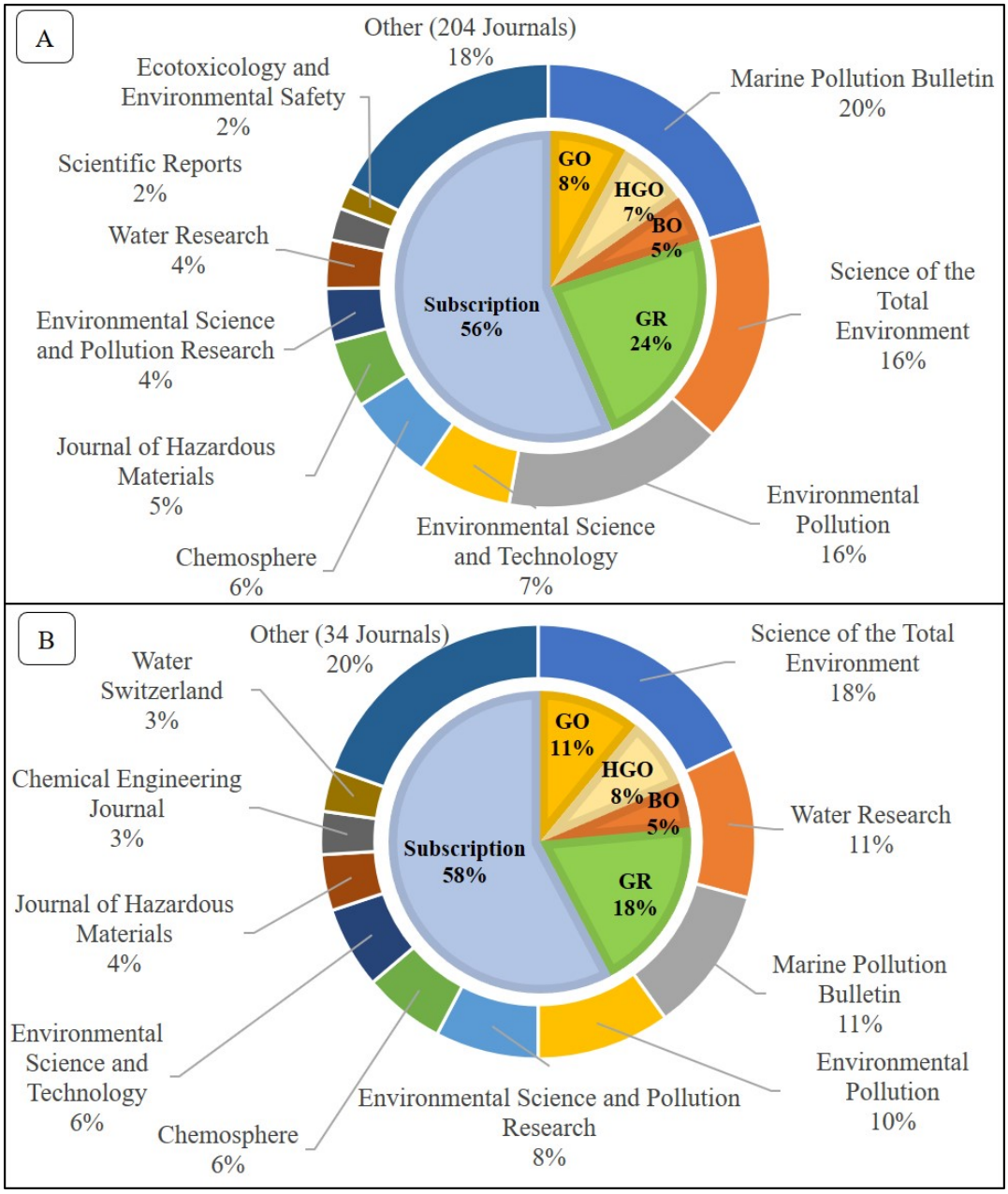
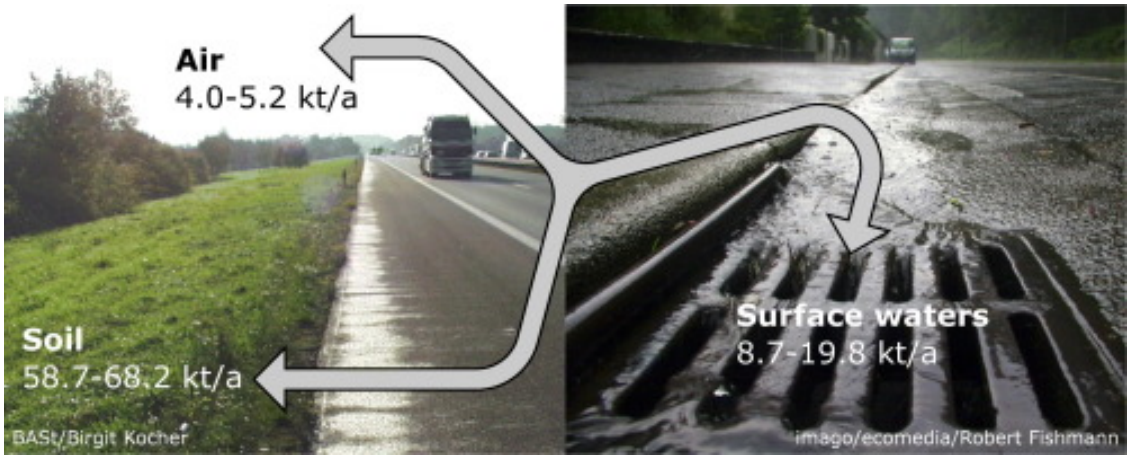


Figure A5.2. (**A**) Top journals reporting on microplastics and (**B**) top journals reporting on microplastics and wastewater. Donuts represent top journals publishing articles, and pie charts inside donuts represent accessibility of articles. (Scopus keyword search January 31, 2021). GO, gold open access; HGO, hybrid gold open access; BO, bronze open access; GR, green repository. GO journals only publish open access; HGO journals allow authors to choose open access; BO journals provide temporary or permanent free access; GR indicates that manuscripts are accepted for publication and available at a repository.

The accessibility options of published literature are given in pie charts in Figure 1.3. In both cases, microplastics and microplastics and wastewater, the subscription-based

sources share most of the published articles. However, there is a considerable proportion available for readers without subscriptions due to the options of BO and GR. This provides opportunities for researchers in developing countries where subscriptions may not always be available. Moreover, the benefits are extended to project managers, policymakers, staff of funding organizations, etc., whose associated organizations are not often subscribers.



ANNEXURE 6

Table A6.1. Microplastics potential toxic element effects reported in literature

Microplastic/plastic			Plant	Effect	Reference
Types	**Size (mm)**	**Concentration (% w/w)**			
Polystyrene (PS)	0.55~56	2	Onion (*Allium fistulosum*)	Increase root biomass, total root length and mean diameter	de Souza Machado et al., 2019
	<0.00005	-	Garden onion (*Allium cepa*)	The root length was inhibited	Giorgetti et al., 2020
	0.1-0.15	0.1-10	Corn (*Zea mays*)	Plant root biomass decreased	Wang et al., 2020a
	D= 0.001	-	Garden Lettuce (*Lactuaca sativa*)	The PS is transported through the vascular system to the stem and leaves	Li et al., 2020
	0.0001	5	Wheat (*Triticum aestivum*)	The root length increased, the root/shoot ratio decreased, and the biomass increased	Lian et al 2020
	5	-	Broad bean (*Vicia faba*)	Decreased biomass and catalase enzymes activity, blocked cell connections or cell wall pores for transportof nutrients in roots	Jiang et al 2019
	D=<0.001	-	Rice (*Oryza sativa*)	PS mostly aggregated in the vascular systems of the roots, stems and leaves and high possibility to enter food chain	Liu et al 2022
	<50	-	Rice (*Oryza sativa*)	Higher doses of PS caused ≈40% decrease in shoot biomass	Wu et al., 2020
	0.01	-	Rice (*Oryza sativa*)	Affect transpiration and stomata of rice seedlingsmainly via inhibiting their root vigor	Dong et al., 2020
	<0.048	-	Garden cress (Lepidium sativum)	Significant decline in germinate rate and plant growth inhibition	Bosker et al., 2019

	<0.001	-	Carrot (*Daucus carota L.*)	Enter to the roots and accumulated in intercellular layer and particles were able to translocate to the leaves	Dong et al., 2021
	0.001	-	Lettuce (*Lactuca sativa L., Rosa*)	Adherence, uptake, accumulation, and translocation of PS in the vascular tissue	Li et al. 2019
Low Density Polyethylene (LDPE)	L: 4-10	1	Garden Lettuce (*Lactuaca sativa*)	The total biomass decreased and the composition of rhizosphere bacterial community changed	Li et al., 2020
	L= 6.9 ;W= 6.1	1	Wheat (*Triticum aestivum*)	The fruit biomass and leaf number decreased	Ze-quan et al., 2010
	-	1	Wheat (*Triticum aestivum*)	Effect to Vegetative and reproductive growth	Qi et al 2018
	0.053-1	0.2-2.5	Bean (*Phaseolus vulgaris*)	Aboveground and root biomass affected but effect it non significant	Meng et al. 2021
	L=5 , W= 5	0.1-0.4	Carrot (*Daucus carota*)	Aboveground biomass and root mass decreased with increasing concentration	Lozano et al., 2021
Poly Lactic Acid (PLA)	0.1-0.15	0.1-10	Corn (*Zea mays*)	High concentration significantly reduced plant biomass	Wang et al., 2020a
	0.065	0.1-0.001	Perennial ryegrass (*Lolium perenne*)	Reduction in shoot height and biomass	Boots et al 2019
	-	-	Bean (*Phaseolus vulgaris*)	Root and aboveground biomass redued	Meng et al. 2021
Polyethylene (PE)	D= 0.2-0.25	0.5-8	Wheat (*Triticum aestivum L.*)	High concentration of PE is damage to the antioxidant system in wheat roots	Liu et al 2021
	0.003	-	Corn (Zea mays)	PE reduced or block water and nutrients uptake as well as growth of maize plant	Urbina et al., 2020
	0.5	0.1-10	Lettuce (*Lactuca sativa L.*)	Increase the toxicity, uptake, accumulation, and bioavailability of heavy metals	Wang et al 2021

High Density Polyethylene (HDPE)	-	0.1-0.001	Carrot (*Daucus carota*)	Shoot height and biomass is reduced, fewer seeds germinated	Boots et al 2019
	0.01-0.15	0.1-10		There was no significant change in plant biomass	Lozano et al., 2021
Polyamide (PA)	0.015-0.02	2	Onion (*Allium fistulosum*)	Significant effect in plant biomass, root traits, tissue elemental composition, soil microbial activity	de Souza Machado et al., 2019
	0.015-0.02	2	Wheat (*Triticum aestivum*)	The total biomass increased, and the total root length and mean diameter increased	Lian et al., 2020
Polypropylene (PP)	-	0.02	Garden cress (*Lepidium sativum*)	Occurrence of oxidative burst	Pignattelli et al 2020
	L=5, W= 5	0.1-0.4	Carrot (*Daucus carota*)	Aboveground biomass and root mass decreased with increasing concentration	Lozano et al., 2021
Polyester fibers (PFs)	L=5, D= 0.008	0.2	Onion (*Allium fistulosum*)	Significant changes in plant biomass, root traits, tissue elemental composition, soil microbial activity	de Souza Machado et al., 2019
	L=1.3, D=0.03	-	Grasses (*Festuca brevipila*) and Herbs (*Achillea millefolium*)	Decreased in biomass	Lozano and Rilling 2020
Polyether Sulfone (PES)	L=5, D=0.008	0.2	Onion (*Allium fistulosum*)	The total biomass and root biomass increased, the total root length and mean diameter increased, and the root microbial activity increased	de Souza Machado et al., 2019
	L=1.3, D= 0.03	0.4	Wood small-reed (*Calamagrostis epigejos*)	The root biomass increased	Lozano and Rilling, 2020
Expandable Polystyrene (EPS)	8.3	-	Mung bean (*Phaseolus radiates*), Lettuce (*Lactuca sativa*), and Rice (*Oryza sativa*)	Low levels of interaction with the crop dependent and water absorption rate	Kim et al 2019

Polyvinyl chloride (PVC)	0.018-0.15	0.5-2	Lettuce (*Lactuva sativa L.*)	PCV-a promoted carotenoid synthesis, whereas PVC-b inhibited it	Li, et al., 2020
Melamine Phenolic (MP)	0.0048	-	Garden cress (*Lepidium sativum*)	Accumulated on the root hairs, germination rate was significantly reduced, physical blockage of the pores in the seed capsule	Bosker et al 2019
Polyetherimide (PEIs)	-	0.01-0.1	Oat (*Avena sativa*), Radish (*Raphanus sativus*)	Nitrogen released from the tested PEIs but no harmful effect, harmful to plants only at high concentrations	Rychter et al 2019

Table A6.1 The elemental analysis using XRF for the Micropalstics

Sample #	P	S	Cl	Ca	Ti	Fe	Ni	Cu	Zn	Br	Sb	I	Ba	Cd	Pb	Cr	Hg	As
0132-0002	31.94	77.57	78	0	172.77	1.01	0.03	0	0.48	0	0.27	0	0	0	0	0	0	0
0132-0009	2.41	30.46	221	3.63	2.71	0.71	0.09	0.01	0	0	0.63	0.97	0	0	0	1	0	0.04
0133-0002	25.81	17.58	6	0	0.41	0	0.07	0.04	0.04	0	0.87	0.11	0	0	0	0	0	0
0133-0005	10.07	29.61	0	0	95.03	0.08	0.1	0	0	0	0.67	0.07	0	0	0	0	0	0
0133-0006	9.17	158.7	849	3.9	0.63	3.32	0.1	0	0.03	1	0.78	0	0	0	0	1	0	0
0133-0005	8.79	129.61	50	55.06	71.44	5.82	0.44	0.04	0.8	0	0.86	0	0	0	0	1	0	0
0112-0001	3.37	198.2	13844	41.83	234.46	198.18	0	0.08	0.23	6	1.07	2.05	7.02	0	0	4	0	0.01
0112-0002	0	247.45	28852	56.34	160.79	230.23	0.51	0	0.33	20	0.12	2.76	7.77	0	1	4	0	0
0112-0011	36.76	99.7	294	4.82	30.21	6.41	0.04	0.02	0.02	0	0.28	1.28	0.62	0	0	1	0	0

0113-0003	35.57	123.12	2337	537.36	603.06	7.84	0	0.16	0.05	2	0.11	0.76	4.46	0	0	0	0	0
0113-0005	0	129.99	744	18.46	282.48	6.93	0	0.13	7.22	1	0	0.31	4.98	0	0	13	0	0
0113-0006	0.04	118.88	844	23.21	53.78	5.01	0.07	0.04	1.06	1	0	1.57	2.91	0	0	0	0	0
0113-0008	16.81	260.15	358	37.91	68.76	6.92	0.07	0	0.32	0	0.71	0.58	0	0	1	5	0	0
0115-0001	5.67	33.54	41	14.38	62.2	0	0.05	0	0.23	0	0.94	0	1.63	0	0	0	0	0
0116-0001	57.89	151.51	1876	288.73	172.58	4.03	0.11	0	0.46	0	0.64	0.19	1.53	0	0	0	0	0.01
0154-0001	7.41	539.91	753	4.9	11.11	18.12	0.13	0.16	0.24	1	0.8	0	0.1	0	0	0	0	0
0154-0013	1.4	228.51	1692	7.65	29.35	58.96	5.28	0	0.01	1	0.8	0	0	0	0	16	0	0.01
0165-0024	2.65	119.55	95	0	3.72	6.56	0.09	0.09	0	11	0.63	1.35	0.59	0	0	1	0	0
0180-0018	0	277.59	13425	144.11	103.11	73.03	0.03	0.81	0.05	1179	0.28	0.67	3.83	0	2	2	1	0
0184-0021	2.93	156.05	11632	32.46	31.9	21.88	0	0	0.3	21	0.86	0.07	0	0	0	0	0	0
0183-0022	0	74.42	3417	9.77	27.57	16.87	0	0.1	0.25	2	0.04	0.18	1.06	0	0	1	0	0.02
X47	115.3	231.83	221	7.57	35.37	949.71	0.12	5.1	3.3	0	0	0	1.48	0	0	0	0	0
X48	66.06	242.08	556	27.56	1382.6	859.69	0.05	7.69	9.83	0	0	0.62	2.68	0	0	0	0	0
X49	238.06	604.52	105	34.59	189.22	480.34	1.51	1.96	19.87	0	0	0.57	0	0	0	1	0	0
0121-0005	11.16	170.18	514	14.25	94.88	2.18	0	0.09	0.21	0	0.7	1.27	0	0	0	0	0	0
0121-0006	13.3	274.79	0	1.09	2.15	1.12	0.05	0.04	0	0	0.43	0.21	0	0	0	1	0	0.02
0121-0008	30.21	1469.5	2575	393.38	227.35	13.12	0.03	0.08	4.38	0	0.56	0	2.27	0	0	0	0	0
0117-0002	27.54	242.27	1048	0.54	0.89	1.44	0.05	0	0	0	0	0	0	0	0	1	0	0
0117-0004	25.02	64.78	3159	23.1	34.27	3.37	0.03	0.1	8.29	0	0	0.55	2.05	0	0	0	0	0
0119-0002	42.7	150.79	914	3.77	4788.58	6.36	0	0.36	0.23	2	0	0	2.69	0	0	0	0	0
0119-0004	14.7	20.57	242	4.6	0.82	0.2	0.03	0.06	0	0	0.66	0.52	0	0	0	0	0	0

0119-0007	7.85	16.11	41	1.68	0.56	0.12	0.04	0	0.05	0	0.51	1.01	0.14	0	0	0	0	0
0120-0001	1.15	382.45	1355	3.94	1.38	2.07	0.02	0	0.04	1	0.43	0	0.45	0	0	0	0	0
0120-0006	21.75	59.82	667	79.82	203.06	4.45	0.31	3.4	1.21	0	0	0	0.08	0	0	0	0	0
0122-0001	0	447.23	33709	391.68	282.03	330.69	1	0.2	2.97	3	0.63	2.89	5.05	0	1	0	0	0
0122-0023	1.82	530.37	813	9.25	1.47	0.39	2.92	0.57	0.03	0	0.43	1.39	0	0	12	30	0	0
0124-0002	119.53	149.29	4697	0	6.66	341.75	0	1.13	0.05	0	0.36	0	0	0	0	0	0	0
0124-0008	4.01	716.72	28	3	109.95	0.74	0.11	0	0.04	0	0.75	1.51	3.11	0	0	0	0	0.02
0124-0010	1.51	25.74	771	2.5	0	2.6	0.09	0.04	0.01	0	0.63	1.33	0	0	0	0	0	0.01
0126-0001	8.11	481.56	561	0	8.92	1.41	0.05	1.06	0.02	1	0.71	0	0.19	0	17	43	0	0
0126-0004	10.43	25.04	19	17.21	27.7	0.15	0.07	0.04	0.03	0	0.54	0.45	0	0	0	0	0	0
0129-0001	85.56	203.68	3048	695.76	31.33	3.46	0.28	0.42	0.01	0	1.08	0	0	0	0	0	0	0
0129-0008	25.8	88.01	1993	12.09	1717.24	0.91	0.17	0.18	0.06	0	0.44	0	0.25	0	0	1	0	0.02
BLK	18.62	16.67	0	0.49	0	0.14	0.04	0	0.03	0	0.23	0.63	1.17	0	0	0	0	0

備考1	0112-0001
備考2	
備考3	
オペレータ	Staff
測定時間	217

[測定条件]

レシピ	001 3mm_4Cond_Vac_FP-C-Bal.			
	条件1	条件2	条件3	条件4
測定時間 (秒)	30	30	30	30
デッドタイム (%)	12	1	4	3
管電圧 (kV)	15	15	50	50
管電流 (uA)	1000	1000	1000	1000
フィルター	OFF	Cr用	Pb用	Cd用
雰囲気	真空	真空	真空	真空
機種名	EA1400 ID0004 (11.07.0.0)			
コリメータ	φ3.0mm			
測定法	バルクFP C:¥XRAY¥HS¥025¥3mm_4条件.bfp			
備考	PeakingTime=1.0usec / フィルム=FILM			

[分析結果]

着目元素		濃度 (ppm)	統計誤差 3σ(ppm)	X線強度 (cps)	強度誤差 3σ(cps)	着目元素		濃度 (ppm)	統計誤差 3σ(ppm)	X線強度 (cps)	強度誤差 3σ(cps)
P	Ka	3.37	51.68	15.18	7.2	Cd	Ka	0.13	0	0.1	0.2
S	Ka	198.2	32.48	63.76	10.58	Pb	Lb	0.06	1	0.2	2.3
Cl	Ka	13843.7	1349.83	32.24	[illegible]	Cr	Ka	3.91	[illegible]	0.5	1.1
Ca	Ka	41.83	59.49	0.74	1.06	Hg	La	0	0	0.0	0.9
Ti	Ka	234.46	38	11.9	1.9	Br	Ka	5.93	1	44.3	3.9
Fe	Ka	198.18	14	68.4	4.7	As	Ka	0.01	0	0.2	1.0
Ni	Ka	0	1	0.0	1.2						
Cu	Ka	0.08	1	0.1	1.1						
Zn	Ka	0.23	1	0.4	1.2						
Sn	Ka	0.41	1.81	0.3	1.32						
Sb	Ka	1.07	2.37	0.76	1.62						
I	Ka	2.05	4.12	1.2	2.28						
Ba	Ka	7.02	8.78	2.34	2.68						

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備考1	0112-0002
備考2	
備考3	
オペレータ	Staff
測定時間	197

[測定条件]

レシピ	001 3mm_4Cond_Vac_FP-C-Bal.			
	条件1	条件2	条件3	条件4
測定時間 (秒)	30	30	30	30
デッドタイム (%)	13	1	4	3
管電圧 (kV)	15	15	50	50
管電流 (uA)	1000	1000	1000	1000
フィルター	OFF	Cr用	Pb用	Cd用
雰囲気	真空	真空	真空	真空
機種名	EA1400 ID0004 (11.07.0.0)			
コリメータ	φ3.0mm			
測定法	バルクFP C:¥XRAY¥HS¥025¥3mm_4条件.bfp			
備考	PeakingTime=1.0usec / フィルム=FILM			

[分析結果]

着目元素		濃度 (ppm)	統計誤差 3σ(ppm)	X線強度 (cps)	強度誤差 3σ(cps)	着目元素		濃度 (ppm)	統計誤差 3σ(ppm)	X線強度 (cps)	強度誤差 3σ(cps)
P	Ka	0	54.38	10.42	7.64	Cd	Ka	0.14	0	0.1	0.2
S	Ka	247.45	36.54	80.14	12	Pb	Lb	0.59	1	1.8	2.4
Cl	Ka	28852.1	1915.83	67.92	4.5[illegible]	Cr	Ka	4.22	[illegible]7	0.5	0.9
Ca	Ka	56.34	52.79	0.92	0.88	Hg	La	0	1	0.0	0.9
Ti	Ka	160.79	32	7.6	1.5	Br	Ka	20.46	1	139.2	6.9
Fe	Ka	230.23	15	73.4	4.9	As	Ka	0	0	0.0	1.1
Ni	Ka	0.51	2	0.4	1.1						
Cu	Ka	0	1	0.0	1.2						
Zn	Ka	0.33	1	0.5	1.2						
Sn	Ka	0.16	1.8	0.12	1.27						
Sb	Ka	0.12	2.66	0.1	1.77						
I	Ka	2.76	4.34	1.53	2.35						
Ba	Ka	7.77	9.99	2.57	3.01						

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備考1	0122-0001
備考2	
備考3	
オペレータ	Staff
測定時間	197

[測定条件]

レシピ	001 3mm_4Cond_Vac_FP-C-Bal.			
	条件1	条件2	条件3	条件4
測定時間(秒)	30	30	30	30
デッドタイム(%)	15	1	4	3
管電圧(kV)	15	15	50	50
管電流(uA)	1000	1000	1000	1000
フィルター	OFF	Cr用	Pb用	Cd用
雰囲気	真空	真空	真空	真空
機種名	EA1400 ID0004 (11.07.0.0)			
コリメータ	φ3.0mm			
測定法	バルクFP C:¥XRAY¥HS¥025¥3mm_4条件.bf			
備考	PeakingTime=1.0usec / フィルム=FILM			

[分析結果]

着目元素		濃度(ppm)	統計誤差3σ(ppm)	X線強度(cps)	強度誤差3σ(cps)	着目元素		濃度(ppm)	統計誤差3σ(ppm)	X線強度(cps)	強度誤差3σ(cps)
P	Ka	0	69.37	9.65	8.27	Cd	Ka	0.1	0	0.1	0.2
S	Ka	447.23	47.42	122.43	13.14	Pb	Lb	0.73	1	1.8	2.4
Cl	Ka	33708.9	2291.79	66.73	4.62	Cr	Ka	0.01	7	0.0	0.7
Ca	Ka	391.68	104.8	5.34	1.45	Hg	La	0	1	0.0	0.9
Ti	Ka	282.03	47	11.0	1.9	Br	Ka	2.74	1	15.2	2.8
Fe	Ka	330.69	20	86.3	5.3	As	Ka	0	0	0.0	1.2
Ni	Ka	1	2	0.6	1.1						
Cu	Ka	0.2	1	0.2	1.2						
Zn	Ka	2.97	1	3.8	1.5						
Sn	Ka	0.57	2.23	0.38	1.45						
Sb	Ka	0.63	2.87	0.41	1.77						
I	Ka	2.89	4.74	1.55	2.41						
Ba	Ka	5.05	11.29	1.68	3.25						

Page 1

備考1	0154-0013
備考2	
備考3	
オペレータ	Staff
測定時間	194

[測定条件]

レシピ	001 3mm_4Cond_Vac_FP-C-Bal.			
	条件1	条件2	条件3	条件4
測定時間(秒)	30	30	30	30
デッドタイム(%)	4	1	2	4
管電圧(kV)	15	15	50	50
管電流(uA)	1000	1000	1000	1000
フィルター	OFF	Cr用	Pb用	Cd用
雰囲気	真空	真空	真空	真空
機種名	EA1400 ID0004 (11.07.0.0)			
コリメータ	φ3.0mm			
測定法	バルクFP C:¥XRAY¥HS¥025¥3mm_4条件.bf			
備考	PeakingTime=1.0usec / フィルム=FILM			

[分析結果]

着目元素		濃度(ppm)	統計誤差3σ(ppm)	X線強度(cps)	強度誤差3σ(cps)	着目元素		濃度(ppm)	統計誤差3σ(ppm)	X線強度(cps)	強度誤差3σ(cps)
P	Ka	1.4	18.83	1.92	5.55	Cd	Ka	0	0	0.0	0.0
S	Ka	228.51	15.85	162.39	11.34	Pb	Lb	[illegible]	0	0.2	1.1
Cl	Ka	1692.17	309.32	8.91	1.65	Cr	Ka	15.54	[illegible]	5.8	1.4
Ca	Ka	7.65	20.57	0.39	0.95	Hg	La	0.07	0	0.4	0.3
Ti	Ka	29.35	9	4.1	1.2	Br	Ka	0.92	0	20.9	2.7
Fe	Ka	58.96	4	60.7	4.3	As	Ka	0.01	0	0.3	0.4
Ni	Ka	5.28	1	13.3	2.0						
Cu	Ka	0	0	0.0	0.9						
Zn	Ka	0.01	0	0.1	1.1						
Sn	Ka	0.52	0.48	0.49	0.44						
Sb	Ka	0.8	0.62	0.69	0.53						
I	Ka	0	1.7	0	1.12						
Ba	Ka	0	4.12	0	1.48						

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備考1	0133-0005
備考2	
備考3	
オペレータ	Staff
測定時間	191

[測定条件]

レシピ	001 3mm_4Cond_Vac_FP-C-Bal.			
	条件1	条件2	条件3	条件4
測定時間 (秒)	30	30	30	30
デッドタイム (%)	3	1	2	4
管電圧 (kV)	15	15	50	50
管電流 (uA)	1000	1000	1000	1000
フィルター	OFF	Cr用	Pb用	Cd用
雰囲気	真空	真空	真空	真空
機種名	EA1400 ID0004 (11.07.0.0)			
コリメータ	φ3.0mm			
測定法	バルクFP C:¥XRAY¥HS¥025¥3mm_4条件.bf			
備考	PeakingTime=1.0usec / フィルム=FILM			

[分析結果]

着目元素		濃度 (ppm)	統計誤差 3σ(ppm)	X線強度 (cps)	強度誤差 3σ(cps)	着目元素		濃度 (ppm)	統計誤差 3σ(ppm)	X線強度 (cps)	強度誤差 3σ(cps)
P	Ka	10.07	13.58	3.23	4.33	Cd	Ka	0	0	0.0	0.0
S	Ka	29.61	8.84	22.97	6.89	Pb	Lb	0.03	0	0.4	1.0
Cl	Ka	0	95.41	0	0.56	Cr	Ka	0.08	0	0.0	0.1
Ca	Ka	0	11.94	0	0.63	Hg	La	0.05	0	0.3	0.3
Ti	Ka	95.03	14	15.3	2.2	Br	Ka	0.24	0	6.5	1.7
Fe	Ka	0.08	1	0.1	0.8	As	Ka	0	0	0.0	0.3
Ni	Ka	0.1	0	0.3	0.3						
Cu	Ka	0	0	0.0	0.8						
Zn	Ka	0	0	0.0	0.8						
Sn	Ka	0.52	0.42	0.49	0.39						
Sb	Ka	0.67	0.58	0.59	0.5						
I	Ka	0.07	1.39	0.12	0.93						
Ba	Ka	0	3.88	0	1.36						

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備考1	0126-0001
備考2	
備考3	
オペレータ	Staff
測定時間	192

[測定条件]

レシピ	001 3mm_4Cond_Vac_FP-C-Bal.			
	条件1	条件2	条件3	条件4
測定時間 (秒)	30	30	30	30
デッドタイム (%)	3	1	2	3
管電圧 (kV)	15	15	50	50
管電流 (uA)	1000	1000	1000	1000
フィルター	OFF	Cr用	Pb用	Cd用
雰囲気	真空	真空	真空	真空
機種名	EA1400 ID0004 (11.07.0.0)			
コリメータ	φ3.0mm			
測定法	バルクFP C:¥XRAY¥HS¥025¥3mm_4条件.bf			
備考	PeakingTime=1.0usec / フィルム=FILM			

[分析結果]

着目元素		濃度 (ppm)	統計誤差 3σ(ppm)	X線強度 (cps)	強度誤差 3σ(cps)	着目元素		濃度 (ppm)	統計誤差 3σ(ppm)	X線強度 (cps)	強度誤差 3σ(cps)
P	Ka	8.11	15.26	2.67	4.84	Cd	Ka	0	0	0.0	0.0
S	Ka	481.56	16.63	372.37	12.87	Pb	[illegible]	17.12	1	180.9	7.7
Cl	Ka	560.96	209.22	3.21	1.[illegible]	Cr	Ka	42.93	[illegible]	18.3	2.4
Ca	Ka	0	12.97	0	0.67	Hg	[illegible]	0	0	0.0	0.9
Ti	Ka	8.92	6	1.4	1.0	Br	Ka	0.57	0	14.2	2.4
Fe	Ka	1.41	1	1.7	0.7	As	Ka	0	1	20.0	8.1
Ni	Ka	0.05	0	0.2	0.5						
Cu	Ka	1.06	0	4.4	1.3						
Zn	Ka	0.02	0	0.2	0.9						
Sn	Ka	0.32	0.33	0.31	0.31						
Sb	Ka	0.71	0.51	0.62	0.44						
I	Ka	0	1.57	0	1.05						
Ba	Ka	0.19	3.91	0.07	1.37						

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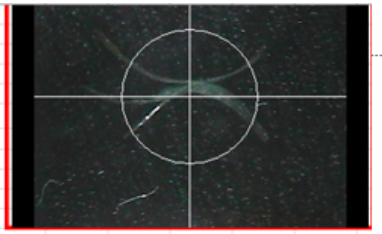
備考1	0122-0023
備考2	
備考3	
オペレータ	Staff
測定時間	196



[測定条件]

レシピ	001 3mm_4Cond_Vac_FP-C-Bal.			
	条件1	条件2	条件3	条件4
測定時間 (秒)	30	30	30	30
デッドタイム (%)	3	1	2	3
管電圧 (kV)	15	15	50	50
管電流 (uA)	1000	1000	1000	1000
フィルター	OFF	Cr用	Pb用	Cd用
雰囲気	真空	真空	真空	真空
機種名	EA1400 ID0004 (11.07.0.0)			
コリメータ	φ3.0mm			
測定法	バルクFP C:¥XRAY¥HS¥025¥3mm_4条件.bfp			
備考	PeakingTime=1.0usec / フィルム=FILM			

[分析結果]

着目元素		濃度 (ppm)	統計誤差 3σ(ppm)	X線強度 (cps)	強度誤差 3σ(cps)	着目元素		濃度 (ppm)	統計誤差 3σ(ppm)	X線強度 (cps)	強度誤差 3σ(cps)
P	Ka	1.82	14.04	0.64	4.46	Cd	Ka	0	0	0.0	0.0
S	Ka	530.37	16.87	410.26	13.09	Pb	[illegible]	[illegible]	1	122.6	6.4
Cl	Ka	812.7	241.89	4.65	[illegible]	Cr	Ka	29.82	5	12.7	2.0
Ca	Ka	9.25	8.29	0.54	0.43	Hg	[illegible]	[illegible]	0	0.0	0.7
Ti	Ka	1.47	2	0.2	0.3	Br	Ka	0	0	0.1	1.3
Fe	Ka	0.39	1	0.5	0.8	As	Ka	0	0	17.3	6.8
Ni	Ka	2.92	1	8.5	1.7						
Cu	Ka	0.57	0	2.6	1.3						
Zn	Ka	0.03	0	0.2	1.1						
Sn	Ka	0.14	0.22	0.14	0.21						
Sb	Ka	0.43	0.4	0.38	0.34						
I	Ka	1.39	0.86	0.96	0.57						
Ba	Ka	0	3.36	0	1.37						

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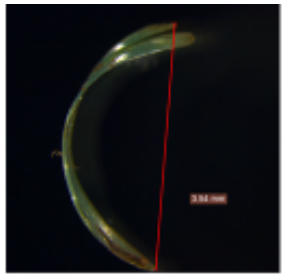
備考1	X48
備考2	
備考3	
オペレータ	Staff
測定時間	193

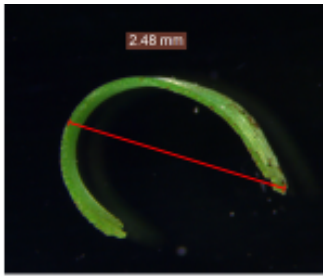
[測定条件]

レシピ	001 3mm_4Cond_Vac_FP-C-Bal.			
	条件1	条件2	条件3	条件4
測定時間 (秒)	30	30	30	30
デッドタイム (%)	11	1	3	3
管電圧 (kV)	15	15	50	50
管電流 (uA)	1000	1000	1000	1000
フィルター	OFF	Cr用	Pb用	Cd用
雰囲気	真空	真空	真空	真空
機種名	EA1400 ID0004 (11.07.0.0)			
コリメータ	φ3.0mm			
測定法	バルクFP C:¥XRAY¥HS¥025¥3mm_4条件.bfp			
備考	PeakingTime=1.0usec / フィルム=FILM			

[分析結果]

着目元素		濃度 (ppm)	統計誤差 3σ(ppm)	X線強度 (cps)	強度誤差 3σ(cps)	着目元素		濃度 (ppm)	統計誤差 3σ(ppm)	X線強度 (cps)	強度誤差 3σ(cps)
P	Ka	66.06	25.87	20.37	7.66	Cd	Ka	0.03	0	0.0	0.1
S	Ka	242.08	17.75	172.8	12.78	Pb	Lb	0.05	0	0.4	2.1
Cl	Ka	555.76	229.86	3.21	1.24	Cr	Ka	0.25	3	0.1	0.9
Ca	Ka	27.56	23.78	1.3	1.14	Hg	La	0	0	0.0	0.7
Ti	Ka	1382.6	55	197.2	7.8	Br	Ka	0.03	0	0.6	1.6
Fe	Ka	859.69	17	867.2	16.5	As	Ka	0	0	0.0	1.0
Ni	Ka	0.05	0	0.1	1.0						
Cu	Ka	7.69	1	25.7	3.0						
Zn	Ka	9.83	1	49.5	4.2						





0119-0007	7.85	12.1	16.1	8.15	41	67	1.68	8.75	0.56	1.1	0.12	0.66	0.04	0.07	0	0.19	0.05	0.15	0	0	0.51	0.43	1.01	0.72	0.14	2.74	0	0	0	0	0	1	0	0	0	0
0120-0001	1.15	17.2	382	17.4	1355	271	3.94	7.97	1.38	1.74	2.07	0.85	0.02	0.18	0	0.21	0.04	0.14	1	0	0.43	0.93	0	2.04	0.45	5.01	0	0	0	0	0	1	0	0	0	0
0120-0006	21.8	18.3	59.8	11.8	667	214	79.8	25.7	203	20.2	4.45	1.13	0.31	0.42	3.4	0.57	1.21	0.28	0	0	0	1.31	0	1.95	0.08	4.94	0	0	0	0	0	1	0	0	0	0
0122-0001	0	69.4	447	47.4	38188	2292	392	105	282	47.5	331	20.3	1	1.77	0.2	1.41	2.97	1.19	3	1	0.63	2.87	2.89	4.74	5.05	11.3	0	0	1	1	0	7	0	1	0	0
0122-0023	1.82	14	530	16.9	813	242	9.25	8.29	1.47	1.72	0.39	0.69	2.92	0.6	0.57	0.32	0.03	0.19	0	0	0.43	0.4	1.39	0.86	0	3.36	0	0	12	1	30	5	0	0	0	0
0124-0002	120	20.5	149	13.7	4697	503	0	11.6	6.66	7.83	342	10.1	0	0.29	1.13	0.39	0.05	0.22	0	0	0.36	0.92	0	1.96	0	4.11	0	0	0	0	0	2	0	0	0	0
0124-0008	4.01	14.3	717	18.7	28	43	3	4.32	110	15.1	0.74	0.59	0.11	0.11	0	0.2	0.04	0.14	0	0	0.75	0.52	1.51	0.86	3.11	4.28	0	0	0	0	0	2	0	0	0.02	0
0124-0010	1.51	13.8	25.7	8.62	771	205	2.5	14.3	0	3.58	2.6	0.88	0.09	0.1	0.04	0.22	0.01	0.15	0	0	0.63	0.43	1.33	0.9	0	3.24	0	0	0	0	0	0	0	0	0.01	0
0126-0001	8.11	15.3	482	16.6	561	209	0	13	8.92	6.27	1.41	0.61	0.05	0.16	1.06	0.32	0.02	0.15	1	0	0.71	0.51	0	1.57	0.19	3.91	0	0	17	1	43	6	0	0	0	0
0126-0004	10.4	13.9	25	8.51	19	118	17.2	18.7	27.7	7.33	0.15	0.63	0.07	0.11	0.04	0.21	0.03	0.15	0	0	0.54	0.44	0.45	1.1	0	3.32	0	0	0	0	0	0	0	0	0	0
0129-0001	85.6	20.3	204	14.9	3048	398	696	64.7	31.3	8.04	3.46	1.03	0.28	0.36	0.42	0.33	0.01	0.21	0	0	1.08	0.63	0	1.72	0	4.07	0	0	0	0	0	1	0	0	0	0
0129-0008	25.8	18	88	12.6	1993	345	12.1	16.3	1717	59.4	0.91	0.8	0.17	0.15	0.18	0.28	0.06	0.15	0	0	0.44	0.8	0	1.9	0.25	4.18	0	0	0	0	1	1	0	0	0.02	0
BLK	18.6	11.9	16.7	7.63	0	0	0.49	1.91	0	0	0.14	0.51	0.04	0.07	0	0.14	0.03	0.08	0	0	0.23	0.29	0.63	0.59	1.17	2.18	0	0	0	0	0	0	0	0	0	0