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## Waste Management

journal homepage: [www.elsevier.com/locate/wasman](http://www.elsevier.com/locate/wasman)

# Physico-chemical properties of excavated plastic from landfill mining and current recycling routes

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## ARTICLE INFO

## Article history:

Received 24 July 2017

Revised 20 March 2018

Accepted 27 March 2018

Available online xxxx

## Keywords:

Enhanced landfill mining

Recycling

Excavated plastics

Pyrolysis

## ABSTRACT

In Europe over 5.25 billion tonnes of waste has been landfilled between 1995 and 2015. Among this large amount of waste, plastic represents typically 5–25 wt% which is significant and has the potential to be recycled and reintroduced into the circular economy. To date there is still however little information available of the opportunities and challenges in recovering plastics from landfill sites. In this review, the impacts of landfill chemistry on the degradation and/or contamination of excavated plastic waste are analysed. The feasibility of using excavated plastic waste as feedstock for upcycling to valuable chemicals or liquid fuels through thermochemical conversion is also critically discussed. The limited degradation that is experienced by many plastics in landfills (>20 years) which guarantee that large amount is still available is largely due to thermooxidative degradation and the anaerobic conditions. However, excavated plastic waste cannot be conventionally recycled due to high level of ash, impurities and heavy metals. Recent studies demonstrated that pyrolysis offers a cost effective alternative option to conventional recycling. The produced pyrolysis oil is expected to have similar characteristics to petroleum diesel oil. The production of valuable product from excavated plastic waste will also increase the feasibility of enhanced landfill mining projects. However, further studies are needed to investigate the uncertainties about the contamination level and degradation of excavated plastic waste and address their viability for being processed through pyrolysis.

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**Abbreviations:** ABS, acrylonitrile–butadiene–styrene; APCr, air pollution control residues; BOD, biological oxygen demand; BPA, bisphenol A; COD, chemical oxygen demand; DEHP, diethyl–hexyl phthalate; DMP, dimethyl phthalate; DoU, degree of unsaturation; DTG, differential thermogravimetric;  $E_a$ , activation energy; ELFM, enhanced landfill mining; FCC, fluid catalytic cracking; GC–MS, gas chromatography–mass spectrometry; GCV, gross calorific value; HDPE, high-density polyethylene; HOCs, hydrophobic organic contaminants; IW, industrial waste; LDPE, low-density polyethylene;  $M_n$ , number average of molecular weight; MBT, mechanical biological treatment; MSW, municipal solid waste; NCV, net calorific value; NP, nonylphenols; PA, polyamide; PAHs, polycyclic aromatic hydrocarbons; PC, polycarbonate; PCDFs, polychlorinated dibenzofurans; PE, polyethylene; PET, polyethylene terephthalate; PMMA, polymethyl methacrylate; PP, polypropylene; PS, polystyrene; PTFE, polytetrafluoroethylene; PU, polyurethane; PVC, polyvinyl chloride; RDF, refused derived fuel; SD, standard deviation; SEM, scanning electron microscopy; SRB, sulphate-reducing bacteria; TDPA, totally degradable plastic additives; TGA, thermogravimetric analyser; TOC, total organic carbon; VOCs, volatile organic compounds; WtE, waste to energy; WtM, waste to material; XRF, X-ray fluorescence.

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## 1. Introduction

Over the last two decades, the amount of waste being managed by landfill disposal each year has decreased across Europe and the UK. According to the Eurostat (2016) waste estimation, 473 kg per capita of MSW was generated in 1995 in EU-27, of which 64 wt% were disposed in landfill and 11 wt% were recycled. In 2015, the EU-27 reported 477 kg of MSW per capita of which 28 wt% were recycled, 26 wt% incinerated (including energy recovery), 25 wt% landfilled, and 16 wt% were processed by composting and digestion (Eurostat, 2016). New legislation, such as Council Directive 1999/31/EC (European Parliament. Council of the European Union, 1999) and Waste Framework Directive 2008/98/EC (European Parliament. Council of the European Union, 2008), has driven the changes in the management of landfill, encouraged sustainable waste management and resulted in the closure of many landfills (Hogland et al., 2011). The waste management hierarchy included in Framework Directive 2008/98/EC has become part of the European waste management, defining different aspects of this topic such as waste, recycling, recovery, secondary raw materials and by-products (European Commission, 2016).

Recently, Europe has moved towards the ‘new’ concept of a Circular Economy, aiming to recycle 65 wt% of MSW and reduce the amount of MSW disposed in landfill by 10 wt% before 2030 (European Parliament. Council of the European Union, 2015). However, there are between 125,000 and 500,000 landfills (EURELCO, 2017) in Europe, many of which are now closed; waste in landfills represents an important legacy that needs to be addressed. It is estimated that over 5.25 billion tonnes of waste were deposited in landfills between 1995 and 2015 across the EU-27 countries (Eurostat, 2016). The first landfill mining (LFM) project was carried out in Israel in 1953 (Savage et al., 1993), but only until the late 1980s, interest began to increase, especially in USA and Europe (Hogland et al., 2004). The concept of enhanced landfill mining (ELFM), which started to develop in 2008 (Jones et al., 2013), focuses on maximising the valorisation of waste found in landfills and dump-sites as material (WtM) and energy (WtE) (Jones et al., 2012). Landfills can be considered as temporary storage for waste while the technologies for their valorisation are improved and achieve large-scale deployment (Bosmans et al., 2013). Landfills operating between the 1950s to the mid-1990 s have been identified as the most suitable for ELFM, because they were not affected by the directives that lead to a minimization and pre-treatment of waste disposed in landfills and have higher content of valuable and combustible materials (Hogland et al., 2011; Van Passel et al., 2013).

A key challenge exists in the recovery of value from materials excavated from landfills, which has been partially addressed in previous academic publications, however this review specifically focuses on plastics. Here there is an opportunity to explore alternative methods of recovering value from plastics as conventional recycling/recovery methods will not be viable. Similarly, recovery of energy from waste and advanced conversion processes require further research and development due to the pollution and the unknown effects of landfill contamination on the chemical transformation pathways.

A variety of the landfilled materials can theoretically be recycled or used for energy recovery, which can contribute to the secu-

rity of energy supply and substitute raw materials (Greedy, 2016). For example, recovery of secondary raw materials available within landfills such as valuable metals (Gutiérrez-Gutiérrez et al., 2015) can mitigate the increasing concern about the availability and security of critical raw materials (European Commission, 2017). Opportunities also exist in the recovery of plastics, which represent between 5–25 wt% of the total waste deposited; the proportion increases in landfills during time due to the degradation of organic matter and its consequent weight loss (García et al., 2016; Sel et al., 2016; Münnich et al., 2015; Quaghebeur et al., 2013; Jones et al., 2013; Van Passel et al., 2013; Van Vossen and Prent, 2011).

The annual worldwide plastic production has increased from 1.5 Mt in the 1950 s to 322 Mt in 2015 (PlasticsEurope, 2016). In 2014 59 Mt and 311 Mt of plastics were generated in Europe and in the world respectively (PlasticsEurope, 2016). In the same year, of 25.8 Mt of plastic waste produced, 29.7 wt% were recycled, 39.5 wt% used for energy recovery and 30 wt% were landfilled (PlasticsEurope, 2016). Over the years, the inadequate plastic waste management has led to the accumulation of plastics in the environment, causing pollution and consequent health risks (Singh and Ruj, 2016; Thompson et al., 2009). The conjunction of increasing energy demand and scarce resources such as fossil fuel has resulted in a need for sustainable secondary fuels and chemical resources (Sharma et al., 2014; Singh and Ruj, 2016). Plastics from landfills can potentially be reprocessed to other plastic products, used as part of a waste-derived fuel for energy or used as a feedstock to produce valuable base petrochemicals (Al-Salem et al., 2009; Mastellone, 1999). Because 90% of the plastic are produced from petroleum, pyrolysis of plastic waste is considered a feasible process to recover chemical building blocks and a valuable alternative to the ordinary plastics disposal routes, such as landfill (Al-Salem et al., 2009; Al-Salem and Lettieri, 2010).

Critical reviews and studies on technical and economic aspects of LFM and ELFM has been previously published (Krook et al., 2012; Jones et al., 2013; Bosmans et al., 2013; Van Passel et al., 2013). However, these do not focus on the excavated plastic waste fraction and therefore do not consider its chemical characteristics. This paper reviews the research focusing on excavated plastics and the physico-chemical properties of this fraction along with the gaps in scientific knowledge that need to be filled to consolidate and enable development of upcycling technologies. The aim of the work is to critically review the likely impacts of landfill chemistry on the degradation and/or contamination of plastic waste and its properties, and assess the viability of using excavated plastic waste as feedstock for upcycling to valuable chemicals or liquid fuels via thermochemical conversion.

## 2. Plastic components of landfill waste and factors affecting their degradation

### 2.1. Plastic components of landfill waste

Plastics can be thermoplastic, which are capable of melting and flowing at a certain temperature without undergoing chemical changes, and thermoset such as bakelite, which are characterised by irreversible cross-linked polymer chains formed at high-temperature treatments (Jasso-Gastinel et al., 2017). The molecular

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