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Contamination in plastic recycling: Influence of metals on the quality of reprocessed plastic

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ABSTRACT

The global consumption of plastic continues to increase, and plastic recycling is highlighted as crucial for saving fossil resources and closing material loops. Plastic can be made from different polymers and contains a variety of substances, added intentionally to enhance the plastic's properties (metals added as fillers, colourants, etc.). Moreover, plastic can be contaminated during use and subsequent waste management. Consequently, if substances and contaminants are not removed during recycling, potentially problematic substances might be recycled with the targeted polymers, hence the need for quantitative data about the nature and presence of these substances in plastic. Samples of selected polymers (PET, PE, PP, PS) were collected from different origins; plastic household waste, flakes/pellets of reprocessed plastic from households and industry, and virgin plastic. Fifteen selected metals (Al, As, Cd, Co, Cr, Cu, Fe, Hg, Li, Mn, Ni, Pb, Sb, Ti, Zn) were quantified and the statistical analysis showed that both the polymer and origin influenced the metal concentration. Sb and Zn were potentially related to the production stage of PET and PS, respectively. Household plastic samples (waste and reprocessed) were found to contain significantly higher Al, Pb, Ti and Zn concentrations when compared to virgin samples. Only the concentration of Mn was reduced during washing, suggesting that parts of it was present as physical contamination. While most of the metals were below legal limit values, elevated concentrations in reprocessed plastic from households, aligned with increasing recycling rates, may lead to higher metal concentrations in the future.

1. Introduction

Continuously increasing resource consumption in modern societies, where resources are often lost after the first life cycle, places pressure on the finite resources of the globe. As a measure to partially mitigate this, and to ensure more sustainable development, the concept of a circular economy, where resources are recirculated into society after being used, has been adopted by many legal authorities (e.g. EC, 2015; Government of Japan, 2010). The European Union (EU) has presented a strategy towards a circular economy that has a strong focus on material flows and subsequent recycling, to keep materials in the loop (EC, 2015). In the strategy, plastic has been highlighted as a priority area, since it is an important material for modern societies, is used in large quantities and is predominantly made from fossil fuels. Consequently, the strategy dictates increases in European recycling rates for both plastic packaging and plastic from household waste (HHW). In addition, the EU has developed a circular economy strategy solely for plastic, further stressing the importance of product design and high-quality recycling (EC, 2018a).

Plastic waste is a complex and heterogeneous material stream, due to several factors. First, plastic as material refers to numerous different polymers with different chemical properties that need to be separated from each other prior to recycling. The main polymers found in plastic from HHW are polyethylene terephthalate (PET), polyethylene (PE), polypropylene (PP) and polystyrene (PS) (Edjabou et al., 2015), collectively representing 63.2% of the plastic demand in Europe (PlasticsEurope and EPRO, 2017). Second, many different additives can be introduced during the production phase to control the properties of the plastic and make it fulfil the requirements for use in specific applications. These include additives such as colourants, fillers, plasticisers, lubricants, antioxidants (Hahladakis et al., 2018), commonly used in plastic

Abbreviations: FOD, frequency of detection; HDPE, high density polyethylene; HHW, household waste; IW, industrial waste; LOD, limit of detection; MANOVA, multivariate analysis of variance; PE, polyethylene; PET, polyethylene terephthalate; PP, polypropylene; PS, polystyrene.

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packaging (Lahimer et al., 2013), as well as additives such as flame retardants, commonly used in plastic for electronics (Hansen, 2013). While plastic packaging represents a significant share of plastic in HHW (Edjabou et al., 2015), waste electronics end up in HHW primarily due to miss-sorting (Edjabou et al., 2015). The type and content of these additives is regulated to varying degrees when it comes to use in specific applications. As an example, plastic used in food packaging needs to comply with the most strict and comprehensive legislation with respect to chemical composition and migration of potentially problematic substances (EU, 2011b). Consequently, plastic applicable for food contact is in this study defined as "high-quality". Thereby, this term refers to the potential applicability (and "circularity" of the plastic) with respect to legal requirements for chemical composition, rather than the physical and mechanical properties of the material (melt flow index, impact strength, etc.). In contrast to high-quality plastic, "low-quality" is used to characterize plastic applicable only for applications with less strict requirements in relation to chemical composition or migration (electrical and electronic equipment, non-food packaging, etc.). Hence, plastic in such applications might contain higher concentrations of potentially problematic substances. Consequently, the chemical properties and the quality of the plastic can vary depending on the specific product and its application. Third, substances can be added non-intentionally, either in the production phase (e.g. residues from catalysts, metal impurities in non-metal additives (Lahimer et al., 2013)) or as contamination through potential sorption during use and waste management (Pivnenko and Astrup, 2016). Some contaminants might be chemically embedded in the plastic matrix rather than being present as physical contamination ("dirt") that can be removed during recycling, e.g. during washing of the plastic waste. Consequently, there is a risk of recycling not only the desired plastic material, but also potentially problematic substances, ultimately affecting the applicability and quality of the reprocessed plastic material. This phenomenon has been demonstrated quantitatively for other materials (e.g. paper (Pivnenko et al., 2016a)), and several sources have underlined the importance of a "clean" circular economy in relation to plastic recycling (Ellen MacArthur Foundation, 2016; Goldberg, 2017).

In regards to available literature on plastic from households, the most analysed items are PET water bottles approved for food contact, with both the total content and migration of potentially problematic substances being studied. The findings are summarised in a review by Bach et al. (2012) which reveals that several substances have been shown to migrate from PET bottles into bottled water, such as Sb, acetaldehylde and selected phthalates, despite the fact that e.g. phthalates are not added intentionally during PET plastic production. However, the migration never exceeded either European or American limit values. PET bottles for mineral water or carbonated drinks represent a relatively homogeneous material stream in which all the items are similar in shape and design and comply with strict legislation related to food packaging. Streams of mixed plastic waste, however, are much more heterogeneous and contaminated (Ragaert et al., 2017), and not all items have to meet the aforementioned strict legislation. Nonetheless, very few studies have chemically analysed HHW streams including plastic items other than bottles, or plastic recovered from HHW and reprocessed into pellets or flakes, representing recycled raw plastic material. Camacho and Karlsson (2001) analysed samples of high-density PE (HDPE) and PP waste sorted from mixed HHW for low-molecular weight organic substances. They identified the presence of several substances related to cosmetics, cleaning agents and personal hygiene in the HDPE waste that was not present in virgin HDPE (including selected acids, esters and alcohols). Moreover, Pivnenko et al. (2016b) found that the concentration of selected phthalates was higher in reprocessed plastic and plastic from HHW, compared to virgin plastic and reprocessed plastic from industrial waste (IW). Similarly, Huber and Franz (1997) found high concentrations of phthalates in reprocessed HDPE made from HHW bottles. All the literature sources mentioned above suggest that plastic from HHW might contain contaminants with the potential to impair the quality of recycled plastic material.

Most studies focused on the presence or migration of organic substances as contaminants. While organic substances may degrade or migrate during use and recycling, inorganic substances, such as metals, are in most cases expected to persist in the material after recycling (Hansen, 2013), though small amounts might migrate during use (Whitt et al., 2016; Bach et al., 2012). Several metals are currently intentionally added during plastic production (often as oxides, acids, etc. (Hahladakis et al., 2018; Hansen, 2013)). These include additives such as colourants (containing Ti, Cr, Co, Cd, Pb, Zn, Fe, Al, Cu), antioxidants and stabilisers (containing Cd, Pb, Zn) or other additives (containing As, Li, Pb, Cd, Zn, Sb, Al) (Hahladakis et al., 2018; Hansen, 2013). Moreover, metals in plastic can originate from catalysts used in plastic production (e.g. Sb, Ti, Cr, Hg, Mn), or contamination added or sorbed to the plastic during production, use and waste management (e.g. Fe, Al, Cu, Mn, Zn, Ni) (Hahladakis et al., 2018; Hansen, 2013; Bach et al., 2012; Romão et al., 2010; EC, 2008). As most of these metals have well-documented toxic effects and/or can be classified as persistent and bioaccumulative (Goldberg, 2017; EC, 2008), it is desirable to reduce recycling of metals in plastic, in order to minimise potential health risks and deterioration of material quality. Where the form of the metals, their hazardousness and exposure influence the potential risk to human health and the environment, the total metal content can be used to identify potential deterioration of material quality, which affects the applicability of the recycled plastic and thereby the circularity (Eriksen et al., 2018). Currently very limited knowledge exists about the fate of metals during plastic recycling, and metal contamination in plastic has been assessed previously only by focusing on one single source of plastic, polymer or metal (Götze et al., 2016; Bach et al., 2012; Romão et al., 2010). Consequently, there is a need to quantify and document the total metal content in conjunction with the plastic material quality, rather than potential health risks, as a first step towards coherently assessing the metal content in plastic collected from various steps in the recycling chain.

The aim of this work was to provide consistent quantitative data on the presence of selected metals in plastic samples of different origins and polymer types with the purpose of identifying potential deterioration of material quality and applicability. The waste plastic samples were further divided into food and non-food contact items, as the substances added to the plastic are expected to be different for the two product groups due to stricter legislation for food contact items. Finally, the impact of pre-treatment through washing was assessed on the waste samples. The differences in metal concentrations between the sample groups were analysed statistically and the effects of metal concentration on the applicability and quality of the reprocessed plastic were evaluated. This was achieved through the following specific objectives: 1) obtain samples of relevant polymer types (PET, PE, PP and PS) from different origins; plastic from household waste (HHW), reprocessed plastic waste (pellets or flakes) from households and industry as well as virgin plastic; 2) pre-treat waste plastic samples for analysis, including separation into food and non-food contact items, sample shredding and washing; 3) analyse the concentration of selected metals (Al, As, Cd, Co, Cr, Cu, Fe, Hg, Li, Mn, Ni, Pb, Sb, Ti, Zn) in all samples; 4) assess statistical differences in metal concentrations between the different sample groups; and 5) evaluate levels of metal contamination and their potential influence on the applicability of reprocessed waste plastic in material recycling.