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Article

Plastic Bottles for Sorting Floating Microplastics in Sediment

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Abstract: Plastic waste's near-permanent pollution of the natural environment is becoming an increasing concern. It is necessary to determine the amount of microplastics (MPs) present in the natural environment and reduce the amount of plastic waste. In this study, the author developed a simple sorting method for MPs in sediment, which can eliminate sediment and does not require filtration, using an apparatus available to the public. This sorting method, called the "bottle method", can shut off floating matter from sediment and be used for sorting and washing at the same time. When the density of the solid sample is lower than the liquid density, the recovery rate is almost 100%, as theoretically predicted. The recovery rate of MPs by the bottle method is comparable to that by the other two methods, i.e., the spoon method and the overflow method, and the sorting time is reduced by approximately half. As for the dilution of the liquid by the bottle method, the ratio of actual concentration to theoretical concentration is in the range 0.86 to 1.36, and the dilution and washing of the liquid proceeds as theoretically predicted.

Keywords: microplastic; sediment; flotation; plastic bottle; density

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1. Introduction

Since the beginning of the industrial production of plastics in the early 20th century, the production of plastics has been increasing. The global production of plastics in 2015 exceeded 300 million metric tons [1]. Polyethylene (PE) [2], polypropylene (PP) [2], polyvinyl chloride (PVC), polyurethane (PUR), polystyrene (PS), and polyethylene terephthalate (PET) are the six major types of plastic [3]. The cumulative production of plastics is estimated to reach 33 billion metric tons by 2050 [4]. This massive production has resulted in global plastic waste pollution [5]; in 2010 alone, it was estimated that between 4 and 12 million metric tons of plastic waste generated on land entered the marine environment [6], and this amount is expected to increase to a cumulative 12 billion metric tons by 2050 [7]. Cole et al. (2011) stated that plastic is a significant component of marine litter [8]. As commonly used plastics are not biodegradable, they accumulate in landfills and the natural environment without decomposing [9]. Therefore, plastic waste's near-permanent pollution of the natural environment is becoming an increasing concern.

The UV rays of sunlight promote the degradation of plastic waste into small fragments called microplastics (MPs) [10]. MPs have been found in major oceans and coastal areas [9]. MPs have the potential to accumulate organic pollutants such as carcinogenic polychlorinated biphenyls (PCBs) [11,12], polycyclic aromatic hydrocarbons (PAHs), and others [13,14], which eventually enter the marine food web [15]. Polybrominated diphenyl ethers were detected in plastic fragments found in the stomachs and abdominal adipose tissues of oceanic seabirds [16]. MP is a general term for plastic particles smaller than 5 mm [9,17]. On the other hand, particles smaller than 1 µm are called nanoplastics [18]. Although MPs are often detected in the environment, little is known about their risks. The identification of MPs in the environment is complex, and no standardized methods are available [19]. The primary sources of MPs are generally the fragmentation of large plastics and the wearing of products, but the fragmentation rate under natural conditions is unknown [20]. These problems and uncertainties hinder exposure and risk assessment [21].

Thus, it is difficult to assess the risks posed by MPs. However, given the potential for contaminants to accumulate in MPs, it is necessary to determine the amount of MPs present in the natural environment and reduce the amount of plastic waste.

In September 2015, the United Nations General Assembly adopted 17 Sustainable Development Goals (SDGs) to address the planet's complex challenges. Sustainable Development Goal 14 (SDG 14) aims to "conserve and sustainably use the oceans, seas, and marine resources for sustainable development." Goal 14.1 of SDG 14 is to prevent and significantly reduce marine pollution of all kinds. One indicator is plastic debris density (14.1.1b). The EU published "*A European Strategy for Plastics in a Circular Economy*" in January 2018, followed by new recycling targets and proposed regulations for single-use plastics (DIRECTIVE OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL on the reduction of the impact of certain plastic products on the environment) in May 2018. In this way, measures to prevent marine pollution and reduce the use of plastics are being taken worldwide. As these measures take effect, the amount of MPs in the environment will probably decrease in the long term, although it may be necessary to collect existing plastic wastes.

MPs in marine sediment such as sand have been intensively investigated [22–27]. However, suppose we are to test the hypothesis that MPs will decrease in the long term as plastic use is reduced; in that case, we need to develop a simple survey method that can be implemented in high schools and increase MP data accumulation rate, rather than relying on costly survey results from professional researchers.

To analyze MPs, it is necessary to sort MPs from marine sediment. MPs can be picked up visually or using the characteristic that MPs float relatively easily in liquid (flotation medium).

Concentrated saline solution is added to marine sediment, and the mixture is stirred. The marine sediment divides into floating matter such as MPs and sediment such as sand. The MPs can be obtained by decanting floating matter and liquid, filtering them through a sieve or filter paper, and washing them. Many researchers have used this method [28–32]. The targeted floating matter could be collected with a spoon, but no studies using this method were found.

When the MPs are floating in liquid in the container and more liquid is added, the MPs flow out of the container. In this overflow method, a cylindrical container is often used. Claessens et al. (2013) used a 1.7-m-high PVC cylindrical container with an outlet on top to facilitate the collection of floating matter by overflow [33]. The same apparatus was used by Zhu (2015) [34]. Vermeiren et al. (2020) reduced the container height to 30 cm [35]. Hengstmann et al. (2018) used a 1.1-m-high glass cylindrical vessel to avoid plastic contamination from the apparatus [36]. Kedzierski et al. (2016, 2017) investigated the relationship between rising liquid speed and recovery rate [37,38]. Nuelle et al. (2014) placed a small glass beaker inside a large glass beaker, added liquid and sediment to the small beaker to induce the flotation of MPs, and further added liquid to cause overflow MPs into the outer beaker for collection [39]. The MPs collected are washed by filtration.

The essence of sorting MPs from sediment is that the MPs and the sediment are separated into two fractions. However, even if separation is possible, the remixing of the MPs and the sediment makes the sorting process difficult. Whether by decantation or overflow, the resurfacing of the sediment interferes with the operation. If the sediment disappears during the sorting process, the MPs can be taken out from the sorting container in a simple way. In the container, the MPs move to the liquid surface, and the sediment moves to the lower layer and accumulates, thereby producing a partition in the middle. Imhof et al. (2012) developed a metal cone sorter (Munich Plastic Sediment Separator, MPSS) [40] in which a ball valve is installed in the container to separate the floating matter from the sediment in the middle of the liquid. However, the commercialized separator is made of aluminum and weighs 22 kg, making it difficult to carry. It also needs to be disassembled and washed after each use [41]. A similar sorting apparatus was used by Knutsen et al. (2020) [42]. Coppock et al. (2017) installed a ball valve in a cylindrical

container made of PVC [43], which weighs only 1.5 kg and is portable. However, because this container has a valve, it must be disassembled and washed. Nakajima et al. (2019) developed a sorting apparatus with a partition plate made of glass (JAMSS) [44]. It is palm-sized and easy to carry, and its simple structure makes it easy to wash. The collected MPs are washed by filtration.

As described above, the sorting apparatuses for MPs have been vastly improved. However, those apparatuses were specially designed for this purpose. In addition, the MPs are washed by filtration through a fine sieve or filter paper as a processing procedure. The need for a specialized apparatus and the filtration step hinders MP research by nonprofessional researchers. By doing away with the need for a specialized apparatus and the filtration step, it would be possible to involve high school students in MP research, and the accumulation of MP data accelerated.

In this study, the author developed a simple sorting method for MPs in sediment, which can eliminate the sediment and does not require filtration, using an apparatus that is available to the public. This method, called the "bottle method", can shut off floating matter from sediment and be used for sorting and washing at the same time. The purpose of this study is to show that the bottle method is more effective than other sorting methods, i.e., the spoon method and the overflow method, and the following points are investigated:

(1) If MP sorting proceeds as theoretically predicted in the three sorting methods;
(2) If the recovery efficiency of MPs by the bottle method is equivalent to that of the other two methods;
(3) If washing of liquid by the bottle method proceeds as theoretically predicted.

2. Materials and Methods

2.1. Experimental Equipment

Commercially available scissors, nippers, or cutters were used to shred plastic samples. Stainless steel sieves (SANPO) with 1 mm and 4.75 mm mesh openings were used to adjust particle size distribution. For density measurements, a 50 mL pycnometer, a plastic bucket, a thermometer (AD-5625, A&D), a water bath (HWA-50D, AS ONE), an electronic balance (ATY124, Shimadzu), and a water purifier (RFP841AA, ADVANTEC) were used. For the sorting experiment, a 500 mL glass beaker, a 500 mL plastic (PET) beverage bottle, a stainless steel spoon, a stainless steel tray, a plastic bucket, and a dryer (DRD420DA, ADVANTEC) were used. A 500 mL plastic beverage bottle, a silicone tube, and an electrical conductivity (EC) meter (B-173, HORIBA) were used for the dilution washing experiment.

2.2. Sample

The details of the plastic samples used in this study are shown in Table 1. The plastic materials used in this study were PE, PP, PS, PVC, PET, and phenol-formaldehyde resin (PF). The plastic samples were prepared by buying new materials or collecting disposable containers. All the samples were new and not exposed to sunlight or rubbed. The major categories in global nonfiber plastic production are PE (36%), PP (21%), and PVC (12%), followed by PET, PUR, and PS (<10% each) [7]. These groups account for 92% of plastic production. Approximately 42% of nonfiber plastics are used for packaging, mainly PE, PP, and PET [7].

Table 1. Details of plastic samples. For example, SB is an abbreviation for shopping bag.

		Description	Hardness *1	Shape *2	Density ρ_s (g/cm^3) *3		
					Mean	SE	Literature Value *4
PE	SB	Shopping bag	F	S	0.908	0.016	0.91–0.97
PE	GV	Disposable gloves	F	S	0.871	0.011	
PE	RP	Rope	F	R	0.754	0.004	
PE	PB	Polybottle	S	M	0.934	0.005	
PE	FB	Freezer bag	F	S	0.919	0.006	
PP	BC	Cap of PET-BB	S	M	0.925	0.005	0.90–0.91
PP	OP	OP *5 bag	F	S	0.888	0.009	
PP	BD	Board	S	S	0.867	0.003	
PP	CP	Clothespin	S	M	0.905	0.003	
PP	RP	Rope	F	R	0.486	0.010	
PS	EP	Expanded polystyrene	S	M	0.018	0.001	1.04–1.07
PS	BD	Board	S	S	1.084	0.007	
PS	LB	Label of PET-BB	F	S	1.031	0.007	
PS	DC	Compact disk case	S	M	1.054	0.005	
PS	FT	Food tray	S	S	0.981	0.007	
PVC	PI	Pipe	S	M	1.424	0.005	1.16–1.45
PVC	BD	Board	S	S	1.333	0.010	
PVC	CP	Corrugated plate	S	S	1.375	0.007	
PVC	AS	Antislip sheet	F	S *6	0.884	0.029	
PVC	TC	Table cloth	F	S	1.305	0.015	
PET	BB	Beverage bottle	S	S	1.378	0.006	1.38–1.39
PET	EG	Egg pack	S	S	1.315	0.017	
PET	LF	Lumirror® film	S	S	1.390	0.007	
PET	FC	Fruit container	S	S	1.336	0.017	
PC	CD	Compact disk	S	M	1.163	0.005	1.2
PC	SG	Safety glasses	S	M	1.166	0.007	
PC	BD	Board	S	S	1.166	0.006	
PF	KP	Knob of pot lid	S	M	1.469	0.016	1.25–1.32

*1: Flexible/Solid, *2: Sheet/Rope/Mass, *3: n = 6, *4: Handbook of Chemistry [45], *5: Oriented Polypropylene, *6: Net.

Only table cloth (PVC-TC) has a heterogeneous appearance. Although it is made entirely of PVC, it has white lace attached to the back of a transparent sheet. The other plastic samples are homogeneous in appearance. Only rope (PE-RP, PP-RP), expanded polystyrene (PS-EP), antislip sheet (PVC-AS), and PVC-TC are not dense and have voids. All other plastic samples are dense materials.

The hardness and shape of the plastic materials are shown in Table 1. Each material was cut into 10 cm square pieces (10 cm long for rope), and the material was classified as flexible (F) if it hung down when the edge was held, and solid (S) if it retained its shape. The shapes were classified as sheet (S, less than 0.5 mm thick), rope (R), and mass (M, more than 1 mm thick).

The plastic samples were shredded with scissors, nippers, or cutters and sieved through a stainless steel sieve to particle sizes of 1 mm to 4.75 mm (hereinafter referred to as MPs).

Sand (TOYOURA silica sand) was used for the density measurement.

Tap water, seawater (Nagasaki City, 32°82′14″ N, 129°76′14″ E, sampled on 4 September 2020), and calcium chloride (first grade, Wako Pure Chemical Industries, Ltd., Osaka, Japan) saturated solution (hereinafter referred to as SCC solution) were used as liquid samples.

2.3. Density

The density was measured as the fundamental property of the sample.

2.3.1. Liquid Sample

The liquid sample's temperature T (°C) was varied from 5 to 40 °C using a refrigerator or a water bath. The density $\rho_l(T)$ (g/cm^3) of the liquid sample was measured using a pycnometer (n = 6). The calculation formula follows:

$$\rho_l(T) = \{(m_l - m_p)\,\rho_w(T)\}/(m_a - m_p) \quad (1)$$

where m_l: mass of pycnometer filled with the liquid sample at T (°C) (g), m_p: mass of pycnometer (g), m_a: mass of pycnometer filled with pure water at T (°C) (g), and $\rho_w(T)$: density of pure water at T (°C) (g/cm^3).

2.3.2. Solid Sample

The density ρ_s (g/cm^3) of the solid samples (MPs and sand) was measured using a pycnometer (n = 6). Pure water was used as the liquid phase. The calculation equation follows:

$$\rho_s = (m_s\,\rho_w(T))/\{m_s + (m_a - m_b)\} \quad (2)$$

where m_s: mass of dry solid sample (g), m_a: mass of pycnometer filled with pure water at T (°C) (g), m_b: mass of pycnometer filled with pure water and sample at T (°C) (g), and $\rho_w(T)$: density of pure water at T (°C) (g/cm^3).

A solid sample with a density lower than that of pure water floats to the mouth of the pycnometer, making it difficult to use a stopper (Figure 1a). If the sample is sufficiently large, the stopper can be placed in the pycnometer mouth after introducing the sample, but air bubbles enter if this process takes too long. Air bubbles are a cause of error in density measurement. In addition, small pieces of the sample are caught between the stopper and the pycnometer mouth, making it impossible to use the stopper. This problem occurs because the solid sample floats on the liquid. A net was used to hold the solid sample in the pycnometer, but this was a cumbersome task. If the pycnometer was turned upside down, the problem of solid sample flotation is solved; however, the liquid leaked out because the outside is the gas phase. To solve this problem, the pycnometer was turned upside down in the liquid phase to prevent leakage (Figure 1b).

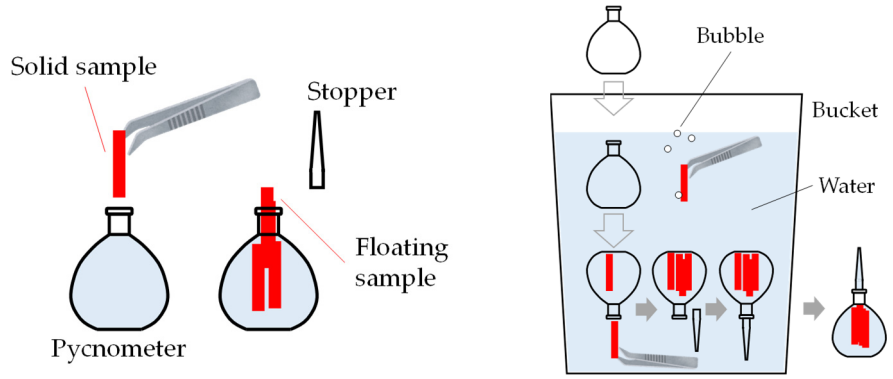


Figure 1. (**a**) Density measurement by the pycnometer method for a solid sample with a density lower than that of pure water. The solid sample floats, and the pycnometer cannot be capped. (**b**) Density measurement by turning the pycnometer upside down in the water.

The procedure follows. Fill a bucket with water. Place a pycnometer in the bucket, fill it with water, and turn it upside down. Shake a solid sample in the water to remove air bubbles, and float it under the pycnometer. Place the stopper in the water, fill it with water, and put it into the mouth of the pycnometer. Turn the pycnometer upside down again and remove it from the bucket. This method has other advantages. It is challenging to degas the

pycnometer with a pump after placing the solid sample in the pycnometer. Still, air bubbles can be easily removed by shaking the solid sample in the bucket of water. Degassing in this manner is easy even for samples with a density that is higher than that of water and in which the problem of flotation does not occur. It is not necessary to turn the pycnometer upside down in this case. It is possible to remove the air bubbles in the bucket of water and place a soft sheet of sample in the pycnometer without cutting the sample into small pieces.

2.4. Sorting Test

2.4.1. Overview of the Sorting Test

The mixture of sand and MPs is referred to as P-sand in the following description. This section explains how to recover MPs contained in coastal P-sand by flotation sorting. Because the density of most sand is higher than that of water or seawater, the sand sinks when added into water, whereas MPs in P-sand, with a density that is lower than that of water or seawater, float to the surface (Figure 2). Stirring with a spoon accelerates the separation of sand and MPs. In this way, MPs with low density in P-sand can be quickly sorted by flotation sorting.

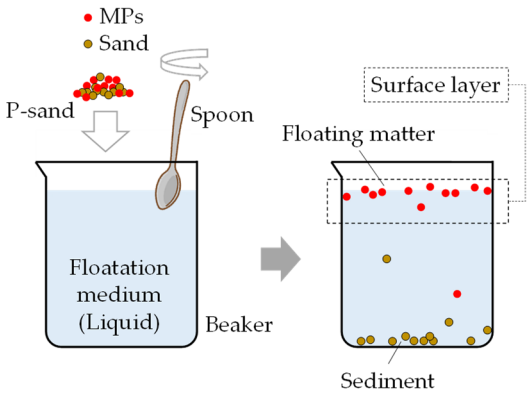


Figure 2. Input of P-sand and floating of MPs with a density that is lower than the liquid.

The recovery rate was measured for the method of scooping floating matter (spoon method) and the method of adding liquid to induce the overflow of floating matter (overflow method). We also measured the recovery rate of the bottle method, which prevents the remixing of floating matter and sinking matter. In this experiment, only MPs were used in the primary experiment, and no sand was used. In addition, filtration was omitted to simplify the sorting process. Figure 3 shows the measurement flow of MP content in P-sand proposed in this study. An initial amount of approximately 0.5 g of MPs was weighed out, added into the flotation medium (liquid), and stirred with a spoon for 30 s. Then, the floating matter was collected into a stainless steel tray. Hereinafter, the liquid in the tray is called the accompanying water. The time between the introduction of MPs into the flotation medium and their collection into the tray is called the required time. Tap water was used as the flotation medium. For the spoon method only, SCC solution (1.45 g/cm^3 at 25 °C) was also used. The recovered matter was dried at 80 °C overnight and weighed to calculate the recovery (n = 6). In other words, the drying process was rapidly completed when there was less accompanying water. The ratio of the amount recovered to the initial amount is the recovery rate (%) (Equation (3)).

$$\text{Recovery rate (\%)} = \text{Amount recovered (g)}/\text{Initial amount (g)} \times 100 \quad (3)$$

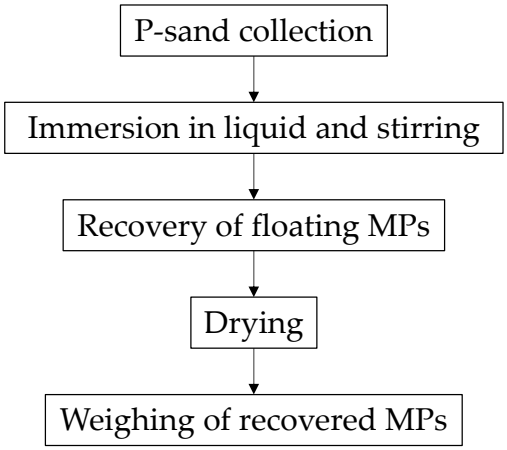


Figure 3. Measurement flow of MP content in P-sand proposed in this study.

Most MPs with a density lower than the liquid will float (Figure 4a). Theoretically, the recovery rate is 100%. Some MPs are left behind, as observed visually. If the plastic material is heterogeneous and has some dense parts, the MPs sinks and are left behind. MPs that adhere to the inner wall of the beaker when stirred and do not float to the surface are treated as sediment. This often happens with MPs in sheet form.

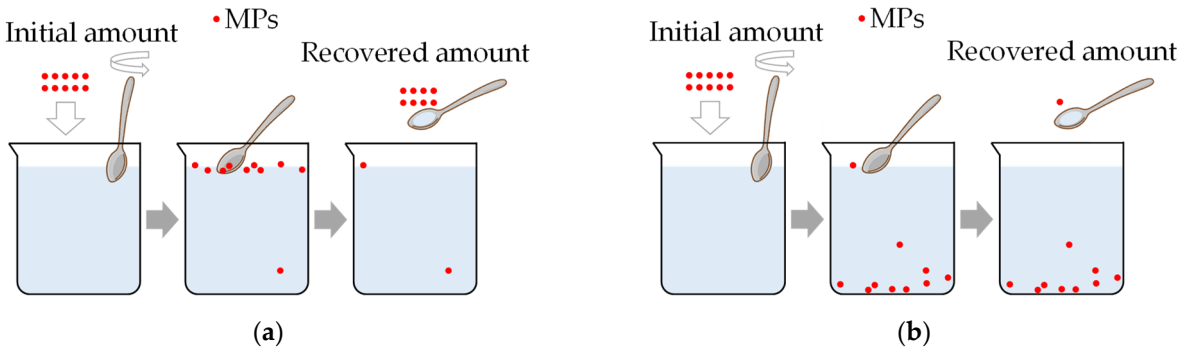


Figure 4. (**a**) Recovery of floating MPs; (**b**) recovery of sinking MPs.

Most MPs with a density higher than the liquid will sink (Figure 4b). Theoretically, the recovery rate is 0%. When there is no floating matter, the surface liquid is collected for 30 s. On the other hand, even MPs with a density higher than the liquid will float if they have a low-density part due to heterogeneity. In addition, MPs that are water-repellent will float on the surface of the liquid and do not sink. In this way, even MPs with a density higher than the liquid do not necessarily have a recovery rate of 0%.

2.4.2. Spoon and Overflow Methods

In the spoon method, the floating matter is scooped out together with the flotation medium from the surface layer using a spoon and transferred to a tray (Figure 5a). The entire beaker used for flotation sorting is placed in a tray in the overflow method. A flotation medium is added to induce overflow of floating matter into the tray (Figure 5b).

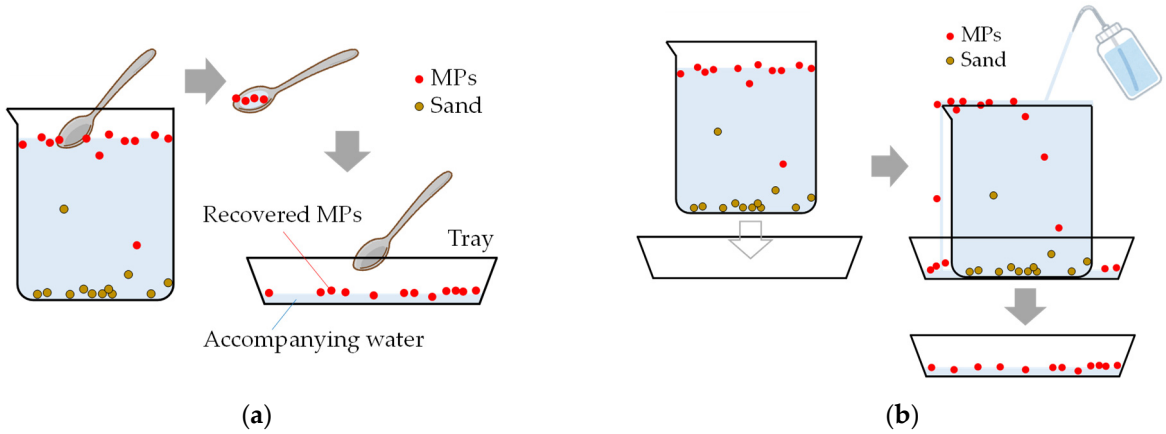


Figure 5. Recovery of MPs: (**a**) spoon method; (**b**) overflow method.

2.4.3. Bottle Method

The spoon method and the overflow method were adopted in the preliminary experiments. One of the issues was the remixing of floating matter and sinking matter. By scooping the surface layer of the liquid with a spoon, the surface layer is disturbed, and floating matter sinks. In the overflow method, when liquid is squeezed out of a wash bottle, the liquid phase is greatly disturbed, and both sinking of floating matter and floating of sediment are likely to occur. In order to avoid this, the liquid should be gently added from the inside of the flotation medium. However, this will reduce recovery efficiency because the MPs that have floated to the edge of the beaker need the force of water to flow out of the beaker.

Therefore, in order to prevent remixing, a method was devised to isolate floating matter from sinking matter. Shut-off methods using valves have also been reported [40,42,43]. The apparatus can be simplified by using a partition plate inserted horizontally in the middle of the liquid phase after separating floating matter from sinking matter. To realize the partition plate, a combination of two cylindrical containers was used (only one of them has a bottom plate) (Figure 6a). However, although it may be possible to prevent water leakage during the sorting process, it is difficult to do so during the insertion of the partition plate. Inserting a closed umbrella or a balloon in the middle of the liquid phase and deploying it inside was considered, but a simple design could not be devised. A similar case is the test of the water retention curve of soil. After filling the stacked rings with soil, water is added and drained, the rings are dismantled, and the soil moisture content is measured. The soil does not spill when the rings are dismantled because the watered soil is compacted. To apply the same method to MP sorting, the liquid can be frozen and cut with a saw. Although this method is cumbersome, freezing and cutting are a promising method to accurately determine the distribution of MPs by height without disturbance.

On the other hand, Nakajima et al. (2019) used a different method to prevent water leakage during partition plate insertion [44]. They made a container having a built-in partition plate instead of inserting it later, and moved the container instead of the partition plate (Figure 6b) [44]. After separation into floating matter and sinking matter, the floating matter can be collected by decantation. If the smooth partition plates are wet, the plates will be in close contact with each other, and water leakage will be prevented. The author was impressed by this new method.

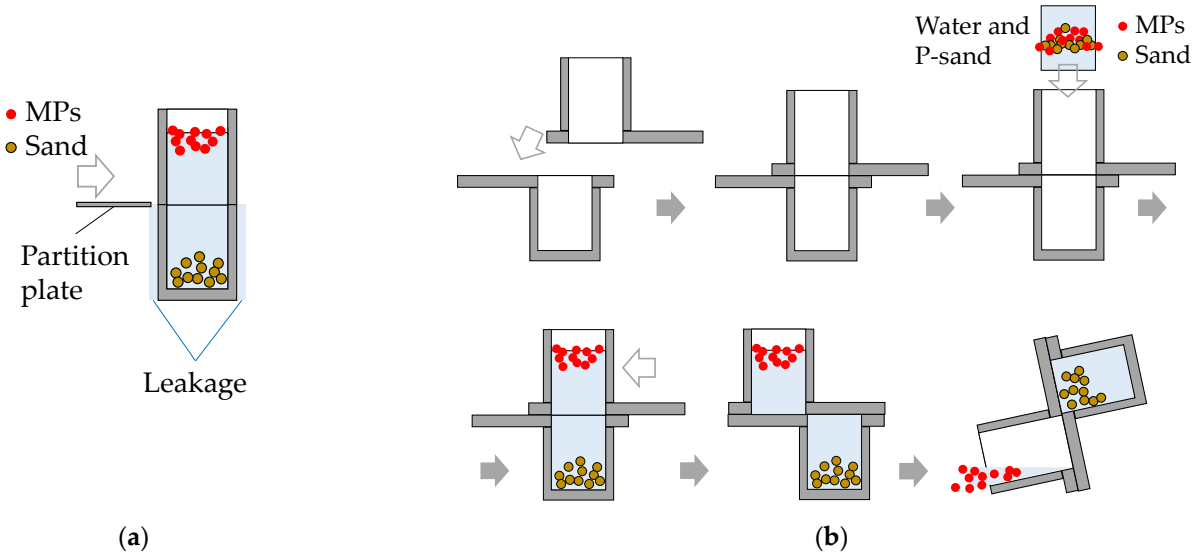


Figure 6. Insertion of partition plate to prevent remixing of floating matter and sinking matter: (**a**) insertion of partition plate in the gas phase causes water leakage; (**b**) method of Nakajima et al. (2019) [44].

On the other hand, it was observed that in the density measurement of a solid sample, the pycnometer is turned upside down to prevent a low-density solid sample from floating to the mouth of the pycnometer. However, the liquid leaks when carried out into the air. On the other hand, there is no leakage if the pycnometer is in water (Figure 1b). Therefore, if the partition plate is inserted into a container immersed in the liquid phase, no liquid leaks from the container (Figure 7a). Moreover, the partition plate itself is not always necessary in this method. For example, if a pipe with an inner diameter of 20 cm and a length of approximately 1 m (open at the top and bottom) is half exposed to air and half immersed in seawater, a large amount of sand can be injected into it, and the MPs contained in the sand can be concentrated. The pipe can be floated or placed on the bottom of the sea. Because sand is removed from the bottom of the pipe, the partition plate is not necessary.

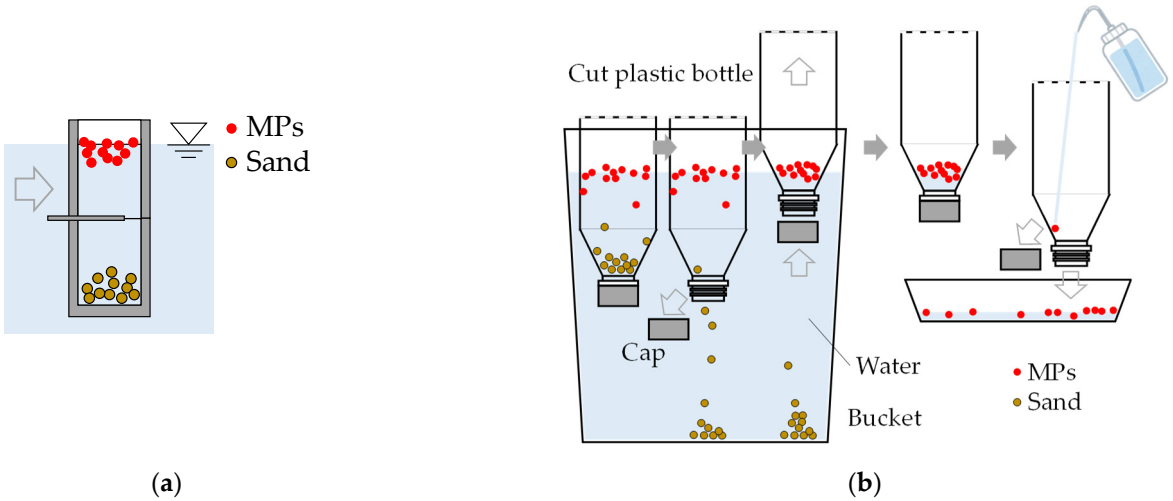


Figure 7. Method of immersing the sorting container in the liquid phase: (**a**) water leakage prevention by inserting a partition plate into the sorting container in the liquid phase; (**b**) MP sorting using the bottle method.

However, the partition plate is convenient for the final collection of floating MPs. Again, it was observed that a stopper was put into the mouth of a pycnometer to hold

the solid sample inside when measuring the density of the solid sample (Figure 1b), and a cap could be used instead of a plate. Special laboratory equipment was expected to be necessary, but a plastic beverage bottle that is available to the public was used instead. Cutting off the bottle bottom made adding P-sand easier and for performing additional operations. In this way, the bottle method was devised (Figure 7b).

Hereinafter, a plastic beverage bottle with a cut bottom is referred to as the bottle. The procedure follows. Stir water and P-sand in the bottle and immerse the bottle halfway in a bucket of water. After P-sand separates into floating matter and sinking matter, remove the cap and drain the sinking matter from the bottle. By placing a beaker under the bottle, the sinking matter can be collected. Pull the bottle up to the surface to remove the liquid in the bottle. Take the bottle out from the bucket with the cap back on, remove the cap while keeping the bottle above the tray, and transfer floating matter and accompanying water into the tray. Because floating matter tends to adhere to the bottle's inner wall, it is necessary to rinse it off with water.

2.5. Dilution and Washing Test of Flotation Medium by Bottle Method

A sorting method that does not require washing by filtration is used in this study. Solid and liquid phases can be washed by repeated drainage and addition of pure water while the bottle is lifted and recapped (Figure 7b). When the liquid (flotation medium) becomes equivalent to pure water, it can be dried and evaporated, and washing by filtration can be omitted. If volume $(x-1)/x$ of the liquid with concentration C_0 of a certain component is eliminated and the same amount of pure water is added, the concentration becomes C_0/x. When this elimination and addition is repeated n times, the concentration C_n is as follows:

$$\begin{aligned} C_n &= C_0/(x^n) \\ C_n/C_0 &= 1/(x^n) \end{aligned} \quad (4)$$

This dilution and washing procedure was checked to see whether it proceeded according to theory. The linearity between concentration and EC was confirmed by measuring the electrical conductivity (EC) of several calcium chloride solutions with different concentrations. Next, a 5% calcium chloride solution was prepared and added until the volume reached approximately half of the bottle volume. Then, the drainage and addition of pure water were repeated. After adding pure water, the mixture was stirred with a spoon and EC was measured. In Figure 8, MPs and sand are shown, but only the MPs were used in the experiment. To facilitate the drainage, a hole was made on one side of the bottle with a soldering iron and a silicone tube was inserted to make a drain. Pulling the silicone tube makes it thinner and loosening makes it thicker, so it easily fits the drain. Neither floating matter nor sinking matter flow out if the drain is set at the middle height of the bottle. Even if MPs floating in an SCC solution sink due to dilution of the solution, they will not flow out of the middle drain. For certainty, the actual outflow amount needs to be tested.

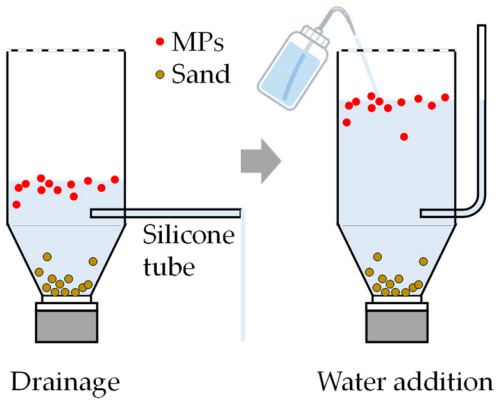
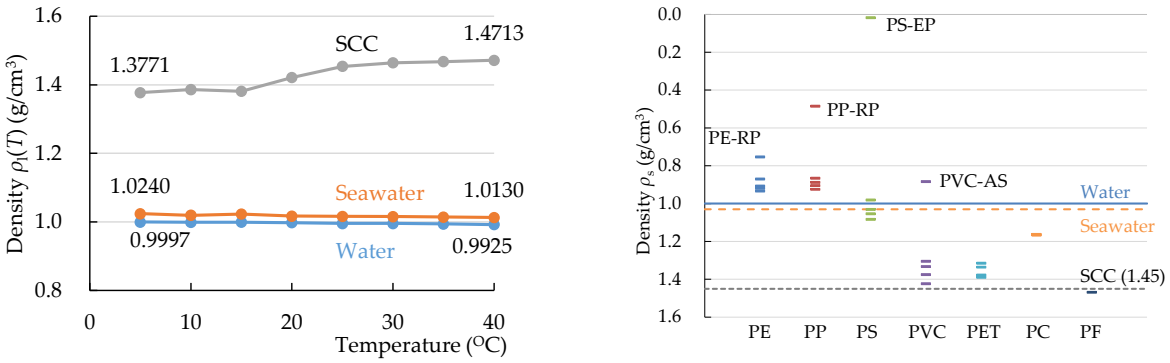


Figure 8. Dilution and washing of flotation medium by bottle method.

3. Results

3.1. Density

The density of liquid samples $\rho_l(T)$ is shown in Figure 9a. The $\rho_l(T)$ of tap water was approximately 0.99 to 1.00 g/cm^3, and that of seawater was approximately 1.01 to 1.02 g/cm^3. The $\rho_l(T)$ decreased with increasing temperature, and $\rho_l(40°C)/\rho_l(5°C)$ was 0.99. The $\rho_l(T)$ of the SCC solution ranged from 1.38 to 1.47 g/cm^3. The density increased with increasing temperature, and $\rho_l(40°C)/\rho_l(5°C)$ was 1.07. In particular, the $\rho_l(T)$ increased from 15 °C and was 1.45 g/cm^3 at 25 °C, which is almost the same as that of 1.47 g/cm^3 at 40 °C.



(**a**) (**b**)

Figure 9. (**a**) Relationship between temperature and density of liquid samples $\rho_l(T)$ (n = 6, mean). (**b**) Densities of solid samples ρ_s (n = 6, mean).

The density of solid samples ρ_s is shown in Table 1 and Figure 9b. In the figure, the densities of the liquid samples are added. The vertical axis is inverted to make it easier to imagine whether the solid samples float or sink in the liquid samples. However, whether a solid sample floats or not was confirmed by experiments. All PE and PP samples had lower densities than tap water and seawater; some PS and PVC samples had lower densities than tap water and seawater, and all samples had lower densities than SCC; the densities of PET and PC samples were between those of water (tap water and seawater) and SCC. PF had a higher density than the SCC solution. The density range of PE, PP, PS, and PVC samples was large because the foaming process reduced the densities of these samples. In other words, the densities measured in this study were not the true densities (outer and inner pores saturated with water) but the envelope densities (outer pores saturated with water and air-filled inner pores) and the bulk densities (air-filled outer and inner pores). The density of sand is 2.620 ± 0.008 g/cm^3 (n = 6), which is larger than that of SCC.

3.2. Recovery Efficiency of Each Sorting Method

Using tap water as the flotation medium, the recovery rate, required time, and amount of accompanying water for each sorting method were determined, and the results are shown in Table 2. PVC-PI and PF-KP were too hard to be shredded into the amount required for the experiment. The recovery rates mainly were around 100% or 0%. The required time varied from 16.2 to 205 s for the spoon and overflow methods. On the other hand, the required time for the bottle method ranged from 22.8 to 63 s, which is a relatively small range.

Table 2. The recovery efficiency of each sorting method (recovery rate, required time, amount of accompanying water) (tap water, n = 6).

		Spoon						**Overflow**						**Bottle**					
		Recovery %		**Time s**		**Acc. Water mL**		**Recovery %**		**Time s**		**Acc. Water mL**		**Recovery %**		**Time s**		**Acc. Water mL**	
		Mean	**SE**	**Mean**	**SE**	**Mean**	**SE**	**Mean**	**SE**	**Mean**	**SE**	**Mean**	**SE**	**Mean**	**SE**	**Mean**	**SE**	**Mean**	**SE**
PE	SB	99.8	0.7	205.3	9.6	161.9	7.6	96.2	0.3	191.7	3.7	239.5	14.1	97.5	0.4	57.3	2.8	90.9	3.0
PE	GV	99.5	0.2	111.2	2.7	20.2	1.8	99.4	0.2	167.5	6.2	206.1	10.8	100.3	0.2	53.0	2.0	78.4	3.3
PE	RP	99.6	0.3	57.2	1.1	6.4	0.3	99.9	0.1	49.3	3.4	136.6	9.8	99.9	0.1	30.3	1.0	81.8	8.7
PE	PB	99.9	0.1	53.2	1.9	3.3	0.2	99.9	0.2	63.0	5.7	33.0	1.6	100.0	0.0	44.3	1.9	35.1	1.5
PE	FB	99.3	0.3	98.0	3.4	18.8	0.9	99.8	0.1	80.0	13.8	125.4	4.3	99.7	0.2	35.0	0.6	65.5	5.2
PP	BC	99.1	0.6	18.8	0.9	1.9	0.1	100.1	0.1	28.0	2.7	25.2	0.6	100.0	0.0	30.2	1.4	31.7	1.0
PP	OP	99.0	0.3	201.8	11.2	168.8	9.9	98.5	0.3	148.2	14.2	118.4	8.7	96.7	1.2	48.8	2.7	93.7	3.8
PP	BD	99.9	0.1	24.8	1.0	1.9	0.1	99.9	0.1	23.7	2.2	28.1	2.1	100.0	0.0	35.0	1.3	32.1	1.6
PP	CP	100.0	0.0	52.2	2.3	2.8	0.2	99.0	0.1	53.8	7.3	32.2	2.1	99.9	0.0	62.8	4.3	36.1	2.7
PP	RP	94.7	2.6	60.0	0.0	29.1	1.6	99.9	0.1	49.7	1.6	60.3	3.2	99.8	0.1	18.5	0.6	31.6	2.9
PS	EP	100.8	0.3	75.0	12.6	177.7	6.5	101.0	0.5	110.7	4.7	124.4	5.3	99.8	0.6	50.2	1.3	128.2	11.1
PS	BD	0.6	0.5	30.0	0.0	5.1	1.6	9.6	0.9	32.5	5.2	55.7	5.0	8.7	1.2	31.2	1.0	64.7	5.0
PS	LB	53.4	2.9	101.5	3.8	17.6	0.9	52.9	2.7	46.8	4.1	146.9	14.8	56.2	3.3	44.3	1.8	69.6	4.8
PS	DC	0.0	0.0	30.0	0.0	2.5	0.4	0.0	0.0	30.0	0.0	11.6	1.2	0.0	0.0	26.8	0.9	28.8	1.3
PS	FT	98.1	0.6	67.8	7.3	142.0	9.0	99.2	0.3	52.3	4.3	40.7	4.2	99.5	0.3	30.7	0.6	60.1	6.5
PVC	PI	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
PVC	BD	0.0	0.0	30.0	0.0	6.5	0.2	0.0	0.0	30.0	0.0	47.0	1.9	7.2	2.0	34.2	1.1	47.5	4.5
PVC	CP	0.0	0.0	30.0	0.0	6.4	0.4	0.0	0.0	30.0	0.0	41.7	0.3	0.0	0.0	31.5	1.7	49.0	3.1
PVC	AS	98.6	1.2	53.7	0.9	8.2	2.1	99.4	0.1	49.3	3.6	44.7	3.8	100.0	0.0	35.3	1.3	81.1	8.5
PVC	TC	6.9	0.9	16.2	1.0	1.2	0.3	6.5	0.6	20.7	2.5	39.5	4.0	3.2	1.1	25.7	0.8	22.5	3.0
PET	BB	0.0	0.0	30.0	0.0	6.6	0.2	0.0	0.0	30.0	0.0	43.9	1.8	0.2	0.1	22.8	0.8	26.6	4.0
PET	EG	16.2	1.4	46.7	3.9	4.9	0.4	19.7	1.7	48.7	4.1	95.0	7.8	6.0	0.7	29.7	0.8	46.3	3.2
PET	LF	2.9	1.0	18.2	2.9	2.3	1.4	3.4	1.0	18.3	3.9	24.8	5.8	0.4	0.3	26.2	0.9	45.8	3.4
PET	FC	0.0	0.0	30.0	0.0	7.0	0.3	0.0	0.0	30.0	0.0	50.2	1.3	0.4	0.3	27.2	0.8	40.6	2.1
PC	CD	0.0	0.0	30.0	0.0	8.3	0.3	0.0	0.0	30.0	0.0	39.8	2.0	0.0	0.0	27.3	0.5	36.4	1.8
PC	SG	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
PC	BD	0.0	0.0	30.0	0.0	7.0	0.3	0.0	0.0	30.0	0.0	47.5	1.5	0.0	0.0	27.3	0.7	29.4	5.0
PF	KP	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-

The relationship between the density of solid samples ρ_s and the recovery rate is shown in Figure 10. The results obtained when tap water was used as the flotation medium are described below. When ρ_s was lower than the density of the flotation medium, the recovery rate was almost 100%. On the other hand, when ρ_s was higher than the density of the flotation medium, some recoveries were 0%, whereas others were higher than 5%. In particular, the recovery of PS-LB (labeled PET-BB, 1.031 g/cm^3), which has a slightly higher density than the flotation medium, was approximately 50% to 60%. Thus, even when ρ_s was higher than that of the liquid, not all samples necessarily sank, which is typical for thin sheet-like materials such as PET-EG. Samples expected to float actually floated, but those expected to sink did not do so.

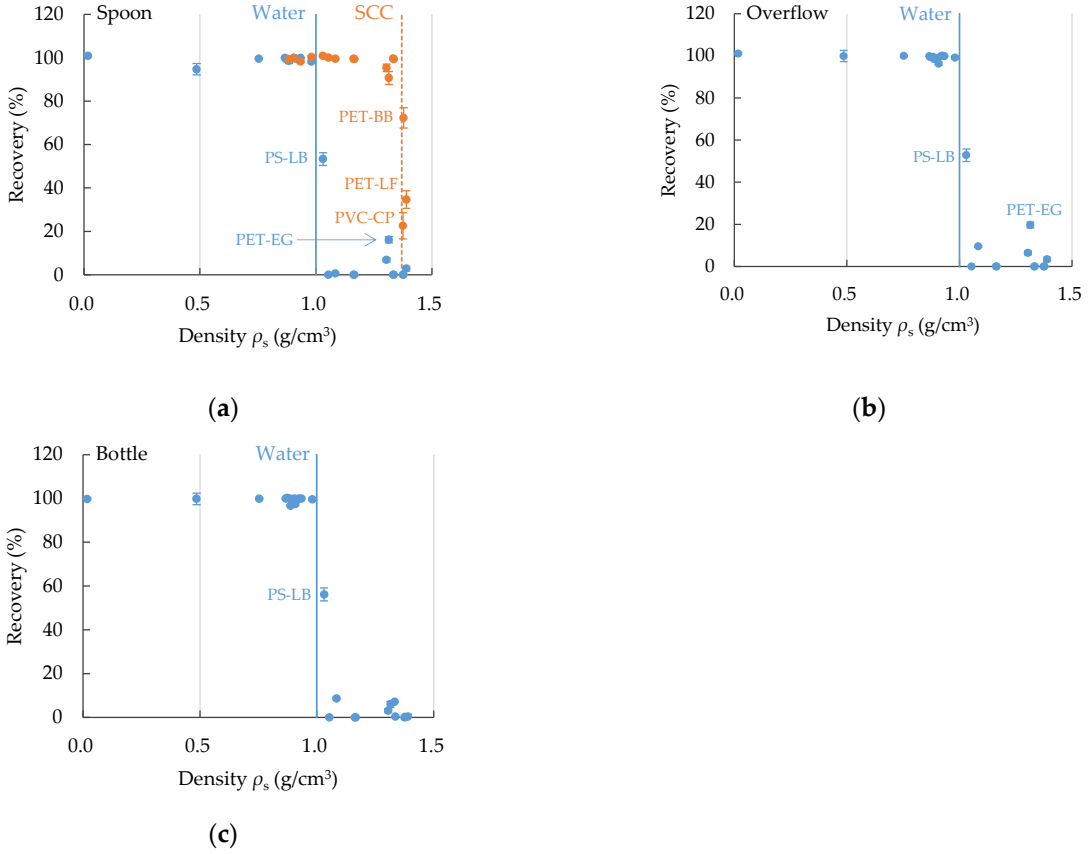


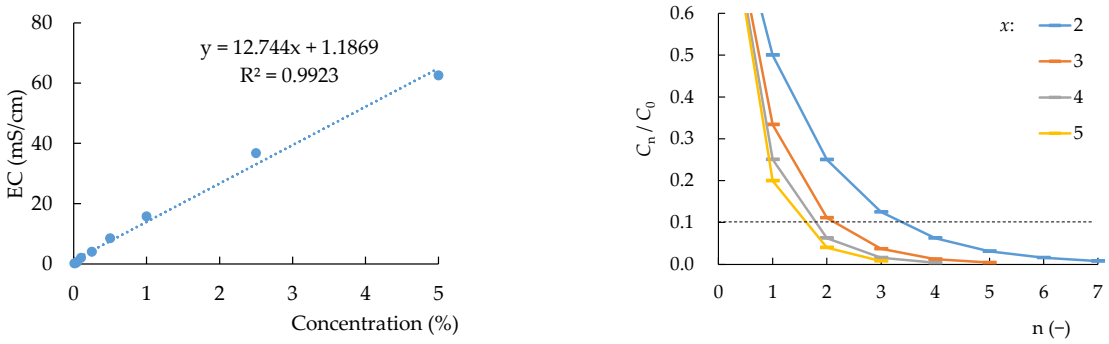
Figure 10. Density ρ_s and recovery rate of solid samples (n = 6, mean ± SE): (**a**) spoon method; (**b**) overflow method; (**c**) bottle method. The flotation media were water and SCC solution for the spoon method only.

In the spoon method, SCC was used for some solid samples (Figure 10a). As in the case of tap water, when ρ_s was lower than the density of SCC, the recovery rate was almost 100%. When ρ_s was slightly higher than that of the flotation medium, the recovery rate was approximately 20% to 70%.

3.3. Dilution and Washing Test of Flotation Medium by Bottle Method

Because there was a linear relationship between calcium chloride concentration and EC (Figure 11a), the calcium chloride concentration was calculated using EC. Using Equation (4), the theoretical relationship between the number of dilutions and the concentration ratio in the bottle method is shown in Figure 11b. To reduce the concentration to 1/10 of the initial concentration, more than four dilutions in 2-fold dilution, more than three dilutions in 3-fold dilution, and more than two dilutions in 4- and 5-fold dilutions were

required. Table 3 shows the ratios of actual concentration to theoretical concentration when actual dilutions were carried out using the bottle method. The concentration ratios were in the range 0.86 to 1.36, and there was no significant difference between the actual and theoretical concentrations.



(**a**) (**b**)

Figure 11. (**a**) Relationship between concentration and electrical conductivity of calcium chloride solution. (**b**) Theoretical relationship between the number of dilutions and concentration ratio in the bottle method.

Table 3. The ratio of actual concentration to theoretical concentration for dilution of flotation medium by the bottle method (n = 1).

n	C_n-Actual/C_n-Theoretical			
	x = 2	3	4	5
0	0.98	0.96	1.05	1.07
1	0.98	1.14	1.08	1.01
2	0.86	0.99	1.36	1.12
3	0.89	0.98	1.25	1.14
4	1.25	1.31	1.10	
5	1.12	1.12		
6	1.21			
7	1.00			

4. Discussion

4.1. Recovery Efficiency of Each Sorting Method

When the density of the solid sample was lower than the liquid density, the recovery rate was almost 100%, as theoretically expected. On the other hand, when the density of the solid sample was higher than the density of the liquid, the sample did not always sink completely, as the theory suggested.

The target MPs, sorting methods, and recovery rates of other researchers follow. Vermeiren et al. (2020) used the overflow method for PP, PE, PVC, and PET [35]. Imhof et al. (2012) sorted PP, PE, PS, PVC, and PET [40], and Coppock et al. (2017) sorted PP and PVC by shutting off the liquid phase with a ball valve [43]. Nakajima et al. (2019) sorted PP, PE, PS, PVC, and PET by shutting off the liquid phase with a partition plate [44]. In all cases, the density of the flotation medium ranged from 1.5 to 1.6 g/cm^3, and the recovery rate was higher than 90%. Similarly, high MP recovery was obtained in this study.

Table 4 shows the ratios of the recovery efficiencies (recovery rate, required time, and amount of accompanying water) of the spoon method and the overflow method to that of the bottle method for solid samples with lower density than tap water. The recovery rates of the three sorting methods were almost the same. Although there was a large variation, the required time for the spoon method and the overflow method was approximately twice that of the bottle method. This means that the bottle method can reduce the required time for sorting. Although the variation was large, the amount of accompanying water for the

spoon method and the overflow method was approximately 0.7 and 1.4 times that for the bottle method.

Table 4. Ratios of recovery efficiencies (recovery rate, required time, and amount of accompanying water) of the spoon method and the overflow method to that of the bottle method (n = 6).

		Recovery			Time			Accompanying Water		
		Spoon	**Overflow**	**Bottle**	**Spoon**	**Overflow**	**Bottle**	**Spoon**	**Overflow**	**Bottle**
PE	SB	1.02	0.99	1.00	3.58	3.34	1.00	1.78	2.63	1.00
PE	GV	0.99	0.99	1.00	2.10	3.16	1.00	0.26	2.63	1.00
PE	RP	1.00	1.00	1.00	1.88	1.63	1.00	0.08	1.67	1.00
PE	PB	1.00	1.00	1.00	1.20	1.42	1.00	0.09	0.94	1.00
PE	FB	1.00	1.00	1.00	2.80	2.29	1.00	0.29	1.92	1.00
PP	BC	0.99	1.00	1.00	0.62	0.93	1.00	0.06	0.80	1.00
PP	OP	1.02	1.02	1.00	4.13	3.03	1.00	1.80	1.26	1.00
PP	BD	1.00	1.00	1.00	0.71	0.68	1.00	0.06	0.88	1.00
PP	CP	1.00	0.99	1.00	0.83	0.86	1.00	0.08	0.89	1.00
PP	RP	0.95	1.00	1.00	3.24	2.68	1.00	0.92	1.91	1.00
PS	EP	1.01	1.01	1.00	1.50	2.21	1.00	1.39	0.97	1.00
PS	FT	0.99	1.00	1.00	2.21	1.71	1.00	2.36	0.68	1.00
PVC	AS	0.99	0.99	1.00	1.52	1.40	1.00	0.10	0.55	1.00
Mean		1.00	1.00	1.00	2.03	1.95	1.00	0.71	1.36	1.00
SD		0.02	0.01	0.00	1.13	0.91	0.00	0.83	0.72	0.00

Among the samples listed in Table 2, those with densities lower than that of tap water (i.e., the samples shown in Table 4) were used to test whether there is a difference in the recovery efficiency among the sorting methods. First, the Friedman test showed no significant difference between any of the groups in the recovery rate and the amount of accompanying water ($p > 0.05$). Still, there was a significant difference in the required time ($p < 0.05$). Next, p-values were obtained by the Wilcoxon signed-rank test, comparing three groups in terms of the required time (n = 13, 3 combinations). The p^*-values after Bonferroni correction were calculated by multiplying the p-values by the number of combinations. There was no significant difference between the spoon and overflow methods ($p^* > 0.05$). Still, there was a significant difference between the spoon method and the bottle method ($p^* = 0.02$), and between the overflow method and the bottle method ($p^* = 0.01$). The effect size d (= combination with short bottle time/all combinations) was 0.77 between the spoon and bottle methods and 0.77 between the overflow and bottle methods.

These results indicate that the bottle method has a similar recovery rate and shorter sorting time than the spoon method and the overflow method. There is no significant difference in the amount of accompanying water.

4.2. Characteristics of Each Sorting Method

The spoon method requires purposeful and subjective work aimed at the target floating matter. Specifically, there is a need to keep chasing the floating target matter with a spoon until the floating target matter is scooped out, and there is a need to judge whether the entire scooping process has been completed. Therefore, the spoon method is considered a precise method. On the other hand, the overflow method can be used relatively without the need to target specific floating matter. However, floating matter in the dead space outside the overflow is difficult to discharge, so it is necessary to judge whether the discharge of floating matter has been completed. In the end, it is necessary to target specific floating matter with water from the wash bottle. In contrast, there is no need to target specific floating matter in the bottle method. The advantage of the bottle method is that it can move the entire floating matter fraction. This is the reason why the required time was reduced. In summary, the partition plate method effectively sorts MPs because it is difficult

to transfer the floating matter to another container, and the floating matter is already in another container when the partition plate is inserted.

The following are the points to be improved. The spoon method should be used with the overflow method to promote the outflow of floating matter. In the overflow method, it is better to have a small opening at the top of the container because it is easier to discharge floating matter if they are grouped together. In the sorter used by Imhof et al. (2012) [40] and Knutsen et al. (2020) [42], the upper part is thinner.

The advantages of the bottle method are summarized as follows. The first advantage is the prevention of remixing. After sinking matter is eliminated and the bottle is recapped, floating matter can be shut off from the sinking matter.

The second advantage is the washing of the contents. Both solid and liquid phases (seawater and heavy liquid) can be washed by repeated drainage and addition of pure water. As shown in Table 3, the dilution and washing of the liquid proceeded as theoretically expected. This method can also be applied to rinsing after washing solids with detergent. When heavy liquid is used in the spoon method or the overflow method, it is necessary to wash the liquid after collecting MPs. On the other hand, the bottle method can be used for sorting and washing simultaneously.

The third advantage is the high recovery efficiency of MPs after sorting. In principle, all the MPs in the bottle are transferred to the tray, unlike the spoon method or the overflow method. As the spoon method and the overflow method collect MPs visually, these methods do not collect particles that are not visible. In other words, there is a possibility of missing transparent or fine particles. However, in the bottle method, the entire MP fraction can be transferred to a tray. Furthermore, it is possible to dry the entire bottle without a tray.

The fourth advantage is the possibility of concentration. After the cap is removed to eliminate sediment and the bottle is recapped, sand can be added again. Whereas the spoon method and the overflow method can only sort MPs until the container is full of sand, the bottle method can process a large amount of sand because the sand is removed. The bottle method can also be applied to the removal of accompanying water generated by the spoon method and the overflow method because the flotation medium can also be removed. In other words, the bottle method can be applied as the subsequent process of the spoon method using buckets, ladles, seawater, and sand on the beach.

The disadvantage of the bottle method is that during the process of transferring MPs to the tray, some MPs adhere to the bottle's inner wall and need to be pushed out by water flow, which results in a large amount of accompanying water.

4.3. Problems, Future Research, and Significance of the Study

Only MPs larger than 1 mm were used. Therefore, there is a need to confirm whether particles finer than 1 mm will not cause problems in the bottle method. In addition to MPs, plant parts such as wood and inorganic materials such as pumice float on the liquid. It is necessary to develop a removal method that does not require visual inspection. Because sand was not used, it is necessary to check whether sand affects floating matter. This study was conducted under ideal conditions. Future research should be conducted using actual marine sediments, degraded plastics, and seawater to simulate sorting operations under actual conditions.

Calcium chloride is one of the major components of seawater, so there is little risk of environmental pollution in coastal sorting operations. MPs can be sorted into two density ranges if SCC is used after tap water as the flotation medium. Furthermore, multistep sorting by density is possible if SCC is diluted. However, as shown in Figure 9b, the minimum density is not determined by the type of plastic material, so it is not possible to sort by plastic material. It may be possible to estimate the ratio of plastic materials that float to the surface of seawater to those that sink.

Because the bottle proposed in this study is made of plastic, debris from the bottle may be mixed into the sample as MPs, which may lead to overestimating the measurement values. Conversely, the electrostatic charge of the plastic bottle may trap fine microplastics,

leading to a smaller estimation. Despite this drawback, the bottle method is recommended because it is simple and requires no special tools or processing. If data on the amount of MPs in the environment can be accumulated by not only professional researchers but also high school students, it will be possible to clarify the long-term trend of MPs.

5. Conclusions

In this study, a simple sorting method for MPs in sediment is developed, which can eliminate sediment and does not require filtration, using an apparatus that is available to the public. This method, called the "bottle method", can shut off floating matter from sediment and be used for sorting and washing at the same time. The results demonstrated that the bottle method is more effective than the spoon and overflow methods. The main findings follow:

(1) When the density of the solid sample was lower than the liquid density, the recovery rate was almost 100%, as theoretically predicted.
(2) The recovery rate of MPs by the bottle method was comparable to that by the other two methods, and the sorting time was reduced by approximately half.
(3) As for the dilution of liquid in the bottle method, the ratio of the actual concentration to the theoretical concentration was in the range 0.86 to 1.36, and the dilution and washing of the liquid proceeded as theoretically predicted.

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