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Benzotriazole-type ultraviolet stabilizers and antioxidants in plastic marine debris and their new products

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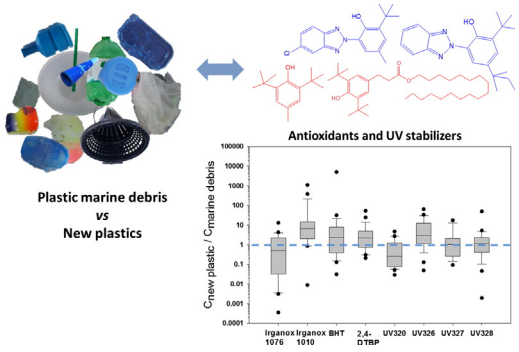
HIGHLIGHTS

- Quantitative measurement of antioxidants and UV stabilizers in plastic marine debris
- A comparative study of additive contents between marine debris and new plastics
- Plastic marine debris has the potential to act as vectors and carriers of hazardous chemicals in the marine environment.
- High and irregular use of additive chemicals in short-term use plastics.

GRAPHICAL ABSTRACT



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ABSTRACT

Ultraviolet stabilizers (UVSs) and antioxidants are the most widely used additives in plastics to enhance the lifetime of polymeric materials. There is growing interest in the roles of plastic marine debris and microplastics as source or vector of toxic substances to marine environment and organisms. However, there is limited information available on plastic associated chemicals, particularly additive chemicals. Therefore, to evaluate their extent of exposure from plastics to the marine environment, we determined UVSs and antioxidants in plastic debris ($n = 29$) collected from beaches along with their corresponding new plastic products in markets ($n = 27$) belonging to food, fisheries, and general use. Antioxidants were present at higher concentrations than UVSs in both plastic debris and new plastics, indicative of their high use over UVSs. Irganox 1076 and Irganox 1010 were more commonly used than other chemicals investigated. The irregular use with high concentration of additive chemicals was observed in short-term use plastic products. Except for Irganox 1076 and UV 326, most antioxidants and UVSs were relatively high in new plastics compared to corresponding plastic marine debris, implying their potential leaching or degradation during use or after disposal. The present study provides quantitative information about additive chemicals contained in plastic marine debris and their new products. These results could be useful for better understanding of environmental exposure to hazardous chemicals through plastic pollution.

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1. Introduction

Plastic resins, such as polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), polystyrene (PS), polyethyl terephthalate (PET), and

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polyurethane (PUR), have lightweight, strong, durable, and cheap characteristics that make them serious hazards to the environment (Laist, 1987; Pruter, 1987). The versatility of these materials has led to an increase in their production and use over the past three decades (Laist, 1987; Hansen, 1990; de la Puente et al., 2002). They have rapidly become important in all aspects of everyday life such as packaging, clothing, appliances, electronics, and vehicles to insulations, industrial applications, greenhouses, automotive parts, aerospace, and mulches (Al-Salem et al., 2010). The global production of plastic is growing at a rate of about 5% per year (APC, 2008) that was reached 288 MT in 2012, with projections of 400 MT by 2050 (Plastic Europe, 2013).

Pollution with plastic debris originating mainly from anthropogenic activities has been recognized as a major problem in fresh and marine water systems (Derraik, 2002; George et al., 2010; Andrady, 2011; Koelmans et al., 2014). In 2013, it was estimated that there was approximately 4.2 million tons of plastic debris that entered the oceans with a stock of 86 million tons at the end of year (Jang et al., 2015). Negative effects of plastics may be associated with entanglement or ingestion, which has been reported for benthic invertebrates, fish, turtles, birds and mammals (Chiras, 2004; Wegner et al., 2012; Besseling et al., 2013; Foekema et al., 2013). It is generally believed that microplastics may increase exposure of marine aquatic organisms to chemicals associated with plastic, such as persistent organic pollutants (POPs) or plastic additives (Teuten et al., 2009; Gouin et al., 2011; Hammer et al., 2012; Browne et al., 2013). These additives such as plasticizers, antioxidants, and anti-UV products are introduced (generally ranging from 0.1 to 1%) in plastics to improve their physical (mechanical, thermal, etc.) and chemical properties (Saint-Laurent and Rhainds, 2004; Arias et al., 2009). Plastic additives have relatively low molecular weights; thus, migration mechanisms from food, drugs, and cosmetic plastic packages (Figge and Freytag, 1980; Pellerin et al., 1984; Yagoubi et al., 1993; Garde et al., 1998; Kenshi et al., 2015) are often a major concern. Among them, benzotriazole ultraviolet stabilizers (UVSs) and synthetic phenolic antioxidants have attracted increasing public and scientific concern because of their occurrence and contamination of rivers, lake waters (Jungclaus et al., 1978; Fent et al., 2010; Giokas et al., 2005; Kameda et al., 2011), sediments (Nakata et al., 2009; Zhang et al., 2011), aquatic organisms (Kameda et al., 2011; Bachelot et al., 2012; Gago-Ferrero et al., 2013), tap water, and well water (Diaz-Cruz et al., 2012). Due to their release from consumer products, discharge through wastewater treatment plant (WWTP) effluent, adsorption onto marine sediments, and bioaccumulation, they have become emerging contaminants (Jurado et al., 2014). The first reports describing the presence of benzotriazole UVSs in the environment were presented by Jungclaus et al. (1978).

UV 326, UV 327, UV 328, UV 329, butylated hydroxytoluene (BHT), Irganox 1076, Irganox 1010, and 2,4-di-tert-butylphenol(2,4-DTBP or antixoidant 33) are commonly used. The 2,4-DTBP is also used as a chemical intermediate for the synthesis of UVSs or antioxidants and other chemical intermediates. Antioxidants are typically added to polymers at a concentration of 0.05–1% w/w (Drobny, 2007). BHT and 2,4-DTBP can also be present in plastic due to the degradation of bulky structured Irganoxs during processing of products (Brocca et al., 2002). The recent data on their global production and demand is not exactly available. Total plastic additive global demand (phthalate, antioxidants, heat stabilizers, UVSs, flame retardants, and antistatic reagent) was 3,279,113 t/y in 1996, with 316,462 t/y in Korea (STEPI, 1998). Among them, the global market demand for UVSs and antioxidants was 14,770 t/y and 85,300 t/y in 1996, respectively. The domestic demand for UVSs and antioxidants was 1330 t/y and 5000 t/y in 1996, respectively. The production, usage, and import of UV 320 has been prohibited in Japan since 2007 due to its potential for bioaccumulation (BCF: up to 10,000; NITE, 2009) and toxicities to the liver and other tissues in laboratory mammals (Ema et al., 2008). UV 327 and UV 328 accumulate in marine mammalian tissues (Nakata et al., 2010) due to their lipophilic properties (log Kow: 6.95 for UV 327; 7.25 for UV 328). The BCF of UV 327 between seawater and marine mammals was estimated to be 33,300 (Nakata et al., 2010), which was similar to that reported for a legacy persistent organic pollutant, hexachlorocyclohexanes (BCF: 37,000; Tanabe et al., 1984) in a western North Pacific food chain. Several long-term experimental studies showed that consuming excessive quantities of synthetic antioxidants can have negative effects on human health. Normally, synthetic antioxidants are polyphenolic compounds that are liposoluble and suspected of being responsible for liver damage and carcinogenesis (Grice, 1988), typically at concentrations of up to 100–200 mg/g in foods, either alone or in combination (Chang et al., 2013). BHT is of low acute toxicity, and Irganox 1010 and 1076 have very low orders of oral toxicity. Thermal migration of Irganox 1010 from various polyolefinic plastics into oleaginous vehicles has been reported (Marcato et al., 2003). Boonnorat et al. (2014) found that BHT and 2,6-DTBP can leach out up to 99.5% and 99.0%, respectively, within 500 days due to biodegradation in landfill leachate. BHT was often identified as the most abundant organic component in seawater (maximum concentration at 2.5 μg/L), river water, and wastewater samples (Kolpin et al., 2002; Oros and David, 2002). Wang et al. (2003) found BHT and 2,4-DTBP in sediments of Taihu Lake, China, in the range of 0.28 to 0.39 mg/kg (mean: 0.33 mg/kg) and 0.67 to 1.69 mg/kg (mean: 1.18 mg/kg), respectively, which was indicative of the high consumption in China. Environmental concentrations of Irganox 1076 and 1010 have not yet been reported. The above-mentioned studies emphasize the importance of understanding the fate of organic UV absorbers and antioxidants in the marine environment. It was interesting to find their high frequency during gas chromatography-mass spectroscopy (GC–MS) screening of chemical additives in plastic debris collected from coastal beaches, which may indicate that plastic is an important source of these hazardous chemicals (Rani et al., 2015).

Based on previous non-target screening analysis of marine plastic debris (Rani et al., 2015), four benzotriazole UVSs (UV 320, UV 326, UV 327, and UV 328) and four antioxidants (BHT, Irganox 1010, Irganox 1076, and 2,4-DTBP) were selected as target analytes in this study, and quantitative analysis was performed for marine plastic debris and their new products. To understand the environmental exposure of plastic additives through marine debris, we collected plastic debris along the Korean coasts. Additionally, the same or similar new plastics were purchased in local markets for comparison with marine debris. PE, PP, and PET are commonly used polymer materials for food packing/serving, fisheries, and general use, and are commonly present on the marine coast as debris. A total of 30 types of samples (plastic debris = 29, new products = 27) of marine debris (ID 1–29), and the corresponding new plastic (ID 1–22, 25–27 and 29–30) products of different categories (food-related, ID 1–15; fisheries, ID 16–24; and general use, ID 25–30) were analyzed (Table 1, Fig. 1). The levels and profiles of UVSs and antioxidants in debris and fresh products may give useful information about chemical exposure through plastics debris in the marine environment.

2. Material and methods

2.1. Chemicals and reagents

Reference standards of 2-tert-Butyl-6-(5-chloro-2H-benzotriazol-2-yl)-4-methylphenol (Bumetrizole or UV326) >98%, 2-(2′-hydroxy-3′,5′-di-tert-butylphenyl)-5-chlorobenzotriazole (UV 327) >98%, pentaerythritol tetrakis(3-(3,5-di-tert-butyl-4-hydroxyphenyl) propionate) (Irganox 1010) >98%, octadecyl 3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate (Irganox 1076) >98%, 2,4-DTBP >99%, and BHT ≥99% were purchased from Sigma Aldrich. Compound 2-(3,5-di-tert-butyl-2-hydroxyphenyl)2H-benzotriazole (UV 320) >98% and 2-(2H-benzotriazole-2-yl)-4,6-di-tert-pentylphenol (UV 328) >98% were purchased from Accu Standard Inc. (New Haven, CT, USA). The isotope labelled standard of 2,6-di(tert-butyl)-4-methyl phenol (d_{21}-BHT) >98% was purchased from Cambridge Isotope Laboratories

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