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Relevance of coplanar PCBs for TEQ emission of fluidized bed incineration and impact of emission control devices

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Abstract

Five fluidized bed incinerators combusting municipal solid waste were assessed for the impact of coplanar PCBs on total TEQ emission. In 17 stack measurements, the coplanar PCBs contributed on average less than 3% to total TEQ with a maximum contribution of 7.5% to total TEQ in one measurement. Differences in the design of the flue gas cooling section did not show an effect on the impact of coplanar PCBs on total TEQ.

The effect of emission control devices on the impact of coplanar PCBs on the total TEQ was studied in more detail at one incinerator. The relative contribution of PCBs to total TEQ increased along the flue gas line. This was caused by a slightly higher removal efficiency for TEQ relevant PCDDs/PCDFs compared to coplanar PCBs by the bag filters and a higher destruction efficiency for PCDDs/PCDFs compared to PCBs by the SCR catalyst.

Additionally, the removal efficiencies of the emission control devices (bag filters and catalyst) for other chlorinated aromatic compounds which have been proposed as TEQ indicator compounds (polychlorinated benzenes and polychlorinated phenols) were compared with those for PCDDs/PCDFs and coplanar PCBs. Removal efficiencies for polychlorinated benzenes or polychlorinated phenols considerably differed from those of PCDD/PCDF and coplanar PCBs. Implications for TEQ assessments using indicator compounds as proposed in the literature are discussed.

Keywords: PCDD; PCDF; PCB; Air pollution control; Removal efficiency; TEQ indicator; TEQ surrogate

1. Introduction

Stringent limits for polychlorinated dibenzodioxin and dibenzofuran (PCDD/PCDF) emissions from municipal solid waste incinerators have been imposed in several European countries and in Japan (Ministry of Health and Welfare of Japan, 1997; The European Parliament, 2000). Additionally in Japan, a "Law Concerning Special Measures against Dioxins" was recently endorsed by the Environmental Agency of Japan (1999). This regulation includes non-ortho and mono-ortho substituted polychlorinated biphenyls (coplanar PCBs) for TEQ calculation. The guidelines follow the assignment of toxicity equivalent factors for coplanar PCBs by the WHO (Van den Berg et al., 1998).

To date, all studies reporting on the contribution of PCBs to total TEQ emissions from waste incinerators are restricted to investigations of grate fired incinerators. The PCB emission from grate fired incinerators are reported generally less than 10% of the total TEQ emission (Sakai et al., 1993, 1994; Alcock et al., 1998;

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Johnke et al., 2001). No data is available in the literature, however, concerning the impact of coplanar PCBs on the TEQ-emission from fluidized bed incinerators (FBIs) combusting municipal solid waste. The contribution of PCDDs and PCDFs to the total TEQ value can differ slightly in fluidized bed incinerators compared to grate fired incinerators (Weber and Sakurai, 2001), and the isomer patterns of PCDF and PCN show some differences between the two types of incinerators (Weber and Hagenmaier, 1999; Imagawa and Lee, 2001). Therefore, the aim of this study was to investigate the impact of coplanar PCBs on total TEQ emission from FBIs and to evaluate the relevance of the PCB emission.

Another actual topic with respect to the evaluation of PCDDs/PCDFs and TEQ emission from municipal solid waste incinerators is the determination of indicator compounds suitable to provide a TEQ estimation (Tuppainen et al., 2000; Blumenstock et al., 2001; Gullett et al., 2001; Kato et al., 2001; Urano and Kato, 2001). Gullett et al. (2001) proposed that an understanding of the relationships of PCDDs/PCDFs and indicator compounds must be established from reaction mechanisms, based on a combination of statistical, theoretical and experimental efforts. However, the impact of the emission control devices on PCDD/PCDF removal were not included in these considerations. Therefore, we provide preliminary evaluations of the effect of emission control devices on PCDDs/PCDFs compared to some indicator compounds proposed for TEQ correlation.

2. Materials and methods

2.1. Fluidized bed incinerators

Five FBIs combusting municipal solid waste were analysed for PCDD/PCDF and PCB emission. One of the FBIs was sampled along the flue gas line and evaluated for PCDD/PCDF, PCB, polychlorophenol (PxCP) and polychlorobenzene (PCBz) emission. All plants were full scale incinerators with a capacity of 60–200 t/day. A flow diagram of the FBIs sampled is shown in Fig. 1. The systems used in the cooling section differed between the incinerators (use of boiler, economiser and cooler). All incinerators were equipped with bag filters for effective removal of particles and operated at a bag filter inlet temperature below 200 °C. Some of the incinerators used activated carbon injection for PCDD/PCDF removal. Plant A was additionally equipped with a SCR-catalyst for removal of nitrogen oxides (NO_x) (Fig. 1, Table 1).

2.2. Sampling and analysis

Sampling and analysis were carried out according to the Japanese Industrial Standard JIS K 0311 (Ministry of Economy, Trade and Industry, 1999). The PCBz and PxCP were sampled together with the PCDD/F. Concentrations were normalised to 12% oxygen. The GC/MS analysis were performed on a Micromass Autospec coupled to a HP 6980 gas chromatograph. For the isomer specific analysis of T_4CDD/F to H_6CDF an SP-2331 column (60 m, 0.25 mm i.d., 0.2 μm film thickness, Supelco, Bellefonte/USA) was used. For analysis of H_7CDD/F, OCDD/F, isomer specific analysis of PCBs, PCBz and PxCP a DB-5 fused silica column (30 m, 0.32 mm i.d., 0.25 μm film thickness, J&W Scientific, Folsom/USA) was used.

3. Results and discussion

3.1. Impact of coplanar PCBs in stack emission in the different FBIs

The distribution of TEQ in the five FBIs is shown in Table 1. The coplanar PCBs contributed on average between 1.6% and 5.3% to the total TEQ value in the

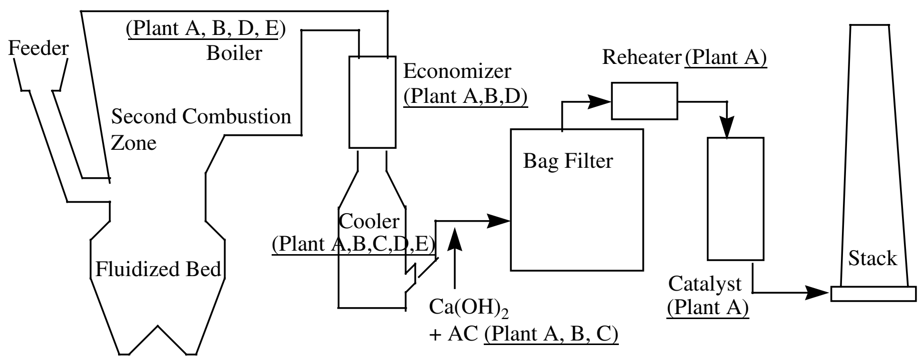

Fig. 1. Flow diagrams of the fluidized bed incinerators including flue gas line and emission control devices of the respective plants (A–E).

Table 1
Average TEQ concentration of PCBs, total TEQ and %-contribution of coplanar PCBs to TEQ in stack gas of the different fluidized bed incinerators and in fly ash in incinerator A

FBI (number of measurements)	Plant A (4)	Plant A (2)	Plant B (5)	Plant C (3)	Plant D (3)	Plant E (2)
Sample	Stack (gas)	Fly ash	Stack (gas)	Stack (gas)	Stack (gas)	Stack (gas)
Air pollution control	BF/AC; Cat		BF/AC	BF/AC	BF	BF
Co-PCB (ng TEQ/N m^3)	0.0006	0.0116[a]	0.002	0.004	0.04	0.09
PCDD/F + PCB(ng TEQ/N m^3)	0.012	1.125[a]	0.071	0.26	1.41	4.05
Co-PCB to total TEQ (%)	5.3%	1.02%	2.5%	1.6%	2.8%	2.3%
Average and (single runs%)	(3.5, 4.8, 5.5, 7.5)	(1.15, 0.9)	(1.8, 1.8, 2.4, 2.5, 4.0)	(1.0, 1.9, 1.9)	(1.9, 2.8, 3.6)	(2.0, 2.6)

[a] ng TEQ/g fly ash.

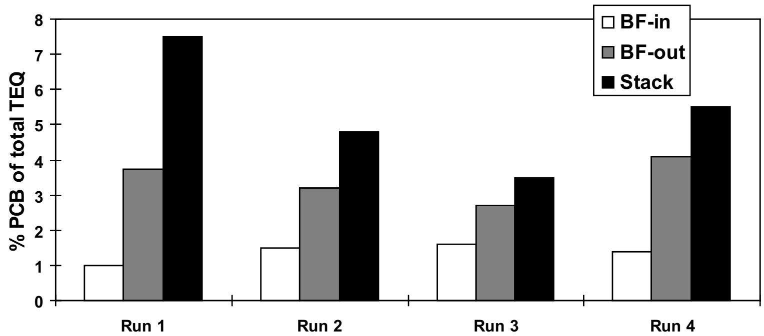

Fig. 2. Contribution of coplanar PCBs to total TEQ at the inlet (BF-in) and outlet (BF-out) of the bag filter and in the stack in plant A.

different plants. This is comparable to the TEQ distribution in grate fired incinerators (Sakai et al., 1993, 1994; Alcock et al., 1998; Johnke et al., 2001).

No correlation was found between the total TEQ emission of the different FBIs and the %-contribution of PCB to total TEQ (Table 1). The use/not use of activated carbon (AC) spray (entrained flow process) did not show a significant effect on the contribution of PCDD/PCDF and PCB to total TEQ (Table 1).

3.2. Impact of coplanar PCB emission on TEQ in incinerator A—effect of the emission control devices

Four of the FBIs sampled (incinerator B–E), showed similar contributions of coplanar PCB to total TEQ (Table 1). The only "outlying" was Plant A, which was found to emit coplanar PCB concentrations at the lower end of all five incinerators, yet their contribution to total TEQ was found to be the highest with 5.3% (Table 1). Therefore we analysed the emissions in plant A in more detail. The flue gas was sampled at three positions: at the inlet of the bag filter (BF-in), at the outlet of the bag filter (BF-out) and in the stack. Fig. 2 shows the contribution of PCBs to total TEQ along this flue gas line. At BF-in, the coplanar PCB amount in all runs to less than 1.7% of total TEQ (average 1.4%). Their contribution increased to more than 2.5% (average 3.3%) at the bag filter outlet. This is comparable to the stack concentration of the other four FBIs (Table 1). Therefore, the differences in the cooling process of the incinerators did not have a significant influence on the ratio of PCDDs/PCDFs and coplanar PCBs formed.

A further increase of PCB contribution to TEQ was found between the BF-out position and the stack with an average contribution of PCBs to total TEQ of 5.3% and a maximum contribution of 7.5% (Fig. 2).

To understand these shifts, a closer analysis of the impact of individual PCBs and PCDDs/PCDFs on TEQ, in combination with the evaluation of the removal efficiency of emission control devices on the relevant congeners is necessary. PCB#126 (3,3′,4,4′,5-P_5CB) amounted to more than 90% of the TEQ contributed by the coplanar PCBs (Fig. 3) and together with PCB#169 (3,3′,4,4′,5,5′-H_6CB), these two non-ortho-substituted coplanar PCB amounted to more than 98% of PCB-TEQ (Fig. 3). This distribution did not significantly

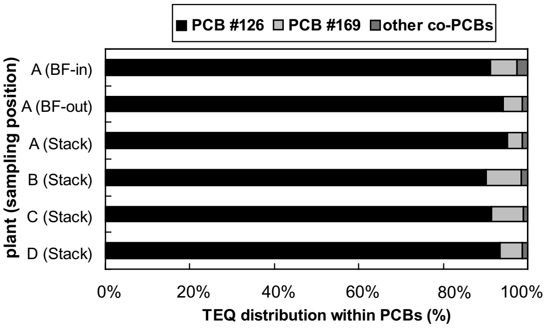

Fig. 3. Distribution of TEQ within the coplanar PCBs at BF-in, BF-out position and stack gas of plant A and at stack position of plants B–D.

change at the different sampling positions (nor for the different incinerators [1]) (Fig. 3). Among PCDDs/PCDFs, the main contribution to total TEQ was found to derive from the 2,3,7,8-substituted P_5CDF and H_6CDF (Fig. 4). Bag filters show a high removal efficiency for PCDDs/PCDFs and PCBs due to the adsorption quality of fly ashes and/or the use of activated carbon spray (Blumbach and Nethe, 1994; Tejima et al., 1994; Sato et al., 1999). The shift in the TEQ distribution between BF-in and BF-out position is a result of the removal efficiencies for the respective compounds. [2] Therefore, less volatile compounds are removed with a higher efficiency. This results for example in an increasing removal efficiency (RE) of PCBs with increasing chlorination degree (Fig. 5), or a considerably lower removal efficiency for PCBz and PxCP compared to PCDDs/PCDFs. With respect to TEQ, the P_5CB#126 is removed with a lower RE compared to the slightly lower volatile 2,3,7,8-substituted P_5CDF and H_6CDF (Fig. 5). This results in an increased contribution of coplanar PCBs to TEQ between BF-in and BF-out (Fig. 2). In addition, we measured two fly ash samples from plant A. The contribution of coplanar PCBs to TEQ was 1.17% and 0.86%, respectively (Table 1), and therefore lower compared to the average contribution of 1.4% of PCBs to TEQ at the bag filter inlet in the raw gas. This further demonstrates that the phenomenon of increasing contribution of PCBs to TEQ between bag filter inlet and outlet is preliminary caused by an adsorption effect, which determines solid phase and gas phase distribution of PCDDs/PCDFs and PCBs (and other semi-volatile compounds).

The second increase of the PCB contribution to total TEQ between BF-out and stack (Fig. 6) was mainly caused by the SCR catalyst. Plant A was equipped with a SCR-catalyst (Table 1) for NO_x removal operated at 210–230 °C. It has been shown that these catalysts are also very effective in the decomposition of PCDDs/PCDFs (Hagenmaier et al., 1990; Fahlenkamp et al., 1991; Ide et al., 1996). The destruction efficiency (DE) of the SCR catalyst for PCBs (DE = 79%) [3] was slightly smaller than for the PCDDs/PCDFs (DE = 91%)[3], which additionally resulted in a higher impact of coplanar PCBs on total TEQ (Fig. 2). The DE of a chlorinated aromatic compound by an SCR catalyst depends here also on the volatility of the respective compound and additionally on the susceptibility towards oxidation (Weber et al., 1999, 2001). The lower DE of the catalyst for PCBs compared to PCDDs/PCDFs can mainly be attributed to the higher volatility of the PCBs. The influence of the volatility on the DE of the catalyst for chlorinated aromatics is further demonstrated by the low removal of PCBz by the catalyst (Fig. 6). Less than 20% of the high volatile D_2CBz and T_3CBz and around 30% of the low volatile P_5CBz and H_6CBz were destroyed (Fig. 6). However, the DE of the catalyst for chlorophenols were similar to the DE of PCBs and PCDDs/PCDFs, although the volatility of chlorophenols and chlorobenzenes is comparable when considering their boiling points. However, due to the hydroxy-group, chlorophenols are more susceptible towards oxidation and chemisorption and therefore they are removed with considerably higher efficiency compared to chlorobenzenes.

An additional influence resulting in the higher impact of coplanar PCBs on TEQ along the flue gas line was due to the standard procedure of GC/MS analysis: While the major contributors to the total TEQ among the PCDFs (2,3,7,8-substituted P_5CDF/H_6CDF) and PCBs (P_5CB#126, H_6CB#169) were detected in all samples, some PCDD congeners (e.g. 2,3,7,8-substituted P5CDD/H6CDD, together contributing between 10% and 20% to total TEQ in plant A (Fig. 4)), were below detection limit in some samples at BF-out and in the stack.

3.3. Implications for measurements of indicator compounds proposed for a PCDD/PCDF (TEQ value) estimation

Recently, several indicator compounds were proposed for an estimation of PCDD/PCDF/PCB (TEQ)

[1] A similar distribution of TEQ within the coplanar PCBs was reported for off gas and fly ash of grate fired municipal solid waste incinerators (Shibayama et al., 2000).

[2] The bag filter was operated below 160 °C and therefore *de novo* synthesis had no significant impact.

[3] Since the catalyst was not optimised for the removal of PCDDs/PCDFs (amount of NH_3 and temperature profile), the destruction efficiency was below the values reported in literature (Hagenmaier et al., 1990; Ide et al., 1996).

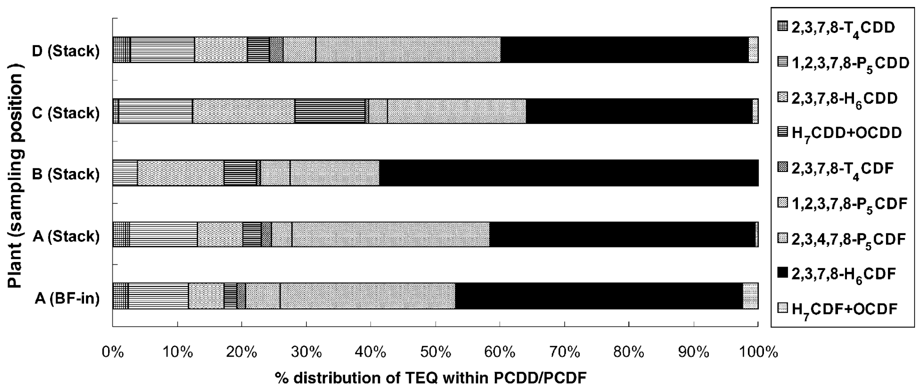

Fig. 4. Distribution of TEQ within the PCDDs/PCDFs in stack gas of plants A–D and at bag filter inlet (BF-in) of plant A.

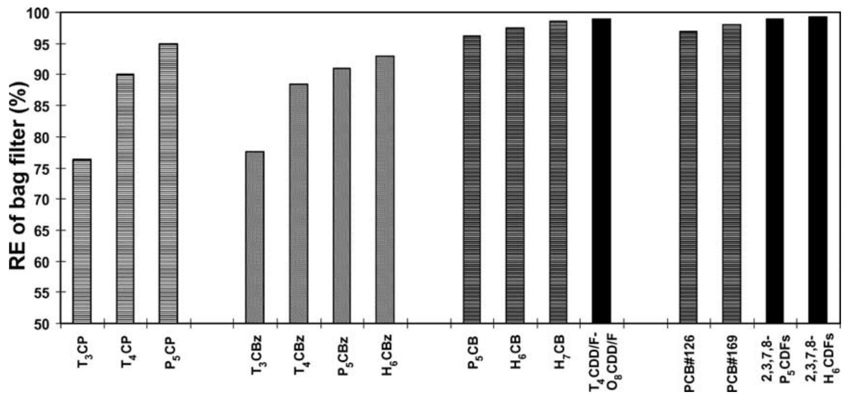

Fig. 5. Removal efficiency of bag filter for chlorophenols, chlorobenzenes, P_5CB to H_7CB and PCDDs/PCDFs and comparison of removal efficiency for PCB#126, PCB#169 and 2,3,7,8-substituted P_5CDF and H_6CDF.

emission from incinerators. These include for example chlorobenzenes (Kaune et al., 1996; Blumenstock et al., 2001; Kato et al., 2001; Urano and Kato, 2001), chlorophenols (Tupparainen et al., 2000; Blumenstock et al., 2001), lower chlorinated PCDD/F (Gullett et al., 2001; Blumenstock et al., 2001) or sum-parameters like semi- and non-volatile organohalogen compounds (Kato et al., 2001; Urano and Kato, 2001). However, the impact of emission control devices was not evaluated or considered in most of these studies with the exception of Kaune et al. (1996) and Blumenstock et al. (2001). Blumenstock et al. (2001) reported on a crucial impact of a wet scrubber on the correlation of chlorophenols to TEQ (especially low chlorinated chlorophenols) and concluded that chlorophenols were not a reliable indicator in stack gas of the investigated incinerator. Kaune et al. (1996) investigated the effect of activated carbon on the correlation of pentaCBz to PCDDs/PCDFs (TEQ value) and found a considerable impact from the use/not use and even the way of application of activated carbon on the correlation.

The emission control devices are normally optimised for PCDD/PCDF removal to comply with the respective emission limits, and the removal efficiency for the indicator compounds can differ considerably from those for PCDDs/PCDFs. For example in our investigation, the removal efficiency of the bag filter for PCDDs/PCDFs in plant A was more than 99% while the removal efficiency for chlorophenols ranged from 75% to 95% (Fig. 5). Further, the destruction efficiency of the SCR catalyst for PCDDs/PCDFs (PCBs) was around 90% (80%) while the DE for the PCBz ranged from 17% to 30% (Fig. 6). Therefore in our opinion, the impact of emission control devices have to be considered and evaluated for each indicator compound when establishing any correlation of the emission of PCDDs/PCDFs/PCBs (TEQ values) and indicator compounds.

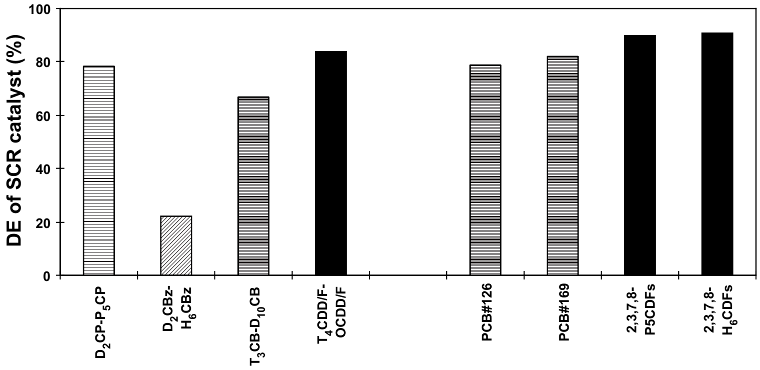

Fig. 6. Destruction efficiency of the SCR-catalyst (225 °C) [4] for chlorophenols, chlorobenzenes, PCBs and PCDD/PCDF and comparison of removal efficiency for PCB#126, PCB#169 and 2,3,7,8-substituted P_5CDF and H_6CDF.

4. Conclusions

In FBIs combusting municipal solid waste, coplanar PCB contribute an average of about 3% to total TEQ. This is comparable with results from grate fired incinerators. The impact of coplanar PCBs from the combustion process (BF-in) was below 2%. Bag filters (with or without carbon injection) or SCR catalysts, also decrease the PCB emission. However, the PCBs are removed slightly less efficiently compared to PCDDs/PCDFs. This results in a relative increase in the impact of coplanar PCBs to total TEQ along the flue gas cleaning line. Since the difference between the volatility of the main contributors to total TEQ (2,3,7,8-substituted P_5CDF/H_6CDF, P_5CB#126 and H_6CB#169) is small, the total impact changes only slightly. Therefore, PCB amounted to less than 8% (on average 2.7%) of total TEQ at the stack position of all measured FBIs. According to reports from the literature, this is also the case for grate fired incinerators. Consequently, the PCDDs/PCDFs still contributed more than 90% (in average 97.3%) to total TEQ in the stack emission, suggesting a minor impact of PCBs from combustion sources [5] on total environmental TEQ burden. In this respect, Johnke et al. (2001) discussed the relevance of coplanar PCB measurements for the evaluation of total TEQ output from municipal solid waste incinerators and concluded that they do not recommend an additional measurement of PCBs for grate fired incinerators in Germany. Also, the present study on FBIs suggests that an obligatory analysis of coplanar PCBs in municipal solid waste incineration samples does not seem to justify the measurement effort of an additional analysis. One possibility might be to include the analysis of PCB#126 (and PCB#169)—contributing more than 90% (>98%) of PCB-TEQ—within the PCDD/PCDF analysis in one GC/MS-injection.

The removal efficiency of air pollution control devices for PCDDs/PCDFs and other chlorinated aromatic compounds differed considerably depending on their volatility or their susceptibility towards destruction by the SCR catalyst. This has to be carefully considered when correlations for PCDDs/PCDFs (TEQ value) and indicator compounds are established and proposed as indicators for a TEQ estimation of stack gas emission.

[4] The catalyst was not optimised for the removal of PCDDs/PCDFs (temperature profile and amount of NH_3) since the emission limit of 0.1 ng TEQ/N m^3 was already achieved at BF-out position. Therefore the DE was considerably below the values reported in literature (Hagenmaier et al., 1990; Ide et al., 1996).

[5] Since the contribution of coplanar PCBs to total TEQ in fish (Scientific Committee on Food, 2000), dairy products (Fürst, 2001) or butter (Santillo et al., 2001; Weiss et al., 2001) amount in average to more than 50%, other sources of coplanar PCB for human intake have to be evaluated and reduced.

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