EMISSION FACTORS FOR UNCONTROLLED BURNING AND SIMULATION OF PCDD/F CONTAMINATION IN OPEN DUMPING SITES

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Introduction

Uncontrolled waste burning, especially in open dumping sites, has recently been focused as a potential source of persistent organic pollutants (POPs) including PCDD/Fs[1-3]. To cope with this issue, previous studies have focused on the following two topics: characterization of PCDD/Fs emission factor for uncontrolled waste burning by lab-scale combustion experiments[4,5]; and elucidation of POPs contamination in soils and mother's milk in open dumping sites[1-3]. However, quantitative relationship between these two research results has not been studied so far. The purpose of this study is to relate the emission factors with the field observations and to obtain some insights on the formation and behaviour of PCDD/Fs from uncontrolled waste burning in open dumping sites.

(1) Using the emission factor for uncontrolled burning of waste, PCDD/Fs concentrations in soils of open dumping sites were estimated and then compared with the actual measurements.
(2) Starting from the PCDD/Fs concentrations in soils, concentrations in cow milk bred in the dumping sites and concentrations in human breast milk exposed through soil ingestion and milk consumption were predicted. The predicted concentrations were compared with the actual measurements to confirm the major exposure route.

Methods

Estimation of PCDD/Fs concentrations in soils of open dumping sites

The relationship between the PCDD/Fs emission factor for uncontrolled burning of waste, EF_{total}, and the PCDD/Fs concentration in soil of open dumping site, C_{soil}, is expressed by:

$$C_{soil} = \frac{(1-R_{burned})(1-MC_{waste})C_{waste} + R_{burned}EF_{total}R_{dr}}{(1-R_{burned})(1-MC_{waste}) + R_{burned}R_{residue}} \quad \ldots(1)$$

C_{soil}: PCDD/Fs concentration in soil of the open dumping site [pg/g-dry]
EF_{total}: PCDD/Fs emission factor for uncontrolled burning of waste [pg/g-wet]
R_{burned}: Fraction of waste burned in the open dumping site [g-wet/g-wet]
R_{dr}: Ratio of PCDD/Fs that remains in the open dumping site to the PCDD/Fs that is formed during the uncontrolled burning of waste [-]
C_{waste}: PCDD/Fs concentration in solid waste [pg/g-dry] (2 pg-TEQ/g-dry)
MC_{waste}: Water content in solid waste [g-water/g-wet] (50%)
$R_{residue}$: Ash content in solid waste [g-dry/g-wet] (10%)

The range of EF_{total} was set from 100 to 6,000 pg-TEQ/g-waste, based on the results of lab-scale combustion experiments by Sakai et al.[4] and the barrel burn experiments by Gullett et al.[5]

The range of R_{burned} was set from 1% to 5%, based on a survey on landfill fires in Finland by Ettala et al.[6] and an estimation of the waste combustion rate in dump sites in India by Gupta et al.[7]

R_{dr} can be described as the sum of PCDD/Fs found in incineration residue ($EF_{residue}$) and PCDD/Fs that deposit on the dump site after emitted to the air (EF_{gas} $R_{deposition}$), and satisfies the condition (3).

$$R_{dr} = \left(EF_{residue} + EF_{gas} R_{deposition}\right) \Big/ EF_{total} \quad \ldots(2)$$

$$R_{dr} > EF_{residue} \Big/ EF_{total} \quad \ldots(3)$$

Our lab-scale combustion experiments showed that the ratios of $EF_{residue}$ to EF_{total} ($EF_{residue}/EF_{total}$) were mostly below 10% when the excess air ratio was 7.0, and around 30% when the excess air ratio was 1.5. These results indicate that the R_{dr} is inversely correlated to the excess air ratio of the waste combustion. We can conjecture that the R_{dr} is relatively high under smoldering combustion often seen in landfill fires[5].

Prediction of the PCDD/Fs concentrations in cow milk and human breast milk

The daily intakes of PCDD/Fs by residents near dump sites were calculated for the three exposure routes: (1) inhalation; (2) ingestion of soil; and (3) drinking of cow milk bred in open dumping sites in India. As for (3), the concentration in cow milk was calculated by the equation (4).

$$P_{cow-milk} \times C_{cow-milk} = A_{feed} \times R_{soil} \times C_{soil} \times COR \quad \ldots(4)$$

$P_{cow\text{-}milk}$: Daily milk production by a cow [L/day.head] (20 L/day.head)
$C_{cow\text{-}milk}$: PCDD/Fs concentration in cow milk [pg/L]
A_{feed}: Daily feed consumption by a cow [g-dry/day.head] (18,000 g-dry/day.head)
R_{soil}: Ratio of soil ingestion to feed consumption [g-dry/g-dry] (0.07: for buffalos) [8]
COR: Congener-specific carry-over rate from feed to cow milk [-] [9]

The PCDD/Fs concentration in human breast milk was calculated by the equation (5). The following conditions were assumed: (1) The mother gave the first birth at 25 years old; (2) The daily intake of PCDD/Fs has been constant for the 25 years; (3) PCDD/Fs concentration in body fat equals in each part of the body including breast milk.

$$C_{mother-milk} \times 1000 \times BW \times R_{fat} = \int_{t=0}^{25} 365 \times DI \times \left(\frac{1}{2}\right)^{\frac{t}{DT50_{human}}} dt \quad \ldots(5)$$

$C_{mother\text{-}milk}$: PCDD/Fs concentration in human breast milk (per fat) [pg/g-fat]
DI: Daily intake of PCDD/Fs [pg/person.day]
$DT50_{human}$: Congener-specific half life in human body [year] by Hashimoto et al.[10]
BW: Body weight of mother [kg] (50 kg)
R_{fat}: Body fat percentage of mother [-] (20%)

Results and Discussion

Figure 1 shows the ranges of EF_{total} and R_{dr} which correspond to the predicted soil concentrations of 52 pg-TEQ/g (the average of measurements in India)[1] and 400 pg-TEQ/g (the average of measurements in Cambodia)[1]. To explain the soil concentration observed in India (52 pg-TEQ/g), the emission factor (EF_{total}) must be higher than 500 pg-TEQ/g (point A in Figure 1). In case of Cambodia (400 pg-TEQ/g), EF_{total} must be higher than 4,000 pg-TEQ/g (point B in Figure 1). These minimum estimates for the emission factor are higher than the previous estimate for the emission factor (140 pg-TEQ/g) used in the US EPA's draft inventory. [11]. This result suggests that

the uncontrolled burning of waste as a source of PCDD/Fs is more important than was previously thought.

The ratio of PCDD/Fs that remains in the open dumping site (R_{dr}) was more than 8% in India (point C in Figure 1) and more than 60% in Cambodia (point D in Figure 1). On the other hand, Lorber et al. (1998)[12] estimated the PCDD/Fs concentration in soil within a 3km radius of incineration furnace at less than 2% of the total PCDD/Fs released. Lohman et al. (2001)[13] conducted a model simulation to assess the fraction of emitted PCDD/Fs that would deposit within 100 km from the source, and found that the fraction ranged from 8% to 16% in case of municipal solid waste incinerators (with stack height = 64m) and from 18% to 40% in case of uncontrolled medical waste incinerators (with stack height = 8.5m). Comparison of these results suggests that the local impact of PCDD/Fs formed in the open dumping sites is more significant than that of municipal solid waste incinerators.

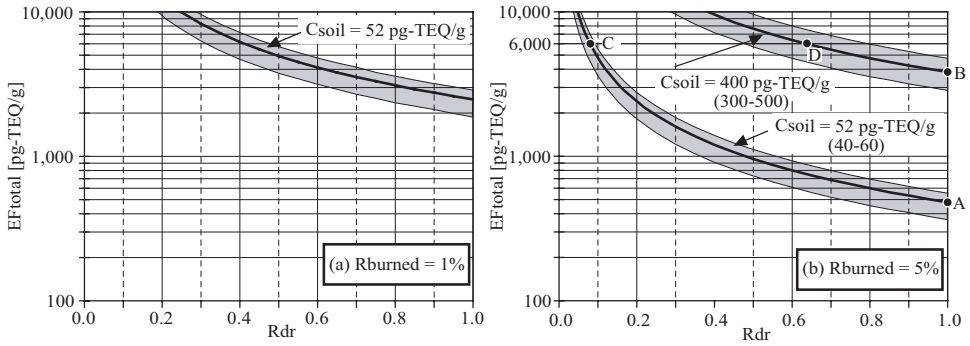

Figure 1: Ranges of EF_{total} and R_{dr} which correspond to the predicted soil concentrations of 52 pg-TEQ/g and 400 pg-TEQ/g for (a) R_{burned} = 1% and (b) R_{burned} = 5%

Table 1 shows the predicted and observed PCDD/Fs concentrations in cow milk bred in open dumping sites and control sites. The predicted concentration for cow milk in dumping site was between the observed concentrations for buffalo milk and cattle milk. The predicted concentration for control site was 2 orders of magnitude smaller than the observed values. This underestimation is reasonable because only the soil ingestion is modeled as an exposure route to the cows. Taking into account uncertainty of the model and variance of the concentrations observed, we can say that the predictions and the observations agreed well. This result is evidence to support the interpretation that the increased concentrations in cows milk in dumping site is caused by the ingestion of dump site soils.

Table 1: Predicted and observed PCDD/Fs concentrations in cow milk.

Site	Soil (observed)[1] [pg-TEQ/g-dry]	cow milk (predicted) [pg-TEQ/L]	cattle milk (observed)[3] [pg-TEQ/L]	buffalo milk (observed)[3] [pg-TEQ/L]
dumping site (India)	52	600	400	1400
control site (India)	0.22	3.1	130	160

Table 2 shows the predicted and observed PCDD/Fs concentrations in human breast milk. The predicted concentrations for control sites both in India and in Cambodia were about 100 times lower than the observed concentrations. This means that the main source of PCDD/F in control

sites are not soils but those omitted in this model, for example fish and meat. The predicted concentrations for dumping sites both in India and in Cambodia were close to the observations. The agreement in Cambodia, however, means that the exposure through soil ingestion is probably overestimated by the model. Nonetheless, the model successfully reproduced the contrast in observed concentrations between the dumping site soils (India < Cambodia) and the human breast milk (India > Cambodia). This result quantitatively supports the speculation that the feeding of cows in the open dumping site is the main factor to cause high concentrations in human breast milk of the residence in the dumping site in India.

Table 2: Predicted and observed PCDD/Fs concentrations in human breast milk

Site	soil (observed)[1] [pg-TEQ/g-dry]	human breast milk (predicted) [pg-TEQ/g-fat]	human breast milk (observed)[2] [pg-TEQ/g-fat]
dumping site (India*)	52	33	38
control site (India*)	0.22	0.16	12
dumping site (Cambodia**)	400	11	9.2
control site (Cambodia**)	1.9	0.054	7.8

* Exposure through cow milk is included. ** Exposure through cow milk is not included.

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