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科学研究費助成事業　　研究成果報告書

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研究課題名（和文）Tracking the behavior of environmental microplastic aggregates using multiple analytical approaches

研究課題名（英文）Tracking the behavior of environmental microplastic aggregates using multiple analytical approaches

研究代表者

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研究成果の概要（和文）：過去十年間において、水圏環境のマイクロプラスチックの地球規模の調査は、科学的に幅広く関心が持たれている。本研究は、同位体分析及び示唆熱天秤-質量分析法を含む多次元分析を行い、環境中のマイクロプラスチックの挙動及び特性を明らかにしたものである。マイクロプラスチックサンプルは、日本の対照的な二つの沿岸域で採取した。本研究により、マイクロプラスチックの劣化が水圏環境における挙動及び特性に重要な役割を担っていることが示唆された。

研究成果の学術的意義や社会的意義
Considering that plastic debris can distribute various pathways around the world, the research outcome and the multidimensional analytical approaches demonstrated in the present study could lay the foundation for further research to elucidate their characteristics and behavior in the world.

研究成果の概要（英文）：The global survey for the presence of microplastics (MPs) in aquatic environments has attracted widespread scientific attention over the past decade. However, evaluating the composition and characteristics of these anthropogenic debris using highly sensitive techniques is still under consideration. This study demonstrates a multidimensional analytical approach, including isotopic, radiotracers, and thermogravimetric analyses to evaluate characteristics and behavior of MPs in the environment. The present study provides fundamental data of environmental MPs from the isotopic and thermogravimetric aspects and highlights the usefulness of the approach for advances in MP research.

研究分野：Environmental Science

キーワード：Microplastic　biofilm　coastal sediment　stable isotopic ratio　radiocesium　thermal analysis　coastal environments

１．研究開始当初の背景

Plastic debris in the aquatic environment has become a growing concern due to its long-term ecological consequences (Thompson et al., 2004). Since it is well recognized that plastics provide surfaces for microbial colonization and biofilm formation in the aquatic environment (**Fig. 1**), organic matter cycling can be altered by plastic-associated biofilm due to adhered distinctive microbial functional diversity. Furthermore, the altered surface of plastics enhances their interaction with pollutants (Battulga et al., 2022b) including radionuclides in the environment.

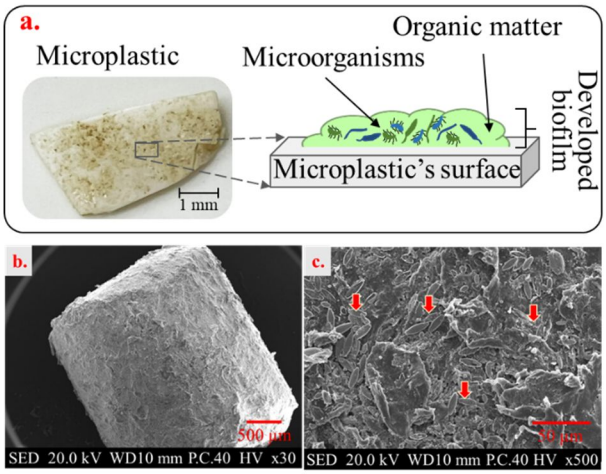

Fig. 1 Developed biofilms on microplastic (a) and SEM images of surface of microplastic (b-c).

Although microplastics (<5 mm in size) have received special attention from the scientific community due to their widespread distribution and relatively large specific surface area, the characteristics of the environmental microplastics and their possible ecological consequences are not yet fully understood. To account for the properties of plastics, various techniques, including microscopy and spectroscopy, have been broadly used to track their characteristics (Birch et al., 2021). However, the application of highly sensitive techniques including isotopic, radiotracer, and thermogravimetric approaches is still under consideration.

２．研究の目的

The main purpose of the research was to evaluate sources and differences of plastic-associated biofilms in the coastal environments of Japan using multiple analytical approaches. Given the potential impacts of the occurrence and composition of microplastics on the aquatic ecosystem, the research first addressed the distribution and properties of microplastics in two contrasting coastal areas of Japan. Secondly, the stable isotope signatures ($\delta^{13}C$ and $\delta^{15}N$) of plastic-associated biofilms were determined based on a newly developed extraction method. Furthermore, the interaction mechanism between environmental plastics and radionuclides especially radiocesium (^{137}Cs) was targeted to understand the radioactivity of environmental plastics. In terms of an interdisciplinary perspective to understand the behavior and degradation of the plastics, spectroscopic and thermogravimetric analyses were also conducted.

３．研究の方法

During the research project, environmental plastics and surrounding compartments (surface waters, bottom sediments, and coastal sands) were collected from the coastal areas of Ibaraki, Niigata, and Fukushima prefectures in Japan. Collected surface water, sediment, and sand samples were used to extract microplastics and evaluate the seasonal variation and occurrence of microplastics (**Fig. 2**). Visible plastic samples (> 5 mm in size) were used to extract biofilms and characterize their properties using isotopic and thermogravimetric approaches. The extraction of biofilms from plastic surfaces was conducted with a simple extraction technique, using an ultrasonic bath, disposable plastic syringes, and a freeze dryer (**Fig. 3;** Battulga et al., 2022a). Then extracted biofilm samples were prepared for the measurements of ^{137}Cs, ^{13}C, and ^{15}N.

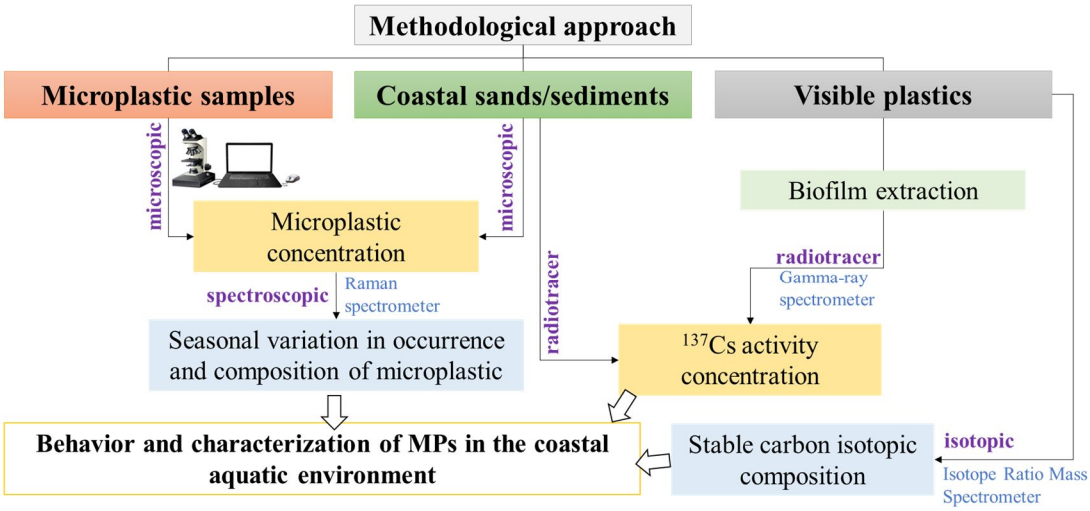

Fig. 2 Methodological approach of the current study

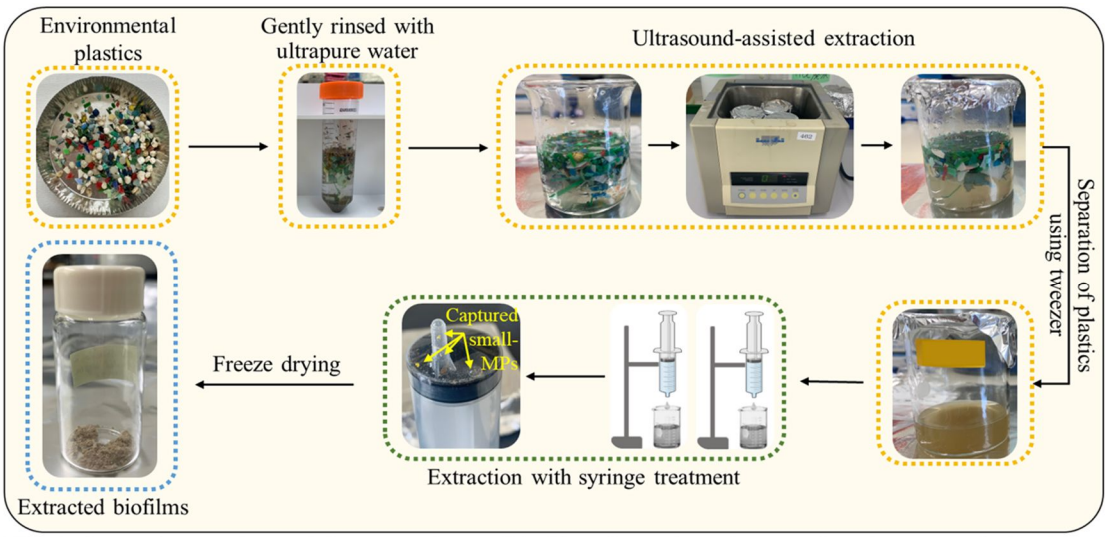

Fig. 3 General procedure of the extraction of biofilms from the mixture of environmental plastics.

4．研究成果

The seasonal patterns in the MP concentration differed between the two sites, suggesting the importance of the various environmental factors (i.e., microbial colonization, changes in density through biofilm formation, degradation, and fragmentation of microplastics) to influence the occurrence of microplastics (**Fig. 4**).

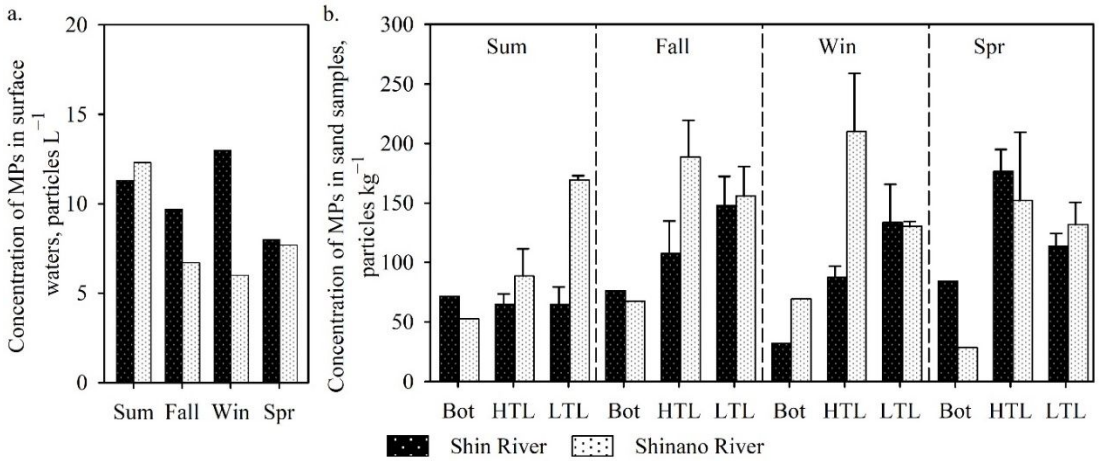

Fig. 4 Microplastic concentrations in surface waters (a) and bottom sediment and coastal sand samples (b) from the Shin and Shinano River sites. The results represent seasonal (Sum, summer; Fall, fall; Win, winter; Spr, spring) data for bottom sediments (Bot) and coastal sands in the high tide line (HTL) and low tide line (LTL).

Furthermore, the ocean circulation system may have an impact on the distribution pattern of plastic particles. The movement of microplastics in the coastal environment can be influenced by dynamic forces, which include ocean current systems, resulting in hotspot distribution in the coastal areas.

Using the newly developed extraction method (**Fig. 3**), the activity concentration of ^{137}Cs in plastic-associated biofilms was detected in several river mouths in Japan, located approximately 150 km from the Fukushima Daiichi Nuclear Power Plant (FDNPP). This observation demonstrates that environmental plastics serve as a carrier for ^{137}Cs in the coastal river environment and are mediated by developed biofilms.

Stable isotopic signatures (**Fig. 5**) demonstrated that plastic-associated biofilms may have an impact on the biogeochemical cycles of organic matter in linking terrestrial and aquatic ecosystems (Battulga et al., 2022a). Despite the particular attention and effort that have been directed at the extraction and evaluation of biofilms from the environmental plastics in this study, further research will be required to address the analysis of nanoplastics in the plastic-associated biofilms and considerable advances for the separation technique for removal of nanoplastics from the environmental samples.

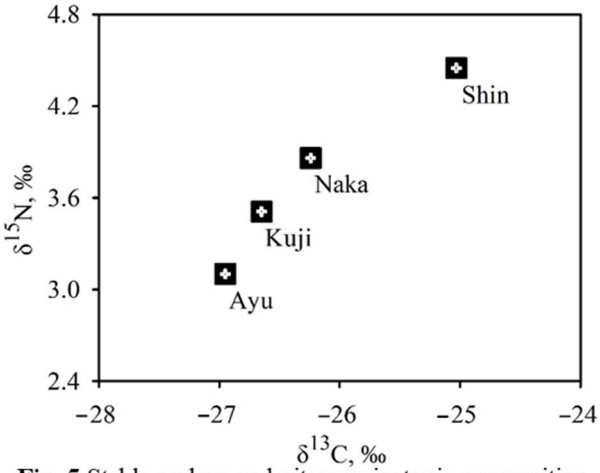

Fig. 5 Stable carbon and nitrogen isotopic composition in biofilm samples extracted with from the environmental plastics.

Further evaluation considering the microbial communities in the Plastisphere is needed to demonstrate the biofilm-associated ecological processes on plastics which would advance our understanding of plastic-altered biogeochemical cycling in the aquatic environment.

References:

Battulga, B., Atarashi-andoh, M., Nakanishi, T., Koarashi, J., 2022a. A new approach to extracting biofilm from environmental plastics using ultrasound-assisted syringe treatment for isotopic analyses. Sci. Total Environ. 849, 157758. https://doi.org/10.1016/j.scitotenv.2022.157758

Battulga, B., Kawahigashi, M., Oyuntsetseg, B., 2022b. Characterization of biofilms formed on polystyrene microplastics (PS-MPs) on the shore of the Tuul River, Mongolia. Environ. Res. 212, 113329. https://doi.org/10.1016/j.envres.2022.113329

Birch, Q.T., Potter, P.M., Pinto, P.X., Dionysiou, D.D., Al-Abed, S.R., 2021. Isotope ratio mass spectrometry and spectroscopic techniques for microplastics characterization. Talanta 224, 121743. https://doi.org/10.1016/j.talanta.2020.121743

Thompson, R.C., Olson, Y., Mitchell, R.P., Davis, A., Rowland, S.J., John, A.W.G., McGonigle, D., Russell, A.E., 2004. Lost at Sea: Where Is All the Plastic? Science (80-.). 304, 838. https://doi.org/10.1126/science.1094559

５．主な発表論文等

〔雑誌論文〕　計1件（うち査読付論文　0件／うち国際共著　0件／うちオープンアクセス　0件）

１．著者名 Battulga Batdulam、Atarashi-Andoh Mariko、Nakanishi Takahiro、Koarashi Jun	４．巻 849
２．論文標題 A new approach to extracting biofilm from environmental plastics using ultrasound-assisted syringe treatment for isotopic analyses	５．発行年 2022年
３．雑誌名 Science of The Total Environment	６．最初と最後の頁 157758～157758
掲載論文のDOI（デジタルオブジェクト識別子） 10.1016/j.scitotenv.2022.157758	査読の有無 無
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 －

〔学会発表〕　計6件（うち招待講演　0件／うち国際学会　4件）

１．発表者名 Batdulam Battulga, Mariko Atarashi-Andoh, Jun Koarashi
２．発表標題 A new extraction method to obtain organic coatings from environmental plastics
３．学会等名 7th International Symposium on Strategies for Sustainability in Food Production, Agriculture, and Environment 2021 (ISFAE 2021 NIIGATA)
４．発表年 2021年

１．発表者名 Batdulam Battulga, Mariko Atarashi-Andoh, Jun Koarashi
２．発表標題 A new approach for biofilm extraction from environmental plastics using ultrasound-assisted syringe treatment for isotopic analyses
３．学会等名 European Geoscience Union (EGU) General Assembly 2022（国際学会）
４．発表年 2022年

１．発表者名 Batdulam Battulga, Mariko Atarashi-Andoh, Jun Koarashi
２．発表標題 Preliminary study to characterize microplastics in the coastal environment using multiple analytical approaches
３．学会等名 Joint Conference on Environmental Chemicals
４．発表年 2022年

１．発表者名 Batdulam Battulga, Masataka Nakayama, Mariko Atarashi-Andoh, Jun Koarashi
２．発表標題 Characterizations of formed biofilms and microbial communities on microplastics in coastal rivers of Japan
３．学会等名 MICRO 2022: Plastic pollution from macro to nano（国際学会）
４．発表年 2022年

１．発表者名 Batdulam Battulga, Masataka Nakayama, Mariko Atarashi-Andoh, Jun Koarashi
２．発表標題 Microbial life on plastics and its implication on elemental cycling in the aquatic environment
３．学会等名 Japan Geoscience Union Meeting 2023（国際学会）
４．発表年 2023年

１．発表者名 Batdulam Battulga, Takahiro Nakanishi, Mariko Atarashi-Andoh, Jun Koarashi
２．発表標題 Characterizing the plastic-associated biofilms by a multi-isotope approach: Insight from visible plastics in two contrasting coastal areas of Japan
３．学会等名 Goldschmidt 2023（国際学会）
４．発表年 2023年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

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６．研究組織

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７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関