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Qualitative assessment to determine internal and external factors influencing the origin of styrene oligomers pollution by polystyrene plastic in coastal marine environments



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ABSTRACT

The objective of this study is to investigate the qualitative contribution of internal and external factors of the area contaminated by polystyrene (PS) in coastal marine environments. This study is based on the extensive results of monitoring the styrene oligomers (SOs) present in sand and seawater samples along various coastlines of the Pacific Ocean. Here, anthropogenic SOs is derived from PS during manufacture and use, and can provide clues about the origin of SOs by PS pollution. The monitoring results showed that, if the concentration of SOs in water is higher than those concentrations in beach sand, this area could be affected by PS plastic caused by an external factor. On the other hand, if the concentration of SOs is higher in the beach sand, the region can be mainly influenced by PS plastic derived from its own area. Unlike the case of an external factor, in this case (internal influence), it is possible to take policy measures of the area itself for the PS plastic problem. Thus, this study is motivated by the need of policy measures to establish a specific alternative to the problems of PS plastic pollution in ocean environments.

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1. Introduction

Since mass production began in the 1940s (Halden, 2010; Cole et al., 2011), the amount of plastic manufactured globally has increased rapidly from 1.7 million tons in 1950 to 311 in 2014 (PlasticsEurope, 2015a). Meanwhile, the amount of plastic waste being generated globally was estimated 275 million tons in 2010, with 4.8–12.7 million tons entering the ocean (Jambeck et al., 2015), where they can persist and accumulate. More unfortunately, plastic debris derived from the breakdown of discarded macro-plastic is found everywhere in marine environment (Barnes et al., 2009; Cole et al., 2011; Eriksen et al., 2013; Barboza and Gimenez, 2015). Thus, the global problem of plastic pollution in marine environments is a current and growing concern.

Plastics, including micro-plastics and nano-plastics, discarded in

the ocean break into smaller plastic debris causing death to marine wildlife through entanglement, smothering, and ingestion (Thompson et al., 2004; Browne et al., 2008; Cole et al., 2011; Hidalgo-Ruz et al., 2012; Avery-Gomm et al., 2012; Derraik, 2002; Laist, 1987; Stephanis et al., 2013; Barboza and Gimenez, 2015; Bergami et al., 2016). In addition, this plastic debris can also act as a media for both transporting persistent organic pollutants (POPs) and leaching plasticizers (often termed "plastic additives") that are considered toxic in the oceans (Thompson et al., 2004; Teuten et al., 2009; Ogata et al., 2009; UNEP, 2011; Lithner et al., 2011; Engler, 2012; Rochman et al., 2013; Koelmans et al., 2013, 2014).

In order to qualitatively and quantitatively explain these plastic marine pollution problems mentioned above, various methods have been attempted, of which the concentration (density or abundance) of plastic debris using counting techniques is the typical indicator (Law et al., 2010; Rees and Pond, 1995; Ryan, 2013; Thompson et al., 2004). In this technique, the concentration is generally expressed as the number or the amount of plastic items of debris per unit area, in which the sizes and shapes of the plastic debris have often been considered in recent studies (Rees and Pond,

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1995; Thompson et al., 2004; Browne et al., 2010; Ryan, 2013). However, according to Jambeck et al. (2015), in the visual counting method for plastic debris, the source of the debris cannot be traced. In addition, effective mitigation strategies and/or policy implications to remove the debris from the ocean environment are also not offered, due to the smaller size of the debris as a resulting of weathering. Further, for practical reason, it is difficult to assess the accumulation of plastic debris as a sink, i.e., on the seaboard, owing to not only the vastness of the oceans compared to the size of the plastic debris, but also oceanic currents and seasonal patterns (Law et al., 2010; Cole et al., 2011; Jambeck et al., 2015).

Although it is generally recognized that approximately 80% of plastic debris originates from land sources (Barnes et al., 2009; Cole et al., 2011), there is a lack of sufficient evidence to support this (Jambeck et al., 2015). In addition, there is some debate as to whether levels of plastic debris are still increasing or have stabilized (Thompson et al., 2004; Law et al., 2010; Cole et al., 2011). For example, due to the largely spatial and temporal variability caused by the very patchy distribution of plastic debris (Barnes et al., 2009; Cole et al., 2011), the visual counting technique of the plastic debris collected may not provide an accurate indicator of plastic pollution in the marine environment (Cole et al., 2011).

Despite significant concerns of plastic marine contamination as an emerging major environmental problem, the large gaps in the research need to be addressed, particularly in terms of the factors related to plastic debris within specific areas and the evaluation of potential mitigation strategies for clean-up operations in all marine environments (Cole et al., 2011; Barboza and Gimenez, 2015). Furthermore, the degree of contribution of the internal or external factors of an area contaminated by plastic waste in a marine environment needs to be assessed across a wide region, which is the focus of this study. In other words, research of plastic pollution that indicates whether or not the debris originates in the area itself has seldom been considered. It is therefore necessary to assess the present cleaning efforts along various coastlines (Leite et al., 2014).

The objective of this study is to report on the degree of qualitative contribution of the internal and external factors of an area contaminated by polystyrene (PS) to the coastal marine environment, assessing whether or not the debris originates from the area itself. In order to meet this objective, this study monitors the concentrations of styrene oligomers (SOs) as an indicator of PS plastic pollution along the coastlines of the Pacific Ocean, focusing on styrene trimer (ST: 2, 4, 6-triphenyl-1-hexene), styrene dimers (SDs; 2,4-diphenyl-1-butene, SD1; 1,3-diphenyl propane, SD2), and styrene monomer (SM), derived from PS (Saido et al., 2014; Kwon et al., 2014, 2015). These SOs are composed principally of two to three benzene rings. Hereafter, the sum of the concentrations of SM, SDs, and ST will be referred to as SOs.

2. Materials and methods

2.1. Study areas and sampling methods

Considering safety, all sand and surface seawater samples were collected from each coastal beach in the cities and islands of the Pacific Ocean, which included 3 nations (Korea, Japan, and USA) and 15 sampling sites (Fig. 1). We have already taken a number of additional samples from the same location mentioned in our previous studies (Saido et al., 2014; Kwon et al., 2014, 2015). The sampling locations are detailed in Table 1. The sampling period was from April 2009 to November 2010. In this study, material containing PS was excluded from all sampling and extraction procedures to eliminate any errors.

Sand samples were taken from the surface of the coast and from a 30 cm depth along the seashore. At two sampling points, about

100 g of sand was collected using a stainless steel shovel, and was stored in a glass container.

Surface seawater samples were taken at a water depth of 40 cm and subjected to cotton plug filtration using a stainless steel beaker. The volume of the water sampled was 5–10 L, which was then extracted in the field with dichloromethane (DCM), with biphenyl (BP) added as the surrogate standard for the recovery test. The water samples in the field were immediately extracted using a portable shaker (Saido et al., 2014; Kwon et al., 2014, 2015). Unlike our previous study, in this study, the water sample volume was taken five to ten L to measure the very small amount of SOs. Other procedures for sampling used in this study are described in details in our previous studies (Saido et al., 2014; Kwon et al., 2014, 2015).

2.2. Sample preparation in the laboratory

In the preparation of seawater, about 100 mL of the DCM extract was mixed overnight with approximately 10 g anhydrous sodium sulfate (ACS grade, Sigma-Aldrich, USA). The extract was evaporated to a dry state by a rotary evaporator at 30 °C. After adding 0.5 mg L⁻¹ phenanthrene as an internal standard, the eluate was completely dissolved into 1 mL of benzene. Further details on other sampling methods in the field and sample preparation in the laboratory have been provided in previous studies (Saido et al., 2014; Kwon et al., 2014, 2015).

In the preparation of the sand sample transported to the laboratory, the water contained in the sand sample collected from all the beach sites needed to be removed. To remove the water from the sand sample, about 10 g of sand was placed in a flask and was freeze-dried overnight. Among this sand sample, 5 g of sand was placed in a flask. Ten mL DCM was added to the sand sample to obtain an extract which was ultrasonificated for 1 min. This process was repeated three times with 10 mL DCM. Finally, the total DCM extract was 30 mL. The subsequent process was treated in the same way as the preparation of the seawater sample mentioned above.

2.3. Analytical method and QA/QC

This analytical method was applicable to the measurement of SOs extracted by DCM. The actual measurement of SOs was based primarily on gas chromatography/mass spectrometry using HP 6890 GC with a JEOL Auto MS-II. Further details on the analytical methods in the laboratory have been provided in previous studies (Saido et al., 2014; Kwon et al., 2014).

The detection limit for each SO was determined to be about $3.0~\mu g~L^{-1}$, which was defined as the concentration based on the signal-to-ratio of 3. The recoveries of the SOs were examined by spiking the SOs with a known amount of surrogate biphenyl $1~\mu g~mL^{-1}$. The mean recovery was 94%. In particular, no equipment made of PS plastic was used in this course. Further details on the QA/QC including field blanks for water (Millipore ultra-purification system) have been provided in previous studies (Saido et al., 2014; Kwon et al., 2014).

3. Results and discussion

3.1. Distribution of SOs in the coastal environments

As shown in Fig. 1, the concentrations of ST and SD in all sand samples collected from coastal beaches selected in this study are mostly higher than the concentrations of SM measured in the same sampling places. Likewise, the concentrations of ST and SD in seawater were observed to show the same trend. In sand and water samples, the contribution to anthropogenic SOs decreased mostly in the following order: ST (or SD) > SD (or ST) > SM. As a result, the

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