TRANSPORTATION PATHWAYS OF LAND SOURCE BASED MICROPLASTICS INTO THE MARINE ENVIRONMENTS: THE CONTEXT OF RIVERS

A. H. M. ENAMUL KABIR

Division of Environmental Engineering, Graduate School of Sciences and Technology for Innovation, Yamaguchi University, Ube, Yamaguchi, Japan. Email: shimul.enamul778@gmail.com

MASAHIKO SEKINE

Division of Environmental Engineering, Graduate School of Sciences and Technology for Innovation, Yamaguchi University, Ube, Yamaguchi, Japan. Email: ms@yamaguchi-u.ac.jp

TSUYOSHI IMAI

Division of Environmental Engineering, Graduate School of Sciences and Technology for Innovation, Yamaguchi University, Ube, Yamaguchi, Japan. Email: imai@yamaguchi-u.ac.jp

KOICHI YAMAMOTO

Division of Environmental Engineering, Graduate School of Sciences and Technology for Innovation, Yamaguchi University, Ube, Yamaguchi, Japan. Email: k_yama yamaguchi-u.ac.jp

ABSTRACT

Marine microplastics pollution has been a global emerging threat. Rivers have been identified as the major transporting pathways of microplastics from land sources into marine environments. This study investigated microplastics in the two rivers i.e. Awano River (AR) and Majime River (MR) which are flowing into the Sea of Japan (SJ) and the Seto Inland Sea (SIS) respectively in the Yamaguchi prefecture, Japan. River surface water samples were collected from the selected stations (n=12) from the upstream to downstream directions. Filtration, wet peroxidation, and density separation methods were applied to extract microplastics. Polymers were identified through ATR-FTIR analysis. The mean microplastics abundance results (AR-131±17.12 n/L, MR- 272.5±299.15 n/L) revealed high-level pollution. Characterization demonstrated that small microplastics (<1 mm) in sizes, fibers and fragments in shapes were predominant characteristics for these rivers. Polymer identification results revealed that polyethylene, polypropylene, vinylon and polyethylene terephthalate were the major polymers. Both the point and non-point sources could release MPs. The estimated loadings (AR-92.36 billion particles per day and MR- 3.48 billion particles per day by numbers) into the SJ and SIS marine environments represented the fate of large quantities Japan land originated MPs as well as same major polymer types in these rivers and marine environments indicated that the Japan land contributed to microplastics pollution largely. Overall, the river freshwater systems were found to be highly MPs polluted and the prominent MPs transporting pathways from land-sources to the marine environments.

Key Words: Japan, microplastics, marine environment, river, transportation.

1. INTRODUCTION

Plastics, the synthetic polymers, have entered in all aspects of man-made uses. There are ever-growing plastic uses and productions from the mid-20th century to the current date (Plastic Europe, 2019). However, today, the disadvantages of plastics are visible. Aquatic environments are polluted with plastics globally. The microplastics (MPs) i.e. the tiny plastic particles (~5 mm) in size, has been regarded as the highly threatening form of plastic pollution in the aquatic systems. The marine MPs pollution is ubiquitous worldwide. The MPs abundances, distributions, types, and characters (shape-size-color-polymer), ecotoxicological threats across the marine environments are well speculated in recent times (GESAMP 2015; Cole et al. 2011; Andrady 2011; Auta et al. 2018). However, the freshwater MPs pollution knowledge are in lack relatively, currently growing attentions of scientific the community. Specifically, river freshwater studies are highly important to understand the freshwater pollution along with their occurrences and transformation from land sources to ocean, threats, and impacts in freshwater ecosystems alongside the marine systems (Li et. al. 2018; Kataoka et al. 2019; Mendoza et al. 2018).

It is known that the MPs are of anthropogenic origin from land-based environments- occurs from manufactured plastic particles in various products (primary microplastics) and the fragmentation of larger plastics (secondary microplastics) under different environmental processes (weathering, UV exposure,

biodegradation, and physical stress etc.) Considering the primary MPs, 98% of them are originated from the land-based sources (Cole et al. 2011; Andrady 2011; Boucher and Friot, 2017). This might be because of ubiquitous man-made uses in all aspects- domestics, personal care, industries, agriculture, packaging, and transportations, building and constructions, electrical and appliances and many more (Plastic Europe 2019; GESAMP 2015). And the aquatic systems receive MPs from these land-based sources (Jambeck et al. 2015; Lebreton et al. 2017). However, less well understandings have been established yet regarding the influences of land uses over point and non-point source types behind MPs pollution occurrences in both the river freshwater and marine aquatic systems alongside for the pollution transformation by the river MPs emissions to the receiving marine systems. Studies indicated that anthropogenic land uses over point sources (i.e. population density, domestic sewages, WWTPs, industries releases etc.) might affect the MPs pollution occurrences (Murphy et al. 2016; Lechner and Ramler 2015) while several other studies indicated no significant relation in such contexts (Nel et al. 2017; Klein et al. 2015; Vaughan et al., 2017). Besides, non-point sources (agricultural, urban, atmospheric fall outs and airborne etc.) might also be the major contributors of aquatic MPs (Dris et al. 2016; Driedger at al. 2015). Hence, considering the ubiquitous uses of plastics in the terrestrial environments, we hypothesized that land uses over both the point and non-point sources affects the MPs pollution occurrences in the aquatics systems. Furthermore, these knowledge are of higher importance for the pollution management furtherly (Kataoka et al. 2019; McNeish 2018).

On the other hand, the marine aquatic environments have been thought to be the ultimate sink of these MPs. And these are being transported from these land-based sources to the marine aquatic systems through various pathways. Rivers connect lands to seas presenting a prominently major MPs transportation pathway from land to seas (Jambeck et al. 2015; Lebreton et al. 2017; Andrady, 2011; Lechner et al. 2014). Andrady et. al. estimated 80% of plastic in the sea originates from inland sources and were emitted by rivers. Lebreton et al. 2017 estimated 1.15 and 2.41 million tons entering the ocean by rivers. Already although it is acknowledged that the river environments feed inland terrestrial MPs to the marine, however, still the quantitative and qualitative information on abundance, distribution and characteristics, their hotspots, fate and loadings into the marine aquatic environments in association with land use-based sources are poorly understood (Eo et al. 2019; Kataoka 2019; Lechnar 2014; Mani 2015; Horton et al. 2017b).

Moreover, less MPs pollution information are known for marine systems while only a very few for river freshwater aquatics ecosystems in Japan. The East Asian seas 'Sea of Japan (SJ)' and 'Seto Inland Sea (SIS)' have been speculated as hot spots of MPs (Isobe et al. 2015). Also, our previous MPs study along the SIS and SJ surrounded Yamaguchi prefecture areas revealed a medium to high level marine MPs pollution. We also thought that these marine areas are affected by both the Japan and foreign land originated MPs (Kabir et al. 2020). These findings emphasized the river MPs studies to know the Japan land originated MPs, their abundances and characteristics, land use-based sources, fate and loadings to understand the influence of Japan land behind the marine MPs pollution occurrences alongside securing the freshwater pollution knowledge. Hence, the main objective of this study was to investigate MPs pollution in the selected Seto Inland Sea and Sea of Japan flowing river environments of Yamaguchi prefecture. Moreover, these investigations were important to address these existing knowledge gaps in and between marine and freshwater aquatic MPs pollution, transportation pathways from the influencing land sourced MPs as well as required inevitably for the pollution control, mitigation and management strategies development towards the environmental protection and sustainability.

2. MATERIALS AND METHODS

2.1 Study Area and Selection of Sampling Stations

The two rivers i.e. Awano River (AR) and Majime River (MR) were selected which are flowing into the Sea of Japan and the Seto Inland Sea respectively in the Yamaguchi prefecture of Japan. The land use and river basin vector data for every square of a 100-m mesh were obtained from the National Land Numerical Information (NLNI) services. And the population vector data for every square of a 250-m mesh were obtained from 'e-Stat, Statistics of Japan'. ArcGIS (version 10.6.1) was used to compute. The major proportions of the AR catchment areas were occupied by non-point sources i.e. the forest and agricultural areas, less populations and urban land uses while the MR was occupied by the higher population inhabitants and urban sites as well as covered by forest and agricultural land uses. The sampling stations were selected focusing on the point and non-point sources as well as upstream and downstream sites following- the upstream areas (AR01 from AR, MR01 and MR02 from MR) were less populous, predominated by the agricultural and forest land uses while all the downstream stations (AR02–AR04 and MR03–MR08) were high number of inhabitants, urban uses as well as partly occupied by the agricultural uses and forest areas (Fig. 1).

2.2 Sample Collection

For surface water sampling– there exists both net based (e.g., plankton nets, neuston nets, bongo nets, manta trawls) and bulk/grab sampling methods. The grab sampling method was employed in this study following the BASEMAN Standardized Protocol and published researches to avoid the underestimation of MPs abundances due to the commonly used mesh sizes >300 μ m of the trawl-nets in numerous studies in which the predominant small MPs less than the used mesh sizes couldn't be identified. Moreover, the grab method is suitable for identification of microplastic fibers (Frias et al. 2018; Barrows et. al. 2017; Green et al. 2018; Dai et al. 2018; Whitaker et al. 2019). We took 1 L of river surface water from each selected station. A total of 12 samples were collected from 12 sites for laboratory analysis. The AR and MR samples were collected on 09 and 10 September 2019, respectively. All sampling materials and apparatus were clean and prepared as well as any possible external contamination was avoided during sample collection.



Figure 1. Study Sites of the Seto Inland Sea and Sea of Japan Flowing Awano (AR) and Majime (MR) Rivers in the Yamaguchi Prefecture of Japan.

2.3 Sample Preparation and Laboratory Analysis

We followed the membrane filtration, density separation and wet peroxidation (WPO) methods for MPs extraction (Frias et al. 2018; Barrows et al. 2017). In brief, 1 L of sample was filtered using 1 μ m PTFE membrane using the vacuums filtration system. After that, the WPO was employed to remove any organic matter from the extracted particles by 20 mL FeSO₄·7H₂O solution, 20 mL of 30% H₂O₂ solution, and digested at 70^oC on hotplate. Then, density separation was done pouring prepared ZnCl₂ solution of obtained density 1.5 g/cm³ was poured into the beaker and allowed at least 24 hours for settling of MPs particles (Coppock et al. 2017). Then, the supernatant was passed through stainless steel sieves (50–250, –500, –1000, –5000 μ m) for size categorization and finally membrane filtration was done to retain the frequently passed fibers through the sieves. Then, the counting measures were taken. Procedural blank tests and control measures were carried out during experiments in the laboratory to avoid any possible external contamination.

2.4 Identification and Characterization

After extraction, all particles were visually identified, counted, and measured under the microscope (BH2, OLYMPUS, Japan) at 40X magnification. Micro-forceps were used for MPs separation according to shapes, sizes and colors-based characteristics and categories. All the sizes were categorized into the small MPs (SMPs) (<1 mm) and large MPs (LMPs) (1–5 mm) (Frias et al. 2018). All the images were photographed using the 'OLYMPUS E-500' camera. Finally, the polymer types of the identified particles were confirmed by Fourier Transform Infrared Spectroscopy (FTIR) (FT/IR-4600, JASCO Incorporation Ltd., Japan) equipped with attenuated total reflection (ATR) unit.

2.5 Calculation of Loadings

The loadings of MPs into the SIS and SJ by these rivers were calculated along the river stations based on the available hydrographic data collected from the 'River Planning Division, Civil Engineering Department, Yamaguchi Prefecture' at our study period using the following equation from Miller et al. 2017. No rainfall was found during the sampling date, thus we thought that the river flow rates were not affected by the rainfall. Preliminarily, this study was conducted regardless of rainfall intensity affecting MPs abundances, river flow rate and loadings. The loadings calculation equation is given following below-

Whereas, N_{MPs} = loadings by number (billion number particles); F_m = the flow rate [The flow rate was converted from m³s⁻¹ to Ls⁻¹ multiplying by 1000]; C_n = MPs number concentration per liter (n/L)

3. RESULTS

3.1 Abundances and Distribution

All the sampling sites of all rivers were contaminated with MPs. The MPs abundance ranges varied from 102 to 146 n/L with an average value of 131 ± 17.12 n/L for AR, and 99 to 1061 n/L with an average value of 272.5 ± 299.15 n/L for MR. All the extracted MPs particles were yielded in 2704 MPs particles in numbers. Although the comparative following orders MR>AR were observed regarding MPs abundances in these rivers, no significantly large fluctuations were found (p > 0.05), thus indicating the similar level of pollution in these rivers of similar geographical and environmental contexts of Yamaguchi Prefecture in Japan.

Overall, the MPs abundances varied across the upstream and downstream stations. The downstream stations–AR02–AR04 (140.67±4.11 n/L) of the AR and MR03–MR08 (321.67±330.80 n/L) of the MR were found having relatively one to two times higher MPs abundance than all the upstream areas– AR01 (102 n/L) from AR and MR01–MR02 (125±26 n/L) from MR. Overall, an inconsistent trend and insignificant variations in abundances were observed from upstream to downstream areas. Also, the upstream stations showed a considerably large number which were merely covered by non-point agricultural and forest areas. On the other hand, statistical analysis suggested no significant correlations (p > 0.05) among the populations and land uses patterns over point and non-point sources from the upstream to downstream stations.

3.2 MPs Pollution Characteristics (Shape, Size, Shape-Size, Colors and Polymers)

The shapes of the observed MPs particles were sorted into visually obvious– fragments, films, and fibers. The fragments (49.87%) were the predominant shape characteristic followed by fibers (47.7%) > films (2.43%) of the totally extracted MPs numbers. From the environmental distribution point of view, the AR was found with higher proportion of fibers (63.79%) following the fragments (33.42%) > films (2.79%) while the fragments (66.33%) predominated the MR followed by fibers (31.61%) > films (2.06%). Then, each of the water samples were contaminated with both the small MPs (<1 mm) and large MPs (1 – 5 mm).



Figure 2. The MPs proportions and distribution of the totally extracted particles numbers in the river surface water– a) shape and size, and b) polymers-based proportions and distributions.



Figure 3. Microscopic view of different types of MPs extracted from the river surface water

The results revealed that the small MPs i.e. <1 mm occupied the major proportion (80.14%) in the river surface water followed by large MPs i.e. 1–5 mm (19.86%) of totally identified MPs particles numbers. And the distribution point view revealed that the small MPs (<1 mm) were predominant following 73.63% for the AR and 86.65% for the MR. The shape-size based characterization revealed that small sized MPs (<1 mm) for all the shapes were predominant for both AR and MR. The visually obvious color-based analysis results showed that transparent, blue, and white MPs were predominant followed by the green, red, black colored particles indicating the abundances of colorful MPs in these river surface water (data not mentioned) (Fig. 3). The ATR-FTIR analysis revealed ten different polymers in this study (Fig. 2). The results showed that the PP (31.91%), PE (20.76%), PS (7.80%), Vinylon (6.92%), and PET (6.47%) (first reported in this study, similar polymer to PVA of the same monomer vinyl acetate) were the commonly predominating polymers overall. From the distribution point of view, the AR was predominated by PE (30.10%), PP (19.70%), Nylon

(18.50%), Vinylon (10.82%) followed by PET (2.80%), PS (5.92%), PPS (5.67%), Epoxy Resin (6.48%) while the MR was predominated largely by PP (44.13%), PE (11.42%), PET (10.14%), PS (9.68%), followed by PPS (8.62%), EPDM (7.16%), PAN (5.82%) and Vinylon (3.03%).

4. DISCUSSION

4.1 Abundance Comparison and Distribution

Due to inconsistencies and dissimilarities in analytical methods, we compared the results with those other studies of similar analytical methods of grab sampling. Overall, the MPs abundances were in similar order of magnitude the Amsterdam Rivers, Netherlands; two to three times higher than the Gallatin River, USA; five to seven times higher than Pearl River, Taihu Lake, China. Besides, the abundances were one to two times smaller than the Lake Superior and Lake Michigan, USA; Saigon River, Vietnam (Table 1). Thus, overall, a higher MPs abundance level in these the SIS and SJ flowing AR and MR rivers than others around the world indicated a high-level MPs pollution. On the other hand, the used grab sampling method led to be resulted in several times higher MPs abundance quantification in this study consistent with evidence from other studies. Thus, the MPs abundances might be much higher in the aquatic systems than thought in earlier studies, hence, the grab sampling and filtration-based quantification method are recommended to avoid the underestimation of MPs abundance and pollution level (Barrows et. al. 2017; Green et al. 2018; Dai et al. 2018; Whitaker et al. 2019).

Study Areas	Water (n/L)	Shape	Polymer	Ref.
Awano River, Japan	102–146	Fibers	PE, PP	This study
Majime River, Japan	99–1061	Fragments	PE, PP,	This study
Ditch River Delta, Netherlands	48–187	Fibers, Spheres	—	Leslie et al. 2017
Lake Superior, Michigan	30–409	Fibers	PS	Whitaker et al. 2019
Gallatin River, USA	0–67.5	Fibers	PET	Barrows et al. 2018
Saigon River, Vietnam	172–519	Fibers, Fragments	PET, PE, PP	Lahen et al. 2018
Hudson River, USA	0.38-12.38	Fibers	PET, Teflon, PP	Miller et al. 2017
Pearl River	8.7–53	Films, Fibers	PA, Cellophane	Yan et al. 2019
Taihu Lake, China	3.4-25.8	Fiber	Cellophane	Su et al. 2016

Table 1. Summary of MPs abundances in river water around the world

The higher MPs abundances induced high level pollution, abundances variations in different upstream and downstream points were thought to be influenced by land uses over point and non-point sources (Mani et al. 2015). However, we did not find any significant relationship neither with the individual point sources nor with the non-point sources consistent with previous studies. These findings also suggested that the considerably large number MPs releases might happen from the non-point sources alongside point sources. And the areas which were affected both by the point and non-point sources, resulted in having higher MPs abundances. These clearly suggested that the higher MPs abundances were affected by the land uses over both the point sources and non-point sources. Furthermore, these might be the alternative indications of ubiquitous MPs existence in the different environmental compartments and the land use sources are required for further assessments.

4.2 Potential Sources of MPs Pollution Occurrences

All the identified polymers are made for our modern daily life uses (Plastic Europe, 2019). The PE, PP, PET, Vinylon and PS were the predominant polymers in all the stations of all rivers consistent with previous studies worldwide. Preliminarily, the plastics productions and consumptions, and MPs polymers occurrences might be relative both in the regional Japan and global context. The Japan Plastics Industry Federation (JPIF) statistical report of 2019 revealed that the PE, PP, PS, PET, Nylon (PA) and PVA productions were 61.1% of total production in the year of 2019 consistent with previous years. Plastic Europe 2019 reported that PE, PP, PS and PET constituted 65.2% of total produced and consumed plastics. Thus, to indicate primarily, the widespread productions, and higher consumption caused higher MPs polymers occurrence in the aquatic environments similarly both in the regional Japan and global context.

However, based on the distribution point of view in this study sites, the presence of PE, PP, Vinylon, PVA, PET, PS plastics indicated the release from all the point and non-point sources according to their widespread uses and applications followings– packaging and decorations, industries, agriculture, buildings and constructions, electrical and appliances. PE, PP, PS are often said as the single use plastics as carry bags, wrapping packages, have a relatively short useful lifetime, and end up in the waste stream, thus contribute to appear as a MPs polymer. The major proportions of these polymers were found as both fibers, fragments, and films. The fragments are mainly attributed to the breakdown of larger plastic products. Films are mainly produced by the fragmentation of plastic carry bags, indicating their disposal, transportation from other areas (Andrady 2011; GESAMP 2015). Further to that, the Vinylon, PET, PAN, PPS, Nylon were mainly found as

fibers. PAN was found in mainly the urban populated areas of MR. The domestic sewage, laundry drain from washing machines, WWTPs and textile sources might release a vast quantity of PET, PAN, PPS microplastic fibers (Falco et al. 2019; Napper and Thompson 2016). Nylon, PPS were abundant in the remote areas of AR and MR affected by the agricultural and forest areas. Nylon might come from ropes used in the agricultural firms in these areas. Another source of fiber is deposition from the atmosphere Urban atmospheric fallouts and airborne MPs have been reported to contribute a lot of MPs into the aquatic environments (Dris et al., 2016). EPDM is the synthetic rubber, has application in automotive indicating tire wears abrasion of vehicles induced sources from the urban areas (Leads et al. 2019; Ziajahromi et.al. 2020). MPs particles might be fate through roadside, agricultural runoff, and stormwater likely to transport tire particles to the aquatic environment. Moreover, MPs are intentionally added during plastics productions for daily life domestics and households, personal care products, industrials, agricultural, that might be released into the aquatic environments (Ziajahromi et.al. 2020; Piñon-Colin et al. 2020; Anderson et al. 2016; Dris et al. 2016; Driedger at al. 2015). And, overall, these indicated for emphasizing the logical reasons to the further investigations for comprehensive sources assessments, control, and management regarding pollution mitigation.

4.3 Fate, Export and Loadings into the SIS and SJ Marine Environments

Rivers transport waste plastics into the sea, the plastics get breakdown due to environmental processes and thus the MPs might get produced secondarily (Lebreton et al. 2017; Jambeck et al. 2015; Andrady 2011). In addition, rivers are also have been reported as the major pathways to transport primarily land sourced MPs (Eo et al. 2019; Boucher and Friot 2017; Eerkes-Medrano et al. 2015). In support to this, this study also speculated that a greater number of terrestrial/land originated MPs of different characteristics (shape-size-color-polymer) are being discharged into the SIS and SJ sea by these AR and MR rivers before the happening of macro plastics degradation into MPs following- the estimated load per day in the SJ were 92.36 billion particles by number by the AR while the load was estimated at 3.48 billion particles by number by the MR respectively. The loadings were one to three times lower than the Nakdong River, South Korea (Eo at al. 2018), and twenty times higher than the Hudson River, USA (Miller et al. 2017). Thus, these rivers feed many MPs to the SIS and SJ. Lechner et al. 2014 estimated 4.2 t per day the micro- and meso-plastic litter input from the River Danube into the Black Sea while Lebreton estimated 2.41 million tons of plastics waste enters the ocean every year from rivers. In this study, overall loadings indicated that these AR and MR rivers fed a large amount of Japan land originated MPs into the SIS and SJ originated as the prominent MPs transportation pathway. Besides, the loadings varied largely under the hydrological regimes- although the MR had comparatively higher abundances, however resulted in less loadings into the receiving marine environments due to the discharge flow rate in this study (Table 2). Hence, we thought that the riverine MPs exportation are affected by other hydrological and meteorological conditions. Furthermore, rainfall significantly affects MPs concentrations in the inland aquatic systems (Xia et al. 2020). Thus, it is possible to overestimate or underestimate the riverine MPs load due to the river hydrodynamic regime- flow rate and turbulences, water column transportation as well as other meteorological factors including- rainfall, seasonal variation, weather influences and climatic events etc. (Lebreton et al. 2017; Eo et al. 2019). Thus, it is recommended to yield a more realistic estimate of the annual load of MPs released to the ocean as well as MPs pollution information in the aquatic systems considering these above factors for future studies.

Furthermore, these findings also suggested that the Japan land sources influenced the SIS and SJ MPs pollution occurrences. Further to verify the influences of Japan land originated MPs in the SIS and SJ, we also compared our current river MPs polymer results with the MPs polymers from SIS and SJ marine areas. Overall, the results showed a predominance of the similarly same MPs polymers (PE, PP, Vinylon, PVA, PET) in these river surface waters and the receiving SIS and SJ marine areas (Kabir et al. 2020). As Japan land-based sources are thought affecting to cause SIS and SJ MPs pollution, then this led us to think and suggested that the terrestrial sources control and interventions of MPs releases alongside the releases of plastic wastes releases are required for further pollution control, mitigation and management.

T 11 A				1. 1	1	1 1.	1			•	
Table 2.	The MPs	abundances.	river	discharge.	and	loadings 1	to the	receiving	marine	environm	ients

Rivers	Flow Rate (L/s)	Loadings by Numbers (Billion/Day)	Receiving Marine Environments	References
Awano River (AR)	7860	92.36	Sea of Japan (SJ)	This Study
Majime River (MR)	210	3.48	Seto Inland Sea (SIS)	This Study

4.4 Potential Impacts on Freshwater and Marine Aquatic Ecosystems

The predominance of the MPs fibers, fragments and films in the same aquatic environment posed higher MPs encounter potential with numerous negative impacts in the inhabiting organisms while the predominance of small MPs (<1 mm) of different shapes might have higher probability of ingestion. Studies indicated different shapes induced uptake and biological effects while small MPs fibers are more serious threat potential to aquatic organisms than fragments, films, and microbeads (Ziajahromi et al. 2017). Thus, the higher fibers predominance in these AR and MR raised a cause of concern. Studies also reported the higher abundance of

the ingested small MPs fragments, films, and fibers in biota- shellfish and fish, were and their numerous negative effects (Tanaka and Takada 2016; Lusher et al. 2016). Also, the large MPs become small MPs periodically due to breakdown. Apart from that, MPs ingestion induced physical damages and intestinal blockages were also evident (Andrady 2011; Cole et al. 2011; Wright 2013). Besides, the large variety of colors for MPs particles are like some natural aquatic foods, therefore, mistaken ingestion by aquatic organisms might happen (Wright 2013). In addition, polymers might cause hazards and pose chemical risks in the water environments that threaten the ecosystems and human health. The found polymers in this study-Epoxy Resin, PAN, nylon, PS, PET were highly hazardous potentials. Once these polymers enter the water environment, it may release carcinogenic monomers and intrinsic plasticizers resulting in lethal effects, bioaccumulation for aquatic organisms, thus entrance in the food web threatening human health (Lithner et al. 2011). Other chemical contaminants, such as POPs, heavy metals, and other hydrophobic contaminants can be easily absorbed onto the surface of polymers causing the enhanced chemical toxicity in the water environment are affected by the polymers, shapes and sizes heavy, molecules producing composite ecological effects (Smith et al. 2018), while more studies are required to know for the other toxic polymers. Moreover, MPs polymers induced numerous ecotoxicological effects have been evident in studies. Moreover, these polymers might cause long-term pollution in the aquatic systems (Auta et al. 2018; GESAMP 2015). Thus, MPs shapesize-color-polymer characteristics might cause hazards, chemical toxicity in the AR and MR river freshwater aquatic ecosystems in implications with the receiving SIS and SJ marine aquatic ecosystems.

5. Conclusion

The higher abundances of MPs in the AR and MR indicated a high-level pollution in the river freshwater ecosystems. The land uses indicated both the point and non-point sources could release the MPs into these river aquatic systems. The estimated loadings revealed a large number of MPs discharge into the receiving SJ and SIS marine aquatic environments. These two rivers were found to be the prominent pathways of land-sourced MPs transportation into these marine environments. The MPs characteristics (size-shape-color-polymer) demonstrated toxicity potentials induced impacts on these aquatic ecosystems. Overall this study provided an insight of river freshwater MPs pollution, fulfilled the preliminary knowledge gaps of pollution occurring land sources, in and between the river freshwater and marine systems, facilitated understanding of MPs transportation pathways, fate and loadings into the receiving marine environments. Hence, the study facilitated further rigorous source assessments towards the development of MPs pollution control, mitigation and management strategies for environmental protection and sustainability in the regional Japan as well as global context.

References

Anderson, A.G.,Grose, J., Pahl S., Thompson, R.C., and Wyles K.J. (2016). Microplastics in personal care products:
Exploring perceptions of environmentalists, beauticians and students. *Marine Pollution Bulletin*, 113:454-460.
Andrady, A. L. (2011). Microplastics in the marine environment. *Marine Pollution Bulletin*, 62(8):1596-1605.
Auta, H. S., Emenike, C. U., and Fauziah, S. H. (2017). Distribution and importance of microplastics in the marine environment: A review of the sources, fate, effects, and potential solutions. *Environment International*, 102: 165-176.
Barrows, A. P. W., Christiansen, K. S., Bode, E. T., and Hoellein, T. J. (2018). A watershed-scale, citizen science approach to quantifying microplastic concentration in a mixed land-use river. *Water Research*, 147: 382-392.
Boucher, J., Friot D. (2017). Primary Microplastics in the Oceans: A Global Evaluation of Sources. Gland, Switzerland: IUCN., pp.44.

Cole, M., Lindeque, P., Halsband, C., and Galloway, T. S. (2011). Microplastics as contaminants in the marine environment: a review. *Marine Pollution Bulletin*, 62(12):2588-2597.

Coppock, R. L., Cole, M., Lindeque, P. K., Queiros, A. M., and Galloway, T. S. (2017). A small-scale, portable method for extracting microplastics from marine sediments. *Environmental Pollution*, 230:829-837.

Dai, Z., Zhang, H., Zhou, Q., Tian, Y., Chen, T., Tu, C., Luo, Y. (2018). Occurrence of microplastics in the water column and sediment in an inland sea affected by intensive anthropogenic activities. *Environmental Pollution*, 242: 1557-1565. De Falco, F., Di Pace, E., Cocca, M., and Avella, M. (2019). The contribution of washing processes of synthetic clothes to microplastic pollution. *Scientific Reports*, 9(1):6633.

Ding, L., Mao, R. F., Guo, X., Yang, X., Zhang, Q., and Yang, C. (2019). Microplastics in surface waters and sediments of the Wei River, in the northwest of China. *Science of the Total Environment*, 667:427-434.

Driedger, A. G. J., Dürr, H. H., Mitchell, K., and Van Cappellen, P. (2015). Plastic debris in the Laurentian Great Lakes: A review. *Journal of Great Lakes Research*, 41(1):9-19.

Dris, R., Gasperi, J., Saad, M., Mirande, C., and Tassin, B. (2016). Synthetic fibers in atmospheric fallout: A source of microplastics in the environment? *Marine Pollution Bulletin*, 104(1-2):290-293.

Eo, S., Hong, S. H., Song, Y. K., Han, G. M., and Shim, W. J. (2019). Spatiotemporal distribution and annual load of microplastics in the Nakdong River, South Korea. *Water Research*, 160:228-237.

Frias, J. P. G. L., and Nash, R. (2019). Microplastics: Finding a consensus on the definition. *Marine Pollution Bulletin*, 138:145-147.

GESAMP (2015). Sources, Fate and Effects of Microplastics in the Marine Environment: A Global Assessment. International Maritime Organization 4 Albert Embankment, London SE1 7SR.

Green, D. S., Kregting, L., Boots, B., Blockley, D. J., Brickle, P., da Costa, M., and Crowley, Q. (2018). A comparison of sampling methods for seawater microplastics and a first report of the microplastic litter in coastal waters of Ascension and Falkland Islands. *Marine Pollution Bulletin*, 137:695-701.

Horton, A. A., Walton, A., Spurgeon, D. J., Lahive, E., and Svendsen, C. (2017). Microplastics in freshwater and terrestrial environments: Evaluating the current understanding to identify the knowledge gaps and future research priorities. *Science of the Total Environment*, 586:127-141.

Isobe, A., Uchida, K., Tokai, T., & Iwasaki, S. (2015). East Asian seas: A hot spot of pelagic microplastics. *Marine Pollution Bulletin*, 101(2):618-623.

Jambeck, J. R., Geyer, R., Wilcox, C., Siegler, T.R., Perryman, M., Andrady, A., Narayan, R., Law, K.L. (2015). Plastic waste inputs from land into the ocean. *Science*, 347:768–771.

Kabir, A. H. M. E., Sekine, M., Imai T., Yamamoto K. (2020). Microplastics Pollution in the Seto Inland Sea and Sea of Japan Surrounded Yamaguchi Prefecture Areas, Japan: Abundance, Characterization and Distribution, and Potential Occurrences. *Journal of Water and Environment Technology* 18(3): 175-194.

Kataoka, T., Nihei, Y., Kudou, K., and Hinata, H. (2019). Assessment of the sources and inflow processes of microplastics in the river environments of Japan. *Environmental Pollution*, 244:958-965.

Klein, S., Worch, E., and Knepper, T. P. (2015). Occurrence and Spatial Distribution of Microplastics in River Shore Sediments of the Rhine-Main Area in Germany. *Environmental Science and Technology*, 49(10):6070-6076.

Lebreton, L. C. M., van der Zwet, J., Damsteeg, J. W., Slat, B., Andrady, A., and Reisser, J. (2017). River plastic emissions to the world's oceans. *Nature Communications*, 8:15611.

Lechner, A., and Ramler, D. (2015). The discharge of certain amounts of industrial microplastic from a production plant into the River Danube is permitted by the Austrian legislation. *Environmental Pollution*, 200:159-160.

Leslie, H. A., Brandsma, S. H., van Velzen, M. J., and Vethaak, A. D. (2017). Microplastics en route: Field measurements in the Dutch river delta and Amsterdam canals, wastewater treatment plants, North Sea sediments and biota. *Environmental International*, 101:133-142.

Li, J., Liu, H., and Paul Chen, J. (2018). Microplastics in freshwater systems: A review on occurrence, environmental effects, and methods for microplastics detection. *Water Research*, 137:362-374.

Lithner, D., Larsson, A., & Dave, G. (2011). Environmental and health hazard ranking, and assessment of plastic polymers based on chemical composition. *Science of the Total Environment*, 409(18):3309-3324.

Lorena M. Rios Mendoza, M. B. (2019). Microplastics in freshwater environments: A review of quantification assessment. *Trends in Analytical Chemistry*, 119:402-408.

Lusher, A. L., McHugh, M., and Thompson, R. C. (2013). Occurrence of microplastics in the gastrointestinal tract of pelagic and demersal fish from the English Channel. *Marine Pollution Bulletin*, 67(1-2):94-99.

McNeish, R. E., Kim, L. H., Barrett, H. A., Mason, S. A., Kelly, J. J., and Hoellein, T. J. (2018). Microplastic in riverine fish is connected to species traits. *Scientific Reports*, 8(1):11639.

Murphy, F., Ewins, C., Carbonnier, F., & Quinn, B. (2016). Wastewater Treatment Works (WwTW) as a Source of Microplastics in the Aquatic Environment. *Environmental Science and Technology*, 50(11):5800-5808.

Napper, I. E., and Thompson, R. C. (2016). Release of synthetic microplastic plastic fibres from domestic washing machines: Effects of fabric type and washing conditions. *Marine Pollution Bulletin*, 112(1-2):39-45.

Nel, H. A., Hean, J. W., Noundou, X. S., and Froneman, P. W. (2017). Do microplastic loads reflect the population demographics along the southern African coastline? *Marine Pollution Bulletin*, 115(1-2):115-119.

Pinon-Colin, T. J., Rodriguez-Jimenez, R., Rogel-Hernandez, E., Alvarez-Andrade, A., and Wakida, F. T. (2020). Microplastics in stormwater runoff in a semiarid region, Tijuana, Mexico. *Science of the Total Environment*, 704:135411. Plastics Europe (2019). Plastics – the Facts 2019 An analysis of European plastics production, demand and waste data. In: Plastics Europe Belgium.

Smith, M., Love, D. C., Rochman, C. M., & Neff, R. A. (2018). Microplastics in Seafood and the Implications for Human Health. *Current Environmental Health Reports*, 5(3):375-386.

Su, L., Xue, Y., Li, L., Yang, D., Kolandhasamy, P., Li, D., and Shi, H. (2016). Microplastics in Taihu Lake, China. *Environmental Pollution*, 216:711-719.

Tanaka, K., and Takada, H. (2016). Microplastic fragments and microbeads in digestive tracts of planktivorous fish from urban coastal waters. *Scientific Reports*, 6:34351.

Vaughan, R., Turner, S. D., and Rose, N. L. (2017). Microplastics in the sediments of a UK urban lake. *Environmental Pollution*, 229:10-18.

Whitaker, J. M., Garza, T. N., and Janosik, A. M. (2019). Sampling with Niskin bottles and microfiltration reveals a high prevalence of microfibers. *Limnologica*, 78.

Xia, W., Rao, Q., Deng, X., Chen, J., and Xi, P (2020). Rainfall is a significant environmental factor of microplastic pollution in inland waters. *Science of the Total Environment*, 732:139065.

Yan, M., Nie, H., Xu, K., He, Y., Hu, Y., Huang, Y., and Wang, J. (2019). Microplastic abundance, distribution and composition in the Pearl River along Guangzhou city and Pearl River estuary, China. *Chemosphere*, 217:879-886. Ziajahromi, S., Kumar, A., Neale, P. A., and Leusch, F. D. L. (2017). Impact of Microplastic Beads and Fibers on Waterflea (Ceriodaphnia dubia) Survival, Growth, and Reproduction: Implications of Single and Mixture Exposures. *Environmental Science and Technology*, 51(22):13397-13406.