Destruction and formation of dioxin-like PCBs in dedicated full scale waste incinerators.

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Abstract

Destruction and formation of dioxin-like PCBs in full scale waste incinerators is studied by analysing input waste streams and boiler and fly ash of a grate furnace incinerator (GFI) incinerating MSW, of a Fluidised Bed Combustor (FBC) incinerating a mix of 50% sludge, 25% refuse derived fuel (RDF) and 25% automotive shredder residue (ASR) and of a rotary kiln incinerator (RKI) incinerating hazardous waste. The dioxin-like PCB fingerprints of the waste inputs show that PCB oils Aroclor 1242 and Aroclor 1254 late are the major dioxin-like PCB contamination source of sludge, RDF and ASR. The dioxin-like PCB fingerprints of the waste inputs are clearly different from the fingerprints of the outputs i.e. boiler and fly ash, indicating that in full scale waste incinerators dioxin-like PCBs in the input waste are destroyed and other dioxin-like PCBs are newly formed in the post combustion zone. The dioxin-like PCB fingerprint of boiler and fly ash of all three incinerators corresponds well to the fly ash fingerprint obtained in lab scale de novo synthesis experiments, indicating that dioxin-like PCBs are mainly formed through this mechanism. The high PCB concentration in the input waste mix of the RKI does not promote the formation of dioxin-like PCBs through precursor condensation.

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1. Introduction

In the post combustion zone of waste incinerators, at temperatures between 800 and 200 °C, toxic, persistent organic pollutants (POPs) are formed due to the presence of products of incomplete combustion, oxygen and chlorine (Altarawneh et al., 2009; Van Caneghem et al., 2010a). Over the last two decades, most research on POP formation in waste incinerators focussed on polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs), partly because of their high toxicity. Indeed, of all know chemicals, 2,3,7,8-TCDD is considered the most toxic to humans (Hites, 2011). Seventeen PCDD/Fs have been shown to have toxic responses similar to those caused by 2,3,7,8-TCDD and their toxicity relative to 2,3,7,8-TCDD is expressed by a toxic equivalency factor or TEF. Their total toxic equivalent (TEQ) concentration corresponds to the sum of the concentrations of the individual congeners multiplied by the respective TEF value (Vanden Bergh et al., 1998). In the post combustion zone of waste incinerators also polychlorinated biphenyls (PCBs) are formed (Lemieux et al., 2001; Ishikawa et al., 2007; Van Caneghem et al., 2010a,b). Twelve PCB congeners with no or only one chlorine in the ortho position show toxic responses similar to those of 2,3,7,8-TCDD and have also been assigned TEF values by the WHO (Vanden Bergh et al., 1998). The TEQ mass of these so called dioxin-like PCBs accounts for 3 - 20% of the total TEQ mass (sum of PCDD/Fs and dioxin-like PCBs) in the output i.e. the emitted flue gas and solid residues of waste incinerators (Dyke et al., 2003; Van Caneghem et al., 2010b). In spite of this relative importance, the destruction and formation of dioxin-like PCBs in waste incinerators received only little attention in the literature.

Generally it is assumed that PCBs are formed by the same mechanisms as PCDD/Fs (Lemieux et al., 2001; Jansson et al., 2011): homogeneous (gas phase) and heterogeneous (gas phase/solid phase) condensation of precursors, and de novo synthesis i.e. formation by chlorination and oxidation of residual carbon in the solid phase (ash particles). There is however a lack of comprehensive, detailed data on dioxin-like PCB congener concentrations in the inputs and outputs of full scale waste incinerators enabling to determine their formation mechanisms in these installations. Therefore this article gives and discusses experimentally measured dioxin-like PCB congener profiles or so called "fingerprints" of relevant waste inputs and of boiler and fly ash of three types of full scale waste incineration facilities. Where possible, the dioxin-like PCB fingerprints of the ashes are compared to the fingerprints typical for the different PCB formation mechanisms obtained in lab scale mechanistic studies. As in full scale installations parameters controlling PCB formation mechanisms, such as waste composition, temperature and oxygen concentration, cannot be controlled within narrow margins, this work does not comprise an in depth mechanistic study of PCB formation. It is merely an attempt to identify the dominant PCB formation mechanism based on analysis of incinerator inputs and outputs. Boiler and fly ash is preferred over other incinerator outputs such as flue gas or flue gas cleaning residue (FGCR), as it is collected in the boiler and in a filter positioned directly after the boiler in which the dioxin-like PCB s are formed, but before any flue gas cleaning e.g. injection of adsorbent that may influence the dioxin-like PCB fingerprint takes place.

2. Materials and methods

2.1 Installations

This paper considers three waste incinerators with various inputs and combustion technologies: a grate furnace incinerator (GFI) for municipal solid waste (MSW) incineration, a fluidized bed combustor (FBC) incinerating a mix of MSW or RDF of MSW, wastewater

treatment (WWT) sludge and non-hazardous industrial waste such as automotive shredder residue (ASR), and a rotary kiln incinerating miscellaneous hazardous waste.

In the considered GFI, located in Doel (North of Belgium), about 400 000 t/year of MSW and comparable industrial waste are incinerated at 850 °C. After heat recovery in a steam boiler, the incineration gases flow through a half-wet flue gas cleaning unit where a CaO-slurry and activated carbon particles are injected. The FGCR is subsequently retained in a baghouse filter and the gases are further cleaned in a wet gas washing system before release to the atmosphere. A more detailed description of the installation is given in Vandecasteele et al. (2007).

In the considered FBC, also located in Doel (North of Belgium), about 470 000 t/year of a mixture of WWT sludge, MSW and non-hazardous industrial waste are incinerated at temperatures of 850 °C. After heat recovery in a boiler, the incineration gases are dedusted in an electrostatic precipitator, after which they undergo further cleaning in the same way as described for the GFI. A more detailed description of the installation is given in Van Caneghem et al (2010b).

In the considered RKI, located in Antwerp (North of Belgium) about 90 000 t/year of hazardous industrial waste are incinerated at temperatures above 1100 °C. After heat recovery in a boiler, the incineration gases are de-dusted in an electrostatic precipitator (ESP) and then cleaned in a four-step wet gas washing unit. The hazardous waste incinerated is very heterogeneous and consists of hospital waste, used solvents, hazardous household waste, contaminated soil and chlorinated organic substances including PCB oils, POP pesticides etc.

2.2 Sampling and analysis

The sampling and analytical procedures were described in detail in previous work of the authors (Van Caneghem et al. 2010a; 2010b).The dioxin-like PCB concentrations were determined on representative input waste types and on boiler and fly ash sampled at the

different installations. From the different waste types, two pooled samples were obtained from samples of each truckload arriving at the incineration facilities on two different days. The analyzed solid residues collected in the post combustion zone are referred to as “boiler and fly ash” for each of the considered incinerators. For the GFI where no ESP is present, “boiler and fly ash” consists of the ash collected at the bottom of the boiler section, for the FBC and RKI it is a mixture of ash collected at the bottom of the boiler and of ash collected in the ESP located directly after the boiler. The boiler and fly ash samples of the FBC were taken when 50% of sludge, 25% of RDF and 25% of ASR were co-incinerated. In each of the considered installations, a boiler and fly ash sample was taken three times a day for five consecutive days during normal steady-state operation. The ashes are very homogeneous fine powders and at the end of the sampling period, for each incinerator, all collected samples were mixed thoroughly in a shaking device to obtain a representative pooled sample. The dioxin-like PCB concentrations were obtained by GC-MS after soxhlet extraction according to the German AbfKlärV, Annex1 (Bundesministerium fur Umwelt, Naturschutz und Reaktorsicherheit, 2006).The detection limit for the individually analysed congeners was 10 pg g_{dw}^{-1}. All analyses were performed by a certified laboratory that has to participate regularly in government-organised inter-comparisons for quality assurance.

3. PCB formation mechanisms

Generally it is assumed that PCBs are formed by the same mechanisms as PCDD/Fs (Fängmark et al., 1995;Lemieux et al., 2001; Jansson et al., 2011): homogeneous (gas phase) and heterogeneous (gas phase/solid phase) “condensation” of precursors (Liu et al., 2001; Hirota et al. 2003; Altarawneh et al., 2009), and de novo synthesis i.e. formation by chlorination and oxidation of residual carbon in the solid phase (ash particles) (Schoonenboom et al., 1995; Pekárek et al., 2001; Weber et al., 2001). This section

summarises information on dioxin-like PCB formation mechanisms from various papers written by different authors over the last decades.

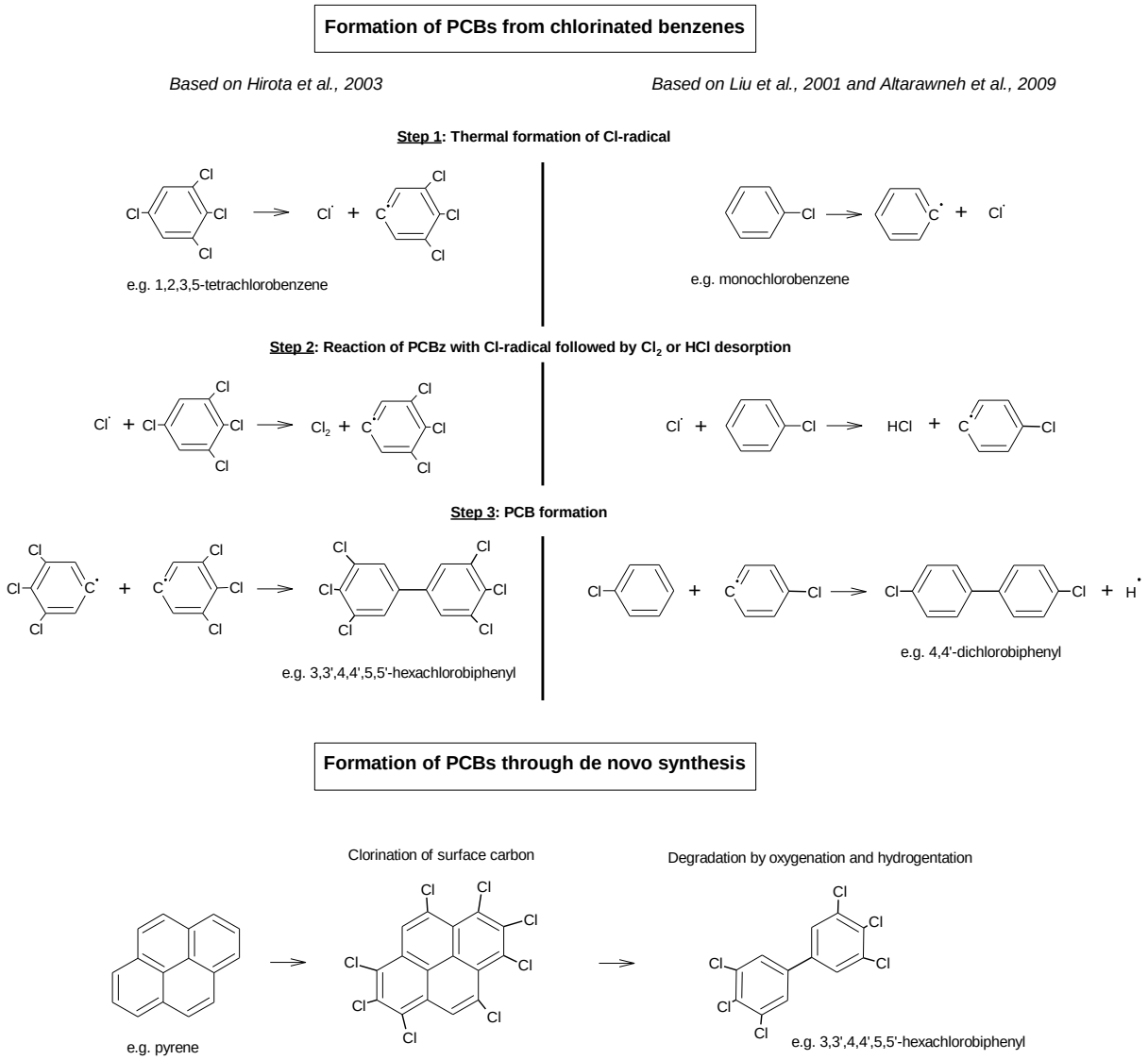


Figure 1: Schematic representation of different dioxin-like PCB formation mechanisms

Figure 1 illustrates the formation of dioxin-like PCBs through homogeneous precursor condensation by means of the reaction of two 1,2,3,5-TCBz molecules to give 3,3',4,4',5,5'-HxCB based on the work of Hirota et al. (2003).The first step involves the breaking of the C-Cl bond at the 5-position in 1,2,3,5-TCBz because at this position the bond dissociation

energy is the lowest. In a second step, the released chorine radical reacts with the chlorine atom at the 5-position of another 1,2,3,5-TCBz molecule to give a radical intermediate and a Cl_2-molecule. Finally, 3,3',4,4',5,5'-HxCB is formed by reaction of two 1,2,3,5-TCBz radicals formed in step 1 and/or step 2. The structural factor determining the formation rate is the C-Cl bond strength in 1,2,3,5-TCBz. Although the bond dissociation energy is the lowest for the C-Cl bond at the 5-position, it remains relatively high (460 kJ/mol), making it unlikely that this homogeneous formation pathway is significant for PCB formation in the post combustion zone of waste incinerators. Hirota et al. (2003) did not limit their study to the formation of 3,3',4,4',5,5'-HxCB, but also listed the chlorinated benzene precursors corresponding to the minimum energy path for the formation of all twelve dioxin-like PCBs. Liu et al. (2001) and Altarawneh et al. (2009) suggested homogeneous formation reactions similar to those of Hirota et al. (2003), where in the second step the chorine radical (formed in the first step) abstracts a hydrogen atom instead of a chlorine atom from the chlorinated benzene (Figure 1). By performing homogeneous precursor condensation experiments at temperatures between 250 and 450°C, Lemieux et al. (2001) showed there is maximum PCB formation at 350°C.

In the heterogeneous precursor reaction mechanism one of the reacting 1,2,3,5-TCBz precursor molecules (see Figure 1) is bound to a copper atom present in the solid particle surface. Hirota et al. (2003) demonstrated that the catalytic role of copper is dual: it stabilises the total energy of the system when the 1,2,3,5-TCBz adsorbs on the copper-cluster and it weakens the C-Cl bond strength due to the charge transfer from the 1,2,3,5-TCBz to the cluster. In this way, heterogeneous precursor condensation could be an important PCB formation mechanism in real waste incinerators. In contrast to PCDD/Fs, for which fingerprints have been reported for flue gas and fly ash formed in numerous lab scale experiments, no comprehensive experimental data on dioxin-like PCB fingerprints typical for

the homogeneous or heterogeneous precursor formation mechanism was found in the literature. Based on the Gibbs free energies of PCB formation reported by Holmes et al. (1993), PCB 156 is expected to be the most abundant in the dioxin-like PCB fingerprint of the homogeneous (gas phase) precursor formation pathway, followed by PCB 189, PCB 167 and 169, PCB 157, PCB 118, 123 and 126, PCB 105 and 114 and PCB 77 and 81 that have the highest formation energy. It should however be kept in mind that Holmes et al. (1993) determined the Gibbs free energies of formation at 25 °C, which is much lower than the gas temperatures at which dioxin-like PCBs are formed in the post combustion zone of waste incinerators.

Schoonenboom et al. (1995) demonstrated the formation of dioxin-like PCBs from carbon in lab scale incineration experiments at temperatures ranging from 300 to 400 °C, with maximum formation at 350°C. They suggest that the polyaromatic carbon structure in incinerator ash particles is broken down by chlorination of the surface carbon by electrophilic aromatic substitution followed by oxidative decomposition resulting in PCB formation and release. Predominantly 3,3',4,4'-substitited PCBs were formed in these de novo experiments. Also Weber et al. (2001) conducted lab scale de novo PCB formation experiments. They concluded that next to chlorination, also hydrogenation is an important step in the degradation of the polyaromatic carbon structure and hence in the de novo formation process of dioxin-like PCBs. As an example of the de novo formation pathway, Figure 1 gives the formation of 3,3',4,4',5,5'-HxCB from pyrene. Pekárek et al. (2001) conducted de novo lab scale experiments and reported typical fly ash congener distributions. In an oxygen rich atmosphere (10% O_2), which reflects the combustion conditions in modern full scale incinerators, PCB 126, 156, 189, 169 and 77 were predominantly formed. All of these dioxin-like PCBs are 3,3',4,4'-substitited, confirming the findings of Schoonenboom et al. (1995). In "combustion" experiments where pure N_2 was used, about 6 times less dioxin-like PCBs

were formed than in the 10% O_2 experiments. This confirms that oxygen plays an important role in the de novo formation mechanism of dioxin-like PCBs. Figure 1 shows that also chlorine radicals plays an important role in formation of PCBs. Gullett et al. (2000) showed that the amount of chlorine radicals present in the gas phase are however not directly correlated to the chlorine content of the waste, but merely depend on the applied combustion conditions.

4. Results and discussion

4.1 Dioxin-like PCB fingerprints of input waste

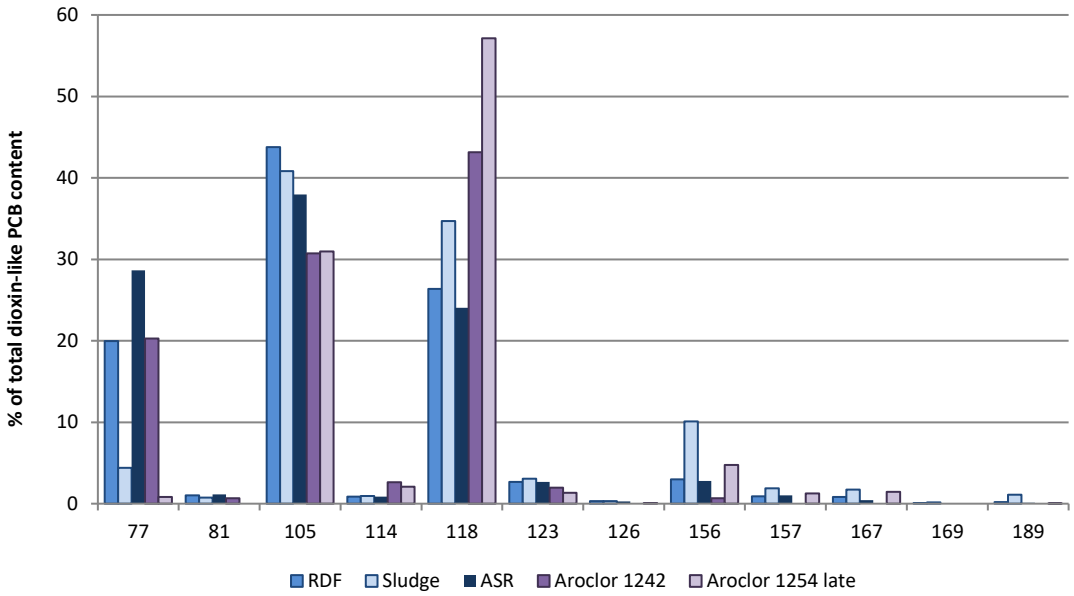


Figure 2: Dioxin-like PCB fingerprints of different input waste types

Figure 2 gives the dioxin-like PCB fingerprints of four types of waste incinerated in the three installations considered in this paper (see Section 2.1): RDF, industrial and municipal wastewater treatment sludge, ASR and the PCB oils Aroclor 1242 and Aroclor 1254 late. The origin and composition of the RDF, sludge and ASR were described in detail in previous work of the authors (Van Caneghem et al., 2010b).The RDF was obtained after MBT of Flemish MSW and is a mixture of mainly dried organic waste and plastics. The sludge is a

mixture of dewatered and digested sludge from industrial and municipal biological wastewater treatment plants. The co-incinerated ASR consists of the "heavy" ASR fraction, which is the remaining fraction after separation of ferrous, non-ferrous and plastic materials from shredded end-of-life vehicles. It contains a mixture of combustible (mainly PU-foam, plastic fibers and foil) and inert materials (sand, small stones, iron and rust particles). Aroclor 1242 and Aroclor 1254 were the main dioxin-like PCB containing Aroclors commercialised in the last years of PCB production by Monsanto (1974-1976) (ATSDR, 2000). As Aroclor was also the most applied PCB brand in Flanders (Covaci et al., 2011), they are considered representative for the obsolete PCB oils incinerated in the RKI in this study. The dioxin-like PCB fingerprint of these PCB oils shown in Figure 2 is based on the composition information given in the Toxicological Profile for PCBs of the ATSDR (2000). From Figure 2 it can be seen that the four waste types contain more or less the same main dioxin-like PCB congeners. Especially the fingerprints of the RDF, the ASR and the Aroclor 1242 correspond well, with PCB 77, 105 and 118 being the most important congeners. The sludge contains, compared to the other inputs, less PCB 77 and more PCB 156, indicating that the dioxin-like PCBs in this waste might also originate from Aroclor 1254 late. A general conclusion from Figure 2 is that PCB-oils such as Aroclor 1242 and Aroclor 1254 late are the main dioxin-like PCB contamination source for the considered incinerator waste inputs. The average total dioxin-like PCB concentration of the two pooled RDF, sludge and ASR samples was 8.57, 4.90 and 1,435 µg kg_{dw}^{-1}, corresponding to 3.84, 2.37 and 556 ng TEQ kg_{dw}^{-1}, respectively. Aroclor 1242 and Aroclor 1254 late contain 1.53 and 24% dioxin-like PCBs, respectively.

4.2 Dioxin-like PCB fingerprints of boiler and fly ash

Figure 3 compares the dioxin-like PCB fingerprint of the boiler and fly ash of the three incinerators. These "output" fingerprints are considerably different from the input waste

fingerprints depicted in Figure 2: the latter are dominated by only a few specific congeners i.e. PCB 77, 105 and 118, while the boiler and fly ash fingerprints show a more even distribution of the twelve dioxin-like PCB congeners. The apparent difference between input and output fingerprints indicates that dioxin-like PCBs in the waste are destroyed during combustion and that the dioxin-like PCBs found in the boiler and fly ash are newly formed.

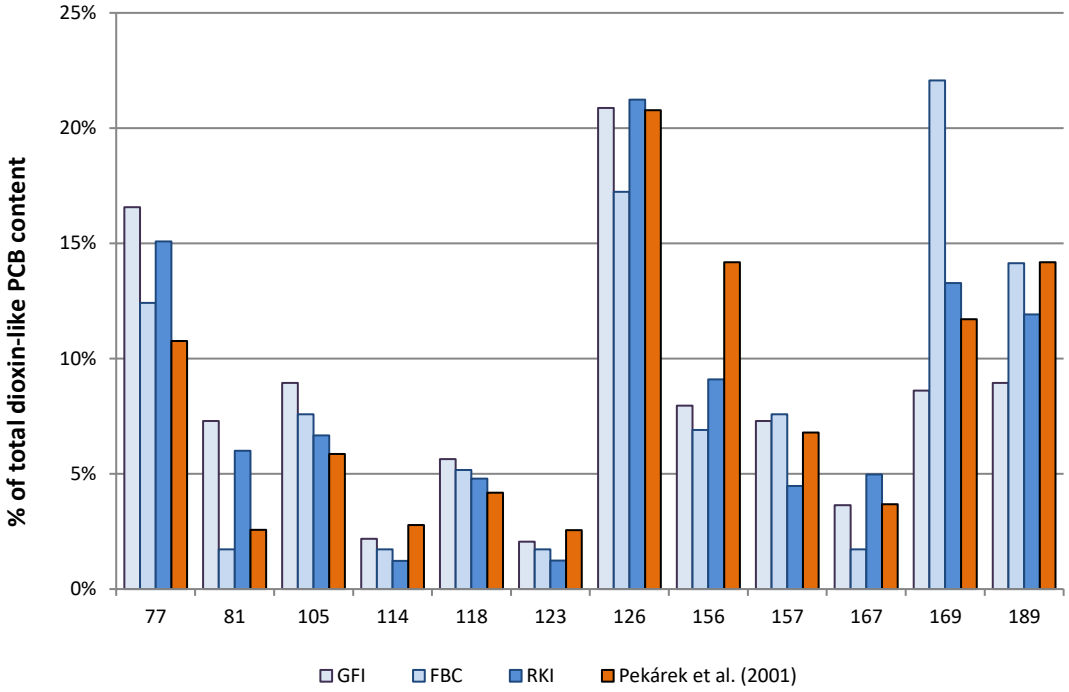


Figure 3: Dioxin-like PCB fingerprints of boiler and fly ash of different full scale waste incinerators and of fly ash of lab scale de novo experiments

The boiler and fly ash fingerprints of the different incinerators are rather similar, PCB 77, 126, 169, and 189 being the most abundant congeners. All these main congeners are 3,3',4,4'-substituted. Seven out of twelve (58%) dioxin-like PCBs are 3,3',4,4'-substituted, but in the boiler and fly ash of the considered incinerators these seven congeners account for at least 80% of the total amount of dioxin-like PCB. The relative abundance of 3,3',4,4'-substituted congeners was also reported by Jansson et al. (2011) for fly ash collected in two MSW incineration plants and confirms the de novo formation mechanism suggested by

Schoonenboom et al. (1995) (Section 3). The fact that the PCBs in the boiler and fly ash of the three full scale incinerators are formed through de novo synthesis is in addition confirmed by the correspondence of the boiler and fly ash fingerprints to the dioxin-like PCB fingerprints of fly ash obtained in lab scale de novo formation experiments by Pekárek et al. (2001) (bar most to the right for each congener in Figure 3). Another explanation for the dominance of de novo synthesis is the good waste burnout achieved in the incinerators resulting in very low chlorinated precursor concentrations in the combustion gas making homogeneous condensation reactions unlikely to take place. For this determination of the dominant formation mechanism it is assumed that precursor condensation formation yields dioxin-like PCB fingerprints that are distinctly different from the de novo PCB fingerprint, as is the case for PCDD/Fs. This seems very likely given the fundamental differences in the formation mechanisms and the relative concentrations that can be expected from the Gibbs free energy of formation (Section 3, Figure 1). Unfortunately, in the literature, no dioxin-like PCB fingerprints were found for ashes from heterogeneous precursor condensation experiments to further sustain this assumption.

In previous work (Vermeulen et al., 2013), the authors showed that in the RKI, because of the high PCB concentration in the input waste mix (Table 1) resulting in higher PCDD/F precursor concentrations in the boiler, heterogeneous precursor condensation was the dominant PCDD/F formation mechanism, in contrast to the GFI and the FBC where PCDD/Fs were predominantly formed through de novo synthesis. Apparently the PCB formation mechanism is not affected by the PCB concentration in the input waste as in all three incinerators it is de novo synthesis (Figure 3).

Table 1: Average dioxin-like PCBs concentration in the input waste (mix)

	PCBs in input waste (mix) %
GFI	$3.95\ 10^{-5}$ [a]
FBC	$1.69\ 10^{-6}$ [b]
RKI	1.5 [c]

[a] Indication based on average PCB concentration in RDF from MSW reported in Van Caneghem et al., 2010b

[b] reported in Van Caneghem et al., 2010b

[c] concentration of PCBs and chlorinated POP-pesticides reported in Van Caneghem et al., 2010a

5. Conclusions

The destruction and formation of dioxin-like PCBs was studied in three full scale waste incinerators: a GFI incinerating MSW, a FBC incinerating a mix of 50% sludge, 25% RDF and 25% ASR and a RKI incinerating hazardous waste containing 1.5% PCBs and chlorinated POP pesticides. The following main conclusion could be drawn:

- The major dioxin-like PCB contamination source of the analysed RDF, industrial and municipal wastewater treatment sludge and ASR are PCB oils Aroclor 1242 and Aroclor 1254 late.
- In full scale waste incinerators dioxin-like PCBs in the input waste are destroyed during incineration and others are newly formed in the post combustion zone.
- The dioxin-like PCB fingerprints of boiler and fly ash of all three full scale incinerators corresponds well to the fly ash fingerprint obtained in lab scale de novo synthesis experiments.

- The high PCB concentration in the input waste mix of the HWI does not promote the formation of dioxin-like PCBs through homogeneous or heterogeneous precursor condensation.

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