

# Emissions of polycyclic aromatic hydrocarbons (PAHs) during hydrothermally treated municipal solid waste combustion for energy generation



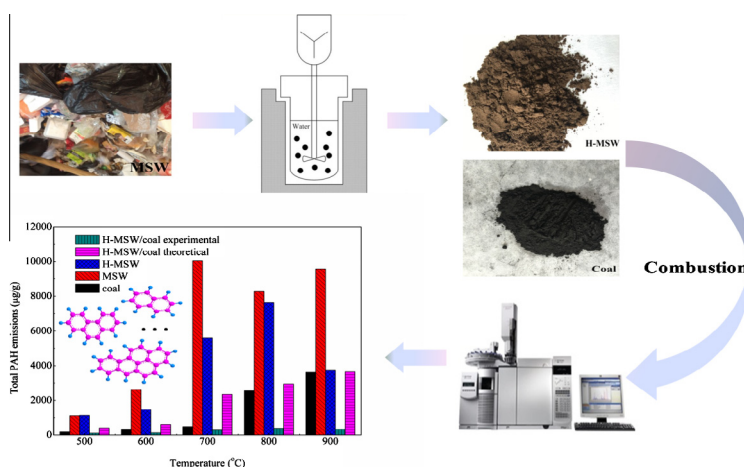
Nana Peng, Zhengang Liu\*, Tingting Liu, Chao Gai

Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China  
University of Chinese Academy of Sciences, Beijing 100049, China

## HIGHLIGHTS

- PAH emissions from H-MSW combustion and H-MSW/coal co-combustion were investigated.
- Compared to MSW, less PAHs were generated from H-MSW combustion.
- Interactions occurred between H-MSW and coal during co-combustion.
- The interactions suppressed PAHs formation and reduced the toxicity of PAHs.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Hydrothermal treatment (HT) is one of the efficient approaches for upgrading municipal solid waste (MSW). In the present study, emission characteristics of polycyclic aromatic hydrocarbons (PAHs) from hydrothermally treated municipal solid waste (H-MSW) combustion alone and H-MSW/coal co-combustion were investigated at different temperatures. The results showed that for all fuel combustion, the majority of PAHs were 3- or 4-ring PAHs. In addition, flue gas had the highest yields of PAHs followed by fly ash and bottom ash, while the ring number of dominated PAHs in fly ash was higher than those in flue gas and bottom ash. Compared to MSW, H-MSW combustion generated less PAHs at the value of 1131.95–7649.24 μg/g. The blending of H-MSW and coal reduced total PAH emissions and positive interactions were observed between H-MSW and coal during co-combustion. The toxicity equivalent quantity (TEQ) values of the PAHs from combustion were in the order MSW > H-MSW > H-MSW/coal, which was consistent with the total PAH emissions. The present study illustrated that significant reduction of PAH emissions and toxicity from combustion could be achieved by HT and the blending of H-MSW and coal.

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\* Corresponding author at: 18 Shuangqing Road, Beijing 100085, China.

E-mail address: [zgliu@rcees.ac.cn](mailto:zgliu@rcees.ac.cn) (Z. Liu).

## 1. Introduction

About 1.7–1.9 billion metric tons of municipal solid waste (MSW) is generated each year in the world [1]. Therefore, the disposal of municipal solid waste (MSW) is emerging to be the most urgent and important task because of the ever-increasing quantities and the harmfulness on environment and health, especially for developing countries [2–4]. The most common disposal method of MSW is landfilling in the world due to the low technical requirements [1]. However, landfill suffers from serious secondary pollution, such as pollution of air, ground water and soil. In addition, the lack of land resource also restricts the application of landfilling. Recently, MSW incineration has received considerable attention as a promising method for effective energy recovery, significant volume reduction (by up to 70% in weight and 90% in volume) and high degree detoxicity [5,6]. However, high moisture content, low calorific value and heterogeneous nature are the main drawbacks associated with the incineration of MSW and as a result, MSW combustion has low energy efficiency and relatively high pollutant emissions [7]. Therefore, pre-treatment is needed to homogenize raw MSW and improve its fuel property prior to combustion.

Hydrothermal treatment (HT), as a pre-treatment process, has been widely applied for upgrading of biomass waste, especially for high moisture content of biomass feedstock [8–10]. Compared to pyrolysis pretreatment, HT exhibits low energy consumption and high conversion efficiency owing to obviating the drying process and conducting at low temperature [11,12]. To date, numerous studies have been reported about HT process on upgrading of lignocellulosic biomass but limited studies are available about HT of MSW. Lu et al. investigated fuel quality and combustion behavior of hydrothermally treated MSW (H-MSW) and the results showed that HT improved fuel property of MSW, such as lowered moisture content, homogenized shapes and increased energy density [7]. Similarly, it was reported that the heating value of MSW was improved by HT through high formation of fixed carbon [13]. Apart from HT, co-combustion of MSW and coal is another effective and economical option, which can not only increase the calorific value but also buffer the impact of the variable qualities of MSW on combustion process [14]. With the increasing fuel quality of H-MSW, further improved combustion behaviors are expected during co-combustion of H-MSW and coal compared to MSW and coal co-combustion. Muthuraman et al. studied the co-combustion behavior of H-MSW and low rank coal using thermogravimetric analysis and the results showed that the blending of H-MSW and coal had improved ignition characteristics and reduced unburned carbon compared to low rank coal [15]. Additionally, nitrogen evolution during co-combustion of H-MSW and coal in a bubbling fluidized bed was investigated, indicating that 30% of H-MSW addition lowered the formation of NO and N<sub>2</sub>O [16]. Similarly, lower CO emission was achieved by 20% H-MSW addition to coal during combustion in a lab-scale fluidized bed reactor [17].

Polycyclic aromatic hydrocarbons (PAHs), mainly emitted from the pyrolysis or incomplete combustion of fuel, is a global environmental issue due to the carcinogenicity, teratogenicity and mutagenicity [18–20]. Nowadays, the PAH emissions standards are increasingly getting strict around the world to reduce their environmental impacts. Therefore, the formation mechanisms of PAHs during coal and MSW combustion alone have been extensively studied. For instance, Shen et al. reported PAH emissions from five coals combustion and the results showed that among the examined factors, the PAH emissions were closely related to the moisture and volatile matter content of the coal [19]. The distribution of PAHs in gaseous and particulate phases from coal combustion using a coal-stove was investigated and it was found that 95% of

the total PAHs were in gaseous phase [21]. In addition, PAHs in bottom ashes from MSW incinerator were analyzed and the results showed that 3- and 4-ring PAHs were dominated in all samples [22]. Zhang et al. determined PAH contents in fly ash from MSW incinerator where 3- and 4-ring PAHs were the main constituents of PAHs [23]. However, to our best knowledge, there is no systematic study on the emission, distribution and toxicity of PAHs from H-MSW combustion alone and H-MSW/coal co-combustion.

With the increasing importance of HT, understanding emission and distribution of PAHs from H-MSW combustion is urgent for its large-scale application. Therefore, the objective of this study aimed to (1) determine the emission, distribution and toxicity of PAHs in bottom ash, fly ash and flue gas during H-MSW combustion, (2) investigate the effect of HT on PAH emissions during MSW combustion and (3) examine the effect of the blending of H-MSW and coal on PAH emissions during co-combustion. The ultimate goal of the present study was to realize the effective and clean energy recovery from MSW.

## 2. Materials and methods

### 2.1. Materials

In this study, bituminous coal was obtained from a coal mine in Inner Mongolia, China. The simulated MSW sample was chosen in the present study and its composition can be found in our previous study [24]. Briefly, MSW was comprised of food residue (64.93 wt.%), wood waste (1.48 wt.%), paper (12.94 wt.%), textiles (3.11 wt.%), and PVC (15.07 wt.%). All samples were dried in an oven at 105 °C for 24 h before use.

HT of MSW was carried out in a laboratory semi-batch 2.5-L autoclave reactor. The mixture of MSW and deionizer water (1:3 mass basis) was loaded into the reactor, and the reactor was sealed and heated to 200 °C. After remaining for 30 min, the reactor was cooled rapidly to room temperature. The mixture was separated by vacuum filtration and the solid product (H-MSW) was dried in oven at 105 °C for 24 h. The H-MSW/coal blend was prepared using coal weight fraction of 75%. The metal contents of coal and H-MSW were determined by inductively coupled plasma-optical emission spectroscopy (ICP-OES). Around 0.1 g of dry sample was digested in mixed acids (4 ml 65% HNO<sub>3</sub>, 4 ml 30% H<sub>2</sub>O<sub>2</sub>, 2 ml 70% HClO<sub>4</sub> and 4 ml 48% HF) at 170 °C for 12 h. The digestion liquid was evaporated to near dryness and then dissolved in 1:1 HNO<sub>3</sub> for ICP-OES analysis. The elementary analyzer (a Vario MACRO cube Elemental Analyzer) was used to determine the ultimate analysis of the samples. Proximate analysis was obtained according to standard GB/T212-2008. Table 1 illustrates proximate analysis, ultimate analysis and metal contents of the coal and H-MSW.

### 2.2. PAHs sampling and analysis

The laboratory scale fixed-bed tubular quartz reactor (an internal diameter of 55 mm and a length of 1000 mm) was applied for combustion experiments in this study. When the reactor was heated to the desired temperature, the fuel sample (around 1 g) placed in the quartz boat was inserted into the center zone of reactor. The fuel was combusted at the desired temperature for 30 min with air flow rate of 500 ml/min. Quartz fiber filters (QFFs) was used to collect fly ash. The PAHs in flue gas was determined by adsorption method. The adsorption apparatus were composed of a glass holder filled with XAD-2 and two bottles of dichloromethane (DCM), which were placed in ice baths. The residue in quartz boat was collected as bottom ash. After complete combustion and cooling down to room temperature, QFFs, XAD-2, absorption liquid and the bottom ash were analyzed for PAHs.

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