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Research Paper

Unlocking circular economy potential: Evaluating high-value chemical production from pyrolysis of plastics recovered from landfilled municipal solid waste☆

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ABSTRACT

This study evaluates the potential for producing valuable chemical products from plastics recovered from municipal solid waste (MSW) landfills and therefore contributing to waste reduction and resource recovery. Using pyrolysis–gas chromatography-mass spectrometry (Py-GC–MS) we analysed the decomposition products of 10 plastic samples including fresh and excavated samples of different landfill periods from 4 landfill sites. The samples comprised polyethylene (PE), polypropylene (PP), two mixed excavated plastic materials containing PE, PP, polystyrene (PS), poly(ethylene terephthalate) (PET) and poly(vinyl chloride) (PVC). All samples were pyrolyzed at 350, 500, 650 and 800 °C to semi-quantitatively identify the range of compounds produced at each temperature. Hydrocarbon production was the highest at 500 °C and 650 °C whit a significant proportion of the products falling within the naphtha range (C6-C10), which is a critical feedstock in the plastic industry. Notably, naphtha can be cracked to produce 55 wt% high-value chemicals, such as ethylene and propylene. Additionally, aromatic compounds such as benzene, toluene and styrene, especially in the mixed samples demonstrate potential for use in the chemical industries for polymers, solvents and dyes. These results provide encouraging evidence that excavated landfill plastics can serve as a viable source of valuable chemical intermediates, supporting both resource recovery and the advancement of a circular plastics economy. While this study offers an important initial assessment, primarily qualitative and focused on single polymers and controlled artificial mixtures, it establishes a solid foundation for future research. Further investigations using heterogeneous and larger-scale processing are recommended to fully demonstrate the techno-economic feasibility and environmental benefits of integrating pyrolysis of aged plastics into sustainable waste management and resource recovery strategies.

1. Introduction

The yearly global production of plastic has grown from 1.5 million tonnes in 1950 to 400 million tonnes in 2022, with half of all plastics produced designed for single use (PlasticsEurope, 2024; PlasticsEurope, 2016). It has been estimated that more than 4.98 bn tonnes of plastics ended up either in landfill or in the natural environment (Geyer et al., 2017). Despite a decline in fossil-based plastic production since 2018, over 80 % of plastics continue to be manufactured using chemicals derived from fossil fuels (PlasticsEurope, 2024). In Europe, over 400,000 municipal solid MSW landfill sites have been reported (EURELCO, 2018) and the average percentage of plastic ranged between 9 and 25 wt% (Quaghebeur et al., 2013; Zhou et al., 2014; Jones et al., 2013; Wagland et al., 2019; Canopoli et al., 2020). This amount of plastic can potentially be recycled through enhanced landfill mining (ELFM) and can be reintroduced into the market sector, therefore, embracing the circular economy concept.

The Global Commitment launched in 2018 by the Ellen MacArthur

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Foundation in collaboration with UN Environment Programme, promotes a circular plastics economy focused on the production, consumption and recycling of plastics to prevent them from becoming waste (Ellen MacArthur Foundation and UN Environment Programme, 2019). However, significant challenges exist in upgrading excavating plastics into sustainable resources. Plastics buried in landfills for several years undergo weathering processes that alter their chemical and mechanical properties, increasing impurities such as soil, surface alteration, higher oxidation level, silicon and aluminium content, and a higher degree of crystallinity (Quaghebeur et al., 2013; Zhou et al., 2014; Canopoli et al., 2020). Considering these challenges, pyrolysis technologies have been identified as a promising technology for converting recovered landfill plastics into valuable chemicals and transportation fuels (Canopoli et al., 2020; Du et al., 2024; Rahman et al., 2023; Kumar et al., 2023). While most studies have focused on the pyrolysis of virgin plastics, few have explored the pyrolysis of plastics recovered from landfills (Bosmans et al., 2014; Breyer et al. 2017). This study aims to address the knowledge gap regarding the effect of polymer ageing on pyrolysis performance by investigating the pyrolysis behaviour of specific polymers (PE, PP, PS, PET, and PVC) recovered from excavated landfill waste. Rather than analysing the pyrolysis of mixed excavated waste as a whole, the research focuses on how the degree of ageing, linked to the time these polymers have been buried, affects their thermal degradation pathways and the yield of valuable products, particularly small-chain hydrocarbons (<C10). It is hypothesised that prolonged environmental exposure in landfills leads to degradation of polymer chains, which may reduce the efficiency of producing desirable petrochemical feedstocks via pyrolysis. Understanding this relationship for individual polymers provides insight into the feasibility and optimisation of pyrolysis processes for converting aged, excavated plastics into high-value chemicals, and informs the design of future large-scale plastic recovery and upcycling strategies within a circular economy.

2. Materials and methods

2.1. MSW samples, plastic materials reference and characteristics determination

A total of 154 kg of waste, in 30 samples, were excavated from four MSW landfills at depths of between 5 and 55 m (see supplementary data Table S1). The non-landfilled plastic waste samples were collected from domestic waste. The samples from landfills appeared generally wet with much of the soil and fine fraction, primarily degraded organic matter, dirt, and woody material, attached to the plastic samples. The plastic samples were pre-dried in an oven, at 60 °C for 2 h, before the identification of plastic types and to easily separate the soil and fine fraction from their surface. Seven plastic material types were identified using an attenuated total reflectance-Fourier transform infrared spectroscopy (ATR-FTIR) and the main fractions with the highest percentages (more details are given in supplementary data Table S1 and Canopoli et al., 2020). The FTIR spectra were recorded over 16 scans at a spectral resolution of 4 cm^{-1} in a wavenumber range of 4,000 to 400 cm^{-1}. After the identification, the plastic materials were manually washed with cold water and air-dried at room temperature. The gross calorific value (GCV), also known as the higher heating value (HHV), refers to the total amount of heat released when a unit mass of a fuel sample is completely combusted in oxygen. This value includes the latent heat of vaporization of water formed during combustion. In this study, the GCV was determined by the combustion of ~1 g of the sample in a bomb calorimeter (British Standards Institute, 2011e). About 1 g of sample was placed inside a crucible with a lid and this was placed in a furnace at 600 °C for 7 min to measure the volatile matter (Zhou et al., 2014). Ultimate (elemental) and metals analyses were carried out following the British Standards methods (2011b, 2011c, 2011d). The samples of fresh and excavated polyethylene (PE) and polypropylene (PP) were analysed in triplicate.

2.2. Plastic materials stability using thermogravimetric analysis

The recovered plastic with more than 10 years of storage presented a generally higher degree of degradation that can affect their thermal decomposition (Canopoli et al., 2020). Thermogravimetric analysis (TGA) was used to quantitatively identify differences in the thermal decomposition of plastic samples of the same type but with different storage ages. This analysis helped to evaluate if the characteristics of excavated plastics affect their thermal behaviour, which could have consequences on the pyrolysis products. PE and PP, fresh and excavated, <10 years and >10 years, were thermally characterised in duplicate using TGA. PE samples were mainly represented by soft plastic, such as plastic bags, while PP was, for the majority, hard pieces of plastics. Due to this difference in density, the PE and PP samples studied were 5 mg and 10 mg, respectively, to maintain a similar surface area. A sample mass range of between 5 and 10 mg is commonly used for TGA of plastic samples (Sørum et al. 2001; Klein-Bendavid et al., 2014; Gallo and Severini, 2017). The samples were placed in a ceramic crucible, heated at 10 °C/min from 50 °C to 600 °C in a nitrogen atmosphere with a flow rate of 40 mL/min (Breyer et al., 2017). The first derivative of the weight loss was calculated from the TGA results to identify the temperature of greatest weight loss. The data reproduced in this paper are the mean values of duplicate runs per each sample.

2.3. The identification of pyrolysis compounds from excavated plastics by pyrolysis-gas chromatography-mass spectrometry

The fresh and excavated plastic waste were pyrolyzed using a Pyrola 2000 (PyroLab AB, Sweden) connected to a quadrupole gas chromatograph-mass spectrometer (GC–MS) (Agilent Technologies 7890A GC − 5975c inert xl EI/CI MSD). A sample between ~150 and 200 μg for a single plastic and between ~300 and 550 μg of mixed plastics, was used for each analysis.

In the Pyrola, a sample is placed on a resistively heated platinum filament, where the pyrolysis of the sample takes place. The filament is surrounded by a glass cell. Temperature is one of the main parameters during plastic pyrolysis because it highly affects the kinetics of the reactions taking place and therefore the final product composition (Anuar Sharuddin et al., 2016). The Pyrola 2000 measures the temperature in two different ways, increasing the accuracy and reproducibility. The exact temperature the sample has reached during pyrolysis is recorded as a temperature–time profile (TTP). Above 600 °C measurement is taken by a highly accurate photodiode, while the resistance of the filament is used for temperatures below 600 °C. A calibration was performed at the 26 mL/min flow rate through the cell on the filament used for the sample analysis to determine the current required to reach and hold the pyrolysis temperatures that would be used for the sample analysis. The platinum filament and glass cell were cleaned after each sample by heating them with a micro-torch. Helium was used as a carrier gas with a flow rate of 26 mL/min through the glass cell when pyrolysis was taking place. To assess the variance in product composition at different temperatures, fractionated pyrolysis was performed for 2 s at each temperature, with a sequential temperature rise time of 8 ms, at each temperature: 350, 500, 650 and 800 °C (Westphal et al., 2001; Evangelopoulos et al., 2015). The rapid analysis through the GC column reduces the likelihood of secondary reactions and the formation of aromatic compounds as non-condensable compounds cool. The Pyrola 2000 chamber temperature was set isothermally at 200 °C to transfer volatile products through the needle and into the GC inlet, while involatile products condensed inside the glass cell.

The GC inlet was a standard Agilent split/splitless inlet, installed with a 2 mm internal diameter empty liner. Temperature between the pyrolyzer and the GC inlet was held isothermally at 290 °C (Jin et al., 2016). The GC separation column was an Agilent HP5-MS UI (30 m × 0.25 mm × 0.25 μm) (Aguado et al., 2007; Jin et al., 2016). A GC inlet split ratio of 20:1 was used. The GC oven was programmed from 30 °C (held for 0.5 min) to 350 °C (held for 1.5 min) at 10 °C/min. The running time for the GC was 34 min (Jin et al., 2016; Sophonrat et al., 2017). The MS operated under electron ionisation (EI) of 70 eV with a mass spectral range of between 33 and 500 *m/z*. The National Institute of Standards and Technology (NIST) library was used to support the identification of products.

Fresh and excavated PE and PP (<10 years and >10 years) were pyrolyzed in triplicate. In addition to the single polymer runs, two different mixes of excavated plastic samples were pyrolyzed. The compositions of samples A and B were designed as model mixtures to systematically study the pyrolysis behaviour of the dominant polymer types identified in the excavated waste. Sample A (PS, PE, PP in equal proportions) represents a simplified mix of the most abundant commodity plastics, enabling the investigation of thermal degradation interactions among polyolefins and styrenic polymers under controlled conditions. Sample B (PET, PVC, PE, PP in equal proportions) includes both polyolefins and more challenging polymers such as PET and PVC, which are known to introduce additional reaction pathways and contaminants (e.g., HCl evolution from PVC). While these mixtures do not replicate the exact proportions found in the excavated waste, they serve as representative models to elucidate the influence of mixed plastic compositions on product distribution and yield, complementing the single polymer experiments and providing insights relevant for real mixed waste streams. The pyrolysis of PVC and PET produces undesirable products such as hydrogen chloride and benzoic acid (Anuar Sharuddin et al., 2016). In addition, pyrolysis of PET and PVC produce lower oil yield than the other mentioned plastic type (Singh et al. 2019; Anuar Sharuddin et al., 2016). However, some pyrolysis plants accept a low level of PVC and PET. For these reasons, the pyrolyzed mixed samples were firstly divided in the mix with and without excavated PVC and PET to evaluate their effects in the produced compounds. Then, the different percentages were tested to identify the possible impact on the products from the excavated plastics pyrolysis.

Pyrolysis-gas chromatography-mass spectrometry (Py-GC–MS) was used for the semi-quantitative analyses of the products from recovered plastics from landfill. The repeatability was calculated from the relative abundance of compounds identified (more details available in the supplementary data Table S2-3). The average relative standard deviation is 9 (±6) % which is in the acceptance range (Hermabessiere et al., 2018).

The pyrolysis experiment can be summarised as follows: the sample was reduced in particle size less than 1 mm following the British Standards method (2011f). The shredded sample was placed on the filament within the pyrolysis chamber and helium was used to purge the chamber at a flow rate of 26 mL/min. For each sample analysis, the chamber temperature was incrementally raised to 350 °C, 500 °C, 650 °C and 800 °C. In fractionated pyrolysis, the sample is heated multiple times each at a higher temperature. After each heating which lasts 2 s, the chamber temperature is lowered to 200 °C while the products are analysed by GC–MS. The relative quantity of volatiles produced at each temperature was identified using the National Institute of Standards and Technology (NIST) library version 2014.

3. Results and discussion

3.1. Sample characterisation and thermal degradation stability of PE and PP

Detailed analyses were performed for PE and PP, fresh and excavated (Table 1). In general, PE and PP fresh and excavated, presented a high calorific value (average value 39 wt%). PP < 10 years of storage time showed the lowest calorific value with 28 MJ/kg and the highest oxygen content (10 wt%). The elevated oxygen content in fresh PE could indicate degradation, as well as the presence of additives or contaminants that contain oxygen (Canopoli et al., 2020). The oldest samples, PE > 10 and PP > 10 years of storage, had lower volatile matter than the rest of the samples. The metals analysis (Table 1) revealed a lower content when compared to the results for mixed excavated plastics analysis from other studies (Quaghebeur et al., 2013; Prechthai et al., 2008). In the other studies, the high metals content is likely to be related to the soil attached to the plastics.

Table 1
Analyses of fresh and excavated PE and PP samples.

	PE	PE < 10	PE > 10	PP	PP < 10	PP > 10
Volatile matter (wt %)	98.7 (0.1)	94.5 (0.4)	93.6 (0.2)	98 (0.2)	90.9 (1.0)	88.5 (2.2)
GCV (MJ/kg)	40.4 (0.3)	42.3 (1.5)	42.1 (1.1)	45.7 (0.6)	42.3 (4.7)	36.3 (0.8)
C (wt%)	72.9*	82.6*	79.6*	84.2*	70.9	81.7
N (wt%)	0.3*	0.3*	0.5*	0.2*	0.1	0.2
H (wt%)	11.9*	13.6*	13.1*	13.6*	11.1	13.3
O (wt%) [a]	7.3	0	1.9	0	10	0
S (wt%)	0.006	0.12	0.13	0.024	0.210	0.094
Cl (wt%)	0.024	0.26	1.72	0.004	0.016	0.025
Br (wt%)	0	0.022	0	0	0	0
F (wt%)	0	0.004	0.002	0.001	0.001	0.001
As (mg/kg)	<1*	<1*	<1*	<1*	1.8	2.3
Cd (mg/kg)	<1*	<1*	<1*	<1*	<1	<1
Co (mg/kg)	<1*	<1*	<1*	<1*	<1	<1
Cr (mg/kg)	<1*	<1*	2.2*	1.6*	2.8	2.6
Cu (mg/kg)	11*	8.1*	5.9*	4.8*	30	21
Hg (mg/kg)	<1*	<1*	<1*	<1*	<1	<1
Mn (mg/kg)	4.9*	3*	1.3*	2.7*	2.5	<1
Ni (mg/kg)	<1*	<1*	<1*	<1*	2.9	4.2
Pb (mg/kg)	1.1*	3.2*	6.7*	5.1*	11	8.8
Sb (mg/kg)	<1*	<1*	<1*	<1*	<1	<1
Sn (mg/kg)	<1*	<1*	<1*	<1*	2.6	1.6
Tl (mg/kg)	<1*	<1*	<1*	<1*	<1	<1
V (mg/kg)	<1*	<1*	<1*	<1*	<1	<1

[a] Calculated by difference.
* Adapted from Canopoli et al., 2018b; polyethylene (PE) and polypropylene (PP) with different storage time (<10 years and > 10 years); PE and PP: fresh polymers. (SD): Standard deviation.

TG analysis was performed to compare the degradation temperature ranges of fresh and excavated PE and PP from different years of storage. The average maximum weight loss for fresh PE, PE < 10, PE > 10 was at 491 °C, 494 °C and 493 °C, respectively (Fig. 1). The maximum weight loss for fresh PP, PP < 10, PP > 10 was at 480 °C, 477 °C and 475 °C, respectively. PE > 10 and fresh PP presented the maximum temperature corresponding to the maximum mass loss for plastic type. The maximum mass loss rate is exhibited by PE < 10 and fresh PP. Fresh and excavated PP degrade at a lower temperature than PE. Degradation of fresh and excavated PE samples did not show significant differences. All temperature results were in the ranges identified for PE and PP waste degradation in Yan et al. (2015) study. The landfill environment and storage time did not seem to have a critical effect on the thermal behaviour of excavated plastic.

3.2. Identification of chemical compounds and potential recycling of pyrolysis products of plastics recovered from landfill

The pyrolysis oil from excavated plastics is considered as a resource, due to its high calorific value if used as a fuel, and the production of feedstock for chemical synthesis and plastics production (Khan et al., 2016; Canopoli et al., 2018a; Fox and Stacey, 2019). The relative abundance of produced compounds at different temperatures is in Tables 2–4, Fig. 2a and b. The Py-GC–MS analysis indicates that aliphatic hydrocarbons are the dominant products, followed by aromatics and fluorine containing compounds (Tables 2–4). The area percentage of condensable pyrolysis products at ambient temperature was more than 80 % in most of the samples (see supplementary data Table S21), with the majority being aliphatic and aromatic hydrocarbons. This finding is consistent with previous studies on the pyrolysis of PE and PP, where a high percentage of aliphatic hydrocarbons was

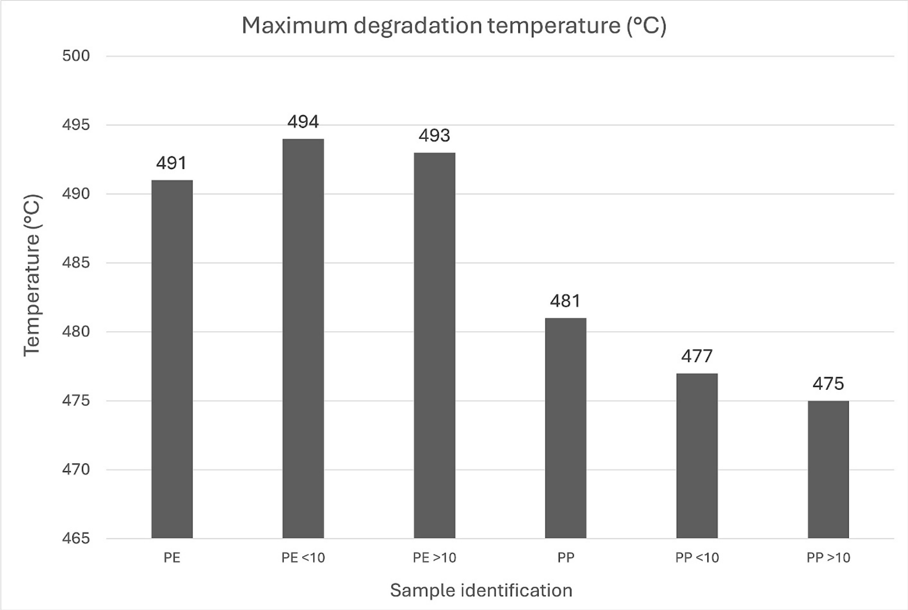


Fig. 1. Maximum degradation temperature of fresh and excavated polyethylene (PE) and polypropylene (PP) with different storage time (<10 years and >10 years).

Table 2
Relative abundance (%) of products at each of the different pyrolysis temperatures for polyethylene.

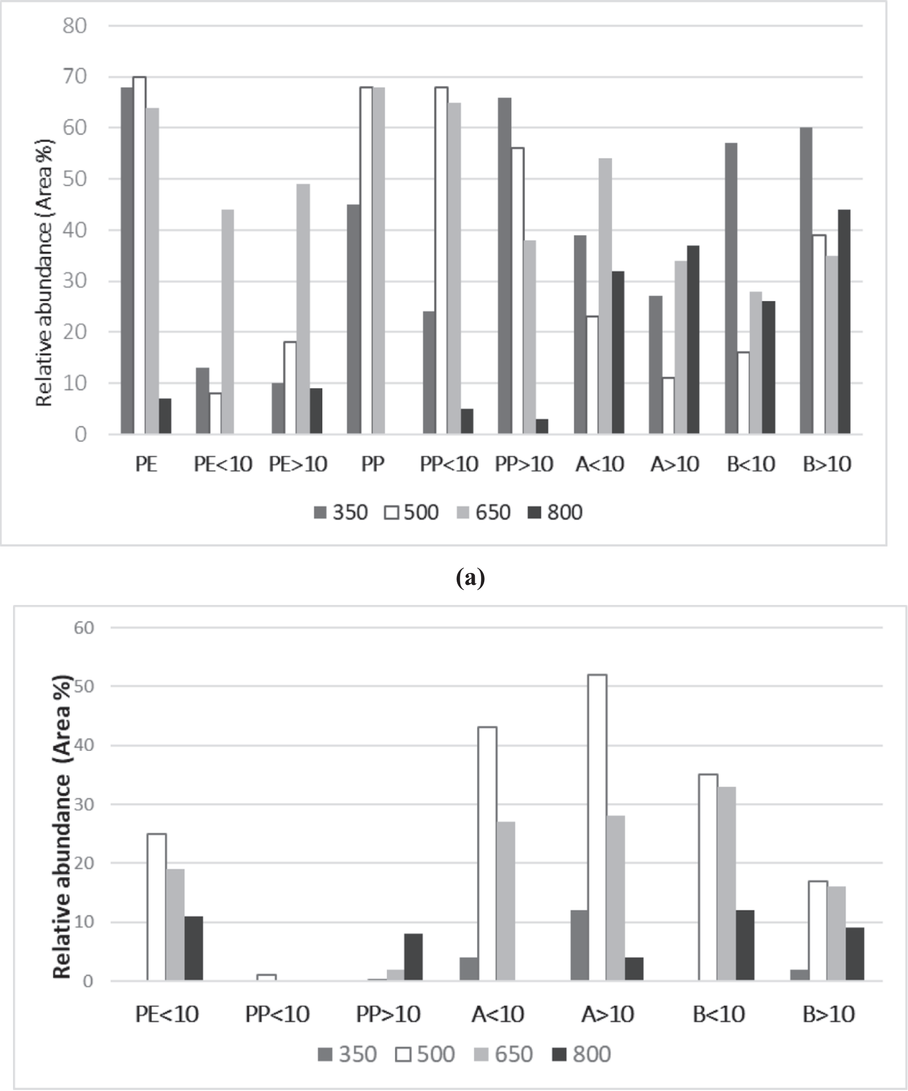
Volatile organic groups produced (% relative conc)	PE fresh				PE < 10 years				PE > 10 years			
	350 °C	500 °C	650 °C	800 °C	350 °C	500 °C	650 °C	800 °C	350 °C	500 °C	650 °C	800 °C
Aliphatic hydrocarbons	68	70	64	7	13	8	44	0	10	18	49	9
Aromatic hydrocarbons	0	0	0	0	0.23	25	19	11	0	0	0	0
Other aromatics	0	9	0	0.3	0.17	0	0.06	0	0	0	0	0
Fluorine containing compounds	0	1	0.04	68	20	25	3	51	90	77	30	69
Bromine containing compounds	0	0	0	0	0	0	0	0	0	0	0	0
Esters	13	5	2	0	56	1	3	16	0	1	0.02	0
Alcohols	0	7	30	4	8	0.1	18	0	0	4	16	4
Amines	0	0		0	2	0	0	0	0	0		0
Organosulfur compounds	0	0	0	0	0	0	0	0	0	0	0	0
Fatty acids	0	0	0	0	0	0.2	0	0	0	0	0	0
Hydrogen halides	0	0	0	0	0	40	0	0	0	0	0	0
Aldehydes	0	2	4	0	0	0	2	0	0	1	1	0
Ketones	0	0	0.01	0	0	0	3	0	0	0	0.01	0
Chlorinated hydrocarbons	0	0	0	0	0	0	0	0	0	0	0	0
Gas (CO_2)	19	6	0	21	0	0.33	8	22	0	1	4	18
Total	100	100	100	100	100	100	100	100	100	100	100	100

Table 3
Relative abundance (%) of products at each of the different pyrolysis temperatures for polypropylene.

Volatile organic groups produced (% relative conc)	PP fresh				PP < 10 years				PP > 10 years			
	350 °C	500 °C	650 °C	800 °C	350 °C	500 °C	650 °C	800 °C	350 °C	500 °C	650 °C	800 °C
Aliphatic hydrocarbons	45	68	68	0	24	68	65	5	66	56	38	3
Aromatic hydrocarbons	0	0	0	0	0	1	0	0.02	0	0.1	2	8
Other aromatics	0	0	0	0	0	0.31	0	0.03	0.34	6	39	22
Fluorine containing compounds	47	14	1	64	65	5	0.5	80	25	0	0.23	61
Bromine containing compounds	7	4	0	2	8	0	0	13	8	16	1	6
Esters	0	1	5	0	0	1	3	0	0	9	4	0
Alcohols	0	13	24	31	0	23	29	0	0	9	11	0
Amines	0	0		0	0	0		0	0	0		0
Organosulfur compounds	0	0	3	0	0	2	1	0	0	0	3	0
Fatty acids	0	0	0	0	0	0	0	0	0	0	0	0
Hydrogen halides	0	0	0	0	0	0	0	0	0	0	0	0
Aldehydes	0	0	0.06	0	0	0	0.21	0	0	0	0.05	0
Ketones	0	0	0	0	0	0	0	0	0	0	2	0
Chlorinated hydrocarbons	0	0	0	0	0	0	0	0	0	0	0	0
Gas (CO_2)	0	0	0	3	0	0	0	2	0	4	1	0
Total	100	100	100	100	100	100	100	100	100	100	100	100

Table 4
Relative abundance (%) of products at each of the different pyrolysis temperatures for mixed plastic samples.

	350 °C				500 °C				650 °C				800 °C			
Volatile organic groups produced (% relative conc)	A <10	A >10	B <10	B >10	A <10	A >10	B <10	B >10	A <10	A >10	B <10	B >10	A <10	A >10	B <10	B >10
Aliphatic hydrocarbons	39	27	57	60	23	11	16	39	54	34	28	35	32	37	26	44
Aromatic hydrocarbons	4	12	0.17	2	43	52	35	17	27	28	33	16	0	4	12	9
Other aromatics	8	6	9	17	0.5	0.2	6	29	1	9	18	29	0.06	0.4	20	11
Fluorine containing compounds	24	29	13	14	6	10	3	2	0	1	0	1	42	45	12	21
Bromine containing compounds	6	9	9	8	4	5	2	2	0	0	0	0	9	0	2	8
Esters	16	10	10	0.08	1	0.4	1	2	2	1	3	3	0	0	0	0
Alcohols	0.1	0.5	0.46	0.05	4	1	1	2	12	16	10	12	13	13	24	3
Amines	0	4	0	0	0	0	0	0					1	1	1	0.37
Organosulfur compounds	2	2	0	0.01	18	20	11	3	2	9	7	4	0	0	0	0
Fatty acids	0	0	1	0.04	0	0	4	3	0	0	0	0	0	0	0	0
Hydrogen halides	0	0	0	0	0	0	16	4	0	0	0	0	0	0	0	0
Aldehydes	0	0	0	0	0	0	0	0	1	2	1	1	0	0	0	0
Ketones	0	0	0	0	0	0	0	0	0.1	0	0	0	0	0	0	0
Chlorinated hydrocarbons	0	0	0	0	0	0	3	0	0	0	0	0	0	0	0	0
Gas (CO_2)	0	0	1	0	0	0	0	0	0	0.32	0.49	0	4	1	3	5
Total	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100



(a)

(b)

Fig. 2. Relative abundance (%) of aliphatic hydrocarbons (a) and aromatic hydrocarbons (b) in each sample at different temperatures (°C).

commonly observed (Al-Salem, 2019; Jung et al., 2010).

Plastics such as PE and PP are often fluorinated to improve and preserve their characteristics, especially when they are intended as chemicals and solvent containers (Kharitonov and Kharitonova, 2009). Fluorine-containing compounds are also used in plastic food containers or wrappers, while bromine is commonly used as a flame retardant (AccuStandard, 2018). Other common plastic additives detected include organosulfur compounds (AccuStandard, 2018). Oxygenated compounds (esters, ketones, alcohols, fatty acids, carboxylic acids, aldehydes and heterocyclic aromatic compounds) were found across all samples, not just in the PET-containing sample B. This finding aligns with previous research, which also reported the formation of oxygenated compounds during plastic waste pyrolysis (Toraman et al., 2014).

Details of the compounds included in each group can be found in supplementary data Table S4-S18.

Regarding the temperature-dependent production of hydrocarbons, aliphatic hydrocarbons (Fig. 2a) reached their highest relative abundance at 650 °C in several samples, including PE < 10, PE > 10 and A < 10. For other samples, such as PP > 10 and B < 10, the highest levels were observed at 350 °C. At 800 °C, these hydrocarbons were most abundant only in A > 10.

Aromatic hydrocarbons (Fig. 2b) were generally most abundant at 500 °C in most samples, except for PP > 10, where 800 °C was more favourable. Styrene was the predominant aromatic hydrocarbon produced in the pyrolysis of A < 10, A > 10, B < 10 and B > 10 at 500 °C (Table 5), consistent with findings from other studies on plastic pyrolysis (Anuar Sharuddin et al., 2016; Miandad et al., 2017; Miandad et al., 2019).

The presence of PET and PVC is known to facilitate the production of oxygenated and chlorinated compounds. A comparison of samples A (without PET and PVC) and B (with PET and PVC) revealed a significant difference in the production of oxygenated compounds, particularly at 650 °C. Sample B had a notably higher oxygenated compound content, with 47 % in B > 10 compared to 36 % in A > 10 and 37 % in B < 10 compared to 17 % in A < 10. Chlorinated compounds, including hydrogen chloride and chlorinated aromatics, were primarily detected in sample B at 500 °C. While the abundance of chlorinated compounds in sample A was minimal (~1%), it was significantly higher in B < 10 (22 %) and B > 10 (10 %), with hydrogen chloride being the dominant halogenated compound in B < 10 (16 %). B > 10 chlorinated compounds were mainly represented by aromatics with 7 %.

Several key compounds identified in the pyrolysis oil (detailed in supplementary data Table S19) include diethyl phthalate (which is used to improve plastic flexibility), 13-Docosen-1-ol, benzene, benzoic acid, styrene and vinyl benzoate, all commonly used in the plastic industry. The temperatures of 500 and 650 °C were identified as the optimal conditions for hydrocarbon production. The hydrocarbon product distribution (see supplementary data Table S20) showed that the C5-C9 range was most prevalent in samples, including PP (fresh and excavated), PE < 10, mixed plastics A and B, while the C10-C15 range was most abundant in PE and PE > 10. Aromatic hydrocarbons were predominantly produced in PE < 10 and the mixed plastic samples A and B. The research hypothesis posited that prolonged environmental ageing of polymers in landfills would degrade polymer chains and reduce the efficiency of pyrolysis in producing valuable small-chain hydrocarbons (<C10). However, the results show that excavated, aged polymers generally produce similar or even higher relative abundances of light hydrocarbons in the gasoline and naphtha ranges compared to fresh

Table 5
Relative abundance (%) of hydrocarbons fractions obtained at pyrolysis temperature of 500 °C and 650 °C.

500 °C		PE	PE < 10	PE > 10	PP	PP < 10	PP > 10	A < 10	A > 10	B < 10	B > 10
LGP, Fuel gas, Refinery gas < C5	Aliphatic	1	0	0	6	8	2	2	0	0	0
	Aromatic	0	0	0	0	0	0	0	0	0	0
Gasoline	Aliphatic	7	2	3	19	21	8	3	3	8	15
C5-C8	Aromatic	0	17	0	0	0	0.1	42	48	32	12
Naphtha	Aliphatic	16	2	7	41	44	24	11	7	8	15
C6-C10	Aromatic	0	22	0	0	0	0	42	48	34	15
Kerosene (Paraffin)	Aliphatic	34	2	9	12	8	5	2	1	1	1
C10-C16	Aromatic	0	6	0	0	0	0	1	3	0.2	0.1
Diesel oil	Aliphatic	24	2	5	1	4	4	0.2	0.4	1	12
C14-C20	Aromatic	0	0	0	0	0	0	1	3	0.2	0.1
Lubricating oil	Aliphatic	16	1	4	0	1	4	0	0.1	2	11
C18-C25	Aromatic	0	0	0	0	0	0	0	1	0.5	0
Fuel oil	Aliphatic	11	0.4	3	0	0.3	5	5	0.5	2	15
C20-C27	Aromatic	0	0	0	0	0	0	0.1	1	0	0
Wax and greases	Aliphatic	2	2	0.4	0	0	11	11	3	7	9
C25-C35	Aromatic	0	0	0	0	0	0	0.1	0	0	0
Bitumen > C35	Aliphatic	1	0	0	0	0	5	1	0	0.2	2
	Aromatic	0	0	0	0	0	0	0	0	0	0
650 °C		**PE**	**PE < 10**	**PE > 10**	**PP**	**PP < 10**	**PP > 10**	**A < 10**	**A > 10**	**B < 10**	**B > 10**
LGP, Fuel gas, Refinery gas < C5	Aliphatic	2	1	1	6	6	4	3	1	3	3
	Aromatic	0	0	0	0	0	0	0	0	0	0
Gasoline	Aliphatic	15	13	8	20	20	10	23	10	6	6
C5-C8	Aromatic	0	5	0	0	0	1	4	9	23	11
Naphtha	Aliphatic	22	17	13	41	42	27	34	15	15	18
C6-C10	Aromatic	0	15	0	0	0	1	11	14	27	12
Kerosene (Paraffin)	Aliphatic	28	11	18	15	16	8	12	11	7	8
C10-C16	Aromatic	0	8	0	0	0	1	14	12	5	4
Diesel oil	Aliphatic	24	15	24	9	7	2	8	8	4	4
C14-C20	Aromatic	0	0	0	0	0	0.2	14	13	5	2
Lubricating oil	Aliphatic	12	12	19	7	4	3	7	5	3	5
C18-C25	Aromatic	0	0	0	0	0	0	3	3	1	0.4
Fuel oil	Aliphatic	8	7	13	1	2	2	5	4	2	4
C20-C27	Aromatic	0	0	0	0	0	0	0	0	0	0
Wax and greases	Aliphatic	0	0	0	1	1	2	4	3	3	5
C25-C35	Aromatic	0	0	0	0	0	0	0.2	0	0	0
Bitumen > C35	Aliphatic	1	0.03	0	0	0	0	0.1	0	0	0
	Aromatic	0	0	0	0	0	0	0	0	0	0

polymers. This suggests that ageing primarily causes chain scission, increasing the formation of smaller hydrocarbon fragments rather than hindering pyrolysis efficiency. Thus, the data do not support the hypothesis that polymer ageing reduces pyrolysis efficiency for producing small-chain hydrocarbons. Instead, ageing appears to facilitate pyrolysis by increasing light hydrocarbon yields, confirming the potential for effective recovery of petrochemical feedstocks from aged landfill plastics. The pyrolysis liquid needs to be upgraded to be used, for example, as fuel. Indeed, the pyrolysis oil from plastics may present ash and heteroatoms such as sulphur and nitrogen, which lower the quality of the fuels (Thahir et al., 2019; Miskolczi et al., 2004). Fractionation, as presented in Tables 5, is a crucial stage for separating fuel fractions (Costa and Santos, 2019), with hydrogenation and cracking steam reforming being important refining processes (Bezergianni et al., 2017; Remón et al., 2014). In Table 5, The relative abundance (%) for each hydrocarbon fraction was calculated by summing the chromatographic peak areas of all compounds within that fraction and dividing by the total detected peak area for the sample, providing a semi-quantitative percentage composition. Because fractions overlap in carbon number ranges, the cumulative values can exceed 100 %. Table 5 shows the relative abundance for different possible uses of the hydrocarbons produced in this study at 500 and 650 °C. The major percentage of compounds for PP, fresh and excavated, PE < 10, mixed A and B, fit in the naphtha range. Pyrolysis products of fresh PE and PE > 10 at 500 °C are mostly represented by kerosene range, while PE > 10 at 650 °C presented more compounds for the diesel range. At 650 °C, the increased cracking intensity can break down not only the long-chain hydrocarbons but also cause secondary reactions that may lead to the formation of heavier, more complex molecules, contributing to a higher diesel fraction.

In 2022, the world produced about 400 million tonnes of plastics (PlasticsEurope, 2024). In Europe, within 58.8 million tonnes of plastics the most in demand polymers were PE and PP (~37 wt%) (PlasticsEurope, 2024). These plastics are produced mostly from virgin fossil resources and in minimal part from mechanical recycling of non-landfilled plastic waste. However, this recycling of non-landfilled plastic waste generally leads to a downcycling where the outputs are of lower value than the feedstock (Nielsen et al., 2019). The use of high-value chemicals (HVCs) from recovered plastics pyrolysis could reduce the needs of virgin fossil resources. The recovered plastics pyrolysis products, such as naphtha, could be used in petrochemical clusters. The naphtha cracking produces around 55 wt% HVCs such as ethylene, propylene, butadiene, aromatics and >C5 (Ren et al., 2006). The unsaturated hydrocarbons and aromatics such as benzene, toluene and styrene, can be sold to produce new plastic and other products (Fox and Stacey, 2019; Miandad et al., 2019). The aliphatic compounds can be used to produce ethylene and propylene. Benzene can be used to produce plastic, detergents, dyes and pesticides. Toluene can be employed as a solvent and a starting material for the synthesis of organic compounds such as benzoic acid, benzaldehyde. The recovered styrene can be reused to make synthetic rubber, polystyrene and expanded polystyrene (PlasticsEurope, 2018). Benzoic acid could be recycled for the synthesis of dyes and other organic compounds. This would lead to the upcycling of recovered plastic which has been disposed of and left unused, sometimes for decades. To provide a preliminary perspective on the possible economic benefit of upcycling disposed plastics, we estimated the potential revenue from converting the mixed PE, PP, and PS fraction of excavated waste into naphtha. The assumptions for this estimation are as follows: 1) the average naphtha market price in January 2024 was 623.38 USD/tonne (Trading Economy, 2024); 2) recovered plastics consisted of ~68 wt% PE, PP, and PS (with an approximate breakdown of PE 50 wt%, PP 15 wt%, PS 3 wt%) identified in recovered plastics (see supplementary data Table S1); 3) the naphtha yield was based on the pyrolysis results of mixed sample A (Table 5); 4) HVCs recovery from naphtha was assumed to be 55 wt% (Ren et al., 2006); 5) 7.6 million tonnes of plastics were disposed in landfill in 2022 in Europe (PlasticsEurope, 2024); and 6) collection, recovery, and processing costs are not included in this estimate.

Based on these assumptions, the potential revenue from naphtha produced from plastics discarded over the last 70 years could range from approximately 602 to 1142 billion USD, with an average estimate of 945 billion USD. For European landfilled plastics in 2022, the estimated revenue could range from 0.9 to 1.8 billion USD, with an average of 1.5 billion USD. While these estimates are preliminary and do not yet account for collection, recovery, and processing costs, potential material degradation, or market variability, they nevertheless illustrate the substantial economic potential inherent in upcycling long-disposed plastic waste. Future detailed techno–economic assessments will be essential to translate this promising potential into practical, scalable solutions.

The cost of a landfill mining project can be variable according to different aspects such as the quantity of waste in the landfill, technologies applied and geographical location of the landfill. The estimated cost for an ELFM project of a landfill in Belgium with a surface area of 2,000 ha was USD ~ 11 billion and comprised of excavation, sorting, pre-treatment, incineration and contingency (Van Passel et al., 2013). The incineration of waste counted about USD 6 billion, and it was considered a worthy process for the recovery of energy which was estimated to be USD ~ 9 billion. In contrast, a similar project in China for a smaller landfill of 11.3 ha was estimated to cost about USD 6 million covering the prices for excavation, screening and sorting, construction of material handling facility, transportation of materials and final waste disposal (Zhou et al., 2015). In this study, the first three potential benefits were obtained from the electricity generated by combustible incineration, reclamation of the land, and recycling of soil-like materials. In addition, the capital investment for the valorisation of the plastic fraction through pyrolysis was estimated to be USD ~ 3.7 million for a pyrolysis plant with a capacity of 1,000 kg of plastic waste per hour (Fivga and Dimitriou, 2018). The estimated annual operating costs for this type of plant were USD 1.5 million (Fivga and Dimitriou, 2018). The introduction of recovered plastics in the new circular plastics economy, could lower the environmental impact related to the mismanagement of this material and offers a variety of HVCs. These products can be sold gaining revenue from current unused materials.

4. Conclusion

The increasing demand for plastics and the environmental challenges associated with their short service life highlight the need for sustainable recovery strategies. The estimated mass of plastics present in landfills and natural environment is 4.9 bn tonnes. As landfills are increasingly remediated for further development, managing the plastics excavated during these projects requires strategies that maximize their value. This study investigated how the storage age of specific polymers recovered from landfills influences their pyrolysis behaviour and product distribution. The results indicate that although some signs of ageing, such as minor variations in product yields, were observed, the thermal behaviour and hydrocarbon yield of aged polymers remained largely comparable to those of non-landfilled plastics. This finding suggests that the degradation occurring during long-term burial does not critically hinder the production of valuable small-chain hydrocarbons (<C10) via pyrolysis. Consequently, excavated plastics can still generate significant quantities of naphtha-range hydrocarbons and aromatic compounds suitable for closed-loop recycling and petrochemical feedstocks. This supports the viability of integrating pyrolysis of aged plastics into existing waste-to-resource frameworks. While this investigation focused on single polymers and controlled artificial mixture, thus providing a more qualitative than quantitative understanding, it establishes an important foundation for further research. Further studies should evaluate the economic feasibility and environmental benefits at scale, including detailed techno-economic analysis and life cycle assessment, to maximise the sustainability and circularity of this approach.

CRediT authorship contribution statement

Stuart T. Wagland: Writing – review & editing, Supervision, Resources, Funding acquisition, Conceptualization. **Luisa Canopoli:** Writing – original draft, Visualization, Methodology, Investigation, Formal analysis. **Frederic Coulon:** Writing – review & editing, Supervision, Formal analysis. **Diane Coral Turner:** Writing – review & editing, Methodology, Investigation, Validation. **Geraint Huw Morgan:** Writing – review & editing, Validation, Resources.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.wasman.2025.115035.

Data availability

Data available through Cranfield data repository

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