

# Regional Studies in Marine Science

## Effects of size and density of microplastic particles on their horizontal distribution on the seafloor of the Otsuchi Bay, a rural coastal area, Japan --Manuscript Draft--

<b>Manuscript Number:</b>	RSMA-D-22-01359R1
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<b>Abstract:</b>	Microplastic (MP) pollution in coastal areas has received increasing attention recently. However, studies focused on MP pollution in rural coastal areas remain limited compared to those in metropolitan coastal areas. This study observed MP particles accumulated on the seafloor of the Otsuchi Bay, a small ria bay located on the Pacific coast, Sanriku, Japan. The MP concentrations in the sediment ranged from $2.6 \pm 0.3$ to $13.6 \pm 9.8$ pcs g <sup>-1</sup> DW and $2.6 \pm 1.4$ to $5.1 \pm 1.2$ pcs g <sup>-1</sup> DW in March and September 2021, respectively. No significant difference in MP concentrations was detected between March and September. The MP concentration in the Otsuchi Bay was lower than that observed in other highly populated coastal areas, but was relatively high considering the population size of the catchment area. MP particles smaller than 1000 $\mu$ m were the most prevalent, accounting for 96.3% of all MP samples. MP size at the bay head was smaller than that at the central bay for high-density MPs; however, the relationship was reversed for low-density MPs. Analysis of the MP distribution pattern using a two-dimensional numerical model suggests that the horizontal distribution of MPs in the Otsuchi Bay depends on the size and density of MP particles. It is also strongly influenced by both the tidal oscillating currents characteristic to the bay and vertical terminal velocity of MP particles. Sedimented MP distributions in a bay with a small catchment population with limited MP sources shed light on our understanding of MP transport dynamics.
<b>Suggested Reviewers:</b>	Tomoya Kataoka, Ph.D Associate Professor, Ehime University tkata@cee.ehime-u.ac.jp His recent publications are related to my study, so he will be familiar with and give fair judgment to my manuscript.  Anthony Andrady, Ph.D NC State University andrady@andrady.com He has many paper in this field  Atsuhiko Isobe, Ph.D Professor, Kyushu University Research Institute for Applied Mechanics aisobe@riam.kyushu-u.ac.jp Professor Isobe is a famous expert in marine plastic debris, especially on transport and formation processes of marine microplastics. He may be proficient in the content that I have written.  Andrés Cózara, Ph.D University of Cadiz

	<p>andres.cozar@uca.es</p> <p>An Expert in the related field.</p>
<b>Response to Reviewers:</b>	<p>Response to the reviewer comments is included in an attached file (RSMA Response letter.doc).</p> <p>We kindly request that you open the document to view the responses.</p>

Dear Dr. Jong Seong Khim, Dr. Huahong Shi, Dr. Kenneth Mei Yee Leung, and Dr. Joanna Staneva,

We would like to submit the manuscript entitled “Effects of size and density of microplastic particles in their horizontal distribution on the seafloor in the Otsuchi Bay, a ria in Sanriku, Japan” by Yehao Wang, Rei Yamashita, Yoshimasa Matsumura, Shin-ichi Ito, and Kosei Komatsu to be considered for publication as an original paper in the *Regional Studies in Marine Science*.

The amount of plastic waste found floating on the ocean surface is small compared with the total amount entering the ocean, suggesting that much of it may be sinking and accumulating on the seafloor. Yet, given the lack of focus on microplastic (MP) pollution in rural coastal areas in past studies, the concentration and composition of MPs on the seafloor are not well known in these areas.

To bridge this knowledge gap, we examined MP contamination accumulated on the seabed sediment in the inner part of Otsuchi Bay which was one of the small bays on the Pacific rias coast of Tohoku, Japan. The concentration of MPs was detected in the range of  $2.6 \pm 0.3$  to  $13.6 \pm 9.8$  pcs g<sup>-1</sup> DW in March 2021 and  $2.6 \pm 1.4$  to  $5.1 \pm 1.2$  pcs g<sup>-1</sup> DW in September 2021, respectively.

Our results will be of great interest to the readers of *Regional Studies in Marine Science*, especially those interested in the extent of MP contamination in rural coastal areas. Determining MPs concentrations in these areas is critical for filling the research blank of the behavior and fate of these particles along the coast, as well as for understanding how they may affect local marine organisms.

The contributions of the authors are as follows. Yehao Wang contributed to analysis and investigation, as well as writing of the original manuscript draft. Rei Yamashita contributed to analysis and the review. Yoshimasa Matsumura contributed to interpretation of results and review. Shin-ichi Ito contributed to project administration, resources, and interpretation of results. Kosei Komatsu was in charge of project administration, as well as investigation, writing, editing, and review of the original draft.

This study was supported by the JSPS KAKENHI Grant no. 20H01969 and from the Nippon Foundation Research Project on Marine Plastics.

We confirm that this manuscript has not been published elsewhere and is not under consideration in whole or in part by another journal. All authors have approved the manuscript and agree with submission to *Regional Studies in Marine Science*. The authors have no conflicts of interest to declare.

We would like to recommend the following researchers as potential reviewers for this paper:

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16 Feb 2022

Dr. Giuseppe Suaria  
Associate Editor  
*Regional Studies in Marine Science*

Dear Editor:

We wish to re-submit the manuscript titled “Effects of size and density of microplastic particles on their horizontal distribution on the seafloor of the Otsuchi Bay, a rural coastal area, Japan” The manuscript ID is RSMA-D-22-01359.

We thank you and the reviewers for your thoughtful suggestions and insights. The manuscript has benefited from these insightful suggestions. I look forward to working with you and the reviewers to move this manuscript closer to publication in *Regional Studies in Marine Science*.

The manuscript has been rechecked and the necessary changes have been made in accordance with the reviewers’ suggestions. The manuscript has been proofread to ensure that the text is grammatically correct. The responses to all comments have been prepared and given below. Revised portions in the manuscript are marked in red.

Thank you for your consideration. I look forward to hearing from you.

Sincerely,  
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## Response to Reviewers' Comments

### Response to Reviewer 1

Thank you for your comments and careful review of the manuscript. The manuscript has been revised based on your comments and suggestions. Revisions in the manuscript are indicated in red.

This manuscript observed MP particles accumulated on the seafloor of the Otsuchi Bay, one of the small ria bays located on the Pacific coast, Sanriku, Japan.

This manuscript could be suitable for RSMS but honestly I cannot see the effects of size and density of microplastic particles in their horizontal distribution. I think this is an interesting MPs assessment but the title is not in accordance with the information presented.

**Response:** Thank you for your valuable feedback on our manuscript. We appreciate your suggestion regarding the need to better justify the appropriateness of the title. We would like to clarify that our study did find significant effects of size and density of microplastic particles on their horizontal distribution. In particular, our results showed that the size of microplastics at the bay head was smaller for high-density particles compared to the central bay, while for low-density particles the reverse relationship was observed. This demonstrates the impact of size and density on the distribution of microplastics, and we believe that these findings reinforce the relevance of our study's title.

Introduction: too generic and I miss more about the area.

**Response:** Thank you for your comment. The study focused on Otsuchi Bay, a small bay located in Iwate Prefecture on the northeastern coast of Japan, and aimed to clarify the horizontal distribution, seasonal changes, and properties of MPs in the sediments on the seafloor in the Otsuchi Bay and investigate their sedimentation dynamics. This is the first study to demonstrate MP pollution in sediment in a rural coastal area in Japan. We have added a detailed description of the study area in the revised manuscript (Lines 82–91).

Materials and methods: I miss more information about the study area. and the section 2.3. Prevention of MP contamination from the laboratory environment during analysis and MP recovery test is not necessary. too much noise!

**Response:** Thank you for your feedback on our manuscript. To address your concern regarding the study area, we have added section 2.1 'study area' in the revised manuscript to provide better context. We have included a detailed description of the study area in lines 106–113.

We have also revised section 2.3 based on your suggestion to improve clarity and conciseness. We have moved this to section 2.4 "Analysis of MPs" as suggested by reviewer 2 (Line 141).

Results: I think there are too subheadings that cut the narrative. a fluid narrative can much better .

**Response:** Thank you for your feedback regarding the structure of our paper. We understand your concern regarding the subheadings potentially disrupting the flow of the narrative. However, we believe that the current structure effectively presents the different aspects addressed in the manuscript. The original manuscript consists of only four sections, each of which addresses a distinct subject area. We feel that this format allows for a clear and organized presentation of our research. As such, we have decided to maintain the current structure. Thank you for your understanding.

Discussion: something is missing. Also I cannot see the real effects of size and density of microplastic particles in their horizontal distribution. Authors can done a better job here.

**Response:** We appreciate your feedback and thank you for pointing out the confusion. We apologize for the lack of clarity in our explanation of the effects of size and density of microplastic particles on their horizontal distribution. To clarify, the main purpose of this numerical experiment was to explain our observation result. The mechanisms of low-density MPs with different size and their horizontal distribution was already discussed by Isobe et al. (2014). Therefore, we tried to fill the gap in the understanding of the high-density MP settling process. Although only two size categories were calculated, we believe this is enough to reproduce our observation result. Nevertheless, as suggested, we will consider using a 3D general ocean model for this numerical experiment in future research. In addition, we also added information about wind and waves to enhance the discussion (Lines 393–405).

Conclusions: poor.

**Response:** Thank you for your feedback regarding the conclusions section of our paper. We apologize if it did not meet your expectations and understand that it may have caused confusion or disappointment. We have taken your feedback into consideration and revised the conclusion section to ensure that it fully covers the content of this paper (Lines 424–429, 437–438).

Note: The Algorithm of numerical experiment must be included in the methodological aspects.

**Response:** Thank you for your suggestion. We have moved the the algorithm of the numerical experiment to the Materials and Methods section (Lines 188–232).

Figures: hard to see. can be much better.

**Response:** Thank you for your comment on the figures in our manuscript. We apologize for any inconvenience this may have caused. We have taken the necessary steps to improve the visibility of the figures; we have increased the resolution and redrawn the figure. In addition, we expect the resolution to be higher when the manuscript is printed and published.

## Response to Reviewer 2

Thank you for your helpful suggestions. We have revised the manuscript based on your comments and suggestions. Revisions in the manuscript are indicated in red.

The manuscript by Wang and colleagues shows the first results of microplastic concentrations in seabed sediments in a rural semi-enclosed bay in eastern Japan. The authors find relatively low concentrations compared to other coastal areas that are "highly-populated" and find evidence of size and density-dependent sorting of microplastics using a 2D horizontal model. Although the study is focused on a specific area of the Japan coast it contributes to increase knowledge on the mechanisms that dominate the settling and horizontal spreading of microplastics in sediments. However, the manuscript lacks cohesion/continuity. The authors should consider obtaining more samples or focus the discussion on the model. Additional data (e.g. winds, currents) can be analyzed to enhance discussion. Discussion of the polymers found can also be improved. The manuscript has multiple typo and grammar errors and must be revised to increase clarity.

**Response:** We greatly appreciate the time and effort you have invested to review our manuscript. We value your feedback and suggestions on improving our study.

You have correctly pointed out that our study, which focuses on a specific rural semi-enclosed bay in eastern Japan, could benefit from a larger number of samples and a more focused discussion. We had initially planned on collecting more samples; however, this was unsuccessful owing to bad weather. Nevertheless, we will consider obtaining additional samples and a 3D general ocean model for future research. In addition, we also added the information about winds and waves to improve our discussion. We apologize for any typos or grammatical errors in the manuscript; we have carefully revised the manuscript to address issues pertaining to language, grammar, clarity, and readability.

Line 28: Significant seasonal difference. The term "seasonal" is inappropriate if samples were only collected in March and September.

**Response:** Thank you for pointing this out. We have removed this term as suggested.

Line 30: Check sentence.

**Response:** Thank you for pointing this out. We apologize for the oversight. We have revised this sentence for clarity (Lines 27–29).

Line 31: Why use 1 mm? Why not use other more common limits for microplastics (5 mm)?

**Response:** Only a few microplastic particles larger than 1 mm were found in this study (Fig. 3).



Therefore, we used 1 mm as it is relevant to the results obtained; we did not intend to define microplastic size.

Lines 57-58: Check references for concentrations of floating MP. You probably meant up to  $10^6$ . You may also want to check more recent publications (e.g. Wilcox et al. 2020, Lebreton et al. 2018).

**Response:** Thank you for your comment. As suggested, we have revised the text and added a recent reference Wilcox et al. (2020). Because we already referred Lebreton et al. (2018) in the previous paragraph, we added van Sebille et al. (2015) instead of Lereton et al. (2018) to make the text narrative (Lines 58–64).

Line 61: Mind point and comma decimal/thousand separator. Units should be preferably written in the format "pieces L<sup>-1</sup>" consistently throughout the manuscript.

**Response:** Thank you for your comments. It was our mistake. We have replaced “pieces L<sup>-1</sup>” to “pieces/L” in the revised manuscript (Line 58).

Lines 64-66: Other potential sinks have been also recently proposed (e.g. see review by van Sebille et al. 2020). In particular, coastal and nearshore areas (e.g. Onink et al. 2021, de Haan et al. 2022).

**Response:** We appreciate your helpful comments. We have added two recent references in the revised manuscript (Onink et al. 2021 and van Sebille et al. 2020) (Lines 65–66).

Lines 72-75: Check writing.

**Response:** Thank you for bringing this to our attention. We apologize for the oversight. We have revised this sentence for clarity (Lines 75–78).

Line 76: Add reference.

**Response:** As suggested, we have added a reference for this in the revised manuscript (van Sebille et al., 2020) (Line 79).

Lines 81-84: I find that these are not actual reasons but a description of the study area.

**Response:** We appreciate your feedback and thank you for pointing out the confusion. We apologize for the lack of clarity in our explanation of the reasons for selecting the Otsuchi Bay as our study site.

To clarify, the main reasons for choosing the Otsuchi Bay were as follows:

- The system is relatively simple.
- The circulation in the bay has been well investigated by previous studies.
- Studying MP concentrations in this region would provide valuable insights into the pollution status of microplastics in rural areas of Japan.

We have also revised the relevant sentences in the manuscript to clarify this (Lines 92–99).

Line 92: "Aim" may be more appropriate.

**Response:** Thank you. We have revised this sentence as suggested for clarity (Line 100).

Lines 94-95: Check writing.

**Response:** Thank you for helping us to improve our manuscript. As suggested, we have revised this sentence for clarity. The manuscript has been proofread to ensure that the text is grammatically correct (Lines 101–103).

Line 108: Check writing.

**Response:** Thank you for bringing this to our attention. As suggested, we have revised this sentence for clarity. The manuscript has been proofread to ensure that the text is grammatically correct (Lines 124–125).

Line 113: Please indicate, what slice of the core was sampled, the first 5 cm? Indicate in this section.

**Response:** Thank you for your suggestion. We used the top 5 cm of the sample in this study. We revised the corresponding sentence in the manuscript to include this information (Line 130).

Line 131: Check writing.

**Response:** Thank you for bringing this to our attention. As suggested, we have revised this sentence for clarity. The manuscript has been proofread to ensure that the text is grammatically correct (Line 165).

Line 132: "diameter".

**Response:** We have revised this as suggested (Line 166).

Line 132: "Petri dish".

**Response:** We have revised this as suggested (Line 166).

Line 134: Did you perform a mapping of the filter using e.g. Liner Array FTIR? What software did you use? How did you analyze the spectra and compare with references? This part of the section could be moved/merged to/with the "Analysis of MPs" section.

**Response:** Thank you for your comment. We used the method described in section 2.4 "Analysis of MPs." We have moved the pointed section into section 2.4 "Analysis of MPs" to avoid confusion.

Line 172: What is the total number of plastics found? Indicate in this section.

**Response:** The suggested information has been added in lines 259–260 of the revised manuscript.

Line 186: Check writing.

**Response:** Thank you for helping us to improve our manuscript. As suggested, we have revised this sentence for clarity. The manuscript has been proofread to ensure that the text is grammatically correct (Line 248).

Line 203: Indicate the % of microplastics < 5 mm.

**Response:** All microplastics observed in this study were smaller than 5 mm. We added the information in the text (Line 266).

Lines 223–224: Check writing.

**Response:** Thank you for helping us to improve our manuscript. As suggested, we have revised this sentence for clarity. The manuscript has been proofread to ensure that the text is grammatically correct (Lines 296–297).

Line 225: Wang et al. (2021) reference missing (<https://doi.org/10.1016/j.ecss.2021.107552>). Please double-check for other missing references throughout the manuscript.

**Response:** Thank you for bringing this to our attention. We have added the reference in the revised manuscript.

Line 229: "This might be attributed to the insufficient smaller number of sampling stations at the Tokyo Bay, which possibly failed to determine the actual MP contaminations". Wang et al. (2021) sampled 3 stations in Tokyo Bay, that is not much of a difference compared with this study (6) in Otsuchi Bay. It seems like your undermining your own results here. In addition, consider checking grammar.

**Response:** Thank you for your valuable feedback and suggestions. As you have mentioned, the number of sampling stations in Tokyo Bay used in the study conducted by Wang et al. (2021) is not considerably different from the number of stations used in our study in Otsuchi Bay. However, we would like to clarify that the purpose of including the comparison with Tokyo Bay in our study was to provide a broader perspective on MP contamination levels in different coastal areas of Japan, rather than to undermine our own results. The manuscript has been proofread to ensure that the text is grammatically correct (Lines 302–306).

Lines 234: "Seasonal" may be inappropriate.

**Response:** We have revised this sentence as suggested (Line 310).

Lines 242-243: Was there a statistically significant difference? Consider assessing this with a Chi-squared.

**Response:** Thank you for your suggestion. There was a statistically significant difference between March and September. The results of Chi-squared test have been added to the revised manuscript (Lines 319–320).

Line 247-248: Explain why.

**Response:** Thank you for your comments. We added the explanation of critical shear stress in the manuscript (Lines 323–324). As you know, a higher flow rate caused an increase in the shear stress value. It was followed by:

$$\tau = (U^*)^2 \times \rho,$$

where  $\tau$  ( $N\ m^{-2}$ ) is the shear stress,  $U^*$  is the shear velocity, and  $\rho$  is the density of seawater. The higher shear stress led to the resuspension of more high-density MPs, as observed in previous laboratory experiments (Ballent et al., 2013). This indicates that high-density MPs were resuspended from the riverbed and flowed into the Otsuchi Bay. To make the discussion narrative, we did not include the explanation of critical shear stress itself in the manuscript.

Lines 252-254: Consider indicating minimum mesh/filter size used by each study.

**Response:** Thank you for your suggestion. Lin et al., 2021 used a 2  $\mu m$  filter with and Wang et al., 2021 used a 1  $\mu m$  filter size. We also used a 1  $\mu m$  PTFE filter in this study. We added the filter size information in the manuscript (Line 333).

Line 255: Consider explaining the meaning of "Enclosed Index". How is it calculated? Consider adding a reference.

**Response:** Enclosed Index (E.I.) is a geographical characteristic which implies the degree of closure of the bay. The value of the E.I. reflects residence time of seawater in a bay (Nakano et al 2021). It was calculated s follows:

$$E.I. = \frac{\sqrt{S} \times D_1}{W \times D_2},$$

where S is the area of the bay, W is the width of the bay mouth, D1 is the maximum depth, and D2 is the depth at the bay mouth. We have revised the corresponding sentence in the revised manuscript (Lines 334–338).

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3 Lines 264-272: It is not really clear to me what you intend to discuss with deposition rates and periods  
4 of sediment particles and MPs.

5 **Response:** Some sediment cores were previously dated using radio nuclides and man-made chemicals  
6 with well-documented emission histories as geochronometers. (Matsuguma et al., 2017). Therefore,  
7 calculating the deposition rates and sinking periods can provide important information regarding the  
8 history of MP pollution in this area. We also revised the corresponding sentence in the revised  
9 manuscript to improve clarity (Line 349–361).  
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15 Line 274: This section is not contemplated in the Materials and Methods and Result sections.

16 **Response:** Thank you for your comment. We moved the algorithm of the numerical experiment to the  
17 Materials and Methods section (Lines 188–232).  
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1 Effects of size and density of microplastic particles on their horizontal distribution on the seafloor of

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## Abstract

Microplastic (MP) pollution in coastal areas has received increasing attention recently. However, studies focused on MP pollution in rural coastal areas remain limited compared to those in metropolitan coastal areas. This study observed MP particles accumulated on the seafloor of the Otsuchi Bay, a small ria bay located on the Pacific coast, Sanriku, Japan. The MP concentrations in the sediment ranged from  $2.6 \pm 0.3$  to  $13.6 \pm 9.8$  pcs  $g^{-1}$  DW and  $2.6 \pm 1.4$  to  $5.1 \pm 1.2$  pcs  $g^{-1}$  DW in March and September 2021, respectively. No significant difference in MP concentrations was detected between March and September. The MP concentration in the Otsuchi Bay was lower than that observed in other highly populated coastal areas, but was relatively high considering the population size of the catchment area. MP particles smaller than 1000  $\mu m$  were the most prevalent, accounting for 96.3% of all MP samples. MP size at the bay head was smaller than that at the central bay for high-density MPs; however, the relationship was reversed for low-density MPs. Analysis of the MP distribution pattern using a two-dimensional numerical model suggests that the horizontal distribution of MPs in the Otsuchi Bay depends on the size and density of MP particles. It is also strongly influenced by both the tidal oscillating currents characteristic to the bay and vertical terminal velocity of MP particles. Sedimented MP distributions in a bay with a small catchment population with limited MP sources shed light on our understanding of MP transport dynamics.

## 1. Introduction

Plastics are ubiquitous in our daily lives, as they are used in various products, such as clothing, food packaging, personal care goods, and technology. The global plastic production was 407 million tons per year in 2015 (OECD (The Organization for Economic Co-operation and Development), 2018). The global discharge of plastic debris was estimated at over 6300 million tons between 1950 and 2015, and the amount of plastic waste generated is steadily increasing (Geyer et al., 2017). Approximately 6–12 million tons of plastic waste enters the ocean each year, and its accumulation will exceed 250 million tons by 2025 (Jambeck et al., 2015; Lebreton et al., 2017). Plastic polymers found on beaches or those that enter the sea are subjected to UV degradation, thermal degradation, biodegradation, and degradation through wave action (Bissen and Chawchai, 2020; Cole et al., 2011).

Plastic particles with a major axis  $\leq 5$  mm are called microplastics (MPs) (Andrady, 2011). There are an estimated  $10^6$  pieces of floating MPs per square kilometer in subtropical ocean gyres (Law and Thompson, 2014). MPs are widespread throughout the oceans worldwide and are also found in both polar regions (Isobe et al., 2019; Lusher et al., 2015). High MP concentrations have been reported from East Asian (Isobe et al., 2015); a summary of the MPs sampled from surface seawater in Southeast Asia revealed a concentration of  $0.13\text{--}11100 \text{ pieces L}^{-1}$  (Curren et al., 2021). In global, the accumulated number of microplastic particles was estimated as 15–51 trillion particles (93–236 thousand metric tons) in the year 2014, but it corresponds “only approximately 1% of global plastic waste estimated to enter the ocean in the year 2010” (van Sebille et al., 2015). Surface plastic concentration was estimated to increase by 0.1 particle per square meter in the western North Atlantic area in the year 2010, but even if extend this value to global ocean, it yields only an increase of 0.2% of global production (Wilcox et al., 2020). The microplastics should be removed from the surface.

In marine ecosystems, the coastlines, coastal waters, and seabed act as sinks for marine plastic debris (Onink et al., 2021; van Sebille et al., 2020). It is also generally accepted that the seafloor is a



large repository where sediments and particle organic matter from the water column eventually settle (Olsen et al., 1982). Thus, with the huge influx of plastic debris into the oceans, the marine sediments serve as a massive MP sink (Jambeck et al., 2015; Woodall et al., 2014; Yao et al., 2019). Owing to their exceptional durability, plastics are likely to be retained in marine sediments (Jorquera et al., 2022). MP concentrations in seabed sediments around the East Asian region have been reported as 92–414 pcs kg<sup>-1</sup> in the Jagir estuary, Indonesia, 0.8 pcs kg<sup>-1</sup> in the Changjiang Estuary, Shanghai, 25–363 pcs kg<sup>-1</sup> in the Gulf of Thailand, and 24–253 pcs kg<sup>-1</sup> in the Hiroshima Bay, Japan (Firdaus et al., 2020; Sagawa et al., 2018; Wang et al., 2020; Zhang et al., 2019).

Previous studies have estimated MP concentrations in various coastal regions, including sites around densely populated metropolitan and rural areas. Although sediment MP concentrations are higher in urban areas than those in rural areas, a variety of polymers have been detected in urban areas, whereas MP composition in rural areas is limited (Jang et al., 2020). However, the tendency of MPs to sink toward the seafloor has not been fully understood (van Sebille et al., 2020). Understanding MP transport dynamics in the densely populated regions is particular challenging because of the high diversity of MP sources around these regions. Conversely, the diversity of MP sources is limited in rural coastal regions, which makes it easier to examine MP transport. Examining MP concentrations in rural coastal regions would simplify the MP source issues and then possibly provide new insights on MP behavior in coastal regions and their accumulation to the sediments. In this study, Otsuchi Bay was chosen as the study area to investigate MP contamination in a rural coastal environment.

Otsuchi Bay is a small, semi-closed bay with an area of 20.2 km<sup>2</sup> (width: 2–3 km, depth: 80 m at the mouth). Three rivers (Otsuchi, Kozuchi, and Unosumai Rivers) flow into the bay. The Otsuchi area is almost entirely covered by forest; however, Otsuchi City, with a population of approximately 15,000 in 2010, is built near the coastal area of Otsuchi Bay. The bay faces the North Pacific and exchanges seawater with the open ocean, facilitating productive inshore commercial fishing; it is one of the major

regions for inshore fisheries where a variety of seaweeds and shellfish are cultured (Tanaka et al., 2017). In this study, Otsuchi Bay was chosen as the study area for the following reasons: First, the system is relatively simple: size of the bay is not large to observe, the bay mouth is not large, the river inputs are limited to the bay head and a small city locates only in the bay head side. Second, the circulation in the bay has been well investigated by previous studies (e.g. Tanaka et al., 2017; Komatsu and Tanaka, 2017). Third, examining MP concentrations in this region would provide new information on the pollution status of MPs in the countryside in Japan because Otsuchi Bay is one of the typical rural fishery areas in Japan. Elucidating MP accumulation processes to the sediments will contribute towards assessing the effects of MPs on the organisms inhabiting the bay.

Therefore, the aim of this study was to clarify the horizontal distribution, and properties of MPs in the sediments on the seafloor in the Otsuchi Bay and to investigate its sedimentation dynamics. To the best of our knowledge, this is the first study to demonstrate sediment MP pollution in a rural coastal area in Japan.

## 2. Materials and methods

### 2.1 Study area

Otsuchi Bay is a small, semi-closed bay located on the Pacific coast of Tohoku, Iwate Prefecture, Japan (Fig. 1). It is a narrow bay with a length of 8 km and a wide mouth of 3.5 km. During the winter and spring seasons, the dominant winds are from the west to northwest, which facilitates the exchange of water masses between the inner and outer parts of the bay (Shikama, 1980). Three small rivers, the Otsuchi (12.5 km in length), Kozuchi (11.8 km in length), and Unosumai Rivers (23.1 km in length) transport nutrients into the Otsuchi Bay (Yoshikawa et al., 2001). The flow rate of the Otsuchi river system is shown in Table S1.

## 2.2. Sample collection

The MP concentration in the sediment core of the Otsuchi Bay was investigated in March and September 2021; the sediment core was sampled using a research vessel (Grand Maillet, 1.8 t, 10 m) of the International Coastal Research Center, AORI, University of Tokyo, Otsuchi. The sampling points included three stations located at different depths of the Otsuchi Bay (Sta. 1, water depth 18.7 m; Sta. 2, water depth 11.7 m; and Sta. 3, water depth 31.2 m) (Fig. 1, Table 1). Sta.1 is located near the mouth of the Otsuchi River (hereafter referred to as ‘bay head’) and Sta. 3 is located at the center of the bay (hereafter referred to as ‘central bay’). Sta.2 is located between Sta.1 and Sta.3.

The seabed sediments at each station were collected using a triple-tube core sampler with a lid system (ASYURA, Rigosha & Co., Ltd., Tokyo, Japan). The sediment cores were sliced onsite at 5-cm depth intervals and collected in PVC containers. The samples were then transported to the laboratory and stored until further analysis.

## 2.3. Extraction of MPs from sediment core samples

The MP concentration was measured as described below (Cai et al., 2018; Masura et al., 2015). Approximately 10 g WW of the top 5 cm of the sample was weighed in a beaker; 20 mL of NaI solution adjusted to a density of 1.6 g cm<sup>-3</sup> was added to the sample and stirred. The mixture was allowed to stand for 60 min, and the supernatant was then collected by pouring it into another beaker without disturbing the sample settled at the bottom. The collected supernatant was sieved through a 10-μm mesh and the particles remaining on the sieve were washed with distilled water. The particles were then treated with 20 mL of 30% H<sub>2</sub>O<sub>2</sub> and incubated for 72 h at 60 °C. After 72 h, the particles were filtered through a 1-μm polytetrafluoroethylene (PTFE) filter (25 mm dia., Merck Millipore Ltd, Billerica, MA, USA). The PTFE filter containing the MPs was then dried at 26 °C for approximately 12 h. The operation was repeated by separating three subsamples from the main sample at each station

and using them as replicates.

## **2.4. Analysis of MPs**

### **2.4.1 Polymer identification**

The infrared absorption spectrum of the particles collected using the PTFE filter was measured using FTIR equipped with an infrared microscope (Nicolet iN10, Thermo Fisher Scientific, Madison, WI, USA). FTIR measurements were obtained in transmission mode using 16 background scans at 4 cm<sup>-1</sup> resolution. The wavenumber range of the measurement was 4000–675 cm<sup>-1</sup> (Peng et al., 2017). The aperture size was 60 µm and the step size was 30 µm.

In this study, the sediment cores sampled contained five typical types of MPs: polyethylene (PE), polypropylene (PP), polystyrene (PS), polyamide (PA), and polyvinyl chloride (PVC). The intensity of the spectra between all candidate particles separated using the filter and the five standard plastics was calculated using a specific wavenumber range (Table S2).

The MP candidate particles were determined using an intensity > 0 as the threshold value. Furthermore, the spectra of the MP candidate particles were compared and confirmed using the infrared library database provided by OMNIC (version 9.12, Thermo Fisher Scientific, USA); the type of MP was determined when the Hit Quality (HQ) of their spectrum was ≥ 60% (Hanke et al., 2013; Zhu et al., 2019). The filter image was saved using Mosaic capture tool provided by OMNIC. The major axis of the MPs was defined by the largest length of each irregular-shaped fragment measured by OMNIC.

### **2.4.2 Quality assurance and quality control**

All instruments and containers were cleaned with MilliQ-water before use to reduce potential contamination owing to MPs generated from them. Sampling and analysis devices were covered with

aluminum foil when not in use (Enders et al., 2015). Method blank tests were also conducted three times to check potential contamination during analysis; all reagents used for analysis were filtered through a 1 µm PTFE filter. Additionally, a laboratory blank test was conducted throughout the treatment process using a 1 µm PTFE filter (47 mm diameter) placed in a Petri dish during the experimental procedure (Quinn et al., 2017). Both filters were examined using the Fourier transform infrared spectroscopy (FTIR) microscope to rule out possible MP contamination from the laboratory environment during analysis.

#### 2.4.3 Standardization of MP recovery percentage estimation

To standardize the procedure for determining the percentage of MPs recovered from core sediments, 20 spherical standard polyethylene particles each of various sizes (45 µm, 100 µm, and 300 µm, density 0.96; Cospheric LLC, Santa Barbara, CA, USA) were mixed with the Sta. 1 sediment sample (10g wet weight). The extraction and recovery procedures followed are described in section 2.3. The number of recovered polyethylene particles was recorded by identifying their spherical shape using an optical microscope (Santa II, Micronet Inc., Saitama, Japan).

#### 2.5. Statistical analysis

MP concentration data in the sediment cores are expressed as the mean ± standard deviation (S.D.). Statistical analysis was performed using SPSS software (version 26.0, IBM Corporation). Nonparametric Kruskal–Wallis test was used as a multiple comparison test to compare MP concentration and average particle size at each observation point because the data failed the normal distribution test. If the test indicated a significant difference, multiple comparisons were performed using a Steel-Dwass post-hoc test. Significant differences for samples collected between March and September 2021 were determined using Steel-Dwass test at a 95% confidence level.

## 2.6. Numerical experiments

To investigate the mechanism determining size dependency of horizontal distribution for high-density MPs, we conducted simple numerical experiments. A numerical model was established in a two-dimensional domain extending in the horizontal ( $x$ ), and vertical ( $z$ ) directions. The  $x$ -axis is extended eastward from the river mouth (0 m) to the central part of the bay (5000 m, Fig. 1), and the  $z$ -axis is extended downward from the sea surface (0 m) to the bottom (30 m). Horizontal positions of pseudo particles  $\mathbf{x}$  at time  $t + \Delta t$  were computed as follows:

$$\mathbf{x}(t + \Delta t) = \mathbf{x}(t) + \mathbf{u}\Delta t, \quad (1)$$

where  $\mathbf{u}$  is the horizontal current vector and  $\Delta t$  (=10 s) is the time increment of the model. The horizontal current vectors were calculated using a combination of the ocean currents observed in the Otsuchi Bay and Stokes drift estimated using the scheme described by Isobe et al. (2014), applying the 4-year averages of wave parameters observed in the Otsuchi Bay (Komatsu and Tanaka, 2017). The Stokes drift velocity  $u_s$  in the finite ocean depth ( $h$ ) was computed as follows using a random walk dispersion (Isobe et al., 2014):

$$u_s = -\frac{a^2 \sigma k \cosh\{2k(h+z)\}}{2(\sinh kh)^2} + \frac{R\sqrt{2K_h \Delta t}}{\Delta t}, \quad (2)$$

where  $a$  denotes the wave amplitude,  $\sigma$  is the angular frequency,  $k$  is the wave number,  $R$  is a random number generated at each time step with an average and standard deviation of 0.0 and 1.0, and  $K_h$  is the horizontal diffusivity.

The vertical positions ( $z$ ) of the simulated particles at time  $t + \Delta t$  were calculated numerically using those at time  $t$  as follows:

$$z(t + \Delta t) = z(t) + \mathbf{w}\Delta t, \quad (3)$$

The vertical velocity ( $w$ ) was calculated using a combination of a terminal sinking velocity based on the experimental trials of Kaiser et al. (2019) and a random walk as follows:

$$w = 11.68 + 0.1991d + 0.0004d^2 - 0.0993\Delta\rho + 0.0002\Delta\rho^2 + \frac{R\sqrt{2K_z\Delta t}}{\Delta t}, \quad (4)$$

where  $d$  is the particle size,  $\Delta\rho$  the density difference between high-density MP particles and seawater.

The vertical diffusivity,  $K_z$ , was computed using the scheme described by Kukulka et al. (2012):

$$K_z = 1.5u_*k_cH_s, \quad (5)$$

where  $u_*$  represents the friction velocity ( $= 0.00012W_{10}$ ),  $k_c$  is the von Karman constant (0.4),  $H_s$  is the significant wave height, and  $W_{10}$  is the 10-m wind speed (Iwasaki et al., 2017). The values of  $H_s$  and  $W_{10}$  are given by the 4-year averages observed in the Otsuchi Bay (Tanaka et al., 2017). The values of the parameters used in Eqns. 2–5 are listed in Table A1.

The computational procedure was as follows. A thousand MP particles were continuously released at  $x = 0$  and  $z = 0$  from  $t = 0$  h through  $t = 12$  h at 5 min intervals. The sizes of the pseudo particles were adjusted to 150  $\mu\text{m}$  and 250  $\mu\text{m}$  to equal the observed high-density MPs size value at Sta. 1 and Sta. 3 (Fig. 4). The horizontal current vector  $\mathbf{u}$  was calculated as follows:

$$\mathbf{u}(\mathbf{t}) = \mathbf{u}_{current} - \mathbf{u}_s, \quad (6)$$

where  $\mathbf{u}_s$  represents Stokes drift and  $\mathbf{u}_{current}$  represents the ambient currents. In the northern part of the bay, the ambient current is reported to flow eastward in the upper layer and reverse periodically in the lower layer (Tanaka et al., 2017). To imitate the ambient currents, we used  $\mathbf{u}_s$  changing at a depth of 10 m as follows:

$$\mathbf{u}_{current} = \begin{cases} \mathbf{u}_a & \text{for } z \leq 10 \text{ m} \\ \mathbf{u}_a \pi \sin\left(\frac{2\pi}{T}t\right) & \text{for } z > 10 \text{ m} \end{cases} \quad (7)$$

where  $\mathbf{u}_a$  is 0.1  $\text{m s}^{-1}$  which is the average current velocity observed in the Otsuchi Bay (Tanaka et al., 2017) and  $T$  is 12 h, representing the M2 tide period. The particles were tracked for 24 h, which was considered an adequate amount of time for particles to reach the seafloor. The model did not include particles with a transport distance lower than 0.

### 3. Results

#### 3.1. The recovery rate

The recovery percentages of the plastic particles measuring 45  $\mu\text{m}$ , 100  $\mu\text{m}$ , and 300  $\mu\text{m}$  were  $85.0 \pm 4.0\%$ ,  $96.7 \pm 4.7\%$ , and  $100 \pm 0\%$ , respectively (Table 3). The results of the recovery test indicate efficient recovery of MPs with a particle size  $\geq 300 \mu\text{m}$ ; the recovery percentage was lower when the particle size was  $\leq 100 \mu\text{m}$  (Table 3). Approximately 80% of the plastic particles of 40–90  $\mu\text{m}$  diameter could be recovered through the recovery operation that was repeated three times. Therefore, the plastic particles in the seabed sediment were considered mostly recovered using the method employed in this study.

#### 3.2. Identification of MPs

FTIR analysis identified a total of seven polymer types including polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polyamide (PA), polystyrene (PS), co-polymers, and other synthetic polymers, such as rubber, resin, and additives (Fig. 2). The polymers were divided into high-density and low-density polymers by comparing their densities in seawater (Kershaw et al., 2019). The densities of different polymer types are shown in Table S3.

Low-density PE and PP were the main polymer types with a proportion of over 60% at Stas. 1, 2 in March 2021 (Fig. 2a). High-density plastic PA and PS constituted 5% or less, followed by PVC, which was 3% at each station in March 2021. Contrarily, the proportions of low-density PE and PP were < 30% at each station in September 2021 (Fig. 2b).

#### 3.3. Spatial and temporal distribution of MPs

The MP concentrations in 0–5 cm seabed sediments at Stas. 1, 2, and 3 in March 2021 were  $13.6 \pm 9.8$ ,  $1.7 \pm 0.6$ ,  $2.6 \pm 0.3$  pcs  $\text{g}^{-1}$  DW and in September 2021 were  $2.8 \pm 0.3$ ,  $2.6 \pm 1.4$ ,  $5.1 \pm 1.2$  pcs  $\text{g}^{-1}$



DW, respectively (Table 2). The MP concentration of Sta. 1 located at the bay head near the Otsuchi River was significantly higher than that of Sta. 2 in March 2021 (Steel-Dwass test,  $p < 0.05$ ). The total number of MPs was 224 and 146 pieces in March and September, respectively.

Nevertheless, there were no significant differences between observations recorded in March and September (Steel-Dwass test,  $p > 0.05$ ), implying that the spatial MP concentration was more remarkable than their temporal distribution.

### 3.4. Size distribution of MPs

All the MP particles were  $< 5000 \mu\text{m}$  and approximately 96.3% of the MP particles were  $< 1000 \mu\text{m}$ ; the most frequently observed MPs were in the range of  $50\text{--}100 \mu\text{m}$  (Fig. 3). These results indicate that the proportion of observed MPs increased with decreasing plastic particle size.

The size ranges (average particles size  $\pm$  standard deviation) of MPs were  $350.8 \pm 323.9$ ,  $267.7 \pm 353.2$ , and  $149 \pm 74.9 \mu\text{m}$  for low-density MPs at Stas. 1, 2, and 3, respectively, whereas those for high-density MPs were  $162.1 \pm 111.8$ ,  $145.3 \pm 81.7$ , and  $278.4 \pm 181.2 \mu\text{m}$ , respectively (Fig. 4). The high-density MP size at the bay head was significantly smaller than that of MPs at the central bay. However, low-density MPs were significantly larger in the bay head than those at the central bay (Steel-Dwass test,  $p < 0.05$ ). The horizontal distribution of average particle sizes of low- and high-density MPs was reversed in the sediments between the bay head and central bay. This suggests that the horizontal distribution of MP particle size is dependent on their particle density.

### 3.5 Numerical simulations

The numerical simulation indicate that the  $250 \mu\text{m}$  MP particles accumulated around  $x = 4500 \text{ m}$  which is near Sta. 3 (Fig. 5). The results of the numerical model matched our observations, as shown in Fig. 4. In contrast,  $150 \mu\text{m}$  MP particles accumulated around  $x = 1500 \text{ m}$ , which is close to Sta. 1.

The essence of this difference is that the sinking speed for 250  $\mu\text{m}$  MPs particles was higher than that of 150  $\mu\text{m}$  MP particles. This is sufficiently fast to allow particles to be subjected to the stirring effect caused by tidal oscillating motion to a lesser degree (Fig. 6). Thus, the 250  $\mu\text{m}$  MPs were transported to the central bay in most cases. In the experiments, the effects of Stokes drift were quite weak and attenuated exponentially with depth as shown in Fig. S1.

## 4. Discussion

### 4.1. MP contamination of seabed sediments in the Otsuchi bay

In this study, MP concentrations in the sediment of Otsuchi Bay ranged between 1.7–13.6  $\text{pcs g}^{-1}$  DW. Xia et al. (2021) investigated the distribution of MPs in sediments of the Liang Feng River, Guilin, South China, which has large catchment populations. They reported MP concentrations of  $33.2 \pm 11.99$   $\text{pcs g}^{-1}$  DW in the sediments, which was several times higher than that of the Otsuchi area. Furthermore, the reported MP concentrations in the sediments of other urban areas were 1,000–30,000  $\text{pcs g}^{-1}$  DW in the Santos and Sao Vicente Estuary in Brazil (Gimiliani et al., 2020) and 200  $\text{pcs g}^{-1}$  DW sediment in Gulf of Thailand (Bissen and Chawchai, 2020). This suggests possible influences of coastal urbanization and catchment population to the MP concentration on the seabed (Shi et al., 2022).

Wang et al. (2021) reported that small MP concentrations ranged from 53.3–145  $\text{pcs g}^{-1}$  DW in the sediments of the Tokyo Bay, which is 10 times higher than that at the Otsuchi Bay. However, the catchment population of the Tokyo Bay is 29 million, which is 2000 times higher than that of the Otsuchi Bay. The concentration ratio of MPs in the sediments does not match the population ratio. This might be attributed to the inadequate number of sampling stations at the Tokyo Bay in the study conducted by Wang et al. (2021). The area of the Tokyo Bay is 1,380  $\text{km}^2$  and 68 times of that of Otsuchi Bay (20.2  $\text{km}^2$ ). More than 100 rivers flow into the Tokyo Bay. Because of the wide area and complex system, it is difficult to estimate the total MPs on the seabed. Again, it supports the

importance of the research on MPs in the rural coastal area. Regardless, it should be emphasized that the MP concentrations are likely not simply proportional to the population in the catchment area; the MP concentrations in rural coastal areas may be higher than expected owing to various associated factors.

No significant difference in MP concentrations was detected between March and September. This result is consistent with that of previous studies, which reported no significant differences among the four seasons in the Sanggou Bay, China (Sui et al., 2020). In addition, a study on MPs in sediments in the Qingduizi bay, North Yellow Sea, China, revealed that there was no significant difference in either MP abundance or shape composition in sediment in offshore areas between summer and winter (Chen et al., 2022). The retention time of MPs in sediments was longer compared with that of floating MPs in water which are more mobile owing to the influence of wind and currents (Woodall et al., 2014). Thus, observing significant differences following a one-year observation period is challenging.

The proportion of high-density MPs, such as PA, increased at each station in September 2021 compared to that in March 2021 (Fig. 2). Chi-square tests ( $\chi^2$ ) indicated a significant difference in polymer type between March and September ( $p < 0.01$ ). This could be attributed to the increase in rainfall which could have increased the influx of river water (Zhao et al., 2019). This would facilitate resuspension of MPs during the high flow rate season, resulting in an increased proportion of high-density MPs (Yan et al., 2021). Critical shear stress which resuspends MPs from the riverbed depends on the MP polymer type (Waldschlager and Schuttrumpf, 2019; Ballent et al., 2013). Moreover, the flow rate of the Otsuchi river increased in September 2021 (Table S1). Thus, the intense flow rate of the Otsuchi river during September likely caused the change in MP proportions observed in the Otsuchi Bay during this period.

Lin et al. (2021) investigated MP accumulation in coastal sediments of the East China Sea and found that MPs  $< 100 \mu\text{m}$  in size were dominant in the upper layer of sediment. Overall, these findings are

in accordance with those reported by another study, which states that MP abundance increases as fragment size decreases (Hale et al., 2020). The average particle size of MPs in the bay head of the Tokyo Bay was 68.3  $\mu\text{m}$  (Wang et al., 2021), which is much smaller than the MP size of 225  $\mu\text{m}$  observed in the Otsuchi Bay (Table 4). Both Wang et al. (2021) and this study used 1  $\mu\text{m}$  PTFE filters. The value of the Enclosed Index (E.I.) reflects the residence time of seawater in a bay (Nakano et al., 2021). It was calculated as follows:

$$E.I. = \frac{\sqrt{S} \times D_1}{W \times D_2}, \quad (8)$$

where S is the area of the bay, W is the width of the bay mouth,  $D_1$  is the maximum depth, and  $D_2$  is the depth at the bay mouth. The E.I. value represents geographic characteristics and the E.I. value of the Tokyo Bay was 1.78, which was larger than the Otsuchi bay's E.I. value of 1.1 (Table 4). The MP particles may remain for a considerably longer duration in the Tokyo Bay because of its higher degree of closure. This implies that the residence time is much longer in the Tokyo Bay, which may cause greater deterioration of particles than that in the Otsuchi Bay. However, as mentioned above, the catchment area of rivers flowing into the Tokyo Bay is much larger. Moreover, the Tokyo Bay is bordered by megacities that generate huge amounts of microplastics that are not only discharged into the rivers but also emitted into the atmosphere (Liu et al., 2019). Therefore, quantifying the degree of MP deterioration between the two bays apart from the effects of deterioration on land and atmospheric transportation is challenging, and these aspects should be further examined in future research.

A previous study reported that the deposition rate of sediment particles off the Tamagawa River in the Tokyo Bay was 0.6  $\text{g cm}^{-2} \text{y}^{-1}$  or 0.89  $\text{cm y}^{-1}$  (Okada et al., 2016). It would take 4–5 years for 3–5 cm (sampling thickness by Wang et al., 2021) of sediment to accumulate. Wang et al. (2021) estimated MP weight concentration as 20.7  $\text{mg g}^{-1} \text{DW}$ . They also estimated the MP deposition rate as 12.4  $\text{mg cm}^{-2} \text{y}^{-1}$ , by deviding the MP weight concentration by the sediment deposition rate in the Tokyo Bay. In addition, Takahashi et al. (1999) reported that the sedimentation rate in the Otsuchi Bay

was 0.17–0.36 cm y<sup>-1</sup>. It would take 14–19 years for 5 cm (sampling thickness by this study) of sediment to accumulate. The dry weight density of the sediment was 0.47 g DW cm<sup>-3</sup>. Thus, a deposition rate of 0.08–0.17 g cm<sup>-2</sup> y<sup>-1</sup> was estimated for sediment particles in the Otsuchi Bay, which is much smaller than that of the Tamagawa River. MPs are mainly composed of PP, PE, and PS, with an approximate density of 1.0 g cm<sup>-3</sup>. Assuming that all particles were spherical, the volume of MPs at Sta. 1 was  $6.3 \times 10^{-2}$  cm<sup>3</sup>; the weight concentration of MPs was  $6 \times 10^{-3}$  g g<sup>-1</sup> DW. The MP deposition rate was estimated as 0.5–1.0 mg cm<sup>-2</sup> y<sup>-1</sup>, which is 12–26 times smaller than that of the Tamagawa River.

#### 4.2. Dependency of the horizontal distribution of high-density MPs on their particle size

Previous studies indicate that small plastic fragments are carried partly by the mass transport (Stokes drift) generated in the uppermost layer in response to wind waves (Iwasaki et al., 2017; Li et al., 2020; Zhang, 2017). In the ocean, the combination of ambient ocean currents, wind, and Stokes drift contributes to the horizontal transport process of MP particles. Both the size and density of MP particles influence the sinking process determined by vertical terminal velocity (Isobe et al., 2014). This study indicated that the particle size of low-density MPs was larger in the bay head (Sta. 1) than that in the central bay (Sta. 3) (Fig. 4). This result corresponds with those of previous numerical experiments, which indicated that smaller MPs tend to spread offshore from the bay head by Stokes drift because of slower vertical terminal velocity, referred to as selective transport process (Isobe et al., 2014).

However, the numerical model in Isobe et al. (2014) did not include the effects of ambient ocean currents and discussed only low-density MPs, whereas our observation revealed that horizontal size distribution was reversed between low and high-density MPs (Fig. 4); the particle size dependency of the horizontal distribution for high-density MPs cannot be reproduced using Isobe's model. Therefore,

a new numerical experiment was conducted not only to discuss the effect of Stokes drift and ocean currents but also to clarify the factors that explain the distribution of high-density MPs.

Our model demonstrated the size dependent effects of tidal currents on high-density MPs. The spatial distributions of modeled particles match the observation results (Figs. 4 and 5). The size dependent distribution of high-density MPs on seabed was explained by the difference in sinking duration between the smaller and larger high-density MPs. In the 30 m depth case of the numerical experiments (Fig. 6), the average sinking duration was  $17.6 \pm 1.9$  h ( $11.6 \pm 1.7$  h at the depth of 10–30 m) for 150  $\mu$ m particles and  $9.4 \pm 0.8$  h ( $6.2 \pm 0.7$  h at the depth of 10–30 m) for 250  $\mu$ m ones, depending on their sinking velocities. The 150  $\mu$ m particles had a slower sinking velocity (the terminal velocity is  $1.0 \times 10^{-3}$  m s<sup>-1</sup>), so they were swept toward the bay head due to the tidal currents that were reversed in direction from the surface currents by the time the particles reached the seafloor. On the other hand, the 250  $\mu$ m particles had a faster sinking velocity (the terminal velocity is  $2.0 \times 10^{-3}$  m s<sup>-1</sup>), so they were less affected by the reversed tidal currents. In coastal areas such as the Otsuchi Bay, where tidal currents are often more influential, the horizontal distribution of MP particles on the seafloor is likely to be dependent on their size. It should be emphasized that this point was revealed by observing in a rural small bay where MP origin was relatively clear. Wave and wind statistics in the Otsuchi Bay are shown in Table S4. Surface waves in the Otsuchi bay are dominated by swells propagating from the offshore region (Komatsu and Tanaka, 2017). Even though wind fluctuations may cause local wind seas to develop temporarily, there is no doubt that the Stokes drift from the mouth to the head of the bay is predominant on average. Surface winds are predominantly southerly to westerly in the bay (Komatsu and Tanaka, 2017), leading to wind drift currents that flow predominantly from the head to mouth of the bay. In the upper layer above the pycnocline, the offshore current dominates on average (Tanaka et al., 2017), as set up in the simple model of this study. In contrast, the current direction in the lower layers changes with both diurnal and semi-diurnal

oscillations of tides (Tanaka et al., 2017). Although the current velocity fluctuates in the range of 0–0.2 m s<sup>-1</sup> in both the upper and lower layers, for simplicity, this study used the average current velocity, 0.1 m s<sup>-1</sup>. We believe that even such a simple model can represent an average depiction of the currents in the bay.

However, the observations and simplified 2D numerical experiments have several limitations. First, there is no direct indication of the duration of sedimentation of 0–5 cm sediment deposition. Second, the model based on the averaged current velocity might be too idealized to reproduce the MP transport process. It is difficult to integrate the model over a long period to evaluate the trends of MPs in the Otsuchi Bay. The current field in the real ocean is more complex due to various factors including wind fluctuations, offshore currents, topography, etc. It is therefore essential to use a 3D general ocean model to reproduce a more realistic velocity field (Sakamoto et al., 2017). In addition, the resuspension of MPs was not considered in the model. Topographic effects, such as changes in the bottom slope of the bay, were also not considered in the model. Notably, slope-induced tidal straining might play a critical role in MP particle transport (Endoh et al., 2016; Schulz et al., 2017). The tidal straining would have an upslope effect on the smaller MP particles and a downslope effect on the larger MP particles. The reproduction of MP transport needs to incorporate more complex mechanisms in a non-hydrostatic ocean model.

#### 4. Conclusions

In this study, we observed the horizontal distribution of the concentrations and composition of MPs in sediments on the seafloor of the Otsuchi Bay, one of the smallest semi-closed bays in Sanriku on the Pacific coast of Tohoku, as a representative of rural coastal area in Japan. The concentrations of seabed sediments near the Otsuchi River mouth were higher than those in the central bay. The MP concentrations in the sediment ranged from  $2.6 \pm 0.3$  to  $13.6 \pm 9.8$  pcs g<sup>-1</sup> DW and  $2.6 \pm 1.4$  to  $5.1 \pm$

1.2 pcs g<sup>-1</sup> DW in March and September 2021, respectively. The MP concentration and estimated MP deposition rate were much smaller than those in the Tokyo Bay. The horizontal distribution of average particle sizes of low- and high-density MPs was reversed in the sediments between the bay head and the central bay. The simple particle-tracking model incorporated horizontal transport owing to both currents (mean estuary circulation and tidal current) and Stokes drift with conventional sinking processes of high-density MPs. The model successfully reproduced a more concentrated sink of larger MP particles near the central bay and revealed the importance of tidal currents and sinking velocity. This study makes several contributions to research on MPs in coastal areas by combining field observations that showed different horizontal size distributions and numerical experiments which reproduced the high-density MP transport process. Our study sheds new light on MP contamination in a rural area. MP concentrations in coastal areas with small populations may be higher than expected if the population ratio is considered. We hope to include a 3D general ocean model in future research to reproduce a more realistic sinking process.

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## Appendix

### A. Supplementary data



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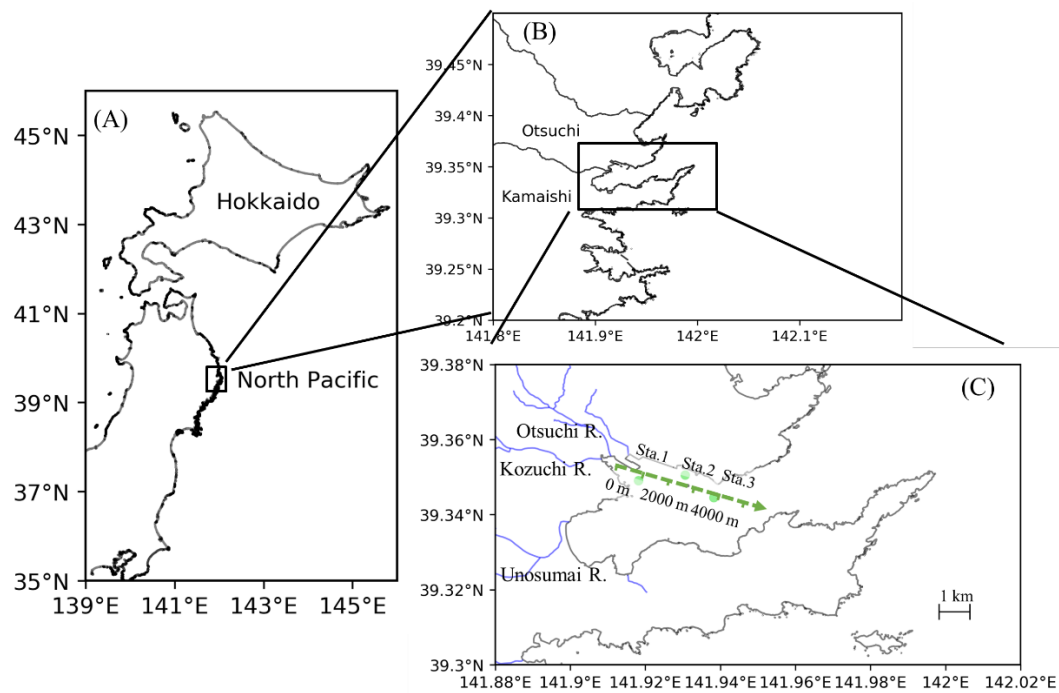


Fig. 1. Map of sampling area and simulation area (green dashed line)

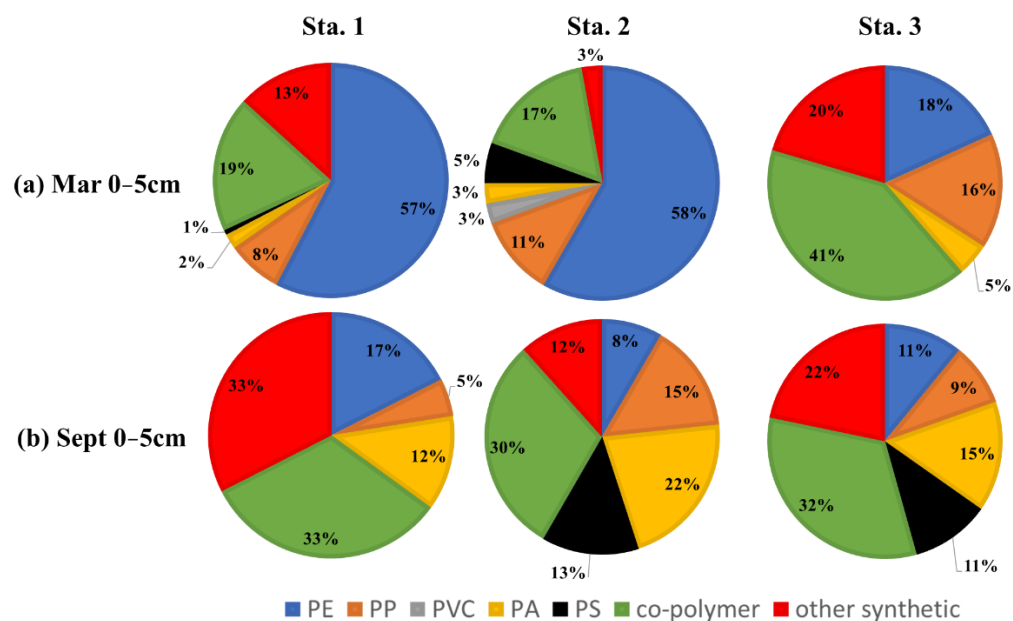


Fig. 2. Proportion of polymer types in the sediments at each station in (a) March and (b) September.

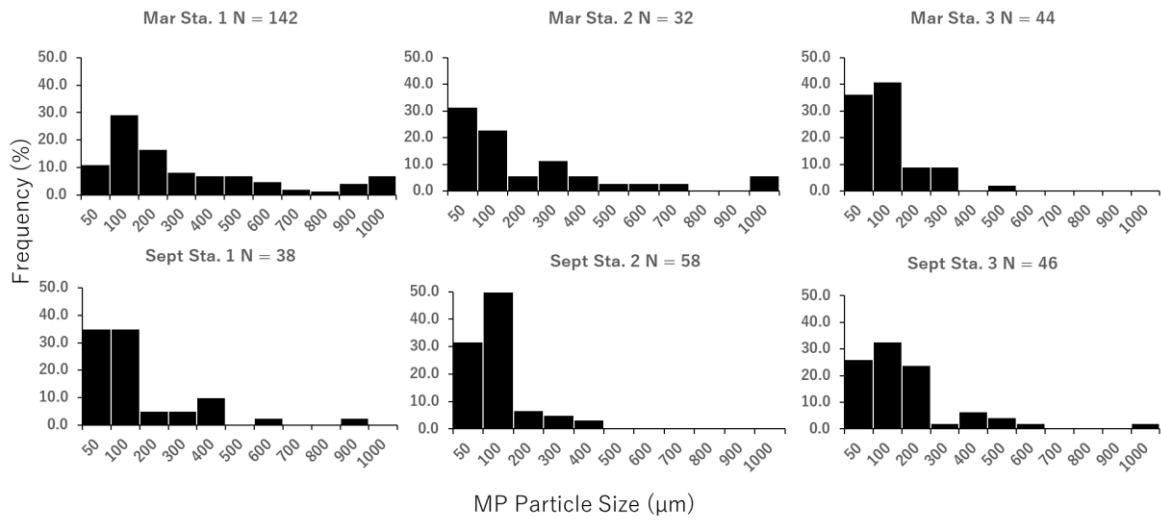


Fig. 3. Size distribution of MPs in the sediments at each station in March (upper) and September (lower).

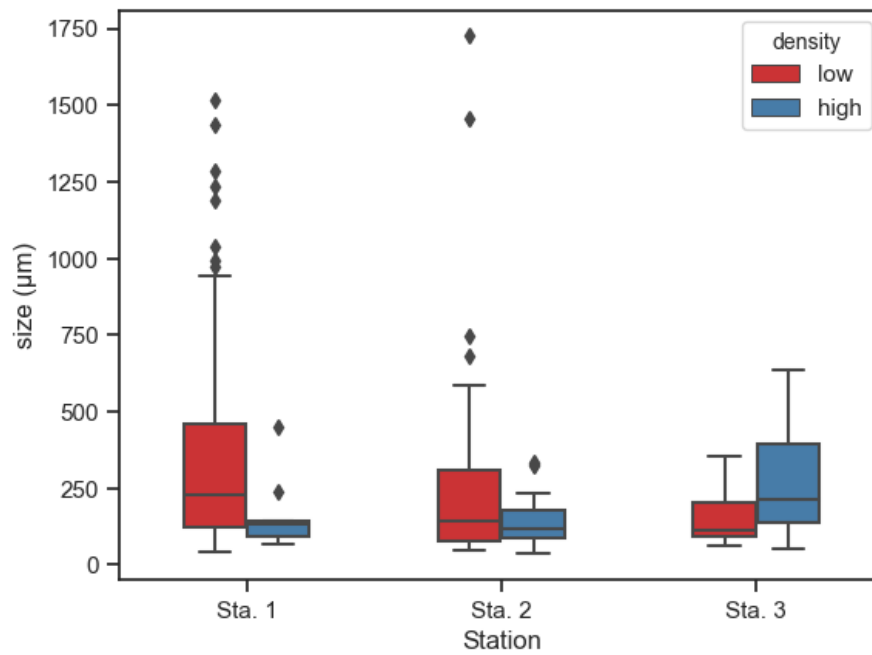


Fig. 4. Size distribution of MPs categorized by the density at each station. The data for March and September were combined.

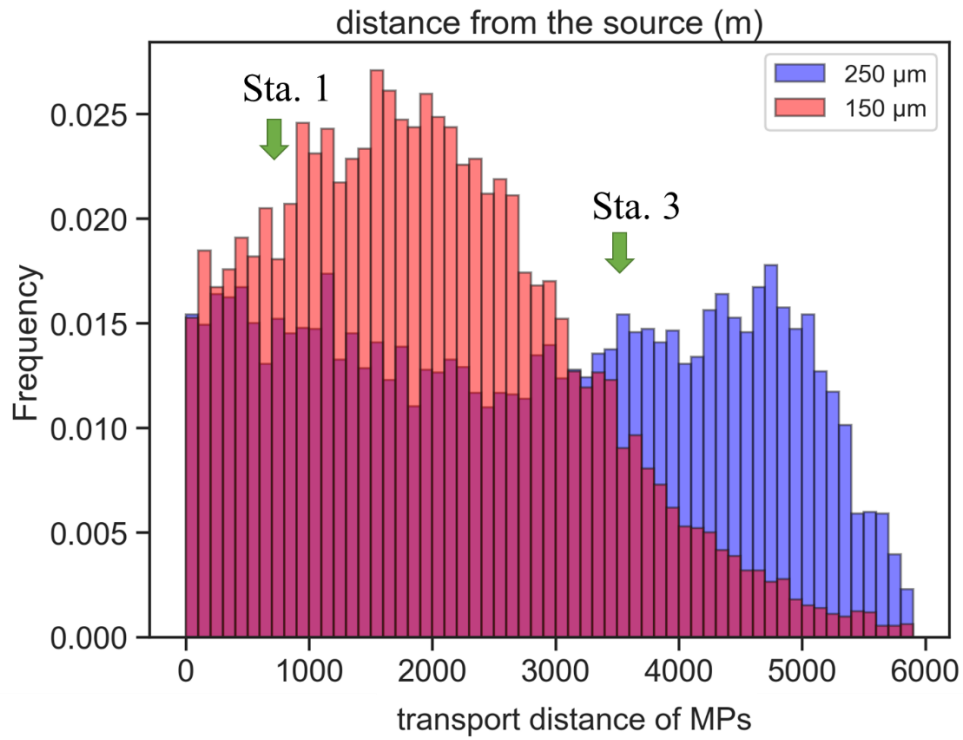


Fig. 5. The frequency histogram of MP transport distance (1150 g/cm<sup>3</sup>)

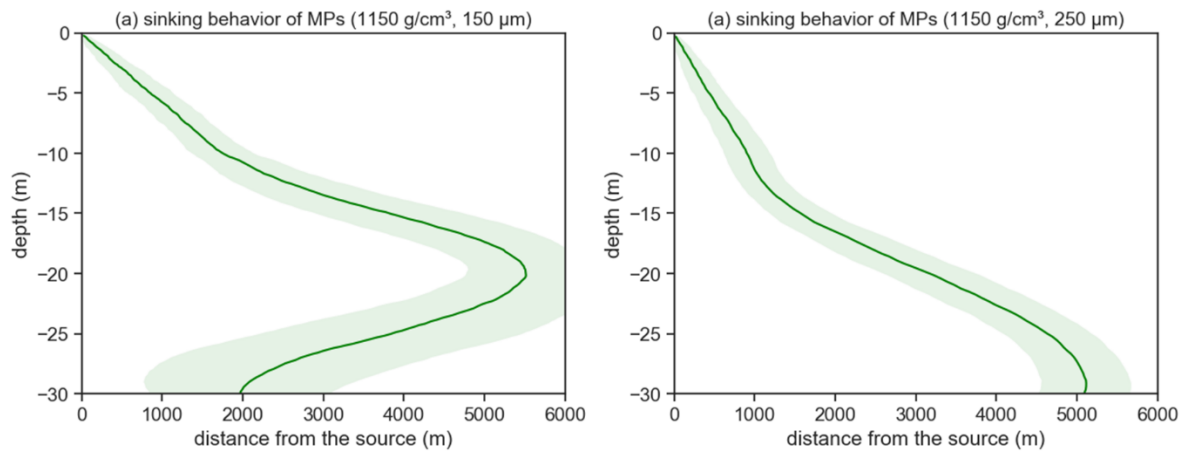


Fig. 6. The typical sinking profile of MPs during high tide ( $1150 \text{ g/cm}^3$ , (a)  $150 \text{ }\mu\text{m}$ , (b)  $250 \text{ }\mu\text{m}$ )



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Table 1. Sampling stations and sampling characteristics

Station	Month	Core depth (cm)	Latitude (°N)	Longitude (°E)	Depth (m)
Sta. 1	Mar	0–5	39.3495	141.9183	18.7
Sta. 2	Mar	0–5	39.3513	141.9308	11.7
Sta. 3	Mar	0–5	39.3464	141.9332	31.2
Sta. 1	Sept	0–5	39.3492	141.9182	18.5
Sta. 2	Sept	0–5	39.3511	141.9307	9.6
Sta. 3	Sept	0–5	39.3462	141.9332	38.5

Table 2. MP Concentration in March and September

Station	N		Mean concentration $\pm$ S.D. (pcs g <sup>-1</sup> DW)	
	Mar	Sept	Mar	Sept
Sta. 1	3		13.6 $\pm$ 9.8	2.8 $\pm$ 0.3
Sta. 2	3		1.7 $\pm$ 0.6	2.6 $\pm$ 1.4
Sta. 3	3		2.6 $\pm$ 0.3	5.1 $\pm$ 1.2

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Table 4. Comparison of MPs in the Tokyo and Otsuchi Bays

Location	Concentration (pcs g <sup>-1</sup> DW)	Average particle size (μm)	Enclosed Index (E.I.)	Reference
Tokyo Bay	53.3–145	68.3	1.78	Wang et al. (2021)
Otsuchi Bay	1.7–13.6	225	1.10	current research

Variables	Physical quantities	Values
$\Delta t$	Time increment	10 s
$a$	Wave amplitude	0.8 m
$\sigma$	Wave frequency	$0.7 \text{ s}^{-1}$
$k$	Wave number	$0.05 \text{ m}^{-1}$
$h$	Ocean depth	30 m
$K_h$	Horizontal diffusivity	$10 \text{ m}^2 \text{ s}^{-1}$
$\Delta\rho$	Density difference	$130 \text{ kg m}^{-3}$
$\eta$	Viscosity of seawater	$1.025\times 10^{-3} \text{ kg m}^{-1} \text{ s}^{-1}$
$H_s$	significant wave height	0.8 m
$W_{10}$	10-m wind speed	$5 \text{ m s}^{-1}$
$g$	Gravitational acceleration	$9.8 \text{ m s}^{-2}$
$K_z$	Vertical diffusivity	$0.01 \text{ m}^2 \text{ s}^{-1}$

Table A1. Values of parameters used in Eqs. 2–5.

### **Declaration of interests**

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

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

## **CRedit author statement**

The contributions of the authors are as follows.

**Yehao Wang:** Conceptualization, Visualization, formal analysis, Software, investigation, Writing - Original Draft, Writing - Review & Editing. **Rei Yamashita:** Formal analysis, Writing - Review & Editing. **Yoshimasa Matsumura:** Software, Writing - Review & Editing. **Shin-ichi Ito:** Project administration, Funding acquisition, Resources, Writing - Review & Editing. **Kosei Komatsu:** Project administration, Resources, investigation, Funding acquisition, Supervision, Writing - Review & Editing.



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