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Is incineration the terminator of plastics and microplastics?

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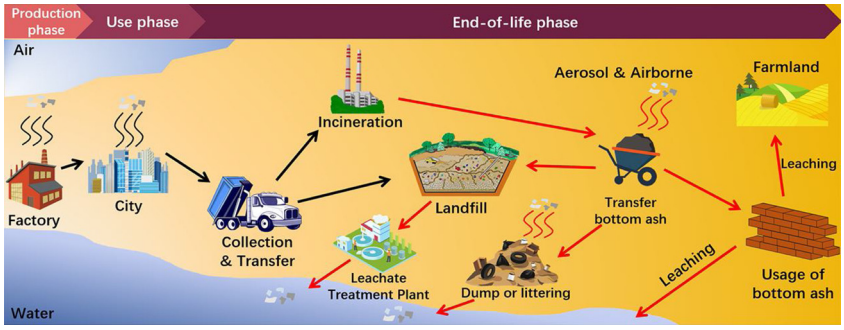
GRAPHICAL ABSTRACT



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ABSTRACT

It is widely accepted that incineration can permanently eliminate plastic waste. However, unburned material still exists in the bottom ash that is a solid residue from incinerators. In this study, microplastics exacted from bottom ash in 12 mass burn incinerators, one bottom ash disposal center and four fluidized bed incinerators were identified by micro-Fourier transform infrared spectroscopy. The results showed that bottom ash was a neglected microplastics source with an abundance of 1.9–565 n/kg, which indicated that per metric ton waste produce 360 to 102,000 microplastic particles after incineration. Nine types of plastics were identified, of which polypropylene and polystyrene were the predominant types. Microplastics sized between 50 μm and 1 mm accounted for 74 %. Granules, fragments, film, and fibers accounted for 43 %, 34 %, 18 %, and 5 % of the microplastics, respectively. The abundance of microplastics differed significantly with whether the local waste was source-separated, the local gross domestic product per capita, and the types of furnace. The global microplastics emission from incineration bottom ash was then estimated. Our observations provide empirical evidence proving that incineration is not the terminator of plastic waste, and bottom ash is a potential source of microplastics released into the environment.

1. Introduction

Microplastics (MPs, with size < 5 mm along their longest dimension) are now ubiquitous around the world (Allen et al., 2019; Bergmann et al., 2019; Brandon et al., 2019; Zhang et al., 2020a), and have drawn increasing global attention for their potential toxicity (Huerta Lwanga et al., 2016; Jacob et al., 2020). To reduce pollution by MPs, two principles are generally followed. One is to reduce plastic

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Table 1
Abundance of MPs, LOI and general information from 17 sites. "W/O" represent without waste source separation, "W" represents with waste source separation.

Furnace type	Site	MPs abundance (n/kg)		Loss on ignition (LOI)		GDP/ per capita (thousand RMB)(2019)	Waste source separation
		Mean	Standard deviation	Mean	Standard deviation		
Mass burn furnace	M1	44.3	67.9	1.52 %	0.15 %	107.6	W/O
	M2	1.9	2.7	1.77 %	0.15 %	174.6	W
	M3	29.6	21.3	2.33 %	0.09 %	119.2	W
	M4	109	154	1.46 %	0.50 %	119.2	W
	M5	44.6	44.8	2.02 %	0.94 %	174.6	W
	M6	15.2	14.7	1.73 %	0.12 %	67.5	W
	M7	453	11.9	1.44 %	0.44 %	122.1	W/O
	M8	3.5	4.9	2.02 %	0.16 %	75.4	W
	M9	2.7	3.9	3.18 %	0.99 %	149.1	W/O
	M10	75.5	11.9	2.99 %	0.03 %	83.2	W/O
	M11	185	52.6	2.94 %	0.17 %	83.2	W/O
	M12	100	77.1	1.28 %	0.93 %	178.2	W/O
	M13	565	195	1.64 %	0.15 %	47.9	W/O
	Mean for M	125	180	2.02 %	0.64 %	115.5	/
Fluidized bed furnace	F1	2.8	3.9	0.75 %	0.41 %	95.8	W/O
	F2	0	0	0.02 %	0.02 %	174.5	W
	F3	0	0	/	/	103.2	W/O
	F4	334	276	0.57 %	0.05 %	65.8	W/O
	Mean for F	84.2	167	0.45 %	0.38 %	109.83	/
	Mean for all sites	116	172	1.73 %	0.87 %	114.18	/

consumption and plastic waste generation at the source, and the other is to increase the recycling and treatment of plastic waste. The first principle seems challenging to implement in the short term for the previous studies reported. Global plastic production increased from 348 million metric tons (MT) in 2017 to 359 million MT in 2018, and 39.9 % of this was used for packaging (PlasticsEurope, 2019). From 1950–2015, the cumulative generation of plastic waste reached 5800 million MT, of which 800 million MT of plastic waste was eliminated by incineration, 100 million MT were recycled and 4900 million MT were landfilled or discarded (Geyer et al., 2017). It was estimated that 28 % of the plastic waste was incinerated in China (Jiang et al., 2020). Globally, plastic waste accounts for about 12 % of municipal solid waste (MSW) by mass (Silpa et al., 2018). Plastic waste in the United States accounted for 13.2 % of the total MSW in 2017 (EPA, 2017). In European countries, plastic waste accounts for 5.4 %–21 %, with an average of 11 %, while in China, it accounted for 11.3 %–17.2 % of total MSW from 2002 to 2011 (Yang et al., 2018), and the proportion increased with economic development. As for the second principle, it is generally believed that poor solid waste management (either non-management or mismanagement), such as littering and dumping, is the primary source of plastic waste into environment (Jambeck et al., 2015), while managed waste treatment systems such as sanitary landfilling, biological treatment and incineration, can stock or eliminate the environmental risks of MPs. However, recent studies have found that plastic waste entering the managed solid waste treatment systems may also produce MPs releasing into the environment. For example, landfilling is a potential source of MPs with the abundance of MPs in Chinese MSW landfill leachate ranged from 0.42 to 24.58 MPs per liter (He et al., 2019). The mean abundance of MPs in refuse from Chinese landfill was 62 n/kg (Su et al., 2019). Biofertilizer processed from organic household waste through composting and anaerobic digestion has been found to be as a vehicle for the entry of MPs into the terrestrial environment. The abundance of MPs ranged from 14 to 895 n/kg of compost, and it has been estimated that compost releases between 35 billion and 2.2 trillion MP particles into the environment annually in Germany (Weithmann et al., 2018). Moreover, instances of plastiglomerate that was generated in the beach by burning plastic waste were found in many locations: like Bali, Indonesia, California, USA, Madeira, Portugal and Ontario, Canada (Corcoran and Jazvac, 2020). The cases indicated that the plastic waste did not end up by landfilling, composting, and burning, but resulted in plastic pollution crisis.

Waste incineration with energy recovery, as an essential part of the circular economy, accounts for a large proportion of solid waste treatment systems in both developed and developing countries. Incineration leads to the production of final bottom ash that is returned to the environment. To evaluate the efficiency of combustion, loss on ignition (LOI) is used as an indicator of the unburned material in the bottom ash. The LOI of bottom ash from two major furnace types, mass burn incineration and fluidized bed incineration, is 3%–5% and 2 %–4 %, respectively. Incineration is generally considered to be the terminator of eliminating plastic waste, which can ultimately convert polymers into CO_2 and mineral fraction (Geyer et al., 2017). However, the unburned material from the bottom ash contained synthetic fibers (Chimenos et al., 1999), which implies that plastics and MPs may still exist in the bottom ash and could be transported into the environment through its reuse or dumping. Therefore, to verify whether incineration can ultimately eliminate MPs and to evaluate the quantity of MPs transported to the environment by bottom ash, this study is the first attempt to extract and identify MPs from the bottom ash produced by MSW incineration.

We investigated the abundance, polymer type and morphology of MPs in bottom ash from 17 sites (16 MSW incinerators and one bottom ash disposal center) in eight Chinese cities. The objectives of the research were 1) to verify whether MPs are present in the unburned material in the bottom ash, 2) if MPs are present, to infer the incineration conditions that may affect the abundance of MPs in bottom ash, and 3) to examine the morphology and types of MPs to explore the reasons for this situation. Based on the abundance of MPs in the bottom ash, we estimated the global quantities of MPs that may be released into the environment from MSW incineration systems.

2. Materials and methods

2.1. Sampling

Bottom ash was collected from 16 MSW incineration plants and one bottom ash disposal center, in eight different cities in China. The disposal center M11 only disposed bottom ash produced from incineration plant M10, so samples from M10 and M11 were produced from the same furnaces in different seasons. Of the 16 MSW incineration plants, 12 sites were equipped with mass burn furnaces and the other four sites were using fluidized bed furnaces. China has been promoting source waste separation and some urban blocks have enforced the rules. Hence, among the 17 sites, the service area of seven sites have

implemented waste source separation while ten sites have not (Table 1). The input waste of the incineration plants was mainly composed of domestic garbage, with some cases also containing some industrial waste, commercial waste and construction and demolition waste (C&D waste). Hence, packaging, plastic pipe, waterproof material and wall paper were mixed in the input waste. The capacities of mass burn incinerators ranged from 330 MT to 867 MT per day and the capacity of fluidized bed furnace incinerators ranged from 350 MT to 400 MT per day. The bottom ash generation rates of mass burn furnaces (15–25 %) were higher than that of fluidized bed furnaces (8–10 %). The selected plants were established or upgraded in the past ten years with advanced technology and can be representative for typical incineration plants in China. In total, 31 bottom ash loads were sampled during stable operation of the incinerators. Detailed information about the incineration plants and sampling sites are shown in Table S1 and Fig. S1. At least 100 kg of bottom ash was grabbed by metal claw randomly and then mixed to form one sample. Subsequently, 5–10 kg of bottom ash was taken for each sample and then homogenized. Two replicates of each sample were taken; one was sealed in a metal box for MPs extraction, and the other was put in a plastic bag for the LOI test.

2.2. Extraction of MPs

The method for extracting MPs from bottom ash was referred to Maniet et al. (Mani et al., 2019) with slight modifications. Samples were dried at 65 ℃ for 24 h, and then screened through a stack of stainless-steel sieves with descending mesh sizes of 2 mm and 50 μm. Suspected MPs on the 2 mm sieve were selected visually, removed with tweezers, and stored in glass petri dishes. Three replicates of fine bottom ash from the 50 μm sieve were mixed with 1.2 g/mL of saturated NaCl solution and stirred for 15 min, then sonicated for 5 min at 215 W/35 kHz. After settling for 2 h, the supernatant was filtered three times through stacked sieves with mesh sizes of 500 μm and 50 μm. Particles on the upper sieve were rinsed three times with deionized water, and then filtered onto nylon membrane filters (Millipore, USA, NY2004700) of 47 mm diameter and 20 μm pore size. To dissolve crystalline particles, the bottom ash from the lower sieve was mixed with 50 mL of 4 % hydrofluoric acid (HF) solution in a bottle made of polytetrafluoroethylene (PTFE), then covered with aluminum foil and fixed on a shaker at 60 rpm, for 24 h at room temperature (Wang et al., 2019). The resulting mixture was vacuum filtered through filters of pore size 20 μm, and rinsed three times with deionized water. All the filter membranes were placed in clean glass petri dishes and dried at room temperature.

2.3. Sorting and identification

Particles on the filter membranes were inspected using a stereomicroscope (Leica S APO, Germany). Particles that were identified as plastic included (but were not limited to) those with a homogeneous texture, an absence of a cellular structure, which did not undergo fragmentation when stress was applied with tweezers, and those where artificial coloring was evident; these were sorted into glass petri dishes. Due to the fact that plastics undergo severe alterations in combustion chamber, the surfaces of some particles were burned into black-carbon like material (Fig.4 A). But micro-Flourier transform infrared spectroscopy (μ-FTIR) in transmission mode (Thermo Scientific Nicolet Continuμm, and Bruker LUMOS) can only identify transparent or semi-transparent particles (Huppertsberg and Knepper, 2020). Therefore, the particles which can not be distinguished with carbon particles by stereomicroscope were excluded for the next identification. This process may thereby cause the underestimation about the abundance of MP particles in bottom ash and this phenomenon was discussed in Section 4.1 Method assessment. The size and shape of each suspected plastic particle were recorded. A digital microscope (VHX-6000, Keyence, Japan) was used to photograph some of the particles.

All suspected MP particles were identified using μ-FTIR in transmission mode. The mid-infrared wavelength range (4000–675 cm^{-1} for Thermo and 4000–600 cm^{-1} for Bruker) was used to scan particles 16 times with a resolution of 4 cm^{-1}. All spectra were matched with the databases from OMNIC and Bruker, respectively. Particles with match values > 70 % were considered to be plastic. As to MP particles with match values between 60 % and 70 %, in previous studies, they were identified individually based on both the morphology of particles inspected by stereomicroscope and the closeness of absorption frequencies to those of chemical bonds in polymers from database (Kim et al., 2018).

2.4. Contamination mitigation

Processing and identification steps were performed with glass, stainless steel and other non-plastic apparatuses in a vertical flow cabinet or ultra-clean stainless steel room to avoid MP pollution, except for the PTFE bottles and PA filtration unit used for processing liquids containing HF. All items were rinsed with deionized water at least three times and sonicated for 15 min before use. Beakers and other receptacles were always covered with glass lids or aluminum foil. A cotton lab coat and nitrile gloves were worn throughout the experiment. Three procedural blanks (30 g of SiO_2) were treated and examined to detect any MPs introduced during the experimental procedures. No plastic particles were found in the blank control group.

2.5. Loss on ignition measurement

Metals were removed and then samples were ground into fine particles (< 2 mm). LOI was conducted in triplicate by heating bottom ash samples in a muffle oven at 600 ℃ for 3 h. the weight loss of cooled residues reflected the unburned material content.

3. Results

3.1. Abundance of MPs in bottom ash from different sites

A total of 892 suspected particles were sorted, and 276 of them were identified as MPs. The mean abundance of MPs in the bottom ash was 116 ± 172 n/kg (Table 1). MPs were identified in all 13 sites with mass burn furnaces with an abundance of 125 ± 180 n/kg. MPs were identified in two of four sites with fluidized bed furnaces with an abundance of 84 ± 167 n/kg. Although a higher mean abundance was observed in bottom ashes sampled from mass burn furnaces than from fluidized bed furnaces, the difference was not significant ($p = 0.37 > 0.05$) as shown in Fig. 1A. Among the sites with mass burn furnaces, the highest abundance of MPs was from M13 (565 ± 195 n/kg), while the lowest abundance was from M2 (1.9 ± 2.7 n/kg). Among the sites equipped with fluidized bed furnaces, the highest abundance of MPs was from F4 with 334 ± 276 n/kg. China has been promoting source waste separation since July 2019. Among the 17 sampling sites, the service areas of seven sites have implemented waste source separation while ten sites have not (Table 1). The mean abundance of MPs from sites with waste source separation (33 ± 79 n/kg) was lower than that from sites without waste source separation (228 ± 264 n/kg) (Fig. 1B). This difference was statistically significant ($p < 0.01$).

Compared with other soil matrices polluted with MPs, the abundance of MPs in bottom ash was significantly higher than that in farmland soil (Liu et al., 2018; Piehl et al., 2018). Additionally, up to 593 n/kg of MPs were found in 26 floodplain soil samples in Switzerland (Scheurer and Bigalke, 2018), which is of the same order of magnitude as bottom ash. Therefore, the bottom ash generated by MSW incinerators is a potential source of MPs. On the other hand, the abundance of MPs in bottom ash is two to three orders of magnitude lower than that in sewage sludge (Mahon et al., 2017) and two to four orders of magnitude lower than that in refuse (Su et al., 2019),

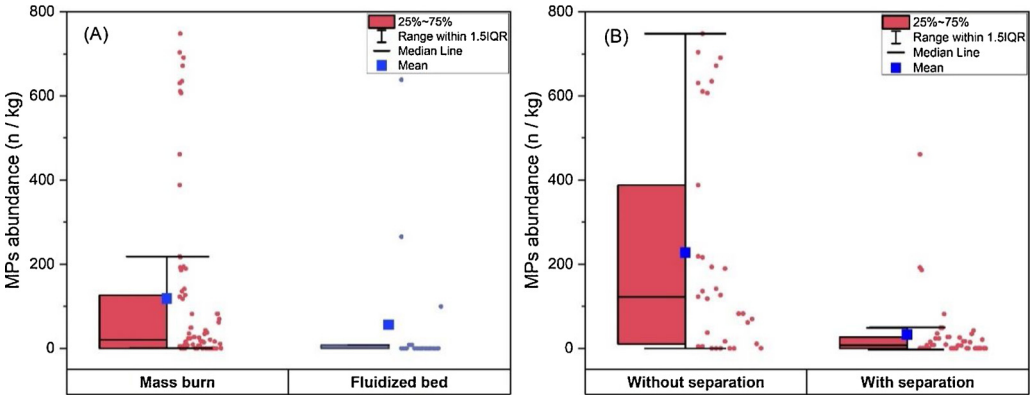

Fig. 1. Abundance of MPs in bottom ash. (A) From different types of incinerators. $n_{Mass\ burn} = 75$, $n_{Fluidized\ bed} = 18$, ● represent mass burn furnaces, ● represent fluidized bed furnaces. (B) From mass burn furnaces in sites whether to implement waste source separation, $n_{without} = 33$, $n_{with} = 42$.

indicating that the incineration process does have an effect in removing MPs.

3.2. Polymer types of MPs

In total, nine kinds of plastic were found in the bottom ash (Table S2): polyethylene (PE), polypropylene (PP), polyethylene terephthalate (PET), polyester (PES), polyamide (PA), polystyrene (PS), polyvinyl chloride (PVC), acrylonitrile butadiene styrene (ABS), polyurethane (PU) and epoxy. The μ-FTIR spectra of several sorted MPs are shown in Fig. S2. Except for samples from M9, the remaining 12 samples from mass burn furnaces contain either PE or PP. It can be demonstrated that PE and PP are two of the most commonly produced and used polymers. Packaging which is the main source of plastic waste is predominant. In samples from M2, M9 and F1, only one type of polymer was found, while in M7, nine types of plastic were found. Epoxy was exclusively identified in M10. PS, which is mainly used for packaging and construction materials, accounts for a high proportion, but mainly concentrated in M7 samples; this may be related to input waste containing about 25 % separated construction waste (Table S1). Among the samples from mass burn furnaces without waste source separation, the dominant categories of MPs were PP and PS (Fig. 2A). For samples from mass burn furnaces with source separation, the highest proportion of MPs was represented by PE, followed by PET/PES (Fig. 2B). Mostly, end-of-life textiles (fiber products) were incinerated or disposed with other solid waste even in source separation area (Geyer et al., 2017), which were the main source of PET/PES MPs in the bottom ash.

3.3. Size and shape

The size refers to the largest dimension of the MP particles. Because that plastic items will eventually undergo fragmentation to form MPs, plastic debris > 5 mm was also included in this research. The size distribution of MPs in the bottom ash is presented in Fig. 3. The size of the identified MPs from mass burn furnaces ranged from 95 μm to 15 cm, while for MPs from fluidized bed furnaces, sizes ranged from 0.3 mm to 6 mm. In total, 8 % of the plastic debris from the samples was larger than 5 mm. MPs were categorized into six classes depending on size, among which the classes of 50 μm–0.5 mm and 0.5–1 mm were dominant, accounting for 48 % and 26 %, respectively. The abundance of MPs increased with decreasing size, which indicates that unburned plastic debris in the bottom ash undergoes fragmentation to form MPs.

To describe the shape of the particles, MPs were categorized into four classes: granule, fragment, film and fiber (Fig. 4). Granules are pieces of irregular, thick plastic, with all three size dimensions comparable. Fragments represent their thickness is apparently lower than other two dimensions. Films are planar objects that are semitransparent and more flexible than fragments. The dominant type of MPs (Fig. 2D) was granules, which accounted for 43 %, followed by fragments and fibers, with 34 % and 18 %, respectively. Granule MPs were mainly PP (Fig. 2D), while fragment MPs were mainly composed of PE and PS, which may be caused by the large amount of packaging used. PET and PES account for 70 % of the fibrous plastics (Geyer et al., 2017) worldwide, which led to these being the dominant fibrous MPs in the bottom ash (Fig. 2D). Similarly to the MPs extracted from landfill leachate, granular and fragment MPs in the bottom ash had irregular shapes and rough edges (He et al., 2019), which also implies that the remaining plastic debris in the bottom ash could gradually undergo fragmentation to form MPs.

4. Discussion

4.1. Method assessment

The aims of this study was to investigate bottom ash for microplastics using μ-FTIR. However, there is no standardized methods available for plastic quantification in solid matrix. MPs can be easily extracted from sediment by floatation using high density solution and organic matter can be degraded by chemical reagent (Klein et al., 2015; Mani et al., 2019). However, bottom ash contains about 0.1 % inorganic lightweight suspended solid that is lighter than saturated NaCl solution, which interferes the MPs exaction. Thereby, we developed and adjusted some methodology for exacting MPs from bottom ash, which include sieving the bottom ash and mixing HF solution with fine particles. Some kinds of polymers like PET denser than 1.2 g/mL and particles adhered on the edges of glass vessel may cause an underestimation of MPs. Besides, the exclusion of partially burned polymers and black MPs when optical sorting contributed to underestimate MPs abundance. Therefore, the present work reported a conservative MPs abundance in bottom ash. The method for exacting and identifying MPs in bottom ash is a challenge and the techniques need to be further developed.

4.2. MPs occurrence patterns under different incineration conditions

The gas distribution inside the bed provides good mixing between wastes and bed materials, such as silica sand, limestone, alumina or ceramic material (Chandler et al., 1997), which improves the heat transfer efficiency and contact probability (Chang and Wey, 2006). The LOI of the fluidized bed furnace bottom ash was lower than that of the mass burn furnace bottom ash (Table 1), which meant that the fluidized bed furnace was more efficient for waste combustion. Therefore, the abundance of MPs from fluidized bed furnaces was lower than that from mass burn furnaces. In addition, the density of the bed materials was about 1.5 g/mL, which is denser than most of the commercial plastics (0.8–1.4 g/mL). Therefore, plastic waste less dense than bed material was circulated and kept burning in the furnace, which caused that there were fewer MPs in the bottom ash from the fluidized bed furnaces.

Furthermore, the low heat value (LHV) of food waste was 14.59 ± 1.55 MJ/kg owing to its high water content (68.0 ± 5.8 %), whereas plastic waste and paper contaminated by other wet materials

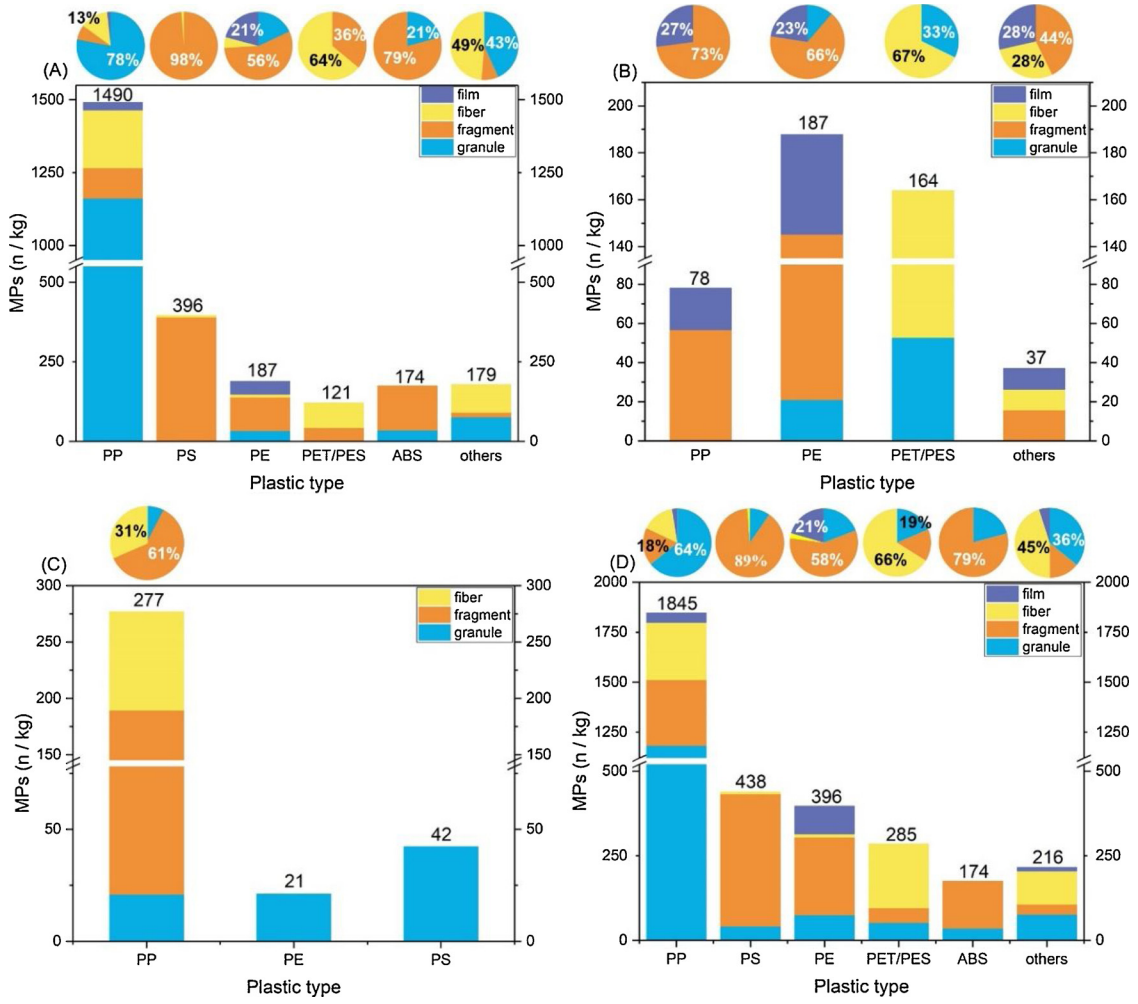

Fig. 2. Abundance of MPs in different shapes and in different material types. MPs from mass burn furnaces in sites without waste source separation (A) and with source separation (B). MPs from fluidized bed furnaces (C). MPs from all (D).

have higher LHV (31.51 ± 2.02 MJ/kg and 15.52 ± 1.14 MJ/kg, respectively) (Yang et al., 2018). With source separation, the proportion of food residue decreased from 64 % to 13 %, and the proportion of plastic and paper with a lower degree of contamination, increased from 28 % to 38 % (Table S1). Obviously, the LHV of MSW increased with the practice of source separation, which resulted in more complete combustion and fewer remaining MPs.

However, the abundance of MPs did not show any positive correlation with LOI (Fig. 5A). There were three possible reasons for this:1) non-plastic organics, such as lignocellulosic plant tissue and cardboard, were present in unburned materials; 2) the bottom ash contained incompletely oxidized metals, especially in fine fraction (Enzner et al.,

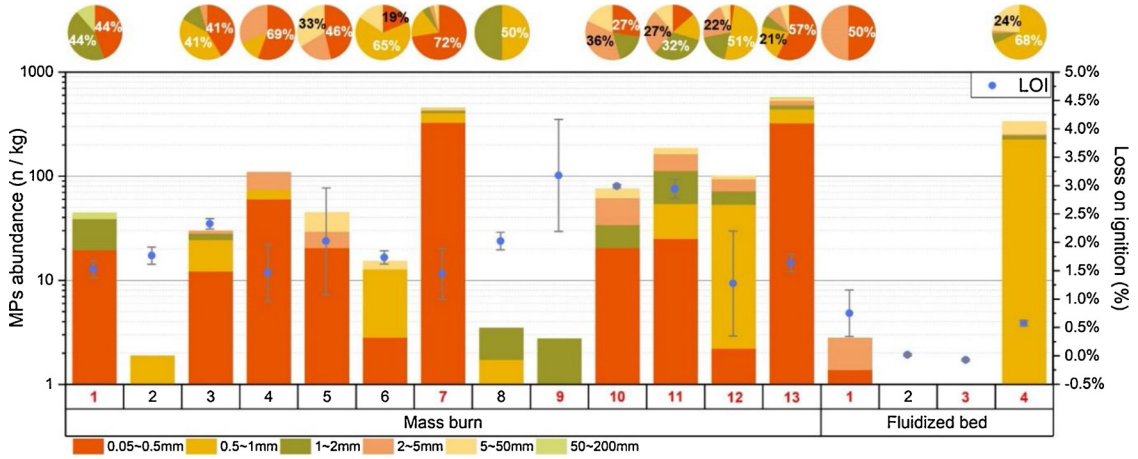

Fig. 3. Size fractions of MPs from different sites. The red number at the bottom line represent the sites without waste source separation, while black number represent the sites with waste source separation.

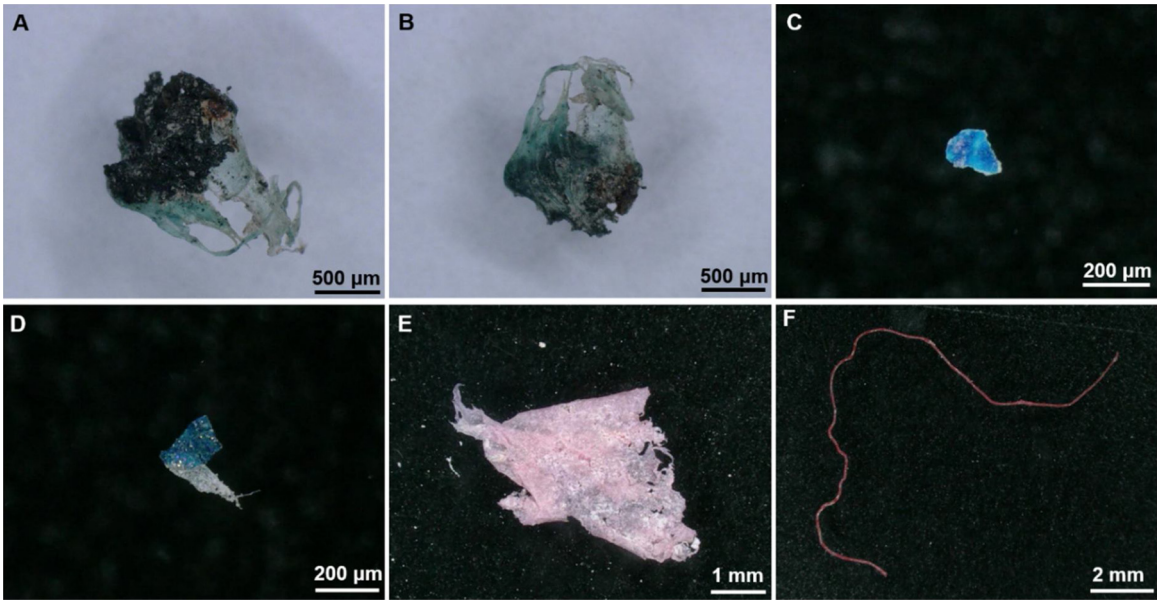

Fig. 4. Shape of MPs observed in bottom ash. Examples of (A, B) a granule in different sides, (C, D) fragment, (E) film, and (F) fiber.

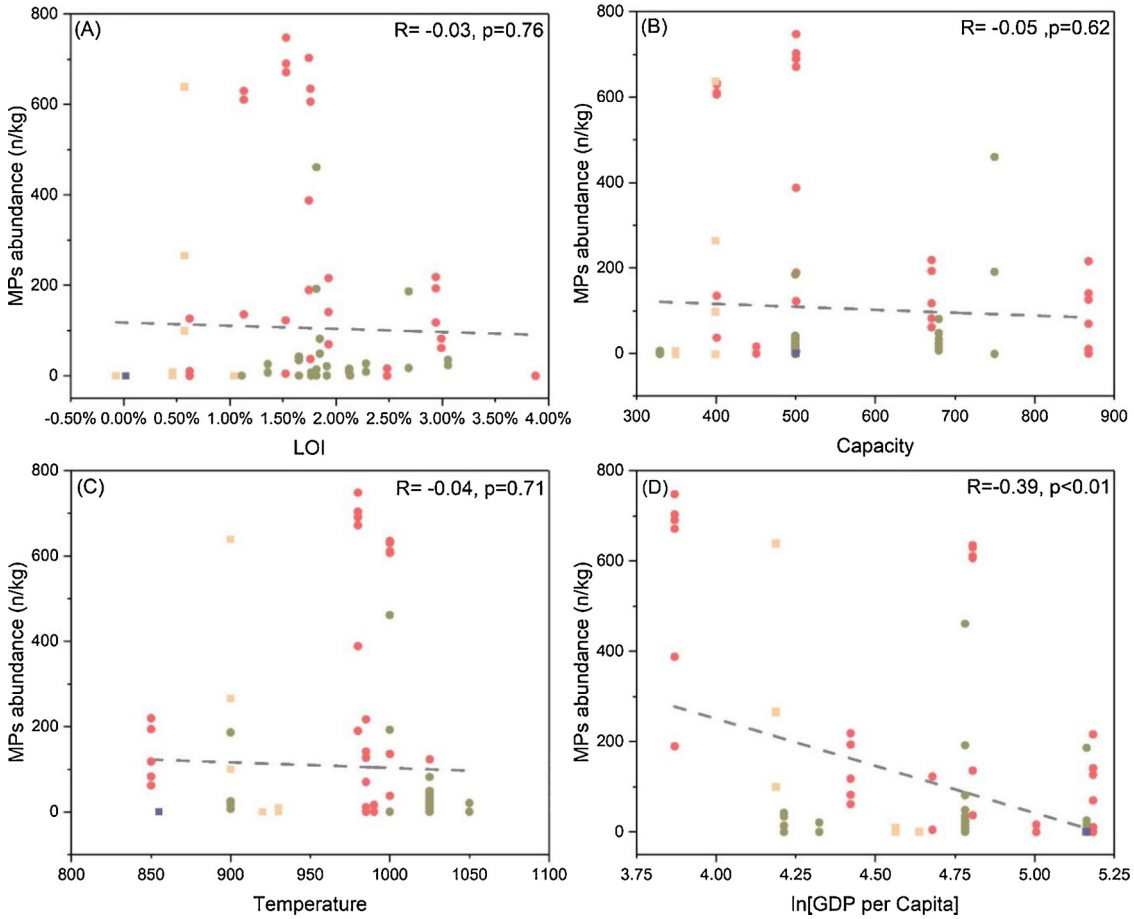

Fig. 5. Relationship between MPs abundance in bottom ash. (A) with LOI, (B) capacity of furnaces, (C) furnace temperature and (D) GDP (gross domestic product, thousand RMB) per capita. (●) and (■) represent mass burn and fluidized bed furnaces from sites without waste source separation. (●) and (■) represent mass burn and fluidized bed furnaces from sites with waste source separation.

2017), which may be responsible for the constant or even increased weight of the samples (e.g. samples from F2 and F3) after ignition; 3) the LOI indicates the weight of the unburned components in each sample. In river shore sediments, the smaller size fractions of MPs contribute most to their total numerical abundance; in contrast, the MPs mass concentration of sediments were determined in the bigger fraction (Klein et al., 2015). Therefore, there was no significant correlation between MPs abundance and the LOI of the samples investigated.

Fig. 5B illustrates the relationship between furnace capacity and abundance of MPs in the bottom ash. In general, owing to their insensitivity to fluctuations of waste input, furnaces with high capacity have an advantage in stable combustion. However, the abundance of MPs varied regardless of the capacity of the furnaces. The amount of unburned material should decrease with furnace temperature. However, no correlation was found between furnace temperature and abundance of microplastic in bottom ash, as shown in Fig. 5C. As shown in Fig. 5D, there was a negative correlation between the abundance of MPs and the GDP per capita of the sampling sites ($p < 0.01$). Although incineration was used to treat waste in low GDP per capita areas, a large amount of MPs would remain in the bottom ash, and this amount may be influenced by the characteristics of the waste accepted in the incineration plants. The waste accepted in the M7 incinerator contained construction and demolition waste, such as plastic tubes, wallpaper, and wood (Table S1). The plastics used for construction are difficult to burn because of the flame retardants they contain, so there was a high abundance of MPs in the bottom ash from M7. Usually, the proportion of food waste decreases as income levels rise, and goods consumed in high-income areas include more plastics than those in low-income areas. The GDP per capita of sites M10, M11 and M13 was relatively low, and with less plastic waste, the LHV of MSW were lower, which contributed to the higher abundance of MPs in the bottom ash. The furnaces in M12 were overloaded, which may cause insufficient incineration time; the waste was not well mixed, and there was insufficient contact with air in the primary combustion chamber (Gidarakos et al., 2009), leading to a high abundance of MPs in the bottom ash. Owing to the low bed pressure in furnace from F4, the plastic waste dropped out of the furnace with the bed material, and underwent fragmentation to form MPs.

4.3. Features of MPs in different MSW treatment systems

Granular MPs accounted for the majority of those in the bottom ash. Previous studies have reported that the dominant shapes of MPs in landfill leachate and compost are flakes and fragments, respectively (Su et al., 2019; Weithmann et al., 2018). This suggests that MPs in bottom ash are markedly different from those in landfill and compost, probably because planar flakes and fragments are more likely to be destroyed by combustion than granules. In addition, other shapes of plastic items will be partially melted into blocks, impeding internal heat transfer, which may contribute to their remaining in the ash. Small particles have larger surface areas, and therefore greater contact with the air, and better heat transfer efficiency than large particles. Therefore, the proportion of small particles should be low. However, the abundance of MPs in the bottom ash increased as sized decrease, which is consistent with the results summarized in landfill (Su et al., 2019), compost (Weithmann et al., 2018), and natural solid matrices (Liu et al., 2018; Piehl et al., 2018). This may be explained by the fragmentation of plastic debris caused by collision with the bottom ash during transportation.

In another study, 17 types of MPs were found in landfill leachate (He et al., 2019), which is more than the types of MPs found in the bottom ash. This result might be attributed to the fact that flotation liquid with a density of 1.7 g/mL (denser than 1.2 g/mL in the present study) was used to extract the MPs. Furthermore, the proportions of PE, PS and PP in the landfill refuse were 17 %, 16 %, and 7 %, respectively (Su et al., 2019). However, PP was found to be the primary MPs in the bottom ash. PP is widely used for food packaging for use in microwaves, owing to its better heat resistance than PE and PS. PP with additives such as flame retardants is also widely used in electronics, construction, and transportation (Hahladakis et al., 2018) and has a higher melting temperature and melt flow index than PE, which may cause the PP waste to melt and to adhere to organic waste, preventing sufficient incineration(Andrady, 2003). The proportion of PS in the bottom ash was also higher than that in landfill refuse. When PS is used in pipelines and building materials, flame retardants are added, which increases the ignition temperature of the product and makes it difficult to burn. Styrene-based polymers (including PS and ABS) accounted for 45 % of the MPs in compost, followed by PP (18 %) and PE (14 %) (Weithmann et al., 2018), while our study found that PS and ABS accounted for 22 %, ranking the second most proportion type of MPs. Their use in food packaging and disposable tableware means that PS, PP and PE are more likely to be mixed with food waste when they go into furnaces at sites without source separation of waste, and the increasing humidity levels may lead to insufficient incineration.

4.4. Estimation of quantities of MPs in the bottom ash and their potential risk

Despite many studies have evaluated the environmental risks of heavy metals and polycyclic aromatic hydrocarbons in bottom ash by leaching tests (Van Gerven et al., 2005). However, no research has been found that assessed the risk to surrounding environment of MPs in bottom ash. Generally, for bottom ash, metal recovery and granulation take place after 4–12 weeks of aging, which may contribute to MPs being atmospherically transported. Similarities between bottom ash and atmospheric PM_{10} collected adjacent to bottom ash dump sites have been found (Rigo et al., 2009). In addition, unburned black fragment MPs have been collected from the marine atmosphere (Liu et al., 2019), and bottom ash reused in coastal areas might be the primary contributor to these.

Many studies assessing mismanaged plastic waste entering the ocean and fresh water have classified plastic waste handled by incineration, sanitary landfill, and recycling as adequately managed waste (Jambeck et al., 2015; Lebreton and Andrady, 2019). However, ignoring these treatment or disposal methods may also cause release of MPs into the environment. He et al. measured the MPs abundance was 0.42–24.58 MPs per liter in leachate from southern China (He et al., 2019). It was 0–4.51 MPs per liter in the leachates from Nordic countries and the mean quantities of MPs released to environment was 334.8 thousand per year (Praagh et al., 2018). Considering the regulations about MSW incineration, China limited the LOI of bottom ash from MSW incinerators to 5 % (Ministry of Ecology and Environment, 2014). In European countries the LOI of bottom ash is also less than 5 % (EU, 2010), while in United States, it is controlled between 3 %–5 % (Lam et al., 2010). Therefore, as long as the LOI is not being zero, unburned material which can contain MPs will exist in bottom ash from incinerators no matter well-managed or not in different countries. We made a conservative estimation of the quantities of MPs in China (China Statistical Yearbook, 2019), Europe (Eurostat, 2018), the United States and some other countries that have established comprehensive solid waste treatment systems, using the available waste management data (see Table S3). Considering that mass burn furnaces account for the majority of thermal disposal furnaces worldwide, that the bottom ash generation rate is recorded as 18 %, and that there are 2–565 MP particles per kilogram of bottom ash, each MT of input-furnace waste will produce 360 to 102,000 MP particles in the bottom ash. The quantities of MPs in composting and anaerobic digestion were also estimated: 12 million MT of biowaste translated into more than 5 million MT of compost in Germany in 2013, and 1 kg compost contains approximately 50 % dry weight content conservatively, with 14–895 MP particles per kilogram dry weight (Weithmann et al., 2018). In recent research, the abundance of MPs in sludge compost were in the range of 150–410 MP particles per kilogram (Zhang et al., 2020b), which was within the range of conclusion by Weithmann et al. (Weithmann et al., 2018). Therefore, each MT of biowaste will produce 2900 to 186,000 MPs. The quantities of MPs from compost in Germany range from 26.48 billion to 1.69 trillion, which is a bit lower than the estimates from recent work (35 billion to 2.2 trillion) (Weithmann et al., 2018), so our estimate is conservative. Therefore, the abundance of MPs produced by waste incineration is lower than that produced by composting, per MT of waste. However, globally, the proportion of

waste incinerated (11 %) is much higher than that composted (5.5 %) (Silpa et al., 2018). Therefore, MPs quantities vary from 107 billion to 30 trillion per year in bottom ash, which is in the same order of magnitude as the quantities in compost, which range from 336 billion to 22 trillion per year worldwide. The quantities of MPs produced annually by waste incineration is estimated to vary from 36.72 billion to 10.37 trillion, which is an order of magnitude lower than those in sludge used as fertilizer in China (Li et al., 2018). Besides, 20.5–1000.3 trillion microbeads from cosmetic products are released annually from waste water treatment plants (WWTPs) to surface waters in mainland China (Cheung and Fok, 2017), surpassing the quantities of MPs in bottom ash by two orders of magnitude. In United States, the quantities of microbeads from facial scrubs (1.1–8.4 trillion per year) discharged to the aquatic environment also approximates the quantities of MPs in bottom ash (Mason et al., 2016). According to a recent study in Korea, the removal rate for WWTPs ranged from 98.7 %–99.99 %, and the per capita production of MPs entering WWTPs was at least 4000 day^{-1} (Park et al., 2020). It is conservatively estimated that at least 7.54 billion MPs are discharged from WWTPs to the environment annually, which is of the same magnitude as those in bottom ash. Therefore, in countries with advanced WWTPs, the quantities of MPs in wastewater effluent and in bottom ash may be comparative.

5. Conclusions

For the first time, the quantities of MPs exacted from MSW incinerator bottom ash were reported. And the abundance of MPs ranged from 1.9–565 n/kg. The results suggested incineration was not the terminator of plastic waste, and bottom ash remained as a potential source of MPs. The predominate shape and types of the MPs in bottom ash were granules and PP, respectively. Besides, the abundance of MPs increased with decreasing size, which indicates that plastic debris may be crushed into MPs during the whole process of incineration. The abundance of MPs from sites with waste source separation was significantly lower than that from sites without waste source separation, suggesting the positive impact of waste source separation for MPs management. The negative correlation between abundance of MPs and GDP per capita indicated that MPs abundance were closely related to the composition of waste. It was estimated that per metric ton of input-furnace waste will produce 360 to 102,000 MPs in the bottom ash, then highlighting the necessity to revise the MPs emission from managed waste.

CRediT authorship contribution statement

Zhan Yang: Investigation, Resources, Data curation, Methodology, Writing - original draft. **Fan Lü:** Investigation, Resources, Visualization, Writing - review & editing, Project administration. **Hua Zhang:** Supervision, Writing - review & editing, Formal analysis. **Wei Wang:** Investigation, Resources. **Liming Shao:** Supervision, Data curation. **Jianfeng Ye:** Investigation, Resources. **Pinjing He:** Writing - review & editing, Funding acquisition, Project administration, Validation.

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 material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.jhazmat.2020.123429.

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