



Characterisation of polycyclic aromatic hydrocarbons in flue gas and residues of a full scale fluidized bed combustor combusting non-hazardous industrial waste



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ABSTRACT

This paper studies the fate of PAHs in full scale incinerators by analysing the concentration of the 16 EPA-PAHs in both the input waste and all the outputs of a full scale Fluidized Bed Combustor (FBC). Of the analysed waste inputs i.e. *Waste Water Treatment (WWT) sludge, Refuse Derived Fuel (RDF) and Automotive Shredder Residue (ASR)*, RDF and ASR were the main PAH sources, with phenanthrene, fluoranthene and pyrene being the most important PAHs. In the *flue gas* sampled at the stack, naphthalene was the only predominant PAH, indicating that the PAHs in FBC's combustion gas were newly formed and did not remain from the input waste. Of the other outputs, *the boiler and fly ash* contained no detectable levels of PAHs, whereas the *flue gas cleaning residue* contained only low concentrations of naphthalene, probably adsorbed from the flue gas. The PAH fingerprint of the *bottom ash* corresponded rather well to the PAH fingerprint of the RDF and ASR, indicating that the PAHs in this output, in contrast to the other outputs, were mainly remainders from the PAHs in the waste inputs. A PAH mass balance showed that the total PAH input/output ratio of the FBC ranged from about 100 to about 2600 depending on the waste input composition and the obtained combustion conditions. In all cases, the FBC was clearly a net PAH sink.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a group of hundreds of chemically related environmentally persistent organic compounds with various structures and varied toxicity (Agency of Toxic Substances and Disease Registry, 1995). PAHs are typical products of incomplete combustion (Vehlow et al., 2006). Some PAHs, including benzo(a)pyrene and chrysene, are classified as probable or possible human carcinogen by the International Agency for Research on Cancer (IARC). In comparison with polychlorinated dibenzo-dioxins and furans (PCDD/Fs), which are other typical products of incomplete combustion, the formation mechanisms and especially the emission of PAHs from waste combustors received little attention so far.

Although in modern waste combustors air supply, mixing of air and waste and gas residence time are carefully controlled, the presence of low concentrations of PAHs in the flue gas and solid

residues cannot be avoided. Only few studies on PAH formation and destruction in waste combustors have been reported. Thomas and Wornat (2008) studied the formation and fate of PAHs from complex solid fuels as a function of temperature (500–1000 °C) and oxygen supply (zero to near stoichiometric). They distinguish two competing effects of oxygen. The first is the “radical enhancement effect”, involving increased formation of free radicals with increasing oxygen rate (OR) (OR equals 1 if a stoichiometric amount of oxygen is supplied), leading to a higher yield of pyrolytic breakdown products, which recombine to form PAHs. The second effect is the “oxidative destruction effect”, involving an increased oxidative breakdown of earlier formed pyrolysis products, including PAHs, with increasing OR. The relative importance of both effects is highly dependent on the combustion temperature: at temperatures below 800 °C and $OR < 1$, the radical enhancement effect is more important and an increase in oxygen supply increases PAH formation. At temperatures above 800 °C and $OR < 1$, oxidative destruction becomes more important and an increase in oxygen supply will decrease the PAH formation rate (Thomas and Wornat, 2008). Herrera et al. (2002) studied the formation of PAHs during thermal degradation of polyurethane elastomers. They burned two different types of polyurethane and

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analysed the combustion gas. Of the 16 EPA PAHs measured, naphthalene occurred in the highest concentration. [Herrera et al. \(2002\)](#) conclude that PAHs are formed by precursor recombination: in a first stage benzene is formed that further “grows” to higher molecular weight PAHs. Indeed, the concentration of naphthalene (2 rings) was higher than the concentration of phenanthrene (3 rings) whose concentration again was higher than that of pyrene (4 rings). The PAH concentrations measured in the combustion gas were very similar for the different polyurethanes, so PAH formation did not seem to depend on the molecular structure of these elastomers. [Kwon and Castaldi \(2012\)](#) studied the thermal degradation of tires in air atmosphere and also in these experiments, the concentration of naphthalene was the highest of all PAHs analysed in the combustion gas.

In waste combustors, the first stage of the combustion process takes place in a furnace where only stoichiometric amounts of oxygen are present. The waste is volatilised and pyrolysed and the pyrolysis products recombine to form PAHs in the raw combustion gas. In the second stage, that takes place in a post combustion chamber, secondary air is supplied to the raw combustion gas and the volatilised products and earlier formed PAHs are destroyed by oxidation, leading to higher temperatures than in the primary combustion stage ([Watanabe and Noma, 2009](#)). The fact that PAHs found in raw combustion gas are newly formed by pyrosynthesis and not directly evaporated from the waste was demonstrated by [Atal and Levendis \(1997\)](#), who added deuterium spiked PAHs to the waste feed of a lab scale furnace. None of these deuterium spiked PAHs were detected in the raw flue gas, so they were destroyed during combustion and the PAHs in the flue gas were newly formed. [Watanabe and Noma \(2009\)](#) determined the concentration of PAHs, expressed in $\text{mg (t waste input)}^{-1}$, in the raw combustion gas of a pilot scale rotary kiln incinerator fed with municipal solid waste (MSW) and showed that the concentration of PAHs was 1 (combustion temperature 890 °C) to 3 (combustion temperature 690 °C) orders of magnitude higher than in the waste input. They come to similar conclusions as [Atal and Levendis \(1997\)](#) and [Thomas and Wornat \(2008\)](#) i.e. that PAHs were formed in the primary combustion zone and at higher rates at 690 °C than at 890 °C. [Watanabe and Noma \(2009\)](#) also showed that the PAH formed in the kiln were largely destroyed in the secondary combustion chamber: the total PAH concentration of the combustion gas leaving the secondary combustion chamber was 3–4 orders of magnitude lower than the PAH concentration in the raw combustion gas entering the chamber, corresponding to a destruction efficiency >99%. Filtration over an active carbon filter and wet flue gas treatment had no significant influence on the total amount of PAHs in the combustion gas.

Besides the combustion air, PAHs are also present in solid residues of waste incinerators. [Wheatley and Sadhra \(2004\)](#) studied PAH in the bottom and fly ash from a modern full scale clinical waste incinerator. They suggest that the PAHs in the bottom ash may be a combination of those present in the waste (which were not completely destroyed during combustion) and those formed in the primary combustion process. In the experiments of [Watanabe and Noma \(2009\)](#), the PAH concentration in the bottom ash of the pilot scale rotary kiln, expressed in $\text{mg (t waste input)}^{-1}$, was 1.5–15% of the PAH concentration in the input MSW. The highest concentrations were detected for the lowest kiln temperature applied (690 °C), which agrees with the results of [Thomas and Wornat \(2008\)](#). The total PAH concentration in the fly ash collected in a bag filter placed downstream of the boiler was much lower than the concentration in the bottom ash and corresponded to about 1% of the MSW input concentration, which was $3000 \mu\text{g kg}^{-1}$.

The findings on PAH formation in waste combustion processes discussed here ([Thomas and Wornat \(2008\)](#), [Herrera et al. \(2002\)](#), [Watanabe and Noma \(2009\)](#), [Atal and Levendis, 1997](#)), were

obtained in lab scale or pilot scale installations in which the parameters influencing PAH formation (e.g. temperature and oxygen rate) can be controlled within narrow margins. It is however not yet clear what the implications of these findings are for full scale installations. Only a few papers study the fate of PAHs in full scale waste incinerators and often these studies only report PAH concentrations in the incinerator outputs i.e. flue gas and/or ashes ([Johansson and Van Bavel, 2003](#); [Wheatley and Sadhra, 2004](#)). It should also be kept in mind that in full scale waste incinerators, in addition to the outputs i.e. ashes and flue gas, also the waste input contains PAHs. Therefore this study reports, to our knowledge for the first time, the concentration of the 16 EPA-PAHs in both the *input* waste and *all the outputs* i.e. flue gas, boiler and fly ash, flue gas cleaning residues and bottom ash of a full scale FBC combusting non-hazardous (industrial) waste. Furthermore, the concentrations of the individual PAHs or so called “PAH Fingerprint” of the waste inputs and outputs are compared in order to find out to which extent PAHs in the outputs remain from the input waste or are newly formed. Finally, a PAH mass balance is drawn up to determine the major PAH input and output streams and to establish whether the FBC is an overall PAH sink or source.

2. Materials and methods

2.1. FBC

The FBC under study was described in detail in previous work of the authors ([Van Caneghem et al., 2010a](#); [Van Caneghem et al., 2012](#)). In the FBC, high calorific RDF from mechanical biological treatment of MSW and comparable industrial waste afford the heat necessary for drying and incinerating low calorific Waste Water Treatment (WWT) sludge, and for steam and electricity generation. In 2011, the installation, which is situated in Flanders, Belgium, incinerated $560 \times 10^3 \text{ t}$ of mixed waste. The FBC is of the ROWITEC internal rotating fluid bed type in which a sand mass is fluidized by hot air. The RDF is together with the WWT sludge introduced and combusted in the fluidized sand bed. In the freeboard, which is the free space above the fluidized bed that acts as post combustion zone, a gas temperature of at least 850 °C, a residence time of at least 2 s and an oxygen excess of at least 6% are maintained to ensure proper burn out. In a vertical boiler, energy is recovered from the hot combustion gases. The produced superheated steam (40 bar, 400 °C) is sent to a turbine (capacity of 34 MW) for electricity production. The flue gas is dedusted in an electrostatic precipitator and flows through a flue gas cleaning installation consisting of a semi-dry reactor, a baghouse filter and a caustic soda scrubber. A mixture of clay and activated carbon particles (Dioxorb®) is injected in the semi-dry reactor to adsorb PCDD/Fs. At the bottom of the FBC, the sand and ashes are removed after which the sand is sieved of and sent back into the incinerator. The boiler and fly ash and the flue gas cleaning residues are solidified and stabilised and subsequently stored on a hazardous waste landfill. The bottom ash goes to a specialised facility for recycling. [Fig. 1](#) gives a schematic overview of the installation, indicating the average temperatures at the different stages of the combustion and flue gas cleaning process.

2.2. Waste types

The two main waste types combusted in the FBC are RDF and WWT sludge. However, occasionally also non hazardous industrial wastes are combusted. One of these wastes is Automotive Shredder Residue (ASR) and is considered in this study.

The RDF combusted in the FBC is one of the remaining fractions of mechanical biological treatment (MBT) of Flemish MSW. The

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