



Potential transfer of organic pollutants from littoral plastics debris to the marine environment[☆]



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ARTICLE INFO

Article history:

Received 22 November 2017

Received in revised form

31 January 2018

Accepted 31 January 2018

Keywords:

Regulated and emerging contaminants

Desorption

Seawater

Littoral plastics

Potential transfer

Marine environment

ABSTRACT

Plastic polymers act as passive samplers in air system and concentrate hydrophobic organic contaminants by sorption or specific interactions, which can be transported to other systems such as the marine environment. In this study plastic debris was sampled in the surrounding area of a Mediterranean lagoon in order to determine the concentration of persistent and emerging organic contaminants. More specifically, desorption of 91 regulated and emerging organic contaminants (polycyclic aromatic hydrocarbons, polychlorinated biphenyls, organochlorinated pesticides, current-use pesticides, personal care products, other pesticides and plastic additives) was characterized for the first 24 h from different polymers to seawater and the remaining content of these contaminants was also extracted by ultrasonic extraction with methanol. All samples were analyzed by Stir Bar Sorptive Extraction coupled to GC/MS. A significant fraction of sorbed contaminants in polymers was desorbed in the first 24 h, particularly for triazines and organophosphorus pesticides due to their lower hydrophobicity than other considered analytes. The remaining contaminants contained in plastics can be also transferred to seawater, sediments or biota. Considering 24 h desorbed fraction plus the remaining methanol extracted fraction, the highest transfer levels corresponded to personal care products, plastic additives, current-use pesticides and PAHs. This is the first study to show the relevance of the transport of organic contaminants on plastic debris from littoral areas to the marine environment.

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1. Introduction

The presence and impact of plastic and microplastics in the marine environment is not a new phenomenon, but in recent years it has received particular attention from the social and scientific communities (Andrady, 2011; Cózar et al., 2015). The presence of plastics in the environment is a consequence of their worldwide use in the majority of human activities (domestic, industry, agriculture, fisheries, etc.) a large proportion of which is as single-use applications that are neither recovered nor recycled. Consequently plastics and plastic debris can reach the marine environment through direct discharges and be transported from continental areas (air, rivers, etc.). Land-based sources, including

beach litter, contribute about 80% of plastic debris (Andrady, 2011). Microplastics are also found in the marine environment due to their direct introduction through runoff, including those used in consumer products, and to the weathering breakdown of larger plastic debris (Andrady, 2011). The most commonly used plastics are polyethylene (PE), polypropylene (PP) and polyethylene terephthalate (PET) and are therefore also those most frequently found in the marine environment (Iñiguez et al., 2017).

Plastic debris causes physical problems in marine fauna, which can confuse it with food (Thomson et al., 2004; Andrady, 2011). Many different organisms ingest plastics and microplastics, including bivalves (Ward and Shumway, 2004), crustaceans (Murray and Cowie, 2011), fish (Boerger et al., 2010; Bellas et al., 2016), marine mammals (Denuncio et al., 2011) and seabirds (Avery-Gomm et al., 2012). The chemical toxicity of microplastics has been also demonstrated on sea urchin (Martínez-Gómez et al., 2017).

[☆] This paper has been recommended for acceptance by Maria Cristina Fossi.

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Plastics also act as passive samplers in continental areas, accumulating hydrophobic organic contaminants (HOCs) which are present in the surrounding environmental compartments (air, water, soil, etc.). Plastic debris may also transport persistent organic pollutants sorbed to these materials to new locations and transfer their chemical components to the surrounding environment (Mato et al., 2001; Teuten et al., 2009; Andrady, 2011; Engler, 2012). However, the low density of some plastics favours their transport not only through water courses but also, and probably more relevantly, by wind transport due to the concentration of human activities in coastal areas. The high accumulation potential suggested that plastic resin pellets serve as both a transport medium and a potential source of toxic chemicals in the marine environment (Mato et al., 2001). The sorption of HOCs to marine plastics may affect the food-web bioaccumulation and bioavailability of HOCs. Several studies have modelled the effect of plastics on the transfer of HOCs to marine organisms (Teuten et al., 2007; Bakir et al., 2016; Paul-Pont et al., 2016; Koelmans et al., 2013, 2014, 2016). The combined intake from food and water was the main route of exposure to DDT, phenanthrene and bis-2-ethylhexyl phthalate, with negligible input from microparticles of polyvinyl chloride (PVC) and PE (Bakir et al., 2016). In this sense polystyrene (PS) microbeads play a minor role in transferring fluoranthene to mussels in comparison with waterborne and foodborne exposures, but direct toxic effects of this plastic were also observed (Paul-Pont et al., 2016). On the other hand positive correlations were found between ingested plastic particles and the concentrations of polychlorinated biphenyls (PCBs) or polybrominated diphenyl ethers in birds (Ryan et al., 1988; Tanaka et al., 2013). However, the ingestion of microplastic by lugworm and cod was expected to result in a marginal contribution to NP and BPA exposure (Koelmans et al., 2014). In this sense a recent critical review suggested that the effects of ingestion of microplastics on bioaccumulation most probably are limited for most marine habitats (Koelmans et al., 2013). Therefore it could be hard to confirm these effects by field data, but it does not imply that plastics do not have deleterious effects on marine life (Koelmans et al., 2016). Further studies are thus required because plastic can accumulate HOCs from air or aquatic environments (Mato et al., 2002) and subsequently transport and transfer them to other compartments. The accumulation of every contaminant depends on the ratio of surface area to volume, the sorption capacity of each material, weathering, the exposure period and its concentration in the surrounding matrix (Endo et al., 2005). In fact, HOCs have a higher affinity for polyethylene than PP (Mato et al., 2002; Karapanagioti and Klontza, 2008), whilst PE, PP and PVC have a greater affinity for these compounds than for natural sediments tested by Teuten et al. (2007). Weathered PE and PP showed higher partition ratios for phenanthrene than unweathered polymers (Karapanagioti and Klontza, 2008). The hydrophobic surfaces of pellets accumulate other compounds in beached plastic pellets, such as DDT and its metabolites, polycyclic aromatic hydrocarbons (PAHs), etc. (Teuten et al., 2009; Frias et al., 2010; Hirai et al., 2011; Van et al., 2012; Zhang et al., 2015). In the majority of studies persistent organic contaminants and PAHs were considered but many more substances can also be sorbed. Moreover, plastics and microplastics can also transfer monomers, base polymers and chemical additives (PBDEs, bisphenol-A, nonylphenol, etc.) contained in the different materials (Teuten et al., 2009). In fact, two recent non-targeted screening analyses looking at the chemicals associated with plastic debris detected a total of 231–251 organic compounds on plastics, including hydrocarbons, UV-stabilizers, anti-oxidants, plasticizers, flame retardants, lubricants, intermediates and compounds for dyes and inks (Gauquie et al., 2015; Rani et al., 2015).

However, to the best of the authors' knowledge no previous

information is available regarding the transfer of HOCs from littoral/continental plastics to coastal areas. Furthermore, the majority of previous studies have mainly focused on the sorption of PAHs, organochlorinated contaminants or plastic additives desorption in seawater (Mato et al., 2001; Teuten et al., 2009), and recently on the sorption of carbamazepine, 4-methylbenzylidene camphor, triclosan and 17 α -ethinyl estradiol to PE (Wu et al., 2016), but no data are available in relation to contaminants of emerging concern such as current-use pesticides (CUPs) and other personal care products (PCPs). In this study the occurrence of plastics is characterized in several areas from the perimeter of a Mediterranean coastal lagoon and the transfer of 91 HOCs (PAHs, PCBs, OCPs, CUPs, PCPs, other pesticides and plastic additives) from different polymers (materials, weathering and origin) to seawater is estimated by stir bar sorptive extraction (SBSE) coupled to thermodesorption unit and GC/MS (Moreno-González et al., 2013). More specifically, the desorption of these pollutants from littoral plastics and microplastics to seawater was characterized for the first 24 h and the remaining organic contaminants content in these materials was also determined by MeOH ultrasonic extraction, as an indicator of the maximum desorbable amount over time and their potential transfer from plastics to the marine environment (seawater, sediments or biota).

2. Materials and methods

2.1. Materials

Seawater used for the preparation of seawater standards and seawater transfer experiments was obtained 10 miles from the coast and filtered (0.22 μ m). The commercial polydimethylsiloxane stir bars employed (Gerstel, Mulheim a/d Ruhr, Germany) were 20 mm \times 0.5 mm. A 15 position magnetic stirrer (Gerstel, Mulheim a/d Ruhr, Germany) was used to stir samples at 750 rpm.

OPPs were purchased from Ultra Scientific (Rhode Island, USA). Standards containing organochlorinated pesticides, PCBs, PAHs, triazines and individual standards of the remaining analytes were purchased from Dr. Ehrenstorfer GmbH (Augsburg, Germany). The complete list of analyzed substances is shown in Table S1 (Supplementary Material section). Atrazine-D5, deuterated PAH mix, triclosan-D10 and chlorpyrifos-D10 were obtained from Dr. Ehrenstorfer (Augsburg, Germany).

2.2. Study area and sampling campaign

The Mar Menor is a Mediterranean coastal lagoon located in southeast Spain. This lagoon is subject to heavy seasonal tourist activity and urban development and is surrounded by Campo de Cartagena, an intensive agricultural area where the use of plastic has greatly increased in recent decades. All these activities are relevant sources of plastic debris in littoral areas.

Plastic and microplastics were sampled in Spring 2015 from 5 coastal areas adjacent to the Mar Menor lagoon (P1–P5, Fig. 1) exposed to a variety of predominantly anthropogenic activities (tourist, agricultural, urban, etc.). Plastic debris were randomly sampled by hand at five coastal areas (P1–P5) in order to obtain a sufficient mass of plastic (0.2–2.0 g) for the analysis of regulated and emerging organic contaminants, being mainly macroplastics. These macroplastic samples corresponded to continental debris transported by air from surrounding areas or deposited directly by tourists in the littoral. Attending to the environmental conditions of the study area and the characteristics of these samples we can consider that they were not previously immersed in seawater and accumulated environmental contaminants mainly from air. Consequently they can be used to simulate the desorption and

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