



A comparison of microscopic and spectroscopic identification methods for analysis of microplastics in environmental samples



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ABSTRACT

The analysis of microplastics in various environmental samples requires the identification of microplastics from natural materials. The identification technique lacks a standardized protocol. Herein, stereomicroscope and Fourier transform infrared spectroscopy (FT-IR) identification methods for microplastics (<1 mm) were compared using the same samples from the sea surface microlayer (SML) and beach sand. Fragmented microplastics were significantly ($p < 0.05$) underestimated and fiber was significantly overestimated using the stereomicroscope both in the SML and beach samples. The total abundance by FT-IR was higher than by microscope both in the SML and beach samples, but they were not significantly ($p > 0.05$) different. Depending on the number of samples and the microplastic size range of interest, the appropriate identification method should be determined; selecting a suitable identification method for microplastics is crucial for evaluating microplastic pollution.

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1. Introduction

Microplastics have been recognized as emerging marine pollutants of significant concern, due to their persistence, ubiquity (Kubota, 1994) and toxic potential (Endo et al., 2005; Engler, 2012). Large plastic debris disintegrates and becomes smaller (<1 mm) microplastics, by photolytic, mechanical and biological degradation processes in the marine environment (Browne et al., 2007; Andrady, 2011; Cooper and Corcoran, 2010). Bioavailability increases with the decrease in size of plastic debris (Gregory, 2009), and microplastics have a greater likelihood of absorbing and desorbing toxic chemicals, due to their increased surface area (Lee et al., 2014). Investigation of their negative impacts on marine environments is based on quantification and qualification of microplastics. For this, it is essential to use reliable analytical methods.

Analysis of microplastics from various environmental samples requires a series of procedures including sampling, separation, cleanup and identification. Although several studies on method development and/or comparison for sampling (Norén, 2007,

2011; Song et al., 2014), separation (Imhof et al., 2012; Claessens et al., 2013), cleanup (Claessens et al., 2013) and identification (Vianello et al., 2013), have been carried out, it is still critical to improve methods to yield more precise and accurate results. Among these identification methods, the most widely used should be evaluated for their relevance to future studies. Recently, small-sized microplastics have been found in the marine environment (Thompson et al., 2004; Frias et al., 2010) and the abundance of microplastics increased exponentially with decreasing particle size (Song et al., 2014). The smaller microplastics are more difficult to identify. Ambiguous characteristics of non-plastics (resembling plastics) and plastics (resembling non-plastics) make it difficult to accurately identify microplastics.

The identification of microplastics using three methods has been investigated. First, only the naked eye and/or microscope (McDermid and McMullen, 2004; Costa et al., 2010; Norén, 2007; Collignon et al., 2012; Boerger et al., 2010; Lindborg et al., 2012; Heo et al., 2013) were used to identify microplastics, and some studies included microplastics of <1 mm. Second, microscope (and the naked eye) and instruments were used together (Martins and Sobral, 2011; Doyle et al., 2011; Nor and Obbard, 2014). The microplastics were identified mainly using a microscope or the naked eye and a limited number of selected particles were identified by a spectroscopic method using a Fourier

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transform infrared spectroscope (FT-IR). Third, all of the counted microplastic particles were identified by a spectroscopic method using an FT-IR or a Raman spectroscope (Vianello et al., 2013; Browne et al., 2011; Ng and Obbard, 2006; Song et al., 2014). However, prior to this study there has been no comparison of the advantages, disadvantages or accuracy of methods of microplastics identification.

In this study, we aimed to assess the microscopic and spectroscopic identification of microplastics. Specifically, we addressed the following questions: (1) How different are the abundances of microplastics determined using the microscopic and spectroscopic identification methods? (2) If the abundances of microplastics differ between the two methods, which component contributes to the measurement error? For the comparison, stereomicroscope and FT-IR microscope identification methods were applied to the same samples from surface microlayer water and beach sand. In addition, microplastic abundances were compared according to type (fragment, fiber, sheet and expanded polystyrene (EPS)) and size.

2. Materials & methods

2.1. Microplastic sampling

2.1.1. Surface microlayer

The microplastics sampling region and methods from the sea surface microlayer (SML) have been published elsewhere (Song et al., 2014) in detail. Water samples were collected near- and off-shore of Geoje Island, which receives riverine discharge from the nearby Nakdong River, South Korea (Fig. 1). The SML water samples were collected at 10 stations in May and July, 2012 (Song et al., 2014). A metal sieve was used for SML water samplers, which

typically collected SML at a depth range of 150–400 μm (Cunliffe et al., 2013). The microplastics and the SML water were trapped within the metal sieve mesh spaces by surface tension. A 2-mm mesh sieve of 20-cm diameter was placed in contact with the sea surface 100 times, covering a 3.14-m² sampling area at each station. The water trapped within the mesh spaces was collected in the stainless tray and transferred to a 1-L polyethylene bottle. The final volume of SML water sampled per station was in the range of 2.2–2.8 L.

2.1.2. Sand beach

Sediment samples were collected from six beaches on Geoje Island, which are affected by riverine discharge from the nearby Nakdong River, in May, 2012 (Fig. 1). At each beach, 10 positions were randomly selected along the high strandline. About 12.5 L of sand samples were collected in a 0.5 \times 0.5 m quadrat with 5-cm depth, using a stainless scoop, through a 1-mm sieve (Tyler sieve, CISA, Spain). The sieved sand samples were well mixed in a stainless tray and 1 L of sand was transferred to a polyethylene bottle. In the case of wet sand samples, sieving was conducted after air-drying in the laboratory to avoid contamination of the mesh screen cover.

2.2. Analysis of microplastics

2.2.1. Microscopic analysis

The SML samples were filtered (GF/F; 0.75 μm ; 47 mm \varnothing) in the laboratory. The filter papers were dried at 60 $^{\circ}\text{C}$ and stored in Petri dishes. The microplastics on the filter paper were identified and counted using a stereomicroscope (ZEISS Model Discovery “SV8”). Microplastics were categorized into four types (fragment, fiber, sheet and expanded polystyrene (EPS)) and again according to six maximal length classes (<50, 50–100, 100–200, 200–500,

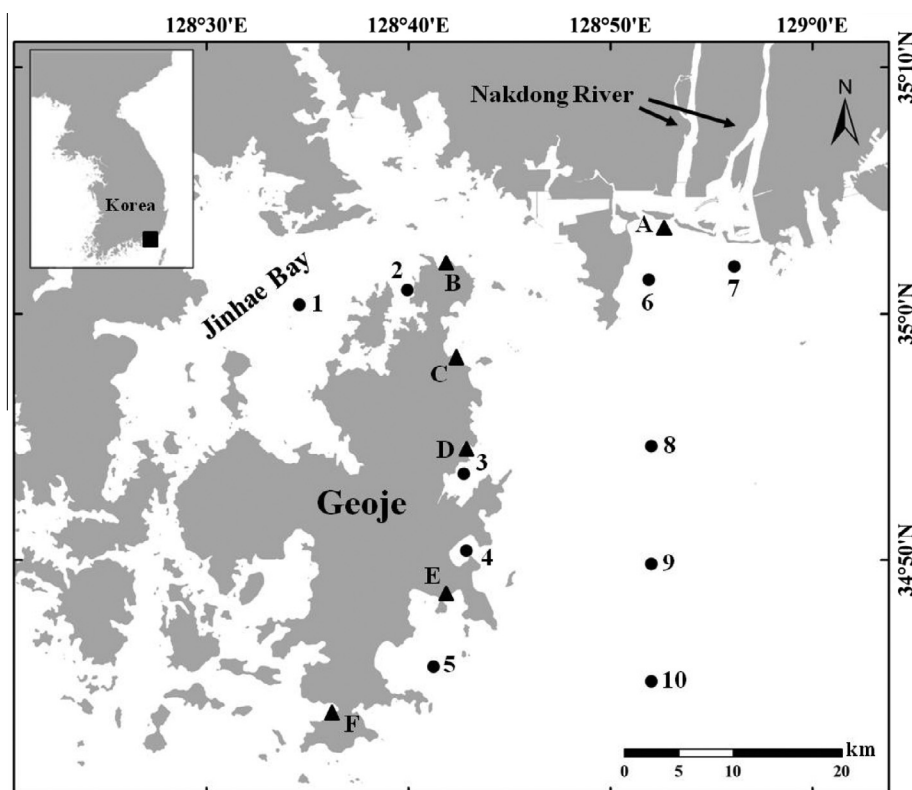


Fig. 1. Map showing the locations of microplastic sampling stations for surface microlayer water (● – Stations 1–10) and beach sediment (▲ – Stations A–F: A; Jinwoodo, B; Guyoung, C; Heungnam, D; Deokpo, E; Wahyun and F; Myoungsa) around Geoje Island, South Korea.

500–1,000, and >1000 μm). Although the EPS particles <1 mm in size in SML and beach sand generally belong to the fragment type and have an irregular shape, they are classified separately due to their exceptionally high abundance in Korea (Lee et al., 2013, 2015). Visual identification of plastic particles followed the criteria reported by the Kommunes Internasjonale Miljøorganisasjon (Local Authorities International Environmental Organisation (Norén, 2007) and empirical research.

For sand samples from beaches, 50-mL sand from each 1-L sample from 10 quadrats was sub-sampled to produce a composite sample (total, 500 mL) representing each beach. The composite sample was well mixed with 330 mL of saturated NaCl solution, vigorously shaken by hand for 1 min and then kept stationary for 10 min. This process was repeated three times for each sample, and the supernatants were collected in a 1-L beaker and stored stationary for 1–24 h, depending on the sample, until the overlying water looked clean. After this time, the supernatant was filtered (polycarbonate membrane filter 1.2 μm ; 47 mm \varnothing). The filters were placed in Petri dishes and dried at 60 °C. The microplastics on the filter paper were identified and counted using a stereomicroscope (ZEISS Model Discovery “SV8”). The microplastics were categorized into the same four types as the SML samples, but not categorized by size.

Two liters of distilled water ($n = 3$) in the polyethylene sampling bottle as that used for the SML water was filtered using a glass fiber filter, and one liter of saturated NaCl solution ($n = 3$) used for density separation for sand samples was filtered using a polycarbonate filter as a blank sample. Particles on the dried filter papers were counted and identified using an FT-IR microscope in the same way as the microplastic particles were counted. None of the plastics were detected in both the blank samples.

2.2.2. Spectroscopic analysis

Microplastic particles on the filter paper from both the SML water and beach sand samples were simultaneously counted and identified using a FT-IR microscope. Particles were counted in five randomly selected squares (SML; $7.9 \times 7 \text{ mm}$; total area = 2.76 cm^2 , beach; $8 \times 7.8 \text{ mm}$; total area = 3.14 cm^2) on the filter paper, which accounted for 25% of the total filtered area. Whole plastic-like particles under an optical microscope ($\times 40$ or $\times 100$ magnification) in each square were selected and immediately identified using the FT-IR (Thermo Nicolet 6700 and Continuum) equipped with a liquid nitrogen-cooled mercury cadmium telluride (MCT) detector and attenuated total reflectance (ATR) consisting of a germanium crystal at an incidence medium angle of 25°. The spectra were recorded as the average of 128 scans in the spectral range of $650\text{--}4000 \text{ cm}^{-1}$ at a resolution of 8 cm^{-1} and an aperture of 100 μm . The type and size classification of microplastics was as for the stereomicroscope method.

2.3. Statistical analysis

All statistical tests were performed using the SigmaPlot version 11 software (Systat Software Inc.). A Mann–Whitney rank sum test was used to assess differences in the abundance of microplastics using the microscope and spectroscopic identification methods.

3. Results

3.1. Comparison by microplastic type and size

The abundance of microplastic particles in SML and beach sand was compared between the two different identification methods, stereomicroscope (hereafter microscope) and FT-IR. The pattern of abundance of microplastics by type was similar in SML and

beach sand (Fig. 2). The mean abundances of fragments and EPS were greater by FT-IR than by microscope, while the mean abundances of fiber and sheet were lower by the FT-IR method. The fragment abundances were 127 ± 111 particles/L (microscope) and 206 ± 117 particles/L (FT-IR) for the SML water, and 152 particles/L (microscope) and 1192 particles/L (FT-IR) for the beach sand. The fragment abundances in the SML and beach by FT-IR identification were significantly ($p < 0.05$) higher than those by microscope. For EPS, 0.1 ± 0.2 particles/L (microscope) and 0.4 ± 1.8 particles/L (FT-IR) were in the SML, and 631 particles/L (microscope) and 1521 particles/L (FT-IR) were in the beach, respectively, but the abundance was not significantly ($p > 0.05$) different between the microscope and FT-IR methods. The abundance of fiber was significantly ($p < 0.05$) lower with FT-IR (4.5 ± 4.1 and 8 particles/L) than microscope (13 ± 15 and 29 particles/L) in the SML and beach samples, respectively. A few sheets were identified using the microscope, but via FT-IR, none was detected. The total abundance of microplastics was offset because of the different increasing and decreasing trends between the microscope and FT-IR methods. The total abundance of microplastics was higher by FT-IR than by microscope both for the SML and beach samples, albeit not significantly so ($p > 0.05$) due to high variability of the abundances according to the sampling stations and periods (see section below).

The abundance of microplastic fragments of all size classes was higher by FT-IR (Fig. 3a). FT-IR showed an obvious trend of increasing abundance with decreasing size, while the microscope method demonstrated a peak of abundance in the 50–100- μm size class. The abundance of fragments in the <50- μm size class was significantly different ($p < 0.01$) between the microscope and FT-IR methods. The abundance of fiber was significantly higher ($p < 0.05$) by the microscope for the 200–500-, 500–1000-, and >1000- μm sizes than by FT-IR, with the exception of the <50- μm class, in which no fiber was detected using either method (Fig. 3b). The microscope method showed a trend of increasing abundance with increasing size of fiber, while FT-IR showed no clear trend. Fibers counted by microscope in the 50–200- μm size range were not detected by FT-IR. There was no difference in abundance of sheet type microplastics by size in between spectroscopic and FT-IR (Fig. 3c). The abundance of EPS of <200 μm was higher by FT-IR, while that of EPS of >200 μm by the microscope method was not confirmed by FT-IR (Fig. 3d).

3.2. Comparison by sampling station

Fragment microplastics were widely distributed throughout the study area in SML (Fig. 4) and beach samples (Fig. 5). The difference in abundance between the microscope and FT-IR methods varied at all stations, but the degree of variation was greater in May than in July in the SML. The fiber abundance was higher by the microscope method than FT-IR at all stations in May, while the abundance at some stations was higher by FT-IR than by microscope in July. The synthetic fibers identified by FT-IR were typically 10–35 μm in diameter and colored, with a few white fibers of slightly thicker, 10–70- μm , diameter.

In the beach sand, the abundance of fragmented microplastics was higher by FT-IR compared to the microscope, except for Wahyun beach (Fig. 5). The abundance of fragments at Deokpo was 52-fold higher by FT-IR than by microscope. Fiber was detected at all beaches with the microscope, while only the Heungnam beach had fibers according to FT-IR, the abundance of which was 1.5-fold higher than that determined by the microscope method. All detected synthetic fibers in SML were of 20- μm diameter. For EPS, the abundance was higher for all beaches using the FT-IR microscope.

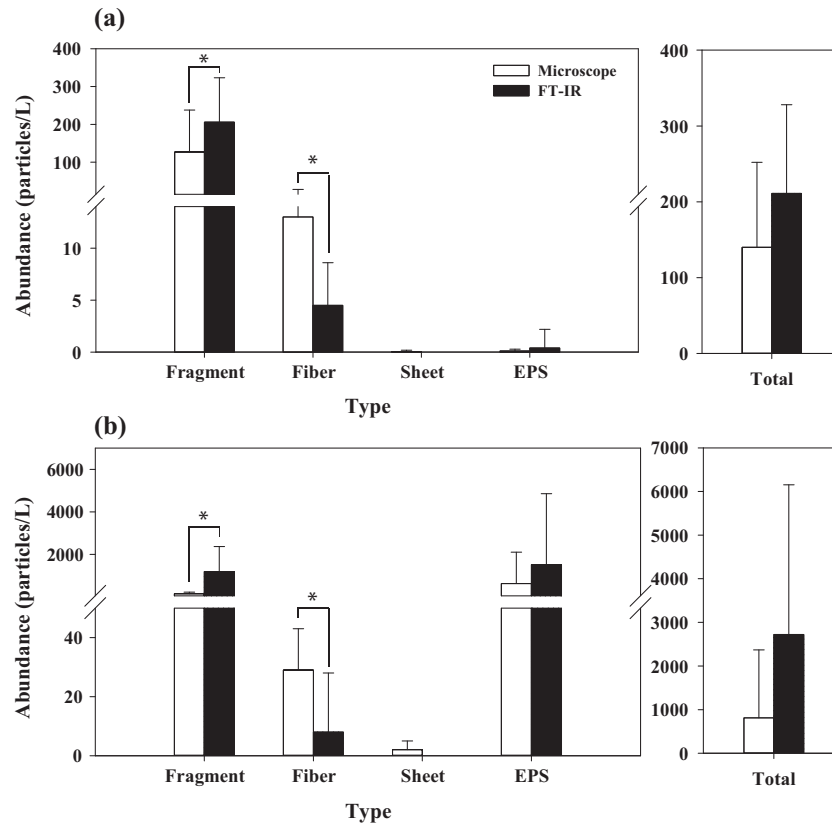


Fig. 2. A comparison of the abundance of microplastics by type in (a) the sea surface microlayer (SML) and (b) beach sand, as identified using a stereomicroscope and Fourier transform infrared spectroscopy (FT-IR). Note: abundance data of microplastics in SML by FT-IR identification were cited from Song et al. (2014) ($*p < 0.05$).

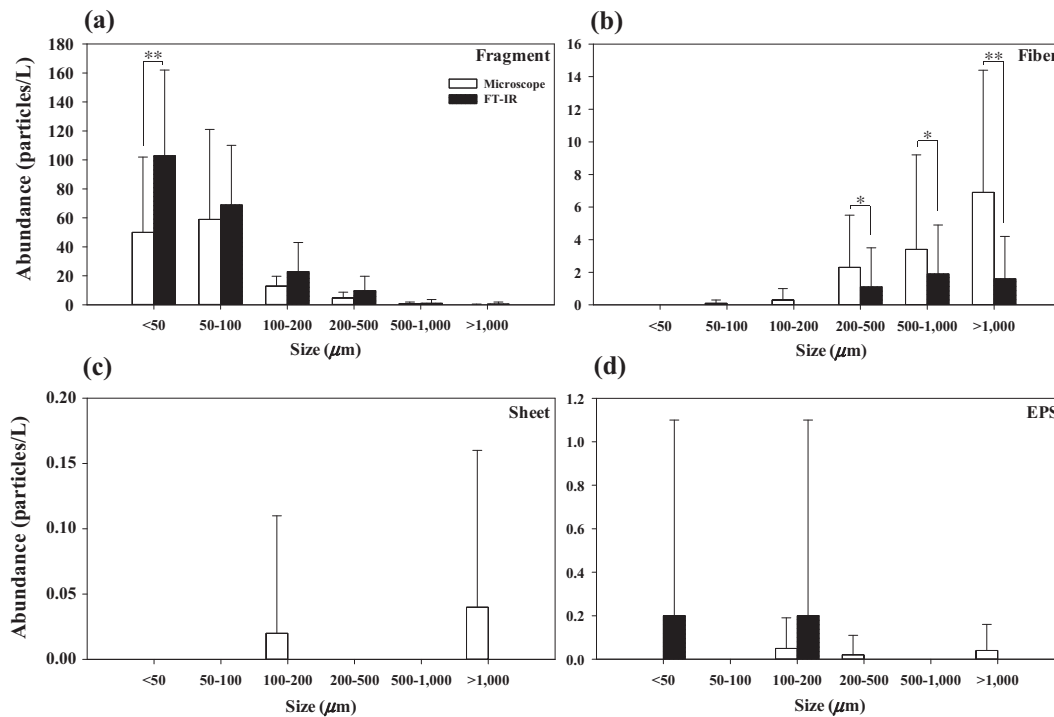


Fig. 3. A comparison of the abundance of microplastics in the sea surface microlayer by size and type ((a) fragment; (b) fiber; (c) sheet; (d) expanded polystyrene) identified using a stereomicroscope and Fourier transform infrared spectroscopy (FT-IR). Note: abundance data of microplastics in SML by FT-IR identification were cited from Song et al. (2014) ($*p < 0.05$, $**p < 0.01$).

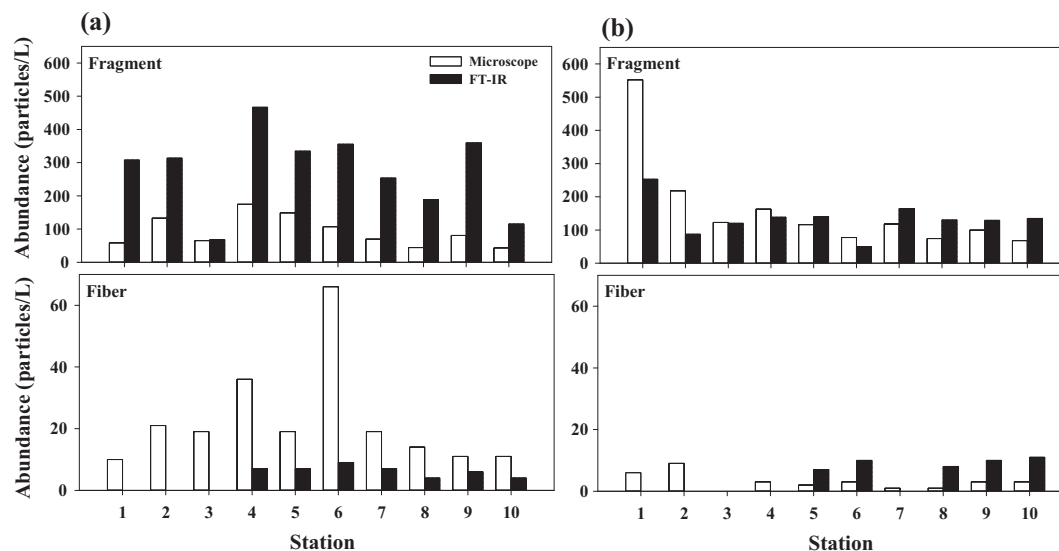


Fig. 4. A comparison of the abundance of microplastics by type (fragment and fiber) in the sea surface microlayer in (a) May and (b) July identified using a stereomicroscope and Fourier transform infrared spectroscopy (FT-IR). Note: abundance data of microplastics in SML by FT-IR identification were cited from Song et al. (2014).

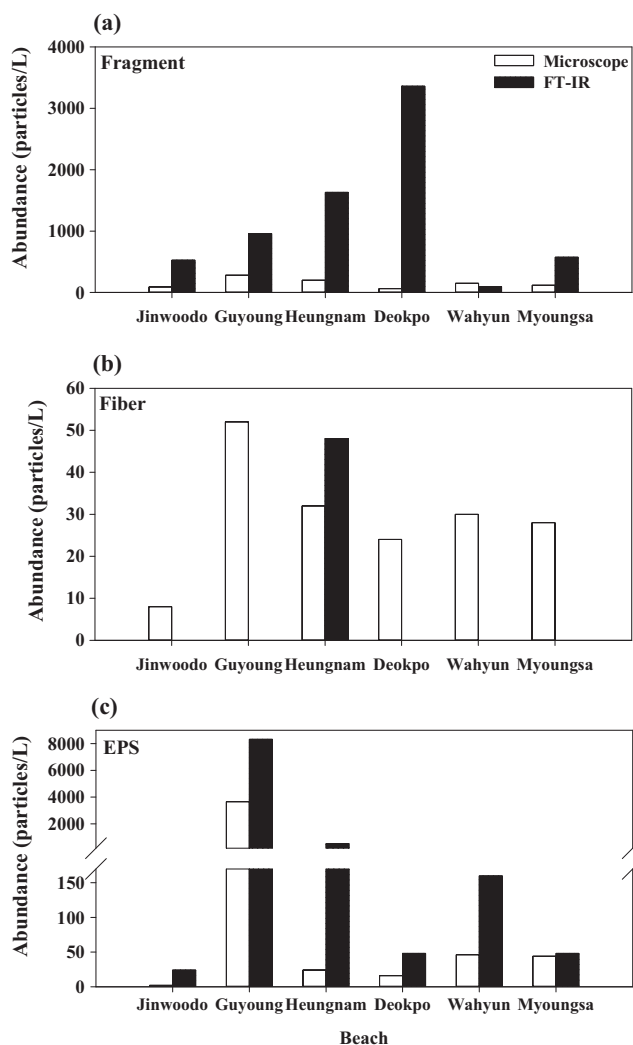


Fig. 5. A comparison of the abundance of (a) fragment, (b) fiber, and (c) expanded polystyrene (EPS) in beach identified using a stereomicroscope and Fourier transform infrared spectroscopy (FT-IR).

4. Discussion

4.1. Underestimating and overestimating microplastics

A significantly ($p < 0.05$) greater number of fragments were detected by FT-IR than by microscope. Two possible explanations exist for the underestimated abundance of fragments using a microscope. First, many transparent or white fragments were identified as synthetic polymers, such as polyethylene (PE) and polypropylene (PP) by FT-IR, but were not counted as microplastics using the microscope (Fig. 6). Particularly in the beach sand, there were many transparent or white PE or PP fragments; thus the number of fragments was 7.9-fold higher by FT-IR. In the SML, many colored fragments were identified as synthetic resin particles from paints and thermoset matrix in fiber-reinforced plastics (FRP) used for ships [alkyd or poly (acrylate: styrene)] by FT-IR (Song et al., 2014); these could relatively easily be considered as microplastics by a microscope, with the exception of particles $< 50 \mu\text{m}$ in size, and thus the number of fragments was 1.6-fold higher by FT-IR. Second, the limitation of identification by microscope for microplastics of which size is less than 1 mm. In practice, the shape and morphology of < 1 - and 1–5-mm microplastics differ to the naked eye. With objects < 1 mm in size, it is more difficult to distinguish between plastic and non-plastic because of their similar features. Using the naked eye, 1–5-mm microplastic can be discriminated from non-plastic, by evaluating their color, hardness, roughness and even the thrum of the plastic. In the case of EPS, its milky white color, slight shine and amorphousness make microscopic identification easier compared to other types of plastic, although detecting EPS $< 200 \mu\text{m}$ was problematic using the microscope. To a lesser extent, it is also difficult to distinguish green and blue resin particles in SML of $< 200 \mu\text{m}$.

The criteria reported by Norén (2007) and practical experience was used for the identification of fiber in this study. Plastic fiber has no cellular or organic structure and should be consistent in thickness, with no taper toward the ends. Also, the fibers demonstrate three-dimensional bending, and are clear and homogeneously colored red, blue, green, gray or dark purple. Even with identification of fiber being conducted using criteria, some faded colored fibers that appeared to be synthetic polymer were cotton,

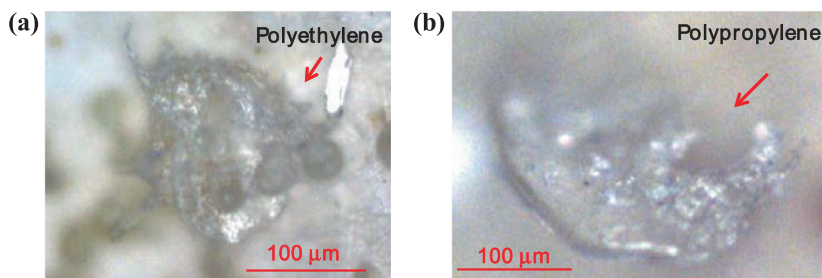


Fig. 6. Pictures of transparent (a) polyethylene and (b) polypropylene fragments on filter paper.

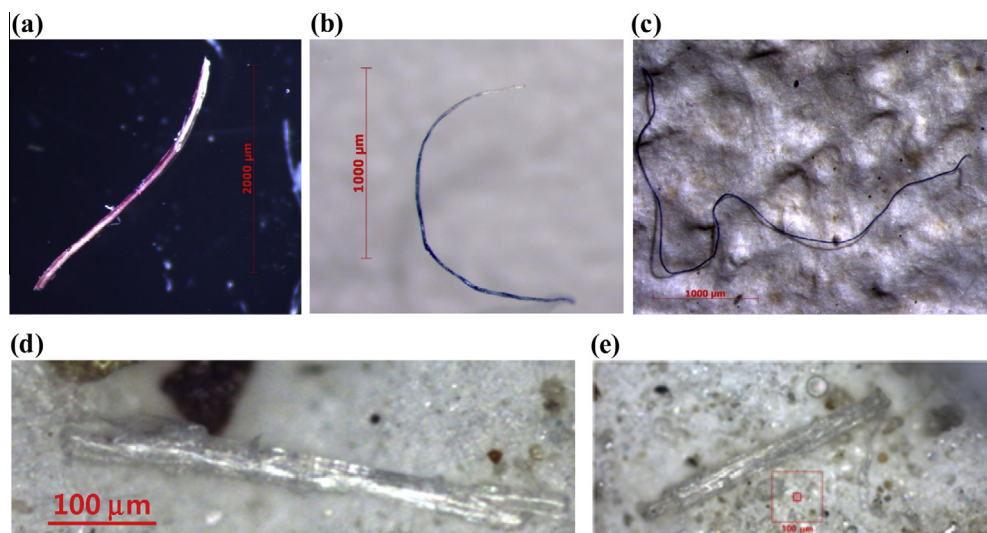


Fig. 7. Pictures of natural fibers; (a) non-plastic (organic), (b) non-plastic (cotton) and (c) non-plastic (rayon) and synthetic fibers; (d) and (e) impact polypropylene.

rayon, or other organic materials (Fig. 7). In particular, when using the microscope for identification of microplastics, plastic-like fibers such as cotton and rayon were apparently counted as microplastics, accounting for 85% of the fiber in the SML water sample via FT-IR in this study. The spectrum of cotton is similar to that of an artificial rayon fabric, so their spectra frequently showed up on the library list with similar match percentages. According to their shape and color, they were assumed to originate from fabric. Although colored cotton and rayon are anthropogenic in origin, they comprise natural polymers or purified cellulose. They are one of the marine debris items, but further discussion is required to determine whether they should be included as microplastics or not. If they are prevalent in the marine environment and causing a harmful impact on the environment, they should be considered microplastics and be monitored. In this study, sea grass and cotton prevailed at all stations in SML water in May, while organic fiber levels were lower in July. Another fiber type that did not comply with the identification criteria was white impact PP (combined PP and PE for better properties). The impact PP was not counted using the microscope because of its white color, irregular shape, and inconsistent thickness (Fig. 7). The white fibers were likely derived from rope debris such as aquaculture buoys, fishing nets, and ship ropes originating from ocean activity around Korea (Jang et al., 2014). All sheets that were counted as microplastics using the spectroscopy turned out to be skeletons of various organisms.

4.2. Advantages and disadvantages of identification methods

The microscope method for identification of microplastics is easier and faster than FT-IR. However, microplastics <1 mm in size

are more likely to be missed or miscounted (non-plastics identified as plastics). Using only microscope identification is therefore suitable for microplastics of >1 mm. In this study, the detected samples contained particles of <1 mm and had a considerable quantity of impurities. In the visual counting, many non-plastic particles with a variety of colors (organisms or minerals) and that appeared to be like plastics, were counted as microplastics. However, many microplastic particles were excluded from the count because of their characteristics (transparency, and colors such as black and white). This tendency resulted in underestimation or overestimation of microplastic abundance, depending on the sample.

In contrast, the spectroscopy method was more accurate than the microscope, substantially reducing the missing and miscount rates. In particular, small microplastics (<1 mm) were detected by FT-IR, including those <50 µm. Because the abundance of detected microplastics increased with decreasing size in the marine environment, accurate identification of microplastics, such as by FT-IR, is essential in the microplastics research field. The most important advantage is the ability of FT-IR to confirm the polymer types of microplastics, which can also provide additional information such as origin and further behaviors (Song et al., 2014). Knowledge of the polymer composition of a microplastic is important. For example, sorption capacities for toxic chemicals change according to polymer type (Lee et al., 2014). However, FT-IR which can detect micro-sized particle is not only expensive but it is also time-consuming to identify the microplastic-like particles one by one. The ATR must contact the surface of the sample, then a beam of infrared light is reflected at least once by passing through the ATR crystal. Consequently, the spectrum is susceptible to variations in the surface condition of the samples (Smith, 2011). Even

if results depended on the sample size, type and condition, several trials are required to obtain a clear spectrum of tiny, rough, and contaminated samples for confirmation. It took approximately 4–5 h for one sample in this study, which was 8–10-fold longer compared with the microscope method (*ca.* 30 min). For this reason, only part of the filtered paper (25% of the total filtered surface) was identified by FT-IR. This is an appropriate method, compared with the whole paper identification method, as it produces very little difference in the distribution of abundance of microplastics (Chae et al., 2014), but it may miss low abundance particles on the filter paper.

Although FT-IR can detect small microplastics down to 10–20 µm, it misses plastics smaller than the aperture size. Raman spectroscopy, using a laser beam, can focus on a smaller area than FT-IR and detect microplastics down to 1–2 µm in size. However, few practical instruments for detecting nano-sized microplastics in real complex field samples are available. Automated mapping of FT-IR at certain typical polymer wavenumbers was conducted by Vianello et al. (2013). This method also analyzes the abundance and composition of microplastics, and several polymer types can be detected in a single run, but in only a limited area of the sample (5.6%). This method will be useful in laboratory experiments using known-polymer samples. In particular, identifying unknown particles in field samples using an automated mapping of FT-IR is difficult. The spectra of samples are difficult to match with those of the library with high percentages, due to the weathered and contaminated surfaces of plastics. For identification of microplastics using the FT-IR, an expert with experience in interpreting spectra is needed.

4.3. Recommendation of identification method

Because the detected and identified microplastics have decreased in size, there is a limitation to quantitative analysis when using only the naked eye or a microscope. The spectroscopic method should be used for identification of small microplastics. If few samples are being assessed, all identification of plastic-like particles by FT-IR or Raman is recommended. This method is relatively accurate for obtaining information on the abundance and polymer types of microplastics. If many samples are being evaluated, use of a combination of both the microscope and spectroscopic methods is recommended. In terms of this combined method, a screening analysis using FT-IR or Raman should be conducted first, to create sample-based criteria for the identification of major and typical microplastics in sample groups, according to matrix, season, and location. Then the stereomicroscope should be used to count microplastics according to these criteria. This is because the characteristics of microplastics could differ among samples, such as the SML and beach sand. When the SML and beach samples were identified using the microscope method by the same person following FT-IR screening, the miscount percentages of non-plastics to plastics were 18% and 22% for SML and beach samples, respectively. Therefore, determination of criteria for each sample by FT-IR screening could reduce the error in counting microplastics between the microscope and FT-IR methods. The appropriate identification method should therefore be determined depending on the number of samples and the size range of interest. Selecting a suitable identification method for microplastics is crucial for evaluating microplastic pollution.

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