POPs Contamination Caused by Use of Plastic Waste as Fuel at Locations in Indonesia

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1 Introduction

Many toxic additives in plastic waste can leak into the environment when disposed of or burned, including chemicals listed in the Stockholm Convention (SC)[1] such as polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCD), short-chained chlorinated paraffins (SCCPs) or perfluorooctane sulfonate (PFOS)[2-4]. Other toxic chemicals are generated when plastic waste is incinerated as fuel, including those listed as unintentionally produced POPs (UPOPs) in Annex C to the Stockholm Convention, for example, polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs)[a], dioxin-like PCBs (dl PCBs), hexachlorobenzene (HCB) or pentachlorobenzene (PeCB)[3]. Contamination by all these chemicals was documented recently at sites where various plastic wastes are burned or incinerated in developing countries, e.g. Ghana, Cameroon[5], Kenya, Tanzania[6], Kazakhstan, China, Thailand[7,8] or Indonesia[9]. In Indonesia, plastic waste imported from developed countries is often used as fuel in the production of tofu or lime[10]. We focused on mapping pollution by POPs in villages of Karawang Regency, West Java, at sites affected by using plastic and rubber waste (including used tires) as a fuel in lime kilns. Four pooled egg samples in this study were already included in a recently published global review of dioxin and dl PCBs in eggs[11], and this study partly builds on its results.

2 Materials and Methods

Pooled samples of four individual egg samples from free range hens were collected at three selected sampling sites in Tamansari, Pangkalan subdistrict of Karawang Regency in order to obtain more representative samples. We also used a sample of a pooled eggs sample from a convenience store located in the same subdistrict as a reference sample to exhibit background levels of POPs, following precedents from other studies[12,13]. The eggs were collected into typical plastic egg packaging and were boiled for approximately 7 minutes. The homogenates from the edible parts of the eggs were used for the analyses in the laboratories. The samples for analysis were mixed from 4 eggs from the same location.

The residual ash from lime kilns and/or ash dumped nearby, and soil samples, all of which were accessible to chickens foraging, were taken. These were also pooled samples from 5 point samples. Soil was taken at a depth of 2 to 10 cm from the surface by a stainless steel shovel. Samples were homogenized in a stainless steel bowl. They were transported in 250 ml PE plastic containers to the laboratory and kept in cool conditions during storage and transportation.

One of the pooled egg samples (KAR-EGG-1) was taken in a separate part of the village marked as locality Karawang I, a little bit further north from samples of ash and eggs labelled sample KAR-EGG-2. The majority of samples (KAR-EGG-2, KAR-ASH-1-3 and KAR-SOIL-1) were taken near the part of the village with concentrated lime kilns, and it is marked on the map at Figure 1 as Karawang II. Most of the samples of plastic and rubber wastes were also taken at this location. Samples of ash, soil and a third egg sample (KAR-ASH-4, KAR-SOIL-2, KAR-EGG-3) were taken further south from the major group of samples, approximately 1 km away and is labelled Karawang III.

All samples were analyzed for their content of individual PCDD/Fs and dl PCBs by GC/HRMS in an ISO 17025 accredited laboratory with a resolution >10,000 using ^{13}C isotope labelled standards. PCDD/F and dl-PCB analysis in eggs followed the methods of analysis for the control of levels of PCDD/Fs and dl-PCBs in foodstuffs according to EU regulations[14]. The results are presented in pg WHO TEQ g^{-1} of fat. TEFs defined in 2005[15] were used to evaluate dioxin toxicity in all samples.

Analyses of PBDEs, HBCD, novel brominated flame retardants (nBFRs), 17 PFASs (including PFOA, PFOS and PFHxS), PeCB, HCB, hexachlorobutadiene (HCBD), and seven indicator PCB (iPCBs) congeners were conducted in a Czech certified laboratory at the Department of Food Chemistry and Analysis of the University of Chemistry and Technology in Prague. The analytes were extracted by a mixture of organic solvents, hexane: dichloromethane (1:1). The extracts were cleaned by means of gel permeation chromatography (GPC). The identification and quantification of the analyte was conducted by gas chromatography coupled with tandem mass spectrometry detection in electron ionisation mode for HCB, PeCB, HCBD, and indicator PCBs. Novel BFRs analysis was described in one of the previous reports from Indonesia[10].The extract was transferred into cyclohexane and diluted. The identification and quantification of SCCPs were performed via gas chromatography/time-of-flight high-resolution mass spectrometry (GC/TOF-HRMS) in the mode of negative chemical ionisation (NCI).

[a] An acronym ‘dioxins’ is used for PCDD/Fs.

Figure 1: Map with group of samples in the middle of lime kilns concentrated around the road, locality Karawang II.

The identification and quanti-fication of PBDEs were performed using gas chromatography coupled with mass spectrometry in negative ion chemical ionisation mode (GC-MS-NICI). The identify-cation and quantification of HBCD isomers and selected PFASs were performed by liquid chromatography interfaced with tandem mass spectrometry with electrospray ionisation in nega-tive mode (UHPLC-MS/MS-ESI).

The results of POPs analyses in eggs were also compared with EU and Indonesian standards set for food stuff[16,17]. The carry-over rates which were determined by the Dutch National Laboratory[18] were used for re-calculation of PCDD/F and PCB congener profiles in eggs for better comparison with profiles in ash and soil samples. The carry-over rates for PCDD/Fs range from 44% (TCDD) to 10% (OCDD). The transfer factors for dl-PCBs are, on average, high and ranged from 80% (PCB-167) to 41% (PCB-81). For some comparison with source patterns, these carry-over rates are used to roughly recalculate the measured PCDD/F and PCB data in eggs. The same method was used in a recently published review of PCDD/Fs and PCBs in eggs[11]. Six samples of tire, plastic and rubber waste used as fuel were also analyzed for some of the analyzed POPs.

3 Results

Results of chemical analyses for pooled samples of free range chicken eggs, ash and soil are summarized according groups of samples from same locations in Table 1, below.

Tire, plastic and rubber waste used as fuel, also partly analyzed for some of the POPs measured in ash, soil and eggs, had mainly exhibited POPs as follows: Tires contained significant level of iPCBs (172 ng/g), and some other plastic wastes contained PBDEs (35 and 170 ng/g respectively) and nBFRs (0.9; 2.2 and 261 ng/g). These POPs were <LOQ in all other measured samples of plastic waste or rubber.

Comparison of PCDD/Fs and dl PCBs patterns are in graphs at Figures 2 and 3 below.

Table 1: Results of the analyses for POPs in pooled samples of eggs, ash and soil from localities Karawang I, II and III and reference sample of eggs. All results for egg samples are expressed per gram of fat, except PFASs expressed per gram of fresh weight. Results are in ng/g if not marked otherwise.

Locality	Karawang II				
Sample ID	KAR-EGG-2	KAR-ASH-1	KAR-ASH-2	KAR-ASH-3	KAR-SOIL-1
Matrix	Eggs	Ash	Ash	Ash	Soil
Fat content (%)	13.5%	/	/	/	/
PCDD/Fs (pg TEQ/g)	178	5.6	536	349	/
dl PCBs (pg TEQ/g)	34	0.19	43	5	/
HCB	16	0.034	991	273	10
PeCB	6.1	0.041	414	225	5.5
HCBD	<0.10	0.12	0.048	0.16	0.24
6 iPCB	98	0.044	0.34	0.36	19
13 PCN congeners	2.4	<LOQ	2.8	0.14	1.9
SCCPs	69	/	/	/	/
sum HBCD	39	<LOQ	<LOQ	<LOQ	1.71
sum of PBDEs	73	<LOQ	0.055	<LOQ	1515
sum of nBFRs	18	1.9	1.3	0.048	1456
sum of PFASs	4.2	/	/	/	/

Locality	Karawang I	Karawang III			Reference
Sample ID	KAR-EGG-1	KAR-EGG-3	KAR-ASH-4	KAR-SOIL-2	KAR-EGG-R
Matrix	Eggs	Eggs	Ash	Soil	Eggs
Fat content (%)	13.8%	16.7%	/	/	17.4%
PCDD/Fs (pg TEQ/g)	11	109	14	72	0.23
dl PCBs (pg TEQ/g)	1.7	9.8	0.2	1.7	0.02
HCB	4.6	4.8	0.15	0.76	5.0
PeCB	0.70	3.5	0.274	1.6	1.9
HCBD	<0.10	0.13	0.034	<0.02	2.1
6 iPCB	4.5	5.7	0.17	0.29	4.8
13 PCN cong.	<LOQ	<LOQ	<LOQ	0.542	<LOQ
SCCPs	<50	237	/	/	151
sum HBCD	47.57	36.47	<LOQ	<LOQ	<LOQ
sum of PBDEs	<LOQ	<LOQ	<LOQ	2.1	<LOQ
sum of nBFRs	<LOQ	<LOQ	0.03	19	<LOQ
sum of PFASs (ng g^{-1})	0.85	2.3	/	/	0.05

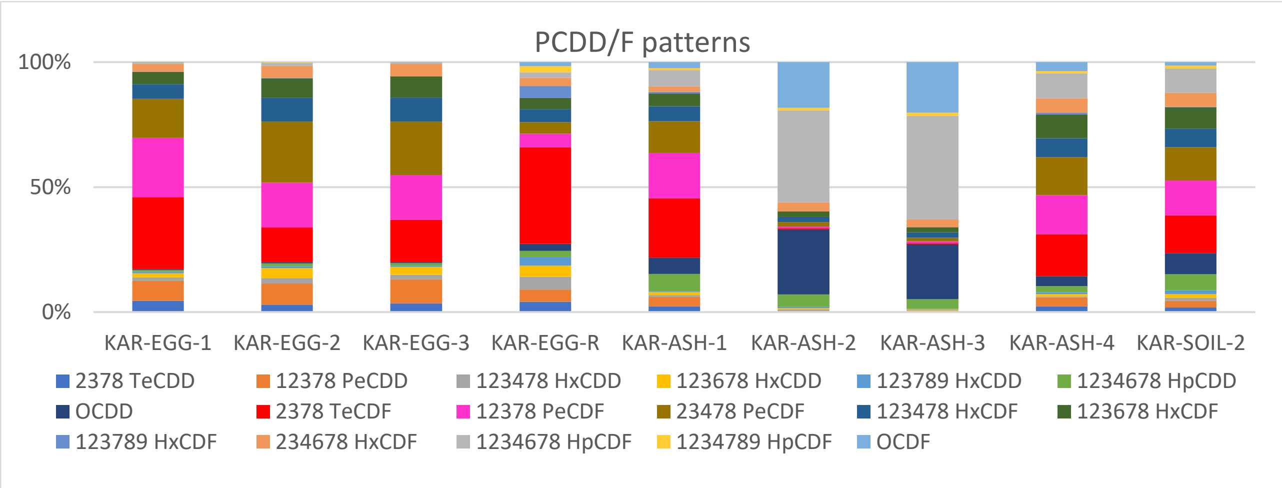

Figure 2: PCDD/F patterns.

High levels of the UPOPs PCDD/Fs, dl PCBs, HCB, and PeCB, were measured in two samples of ash and free range chicken eggs from Karawang II. Free range chicken eggs samples and soil samples from Karawang III contained high levels of PCDD/Fs. Very high level of PBDEs and nBFRs was measured in soil samples from Karawang II. PCDD/Fs levels were much higher in "fresh" ash samples taken in close vicinity (5 to 10 m) of the lime kilns (KAR-ASH-2 and 3) while they were lower in older, weathered ash samples (KAR-ASH 1 and 4). These groups of ash samples had also very different congener patterns. Weathered ash samples had similar patterns to soil which can be caused due to the fact that weathered ash samples were already partly mixed with surrounding soils. PCNs were measured at low levels or they were not detected above LOQ in half of all samples. High levels of six indicator PCBs was measured in eggs and soil samples from Karawang II which is interesting when compared to the significant iPCBs level also found in used tire samples.

All three free range chicken egg samples exhibit a waste incineration pattern of dl PCB congeners (see Figure 3) according to the dl PCB congener profile already established in a previous study[11]. However, egg samples from Karawang II has dl PCB pattern very close to Aroclor 1254[11].

4 Discussion

In two free range chicken egg samples presented in this study KAR-EGG-2 and 3 were measured as the eighth and eleventh highest level of PCDD/Fs in eggs respectively in comparison with other samples included in the global review and from scientific literature[11]. They exhibit the same level of contamination with dioxins as samples from Tropodo, another locality in Java, Indonesia contaminated by incineration of plastic waste used as fuel in tofu production facilities. Dioxin concentrations in these eggs exceeded the EU standard of 2.5 pg TEQ/g fat[17] by more than 71 and 43 fold respectively. The EU standard of 40 ng/g for six iPCB congeners[17] was exceeded in KAR-EGG-2 by more than twice.

Higher levels of dioxins in free range chicken eggs from Asia were observed only in Bien Hoa, a former US Army base in Vietnam contaminated by Agent Orange[19] or in Tropodo[11,20].

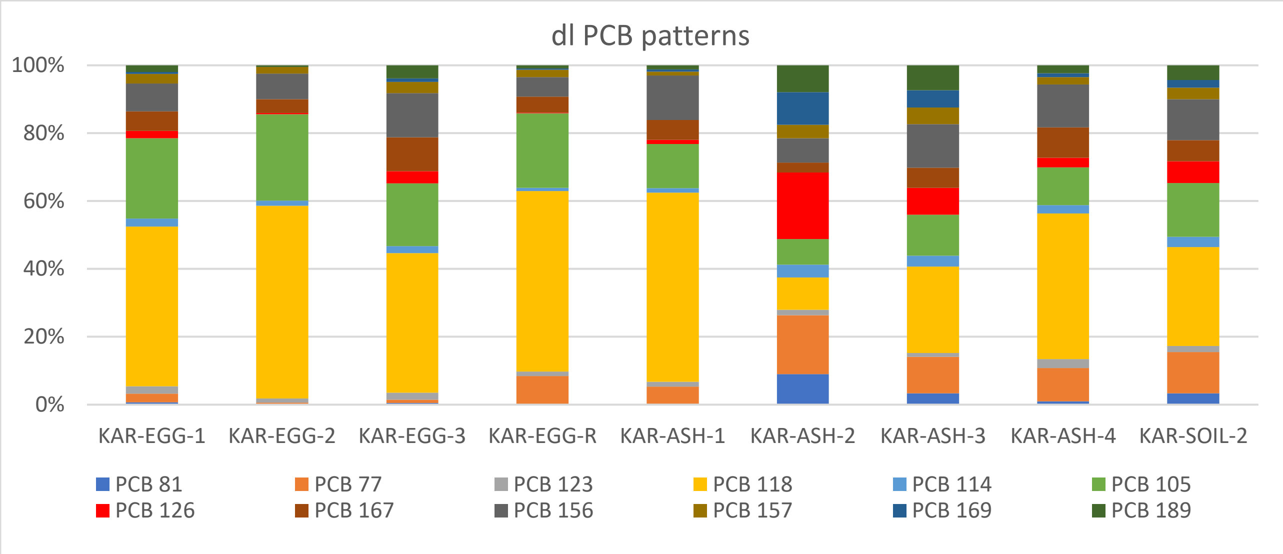

Figure 3: Dl PCB patterns in samples from Karawang.

KAR-SOIL-1 sample had very high concentrations of BFRs, both regulated (PBDEs) and unregulated (nBFRs) by the Stockholm Convention[3]. Their concentration in this sample is much higher in comparison with levels measured in samples of plastic waste from the locality Karawang II but it is not possible to monitor BFRs in large volumes of plastic mixed in fuel for lime kilns. BFRs as well as PCBs most likely only evaporate from plastic and rubber waste under temperatures in lime kilns. The level of PBDEs measured in soil sample KAR-SOIL-1 of 1515 ng/g dw exceeds the level measured in mixed soil and ash samples from a plastic waste scrap yard in Bangun and exceeds the background level of 0.04 ng/g dw in soil samples from a clean reference site in Mbeji forest by almost 38 thousand fold[10].

Two ash samples KAR-ASH-2 and 3 contained very high concentrations of HCB and PeCB. HCB levels in these two samples exceeded the level of 34 ng/g measured in ash from Tropodo[10] by 29 and 8 fold respectively. PeCB and HCB levels in these ash samples exceed levels observed in ashes from small medical waste incinerators in the Czech Republic[21] and Africa[5] by several thousand and almost 250 fold respectively.

Free range chicken eggs and soil in Karawang are contaminated with POPs as consequence of incineration of plastic and rubber waste in lime kilns. Pollution occurs by several pathways, via air pollution but also in ash which is dumped next to lime kilns. This pollution is very serious although the levels of POPs measured in ash produced by lime kilns are below currently established limits for definition of POPs waste by both Basel and Stockholm Conventions, including dioxins (1 or 15 ng TEQ/g), dl PCBs (no limit established yet) or PBDEs (50 or 1000 mg/kg for PBDEs without decaBDE)[22]. Despite that, the ashes which are widespread, represent a serious threat to the environment in Karawang district. This situation repeats similar cases observed in Tropodo (ash from plastic waste incineration in tofu factories), Kendalsari (ash from aluminum smelters), both in East Java, or Accra in Ghana (medical waste incinerator ash).

Plastic waste in Indonesia and other developing countries is also promoted as Refuse-Derived Fuel (RDF) but not controlled[23]. Plastic waste was used as fuel in so called 'community cookers' in Kenya for example. In the vicinity of one of these cookers, dioxin contamination of chicken eggs exceeding EU standard by several folds was also observed[6,24].

5 Conclusions

Very serious contamination of the environment and food chain with POPs as result of using plastic and rubber waste as fuel in lime kilns in Karawang Regency was confirmed by measurements of samples of ash, soil and free range chicken eggs in this study. The contamination of eggs with dioxins represents the highest ever measured levels in Asia and globally. It is necessary to avoid using plastic waste as fuel in facilities like lime kilns or tofu factories in particular. More strict limits for the definition of POPs waste are needed to regulate disposal options for waste produced by waste incineration, including facilities like lime kilns burning plastic waste.

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