Mass balance for POPs in hazardous and municipal solid waste incinerators.

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Abstract

The amount of different persistent organic pollutants (POPs) in the input of waste incinerators was compared to that in the output. Three cases were considered: a rotary kiln incinerating hazardous waste, a grate furnace incinerating municipal solid waste (MSW) and the same grate furnace co-incinerating plastics of waste of electrical and electronic equipment (WEEE) and automotive shredder residue (ASR) with MSW. The mass balance for PCBs in the rotary kiln indicates that these POPs are destroyed effectively during incineration. The grate furnace can be a sink or source of PCDD/Fs and PCBs depending on the concentrations in the incinerated waste. In order to compare the total amount of POPs in input and output, a methodology was developed whereby the amount of POPs was weighed according to

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minimal risk doses (MRDs) or cancer potency factors. For both incinerators the PCDD/Fs, PCBs and polyaromatic hydrocarbons (PAHs) are the main contributors to total weighed POP output. In MSW, the PCDD/Fs, PBDD/Fs and polybrominated diphenylethers (PBDEs) are the main contributors to the weighed POP input. The ratios of the weighed POP-input over -output clearly indicate that the rotary kiln incinerating hazardous waste is a weighed POP sink. The grate furnace incinerating MSW is a weighed POP sink or source depending on the POP concentrations in the waste, but the difference between output and input is rather limited. When e.g. ASR and plastics of WEEE, containing high concentrations of PBDEs and PCBs, are co-incinerated in the grate furnace, it is clearly a weighed POP sink.

32

33 **Keywords**

34 Incineration; PCDD/F; PCB; Mass balance; Waste incinerator

35

36 **1. Introduction**

37 Waste incinerators are usually considered as sources of polychlorinated dibenzo-p-
38 dioxins and –furans (PCDD/Fs) as well as polychlorinated biphenyls (PCBs). Indeed,
39 these persistent organic pollutants (POPs) are unintentionally formed in the
40 incinerator and found in the output (Sakai et al., 1999; Abad et al., 2000; Sakai et al.,
41 2001; Abad et al., 2002). The waste to be incinerated, however, also contains a range
42 of POPs (Wilken et al., 1992; Mark et al., 1998; Sakai et al., 1998; Sakai et al., 1999;
43 Aae Redin et al., 2001; Sakai et al., 2001; Vehlow et al., 2002; Tange and

44 Drohmann, 2005; Ishikawa et al., 2007; Morf et al., 2007; Schlummer et al., 2007). It
45 is generally assumed that the incoming POPs are destroyed during incineration and
46 that during the cooling of the flue gases new PCDD/Fs and PCBs are formed through
47 precursors or through de novo synthesis (Everaert and Baeyens, 2002; McKay,
48 2002). This assumption is supported by the analysis of the isomer and congener
49 distributions or so called “chemical fingerprints” of PCDD/Fs and PCBs; chemical
50 fingerprints of PCDD/Fs and PCBs in waste are characterized by a significantly
51 different isomer and congener distribution than PCDD/F- and PCB-fingerprints in
52 flue gases and ashes of waste incinerators (Sakai et al., 1999; Abad et al., 2000;
53 Sakai et al., 2001; Abad et al., 2002).

54 Waste may contain a variety of toxic substances. Besides the POPs considered by the
55 Stockholm convention (PCDD/Fs, PCBs and polychlorinated pesticides including
56 hexachlorobenzene (HCB)), this paper also considers polybrominated dibenzo-p-
57 dioxins and –furans (PBDD/Fs), polybrominated diphenyl ethers (PBDEs),
58 polyaromatic hydrocarbons (PAHs) and di(2-ethylhexyl)phthalate (DEHP) in
59 different types of waste. All the above-mentioned toxic substances will for the sake
60 of brevity further be referred to as POPs.

61 Some studies compared the amount of PCDD/Fs in the in- and output of municipal
62 solid waste (MSW) incinerators (Abad et al., 2000; Abad et al., 2002; Giugliano et
63 al, 2002; Grosso et al. 2007) and concluded that MSW incinerators can act as
64 PCDD/F sinks, depending on the assumed PCDD/F concentrations in the waste, on
65 the operating conditions and on the flue gas cleaning techniques.

66 This study considers, in addition to PCDD/F, the amount of dioxin-like PCBs, PCBs,
67 PAHs and HCB in the outputs (flue gas, solid residues) of three different
68 installations:

- 69 - a BAT compliant rotary kiln incinerating hazardous waste
- 70 - a BAT compliant grate furnace incinerating MSW
- 71 - the same grate furnace co-incinerating automotive shredder residue (ASR)
72 and plastics of waste of electrical and electronic equipment (WEEE) with
73 MSW.

74 The purpose of the present paper is to answer the question whether the considered
75 waste incinerators are sinks or sources for the above mentioned groups of POPs.

76 In an attempt to estimate and compare the overall input and output of POPs – not just
77 the different POP-groups separately- a POP-weighting methodology is proposed and
78 applied to the three installations.

79 **2. Materials and methods**

80 **2.1 Installations**

81 The POPs in the in- and output are first compared for a rotary kiln, incinerating ca.
82 90.000 tons/year of hazardous industrial waste at temperatures between 1000 and
83 1200 °C. The flue gases are de-dusted with an electrofilter and then washed in a wet
84 gas washing unit. A fixed bed brown coal filter is used to remove PCDD/F. The
85 bottom ashes and the filtercake resulting from the wet gas washing are landfilled on a
86 class 1 landfill for hazardous inorganic waste. The boiler- and fly ashes first undergo
87 stabilization/solidification before landfilling.

88 The POPs in the in- and output are also compared for a grate furnace (Vandecasteele
89 et al., 2007), incinerating ca. 400,000 tons/year of MSW and comparable industrial
90 waste at temperatures of at least 850 °C. The flue gases resulting from the
91 incineration are led through a steam boiler for heat recovery and are then cleaned in a
92 half wet gas washing unit. After injection of activated carbon to adsorb PCDD/F and
93 heavy metals, the gases flow through a baghouse filter and - before release to the
94 atmosphere - are further cleaned in a wet gas washing system consisting of a
95 quenching unit, an acid washing unit and an alkaline washing unit. The bottom ashes
96 are treated in a special wet treatment installation (Vandecasteele et al., 2007). After
97 addition of water, they are sieved and all parts larger than 50 mm are, after separation
98 of ferrous and non-ferrous metals, sent back to the incinerator. A screening and
99 washing installation further separates the particles smaller than 50 mm in 2 granulate
100 fractions (2-6 mm and 6-50 mm) and a sand fraction (< 2 mm). Ferrous and non-
101 ferrous metals are separated from the granulate fractions, which can e.g. be used in
102 construction applications. The sand fraction is applied as construction material on
103 landfills. The boiler ashes and the flue gas cleaning residues are treated by
104 stabilization/solidification before they are landfilled on a class 1 landfill for
105 hazardous inorganic waste. All ferrous and non-ferrous fractions are recycled.

106 **2.2 Sampling and analysis**

107 **2.2.1 POP concentrations in the input waste**

108 In the case of the rotary kiln, the considered POP input consists of 1,235 tons/year of
109 commercial 100% PCB mixtures and 100 tons/year of 100% chlorinated POP

110 pesticides. Their exact composition is not known. The POPs in the other hazardous
111 waste fractions are not considered; their contribution is estimated negligible
112 compared to the contribution of the PCBs and the chlorinated POP pesticides.
113 In MSW incineration the assessment of PCDD/Fs mass balances is complicated
114 because of lack of data on PCDD/Fs concentrations in the waste (Hedman et al.
115 2007). This conclusion holds for all the POPs considered in this paper. An extensive
116 literature search revealed substantial differences in the reported POP concentrations.
117 Therefore it was decided to work with concentration ranges as detailed in the
118 comment column of table 1. For PAHs, dioxin-like PCBs and HCB no representative
119 concentration data were found. All the bromine in the plastics of WEEE is supposed
120 to come from the PBDEs.

121 **2.2.2 POP concentrations in the output**

122 The POP-concentrations in the outputs (flue gas, solid residues, waste water
123 treatment sludge) were determined on samples taken on site of the hazardous and
124 municipal solid waste incinerator plant.
125 The flue gas was monitored with equipment located on a measuring platform at the
126 stack of the respective installations. For PCDD/Fs, PCBs and PAHs, the flue gases
127 were sampled using sampling equipment of the AMESA (Adsorption Method for
128 Sampling) type, according to the EN 1948-1 standard. The wet flue gas was extracted
129 isokinetically with a watercooled titanium probe, passed through an XAD II resin
130 and through a condenser. As earlier research showed that the condensate contains a
131 negligible fraction of the PCDD/Fs, it is not further analysed. The components of

132 interest adsorbed on XAD II resin, spiked with C-13 labelled PCDD/Fs, PCBs and
133 PAHs of all relevant components, were subsequently determined by an accredited
134 laboratory by GC-MS according to the EN 1948-2 standard. Clean-up for PCDD/Fs
135 was performed using a multi-layer column of modified silica gel, followed by an
136 aluminium oxide and activated carbon column. PCBs were cleaned-up over silica gel
137 and PAHs were cleaned-up online by means of a multi-layer alumina and silica gel
138 column. The detection limits were $0.001 \text{ ng (Nm}^3)^{-1}$ for PCDD/F and dioxin-like
139 PCBs and $0.001 \text{ } \mu\text{g (Nm}^3)^{-1}$ for PCBs and PAHs. Part of the extract, obtained during
140 cleaning up, was separated for the determination of HCB by GC-MS according to the
141 CMA/3/I method (Emis, 2008a). The detection limit was $0.001 \text{ } \mu\text{g (Nm}^3)^{-1}$. The
142 accredited laboratory that performed the analyses had to participate regularly in
143 government-organised inter-comparisons in order to demonstrate quality.
144 In the solid residues, PCDD/Fs and dioxin-like PCBs were determined by GC-MS
145 after soxhlet extraction according to the German AbfKlärV, Annex 1
146 (Bundesministerium, 2006). The PCBs and HCB were analysed by GC-MS after
147 extraction as described in the CMA/3/I method (Emis, 2008a). The PAHs were
148 analysed by HPLC-UV fluorescence after soxhlet extraction according to the
149 CMA/3/B method (Emis, 2008b). The detection limits were $1 \text{ pg g}_{\text{dw}}^{-1}$ for PCDD/F, 3
150 $\text{pg g}_{\text{dw}}^{-1}$ for dioxin-like PCBs, $1 \text{ ng g}_{\text{dw}}^{-1}$ for PCBs and HCB and $10 \text{ ng g}_{\text{dw}}^{-1}$ for
151 PAHs.
152 The ashes were sampled in August 2007 and August 2008 (for PCDD/Fs, PCBs and
153 PAH analysis) and in February 2008 (for HCB analysis); the flue gas was sampled

154 from the 26th of March until the 9th of April 2008. The concentrations of the POPs in
 155 the different output fractions are given in Table 2. For the bottom-, boiler- and filter
 156 ashes and for the sludge, the POP concentrations in the two samples (where
 157 applicable) are given, the lowest value first.

158 **2.3 Calculation of the amount of POPs in MSW, ASR and incinerator output**

159 The mass of a POP “i” entering the grate furnace ($POP_i \text{ mass}_{in}$) is calculated by
 160 multiplying the concentration of that POP given in table 1 ($POP_i \text{ concentration}_{waste,j}$)
 161 with the mass of the incinerated waste stream “j” (MSW, ASR or WEEE)
 162 ($mass_{waste,j}$). The masses of the POP in each of the three waste types are added (see
 163 equation 1).

$$164 \quad POP_i \text{ mass}_{in} = \sum_j mass_{waste,j} \times POP_i \text{ concentration}_{waste,j} \quad (1)$$

165 The mass of PCDD/Fs, dioxin-like PCBs, PCBs, and PAHs in the output of the
 166 rotary kiln and the grate furnace ($POP_i \text{ mass}_{out}$) is calculated by multiplying the
 167 concentration of the respective POP in the output fraction “j” (POP
 168 $concentration_{output,j}$) with the mass of that fraction ($mass_{output,j}$) given in table 2. The
 169 total is obtained by adding the amount of the POP in each of the output fractions (see
 170 equation 2).

$$171 \quad POP_i \text{ mass}_{out} = \sum_j mass_{output,j} \times POP_i \text{ concentration}_{output,j} \quad (2)$$

172 Concentrations below the detection limit are assumed zero in the calculations (see
 173 also paragraph 3.4).

174 **2.4 POP-weighting methodology**

175 In order to compare the total amount of all POPs (not just the individual POP groups
 176 separately) in the in- and output of a waste incinerator, a “weighed POP mass” (POP_i
 177 $mass_w$) is obtained for each POP in the input and output, respectively, by multiplying
 178 the mass of each POP “i” with a “weighing factor” (see equation 3 below):

$$179 \quad POP_i \text{ mass}_w = POP_i \text{ weighing factor} \times \sum_j mass_j \times POP_i \text{ concentration}_j \quad (3)$$

180 where “ $mass_j$ ” equals the mass of different waste types (input) or the mass of the
 181 different output fractions (flue gas, bottom ash, boiler ash, filter ash or sludge,
 182 respectively). A total weighed POP mass (Total POP $mass_w$) for the in- and output is
 183 obtained by adding the weighed POP masses for the different POPs:

$$184 \quad \text{Total POP mass}_w = \sum_i POP_i \text{ mass}_w \quad (4)$$

185 The ratio of the total weighed POP mass (equation 5) for the input (Total POP
 186 $mass_{w,input}$) over the total weighed POP mass for the output (Total POP $mass_{w,output}$)
 187 indicates whether a waste incinerator acts as a POP sink or source.

$$188 \quad \text{Ratio} = \frac{\text{Total POP mass}_{w,input}}{\text{Total POP mass}_{w,output}} \quad (5)$$

189 Even though most of the POPs belong to the group of halogenated organic
 190 compounds, they do not necessarily provoke the same health effects, what makes it
 191 difficult to compare their toxicity. Furthermore, it is difficult to quantify the severity
 192 of the different health effects. (Is, for instance, the suppression of the immune
 193 system, making the exposed person more vulnerable to infections, a more or less
 194 severe effect than the development of a learning disability?).

195 The idea of weighing the amount of toxic substances using a toxicity-related
196 parameter was already proposed by Horvath et al. (1995). They described a
197 methodology in which $1/\text{TLV}^1$ is used as a factor to weigh toxic chemicals before
198 adding their masses. The TLV was preferred to other toxicity indices, because it is
199 available for a very broad group of chemicals. Also Davis et al. (1994) presented a
200 model, in which toxic chemical releases are weighed by a factor based on lethal
201 doses for rodents and fish. Hereby only the acute toxicity of the chemicals is taken
202 into account. Forman (1993) developed a chemical index, intended for the ranking of
203 chemical releases, in order to identify facilities estimated to contribute the largest
204 amount of toxic releases to the environment. This chemical index (CI) combines
205 minimal risk doses (MRDs)² with carcinogenic doses calculated from EPA
206 “regulatory risk and hazard levels” and from potency factors³.
207 POPs resist well to biological and chemical degradation and are thus stable over time
208 in normal environmental conditions, so chronic rather than acute exposure is the
209 main issue in the situation considered here. Lethal doses, which are a measure for
210 acute toxicity, are not considered advisable as weighing factor. Mato et al. (2007)
211 showed that in Japan, diet accounted for more than 90% of the intake of PCDD/Fs
212 and dioxin-like PCBs. The fact that food is the primary source of human exposure for
213 most of the POPs is confirmed by toxicological information in the Environmental

¹ TLV stands for Threshold Limit Value and is the time weighted average airborne concentration ($\text{mg (m}^3)^{-1}$) for an 8-hour workday and a 40-hour workweek, to which it is believed that nearly all workers may repeatedly be exposed, day after day, for a working lifetime, without adverse effect

² MRD stands for Minimal Risk Dose and is the estimate of the amount of a substance, expressed in $\text{mg (kg}_{\text{body weight}}\cdot\text{day)}^{-1}$, which can be ingested daily over a lifetime by humans without adverse non cancer health effects

³ For oral exposure to low doses, there seems to be a linear relationship between cancer risk and dose. The potency factor is the slope of the dose response curve at low doses, expressed in $(\text{kg}_{\text{body weight}}\cdot\text{day}) \text{ mg}^{-1}$.

214 Health Criteria of the World Health Organization and the Toxicological profiles of
215 the American Agency for Toxic Substances and Disease Registry (ATSDR 1995a;
216 ATSDR 1995b; ATSDR 1998; World Health Organisation, 1998a; World Health
217 Organisation, 1998b; World Health Organisation, 1998c; ATSDR 2000; ATSDR
218 2002a; ATSDR 2002b; ATSDR 2002c; ATSDR 2003; ATSDR 2004; ATSDR 2005).
219 For all these reasons (chronic rather than acute exposure, major uptake by food rather
220 than from air), in the methodology described and applied in this paper, the “minimal
221 risk dose (MRD)” for non-carcinogenic effects of chronic oral exposure to a given
222 POP is preferred to lethal doses and TLVs for weighing. As shown in equation 6, the
223 weighing factor (see equation 3) is set equal to 1/MRD:

$$224 \quad \text{POP}_i \text{ weighing factor} = \frac{1}{\text{MRD}_i} \quad (6)$$

225 The methodology must be considered as a pragmatic attempt to answer the question
226 whether a waste incinerator should be considered as an overall sink or source of
227 POPs. It takes into account any adverse effect of the POPs, irrespective of its severity
228 and not one particular effect. Table 3 gives an overview of the MRDs (mg/kg body
229 weight.day) selected from literature and applied in this study.

230 PBDE-concentrations are often only reported as a total, without specification of the
231 considered congener(s). For this group of POPs, a maximum and minimum weighed
232 input and output is calculated applying the MRD of octaBDE (lowest MRD) and
233 decaBDE (highest MRD). The same approach is used for PAHs, where the MRD of
234 the most and least toxic of the 16 EPA PAHs are applied. The composition of the
235 chlorinated pesticides incinerated in the rotary kiln selected to illustrate the

236 methodology (see paragraph 3.1) is not exactly known, so a maximum and minimum
 237 is calculated using the MRDs of aldrin (lowest MRD of all 9 Stockholm pesticides)
 238 and mirex (highest MRD of all 9 Stockholm pesticides). The comment column in
 239 table 3 points out if additional assumptions were made for the application of the
 240 mentioned MRDs. Hexachlorobenzene (HCB), which is one of the nine POP
 241 pesticides considered by the Stockholm convention, can also be produced during
 242 combustion processes and found in the output of waste incinerators (Wienecke et al.,
 243 1995). Since the MRD of HCB is of the same order of magnitude as the one of PCBs,
 244 its presence can influence the total weighed POP-output significantly.

245 Information from the World Health Organization (WHO-Environmental Health
 246 Criteria) and the American Agency for Toxic Substances and Disease Registry
 247 (ATSDR-Toxicological profiles) indicates that several of the POPs within the scope
 248 of this study are carcinogenic, are possibly carcinogenic or are not classifiable as to
 249 their carcinogenicity. Therefore the methodology also attempts to weigh the
 250 considered POPs by using the “potency factor” for oral exposure as toxicity factor in
 251 equation (3).

$$\text{POP}_i \text{ weighing factor} = \text{Potency factor}_i \quad (7)$$

253 Table 4 gives the limited number of potency factors available for the relevant
 254 components. The dioxin-like PCB- and PBDD/F-masses expressed in TEQ are
 255 weighed using the potency factor of the PCDD/Fs (in TEQ). This approach is
 256 proposed by the ATSDR (ATSDR, 1998) and was also used by van Larebeke et al.
 257 (2001).

258 Some PAHs are classified by the International Agency for Research on Cancer as
259 possibly or probably carcinogenic. Therefore, the PAHs in the output are considered
260 in the calculations below. They are weighed using the highest and lowest reported
261 potency factor for benzo(a)pyrene, one of the most carcinogenic PAHs
262 (WHO,1998a). The PAHs in the input are not taken into account, as no data are
263 available on their concentration in the considered types of waste (see also paragraph
264 3.4). For PBDEs no potency factors were found in literature, these pollutants are not
265 considered when POPs are weighed with this factor.

266 **3. Results and discussion**

267 The POPs in the in- and output were compared in the following 3 scenarios:

- 268 • Scenario 1: Incineration of hazardous waste in a BAT compliant rotary kiln
- 269 • Scenario 2: Incineration of MSW in a BAT compliant grate furnace
- 270 • Scenario 3: Co-incineration of 3% plastics of WEEE and 5% ASR with MSW in
271 a BAT compliant grate furnace

272 **3.1 Scenario 1: Incineration of hazardous waste in a rotary kiln**

273 The POPs in the input of the considered rotary kiln are given in paragraph 2.2.1.

274 Table 2 gives the yearly mass flows of the different output fractions, the POP

275 concentration-ranges measured and the POP masses in the different output fractions.

276 The total amount of PCBs in the output fractions of the rotary kiln (0.17 to 94 kg) is

277 $7.4 \cdot 10^6$ to $1.3 \cdot 10^4$ times lower than the yearly PCB input ($1.235 \cdot 10^6$ kg). For the other

278 POPs it is not possible to compare the amounts in the in- and output of the rotary kiln

279 as no detailed information is available on the concentrations in the input as explained
280 in paragraph 2.2.1.

281 A maximum and minimum total weighed in- and output are calculated according to
282 equation 3 using the data given in tables 2, 3 and 4. The PCDD/Fs account for ca. 20
283 to 90% of the total MRD-weighted output, the dioxin-like PCBs for ca. 10 to 55%,
284 PCBs for ca. 1 to 25% and HCB for < 0.1 to ca. 3 %. These percentages take into
285 account the POP concentration ranges in the output fractions and the range on the
286 MRDs used for weighing. The contribution of PAHs is negligible (< 0.1%). If the
287 POPs in the output are weighed using the cancer potency factors, the PCDD/Fs
288 contribute for ca. 20 to 85% to the total weighed output, the dioxin-like PCBs for ca.
289 10 to 60% and the PAHs for ca. 5 to 15%.

290 Only ca. 0.6% of the total amount of weighed POPs in the output of the hazardous
291 waste incinerator is directly emitted in the environment via the flue gas. As
292 mentioned earlier, the POPs in the solid residues are landfilled on specially designed
293 landfills to minimize the environmental impact. If in the output only the POPs in the
294 flue gases are taken into account, the POP input over output ratio equals (7.2 to 7.6)
295 10^6 for MRD-weighting and ca. (1.2 to 4.7) 10^5 for weighing with cancer potency
296 factors (see table 5). If the POPs in all the output fractions are taken in to account,
297 the MRD-weighted POP-input exceeds 3,200 to 49,800 times the output; if the POPs
298 are weighed with potency factors, the input exceeds the output by 890 to 16,900
299 times. The hazardous waste incinerator clearly is a weighed POP sink.

300 **3.2 Scenario 2: MSW incineration in a grate furnace**

301 The considered input for the BAT grate furnace selected to illustrate scenario 2 and 3
302 consists of 408,430 tons/year of MSW. A minimum and maximum POP input mass
303 was calculated according to equation 1, using the concentration ranges given in table
304 1. For PCDD/Fs, the estimated yearly PCDD/F input ranges from $(0.32 \text{ to } 2.1) \cdot 10^{-2}$
305 kg TEQ; for PCBs it ranges from 3.4 to 13.5 kg. The minimum and maximum
306 amounts of PCDD/Fs, dioxin-like PCBs, PCBs, PAHs and HCBs in the output were
307 calculated according to equation 2 and using the concentration ranges and mass flow
308 data given in table 2. Only ca. 0.4% of the total amount of PCDD/Fs in the output of
309 the BAT compliant grate furnace come directly in the environment via the flue gas or
310 via the bottom ash granulates used as secondary material. As mentioned earlier, the
311 POPs in the other residues are landfilled in landfills specially designed to minimize
312 environmental impact. The amount of PCDD/F in the flue gas and granulates (5.02
313 10^{-5} kg TEQ) is ca. 60 to 400 times lower than the estimated amount of PCDD/Fs in
314 incinerated waste. When the total PCDD/F output $((1.35 \text{ to } 1.83) \cdot 10^{-2}$ kg TEQ) is
315 compared to the amount of PCDD/Fs in the input, it seems that the grate furnace
316 incinerating MSW can be a sink or source of these POPs, depending on the
317 concentrations in the input waste. For the PCBs, the comparison shows that the
318 amount of these persistent pollutants are ca. 10.000 times lower in the output if the
319 highest concentrations in the waste and the lowest concentrations in the output
320 fractions are considered. (The flue gas and granulates are not considered separately
321 as the latter contain more than 99% of the total PCB output.) On the other hand, the
322 output could be ca. 2 times higher than the input if the lowest concentrations in the

323 waste and the highest concentrations in the output fractions are considered. For
324 dioxin-like PCBs, PAH and HCB, no representative input concentration data were
325 found and hence no comparison between in- and output can be made.

326 As for the rotary kiln in scenario 1, the minimum and maximum total weighed in-
327 and output for the grate furnace described in paragraph 2.1 are calculated according
328 to equation 3 using the data given in tables 1, 2, 3 and 4. The main contributors to the
329 total MRD-weighted input are the PCDD/Fs (ca. 50%) and the PBDEs (ca. 25 to
330 50%). The contribution of the PBDD/Fs reaches ca. 25% if the highest
331 concentrations for these POPs given in table 1 are used in the calculations. In case of
332 weighing with cancer potency factors, PAH become the main contributors (ca. 30 to
333 90%) but PCDD/Fs and PBDD/Fs remain important (ca. 10 to 45% and <0.1% to ca.
334 25%, respectively). MSW contains ca. 0.35% plastics of WEEE (Tange and
335 Drohmann, 2005). For the weighing of total PBDD/Fs in this waste fraction, no
336 specific weighing factors were found in literature. In order to apply the MRD for
337 PCDD/Fs as suggested by the World Health Organisation (WHO, 1998b), TEQ-
338 concentrations were estimated. The lowest values (see table 1), which express total
339 PBDD and PBDF concentrations, were reported by Vehlow et al. (2002) without
340 indication of the individual congeners concentration, making it impossible to convert
341 them to TEQ using TEF-values. Sakai et al. (2001) and Abad et al. (2002) reported
342 PCDD/F concentrations in waste in both mass and TEQ, the former being on average
343 200 times higher than the latter. The minimum PBDD/F mass concentrations given in
344 table 1 were estimated in TEQ by dividing them by this factor. Hereby it is assumed

345 that the congener distribution of the PBDD/F is similar to the one of PCDD/F. The
346 PBDD/F concentration used to calculate the maximum weighed output (see table 1)
347 was reported as the total mass of 8 congeners and isomers, without indication of the
348 individual concentrations (Schlummer et al., 2007). It was multiplied with the
349 average TEF value of the 8 chlorinated analogues to give a PBDD/F concentration in
350 TEQ. The estimation of PBDD/F-concentrations in TEQ introduces an additional
351 uncertainty but it was considered important to include these pollutants, which are
352 considered to have the same MRD and potency factor than PCDD/Fs (WHO, 1998b).
353 Only PCDD/Fs and PCBs contribute significantly to the total MRD-weighted output
354 (ca. 80 to 98% and 2 to 20% respectively). If potency factors are used for weighing,
355 the PCDD/Fs, dioxin-like PCBs and PAHs account for ca. 65 to 85%, ca. 2 to 20%
356 and ca. 10 tot 20% of the total weighed output, respectively. The contribution of
357 HCB is negligible ($< 0.1\%$).
358 If in the output only the POPs in the flue gases and the granulates are taken into
359 account, the input over output ratios in table 5 indicate that the incinerator clearly
360 acts as a weighed POP sink, both in case of weighing with MRDs and potency
361 factors. If the POPs in all the output fractions are considered, the weighed POP-input
362 is comparable to the weighed POP-output (see table 5). Abad et al. (2000, 2002),
363 Giugliano et al. (2002) and Grosso et al. (2007) came to a similar conclusion for
364 PCDD/Fs in MSW incinerators. This can be explained by the fact that PCDD/Fs are
365 one of the main contributors to the total weighed in- and output, as indicated earlier.

366 **3.3. Co-incineration of 3% plastics from WEEE and 5% ASR with MSW in a**
367 **grate furnace**

368 Plastics have become an increasingly important part of E&E equipment: the average
369 proportion has increased from 12% in 1980 to more than 20% today (Vehlow et al.,
370 2002; Schlummer et al., 2007). Selective treatment and recycling of WEEE is
371 foreseen in the European Directive 2002/96/EC. One of the components that have to
372 be removed before recycling according to annex II of this directive are plastics
373 containing brominated flame retardants. Increasing amounts of high calorific waste
374 plastics containing high levels of brominated flame retardants become thus available
375 (Schlummer et al., 2007). By co-combusting these plastics in a BAT compliant MSW
376 incinerator, the hazardous POPs they contain are destroyed and energy from these
377 high calorific wastes is recovered. Table 1 gives the concentration ranges of POPs in
378 plastics of WEEE used to calculate the maximum and minimum POP input flow for
379 scenario 3.

380 Another industrial waste stream that can be co-incinerated in grate furnaces is
381 Automotive Shredder Residue (ASR) (Mark et al., 1998; Aae Redin et al., 2001).
382 After dismantling, end-of-life vehicles are shredded and metals, representing 75 to
383 80% of the mixed waste, are recovered. The remaining 20-25% is referred to as ASR
384 and is a heterogeneous mixture of both inert and combustible materials, including
385 plastics, glass, fabric, wood, rubber, fibres and paper. About 50% of the ASR is
386 combustible. Table 1 gives the concentration ranges of POPs in ASR used to
387 calculate the maximum and minimum POP input flow for scenario 3.

388 In scenario 3 it is assumed that 3% of the 408,430 tons of MSW yearly incinerated in
389 the grate furnace (scenario 2) would consist of plastics of WEEE, and 5% of ASR.
390 These percentages are chosen somewhat arbitrarily, but are below the reported
391 maximum workable percentages (Vehlow et al., 1997; Mark et al., 1998; Aae Redin
392 et al., 2001; Vehlow et al., 2002). For PCDD/Fs, the estimated yearly input ranges
393 from (0.83 to 2.6) 10^{-2} kg TEQ; for PCBs it ranges from 27 to 1,300 kg.
394 The considered POP concentrations in the different output fractions of the grate
395 furnace are the same as in scenario 2 (see table 2) as it is assumed that the incoming
396 POPs are completely destroyed and the formation of POPs in the outputs only
397 depends on process-conditions. This assumption is supported by a recent study by
398 Van Caneghem et al. (2009) where POP-concentrations in the different in- and
399 output fractions of a fluidised bed combustor were compared during normal
400 operation (input consisting of 30% waste water treatment sludge and 70% RDF) and
401 during co-incineration of automotive shredder residue (input consisting of 25% ASR,
402 25% RDF, 50% waste water treatment sludge). Because of the high concentration of
403 POPs in the ASR, the total weighed POP input was on average 16 times higher when
404 this waste was co-incinerated. Nevertheless it was observed that the weighed POP
405 output did not change significantly. So, although the assumption is supported by
406 indirect evidence, the confidence of the results for this scenario is somewhat lower
407 than for scenario 1 and 2. When the yearly amount of PCDD/F in the output given in
408 table 2 is compared to the amount of PCDD/Fs in the input, it seems that the grate
409 furnace incinerating MSW, WEEE and ASR can be a sink or source of these POPs,

410 depending on the assumptions made on the concentrations in the wastes. The mass of
411 PCBs in the input, mainly coming from the plastics of WEEE and ASR, is estimated
412 ca. 20 to 100 times higher than in the output.

413 The main contributors to the MRD-weighted input are the PBDEs from the plastics of
414 WEEE (ca. 40 to 70%), PCDD/Fs (ca. 10 to 20%), PCBs (ca. 5 to 20%) and
415 PBDD/Fs (< 0.1% to ca. 40%). If potency factors are used for weighing, the main
416 contributors are PAHs (ca. 10 to 60%), PBDD/Fs (< 0.1% to ca. 60%) and DEHP
417 (ca. 10 to 30%). PCDD/Fs (ca. 14%) and PCBs (ca. 1 to 10%) are less important.

418 The input over output ratios given in table 5 indicate that when WEEE-plastics (3%)
419 and ASR (5%) are co-combusted, the MSW incinerator acts as weighed POP sink.

420 **3.4. Influence of output concentrations below detection limit**

421 In the calculation of the ratios discussed in paragraph 3, in the output fractions all
422 POP-concentrations below the detection limit were assumed zero. Because of this
423 assumption, the total (weighed) POP output may be underestimated. To investigate
424 the impact of this assumption, the input over output ratios were recalculated with all
425 the output concentrations below the detection limit taken equal to the detection limit.
426 If e.g. the detection limit for the individual PAHs was 10 ng/g_{dw} and the
427 concentration of all of the 16 analysed PAH was below this limit, total PAH-
428 concentration was taken 160 ng/ g_{dw}. This had no significant influence (< 5%
429 decrease) on the minimum input over output ratios of the rotary kiln and the
430 conclusions drawn in paragraph 3.1 remain valid. For the grate furnace, increasing
431 the non-detectable PCDD/F, dioxin-like PCB, PCB and HCB concentrations also

432 was of limited influence (< 15% decrease in minimum input over output ratio).
433 However, if in the output only the POPs in the flue gas and granulate fraction were
434 taken into account, setting the non detectable PAH-concentrations equal to the
435 detection limit decreased the minimum input over output ratios by ca. a factor 2. This
436 decrease of the input over output ratio does however not alter the conclusions drawn
437 in paragraphs 3.2 and 3.3. Furthermore, the PAHs in the input were taken equal to
438 zero in the calculations, because although several data were found, they could not be
439 considered sufficiently representative. Ruokojärvi et al. (1995) reported a PAH-
440 concentration of $2.0 \mu\text{g g}_{\text{dw}}^{-1}$ on household waste in a Finish landfill. The waste was,
441 however, sampled close to a controlled fire, which could have influenced the PAH
442 concentration although the waste itself was not burned. Moeller and Reeh (2003)
443 reported a PAH concentration of $1.2 \mu\text{g g}_{\text{dw}}^{-1}$ for composted Danish MSW. Brändli et
444 al. (2007) reported PAH-concentration of $3.32 \mu\text{g g}_{\text{dw}}^{-1}$, $0.41 \mu\text{g g}_{\text{dw}}^{-1}$ and $1.45 \mu\text{g g}_{\text{dw}}^{-1}$
445 ¹ for Swiss green waste, a mixture of green and organic kitchen waste and digested
446 green- and kitchen waste, respectively (ca 40% of the MSW incinerated in the grate
447 furnace used to illustrate the methodology in paragraph 3.2 and 3.2 consists of
448 organic kitchen waste and green waste). Li et al. reported PAH output over input
449 ratios ranging from 0.00018 to 0.00032 for the incineration of waste plastics (Li et
450 al., 2001). If in scenario 2 (see paragraph 3.2), the PAH-concentration of MSW is
451 taken equal to $2.0 \mu\text{g g}_{\text{dw}}^{-1}$ (Ruokojärvi et al., 1995), this has a negligible influence
452 on the MRD-weighted POP-input, but a significant influence on the POP-input
453 weighed with cancer potency factors: the minimum input over output ratio which

454 ranged from 0.1 to 6 (see table 5) increases to a range of 1.4 to 70. This indicated that
455 accurate PAH concentrations in the input would be required.

456 **4. Conclusion**

457 The amount of different POPs was compared in the in- and outputs of a BAT
458 compliant rotary kiln incinerating hazardous waste, a BAT compliant grate furnace
459 incinerating MSW and the same grate furnace co-incinerating WEEE and ASR with
460 MSW. In order to compare the total amount of POPs, a methodology was developed
461 to weigh the POPs, in the in- and output of waste incinerators based on MRDs or
462 cancer potency factors.

463 The mass of PCBs incinerated in the rotary kiln exceeded $7.4 \cdot 10^6$ to $1.3 \cdot 10^4$ times the
464 mass of PCBs found in the output: the incinerator was clearly a sink of PCBs. For the
465 other POPs it was not possible to compare the amounts in the in- and output, as no
466 detailed information was available on the concentrations in the input. PCDD/Fs and
467 dioxin-like PCBs were the main contributors to the MRD-weighted output. If potency
468 factors were used for weighing, also PAH became important contributors. The total
469 amount of weighed POPs in the input waste exceeded, depending on input and
470 process-conditions and applied weighing factor, $1.2 \cdot 10^5$ to $7.6 \cdot 10^6$ times the amount
471 of weighed POPs emitted in the environment via the flue gas. If the POPs in all
472 output fractions were taken into account, the weighed POP input over output ratio
473 ranged from 890 to 49,800. The studied rotary kiln incinerating hazardous waste was
474 clearly a sink of POPs.

475 For the grate furnace incinerating MSW, the mass of PCDD/Fs in the flue gas and
476 granulates was ca. 60 to 400 times lower than the PCDD/Fs mass in the incinerated
477 waste, depending on the considered concentrations. When the total amount of
478 PCDD/F in the output was compared to the amount of PCDD/Fs in the input waste, it
479 seemed that the grate furnace incinerating MSW could be a sink or source of these
480 POPs, but the difference between output and input is rather limited. Also for PCBs
481 the grate furnace incinerating MSW could be a sink or source depending on the
482 considered concentrations in the input waste and in the output. PCDD/Fs, PBDD/Fs
483 and PBDEs were the most important contributors to the MRD-weighted input. In case
484 of weighing with cancer potency factors, PAH become the main contributors. The
485 weighed POP input over output ratio ranged from 6 to 580 depending on the
486 considered concentrations and weighing factors. The total amount of weighed POPs
487 in the output (all fractions) was about the same as in the input. The conclusion was
488 the same as for the (un-weighted) PCDD/Fs and PCBs.

489 When 5 % of ASR and 3 % of plastics of WEEE would be co-incinerated in the BAT
490 compliant grate furnace incinerating MSW, the mass of PCBs in the input, mainly
491 from the WEEE and ASR, would be estimated ca. 20 to 100 times higher than the
492 mass of PCBs in the output. The PBDE's from the plastics of WEEE would become
493 the main contributors to the MRD-weighted input. The weighed input over output
494 (flue gases and granulates) ratio would range from 50 to 4,100 depending on the
495 considered concentrations and weighing factors. If all the POPs in the output were
496 taken into account, the input over output ratio would range from 1.0 to 23. Both the

497 comparison of PCBs and total weighed POP masses in the in- and output showed
498 that, when wastes containing relatively high levels of POPs are co-incinerated with
499 MSW, the BAT compliant grate furnaces studied acted as a PCB and weighed POP
500 sink.

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507

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Table 1

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Table 1: POP concentration ranges in waste incinerated in the BAT compliant grate furnace

	Concentration			Comments
	Min.	Max.		
MSW				
PCBs	8.2	33.0	μg kg ⁻¹	Measured PCB concentrations in Japanese waste (Sakai et al., 1999, Sakai et al., 2001, Ishikawa et al., 2007) ^a
PCDD/Fs	7.91	50.2	ng TEQ kg ⁻¹	The minimum value is the average concentration in 18 Spanish waste samples analysed in 2000 (Abad et al., 2002). The maximum value is the average concentration in German samples taken in 1980 (Wilken et al., 1992)
WEEE-plastics in MSW		0.35	%	Tange and Drohmann, 2005
Plastics of Waste of Electrical and Electronic Equipment (WEEE)				
Br content		2.0	%	Average Br-concentration of mixed shredded WEEE-plastics (Schlummer et al., 2007)
PCB	0.09	0.15	mg kg ⁻¹	Morf et al., 2007
PBDD	15	7,700	ng kg ⁻¹	Lowest value reported by Vehlow et al., 2002 Highest value reported by Schlummer et al., 2007
PBDF	600	7,700	ng kg ⁻¹	Lowest value reported by Vehlow et al., 2002 Highest value reported by Schlummer et al., 2007
Automotive Shredder Residues (ASR)				
PCBs	1.1	63	mg kg ⁻¹	Lowest value reported by Aae Redin et al., 2001 Highest value reported by Mark et al., 1998
PCDD/Fs		250	ng TEQ kg ⁻¹	Sakai et al., 1998 (Japanese ASR)
DEHP		11,000	mg kg ⁻¹	Sakai et al., 1998 (Japanese ASR)

Table 2

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Table 2: POP concentrations in the output fractions of the considered waste incinerators

	PCDD/Fs	Dioxin-like PCBs ^a	PCBs ^b	PAH ^c	HCB	Mass flow
	pg TEQ g _{dw} ⁻¹	pg TEQ g _{dw} ⁻¹	ng g _{dw} ⁻¹	ng g _{dw} ⁻¹	ng g _{dw} ⁻¹	ton _{dw} year ⁻¹
Scenario 1 (Rotary kiln)						
Flue gas	10 ^d	n.d.	41 ^e	1,710 ^e	0.48 ^e	713,014 ^f
Bottom ash	12- 71	4.0 – 483	5.5 – 4,200	79 – 1,400	91	22,126
Boiler ash	73 - 161	10.4 - 11.3	n.d. – 7.4	n.d.	20	683
Filter ash	246 - 733	0.12 - 48	n.d. – 4.5	n.d. - 22	12	1,829
Sludge	333 - 371	0.2 - 9.7	8.5 - 830	n.d. - 210	110	3,246
Total output (kg year ⁻¹)	(1.14 - 3.63) 10 ⁻³ g	(1.16 – 108) 10 ⁻⁴ g	0.17 - 94	2.96 – 32.6	2.26	
Scenario 2 and 3 (Grate furnace)						
Flue gas	20 ^d	3 ^d	13 ^e	4,950 ^e	0.034 ^e	2,115,376 ^f
Bottom ash	8.1 – 22	1.3 – 47	n.d. – 6.2	570 – 680	n.d.	94,724
Granulate fraction	0.58	0.56	n.d.	220	n.d.	31,723
Boiler ash	37 - 97	0.007 - 2.9	n.d.	n.d.	n.d.	8,660
Flue gas cleaning residue	1,040 – 1,290	17 – 29	n.d.	n.d. - 24	n.d.	11,939
Total output (kg year ⁻¹)	(1.35 – 1.83) 10 ⁻² g	(3.29 – 48.3) 10 ⁻⁴ g	(0.028– 6.2)	65 – 75	7.19 10 ⁻⁵	

^a sum of PCB77, PCB 81, PCB 105, PCB 114, PCB 118, PCB 123, PCB 126, PCB 156, PCB 157, PCB 167, PCB 169, PCB 189

^b sum of PCB 28, PCB 52, PCB 101, PCB 118, PCB 138, PCB 153, PCB 180

^c sum of 16 EPA

^d expressed in pg TEQ (Nm³)⁻¹

^e expressed in ng (Nm³)⁻¹

^f expressed in kNm³ year⁻¹

^g expressed in kg TEQ year⁻¹

n.d. : concentration below the detection limit

Table 3[Click here to download Table: Table 3 - rev3.doc](#)**Table 3: MRDs used for the calculation of weighed POP masses**

	MRD ^a mg (kg bw.day) ⁻¹	Comments
PCDD/Fs Dioxin-like PCBs PBDD/Fs	1. 10 ⁻⁹ TEQ	TDI ^d for dioxin-like compounds with TEF-values (WHO, 1998a) Sufficient data supports the concept of using corresponding TEF values for brominated and chlorinated analogues (WHO, 1998b)
PCBs	2.10 ⁻⁵	MRL ^c for chronic oral exposure to Aroclor 1254 (ATSDR, 2000)
Chlorinated pesticides	3. 10 ⁻⁵	MRL ^c for chronic oral exposure to aldrin (ATSDR, 2003)
	8. 10 ⁻⁴	MRL ^c for chronic oral exposure to mirex (ATSDR, 1995a)
PAHs	2. 10 ⁻²	RfD ^b for naphthalene (ATSDR, 2005)
	3. 10 ⁻¹	RfD ^b for anthracene (ATSDR, 1995b)
HCB	5. 10 ⁻⁵	MRL ^c for chronic oral exposure to HCB (ATSDR, 2002b)
PBDEs	3. 10 ⁻³	RfD ^b for octabromodiphenylether (ATSDR, 2004)
	1. 10 ⁻²	RfD ^b for decabromodiphenylether (ATSDR, 2004)
DEHP	6. 10 ⁻²	MRL ^c for chronic oral exposure to DEHP (ATSDR, 2002a)

^aMRD stands for Minimal Risk Dose and is the estimate of the amount of a substance, expressed in mg (kg body weight.day)⁻¹, which can be ingested daily over a lifetime by humans without adverse non cancer health effects

^bRFD stands for Reference Dose and is an MRD derived by the US Environmental Protection Agency

^cMRL stands for Minimal Risk Level and is an MRD derived by the American Agency for Toxic Substances and Disease Registry

^dTDI stands for tolerable Daily intake and is an MRD derived by the World Health Organisation

^eADI stands for acceptable Daily intake and is an MRD derived by the World Health Organisation

Table 4[Click here to download Table: Table 4 - rev3.doc](#)**Table 4: Potency factors used for the calculation of weighed POP masses**

	Potency Factor ^a (kg bw.day) mg ⁻¹	Comments
PCDD/Fs Dioxin-like PCBs PBDD/Fs	1.56 10 ⁵ TEQ	Potency factor for exposure to dioxin-like compounds with TEF-values (EPA) (ATSDR, 1998) Sufficient data supports the concept of using corresponding TEF values for brominated and chlorinated analogues (WHO, 1998c)
PCBs	2.0	Potency factor for oral exposure to commercial PCB mixtures (EPA) (ATSDR, 2000)
Chlorinated pesticides	3.4 10 ⁻¹	Potency factor for DDT (EPA), the lowest potency factor of all of the considered chlorinated pesticides for which potency factors were derived (ATSDR, 2002c)
	17	Potency factor for aldrin (EPA), the highest potency factor of all of the considered chlorinated pesticides for which potency factors were derived (ATSDR, 2003)
PAHs	4.5 – 11.7	Lowest and highest reported potency factor for benzo(a)pyrene (WHO, 1998b)
HCB	1.6	Potency factor for HCB (EPA) (ATSDR, 2002c)
DEHP	1.4 10 ⁻²	Potency factor for DEHP (EPA) (ATSDR, 2002a)

^a The potency factor is the slope of the dose response curve at low doses (linear part), expressed in (kg_{body} weight.day) mg⁻¹

Table 5[Click here to download Table: Table 5 - rev2.doc](#)**Table 5: Maximum and minimum weighed input/output ratios**

	Input/output Only flue gas^a	POPs weighed with		Input/output All output fractions	POPs weighed with	
		MRDs	Potency factors		MRDs	Potency factors
Scenario 1	Maximum	7,600,000	470,000	Maximum	49,800	16,900
	Minimum	7,200,000	118,000	Minimum	3,200	890
Scenario 2	Maximum	580	55	Maximum	3.2	2.0
	Minimum	90	6	Minimum	0.3	0.1
Scenario 3	Maximum	4,100	290	Maximum	23	11
	Minimum	600	50	Minimum	2.0	1.0

^a in flue gas for scenario 1, in flue gas and granulate fraction of bottom ashes for scenario 2 and 3