TOXIC HOT SPOTS IN JAVA

Research Report

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Toxic Hot Spots in Java and Persistent Organic Pollutants (POPs) in Eggs

Research Report

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December, 2020

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Photo on cover page: Prigi Arisandi, Ecoton.

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Executive Summary

Persistent Organic Pollutants (POPs) contamination in developing countries can include both domestic and foreign sources of pollution. This study focused on sites potentially polluted by such sources on the island of Java, Indonesia, in particular sites affected by plastic and paper waste imports, secondary aluminum production, and waste incineration.

Developed countries sharply increased exports of non-recyclable plastic waste to Indonesia and other Southeast Asian countries after China closed its doors to plastic waste imports in 2018. As a result, Indonesia's plastic waste imports doubled to 320,000 tons in 2018 compared to 2017. Based on observations by Ecoton and Nexus3, between 25% and 50% of the plastic wastes imported by Indonesian plastic and paper recycling companies were mismanaged.

Aluminum is another material used widely in beverage packaging and car production. Waste from primary aluminum production and discarded aluminum scrap, often used for secondary aluminum production, are also sources used in smelters located in the Jombang regency.

There are many toxic additives in plastic wastes that can leak into the environment when they are disposed or burned, including chemicals listed in the Stockholm Convention. Toxic chemicals are also involved in secondary aluminum production and contained in aluminum scrap. These include flame retardants such as PBDEs and SCCPs, surface treatment chemicals such as PFOS, and substances such as dioxins that the treaty requires to be continuously minimized with the aim of elimination.

The Stockholm Convention regulates these substances because they are unmanageable due to their persistence, bioaccumulation, long-range transport, and toxicity to both humans and living organisms. Some of these chemicals also accumulate in ash residues from both waste burned as fuel (waste incineration) and in aluminum smelters. POPs can further leak from ash, which is widely used as material for roads or as construction material, and contaminate food chains.

This study examined toxic chemical contamination of free-range chicken eggs from five sites in Indonesia: Tropodo, where plastics are burned as fuel in tofu factories; Bangun and Tangerang, where plastics are dumped on the ground and burned; Lakardowo, where a privately owned hazardous waste incinerator facility is located; and Kendalsari, where dozens of secondary aluminum smelters operate.

Globally regulated toxic substances contaminating the eggs and analyzed in our study include polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs, called "dioxins" in brief), PCBs, HCB, PeCB, SCCPs, PBDEs, HBCD, and PFAS substances such as PFOS. We also included analyses of novel Brominated Flame Retardants (BFRs) which replaced already regulated PBDEs and HBCD, and polybrominated dibenzo-p-dioxins and dibenzofurans (PBDD/Fs, called "brominated dioxins" in brief), which are not regulated yet but exhibit the same toxicity as PCDD/Fs.

The results of the chemical analyses revealed levels of POPs among the highest ever measured in several pooled free-range chicken egg samples. In the pooled samples of free-range chicken eggs from Tropodo, we found outrageous levels of 200 and 140 pg TEQ g^{-1} fat of dioxins, respectively. The regulatory limit in Indonesia is 2.5 pg WHO-TEQ g^{-1} fat, but includes both dioxins and dioxin-like PCBs. These are the third- and the fourth-highest levels of dioxins in eggs from Asia ever measured and the sixth- and seventh-highest levels of dioxins in eggs found globally. The level of PBDEs in egg samples taken in November 2019 from Tropodo was the second-highest level ever measured of these flame retardants in eggs globally.

The chicken eggs from Kendalsari and Tangerang belong together with two pooled eggs samples from Tropodo to samples from this study, which are among 20 egg samples with the highest ever measured levels of dioxin globally. Very high levels of HBCD, a brominated flame retardant used mainly in polystyrene foams, were measured in egg samples from Bangun and Tangerang. They are among the ten highest ever measured levels of HBCD in eggs globally.

PLASTIC WASTE DUMPSITES

Analyses performed in this study have shown that plastic landfills on the island of Java are not only a waste problem, but they are also a source of environmental contamination from a wide range of persistent organic pollutants. Many of them are already contained in the plastics themselves as additives, but others are created by burning waste to clear space for new waste brought in for sorting.

The level of POPs contamination caused by dumping, incineration, and open burning of plastic waste ranks some sites on Java among the most contaminated in the world, alongside sites heavily affected by industrial production or sites contaminated due to military conflicts.

Dioxin levels in the eggs from plastic waste dumpsites exceeded the EU regulatory limit by 4- to 22-fold. The eggs from Tangerang also had high levels of brominated dioxins and HBCD. The egg samples from Bangun had high levels of PBDEs, HBCD, and PFOS. Currently, there is no limit for perfluorinated compounds, including PFOS, in Indonesia, despite their high toxicity.

E-WASTE

It was most likely plastics from e-waste that contributed significantly to the food chain contamination in Bangun, Tropodo, and Tangerang found during the November 2019 round of sampling. This was reflected in the high concentrations of brominated flame retardants found in free-range chicken eggs.

In the case of Tangerang, we also found a significant contribution of brominated dioxins to the overall toxicity of the chicken eggs samples, due to plastic residues from refrigerator insulation.

WASTE INCINERATION

In the vicinity of the hazardous waste incinerator facility in Sumberwuluh, Lakardowo, we found contamination of hens' eggs, mainly with dioxins and dioxin-like PCBs. In Tropodo, where plastic wastes separated in Bangun village are burned, we also found high concentrations of PBDEs in eggs.

The combustion in the tofu factory's furnace does not reach temperatures that cause PBDEs to decompose. The PBDEs instead accumulate in the dust and enter the food chain. Although the level of dioxins and dl-PCBs was higher in the tofu factory alone compared to the reference sample, it was not heavy contamination.

The analytical results of POPs in tofu samples from Tropodo showed potential trace contamination from the practice of burning plastic waste as fuel. High contamination of tofu as such was not expected, as it was not produced from locally grown soya beans, and also because POPs do not accumulate in the water in which the tofu is boiled in the factories. Thus, the potential pathway may be factory dust and soot that gets into the water, but POPs are not soluble in water. It is, rather, local food of animal origin rich in animal fats that is contaminated by the practice of plastic waste incineration in tofu factories.

The level of dioxin contamination of the food chain in Tropodo has reached the level of sites such as the Bien Hoa former U.S. Army base in Vietnam, a loading site for Agent Orange during the Vietnam war . Another site for comparison is the infamous e-waste scrap processing site in Guiyu, China, which is contaminated with brominated flame retardants, especially PBDE.

SECONDARY ALUMINUM SMELTERS

Secondary aluminum smelters in the Jombang Regency are significant sources of releases of dioxins, dl-PCBs, and possibly PBDEs into the environment. This was demonstrated by analyses of hens' eggs, rice crop, soil, ash, and dust from the villages of Kendalsari and Sidokampir. The level of dioxin contamination of eggs from free-range hens ranked the Kendalsari samples as the seventh and eighth highest among those analyzed from Asia so far and as the 15th and 19th highest among the eggs sampled worldwide. In addition, the very high levels of dl-PCBs, PCDD/Fs and dl-PCBs in the egg samples from Kendalsari exceeded the Indonesian limit for eggs by 24- and 34-fold respectively.

The use of ash from secondary aluminum production as a building material to strengthen roads, flood defenses, and building foundations has been shown to be a significant source of environmental contamination in the Jombang Regency.

ASH RESIDUES

The situation in the Jombang Regency around villages where secondary aluminum smelters are located, and in Tropodo documents that dioxin-containing ash as a result of combustion processes causes or significantly contributes to the contamination of food chains with POPs.

The ash from the combustion processes in both Tropodo (plastic combustion) and Jombang Regency (secondary aluminum smelters) contains dioxin concentrations well below the current provisional Low POPs Content Level of 15 ng g^{-1} dw (= 15 ppb).[1] However, the dioxin levels observed in eggs exceed the recommended acceptable limits for their consumption by many orders of magnitude.

A similar situation has been mapped in several cases in different locations around the world. The overly lenient setting of limits for dioxins

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[1] The limit called "low POPs content" in Article 6 of the Stockholm Convention defines when the waste is considered to be a POPs waste, which has to be managed in special ways defined in Article 6; Stockholm Convention (2010). Stockholm Convention on Persistent Organic Pollutants (POPs) as amended in 2009. Text and Annexes. Geneva: 64. It is established at levels of 1 or 15 ppb (ng TEQ g^{-1} dw) for PCDD/Fs in the last update of the General Technical Guidelines for POPs Waste; Basel Convention (2017). General technical guidelines for the environmentally sound management of wastes consisting of, containing or contaminated with persistent organic pollutants. Technical Guidelines. Geneva. The EU uses the level of 15 ppb of PCDD/Fs in waste, as set in its last update of the POPs Regulation; European Parliament and Council (2019). "Regulation (EU) 2019/1021 of the European Parliament and of the Council of 20 June 2019 on persistent organic pollutants (Text with EEA relevance.)." Official Journal of the European Union: 33.

in waste leads to uncontrolled handling of residual ashes from various combustion sources. Although we monitored the concentrations of dioxins and other POPs only in free-range chicken eggs, the milk and meat of cows and other cattle consumed by the locals may be similarly contaminated.

Locally produced food is of great importance in developing countries and rural locations in developed countries; therefore, the exposure scenario when ash-containing dioxins contaminate the food chain is of particular concern. Two case studies from the sites in Indonesia underline the need for more strict controls of POPs in wastes that are allowed to be used freely at places in direct or indirect contact with agricultural or rural areas where the local food is produced and/or with residential areas. The demonstrated cases also show that waste with dioxin contents even far below 1 ppb should be restricted from direct use in such areas.

POPs IN EGGS

UNINTENTIONALLY PRODUCED POPs (UPOPs)

Analyses of nine pooled samples of free-range chicken eggs have shown widespread contamination by PCDD/Fs and dl-PCBs at all selected hotspots. All free-range chicken egg samples in this study exceeded the Indonesian and EU maximum levels (ML) for PCDD/Fs and the sum of PCDD/Fs and dl-PCBs. All samples were also above the background level of WHO-TEQ measured in eggs from the supermarket by more than 4,060-fold (sample from Sumberwuluh) to 72,500-fold (sample from Tropodo). Brominated dioxins (PBDD/Fs) contributed significantly to the overall toxicity of free-range eggs from the site in Tangerang affected by plastics from e-waste.

None of the egg samples contained levels of HCBD above the laboratory Limit of Quantification (LOQ). Also, levels of HCB and PeCB were relatively low and did not exceed the established EU limit for eggs.

PBDEs AND OTHER BFRs

There was a massive difference in the levels of PBDEs in eggs collected in Bangun and Tropodo in May and November of 2019, respectively. Egg samples collected in November 2019 contained one order of magnitude higher levels of these brominated flame retardants. This concentration is most likely a consequence of different types of plastic waste brought to Bangun and burned in Tropodo in the autumn of 2019, of which a substantial part most likely had its origin in e-waste.

The level of PBDEs measured in the pooled eggs sample from Tropodo is the second highest ever measured in chicken eggs globally. There was a very high level of DBDPE in the same egg samples. DBDPE is a representative of the group of nBFRs, which in products replaced PBDEs and HBCD already regulated by the Stockholm Convention on POPs. It replaced mainly DecaBDE in wire coatings and polystyrene products. Although DecaBDE reached the highest concentrations in samples from Tropodo and Bangun compared to other congeners of PBDEs, other congeners forming previously banned commercial PBDE mixtures were also found in eggs in high concentrations.

Also, the levels of 844 and 538 ng g^{-1} fat in eggs from Tangerang and Bangun are among the ten highest levels of HBCD observed in eggs globally.

PFASs

The highest levels of PFASs were measured in eggs from Bangun, in both the May and November samples of eggs. The PFASs concentration in egg samples collected in November 2019 was five times higher than the level in samples collected in May 2019. These levels are comparable to those found in eggs from areas affected by industry in Europe. Levels in samples from other sites on the island of Java were not so high.

TOLERABLE DAILY INTAKE (TDI) OF SELECTED POPs

We calculated the dietary intake for PCDD/Fs, dl-PCBs, PBDD/Fs, PBDEs, and PFOS.

On average, egg consumption was calculated as half an egg (18 g of egg) per day for an adult person weighing 58 kg. A person who eats free-range eggs from the sites in this study exceeds the tolerable daily intake (TDI) set by the European Food Safety Authority (EFSA) for dioxins and dioxin-like PCBs by 1.5- to 43-fold. This situation should be considered as the most severe finding in 2019 in Tropodo, Kendalsari, and Tangerang.

The tolerable daily intake (TDI) for PCDD/Fs and dl-PCBs can be reached by eating 0.01 to 0.02 of a free-range egg in Tropodo, or one quarter of an egg in Bangun or Sumberwuluh, where contamination by dioxins and dl-PCBs is lower than in Tropodo, Tangerang, and Kendalsari. In comparison, it would be necessary to eat more than 1,350 eggs from the supermarket in Jakarta to reach the level of tolerable daily intake for dioxins and dl-PCBs.

This example shows the vast difference between background/reference contamination by PCDD/Fs and dl-PCBs and contamination at localities affected by improper handling of plastic waste, secondary aluminum smelters, or hazardous waste incineration facilities and waste-to-energy operations.

There is a significant contribution from brominated dioxins to the daily intake of dioxin-like acting chemicals in the sample of eggs from Tangerang, which reached one tenth of the total intake from PCDD/Fs and dl-PCBs in eggs.

The intake of PBDEs from the eggs sample in Tropodo (110 ng kg^{-1} bw) is almost 28-fold higher than the average total daily intake from "the food basket" calculated by the Joint Committee of WHO and FAO in 2006, at a level of 4 ng kg^{-1} bw. The daily intake of PBDEs at the studied locations in 2019 was more than ten times higher than the total daily intake of PBDEs in Finland, Sweden, or Canada more than fifteen years ago.[2]

An adult eating half an egg per day from a free-range chicken foraging in the vicinity of the Bangun dumpsite would exceed the proposed tolerable daily intake (TDI) of PFOS by 3- to 16-fold.

The eggs from Tangerang exhibited the second highest intake of PFOS among the sampled eggs from Java in this study. An adult eating one egg from a free-range chicken in Tangerang's plastic waste yard would reach the TDI limit for PFOS, but in reality, people get exposed to PFOS from a much wider range of foods and drinks than just eggs.

RECOMMENDATIONS

The mismanagement of plastic and paper waste imported by Indonesian companies in Bangun and Tangerang result in complex POPs contamination. Waste present in Bangun and Tangerang obviously includes plastics from dismantled electronics.

Secondary aluminum production seems to be a significant source of POPs contamination in Kendalsari, and the Lakardowo hazardous waste incinerator is most likely the source of dioxin pollution in the nearby village of Sumberwuluh.

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[2] In this comparison decaBDE and eight other PBDE congeners were not included in order to make it more comparable to calculations of PBDE intake done between 2001 and 2004. The highest intake of PBDEs was observed in a pooled eggs sample from Tropodo, taken in October 2019, with an extremely high level of these BFRs. It also exhibits a very high ratio of decaBDE congener intake. The second-highest intake was calculated for a sample taken in November 2019 in Bangun, again with a very high contribution of decaBDE congener.

Two case studies in this report demonstrated that ash and slag residues used for the building of roads, flood defenses, and building foundations contribute significantly to the spread of POPs pollution.

Some of the measures to address these issues include:

1. Prohibit combustion as a disposal option for plastic waste or as an example of the "circular economy," or as a measure to facilitate the illegal cases of wastes import. Burning should not be accepted as a best practice for plastic waste management and disposal.
2. Prohibit the combustion of plastics as fuel for industrial operations due to the dioxin and other halogenated pollution generated in emissions and ash.
3. Restrict the use of halogen-containing synthetic fuels derived from plastics due to the persistent organic pollutants that occur in emissions from burning such fuel.
4. Remediate sites contaminated with dioxins and other POPs to ensure that human health is protected and that no food-chain contamination occurs.
5. Update the Indonesian Stockholm Convention National Implementation Plan, including the new POPs, to evaluate the effectiveness of preventive measures and control of POPs in Indonesia.
6. Strictly apply the new provisions of the Basel Convention on the plastic waste trade to block hazardous waste imports and control the transboundary movement of plastic wastes, or enact a ban on plastic waste imports.
7. Enact a stronger international chemicals framework, Beyond 2020, that includes work to reduce and eliminate PFASs as a class.
8. Reduce and minimize plastic production and avoid the use of halogenated plastics or the addition of halogenated compounds such as bromine, chlorine, and fluorine in plastic production.
9. Implement stricter control of potential imports of e-waste or end-of-life electronics to Indonesia.
10. Set a better system for sorting e-waste and prevent the use of plastics from electronics as fuel.
11. Implement the BAT/BEP guidelines of the Stockholm Convention for secondary aluminum production.
12. Prepare and/or update an action plan that addresses the pollution sources of UPOPs to reduce the total release of these chemicals.[3]
13. Reduce the use of aluminum and do not promote it as a replacement for plastic packaging of beverages or food.
14. Avoid halogenated compounds to fuel the thermal and combustion processes.
15. Use non-combustion alternative methods for treatment of hazardous waste, e.g. for POPs waste or medical waste disposal.
16. Ash contaminated with POPs should be prevented from use as a building material, and managed in a way to avoid generating leachate and dust dispersion.
17. Introduce stricter, more protective limits for POPs in emissions and wastes, within the frameworks of both the Stockholm and Basel Conventions.
18. Prohibit the use of wastes and materials with a concentration of dioxins and dl-PCBs exceeding the level of 50 pg TEQ g^{-1} dw (0.05 ppb) on the soil surface.

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[3] "Develop an action plan or, where appropriate, a regional or subregional action plan within two years of the date of entry into force of this Convention for it, and subsequently implement it as part of its implementation plan specified in Article 7 ... " Article 5 Stockholm Convention (2010). Stockholm Convention on Persistent Organic Pollutants (POPs) as amended in 2009. Text and Annexes. Geneva: 64.

1. Introduction

Toxic contamination in developing countries can originate from both domestic and foreign sources. From the perspective of different persistent organic pollutants (POPs) and their origin, we must differentiate between those that might be mainly in products and wastes, and those unintentionally produced POPs (UPOPs) such as dioxins, dioxin-like PCBs or hexachlorobenzene. The primary sources of UPOPs are listed in Annex C to the Stockholm Convention (Stockholm Convention 2010) and more closely specified in the BAT/BEP Guidelines (Stockholm Convention on POPs 2008), and the Dioxin Toolkit (UNEP and Stockholm Convention 2013).

Some developing countries have become destinations for waste exports, including plastic waste, paper for recycling and/or electronic waste (e-waste), which may contain a whole range of POPs added intentionally to the products. These chemicals are now present in the waste chain, including brominated flame retardants (BFRs), short-chain chlorinated paraffins (SCCPs), and per- and polyfluoroalkyl substances (PFASs).

In this study, free-range chicken eggs were used to investigate POPs contamination at selected hotspots on the Indonesian island of Java. The selected hotspots included imported plastic waste dumpsites (Bangun, Tangerang), tofu factories using plastic waste as fuel (Tropodo), secondary aluminum smelters from aluminum slag and scrap (Kendalsari), and a hazardous waste incinerator facility (Lakardowo). We also included previously published data from two sites, Bangun and Tropodo, which were re-sampled for both free-range chicken eggs as well as for contaminated soil or ash residues.

A previous study based on free-range chicken-egg samples from Bangun and Tropodo was published in December 2019 (Petrlik, Ismawati et al. 2019). This report looks at a broader number and scale of sites, as well as a larger number of samples, including samples of soil, ash and other matrices. The whole range of POPs, from additives in plastic and other waste products, through to the unintentionally produced POPs, were analyzed in the samples. A brief description of the major POPs investigated in this study can be found in sub-chapters 1.2 – 1.5. Organochlorine pesticides (OCPs) and technical PCBs represented by indicator PCBs (i-PCBs) were also included in the analyses. Although they were not expected to be contained in the wastes or target processes of wastes at the selected sampling sites in any significant levels, they could be present as contaminants from other human activities at selected, mostly rural locations.Free-range chicken eggs are sensitive indicators of POPs contamination in soils or dust and represent an important human exposure pathway. As “active samplers,” they can be used to reveal POPs contamination, particularly in areas impacted by dioxins (PCDD/Fs) and PCBs as well as by DDT, or BFRs.

Major waste flows, reprocessing and disposal methods that can lead to the contamination by either intentionally added POPs contained in waste or by UPOPs created by thermal processes are described in sub-chapters 1.6-1.10.

IPEN, in cooperation with its participating organizations Arnika, Nexus3 Foundation and Ecoton, conducted chemical analyses of free-range chicken egg samples collected by their experts and/or by local groups. The investigations aim to assess the potential presence of toxic substances in the environment of villagers in East Java and Banten. The previous study evaluated initial sampling in Tropodo and Bangun, and it was the first study to examine globally regulated substances in communities affected by plastic waste imports in Southeast Asia. (Petrlik, Ismawati et al. 2019).

1.1 TOXIC ADDITIVES IN PLASTICS

Plastics and food packaging contain chemical contaminants from manufacturing, with many additives employed to create various properties of this packaging. Various additives can be used to make a product inflammable (flame retardants), more flexible (plasticizers), grease-resistant (fluorinated chemicals - known collectively as PFASs), or sterile (biocides), to mention a few examples. Many of these additives are toxic, leak from products when they are being used, can be released during recycling, and can also be released from recycled products. As noted by Hahladakis et al., "*sound recycling has to be performed in such a way as to ensure that emission of substances of high concern and contamination of recycled products is avoided, ensuring environmental and human health protection, at all times*" (Hahladakis, Velis et al. 2018).

Some phthalates used as plasticizers are toxic to reproduction (Swan 2008, Lyche, Gutleb et al. 2009), increase the risks of allergy and asthma, and can have an adverse impact on children's neurological development (Jurewicz and Hanke 2011). Many of the additives in plastics have been found to last for a long time in the environment and accumulate in animals. Some of them belong to the group of persistent organic pollutants (POPs) regulated by the Stockholm Convention (Cole, Lindeque et al. 2011, Rochman, Hoh et al. 2013). These include, for example BFRs, SCCPs and/or PFASs, which exhibit severe impacts on human health. These are described in sub-chapters 1.2–1.4.

Substances of concern that are found in plastics were well described in a report prepared for the last meeting of the Conferences of Parties to both the Basel and Stockholm Conventions (Marine Litter Topic Group 2019).

When plastics are burned as fuel, new toxic chemicals can be created. For example, burning chlorine-containing plastics such as PVC forms polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs). These highly toxic substances are commonly known as dioxins. Burning plastics containing brominated flame retardants creates brominated dioxins and furans (PBDD/Fs), a group of toxic chemicals similar to chlorinated dioxins. They are closely described in sub-chapter 1.5.4.

Some plastics additives, such as short-chain chlorinated paraffins (SCCPs), polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCD) as well as byproducts of burning these additives (PCDD/Fs, dioxin-like PCBs or hexachlorobenzene), are already regulated under the Stockholm Convention (Stockholm Convention 2010, Stockholm Convention 2017). Additionally, some chemicals used in food packaging are toxic, and some fluorinated chemicals are also regulated under the Stockholm Convention, notably perfluorooctanesulfonic acid (PFOS) and perfluorooctanic acid (PFOA), including their salts and related substances. All of these chemicals can leach from plastic and paper wastes when dumped or burned.

1.2 BROMINATED FLAME RETARDANTS (BFRs)

Brominated flame retardants such as polybrominated diphenyl ethers (PBDEs) are known as endocrine-disrupting chemicals (EDCs) and adversely impact the development of the nervous system and of children's intelligence (POP RC 2006, POP RC 2007, POP RC 2014).

The indisputable toxicity and persistency of the main representatives of brominated flame retardants, i.e. PBDEs and HBCD, resulted in governments listing them in the Stockholm Convention for global elimination. Scientists have raised serious concerns over substitutes for flame retardant chemicals, but they continue to be used without precautions or restrictions (DiGangi, Blum et al. 2010).

PBDEs are of primary interest for this study due to the fact that these hazardous chemicals were and still are used in many plastic products, including recycled plastics. PBDEs were allowed to be recycled from waste materials into new products despite of their well-known adverse environmental and human health effects. HBCD and a few substitutes for PBDEs, described as new brominated flame retardants (nBFRs), are also investigated in this study. The new flame retardants are being introduced to the market much faster than they are being evaluated, so there is an accumulating worldwide inventory of potentially problematic chemicals.

Only limited information is available on the current global market volume, but approximately 390,000 tons of brominated flame retardants were sold in 2011. This represents 19.7% of the flame retardants market (Townsend Solutions Estimate 2016).

1.2.1 POLYBROMINATED DIPHENYL ETHERS (PBDEs)

Polybrominated diphenyl ethers (PBDEs) are a group of brominated flame retardants that include substances listed in the Stockholm Convention for global elimination such as PentaBDE (2009), OctaBDE (2009) and DecaBDE (2017). PBDEs are additives mixed into plastic polymers that are not chemically bound to the material and therefore leach into the environment. They already have been identified in breast milk in Indonesia in research from more than a decade ago, and *"the levels were in the same order as those in Japan and some European countries, but were one or two orders lower than North America*" (Sudaryanto, Kajiwara et al. 2008).

PBDEs have adverse effects on reproductive health as well as developmental and neurotoxic effects (POP RC 2006, POP RC 2007, POP RC 2014). DecaBDE and/or its degradation products may also act as endocrine disruptors (POP RC 2014).

PentaBDE has been used in polyurethane foam for car and furniture upholstery, and Octa- and DecaBDE have been used mainly in plastic casings for electronics. OctaBDE formed 10%-18% of the weight (Stockholm Convention 2016) of CRT television and computer casings and other office electronics made of acrylonitrile butadiene styrene (ABS) plastic. DecaBDE forms 7%-20% of the weight (POP RC 2014) of many different plastic materials, including high-impact polystyrene (HIPS), polyvinylchloride (PVC), and polypropylene (PP) used in electronic appliances. As this study examines eggs from sites affected by the presence of plastic waste and/or by its incineration, all of the mentioned PBDEs were part of the main focus of our investigation.

1.2.2 HEXABROMOCYCLODODECANE (HBCD)

Hexabromocyclododecane (HBCD) is a brominated flame retardant primarily used in polystyrene building insulation. HBCD is an additive mixed into plastic polymers that is not chemically bound to the material and therefore may leach into the environment. HBCD is highly toxic to aquatic organisms and has negative effects on reproduction, development and behavior in mammals, including transgenerational effects (POP RC 2007a). HBCD is also found in packaging materials, video cassette recorder housings and electric equipment.

HBCD was listed in Annex A of the Stockholm Convention for global elimination with a five-year specific exemption for use in building insulation that expired for most Parties in 2019 (Stockholm Convention 2013). This chemical also belongs among the SVHC substances under the REACH legislation.

1.2.3 NOVEL BFRs (nBFRs)

Novel BFRs (nBFRs) are a group of chemicals that replaced in many cases already restricted BFRs. Different sources list different chemicals among this group, but only a few of them are measured in the environment. Recent studies also show that nBFRs are becoming widespread in the environment, including in food, particularly in some Asian countries (Shi, Zhang et al. 2016). A review of the levels of BFRs in soil concluded that: *“Although further research is required to gain baseline data on nBFRs in soil, the current state of scientific literature suggests that nBFRs pose a similar risk to land contamination as PBDEs“* (McGrath, Ball et al. 2017).

The scientific panel of the EFSA evaluated 17 “emerging“[4] and 10 “novel“[5] BFRs in 2012 and suggested that: *“There is convincing evidence that tris(2,3-dibromopropyl) phosphate (TDBPP) and dibromoneopentyl glycol (DBNPG) are genotoxic and carcinogenic, warranting further surveillance of their occurrence in the environment and in food. Based on the limited experimental data on environmental behaviour, 1,2-bis(2,4,6-tribromophenoxy)ethane (BTBPE) and hexabromobenzene (HBB) were identified as compounds that could raise a concern for bioaccumulation“* (EFSA CONTAM 2012). EFSA’s panel also stated that for most evaluated BFRs, there were not sufficient data about their presence in the environment to draw meaningful conclusions.

Decabromodiphenyl ethane (DBDPE) was introduced in the early 1990s as an alternative to DecaBDE in plastic and textile applications (Ricklund, Kierkegaard et al. 2010). It was used mainly in wire coatings and polystyrene, in both cases as replacement of DecaBDE. This widespread contaminant is a highly hydrophobic compound (with a log Kow of 11.1); (Covaci, Harrad et al. 2011). DBDPE has been identified in sewage sludge (De la Torre, Concejero et al. 2012), indoor dust (Julander, Westberg et al. 2005, Ali, Harrad et al. 2011) outdoor dust (Muenhor, Harrad et al. 2010, Anh, Tomioka et al. 2018), chicken eggs (Tlustos, Fernandes et al. 2010), honey (Mohr, García-Bermejo et al. 2014), food in general (Tlustos, Fernandes et al. 2010, Shi, Zhang et al. 2016), and in sediments and peregrine falcon eggs (Ricklund, Kierkegaard et al. 2009, Ricklund, Kierkegaard et al. 2010). A Chinese “total diet study” (TDS) for 2011 concluded that: *“The levels and estimated daily intake (EDI) of DBDPE in the present study were similar to or higher than those of legacy BFRs (i.e., PBDEs and HBCD) in the TDS 2007”* (Shi, Zhang et al. 2016).

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[4] The group of emerging BFRs included: BEH-TEBP - Bis(2-ethylhexyl) tetrabromophthalate, BTBPE - 1,2-Bis(2,4,6-tribromophenoxy)ethane, DBDPE - Decabromodiphenyl ethane, DBE-DBCH - 4-(1,2-Dibromoethyl)-1,2-dibromocyclohexane, DBHCTD - 5,6-Dibromo-1,10,11,12,13,13-hexachloro-11-tricyclo[8.2.1.02,9]tridecene, EH-TBB - 2-Ethylhexyl 2,3,4,5-tetrabromobenzoate, HBB - 1,2,3,4,5,6-Hexabromobenzene, HCTBPH - 1,2,3,4,7,7-Hexachloro-5-(2,3,4,5-tetra-bromophenyl)- bicyclo[2.2.1]hept-2-ene, OBTMPI - Octabromotrimethylphenyl indane (OBIND in this study), PBB-Acr - Pentabromobenzyl acrylate, PBEB – Pentabromoethylbenzene, PBT – Pentabromotoluene, TBNPA - Tribromoneopentyl alcohol, TDBP-TAZTO - 1,3,5-Tris(2,3-dibromopropyl)-1,3,5-triazine-2,4,6-trione, TBCO - 1,2,5,6-Tetrabromocyclooctane, TBX - 1,2,4,5-Tetrabromo-3,6-dimethylbenzene, and TDBPP Tris(2,3-dibromopropyl) phosphate.

[5] The group of novel BFRs included: BDBP-TAZTO - 1,3-Bis(2,3-dibromopropyl)-5-allyl-1,3,5-triazine-2,4,6(1H,3H,5H)-trione, DBNPG - Dibromoneopentyl glycol, DBP-TAZTO - 1-(2,3-Dibromopropyl)-3,5-diallyl-1,3,5-triazine-2,4,6(1H,3H,5H)-trione, DBS – Dibromostyrene, EBTEBPI - N,N’-Ethylenebis(tetrabromophthalimide), HBCYD – Hexabromocyclododecane (HBCD or HBCDD are more of the used abbreviations for this chemical, listed already in Annex A to Stockholm Convention), HEEHP-TEBP - 2-(2-Hydroxyethoxy)ethyl 2-hydroxypropyl 3,4,5,6-tetrabromophthalate, 4’-PeBPO-BDE208 - Tetradecabromo-1,4-diphenoxybenzene, TTBNPP - Tris(tribromoneopentyl) phosphate, and TTBP-TAZ - Tris(2,4,6-tribromophenoxy)-s-triazine.

BTBPE was first produced in the 1970s and is used as a replacement for OctaBDEs (Hoh, Zhu et al. 2005). It has been identified in various abiotic media (dust, atmosphere, sediment, water) and biotic media (zooplankton, mussel, fish, aquatic bird eggs, honey, chicken eggs or food in general) (Hoh, Zhu et al. 2005, Julander, Westberg et al. 2005, Ali, Harrad et al. 2011, Wu, Guan et al. 2011, Mohr, García-Bermejo et al. 2014, Poma, Volta et al. 2014, Petrlik 2016, Petrlik, Kalmykov et al. 2017, Anh, Tomioka et al. 2018).

This compound has the ability to bioaccumulate and to biomagnify in aquatic food webs (Law, Halldorson et al. 2006, Wu, Guan et al. 2011). Similar to DecaBDE, the commercial mixture of BTBPE was found to contain brominated dioxins (PBDD/Fs) and/or to support their formation during treatment of ABS plastic (Tlustos, Fernandes et al. 2010, Ren, Zeng et al. 2017, Zhan, Zhang et al. 2019). BTBPE has been measured in increased concentrations in Indonesia during passive air sampling conducted in 2005-06 (Lee, Sverko et al. 2016).

HBB has commonly been used for the manufacture of paper, woods, textiles, plastics and electronic goods (Yamaguchi, Kawano et al. 1988, Watanabe and Sakai 2003) and it is *"likely widely distributed, as verified both by chemical analysis and estimated properties"* (Arp, Møskeland et al. 2011). Thermal degradation of the DecaBDE technical mixture and polymeric PBDEs pyrolysis could also be sources of the HBB found in the environment (Thoma and Hutzinger 1987, Gouteux, Alaee et al. 2008).

The laboratory at the Department of Food Chemistry and Analysis of the University of Chemistry and Technology, Prague, routinely measures six nBFRs in environmental samples, including the egg samples for this study: 1,2-bis(2,4,6-tribromophenoxy) ethane (BTBPE), decabromodiphenyl ethane (DBDPE), hexabromobenzene (HBB), octabromo-1,3,3-trimethylpheny-1-indane (OBIND), 2,3,4,5,6-pentabromoethylbenzene (PBEB), and pentabromotoluene (PBT).

Out of this group, BTBPE, DBDPE and HBB are monitored more often in environmental samples (Munschy, Héas-Moisan et al. 2011, Mohr, García-Bermejo et al. 2014, Poma, Volta et al. 2014, Vorkamp, Bossi et al. 2015). Along with OBIND, they also were found in increased levels most frequently out of the group of six nBFRs in samples from Indonesia measured in this study.

1.3 SHORT-CHAIN CHLORINATED PARAFFINS (SCCPs)

Short-chain chlorinated paraffins (SCCPs) are a group of POPs added by governments to the Stockholm Convention for global elimination in 2017. SCCPs are toxic to aquatic organisms at low levels, disrupt endocrine function and are suspected to cause cancer in humans (POP RC 2015). SCCPs are other additives in plastics that might also be expected in waste imported to Java. A 2017 study of 60 plastic children's products from 10 countries found SCCPs in 45% of them (Miller and DiGangi 2017, Miller, DiGangi et al. 2017)

1.4 PER- AND POLYFLUOROALKYL SUBSTANCES (PFASs)

Per- and polyfluoroalkyl substances (PFASs) are a large class (OECD 2018) of more than 4,500 very persistent fluorinated chemicals (including PFOS) that have been widely used in packaging, textiles and plastics. Scientists are concerned with their widespread presence in the environment, and in the Madrid Statement said that they: *"call on the international community to cooperate in limiting the production and use of PFASs and in developing safer nonfluorinated alternatives"* (Blum, Balan et al. 2015). Later, in the Zurich Statement, they called upon regulators to address PFASs in chemically-related groups rather than as individual substances (Ritscher, Wang et al. 2018).

In animal studies, some long-chain PFASs have been found to cause liver toxicity, disruption of lipid metabolism and of the immune and endocrine systems, adverse neurobehavioral effects, neonatal toxicity and death, and tumors in multiple organ systems (Lau, Anitole et al. 2007, Post, Cohn et al. 2012). More health effects are summarized in the Madrid and Zurich statements as well as in the toxicological profiles of PFASs (Blum, Balan et al. 2015, ATSDR 2018, Ritscher, Wang et al. 2018, Fenton 2019).

The EFSA has sharply lowered the permitted intake of PFOS from 150 ng/kg body weight/day to 13 ng/kg body weight/week (EFSA CONTAM 2018b). An investigation of PFASs substances in Indonesia found that they are unregulated and contaminate both coastal sediments and breast milk (BaliFokus/Nexus3 Foundation 2019).

Electrochemical fluorination (ECF) and telomerisation are the two major methods employed to produce PFASs. The manufacturing process of PFASs can help us understand the differences in the presence of their isomers in the environment, and their links to potential sources of contamination. *"The branched isomers of PFASs are mainly manufactured in the ECF method, which has historically been used to produce the major part of the two dominant PFASs, PFOS and PFOA. ECF gives rise to complex mixtures of linear and branched compounds. PFOA produced by this method has typically had an isomer composition of 78% linear (n-PFOA) and 22% branched isomers (br-PFOA). ECF-PFOS shows a distribution of around 70% linear (n-PFOS) and 30% branched (br-PFOS). … the telomerisation process keeps the structure of the starting telogen and a pure linear or isopropyl form is produced".(Benskin, De Silva et al. 2010, Jiang, Zhang et al. 2015)*" (van Hees 2016).

1.4.1 PFOS

Perfluorooctanesulfonic acid (PFOS) and its salts were listed in the Stockholm Convention in 2009 along with perfluorooctane sulfonyl fluoride (PFOSF). The Stockholm Convention expert committee concluded that, "*PFOS is extremely persistent. It does not hydrolyse, photolyze or biodegrade in any environmental condition tested*"(POP RC 2006a). In animal studies, PFOS has been shown to cause cancer, neonatal mortality, delays in physical development, and endocrine disruption (Thomford 2002a, Thomford 2002b, Luebker, York et al. 2005, Jacquet, Maire et al. 2012, Du, Hu et al. 2013). PFOS-related substances have been used in the packaging and paper industries in both food packaging and commercial applications to impart grease, oil and water resistance to paper, paperboard and packaging substrates (KemI 2004).

1.4.2 PFOA

Perfluorooctanic acid (PFOA) is another common member of the PFASs family of substances. Governments added PFOA, its salts and PFOA-related substances to the Stockholm Convention for global elimination in 2019. PFOA and related substances have a large variety of uses, e.g. in the manufacture of many fluoropolymers, for the semiconductor industry, and in firefighting foams, ski waxes, paper packaging for microwave popcorn, and baking papers (POP RC 2016).

Higher maternal levels of PFOS and PFOA are associated with delayed pregnancy, reduced human semen quality and penis size (Fei, McLaughlin et al. 2009, Joensen, Bossi et al. 2009, Di Nisio, Sabovic et al. 2018). In humans, PFOA is associated with high cholesterol, ulcerative colitis, thyroid disease, testicular cancer, kidney cancer, pregnancy-induced hypertension, and immune system effects. IIt is also transferred to the fetus through the placenta and to infants via breast milk (POP RC 2016).

1.4.3 PFHxS

Perfluorohexane sulfonate (PFHxS), with its salts and PFHxS-related substances, is another group of PFASs suggested to be listed in the Stockholm Convention by decision of the POPs Review Committee (POP RC 2019). PFHxS was commonly used as a surfactant (foam formation for reduction of fuel fires) and a surface protector (in metal plating processes, consumer products such as carpets, textiles, and in the leather industry). It belongs to one of the most persistent compounds in the environment. The estimated serum elimination half-life of PFHxS in humans is higher than other PFASs, with an average of 8.5 years (range 2.2–27 years) (POP RC 2019).

The most common exposure pathways for humans are mainly through intake of food and drinking water, but exposure also comes from indoor dust inhalation or from consumer products containing PFHxS or its precursors (POP RC 2019). PFHxS can trigger hypersensitivity and suppression of the immune system (asthma, allergic reactions), changes of lipids and protein metabolism pathways, changes in liver and thyroid functioning, and impacts on the reproductive system (Ali, Roberts et al. 2019, POP RC 2019).

1.4.4 OTHER PFASs

There is a whole range of other PFASs that could be present in wastes imported or locally produced in Indonesia. Samples from Indonesia were analyzed in the laboratory at the University of Chemistry and Technology in Prague, Department of Food Chemistry and Analysis, for 17 PFASs, both individual substances and/or their groups.[6] We have included a more detailed description for another two PFASs which have been found to be more frequently present in samples from Indonesia. We also refer to other scientific references on PFASs as a group, or regarding other ones not described here (Blum, Balan et al. 2015, ATSDR 2018, Ritscher, Wang et al. 2018, Fenton 2019).

1.4.4.1 PFDA

Perfluorodecane acid (PFDA) is a long-chain (C10) alkyl fatty acid. It is a persistent, nondegradable compound. PFDA releases into the environment due to the breakdown of products like stain- and grease-proof coatings on food packaging, couches and carpets (Ghisi, Vamerali et al. 2019). Scientific studies have established the occurrence of PFDA in human blood (Lu, Shi et al. 2014), urine and breast milk (So, Yamashita et al. 2006, Tao, Ma et al. 2008), and in infant formula (Tao, Ma et al. 2008).

The toxicity of PFDA is related to hepatotoxicity, immunosuppression, changes in lipid metabolism pathways, increases in oxidative stress (Xu, Zhang et al. 2019), and influence on thyroid hormone levels and thyroid functioning (Vanden Heuvel 1996).

1.4.4.2 PFDoA

Perfluorododecanoic acid (PFDoA) is a ubiquitous environmental contaminant that is widely spread in water, soil, wildlife and human tissue. The molecule of PFDoA has a long carbon chain (C12) that enables this chemical to persist longer in the environment and in living organisms. The most significant source of PFDoA in the environment is water from the textile industry, where PFDoA is a component of dyes and surfactants (Ayanda, Yang et al. 2018).

The mechanism and toxicity of effects of PFDoA on organisms is not well studied yet. The results of one study on fish indicated that *“sub-chronic exposure of PFDoA caused DNA damage with a simultaneous induction of different erythrocyte abnormalities“* (Ayanda, Yang et al. 2018). The presence of PFDoA causes hepatotoxicity, neurotoxic effects, problems with lipid metabolism pathways, steroidogenesis, and reproductive system impacts (Shi, Feng et al. 2010, Long, Ghisari et al. 2013).

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[6] List of 17 PFASs included in the analysis: PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUdA, PFDoA, PFTrDA, PFTeDA, PFBS, PFHxS, br-PFOS, L-PFOS, PFDS, PFOSA

1.5 DIOXINS (PCDD/Fs) AND OTHER UNINTENTIONALLY PRODUCED POPs

Annex C of the Stockholm Convention lists seven unintentionally produced POPs: HCB, hexachlorobutadiene (HCBD), pentachlorobenzene (PeCB), PCBs, polychlorinated dibenzo-p-dioxins (PCDD), polychlorinated dibenzofurans (PCDF), and polychlorinated naphthalenes. Analyses of eggs in this study covered HCB, HCBD, PeCB, PCBs, and PCDD/Fs. Polychlorinated naphthalenes were not analyzed.

1.5.1 PCDD/Fs and dl-PCBs

Dioxins belong to a group of 75 polychlorinated dibenzo-p-dioxin (PCDD) congeners and 135 polychlorinated dibenzofuran (PCDF) congeners, of which 17 are of toxicological concern. Polychlorinated biphenyls (PCBs) are a group of 209 different congeners that can be divided into two groups according to their toxicological properties: 12 congeners exhibit toxicological properties similar to dioxins and often referred to as "dioxin-like PCBs" (dl-PCBs). The other PCBs do not exhibit dioxin-like toxicity but have a different toxicological profile and are referred to as "non dioxin-like PCBs" (ndl-PCBs) (European Commission 2011). Technical mixtures of PCBs are characterized by six, sometimes seven indicator PCB congeners (i-PCBs). Levels of PCDD/Fs and dl-PCBs are expressed in total WHO-TEQ, calculated according to toxic equivalency factors (TEFs) set by a WHO expert panel in 2005 (van den Berg, Birnbaum et al. 2006). These WHO TEFs were used to evaluate dioxin-like toxicity in the pooled samples of chicken eggs, soils, ash and other samples from Indonesia in this study.

Chlorinated dioxins (PCDD/Fs) are known to be extremely toxic. Numerous epidemiologic studies have revealed a variety of human health effects linked to chlorinated dioxin exposure including cardiovascular disease, diabetes, cancer, porphyria, endometriosis, early menopause, alteration of testosterone and thyroid hormones, and altered immune system response among others (White and Birnbaum 2009, Schecter 2012). Laboratory animals given dioxins suffered a variety of effects, including an increase in birth defects and stillbirths. Fish exposed to these substances died shortly after the exposure ended. Food (particularly from animals) is the major source of exposure for humans (BRS 2017).

Chlorinated dioxins became known to the public in the 1970s as a result of their contamination of Agent Orange, a defoliant pesticide mixture sprayed by the U.S. during the Vietnam War.[7] The production of 2,4,5 T pesticide as a basic ingredient for Agent Orange left one of the most seriously contaminated sites in Europe (Zemek and Kocan 1991, Kubal, Fairweather et al. 2004, Weber, Gaus et al. 2008) and sick workers with many symptoms of exposure to the most toxic of dioxin congeners 2,3,7,8-TCDD (Pelclová, Urban et al. 2006, Bencko and Foong 2013).

1.5.2 PeCB and HCB

Pentachlorobenzene (PeCB) and hexachlorobenzene (HCB) are primarily produced unintentionally during combustion, as well as during thermal and industrial processes. They also occur as a byproduct during the production of chlorinated hydrocarbons such as perchloroethylene, trichloroethylene, carbon tetrachloride or pesticides. In the past, they were produced intentionally as pesticides or technical substances. Perchloroethylene is widely used in drycleaning, and trichloroethylene and carbon tetrachloride have been used extensively as degreasing agents and as solvents for other chlorine-contain-

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[7] According to estimates provided by the Government of Vietnam, 400,000 people were killed or maimed by the pesticide; 500,000 children were born with birth defects ranging from retardation to spina bifida; and an additional 2 million people have suffered cancers or other illnesses, which also can be related to dioxins as impurities in the Agent Orange mixture. It is estimated that in total, the equivalent of at least 366 kilograms of pure dioxin were dropped. York, G. and H. Mick. (2008, April 27, 2018). "Last ghost' of the Vietnam War." Retrieved 19-11-2018, 2018, from https://www.theglobeandmail.com/incoming/last-ghost-of-the-vietnam-war/article1057457/?page=all.

ing compounds. PeCB was used as a component in PCB products, in dyestuff carriers, as a fungicide, as a flame retardant and as a chemical intermediate for the production of the pesticide quintozene (POP RC 2008).

In high doses, HCB is lethal to some animals and, at lower levels, adversely affects their reproductive success. Researchers also found out that HCB, similar to other organochlorinated compounds, has a transplacental transfer (Sala, Ribas-Fitó et al. 2001). HCB has been found in food of all types (BRS 2017).

Although globally, the consumption of HCB-contaminated food is the primary source of HCB exposure, other potential exposure pathways include the inhalation of HCB-contaminated air, skin contact, in utero exposure and from breast milk (Reed, Büchner et al. 2007). The study also found that in addition to cancer, the human health effects associated with HCB exposure encompass systemic impairment (thyroid, liver, bone, skin), damage to the kidneys and blood cells, as well as the immune and endocrine systems. It also causes a teratogenic effect, and impairs nervous systems.

PeCB is very toxic to aquatic organisms and may cause long-term adverse effects in the aquatic environment (POP RC 2007b).

1.5.3 HCBD

Hexachlorobutadiene (HCBD) occurs as a byproduct during the production of the same chlorinated hydrocarbons as PeCB and HCB, as a part of so-called “hexa-residues.” It is also formed unintentionally during incineration processes of such substances as acetylene and chlorine residues. HCBD is very toxic to aquatic organisms, and has been shown to cause kidney damage and cancer in animal studies as well as chromosomal aberrations in occupationally exposed humans (Pohl, McClure et al. 2001, POP RC 2012, Balmer, Hung et al. 2019). Systemic toxicity following exposure via oral, inhalation, and dermal routes may include fatty liver degeneration, epithelial necrotizing nephritis, potentially causing chronic inflammation, central nervous system depression and cyanosis (BRS 2017, Balmer, Hung et al. 2019).

1.5.4 PBDD/Fs

There are also other unintentionally produced POPs that are not yet listed in the Stockholm Convention. With the broad use of brominated flame retardants, the question has arisen about the presence of polybrominated dibenzo-p-dioxins and dibenzofurans (PBDD/Fs)[8] in the food chain, as they are found in different environmental compartments (Kannan, Liao et al. 2012). The WHO expert panel has concluded that polybrominated dibenzo-p-dioxins (PBDDs), dibenzofurans (PBDFs) and some dioxin-like polybrominated biphenyls (dl-PBBs) may contribute significantly to daily human background exposure to the total dioxin toxic equivalencies (TEQs) (van den Berg, Denison et al. 2013).

PBDD/Fs are the most relevant groups of unintentionally produced POPs in the sampled sites with e-waste and/or plastic waste which may contain brominated flame retardants, like those in Agbogbloshie, Ghana, and Samut Sakhon, Thailand, respectively (Teebthaisong, Petrlík et al. 2018, Hogarh, Petrlik et al. 2019). The same applies to plastic waste yards in Tangerang or Bangun, which is why PBDD/Fs were also analyzed in the selected samples from the sites in Indonesia in this study.

PBDD/Fs have been known to be potential byproducts of commercial PBDE mixtures since 1986 (Buser 1986). They were also found to be byproducts of some novel BFRs like DBDPE (Brenner and Knies

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[8] The synonym ”brominated dioxins“ is used for this group of chemicals as well, while ”dioxins“ applies for PCDD/Fs. We use both these shorter synonyms in this report.

1990) or BTBPE (Ren, Zeng et al. 2017, Zhan, Zhang et al. 2019). This is similar to the chlorinated dioxins that have been observed as impurities in PCBs and other chlorinated chemicals. PBDFs have also been found to be formed by sunlight exposure during normal use, as well as during disposal/recycling processes of flame-retarded consumer products (Kajiwara, Noma et al. 2008). Some studies found PBDD/Fs in copper metal recycling (Mei, Guorui et al. 2015), in the air around a waste incinerator plant (Gao, Zhang et al. 2014), around an open burning site (Gullett, Wyrzykowska et al. 2010), and, recently, in children's toys (Budin, Petrlik et al. 2020). PBDD/Fs are similar to the PCDD/Fs, however, they have been studied less extensively than their chlorinated analogues.

PBDD/Fs have been found to exhibit similar toxicity and health effects as their chlorinated analogues (PCDD/Fs) (Mason, Denomme et al. 1987, Behnisch, Hosoe et al. 2003, Birnbaum, Staskal et al. 2003, Kannan, Liao et al. 2012, Piskorska-Pliszczyńska and Maszewski 2014). They can, for example, affect brain development, damage the immune system and fetus, or induce carcinogenesis (Kannan, Liao et al. 2012).

"Both groups of compounds show similar effects, such as induction of aryl hydrocarbon hydroxylase (AHH)/EROD activity, and toxicity, such as induction of wasting syndrome, thymic atrophy, and liver toxicity" (Behnisch, Hosoe et al. 2003).

In general, brominated dioxins are less regulated than chlorinated dioxins. For example, PBDD/Fs are not currently listed under the Stockholm Convention (Stockholm Convention 2010), although there is clear evidence that they contain very similar properties to PCDD/Fs, which have been listed in Annex C of the Convention since its origin in 2001. In 2010, the Stockholm Convention POPs Review Committee recommended further assessment of PBDD/Fs including, *"releases from smelters and other thermal recovery technologies, including secondary metal industries, cement kilns and feedstock recycling technologies"* (POP RC 2010).

Because brominated dioxins are almost entirely unregulated substances, there is less data available on their presence in the environment. There is also very little information about their presence in food and/or consumer products, and where they can have direct impacts on human health, including in vulnerable groups such as children and women of childbearing age. This applies in particular to developing countries like Indonesia.

1.6 WASTE EXPORTS FROM DEVELOPED COUNTRIES TO INDONESIA

A World Bank study identified that recycling in Indonesia accounts for about 15% of the total waste, and it is mostly undertaken by the informal sector, with formal recycling systems capturing less than 5% of wastes generated by the population (Shuker and Cadman 2018). The study also shows that plastic represents a significant proportion of debris extracted from waterways in all towns, ranging from 20% to 38%.

ASEAN countries, including Indonesia, imported recyclables from various countries to be sorted and combined with domestically collected plastic scrap, and then exported it to China. At this time, these countries imported around 3% of the global trade of plastic wastes (2011 data) and exported around 5% of the worldwide business. The discrepancy in those numbers was due to the significant role of intermediate processing or re-exporting (Velis 2014).

Current global recycling rates show that only 10% of all plastic has been recycled more than once (Geyer, Jambeck et al. 2017). Annually, Indonesia generates ~9.5 million tons of plastic waste, or about 15% of the total national waste generated. According to a recent report issued by the Ministry of Environment and Forestry of Indonesia, about 69% of Indonesia's municipal wastes were landfilled, 7% of waste were recycled and 24% unmanaged (KLHK 2020).

After China closed the door to plastic waste imports, Indonesia's plastic waste import volume doubled to 320,000 metric tons in 2018 compared to 2017. Based on observations by Ecoton and Nexus3, between 25% and 50% of plastic wastes imported by Indonesian plastic and paper recycling companies were mismanaged (Ismawati Drwiega, Septiono et al. 2019). Paper scrap imported by paper companies in East Java was found to be mixed with plastic scrap and therefore donated or dumped in several villages, including Bangun and Tropodo (GAIA 2019).

1.6.1 OPEN BURNING OF PLASTIC WASTE

Open burning of remaining plastic waste unsuitable for reuse or recycling and residue of sorting at plastic waste yards (rural dumpsites), is a very common practice. For local communities, it is also the way they prepare space for delivery of new trucks bringing them the waste for sorting.

The Stockholm Convention has identified open burning of waste, including burning of landfill sites and smoldering of copper cables, as a source category where POPs such as dioxins, PCBs, hexachlorobenzene and pentachlorobenzene *"may also be unintentionally formed and released from"* (Stockholm Convention 2010). Open burning also mostly cannot destroy POPs' additives present in the burned material, but it can contribute to further spreading of these chemicals as they bind to the soot and dust, and are simply evaporating from the burned material/waste.

Recent research focused on open burning concluded that *"Anthropogenic sources produce several times or even several orders of magnitude higher emission factors than biomass sources; the main reasons are that more PICs[9] are formed, and more metals and halogens are present. The sequence of importance of those three factors is probably system- and case-dependent,"* and the same research also concluded that *"Smoldering combustion is more proficient in generating dioxins than flaming combustion; main reason could be their longer timescale"* (Zhang, Buekens et al. 2017). The last conclusion might apply to the sites where e-waste, also including wires, could be involved in open burning of wastes in Indonesia.

Other research focused on open burning has underlined the important role that PVC plays in dioxin formation: *"Burn tests of wastes with no added PVC had low emissions of dioxin, even though the wastes presumably contained small levels of chlorine from other sources"* (Neurath 2003).

1.6.2 WASTE TO FUEL OPERATION(S)

Plastic is a mostly flammable material often used as fuel either in some kind of waste incineration operation, when it replaces fuels like wood and coal, or in more sophisticated technologies producing liquid fuel or gas made of plastic wastes (Rollinson and Oladejo 2019, Rollinson and Oladejo 2020). Both scenarios lead to the formation of a whole range of chemicals, as a result of combustion, pyrolysis or depolymerization processes. When chlorine and bromine are present in the burned wastes, the groups of released chemicals can also be PCDD/Fs and PBDD/Fs respectively.

Plastic waste replaced wood for fuel in tofu factories in Tropodo for economic reasons. We consider that process as a kind of "waste-to-fuel" operation, comparable to waste incineration. The Stockholm Convention has identified waste incineration as a sector *"for comparatively high formation and release"* of POPs such as PCDD/Fs,

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[9] PICs = products of incomplete combustion

PCBs, hexachlorobenzene and pentachlorobenzene (Stockholm Convention 2010).

As an open burning activity, the use of waste as fuel in tofu factories is not able to destroy POPs additives present in the burned plastic, instead it contributes to further spreading these chemicals as they bind to the soot and dust. Incineration of waste as fuel has been found to generate higher amounts of dioxins in comparison with open burning (Neurath 2003). Further, the dioxin concentration also depends on the composition of the burned wastes (Costner 2005, UNEP and Stockholm Convention 2013).

1.7 HAZARDOUS AND MEDICAL WASTE INCINERATION

Medical waste incineration is among the major dioxin sources, primarily due to the combustion of PVC plastic, which is a dominant source of organically bound chlorine (Thornton, McCally et al. 1996). The health sector is also a source of mercury pollution due to improper disposal of mercury-containing thermometers and sphygmomanometers.

The Stockholm Convention Guidelines on Best Available Techniques and Guidance on Best Environmental Practices note concerns over small hospital incinerators. The guidelines say: *"Due to the poor design, operation, equipment and monitoring of many existing small hospital incinerators these installations cannot be regarded as employing best available techniques"* (Stockholm Convention on POPs 2008). In developing countries, medical waste is often not segregated by type, and polluting open pit and single chamber incinerators are common. Successful implementation of medical waste management and non-combustion techniques has been demonstrated in developing countries (Stringer, Kiama et al. 2010, UNDP GEF Global Healthcare Waste Project 2010, GEF 2012, UNDP 2015).

The Stockholm Convention has identified waste incineration as a sector *"for comparatively high formation and release"* of persistent organic pollutants such as dioxins, furans, PCBs, hexachlorobenzene and pentachlorobenzene (Stockholm Convention 2010).

1.8 ELECTRONIC WASTE

Global estimates of annual waste of electrical and electronic equipment (in short e-waste production) exceed 40 million tons, with an annual growth rate of 4%-5% (Fobil, Basu et al. 2018). The export of electronic waste from developed countries to developing countries (BAN 2019), under the guise of "recycling", "repair" and/or "reuse" has effectively become a form of hazardous waste dumping that international agreements such as the Basel Convention or Stockholm Convention were created to prevent.

A recent report by the Basel Action Network confirmed that Southeast Asian countries are on the list of destinations for old, used electronic devices from developed countries, devices that contain high levels of PBDEs in the plastic casings and wire insulation (Lee, Offenhuber et al. 2018, BAN 2019). Indonesia is one of the suspected destinations for e-waste exports from developed countries (WorldLoop 2013), and such imports to Indonesia have been identified in past years (Agustina 2007, Yoshida, Terazono et al. 2016, Petridis, Petridis et al. 2020).

Also, the locally produced e-waste stockpiles increase every year. A recent study concluded that: *"It should not be a problem if all of this waste is being collected and recycled properly. Statistics show that only around 20% of the e-waste generated in the world was recycled properly. The situation is even worse in the developing countries, where the population has not yet covered by e-waste legislation, such as Indonesia. The lack of reliable e-waste data is the main reason, as no statistics are available to show that e-waste in Indonesia is grow-*

ing rapidly and will cause problems in the future. … The results show that the average growth rate of e-waste in Indonesia is 14.91% annually. The total amount of electronic waste generated in Indonesia is estimated to reach ±49,627,917 units (±487,416 ton) by 2028“ (Santoso, Zagloel et al. 2019).

As plastics from e-waste were present at some of the sites, we also analyzed free-range chicken eggs, soil, dust and ash for chemicals known to be additives in electronics.

Electronic waste is known to contain short-chain chlorinated paraffins (SCCPs) and flame-retardant chemicals listed in the treaty.

1.9 ALUMINUM DROSS RECYCLING

Large-scale aluminum production plants generate waste or aluminum dross that still contains 20%-45% aluminum in the form of residue. Aluminum dross is either a metallic lump or grey particulate flakes containing various metals and chemical compounds that have high potential to pollute the environment. It is necessary to apply the precautionary principle in the use of dross and its disposal.

During the dross smelting process, aluminum will react with the air to form an aluminum oxide on the soft surface. All aluminum smelter facilities in Jombang use wood to create a strong fire to melt the dross.

Aluminum dross melting requires additive materials and flux to bind the remaining aluminum inside the primary aluminum ash, as well as sulphuric acid and ammonia acid solutions to produce secondary ash or blackish ash and slag waste.

The use of chloride and fluoride salt fluxes in the dross melting process can reduce the formation of aluminum oxide, thereby increasing the yield to produce aluminum ingots, but also producing toxic secondary ash. Ash and slag wastes piled up on community lands produce toxic gases such as ammonia gas, methane gas, and hydrogen sulphide gas when the ash is soaked in water or during the rainy season.

The aluminum dross melting process uses KCl and NaCl as salt additives, reaching as much as 8% of the total composition, followed by washing with sulphuric acid, followed by ammonia acid. After the aluminum is separated and removed, the remaining dross, which is mixed with flux and acid solution, becomes slag waste that has the potential to pollute the soil, groundwater and surface water.

The Stockholm Convention has identified secondary aluminum production as a sector *“for comparatively high formation and release”* ” of UPOPs (Stockholm Convention 2010). Secondary aluminum production is considered to be larger source of dioxin releases in comparison with primary aluminum production (Environment 2004, Schlesinger 2007), which can be possibly explained by the use of chlorinated compounds like KCl or NaCl in the process as described above. They might serve as the donor of chlorine needed for dioxin formation (UNEP and Stockholm Convention 2013).[10] Aluminum is among the metals known to catalyze PCDD/Fs formation (Stockholm Convention on POPs 2008) so the combination of chlorinated substances and aluminum leads to a large formation of dioxins.

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[10] *“PCDD/PCDF formation is influenced by many factors. However, when chlorine is not present, PCDD/PCDF formation cannot occur; when chlorine is present, even as a trace element, PCDD/PCDF formation may occur.“* UNEP and Stockholm Convention (2013). Toolkit for Identification and Quantification of Releases of Dioxins, Furans and Other Unintentional POPs under Article 5 of the Stockholm Convention. Geneva, United Nations Environment Programme & Stockholm Convention Secretariat: 445.

1.10 USE OF ASH RESIDUES FROM COMBUSTION PROCESSES

The combustion processes in both Tropodo (plastic combustion) and Jombang regency (secondary aluminum smelters) produce large volumes of residual ash or slags as waste, which is then packed in big bags and used as construction material for paving roads and sidewalks, building defenses before floods, or in embankments along rivers and water channels. This situation is similar to several other ones documented in different locations around the world. Setting limits for dioxins in waste too loosely leads to uncontrolled handling of residual ashes from different combustion sources (Petrlik and Bell 2017).

In other studies, UPOPs were mainly observed in ash residues in significant levels (Mininni, Sbrilli et al. 2004, BiPRO 2005, Petrlik and Ryder 2005, Mach 2017, Petrlik and Bell 2017a, Peng, Weber et al. 2020). However, there are also cases when other intentionally produced POPs entered combustion processes that were not obviously able to destroy them as required in Article 6 of the Stockholm Convention for POPs waste (Stockholm Convention 2010). They ended up, for example, in waste incineration residues (Petrlik 2006, Wang, Hsi et al. 2010). UPOPs have also been measured in ash residues from different combustion processes, not only waste incineration (Umlauf, Bouwman et al. 2017, Nguyen, Nguyen et al. 2018, Wu, Zheng et al. 2018). Currently, no general consensus appears to exist regarding residue disposal and use solutions on a worldwide level, although the BAT/BEP Guidelines of the Stockholm Convention contain advice on how to avoid POPs releases due to improper handling of Air Pollution Control (APC) residues.

Formation of dioxins in combustion processes is specified in the BAT/BEP Guidelines of the Stockholm Convention as follows: *"There are two main pathways by which these compounds can be synthesized: from precursors such as chlorinated phenols or de novo from carbonaceous structures in fly ash, activated carbon, soot or smaller molecule products of incomplete combustion. Under conditions of poor combustion, PCDD/PCDF can be formed in the burning process itself"* (Stockholm Convention on POPs 2008).

Variables known to impact the thermal formation of PCDD/Fs according these guidlelines include:

- *"Technology: PCDD/PCDF formation can occur either in poor combustion or in poorly managed post-combustion chambers and air pollution control devices. Combustion techniques vary from the very simple and very poor, such as open burning, to the very complex and greatly improved, such as incineration using best available techniques;*
- *Temperature: PCDD/PCDF formation in the post-combustion zone or air pollution control devices has been reported to range between 200 °C and 650 °C; the range of greatest formation is generally agreed to be 200–450 °C, with a maximum of about 300 °C;*
- *Metals: Copper, iron, zinc, aluminum, chromium and manganese are known to catalyze PCDD/PCDF formation, chlorination and dechlorination;*
- *Sulphur and nitrogen: Sulphur and some nitrogen-containing chemicals inhibit the formation of PCDD/PCDF but may give rise to other unintended products;*
- *Chlorine must be present in organic, inorganic or elemental form. Its presence in fly ash or in the elemental form in the gas phase may be especially important;*
- *PCB are also precursors for the formation of PCDF.*

Research has shown that other variables and combinations of conditions are also important." (Stockholm Convention on POPs 2008).

Both processes produce ash and slag residues used as construction materials in Tropodo (plastic waste used as fuel) and Jombang Regency (secondary aluminum smelters) that meet the conditions needed for formation of PCDD/Fs. Some part of them also ends up in the ash residues.

Therefore, we have also included sampling and analyses of ashes in our research and compared the results with previously mapped similar cases when ash utilization or application led to food chain contamination by UPOPs, and dioxins in particular (Pless-Mulloli, Schilling et al. 2001, Katima, Bell et al. 2018).

2. Sampling and analytical methods

The samples of free-range chicken eggs, ash, soil, dust and rice crop discussed in this report were collected during three sampling periods: 1) in April 2018, the first sampling in Kendalsari and Sidokampir; 2) in May 2019, the first sampling of free-range chicken eggs in Bangun and Tropodo; 3) and in October/November 2019, the sampling of free-range chicken eggs, soils, dust and ash in Bangun, Tropodo, Kendalsari, Lakardowo and at the reference sites was done. The analyses were conducted in European laboratories between June 2018 and March 2020 closely following the three sampling campaigns.

Nine pooled samples of free-range chicken eggs were collected at seven hotspots at the island of Java. As performed in other studies, a sample of eggs purchased in a supermarket (in Jakarta) served as a background sample, as it was from not free-range hens (DiGangi and Petrlik 2005). We also used data obtained from analysis of eggs from a supermarket in Bangkok as an additional control sample from Southeast Asia included in the previous studies (Petrlík, Dvorská et al. 2018, Petrlik, Ismawati et al. 2019). Seven sites were expected to be contaminated by POPs and particularly by the unintentionally produced ones to a certain level. A basic description of these seven sites in six localities as well as reference sites can be found later in this report (see chapter 3).

Pooled samples of more individual egg samples were collected at each of the selected sampling sites in order to obtain more representative samples. Table 1 summarizes the basic data about the size of samples and the measured levels of fat content in each of the pooled samples. Table 1 also shows in which months of the years 2018 and 2019 sampling occurred.

The sampling method for soil, ash and other environmental matrices is specified in subchapter 4.3.

Free-range chicken eggs from the four pooled samples (two samples from Bangun, one sample from Tropodo and one sample from Tangerang) and one pooled sample of commercial eggs (non free-range) from Jakarta were analyzed for polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs)[11] as well as for dioxin-like polychlorinated biphenyls (dl-PCBs) using the DR CALUX® method. These were sent to a Dutch ISO 17025 certified laboratory (BioDetection Systems B.V., Amsterdam) performing the cell-based screening analysis DR CALUX® according to the European Standard EC/644/2017. The procedure for the BDS DR CALUX® bioassay has previously been described in detail (Besselink H 2004). Briefly, rat liver H4IIE cells stably transfected with an AhR-controlled luciferase reporter gene construct were cultured in an α-MEM culture medium supplemented with 10%

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[11] The synonym ”dioxins“ is used for this group of chemicals as well, while ”brominated dioxins“ applies for PBDD/Fs, another group of polyhalogenated dibenzo-p-dioxins and dibenzofurans. We use both these shorter synonyms in this report.

Table 1: *Overview of samples of chicken eggs, soil, ash, dust and rice crop from hotspots in Java and reference samples used in this study.*

Locality	Sample ID	Matrix	Month/year of sampling	Number of individual (eggs etc.) in pooled sample	Fat content (%)
Bangun	Bangun 1	eggs	05/2019	3	13
Bangun	BAN-E-1	eggs	11/2019	3	9.45
Bangun	BAN-S-1	soil/ash	11/2019	4	-
Kendalsari	KEN 01	eggs	04/2018	9	27.38
Kendalsari	KEN-E-1/19	eggs	11/2019	6	14.29
Kendalsari	KEN-A-1	ash/soil	11/2019	6	-
Kendalsari	KEN-AD-1	ash/dust	11/2019	6	-
Sidokampir	SA1 - 3	ash	04/2018	3	-
Sidokampir	SID-D-1	dust	11/2019	5	-
Sidokampir	SID 01	soil	04/2018	1	-
Sidokampir	SID-S-1/19	soil	11/2019	6	-
Sidokampir	RICE 01	rice crop	04/2018	2	-
Sumberwuluh	SUM-E-1; SUM-E-2	eggs	11/2019	6	14.12
Tangerang	SEM-E-1	eggs	11/2019	3	16.22
Tangerang	TAN-ESIN-01	eggs	11/2019	5	13.69
Tangerang	TAN-EBUT-01	soil/ash	11/2019	5	-
Tropodo	Tropodo 1	eggs	05/2019	3	15
Tropodo	TROP-E-1	eggs	10/2019	6	13.89
Tropodo	TROP-A-1	ash	10/2019	6	-
Tropodo	TROP-A-2	ash	10/2019	5	-
Ref. sample – Bangkok	Supermarket	eggs	02/2016	6	11.6
Ref. sample – Jakarta	JAK-SUP	eggs	11/2019	6	9.53
Ref. sample - Mbeji Forest	MBEJI-S	soil	11/2019	4	-

(v/v) FCS under standard conditions (37°C, 5% CO2, 100% humidity). Cells were exposed in triplicate on 96-well microtiter plates containing the standard 2,3,7,8-TCDD calibration range, a reference egg sample (analyzed by HRGC-HRMS; for the bioassay apparent recovery), a procedure blank, a DMSO blank and the sample extracts in DMSO. Following a 24-hour incubation period, cells were lysed. A luciferin-containing solution was added and the luminescence was measured by using a luminometer (Mithras, Berthold Centro XS3).

The DR CALUX® bioassay method has been shown to be a cost-efficient, semi-quantitative, effect-based toxicity screening analysis for all kinds of stable dioxin-like compounds (PCDD/Fs, dl-PCBs, PBDD/Fs, PBBs, and chlorinated and brominated polycyclic aromatic hydrocarbons, N-dioxins)[12]; however, for confirmation it is recommended to go for more specific PCDD/Fs and dl-PCBs congener analyses, which also allow examination of the fingerprints of dioxins (PCDD/F congener patterns) specific to different sources of pollution.

Seven pooled egg samples from Java as well as samples of soil, ash, rice and dust were analyzed for content of individual PCDD/Fs and an extended list of PCB congeners by HRGC-HRMS at the accredited laboratory of the State Veterinary Institute in Prague, Czech Republic. Samples of eggs collected in Bangun and Tropodo in May 2019 (Bangun 1 and Tropodo 1) were analyzed for specific PCDD/Fs and dl-PCBs congener in MAS laboratory, Muenster, Germany, simultaneously with brominated dioxins.

Eight samples of free-range eggs and selected samples of ash, dust and soil (see Table 12) were also analyzed for content of non-dioxin-like (indicator) PCBs (i-PCBs), DDT, and its metabolites, hexachlorocyclohexanes (HCHs), hexachlorobutadiene (HCBD), pentachlorobenzene (PeCB) and hexachlorobenzene (HCB), in a Czech-certified laboratory (University of Chemistry and Technology in Prague, Department of Food Chemistry and Analysis).

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 ionization mode.

The eight free-range and reference egg samples as well as selected samples of ash, dust and soil (see Table 12) were also analyzed for PBDEs, HBCD, novel BFRs[13] (nBFRs), and short-chain chlorinated paraffins (SCCPs). All of these analyses were conducted in a Czech-certified laboratory (University of Chemistry and Technology, Department of Food Chemistry and Analysis).

Identification and quantification of PBDEs and nBFRs were performed using gas chromatography coupled with mass spectrometry in negative ion chemical ionization mode (GC-MS-NICI). Identification and quantifi-

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[12] “Bioanalytical methods“ means methods based on the use of biological principles like cell-based assays, receptorassays or immunoassays. They do not give results at the congener level but merely an indication of the TEQ level, expressed in Bioanalytical Equivalents (BEQ), to acknowledge the fact that not all compounds present in a sample extract that produce a response in the test necessarily obey all requirements of the TEQ-principle [European Commission (2012). Commission Regulation (EU) No 252/2012 of 21 March 2012 laying down methods of sampling and analysis for the official control of levels of dioxins, dioxin-like PCBs and non-dioxin-like PCBs in certain foodstuffs and repealing Regulation (EC) No 1883/2006 Text with EEA relevance European Commission. Official Journal of the European Communities: L 84, 23.83.2012, p. 2011–2022.

[13] This group of chemicals is represented by the following chemicals: 1,2-bis (2,4,6-tribromophenoxy) ethane (BTBPE), decabromodiphenyl ethane (DBDPE), hexabromobenzene (HBB), octabromo-1,3,3-trimethylpheny-1-indane (OBIND), 2,3,4,5,6-pentabromoethylbenzene (PBEB), and pentabromotoluene (PBT).

cation of HBCD isomers were performed by liquid chromatography interfaced with tandem mass spectrometry, with electrospray ionization in negative mode (UHPLC-MS/MS-ESI).

The extract, which was prepared same way as for the other analyses, was transferred into cyclohexane and diluted. Identification and quantification of SCCPs was accessed via gas chromatography/time-of-flight high resolution mass spectrometry (GC/TOF-HRMS) in the mode of negative chemical ionization (NCI).

Five pooled samples of free-range chicken eggs (one sample from Bangun, two samples from Tropodo, one sample from Kendalsari and one sample from Tangerang) and the control group chicken egg sample from a supermarket in Bangkok were also analysed for polybrominated dibenzo-p-dioxins and dibenzofurans (PBDD/Fs) in the MAS laboratory, Muenster, Germany. The accredited method MAS_PA002, ISO/IEC 17025:2005 was used to determine PBDD/Fs. The basic steps of the analyses can be summarized as follows:

- Addition of $^{13}C_{12}$-labelled PBDD/F internal standards to the sample extract
- Multi-step chromatographic clean-up of the extract
- Addition of 13C12-labelled PBDD/F - recovery standards
- HRGC/HRMS analysis
- Quantification via the internal labelled PBDD/F-standards (isotope dilution technique and internal standard technique).

3. Description of hotspots

Localities chosen for sampling in Java, Indonesia, were sites where higher exposure to unintentionally produced POPs, such as dioxins, was expected due to the activities on or near the sites. Plastic waste suspected to contain either BFRs, SCCPs or PFASs was dumped at some of these sites as well.

Those sites fell into three categories: 1) plastic waste yards with open burning of waste, including plastic from electronic waste in some cases (rural dumpsites), 2) locations affected by secondary aluminum production, and 3) areas affected in some way by waste incineration, either with plastic waste used as fuel in tofu production or hazardous waste incineration. Most of them were located in East Java, and one was in Banten Province, near the capital of Indonesia, Jakarta.

Figure 1: *Location of Banten Province in Indonesia.*

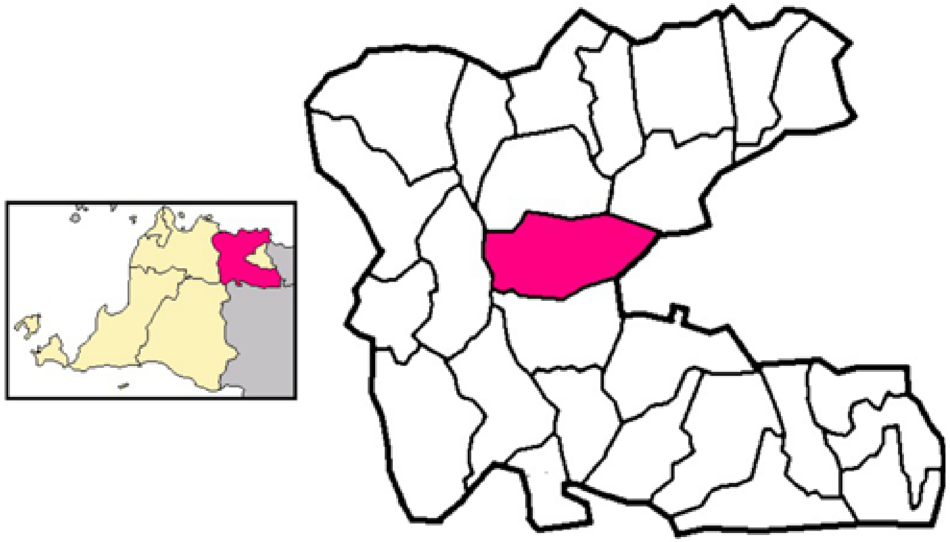

Figure 2: *Map of Tangerang Regency in Banten Province.*

Eggs bought in a supermarket in Jakarta, and soil from the Mbeji Forest in East Java, were chosen as reference samples for comparison with those from potentially contaminated areas.

3.1 BANTEN

3.1.1 TANGERANG REGENCY – PLASTIC WASTE DUMPSITES AND SORTING PLACES

In Tangerang Regency, we choose the Pasar Kemis and Sindang Jaya subdistricts and their surroundings as the sampling sites. In the Pasar Kemis Industrial Estate, there are two plastic manufacturers – PT Harvestindo International (Harvestindo International 2019) and PT New Harvestindo International – that import plastic and metal scraps from various countries as raw materials for their productions. We traced the wastes generated by the two companies around their neighbourhood.

In Pasar Kemis, we found several simple warehouses where the local communities or local recyclers bought unwanted scraps from the Harvestindo group, that they later dumped and burned. In Sindang Jaya, there were several local recycling facilities that also purchased unwanted plastic and metal scraps from the Harvestindo group and then burned the trash that had no value anymore. Some populations, mainly women and children, in the Duta Asri housing complex who were affected severely due to the burning of plastic waste over which they disagreed with the recyclers. We also observed residues of refrigerator insulations at one of the sampled sites.

3.1.2 JAKARTA – SUPERMARKET, REFERENCE SAMPLE FOR EGGS

In Indonesia we bought chicken eggs from a large supermarket as the control egg sample from a big farm. Jakarta is the capital of Indonesia and the largest city in the country, with a population of almost 11 million people (Wikipedia 2020a).

Photo 1: *Metal scrap imported by PT Harvesting sold to a local waste recycler. Photo: Nexus3.*

Photo 2: *Unwanted imported wastes are burned every day in the dumping site, with children and cattle onsite. Photo: Jindrich Petrlik, Arnika.*

Photo 3: *Unwanted plastic scrap sorted by group of recyclers in Tangerang. The residues were burned to reduce the volume and clear the space for the next drop. Photo: Nexus3.*

Photo 4: *Cattle grazing in the illegal dumpsite where plastic waste residues were burned in Tangerang. Photo: Nexus3.*

3.2 EAST JAVA

3.2.1 KENDALSARI, JOMBANG REGENCY - ALUMINUM SMELTERS

The Jombang Regency is located in the East Java Province, situated to the southwest of Surabaya (Wikipedia 2020b). The capital of the regency is the town of Jombang. The regency has an area of 1,159.09 km² (2.3% of the area of East Java). The population of Jombang Regency in 2018 was 1,258,618 people. Jombang Regency is divided into 21 subdistricts, one of which is Sumobito subdistrict, located about 15 km from Jombang city (BPS 2019).

Sumobito and Kendalsari are the centre of small to medium enterprises (SMEs), e.g. aluminum smelters, in Jombang Regency since the 1990s (Rini 2019). The Sumobito and Kesamben subdistricts are also centres of secondary aluminum smelting industry in East Java. The SMEs industry has been processing aluminum waste from several large aluminum smelting companies around Jakarta and West Java since the 1990s.

There are 136 units of secondary aluminum smelting companies in the two districts that produce aluminum ash. After the process, the hot wastes from the aluminum production are disposed of around the villages in big bags. This dumping is done carelessly and spread out in 20 hamlets in the Sumobito and Kesamben Districts.

Based on the Indonesian Government Decree No. 101/ 2014, aluminum waste and slag or residues fall under the category of hazardous wastes (Bahan Berbahaya Beracun or B3), code number B313-2 in the list of B3 waste from specific sources of primary and secondary production processes (Government of Indonesia 2014). Villagers have also used the aluminum waste to strengthen the embankment and irrigation channels in the Sumobito, Kesamben and Peterongan Districts.

Additionally, communities have also used aluminum ash and slag waste to fill their backyards and raise the height of the land to avoid

Figure 3: *Map of the East Java Province*

Figure 4: *Map of Jombang Regency in the East Java Province*

flooding, as well as to widen and raise the level of the roads around sugar cane plantations and rice fields.

When the ash is washed by the rainwater, it will produce leachate, which increases the levels of ammonia, sodium, potassium, chloride and Total Dissolved Solid (TDS) levels in the surrounding environment. This chemical cocktail releases a foul smell and strong, pungent vapour that could make some people dizzy. As the waste potentially also contains POPs, they can leak as well or be spread by the wind with dust.

Photo 5: *Aluminum slags purchased from the large aluminum factories. Photo: Ecoton.*

Photo 6: *A crusher is used to refine the size of the slags. Photo: Nexus3.*

Photo 7: *Fine slags ready to be sent to the smelter plant. Photo: Nexus3.*

Photo 8: *Smelting process using firewood. Photo: Nexus3.*

Photo 9: *Pouring hot melted aluminum into the cast. Photo: Prigi Arisandi, Ecoton.*

Photo 10: *: Aluminum ingots ready to be sold. Photo: Nexus3.*

3.2.2 BANGUN, MOJOKERTO REGENCY – PLASTIC WASTE DUMPSITE AND SORTING PLACE

Bangun village is located in the Mojokerto Regency and is home to about 10,000 inhabitants. The village is situated near a large paper manufacturing company, PT Pakerin. Before the plastic waste import tsunami, the Bangun villagers mainly worked in the agricultural sector, planting seasonal crops such as corn and other types of vegetables.

In the last five years, new economic activity related to plastic wastes has increased and attracted people from other regencies to come and work in the plastic scavenging business. Most families own a pile of plastic waste,

Figure 5: *Map of the East Java Province*

Figure 6: *Map of Mojokerto in the East Java Province*

Photo 11: *In Bangun, open burning is a common practice to get rid of metals, reduce volume and clear the space for the next drop. Photo: Fulli Syafi Handoko, Ecoton.*

Photo 12: *Plastic scrap piles in Bangun village. Fulli Syafi Handoko, Ecoton.*

Photo 13: *Every pile is owned by a family or a group of villagers. Fulli Syafi Handoko, Ecoton.*

Photo 14: *Free-range chicken were foraging between plastic waste piles. Photo: Jindrich Petrlik, Arnika.*

and every day or two they purchase unwanted plastics from the paper factory or the truck driver for IDR 250,000 (~USD $18) per truck (capacity 4 tons), or ask the driver to drop it off in their spots and in return give a tip of IDR 20,000 (~USD $1.40). Such piles can be seen in front of every house in the village, and more piles can be found in a huge sorting site.

When the plastic waste piles are purchased directly from the factory, sometimes the bales still contain metal wire to bind the bales. Some of the recyclers later burn the wire along with the leftover plastics that have not been sold in order to create space for new piles.

3.2.3 TROPODO, KRIAN SUB-DISTRICT, SIDOARJO REGENCY – TOFU FACTORIES BURNING PLASTIC WASTE

Tropodo village, located in the Sidoarjo Regency, has a population of approximately 25,000. There are 50 small-scale tofu makers that use unwanted plastic scrap as fuel to create steam and turn the soybean milk into tofu.

Photo 15: *Plastic scrap piles as fuel feed stock in a tofu factory. Photo: Nexus3.*

Photo 16: *Plastic scraps burned in a simple furnace to create steam. Photo: Nexus3.*

Photo 17: *Daily view in Tropodo: black smoke between houses. Photo: Ecoton.*

Photo 18: *Ashes from the furnace used to fill and reclaim the agricultural land. Photo: Jindrich Petrlik, Arnika.*

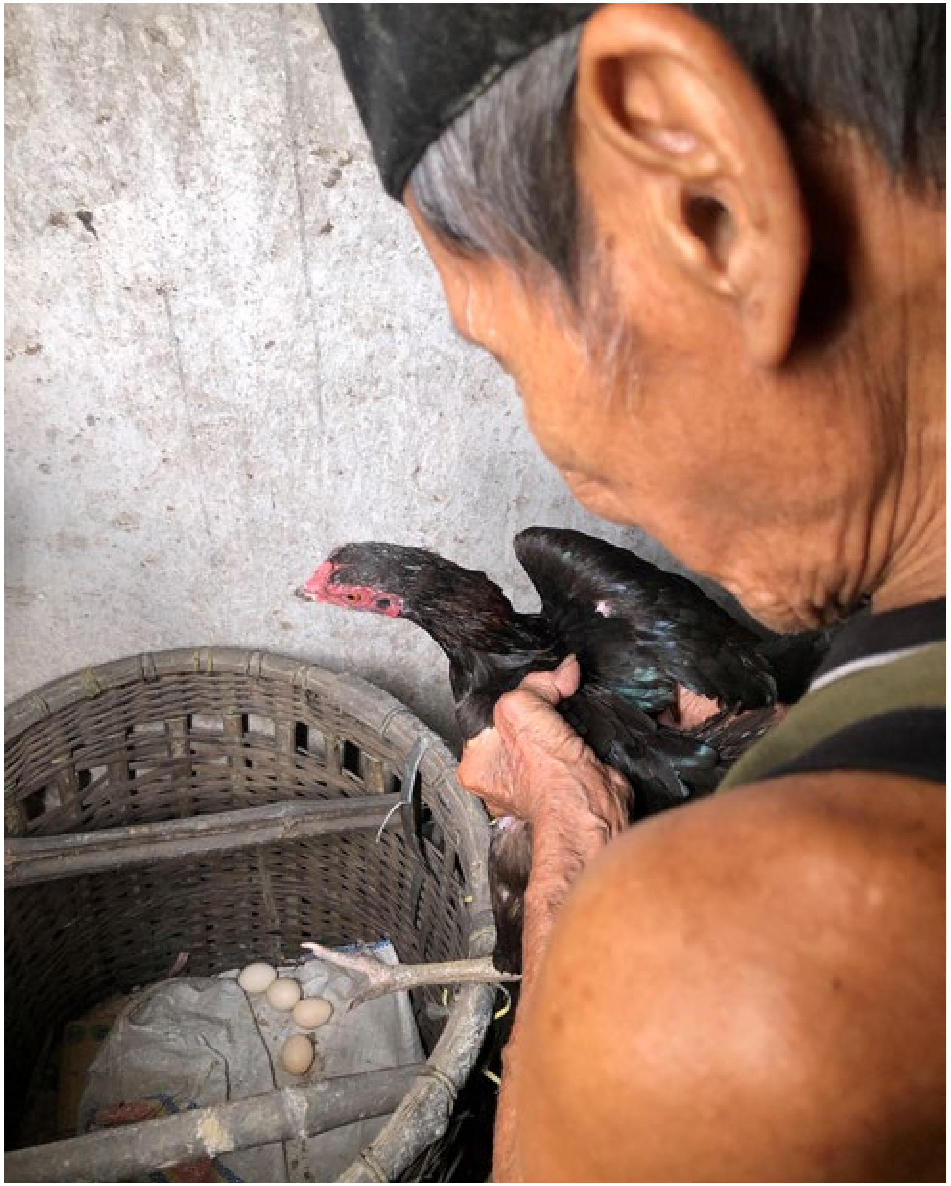

Photo 19: *Chicken eggs from near a tofu factory contaminated with dioxins. Photo: Nexus3.*

Photo 20: *Yuyun Ismawati (Nexus3) is checking ash residues after burning of plastics in one of tofu factories in Tropodo. Photo: Jindirch Petrlik, Arnika.*

Five years ago, most tofu makers used wood to create hot steam. The price of one small truckload of firewood was about IDR 1.5 million (~USD $107). When the paper companies started to receive increased volumes of unwanted plastic scrap, tofu makers saw the opportunity to cut their production costs by replacing the firewood with unwanted low-grade plastic scrap for fuel. The price of one small truck of plastic scrap is IDR 250,000 to 350,000 (~USD $18 to $25). By burning the plastic waste to create steam, tofu makers can cut their production costs by up to 15%-20%.

The combustion of mixed plastic scraps takes place all day long, from 8 a.m. to 4 p.m., releasing thick black smoke. In the morning, people who pass the main road nearby are smothered with smoke-like fog every day. The Primary Health Clinic of the Krian subdistrict has recorded an elevated number of patients with respiratory problems from the Tropodo area, especially children. Some tofu makers donate the ashes from the plastic burning to corn farmers to be used as fertilisers. Others use the ash to reclaim the agricultural land from wetland areas.

3.2.4 LAKARDOWO, MOJOKERTO REGENCY – HAZARDOUS WASTE INCINERATOR

In the last 10 years, the inhabitants of the Lakardowo village in the Jetis subdistrict have suffered from pollution released by a hazardous waste facility, PT Putra Restu Ibu Abadi (PRIA 2019). The population of the Lakardowo village, about 3,500 people, have suffered from various ailments such as skin problems, respiratory problems, and other kinds of non-communicable diseases (Riski 2018).

The company PT PRIA was established in 2010 and has intensively increased its services and expanded its sites since 2016. The company collected hazardous wastes from the Jakarta area, West Java, Central Java, Yogyakarta, the East Java area and Bali, from various sources including health care services facilities (medical wastes).

Photo 21: *Black smokes released by PT PRIA during their daily operation. Photo: Ecoton.*

Photo 22: *PT PRIA buried all the slags and ashes within the fenced area of their property. Photo: Ecoton.*

Photo 23: *Smokes from PT PRIA released almost 24 hours, 7 days. Photo: Ecoton.*

Photo 24: *The slags and fly ash piled under the warehouse potentially contaminated the groundwater and wells of several houses in the nearby village. Photo: Mongabay.*

Photo 25: *Coal fly ash from PT PRIA sold to the community for foundation of houses. Photo: Mongabay.*

Photo 26: *Some houses in the nearby village of PT PRIA used fly ash as the foundation material of their houses. Photo: Ecoton.*

Inside the property, PT PRIA has two units for hazardous waste treatment : an incinerator and an electrocoagulation process. The company have buried the ashes and slags from the incinerator operation underneath their buildings within the company grounds. However, people in Lakardowo have stated that they have very often witnessed black smoke released from the stacks inside the PT PRIA property which did not smell very nice. Some inhabitants in houses with groundwater access have also complained about its quality and say that it has caused them skin irritation and other ailments.

3.2.5 SUMBER BEJI SPRING, PANGLUNGAN JOMBANG, "MBEJI FOREST" – REFERENCE SITE

The field team collected soil samples at the entranceway to the springwater forest Sumber Beji, also called "Mbeji Forest." The reference site is located at Sranten, Panglungan Village, a mountainous area situated at 600 to 800 meters above sea level.

The village area lies at the foot of the Anjasmoro Mountain, surrounded by a teak timber forest that is managed by Perhutani (a state-owned company), bordering the natural forest of Bromo Tengger Semeru National Park.

The village has significant potential for economic development from tourism, due to its stunningly beautiful and natural scenery of forest, mountains, waterfalls and water springs, as well as the iconic local durian, bido and other tropical fruits (Kompasiana 2012).

The Sumber Beji Forest is a natural forest with water springs that feed into Gunting River, a tributary of Brantas River. Sumber Beji is also a sacred forest for the local communities.

These communities have prohibited visitors from taking any plants or animals from the woods and water spring areas to avoid getting lost in the forest, accidents, or bad luck. There are sacred tombs in the forest that local people preserve for cultural rituals, during which they bring offerings to pray for prosperity and safety.

Photo 27: *Forest is used for coffee cultivation and natural forest for spring conservation, sacred forest for cultural rituals and prayers. Photo: Ecoton.*

However, housing developments and poultry farms are threatening the springs and forest. A local community group organized around forest protection, offers an education program for local students and undertakes reforestation efforts with native trees (Riski 2015).

Like other rural areas in Indonesia, the community in Panglungan Village does not have access to a waste management system, which should be provided by the local government. Although most of the wastes generated in the village are organic, waste burning is a common way of managing solid waste in the village.

4. Results and discussion

4.1 FREE-RANGE CHICKEN EGGS

The results of the chemical analyses for various POPs of nine free-range chicken egg samples from Bangun, Tropodo, Kendalsari, Sumberwuluh (close to Lakardowo), and Tangerang, as well as reference egg samples from supermarkets, are summarized in Table 3. Details about the sampling and the sampled localities can be found in chapters 2 and 3. Their evaluation is discussed further in separate subchapters according to the natural groups of POPs.

There is no special subchapter dedicated to organochlorine pesticides in these eggs, DDT and metabolites and HCHs, as they were not found in very high levels in our samples in comparison with samples from some other locations, e.g. eggs from Vikuge, Tanzania, sampled in 2005 or Klatovy – Luby, Czech Republic, sampled in 2003, and Peshawar, Pakistan, sampled in 2005, with observed levels of DDT at 7,041, 2,321 and 2,329 ng g^{-1} fat, respectively (Petrlik, Khwaja et al. 2005, Dvorská 2007, IPEN Pesticides Working Group 2009). The highest level for the sum of DDT measured in eggs from Kendalsari in this study reached 105 ng g^{-1} fat (see Table 3).

The measured levels of POPs in our chicken egg samples were compared with the legislative limits established in Indonesia and in the European Union, although not all measured chemicals in this study have defined limits. For example, the European Union does not currently have a limit for SCCPs, PFASs, brominated flame retardants or PBDD/Fs in chicken eggs. The limit values for eggs are summarized in Table 2. These limits are used for comparison with levels measured in food in many other studies, mainly in developing countries that do not have official limits for dioxins and other POPs in food. Indonesia has set a limit value for dioxins and dl-PCBs in eggs (Badan pengawas obat dan makanan Republik Indonesia 2018).

Table 2: *Limit concentration values for OCPs, PCBs and PCDD/Fs-TEQs in chicken eggs.*

Unit	Indonesia	Hen eggs	
		EU ML[1]	EU MRL[2]
	pg g^{-1} fat	pg g^{-1} fat	ng g^{-1} fresh weight
WHO-PCDD/Fs TEQ	-	2.5	-
WHO-PCDD/Fs-dl-PCB TEQ	2.5[5]	5.0	-
PCBs[3]	-	40	-
HCB		-	20
DDT total[4]		-	50
γ-HCH (lindane)		-	10
γ-, γ-HCH*		-	20, 10

[1] EU Regulation (EC) N°1259/2011. Maximum level (ML) – Food with PCDD/Fs and dl-PCBs concentrations above this level is considered to be contaminated and is not suggested for consumption.

[2] Regulation (EC) N°149/2008. Maximum residue level (MRL) means the upper legal level of a concentration for a pesticide residue in or on food or feed set in accordance with the Regulation, based on good agricultural practice and the lowest consumer exposure necessary to protect vulnerable consumers.

[3] The sum of PCB28, PCB52, PCB101, PCB138, PCB153 and PCB180

[4] The sum of p,p´-DDT, o,p´-DDT, p,p´-DDE and p,p´-DDD

[5] Limit set in TEF that includes both PCDD/Fs and dl-PCBs

*MRL is set separately for each isomer

***Table 3:** Overview of results of the chemical analyses for POPs in nine free-range chicken egg samples from Javanese hot spots, and two egg samples from a commercial farm, one bought in Jakarta, the other one in Bangkok. The samples from Indonesia were taken in 2018-2019. The sample from Bangkok was taken in February 2016. Levels of POPs are in ng g^{-1} fat if not specified otherwise.*

Locality	Bangun	Bangun	Tropodo	Tropodo	Kendalsari	Kendalsari	Sumberwuluh	Tangerang	Tangerang	Jakarta	Bangkok	Indonesia limit	EU limits
Sample ID (eggs)	**Bangun 1**	**BAN-E-1**	**Tropodo 1**	**TROP-E-1**	**KEN 01**	**KEN-E-1/19**	**SUM-E-1 and E-2**	**SEM-E-1**	**TAN-ESIN-01**	**JAK-SUP**	**supermarket**		
Number of eggs in pooled sample	3	3	3	6	9	6	6	3	5	6	6		-
Fat content (%)	13	9.5	15	13.9	27.4	14.3	14.1	16.2	13.7	9.5	11.6		-
PCDD/Fs (pg TEQ g^{-1} fat)	10.8	9.5	200	140	49	41	11.0	54	20.4	0.0012	0.08		2.50
DL-PCBs (pg TEQ g^{-1} fat)	3.1	5.1	32	32	35	20	2.0	18	7.4	0.0020	0.001		-
Total PCDD/F + DL-PCBs (pg TEQ g^{-1} fat)	13.9	14.6	232	172	84	61	13.0	72	28	0.0032	0.1	2.50	5.00
Total PCDD/Fs + DL-PCBs - DR CALUX® (pg BEQ g^{-1} fat)	21	13	560	NA	NA	NA	NA	88	NA	<LOQ <0.6	NA		-
PBDD/Fs (pg TEQ g^{-1} fat)	< 21.3	NA	< 21.3	0.331	NA	0.565	NA	6.93	NA	NA	< 21.3		-
HCB	2.7	3.61	5.5	4.10	1.5	2.49	0.58	6.05	NA	<0.1	< 0.2		-
PeCB	1.1	2.23	1.9	1.67	1.07	1.28	0.26	3.63	NA	<0.1	< 0.4		-
HCBD	< 0.1	<0.1	< 0.1	<0.1	<0.1	<0.1	<0.1	<0.1	NA	<0.1	< 0.4		-
7 PCB[1]	15.4	16.9	5.3	2.9	7.0	3.7	<LOQ	3.4	NA	<LOQ	0.22		-
6 PCB[2]	12.3	14.0	4.4	2.9	5.1	2.9	<LOQ	3.4	NA	<LOQ	0.22		40.00
SCCPs	153	97	65	97	NA	161	50	153	NA	136	-		-
sum HCH	0.9	<LOQ	0.8	<LOQ	<LOQ	<LOQ	<LOQ	4.68	NA	<LOQ	2.2		-
sum DDT	4.3	4.3	10.8	3.4	60	105	5.1	3.9	NA	<LOQ	< LOQ		-
sum HBCD	5.2	538	<LOQ	<LOQ	<LOQ	<LOQ	4.5	844	NA	<LOQ	<LOQ		
sum of PBDEs	91	1,457	65	27,159	6.2	150	8.2	321	NA	1.4	3.1		-
decaBDE (BDE 209)	54	1,265	4.1	24,611	< 2	75	7.0	77	NA	0.5	<1.0		
7 PBDEs [3]	19	32	52	143	6.2	45	1.0	181	NA	1.4	< LOQ		
sum of NBFRs	NA	124	NA	2166	<LOQ	12.2	0.87	33	NA	<LOQ	<LOQ		
sum of PFASs (ng g^{-1} of fresh weight)	26	97	2.7	0.30	NA	0.35	0.46	6.2	NA	0.1	NA		-
PFOA (ng g^{-1} of fresh weight)	0.39	0.05	0.1	<0.01	NA	<0.01	0.01	0.27	NA	<0.01	NA		
br-PFOS (ng g^{-1} of fresh weight)	2.23	16.10	0.14	0.03	NA	0.02	0.04	0.66	NA	<0.01	NA		
n-PFOS (ng g^{-1} of fresh weight)	15	76	0.9	0.11	NA	0.12	0.22	1.8	NA	<0.01	NA		-

[1] 7 PCBs including congeners 28, 52, 101, 118, 138, 153 and 180; [2] 6 PCBs similar to 7 PCBs but congener 118 is excluded in the sum;

[3] The following congeners are included in this sum: BDE 28, BDE 47, BDE 99, BDE 100, BDE 153, BDE 154, and BDE 183

4.1.1 DIOXINS (PCDD/Fs) AND OTHER UNINTENTIONALLY PRODUCED POPs

The basic description of unintentionally produced POPs (UPOPs) is provided in subchapter 1.5. Eggs measured in this study contained HCB, HCBD, PeCB, PCBs, and PCDD/Fs. Polychlorinated naphthalenes were not analyzed.

4.1.1.1 Dioxin-like activity of eggs measured by using bioassay analyses

Several bio-analytical tools are accepted by international standards[14] for measuring dioxin-like activity in environmental and food samples. These methods are an easier and more cost-efficient option for screening larger quantities of environmental, food or human samples, and many studies use them to evaluate contamination by dioxins and dioxin-like substances, e.g. for food (Hoogenboom, Traag et al. 2006, Behnisch Peter A. 2011, Hussain A 2011, Polder, Müller et al. 2016).

The four pooled egg samples in this study were analyzed using the DR CALUX® method. The highest level in BEQs was measured in the sample from Tropodo (560 pg BEQ g^{-1} fat) followed by a sample from the site affected by open burning and dumping of plastic waste in Tangerang (88 pg BEQ g^{-1} fat). Two samples from Bangun were also suspected not to meet neither the Indonesian nor the EU limit for PCDD/Fs + dl-PCBs, of 2.5 and 5 pg TEQ g^{-1} fat, respectively.

All sample results measured using the DR CALUX® method (see Table 3; total PCDD/Fs + dl-PCBs – DR CALUX® in pg BEQ g^{-1} fat) were also in the same order of magnitude in the chemical HRGC/HRMS analysis for PCDD/Fs + dl-PCBs -TEQ (see Table 3; total PCDD/Fs + dl-PCBs in pg TEQ g^{-1} fat):

- Bangun-1 (DR CALUX® 21 vs chemical analysis 13.9; therefore non-compliant according to EU guidelines);
- BAN-E-1, second sample from Bangun (DR CALUX® 13 vs chemical analysis 14.6; therefore non-compliant according to Indonesian and EU guidelines);
- Tropodo-1 (DR CALUX® 560 vs chemical analysis 232; therefore non-compliant according to Indonesian and EU guidelines);
- SEM-E-1, sample from Tangerang (DR CALUX® 88 vs chemical analysis 72; therefore non-compliant according to Indonesian and EU guidelines)
- and finally the egg sample from the Jakarta supermarket which was the only one compliant with both the Indonesian and the EU guidelines (DR CALUX® below LOQ of 0.6 vs chemical analysis 0.0032).

The differences between the results from the DR CALUX® analyses and the chemical HRGC/HRMS analyses could potentially be explained by more chemicals showing dioxin-like activity, which were not analyzed by any of the instrumental analyses in our study[15], and/or which were analyzed in our study but are not included in the WHO-TEQ value.[16]

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[14] Those standards are such as EC/644/2017, EPA 4435/2008, JIS 463/2009, Dutch Specie 07/2005 and the Chinese standard for Solid waste—Screening of PCDD/Fs—Chemical activated luciferase expression, 2018.

[15] Substances with dioxin-like properties that can bind to the AhR like e.g. PCNs, mixed polyhalogenated dioxins, polybrominated biphenyls, chlorinated dibenzothiophenes, and other chemicals; see Behnisch, P., K. Hosoe and S.-i. Sakai (2001). "Bioanalytical screening methods for dioxins and dioxin-like compounds - a review of bioassay/biomarker technology." Environment International 27(5): 413-439, Giesy, J. P., K. Hilscherova, P. D. Jones, K. Kannan and M. Machala (2002). "Cell bioassays for detection of aryl hydrocarbon (AhR) and estrogen receptor (ER) mediated activity in environmental samples." Marine Pollution Bulletin 45(1): 3-16.

[16] These can include PBDD/Fs, some BFRs; see Behnisch, P. A., K. Hosoe and S.-i. Sakai (2003). "Brominated dioxin-like compounds: in vitro assessment in comparison to classical dioxin-like compounds and other polyaromatic compounds." Environment International 29(6): 861-877. According to some older scientific studies this might include HCB as well, but its contribution would be probably negligible in samples presented in this study as *"HCB binds to the Ah receptor about 10,000 times less than TCDD"* van Birgelen, A., P.J.M. (1998). "Hexachlorobenzene as a possible major contributor to the dioxin activity of human milk." Environ Health Perspect 106(11):

Part of that difference can also be explained by a variation in the homogenicity of the sample, although the same homogenate was used for all analyses. The chemical analyses of PCDD/Fs and dl-PCBs were conducted with a generally used certainty of ±40%. Bioassay analyses of eggs and other environmental samples could be a pathway to broader monitoring of dioxin contamination in Asian countries.

4.1.1.2 Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) and dioxin-like polychlorinated biphenyls (dl-PCBs)

All nine free-range chicken egg samples in this study analyzed for PCDD/Fs and dl-PCBs by instrumental analysis exceeded a level of 2.5 pg WHO-TEQ g-1 fat, the limit for dioxin content in eggs set in Indonesia (Badan pengawas obat dan makanan Republik Indonesia 2018) as the sum of PCDD/Fs and dl-PCBs. They also exceeded the EU maximum level (ML) of PCDD/Fs, and the sum of PCDD/Fs and dl-PCBs, expressed as WHO-TEQ (see Tables 2 and 3) (European Commission 2011). The background levels for PCDD/Fs and dl-PCBs measured in chicken eggs from a supermarket in Jakarta were 0.0012 and 0.0020 pg WHO-TEQ g-1 fat, respectively. The highest level of dioxins (200 pg WHO-TEQ g-1 fat) and dl-PCBs (35 pg WHO-TEQ g-1 fat), respectively, were measured in eggs from Tropodo and Kendalsari. Eggs from Tropodo were sampled in the vicinity of tofu factories burning plastic waste, and in Kendalsari they were sampled next to a small aluminum smelter.

In the eggs from Tropodo were measured the highest, as well as the second-highest level of PCDD/Fs and dl-PCBs in this study, 232 and 172 pg WHO-TEQ g^{-1} fat, respectively. The third to fifth highest levels were measured in eggs from Kendalsari, where there were aluminum smelters in the area (85 and 60 pg WHO-TEQ g^{-1} fat), and Tangerang (72 pg WHO-TEQ g^{-1} fat), respectively.

The whole village of Kendalsari is paved with aluminum ash residues, which can contain significant levels of PCDD/Fs. Emissions and ash from open burning of plastic waste can be the source of dioxin contamination in the eggs from Tangerang. In addition, another egg sample from the Tangerang area contained a high level of PCDD/Fs and dl-PCBs (28 pg WHO-TEQ g^{-1} fat).

Samples from Bangun and Sumberwuluh (near Lakardowo) contained between 13 and 15 pg WHO-TEQ g^{-1} fat of total PCDD/Fs and dl-PCBs, which are levels almost triple the EU maximum level (ML), and six times the Indonesian maximum level, respectively.

All samples were above the background level of WHO-TEQ measured in eggs from the supermarket by more than 4,060-fold (sample from Sumberwuluh) to 72,500-fold (sample from Tropodo).

Dioxin levels in five samples from this study are among the 20 egg samples with the highest-ever measured levels of PCDD/Fs (see graph in Figure 7 and data in Table 4). The egg samples from Tropodo with 200 and 140 pg WHO-TEQ g-1 fat of PCDD/Fs are the third- and the fourth-highest ever measured levels of these chemicals in eggs from Asia, right after two egg samples from the Bien Hoa airbase area in Vietnam (Traag, Hoang et al. 2012, Kudryavtseva, Shelepchikov et al. 2020). The level of 54 pg WHO-TEQ g-1 fat of PCDD/Fs measured in eggs from Tangerang (sample SEM-E-1) is the sixth-highest in egg samples from Asia, after the fifth-highest eggs from Samut Sakhon, Thailand (Petrlik, Teebthaisong et al. 2017).

The level of 49 pg WHO-TEQ g-1 fat of PCDD/Fs in eggs from the vicinity of a secondary aluminum smelter in Kendalsari is comparable with levels in another sample from a hospital site in Accra (Petrlik, Adu-Kumi et al. 2019) influenced by residual ash from a small medical waste incinerator (see Table 4), and also comparable to the highest level of PCDD/Fs in free-range eggs from Newcastle allotments, in the area where the incineration ash from the Byker waste incinerator was used to pave the path

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683-688, Ruprich, J. (1999). "Hexachlorbenzen : přispívá k dioxinové toxicitě více než PCB?" Zprávy Centra potravinových řetězců v Brně 8(1): 4-6.

between allotments in 2000 (Pless-Mulloli, Schilling et al. 2001). It is also close to the highest level of PCDD/Fs and dl-PCBs measured in BEQs in pooled egg samples from another site impacted by waste incineration fly ash in a U.K. farm in Bishops Cleeve (Katima, Bell et al. 2018). What these sites all have in common is the potential influence of industrial processes generating ash residues, either from waste incineration or small metallurgical facilities (see Table 16 in this report). Dioxin-like PCBs were lower in comparison with PCDD/Fs in all samples. The highest level of dl-PCBs, 35 pg WHO-TEQ g^{-1} fat, was measured in eggs from Kendalsari.

The lowest dl-PCBs in free-range eggs had a sample from Sumberwuluh, 2 pg WHO-TEQ g^{-1} fat, which exceeded the background level in eggs from the supermarket by 990-fold. It was followed by samples from Bangun with levels of dl-PCBs 3.1 and 5.1 pg WHO-TEQ g^{-1} fat, respectively.

Figure 7: *Graph showing maximum levels of PCDD/Fs measured in chicken eggs in different countries. Samples before 2006 are in WHO-TEQ 1998. Sources of information are listed in Table 4.*

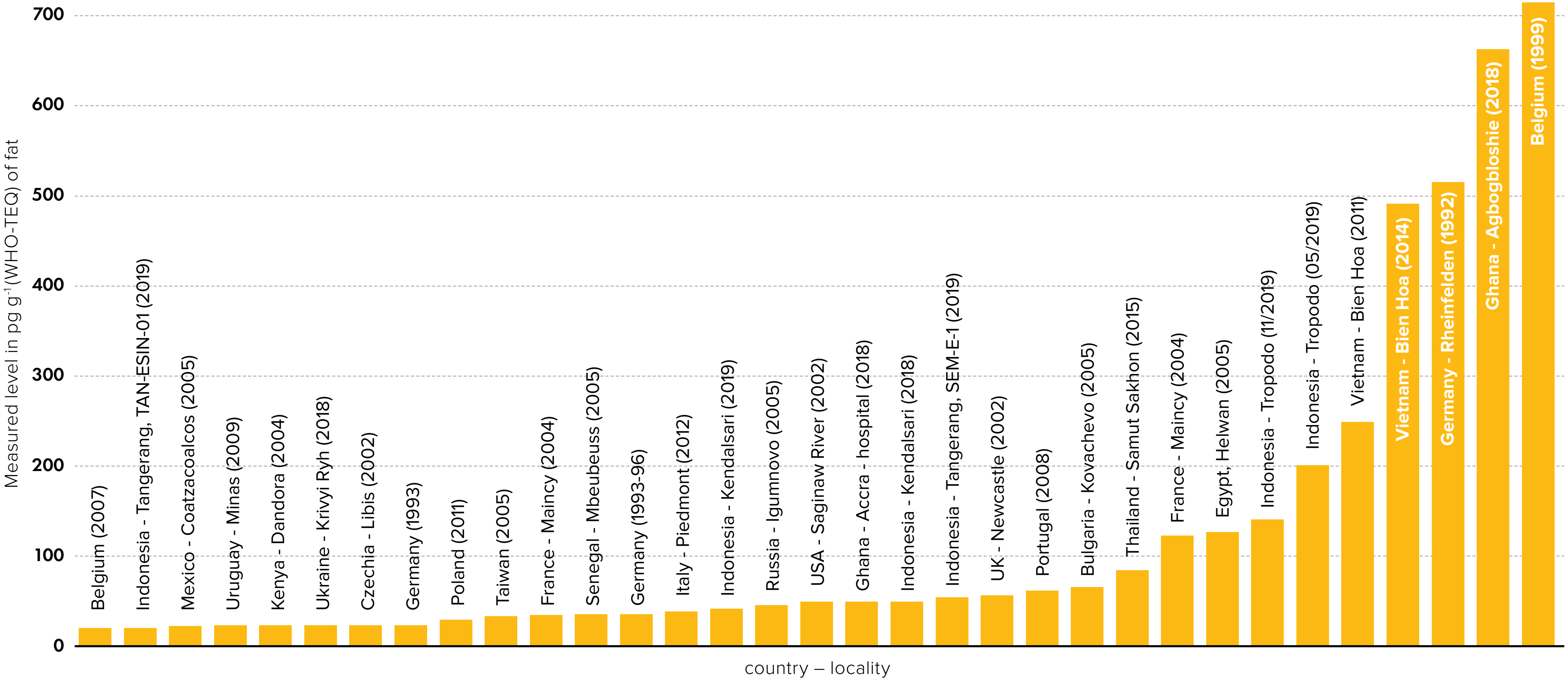


Table 4: *Overview of poultry egg samples with the highest measured levels of PCDD/Fs since the 1990s.*

Country	Year	Locality	PCDD/Fs pg WHO-TEQ g^{-1} fat	Source	Comments
Belgium	2007	Not specified	20	(Van Overmeire, Pussemier et al. 2009)	-
Indonesia	2019	Tangerang	20	This study	Open burning of plastic waste
Mexico	2005	Coatzacoalcos	22	(DiGangi and Petrlik 2005)	Petrochemical complex; hazardous waste incinerator
Uruguay	2009	Minas	23	(Reyes 2010, Uruguay 2017)	Cement kiln co-incinerating PCBs
Kenya	2004	Nairobi - Dandora	23	(DiGangi and Petrlik 2005)	Open burning at dumpsite
Ukraine	2018	Krivyi Ryh	23	(Petrlík, Straková et al. 2018)	Metallurgical and coke plants
Czechia	2002	Libis	23	(Greenpeace CZ 2002)	Chlor-alkali plant, dioxin contaminated site
Germany	1993	Not specified	23	(Fürst, Fürst et al. 1993)	Either PVC burning or PCP - not clear from
Poland	2011	Not specified	29	(Piskorska-Pliszczynska, Strucinski et al. 2016)	PCP treated wood
Taiwan	2005	Changhua county	33	(The Epoch Times 2005)	Metallurgical plants (steelworks); (duck eggs)
Uzbekistan	2001	Chimbay	34	(Muntean, Jermini et al. 2003)	Potential use of 2,4,5-T in cotton cultivation
Senegal	2005	Mbeubeuss	35	(DiGangi and Petrlik 2005)	Mixed waste dumpsite, potential PCP contamination
Germany	1993-96	Not specified	35	(Malisch 1998)	Not specified (free-range chicken eggs)
Italy	2012-13	Piedmont region	38	(Squadrone, Brizio et al. 2015)	Secondary aluminum smelter
Indonesia	2019	Kendalsari	41	This study	Secondary aluminum smelters/contaminated ash
Russia	2005	Igumnovo	45	(DiGangi and Petrlik 2005)	Chlorine chemical industry area; HWI
USA	2002	Saginaw River	49	(MDEQ 2003)	Floodplain downstream from chlorine chemical industry
Ghana	2018	Accra - hospital WI	49	(Petrlik, Adu-Kumi et al. 2019)	Medical waste incinerator ash
Indonesia	2018	Kendalsari	49	(This study, Praha 2018)	Secondary aluminum smelter
Indonesia	2019	Tangerang	54	This study	Open burning of plastic waste and e-waste plastics
UK	2000	Newcastle	56	(Pless-Mulloli, Schilling et al. 2001)	Waste incineration ash
Portugal	2008	Not specified	61	(Cardo, Castel-Branco et al. 2014)	PCP treated wood
Bulgaria	2005	Kovachevo	65	(DiGangi and Petrlik 2005)	Industrial area with coal burning power plants
Thailand	2015	Samut Sakhon	84	(Petrlik, Teebthaisong et al. 2018)	Artisanal e-waste and general waste recycling; open burning
France	2004	Maincy	122	(Pirard, Focant et al. 2004)	Old waste incinerator operating between 1974-2002
Egypt	2005	Helwan	126	(DiGangi and Petrlik 2005)	Metallurgical workshops
Indonesia	2019	Tropodo	140	This study	Plastic waste used as fuel in tofu factories/ash
Indonesia	2019	Tropodo	200	This study	Plastic waste used as fuel in tofu factories/ash
Vietnam	2011	Bien Hoa	249	(Traag, Hoang et al. 2012)	Former U.S. military base, dioxin contaminated site
Vietnam	2014	Bien Hoa	490	(Kudryavtseva, Shelepchikov et al. 2020)	Former U.S. military base, dioxin contaminated site
Germany	1992	Rheinfelden	514	(Malisch, Schmid et al. 1996)	Waste from chlor-alkali chemical plant
Ghana	2018	Agbogbloshie	661	(Petrlik, Adu-Kumi et al. 2019)	E-waste and automobile scrapyard
Belgium	1999	Not specified	713	(van Larebeke, Hens et al. 2001)	Dioxin contamination of feed

4.1.1.3 Dioxin congeners patterns

Major sources of pollution by dioxins were listed in Annex C to the Stockholm Convention, and their emission factors were specified in the Dioxin Toolkit for calculation of dioxin emission inventories. Some of these sources were found to have specific profiles/ratios of PCDD/Fs congeners, which can be found in the literature (Dopico and Gómez 2015, Fiedler, Malisch et al. 2018). However, the fate of specific dioxin congeners in the environment may vary in the sense that we don't necessarily receive an exact match of the original pollution source pattern in eggs and/or other environmental samples. Specific congener patterns act like unique 'fingerprints' for the pollution source and can sometimes be used to trace the source of environmental pollution.

Levels observed in certain environmental matrices can also be caused by the influence of mixed dioxin sources, including general background contamination, although a major source can still be partly recognized in some prevailing congeners. This has been described in many scenarios, like for example for sediments in the Baltic sea (Sundqvist, Tysklind et al. 2009). It applies even more to animals. For example, Malisch and Kotz (2014) concluded that *"bioaccumulation from feed to food of animal origin changes the dioxin patterns considerably.*"

Originally, it was believed that only when we can receive such exact fingerprints, can we prove the contamination source. As science has gained more information about specific dioxin congeners' concentration in different environmental compartments, and releases from different pollution sources, it has become clearer that many conditions play a role in the final pattern that we see, for example, in chicken eggs.

Those conditions include the bioavailability of each of the congeners, their fate in soil or sediments, and/or the individual metabolism of animals (Stephens, Petreas et al. 1995, Henriksson, Bjurlid et al. 2017). The whole transfer and fate of dioxin congeners has not been fully discovered yet. We must keep this limited knowledge in mind when looking for sources of pollution. The similarity of dioxin patterns of different combustion sources also plays a significant role. We can try to find the most likely sources of egg contamination, bearing the aforementioned limitations of such a search in mind.

The bioavailability of dioxin congeners in poultry is "*chlorination-dependent, ranging from 80% for tetrachlorinated to less than 10% for octachlorinated congeners,*" according to a study by Stephens et al. (1995). Furthermore, Kang et al. (2002) reported that the biomagnification factors of PCDD/Fs in wild tufted ducks decreased with an increase in the degree of chlorination. This indicates that the dioxin congener pattern in eggs can be different than the pattern found in the contamination source.

We also tried to take indicative pool samples of soil or ash in places where we found them to be accessible for the hens from which we received egg samples. However, we must admit that our knowledge about the overall contamination at the sites is limited because of the limited number of samples and the lack of measured data about dioxin congener profiles of air emissions. Our ability to fully follow the food chain contamination was limited by the resources available for this study, which has to be considered as pilot information.

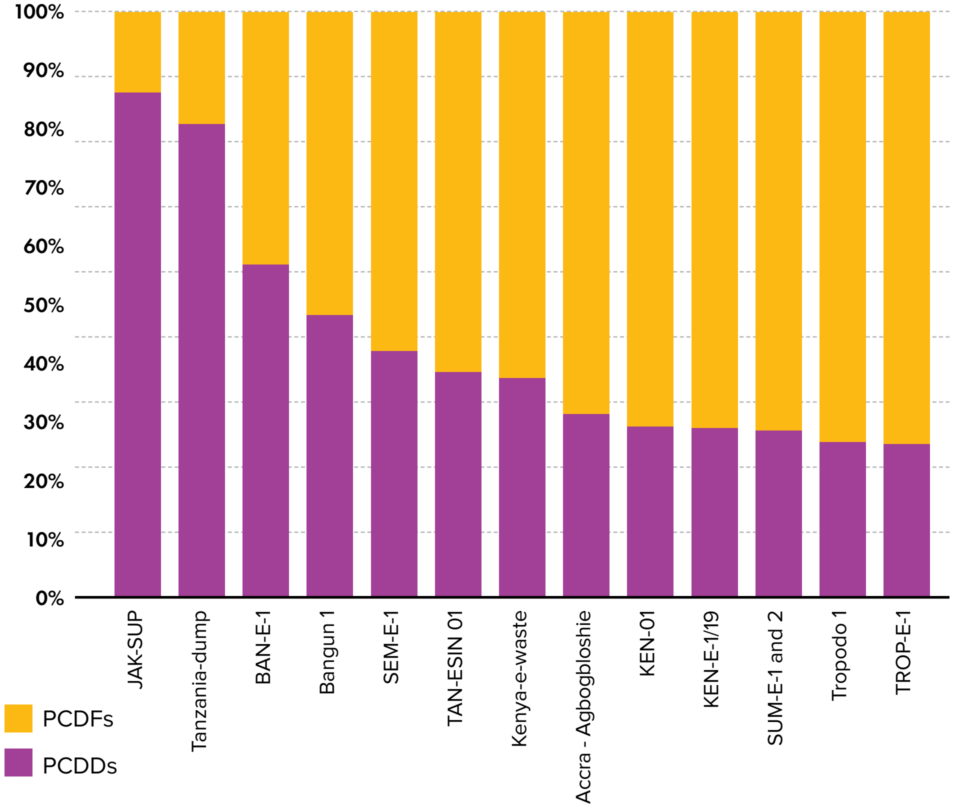
Zhang et al. (2017) concluded with regards to dioxin congener patterns for waste open burning sources: *"Due to the large variability in combustion conditions and composition of fired fuel, the distribution pattern of the seventeen 2,3,7,8-PCDD/Fs varies significantly for different sources. No specific pattern could be established from the reported data."* They also suggested that: *"Some biomass materials, such as lignin, are almost to be considered as a three-dimensional phenol polymer. Truly, these explanations seem solidly supported, yet the emissions of PCDD/Fs still remain largely unpredictable in both their amount and fingerprint.*

Another justification may be in the higher chlorine content of the anthropogenic fuels involved, in which PVC may act as a chlorine donor.

Wang et al. (2003) used principal component analysis (PCA) to compare congener profiles of PCDD/Fs in flue gases from various emission sources and proposed a threshold value of the chlorine content at 0.8–1.1 wt. %. When the chlorine content in fuels is lower, the formation of PCDDs dominates; once above this threshold, the rate of formation of PCDFs increases faster than that of PCDDs“ (Zhang, Buekens et al. 2017).

PCDF/PCDD ratios in the egg samples can be found in the graph in Figure 8. We have also added information obtained from analyses of eggs from African locations where open burning of waste occurs. We can observe the group of samples from Tanzania (dumpsite vicinity) and two samples from Bangun with a ratio between 0.2 and 1, followed by the group of samples from Tangerang plus an e-waste site in Kenya with ratios of 1.4-1.7, samples from an e-waste scrapyard in Agbogbloshie that has a ratio of 2.2, and finally ratios for samples from Kendalsari and Tropodo varying between 2.4 and 2.8. These groups match partly to the conclusions made by Zhang et al. (2017), as the samples start from:

Figure 8: *PCDFs/PCDDs congener ratios in samples from Indonesia in comparison with some egg samples from Africa. Sources of information for samples from Africa: (SVÚ Praha 2019, SVÚ Praha 2020)*



1) the group potentially influenced by open burning of waste at dumpsites; followed by,
2) the group of samples potentially contaminated by open burning of waste containing e-waste plastic; and lastly,
3) the group potentially influenced by combustion sources in “closed” systems and/or ash residues from such processes, aluminum smelters (Kendalsari) and other kinds of waste incineration (Tropodo and Lakardowo).

The congener profiles for PCDD/Fs in the pooled egg samples from Bangun and Tropodo/Sumberwuluh group are presented in Figures 9 and 10. While OCDD is the most prevalent congener in the eggs from Bangun, 2,3,4,7,8 PeCDF is the most prevalent congener in the eggs from Tropodo. In the sample from Sumberwuluh 2,3,7,8 TCDF also has a high ratio. The eggs from Tropodo/Sumberwuluh also had proportionally higher levels of HxCDF and HpCDF congeners in comparison with the eggs from Bangun. Dibenzofuran congeners were more prevalent in the egg samples from Tropodo/Sumberwuluh than in those from Bangun.

Figure 11 shows congener profiles for air samples from open burning versus a municipal solid waste incinerator from a study in China. Congener profiles from previous IPEN egg studies (DiGangi and Petrlik 2005, Petrlik, DiGangi et al. 2005, Petrlik, Lobanov et al. 2005) were used for comparison and are shown in Figures 12 and 13.

Figure 9: *PCDD/Fs congener proportions in pooled free-range chicken eggs from Bangun.*

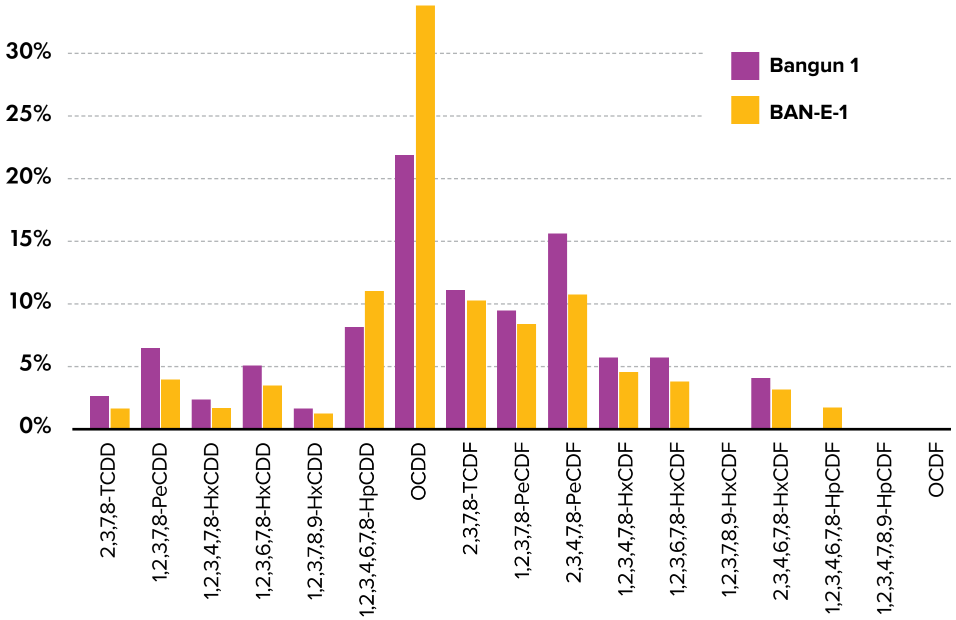


Figure 10: *PCDD/Fs congener proportions in pooled free-range chicken eggs from Tropodo and Sumberwuluh.*

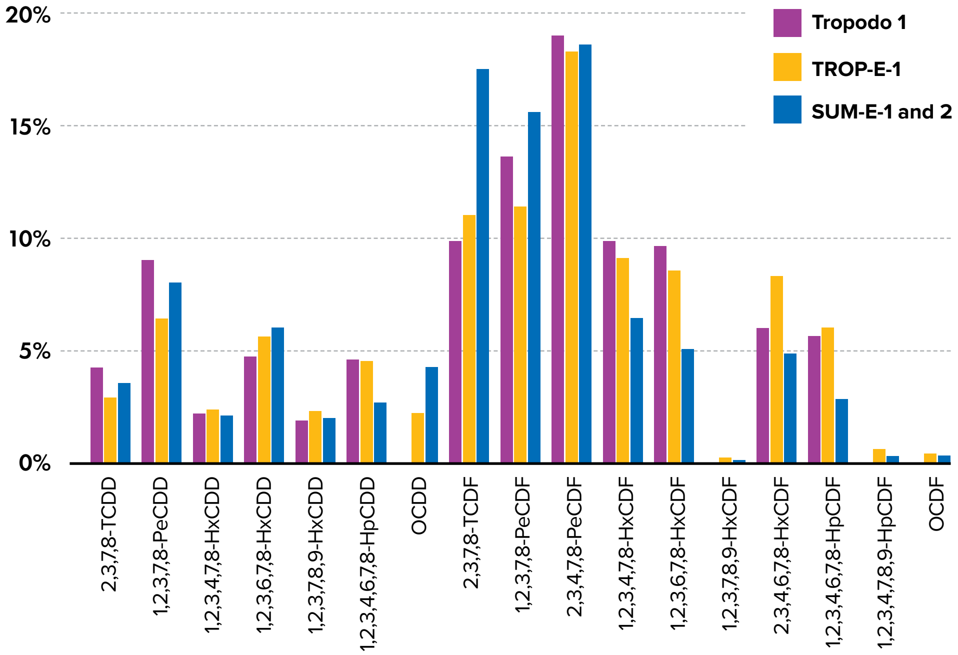


Figure 11: *Congener patterns for open burning of waste and municipal solid waste incineration as observed in air samples, expressed as a percentage relative to dioxin concentration (PCDD/Fs). Source: .(Xu, Yan et al. 2009).*

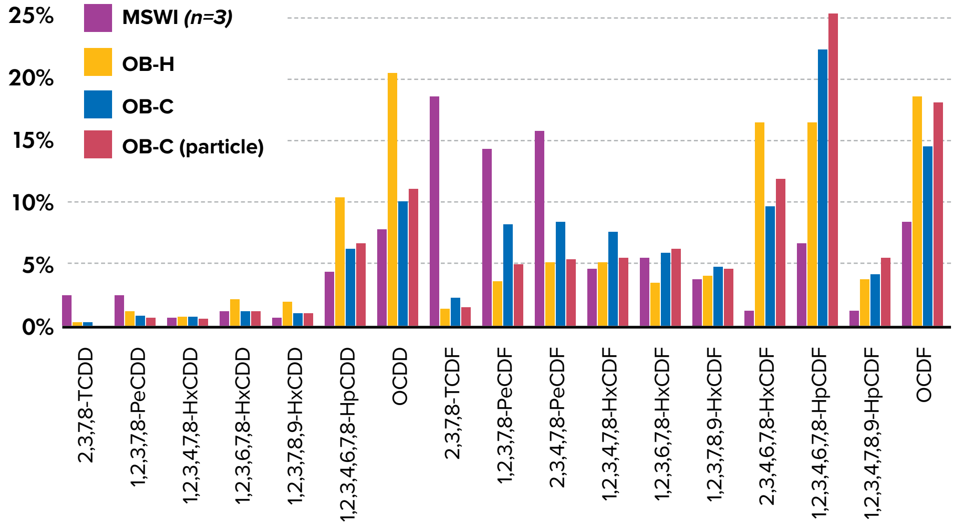


Figure 12: *PCDD/Fs congener profile for pooled free-range eggs from the vicinity of a municipal waste dumpsite in Bolshoi Trostenec, Belarus. (DiGangi and Petrlik 2005).*

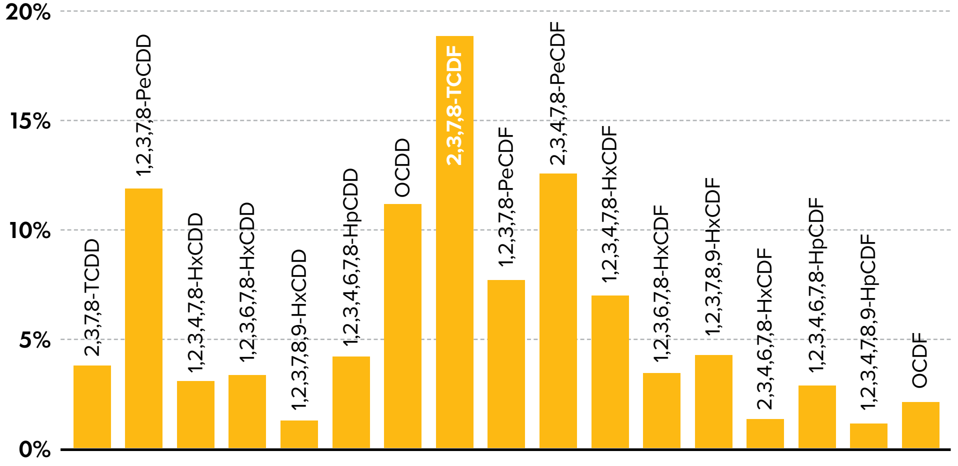
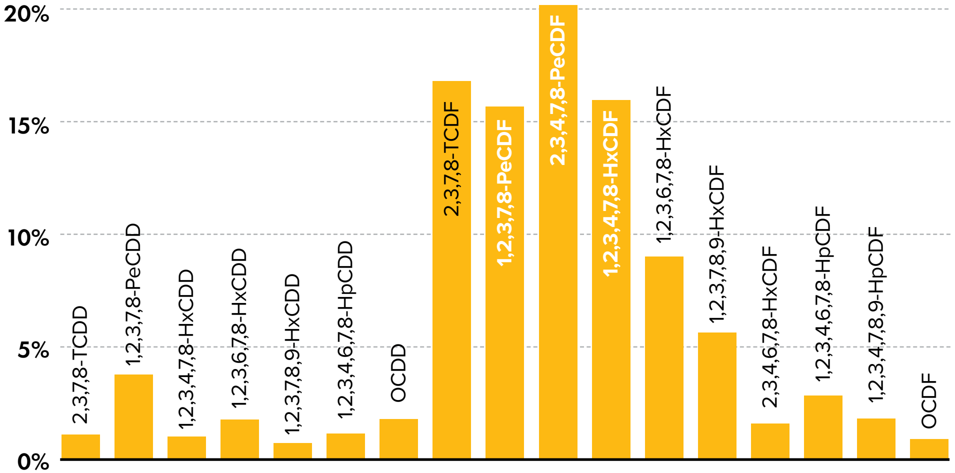
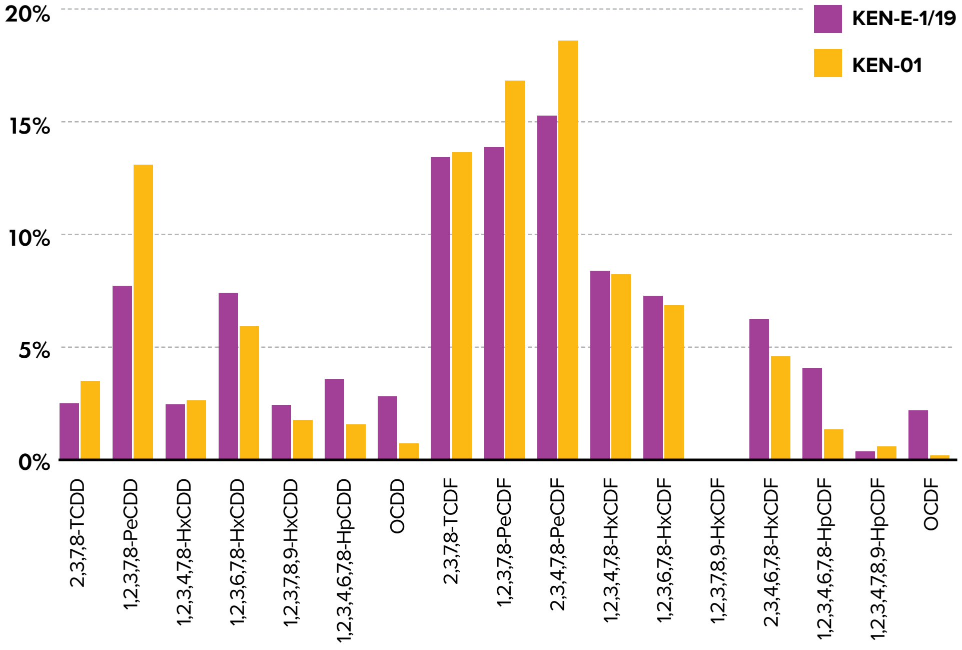


Figure 13: *PCDD/Fs congener profile for a pooled free-range eggs sample from Helwan, Egypt. Source (DiGangi and Petrlik 2005).*



Although the PCDD/Fs congener profiles in the samples from Bangun and Tropodo/Submerwuluh are not identical with either one of the presented profiles, there are similarities to different types of profiles for each of the groups. The congener profile for the Tropodo/Sumberwuluh samples is closer to profiles from "closed" facilities, e.g. waste incineration operations or smelters (for example eggs sampled from Helwan, Figure 13) (Petrlik, DiGangi et al. 2005, Yu, Jin et al. 2006). In contrast, the congener profile for the Bangun sample is closer to those influenced by open air burning of wastes as seen in the sample from Bolshoi Trostenec (Figure 12), which is from an area close to a municipal waste dumpsite in Belarus (Petrlik, Lobanov et al. 2005). Access to ash also plays a role in congener profiles, as waste incineration ash shows different dioxin congener patterns than air emissions (Oh, Chang et al. 2002).

Figure 14: *PCDD/Fs congener proportions in pooled free-range chicken eggs from Kendalsari.*



The dioxin congener pattern in eggs from Kendalsari, visible in Figure 14, is very similar to the pattern in eggs sampled from Helwan, Egypt, at Figure 13 (Petrlik, DiGangi et al. 2005), although there is a partial difference in some congener ratios between the two egg samples from Kendalsari (see Figure 14). They were taken from the same chicken owner and the same place but in different years and seasons. The PCDD/Fs congener pattern in eggs from Kendalsari is also very similar to patterns in the group of samples from Tropodo and Sumberwuluh (see Figure 15).

The two pooled egg samples from Tangerang show a slightly different PCDD/Fs congener pattern, although their PCDF/PCDD ratios are very close to each other. While the sample SEM-E-1 had 2,3,4,7,8 PeCDF as the dominant congener, followed by the 1,2,3,7,8 PeCDD and 1,2,3,6,7,8 HxCDD congeners, TAN-ESIN-01 had 1,2,3,4,6,7,8 HpCDD and 1,2,3,4,6,7,8 HpCDF respectively as dominant congeners (Figure 16). The SEM-E-1 sample is closer to the sample from the Agbogbloshie scrapyard, and the TAN-ESIN-01 sample is closer to the sample from the Kenyan e-waste site when compared to African e-waste sites (see Figures 17–20), which, in turn, had

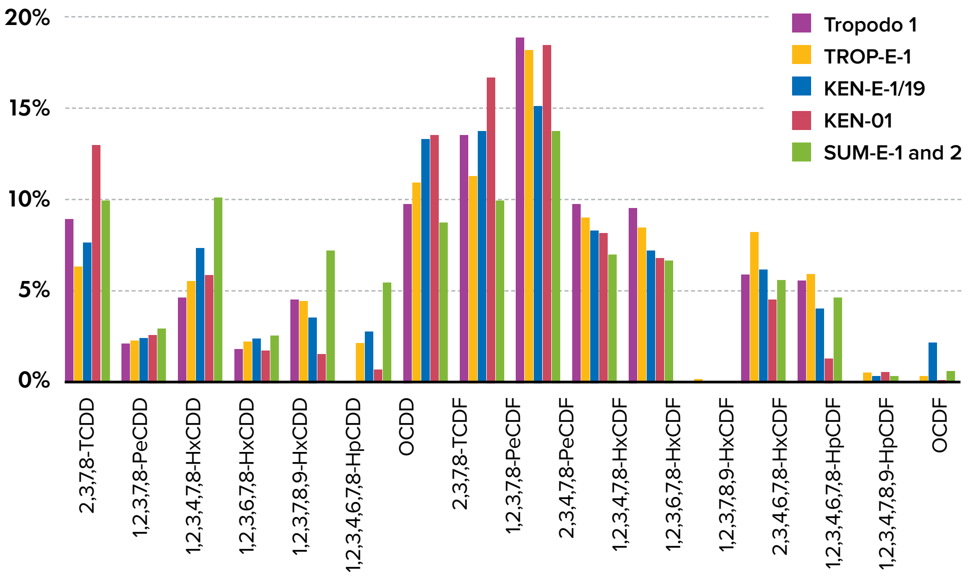


Figure 15: *PCDD/Fs congener ratios for free-range chicken egg samples from Tropodo (Tropodo 1 and TROP-E-1), Kendalsari (KEN-E-1/19 and KEN-01) and Sumberwuluh (sample SUM-E-1 and 2).*

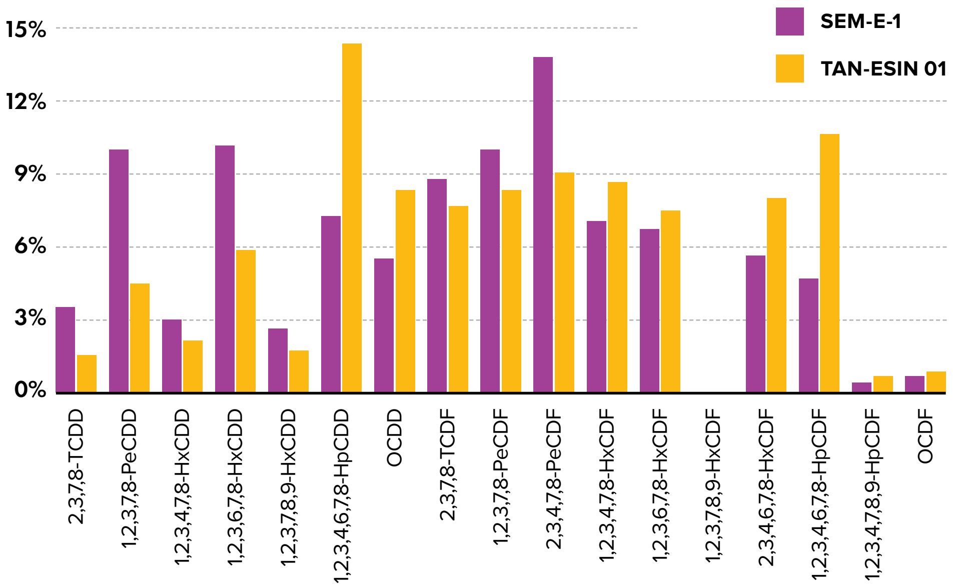
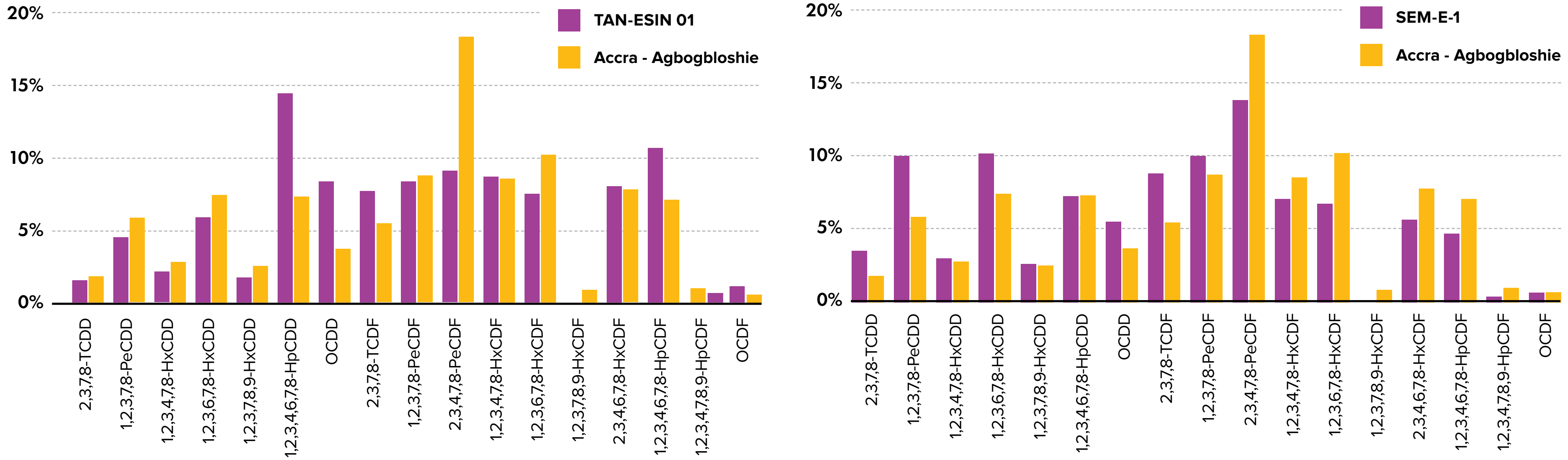
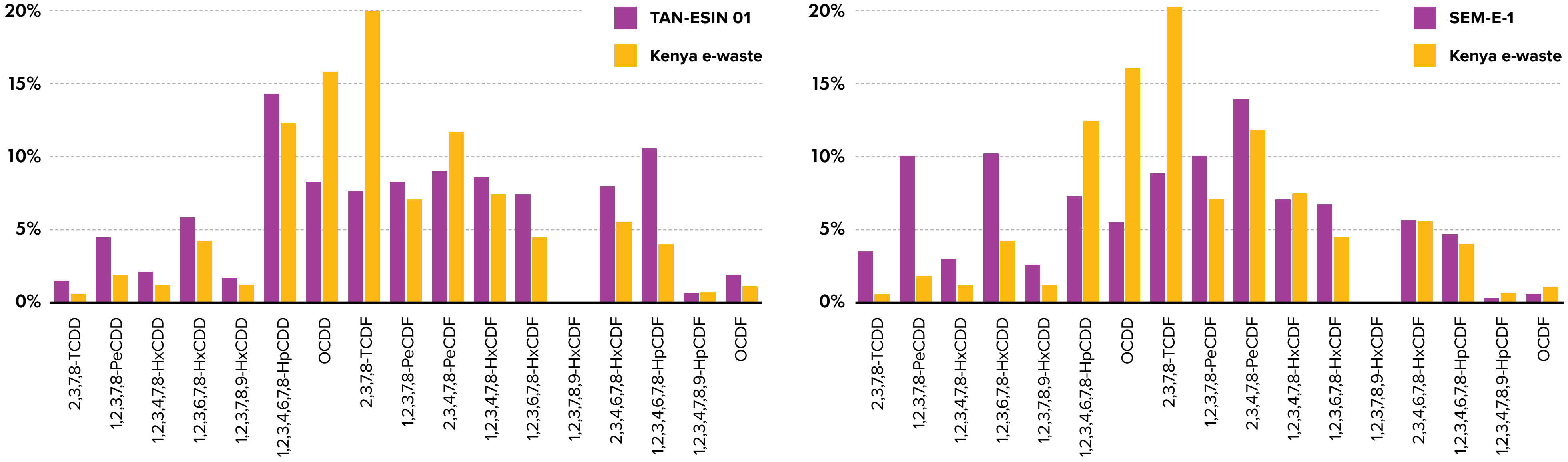


Figure 16: *PCDD/Fs congener ratios in two pooled free-range chicken egg samples from Tangerang marked as SEM-E-1 and TAN-ESIN-01.*



Figures 17 and 18: *Comparison of dioxin patterns between two free-range egg samples from Tangerang (TAN-ESIN-01 and SEM-E-1) and one egg sample from Agbogbloshie, an e-waste scrapyard in Accra, Ghana (Petrlik, Adu-Kumi et al. 2019, SVÚ Praha 2019).*

Figures 19 and 20: *Comparison of dioxin patterns between two free-range egg samples from Tangerang (TAN-ESIN-01 and SEM-E-1) and one egg sample from a Kenyan e-waste site. Source of information for the sample from Kenya: (SVÚ Praha 2020).*



very similar PCDF/PCDD ratios to the samples from Tangerang. Different chemicals in burned wastes as well as different burning conditions and/or even different PCDD/Fs contained in the wastes themselves might be the explanation for this variability. We observed insulation from refrigerators, for example, near the sampling place of sample SEM-E-1, which was absent at another place close to the TAN-ESIN-01 sample, where more non-e-waste plastic was burned constantly (see description in 3.1.1).

4.1.1.4 Hexachlorobenzene (HCB), pentachlorobenzene (PeCB) and hexachlorobutadiene (HCBD)

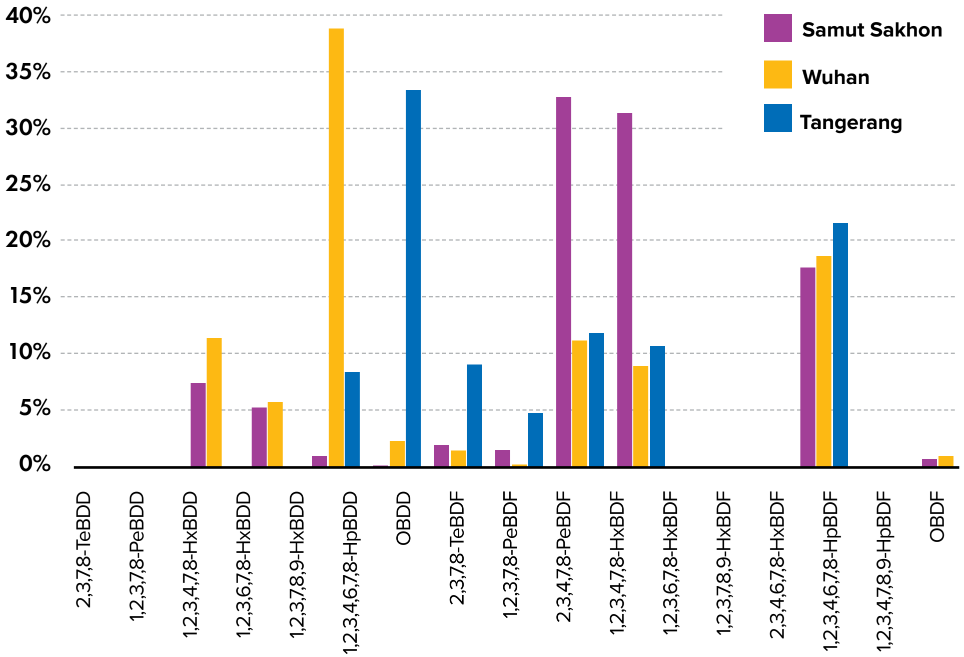
Among the eight free-range egg samples in this study, the highest levels of PeCB and HCB were measured in eggs from Tangerang (sample SEM-E-1) with 3.6 and 6.1 ng g^{-1} fat, respectively. None of the samples were above LOQ for HCBD. The levels of HCB are comparable to those observed in free-range chicken eggs in Kazakhstan (Petrlík, Kalmykov et al. 2016) or in the sample from Yaoundé - Etetak quarter (Petrlik, Adu-Kumi et al. 2019), but they are lower than levels measured in eggs from the e-waste scrapyard in Agbogbloshie, Ghana (Petrlik, Adu-Kumi et al. 2019), or Wuhan, China (Petrlik 2016).

In general, none of the observed levels were extremely high nor exceeded the EU limit values. The same applies to PeCB levels in the eggs in this study.

4.1.1.5 Polybrominated dibezo-p-dioxins and dibenzofurans (PBDD/Fs)

PBDD/Fs are currently not measured in the environment very often, although there are some studies focused on their presence in Southeast Asia, and in particular in China. They were, for example, measured in emissions from a Chinese cement kiln co-processing hazardous waste where PBDD/Fs accompanied deca-BDE (Du Bing, Huang et al. 2011). Simi-

Figure 21: *PBDD/Fs congener patterns in egg samples from Samut Sakhon (Thailand), Wuhan (China) and Tangerang (Indonesia). Source for the data from Samut Sakhon and Wuhan: (Petrlík, Dvorská et al. 2018)*



larly, they were found at an e-waste site in Guiyu, China where high levels of different BFRs were also found: *"The levels of PBDD/Fs in EW disposal area soils were 2.5–17 pg TEQ g^{-1}. 1,2,3,4,6,7,8-HpBDF and OBDF were the dominant congeners, mainly derived from processing, pyrolysis and combustion of BFRs*" (Xu, Tao et al. 2017). Also, other industrial sources such as waste incinerators or metallurgical plants were found to emit PBDD/Fs in China (Du, Zheng et al. 2010). It is obvious from studies in China, Japan, Taiwan and Vietnam that PBDD/Fs are widely present in Asia from pollution sources' releases, and in the environment as well (Suzuki, Someya et al. 2010, Tue, Suzuki et al. 2010, Zhou, Zhao et al. 2014, Gou, Que et al. 2016, Hsu, Arcega et al. 2018). IPEN and Arnika recently found PBDD/Fs in consumer products from recycled e-waste plastic sold in Cambodia and Japan (Petrlík, Adu-Kumi et al. 2019, Budin, Petrlik et al. 2020).

We have found only one study assessing PBDD/Fs in chicken eggs in countries other than China, Thailand and Ghana, from which data are incorporated in Table 5. A report from Ireland showed levels of 0.244-0.415 pg TEQ g^{1} fat. (Fernandes, Tlustos et al. 2009). That is two orders of magnitude lower than the level measured in the free-range chicken egg samples from Wuhan or Samut Sakhon, and three orders of magnitude lower than in the samples from Agbogbloshie. However, the levels of PBDD/Fs in the egg sample from Tropodo and Kendalsari are similar to those measured in Ireland. Eggs sampled in Tangerang showed more than ten times higher levels, but lower than those from Wuhan or Samut Sakhon. The PBDD/Fs congener pattern in an egg sample from Tangerang is very different compared to samples from Wuhan and Samut Sakhon. It shows OBDD as predominant congener followed by 1,2,3,4,6,7,8 HpBDF, which has a comparable percentage in all three discussed samples. Dominance of OBDD congener might be a sign of the open burning of plastics treated with BFRs as the prevailing source of contamination by PBDD/Fs in the egg sample from Tangerang, which is in agreement with the observation of refrigerator insulation present at the site sampled in Tangerang.

4.1.2 NON-DIOXIN-LIKE PCBS

Levels of 6 or 7 indicator PCB congeners represent a potential influence of technical mixtures of PCBs, which is likely not the outcome of unintentional generation but intentional production and use. The EU limit for 6 i-PCB congeners in eggs is set at 40 ng g^{1} fat. None of the samples in this study exceeded that limit value. The highest concentrations of i-PCBs were measured in the eggs from Bangun, which reached levels below 20 ng g^{1} fat, while they were one third or lower than in other samples. Increased levels in Bangun could be explained due to the trucks bringing the plastic waste loads, as oils used in these trucks might still contain technical PCB mixtures and leak. Another explanation could be the content of technical PCBs in the waste brought to Bangun. A recent study from Norway found high levels of i-PCBs present in plastic waste *"even though production and open use of polychlorinated biphenyls (PCBs) have been phased out in Western industrialised countries since the 1980s*" (Arp, Morin et al. 2020).

4.1.3 BROMINATED FLAME RETARDANTS (BFRS)

The laboratory at the Department of Food Chemistry and Analysis of the University of Chemistry and Technology, Prague, routinely measures the following six nBFRs in environmental samples, including the egg samples in this study: 1,2-bis(2,4,6-tribromophenoxy) ethane (BTBPE), decabromodiphenyl ethane (DBDPE), hexabromobenzene (HBB), octabromo-1,3,3-trimethylpheny-1-indane (OBIND), 2,3,4,5,6-pentabromoethylbenzene (PBEB), and pentabromotoluene (PBT). From this group, BTBPE, DBDPE and HBB are monitored more often in environmental samples (Munschy, Héas-Moisan et al. 2011, Mohr, García-Bermejo et al. 2014, Poma, Volta et al. 2014, Vorkamp, Bossi et al. 2015). They were also found in increased levels in samples from Indonesia, together with OBIND more often than other nBFRs measured in this study.

Table 5: *Summarized results of analyses for different BFRs in free-range chicken egg samples from Javanese hotspots (the focus of this study) in comparison with samples from Yaoundé (Cameroon), Agbogbloshie (Ghana), Wuhan (China) and Samut Sakhon (Thailand). Also, results for eggs from supermarkets (background) in Jakarta, Accra, and Beijing, respectively, are included. The table also contains the results of analyses for PBDD/Fs in addition to BFRs. The number of pooled egg samples is in brackets after the locality name. Sources of the data from China, Thailand, Cameroon and Ghana. (Petrlik 2016, Teebthaisong, Petrlik et al. 2018, Hogarh, Petrlik et al. 2019).*

Chemicals	Σ PBDE	Σ HBCD	BTBPE	DBDPE	nBFRs	PBDD/Fs
Units	ng g^{-1} fat					pg TEQ g^{-1} fat
Bangun (n=2)	91 – 1,457	5.2 - 538	NA/2.7	NA/106	NA/124	<LOQ/NA
Tropodo (n=2)	65 – 27,159	<LOQ	NA/32	NA/2,077	NA/2,166	<LOQ/ 0.331
Kendalsari (n=2)	6.2 - 150	<LOQ	<0.1* - 12.2	<5.0*	<LOQ – 12.2	NA/ 0.565
Sumberwuluh (n=1)	8.2	4.5	0.9	<5.0*	0.9	NA
Tangerang (n=1)	321	844	9.7	23.7	33.4	6.9
Yaoundé (n=3)	0.5-2.8	25 - 379	NA	NA	NA	NA
Agbogbloshie (n=1)	1,258	1,961	37.7	<3.3*	38.8	300
Wuhan (n=1)	1,054	NA	51	NA	51	27
Samut Sakhon (n=2)	3.1 - 427	159/NA	< 0.5*	NA/<5.0*	<LOQ	NA/16
Jakarta (supermarket)	1.4	<LOQ	<0.1*	<5.0*	<LOQ	NA
Accra (supermarket)	11	< 12.6*	< 0.3*	<3.3*	<LOQ	< 8.5*
Beijing (supermarket)	0.2	NA	< 0.5*	NA	3.7	< 1.8*

**below LOQ*

Eight out of nine free-range egg samples from Java could be analyzed for three groups of BFRs: 1) eleven congeners of polybrominated diphenyl ethers (PBDEs), 2) three isomers of hexabromocyclododecane (HBCD), and 3) novel BFRs (nBFRs). The results of the analyses for BFRs and PBDD/Fs are summarized in Table 5. For comparison, there are also the results in eggs from Yaoundé, Cameroon; Agbogbloshie, Ghana; Wuhan, China; and Samut Sakhon, Thailand, where PBDD/Fs have previously been found in high levels. The African and Asian sites were chosen for comparison, as they can be similar in nature to those sampled for this study. They are described in studies focused on POPs in eggs from hot spots in Africa (Petrlik, Adu-Kumi et al. 2019), China (Petrlik 2016) and Thailand (Petrlík, Dvorská et al. 2018, Teebthaisong, Petrlik et al. 2018). In Table 5, there are also the results for reference samples from supermarkets in Jakarta, Accra and Beijing.

The highest levels of PBDEs in this study were measured in the eggs from Tropodo and Bangun sampled in October/November 2019. The level in the sample from Bangun is higher than levels observed in eggs from Agbogbloshie, an e-waste scrapyard site, or from Wuhan, in the vicinity of a municipal solid waste incinerator. It is also the seventh-highest ever measured level of PBDEs in free-range eggs. The level exceeding 27,000 ng g^{-1} fat of PBDEs measured in eggs from Tropodo is the second-highest ever measured level in eggs globally (see graph in Figure 22 and data in Table 6). The PBDEs in eggs from Tropodo and Bangun were in the same range as in egg samples from e-waste sites in China.

Interestingly, there is also a big difference between the levels in eggs sampled earlier in 2019 and those from October/November at the same locations. The eggs from Bangun were even from the same chicken owner. It shows that there must have been new waste containing high levels of PBDEs brought to Bangun and afterward incinerated in Tropodo. The temperature in the ovens of the tofu factories is not high enough to destroy POPs like PBDEs, so they most likely bind to particulate matters and fall to the ground in the vicinity of the factories. A relatively high level of PBDEs was also found in soil/ash samples from Bangun (see Table 13 and subchapter 4.4.1).

A relatively high level of 321 ng g^{-1} fat of PBDEs was measured in the sample SEM-E-1 from Tangerang, and it is among the twenty highest levels ever found in free-range eggs. That concentration is higher than the highest level previously measured in eggs from Balkhash, Kazakhstan (Petrlik, Kalmykov et al. 2017), a location with several car wrecks on site, and it is very close to the highest level previously measured in eggs from Arusha, Tanzania (Polder, Müller et al. 2016).

There is a big difference between the samples from Tropodo and Bangun and the sample from Tangerang in their PBDEs congener patterns. The "older" PBDEs (a sum of 7 PBDEs)[17] are dominant in the eggs from Tangerang, while it is decaBDE (BDE 209) that dominates the samples taken in Tropodo or Bangun during October/November 2019. However, the eggs from Tropodo had the second highest sum of 7 PBDEs among the samples in this study (see Table 3) and the sum of 7 PBDEs dominated in the earlier sample from May 2019. The levels of the sum of 7 PBDEs were 181 and 143 ng g^{-1} fat respectively in the samples SEM-E-1 (Tangerang) and TROP-E-1 (Tropodo). It seems that plastic parts or insulation from obsolete electronics probably was the source of contamination of the free-range eggs from the sampled site in Tangerang in November, 2019, and also that the contamination sources in the waste brought there changed rapidly throughout the year in Bangun and Tropodo.

Table 6: *Levels of PBDEs in ng g^{-1} fat measured in free-range chicken eggs in different studies worldwide with a sum of PBDEs above 30 ng g^{-1} fat. Only those samples which were also analyzed for decaBDE (congener BDE 209) have been included. There are two examples of samples from wild birds in Antarctica and some from duck or goose eggs included as well for comparison. All samples other than chicken eggs are marked by specification of the bird species in parentheses after the name of the locality.*

Country (year)	Locality	PBDEs in ng g^{-1} fat	Source of information
Mexico (2004)	Coatzacoalcos	31	(Blake 2005)
Antarctica (2009)	King George Island (chinstrap penguin)	33	(Yogui and Sericano 2009)
Philippines (2004)	Aguado	34	Blake 2005
Indonesia (2019)	Tropodo	65	This study
Indonesia (2019)	Bangun (Bangun-1)	91	This study
Turkey (2004)	Izmit	107	Blake 2005
Indonesia (2019)	Kendalsari	150	This study
South Africa (2009)	Vanderbijlpark	200	(Quinn 2010)
Kazakhstan (2014)	Balkhash – Rembaza	235	(Petrlik, Kalmykov et al. 2017)
Indonesia (2019)	Tangerang (SEM-E-1)	321	This study
Tanzania (2012)	Kwamrefu	347	(Polder, Müller et al. 2016)
Thailand (2016)	Samut Sakhon	427	(Petrlik, Kalmykov et al. 2017)
Antarctica (2009)	King George Island (south polar skua)	558	(Yogui and Sericano 2009)
China (2011)	Wenling	564	(Qin, Qin et al. 2011)
China (2011)	Wenling (duck)	982	(Labunska, Harrad et al. 2013)
China (2014)	Wuhan	1,054	(Petrlik 2016)
Ghana (2018)	Agbogbloshie	1,258	(Hogarh, Petrlik et al. 2019)
Indonesia (2019)	Bangun (BAN-E-1)	1,457	This study
China (2011)	Taizhou (duck)	1,778	(Labunska, Harrad et al. 2013)
China (2012-2013)	Taizhou	3,620	(Labunska, Harrad et al. 2014)
China (2013)	Guiyu (goose)	7,500	(Zeng, Luo et al. 2016)
China (2010)	Quingyuan, Guangdong,	14,100	(Zheng, Wu et al. 2012)
Indonesia (2019)	Tropodo	27,159	This study
China (2013)	Guiyu	46,000	(Zeng, Luo et al. 2016)

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[17] Those seven congeners are BDE 28, BDE 47, BDE 99, BDE 100, BDE 153, BDE 154, and BDE 183

HBCD was measured at high levels of 844 and 538 ng g^{-1} fat in the free-range eggs from Tangerang and Bangun, respectively, both sampled in November 2019. The eggs from Tropodo, Kendalsari and Sumberwuluh, as well as the earlier sample from Bangun, contained levels of HBCD that were either quite low or even below LOQ. The levels of HBCD in the eggs from Tangerang (sample SEM-E-1) and Bangun (sample BAN-E-1) are among the highest levels ever measured in eggs globally. They are higher than the level found in eggs from the vicinity of the municipal waste dumpsite in Baskuduk, Kazakhstan (430 ng g^{-1} fat), but lower than the high level found in chicken eggs from a commercial farm bought in a supermarket in Karaganda, Kazakhstan. HBCD in the eggs from Bangun reached almost half of the concentration observed in eggs from the Agbogbloshie e-waste scrapyard or in the commercial egg sample from Germany(Hiebl and Vetter 2007).

The contamination of eggs with HBCD is related to insulation foams treated with this flame retardant. Insulation from refrigerators was observed near the site where the eggs were sampled in Tangerang. Another potential source of contamination might be polystyrene foam used in obsolete electronic devices or in their packaging (Abdallah, Sharkey et al. 2018). Polystyrene foams were present at the plastic waste dumpsite in Bangun, and can possibly have been present in Tangerang as well.

An increased level of 32 ng g^{-1} fat was measured in a sample from Tropodo analyzed for for BTBPE[18]. This is comparable to the findings in eggs from Agbogbloshie (38 ng g^{-1} fat) or Wuhan (51 ng g^{-1} fat) (Petrlik 2016, Petrlik, Adu-Kumi et al. 2019). They exceeded the background samples by at least two orders of magnitude (see Table 5). A study from Tanzania found four times lower levels of BTBPE in eggs from the Arusha area. An extremely

Table 7: *Levels of HBCD in ng g^{-1} fat measured in chicken or goose eggs in different studies worldwide, above 50 ng g^{-1} fat. Those other than free-range chicken egg samples are marked in parentheses after the name of the locality.*

Country	Locality	HBCD in ng g^{-1} fat	Source of information
Tanzania (2012)	Arusha	63	(Polder, Müller et al. 2016)
Uruguay (2004)	Minas	89	(Blake 2005)
Slovakia (2004)	Kokshov – Baksha	89	(Blake 2005)
Mexico (2004)	Coatzacoalcos	91	(Blake 2005)
China (2013)	Guiyu (goose)	110	(Zeng, Luo et al. 2016)
South Africa (2009)	Vanderbijlpark	136	(Quinn 2010)
Thailand (2016)	Samut Sakhon	159	(Petrlik, Teebthaisong et al. 2017)
Kenya (2004)	Dandora	160	(Blake 2005)
Thailand (2016)	Map Ta Phut	184	(Petrlik, Teebthaisong et al. 2017)
Kazakhstan (2014)	Balkhash – Rembaza	225	(Petrlik, Kalmykov et al. 2017)
China (2010)	South China	350	(Zheng, Wu et al. 2012)
Cameroon (2018)	Yaoundé	379	(Petrlik, Adu-Kumi et al. 2019)
Kazakhstan (2016)	Baskuduk	430	(Petrlik, Kalmykov et al. 2017)
Indonesia (2019)	Bangun	538	This study
Indonesia (2019)	Tangerang (SEM-E-1)	844	This study
Kazakhstan (2015)	Karaganda, supermarket (commercial)	1,036	(Petrlik, Kalmykov et al. 2017)
Ghana (2018)	Agbogbloshie	1,961	(Hogarh, Petrlik et al. 2019)
Germany (2007)	Bavaria (commercial)	2,000	(Hiebl and Vetter 2007)
China (2013)	Guiyu	7,600	(Zeng, Luo et al. 2016)
Kazakhstan (2016)	Shetpe	18,321	(Petrlik, Kalmykov et al. 2017)

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[18] BTBPE stands for 1,2-bis(2,4,6-tribromo-fenoxy)ethane. It is one of the family of novel brominated flame retardants used e.g. in electronics where it has replaced PBDEs. Its accumulation in the eggs highlights the need for more detailed screening of new retardants used as alternatives replacing PBDEs for their potential properties similar to those of POPs. Otherwise we will continue to repeat the same mistake and continue to use new POPs to replace other POPs, which is not the intention of the Stockholm Convention.

Figure 22: *Graph showing levels of PBDEs in ng g⁻¹ fat measured in free-range chicken or duck eggs in different studies worldwide with a sum of PBDEs above 30 ng g⁻¹ fat. Only those which included decaBDE (congener BDE 209) were included. There are two examples of wild birds from Antarctica included as well for comparison. Specific data and sources of information can be found in Table 6.*

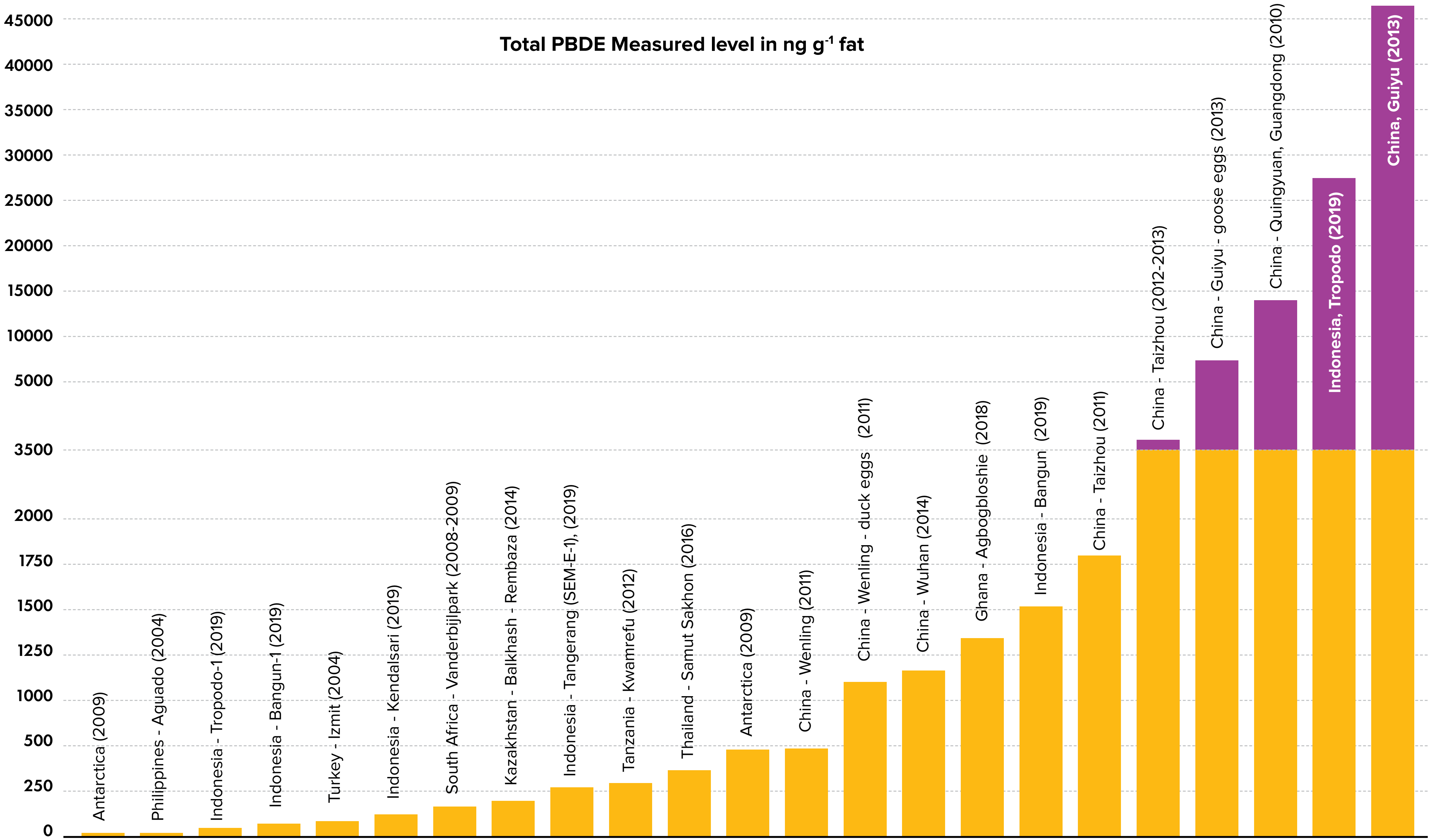
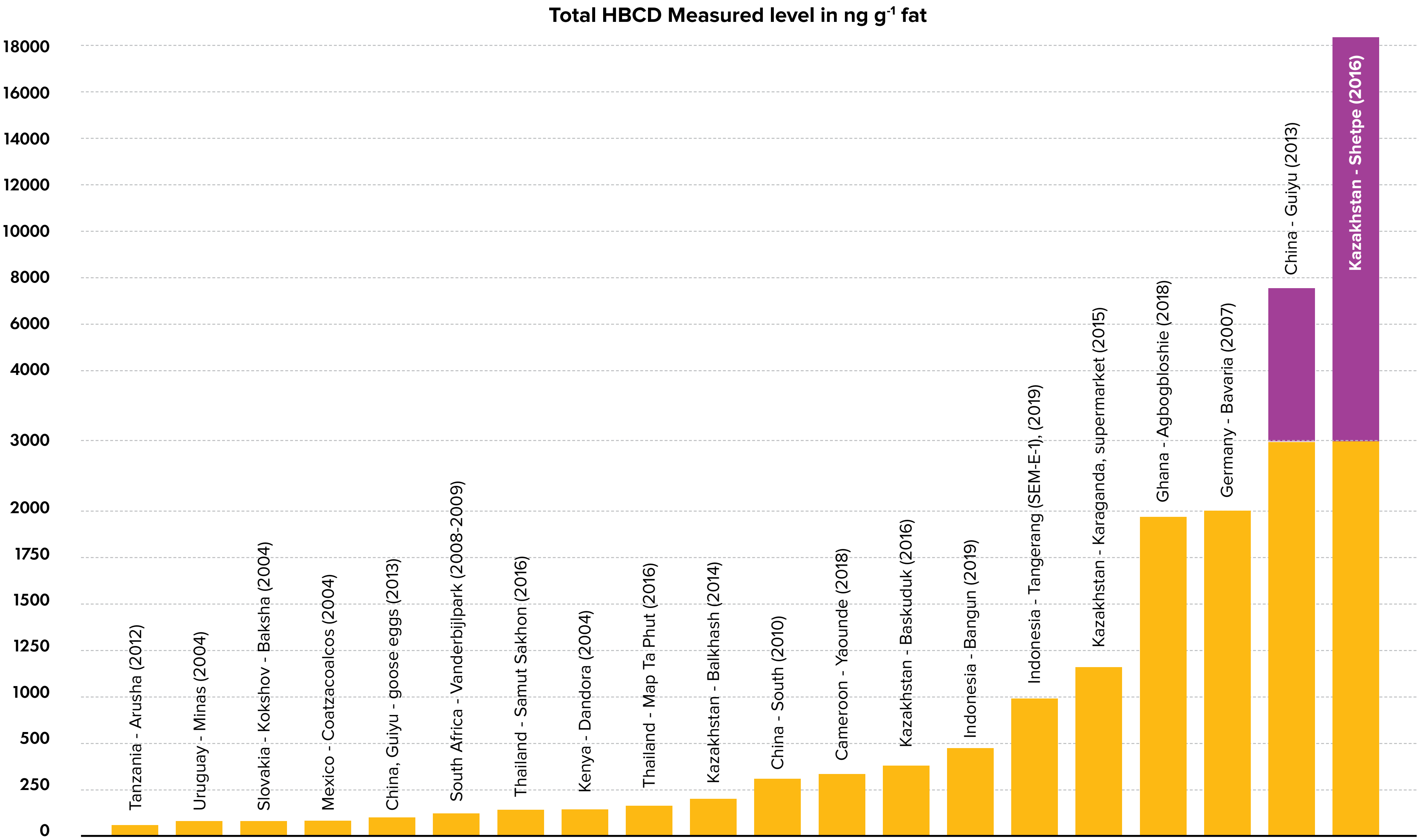


Figure 23: *Graph showing levels of HBCD in ng g⁻¹ fat measured in chicken or goose eggs in different studies worldwide with a sum of HBCD isomers above 50 ng g⁻¹ fat. Specific data and sources of information can be found in Table 7.*



high level above 2,000 ng g^{-1} fat of DBDPE[19] for another nBFR was measured in the same free-range egg sample from Tropodo as BTBPE. An increased level of DBDPE (106 ng g^{-1} fat) was also measured in the egg sample from Bangun. This shows, together with a very high level of PBDEs, that most likely a larger volume of e-waste plastic or other items treated with BFRs was brought to the plastic waste yard in Bangun and then burned in Tropodo. DBDPE was below LOQ in previously studied localities in Kazakhstan, Thailand, China or Africa (Petrlik, Kalmykov et al. 2017, Petrlik, Adu-Kumi et al. 2019) or found in a much lower level of 6 ng g^{-1} fat in eggs from Ireland (Tlustos, Fernandes et al. 2010).

BTBPE was together with HBCD *"detected with the highest frequency and abundance in the global atmosphere,"* including in the location of passive air sampling in Indonesia in 2005-2006 (Lee, Sverko et al. 2016). Air transport can constitute a significant contribution to the BTBPE levels measured in eggs from hotspots in this study.

A sample from Tropodo contained another two nBFRs, HBB and PBT at levels of 3 and 0.7 ng g^{-1} fat respectively, as well as an increased level of 53 ng g^{-1} fat of OBIND.[20]

4.1.4 SHORT-CHAIN CHLORINATED PARAFFINS (SCCPs)

All eight pooled egg samples contained SCCPs at a level equal to or above LOQ[21], including the reference sample from a supermarket in Jakarta. The reference sample in this study coming from the supermarket belonged to those that had higher levels above 100 ng g^{-1} fat (see Table 3). The highest level of 161 ng g^{-1} fat was measured in eggs from Kendalsari, although it was not much higher than the level of 136 ng g^{-1} fat measured in the supermarket reference sample. Other free-range chicken egg samples from dumpsite locations in Kazakhstan or Ghana had 10- to 20-fold higher levels, but they were at almost the same level as in samples from dumps in Yaoundé, Cameroon, and from a metallurgical site in Samut Sakhon, Thailand. (Adu-Kumi, Petrlík et al. 2019).

All eggs from the Javanese locations in this study were well below the minimum levels measured in eggs from one study in South China. The total concentrations of SCCPs in eggs ranged from 477 to 111,000 ng g^{-1} fat from an e-waste-polluted area in South China (Zeng, Huang et al. 2018). The level of 2,067 ng g^{-1} fat of SCCPs in eggs from Agbogbloshie, Ghana, was higher than the minimum level in eggs from South China.

4.1.5 PER- AND POLYFLUOROALKYL SUBSTANCES (PFASs)

Seven out of nine pooled free-range chicken egg samples from Javanese hot spots were analyzed for a range of 17 PFASs[22], including PFOA, PFOS and PFHxS. The results of the analyses are summarized in Table 3, but not all results for all individual PFASs are shown there. The levels of PFPeA and PFHxA, two of these 17 PFASs, were below LOQ of 0.01 ng g^{-1} fw in all samples.

Linear PFOS (n-PFOS) had the highest levels in all samples, and branched PFOS (br-PFOS) contributed to them mostly as the second highest PFASs substance analyzed in the samples (see Table 3). Sample SEM-E-1 from Tangerang exhibited a more equal presence of all PFASs, although n-PFOS still contributed 29% of the total PFASs content, while in samples from Bangun it was 59% and 79%, respectively. Both isomers of PFOS

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[19] DBDPE stands for decabromodiphenyl ethane. It is another chemical from the family of nBFRs, used mainly in polystyrene foams as well as being a replacement for DecaBDE in electronic wires since the 1990s. Its accumulation in the eggs also highlights the need for more detailed screening of new retardants used as alternatives replacing HBCD and PBDEs for their potential properties similar to POPs.

[20] OBIND stands for octabromo-1,3,3-trimethylpheny-1-indane, which is also another nBFR used as a replacement for older BFRs, such as PBDEs or HBCD.

[21] LOQ for SCCPs was set to 50 ng g^{-1} fat for the used analytical method under the conditions for these measurements.

[22] A list of the 17 PFASs included in the analysis: PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUdA, PFDoA, PFTrDA, PFTeDA, PFBS, PFHxS, br-PFOS, L-PFOS, PFDS, PFOSA

represented 68% and 96%, respectively, in egg samples from Bangun, which also had the highest levels of PFASs measured out of the egg samples in this study of 26 and 97 ng g^{-1} fw, respectively, followed by eggs from Tangerang, although their level of 6.2 ng g^{-1} fw is much lower.

If we compare the data obtained from previous research, the levels of PFOS measured in eggs from the rural Bangun dump site are comparable to levels from industrialized areas in European countries, in that they are not directly contaminated by production of perfluorinated compounds, e.g. in eggs from Belgium and Netherlands (D'Hollander W 2011, Zafeiraki, Costopoulou et al. 2016). This fact demonstrates the high impact of waste imported to Bangun, which is most likely the source of the PFASs contamination there. PFOS-related substances have been used in the packaging and paper industries in both food packaging and commercial applications to impart grease, oil and water resistance to paper, paperboard and packaging substrates (KemI 2004).

PFDA was measured in free-range chicken eggs at levels of 0.01 in eggs from Kendalsari up to levels of 0.7 and 0.9 ng g^{-1} fw in eggs from Bangun and Tangerang, respectively. The highest level of 1.7 ng g^{-1} fw was measured in an egg sample from Bangun from May 2019. Similarly, the highest level of PFDoA of 2.3 ng g^{-1} fw was measured in the same egg sample from Bangun, and levels of 1.5 and 1 ng g^{-1} fw in samples from November 2019, from Bangun and Tangerang, respectively. PFDA and PFDoA in the egg samples from the supermarket were below LOQ.

4.1.6 BACKGROUND LEVELS OF POPs IN EGGS

The approach to establishing background levels of POPs in eggs varies among different studies. It is difficult in the current world to find remote sites without any substantial influence of human activity, which is why it was established to use supermarket eggs from large covered chicken farms (sometimes called "battery farms") where poultry does not have access to contaminated soil, as background level samples (Malisch, Schmid et al. 1996, Dvorská 2015).

We sampled chicken eggs from a supermarket in Jakarta from chickens raised on a farm without access to open-air space, in order to obtain information about background levels of POPs in chicken eggs. The results of the analyses of this sample are presented in Table 3. The levels of POPs in this sample were either below the level of quantification (LOQ) of the used analytical methods for most of the POPs, or it was much lower for PCDD/Fs, PCBs (DiGangi and Petrlik 2005, Petrlik, Teebthaisong et al. 2018), and PBDEs (Petrlik 2016). Only in the case of SCCPs (Adu-Kumi, Petrlík et al. 2019) was it higher compared to those observed in the background samples from other studies of POPs in chicken eggs.

The level of PCDD/Fs measured in eggs from the supermarket in Jakarta was one or two magnitudes lower than in egg samples from supermarkets used as a reference in other countries or studies (Petrlik, Teebthaisong et al. 2018, Petrlik, Arkenbout et al. 2019). It is also visible from comparison with an egg sample from Bangkok (see Table 3).

4.2 DIETARY INTAKE OF SELECTED POPs THROUGH CONSUMPTION OF FREE-RANGE CHICKEN EGGS FROM JAVANESE HOTSPOTS

The egg share in total food consumption in Indonesia in 2007 was close to 1% of total food basketper day, according to World Atlas – Food Security data [23] (Knoema 2012), and it has risen by approximately one quarter of its total amount per day (12 g per person per day) every five years. That would mean that for 2017 consumption it would be about 18 g per person per day, if the trend remained. The assumption for 2016 was 470 g of eggs per person, per month, according to the World Food

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[23] The food consumption refers to the amount of food available for human consumption as estimated by the FAO Food Balance Sheets. However the actual food consumption may be lower than the quantity shown, as food availability depends on the magnitude of waste and losses of food in the household. Food consumption per person is the amount of food, in terms of quantity, for each individual in the total population. Food from eggs relates to the quantity of eggs used also for preparation of food such as bakery products.

Table 8: *Summarized results of the calculation of dietary intake of selected POPs by eating half an egg (18 g) from chickens raised at some Javanese hot spots, or eggs bought in the supermarket in Jakarta from chickens raised at a large commercial farm. Half of a chicken egg is the approximate current average consumption per person per day in Indonesia, based on calculations from available data (Knoema 2012, WFP 2017). For this calculation, zero was taken as measured in the concentration in eggs when the level of certain congeners of PCDD/Fs and dl-PCBs was measured as below LOQ; in the case of PBDD/Fs it was not calculated for samples where levels were below LOQ.*

Locality	Bangun		Tropodo		Kendalsari		Sumberwuluh	Tangerang		Jakarta
Sample	**Bangun 1**	**BAN-E-1**	**Tropodo 1**	**TROP-E-1**	**KEN 01**	**KEN-E-1/19**	**SUM-E-1 and 2**	**SEM-E-1**	**TAN-ESIN-01**	**JAK-SUP**
Total content of toxic chemical(-s) in one egg (35 g)										
Total PCDD/F + dl-PCBs (pg TEQ g^{-1} fw)	1.8	1.4	34.8	23.9	23.1	8.6	1.8	11.6	3.8	0.0
PBDD/Fs (pg TEQ g^{-1} fw)	N/A	N/A	N/A	0.046	N/A	0.081	N/A	1.12	N/A	N/A
sum of PBDEs ng g^{-1} fw	11.8	138	9.8	3,772	1.7	21.4	1.2	52.0	N/A	0.13
209-BDE (decaBDE) ng g^{-1} fw	7.1	120	0.6	3,418	N/A	10.7	1.0	12.4	N/A	0.05
PFOS (ng g^{-1} of fw)	17.7	92.4	1.0	0.1	N/A	0.1	0.3	2.5	N/A	0.01
Dietary intake per kg of body weight for an adult person (58 kg on average) by eating half egg (18 g) per sample										
Total PCDD/F + dl-PCBs (pg TEQ kg^{-1} bw)	0.56	0.43	10.80	7.41	7.18	2.68	0.57	3.61	1.18	0.0001
PBDD/Fs (pg TEQ kg^{-1} bw)	N/A	N/A	N/A	0.01	N/A	0.03	N/A	0.35	N/A	N/A
sum of PBDEs (ng kg^{-1} bw)	3.67	42.7	3.03	1,171	0.53	6.63	0.36	16.1	N/A	0.041
209-BDE (decaBDE) (ng kg^{-1} bw)	2.19	37.1	0.19	1,061	N/A	3.31	0.31	3.86	N/A	0.015
PFOS (ng kg^{-1} bw)	5.48	28.67	0.32	0.04	N/A	0.04	0.08	0.76	N/A	0.0031
Exceedance of total daily tolerable intake when eating half an egg (18 g) per day										
PCDD/Fs + dl-PCBs (EFSA 2018)[1]	2.24	1.71	43.20	29.65	28.74	10.73	2.27	14.45	4.73	0.0004
PCDD/Fs + dl-PCBs (WHO 2005)[2]	0.28	0.21	5.40	3.71	3.59	1.34	0.28	1.81	0.59	0.00005
PFOS (EFSA 2018)[3]	3.04	15.93	0.18	0.02	N/A	0.02	0.04	0.82	N/A	0.002

N/A = not applicable; [1] 0.25 pg TEQ kg^{-1} bw ; [2] 2 pg kg^{-1} bw .; [3] 6 ng kg^{-1} bw

Programme, which means approximately 16 g of eggs per person, per day (WFP 2017).. If we count 35 to 40 g per one free-range chicken egg (the typical weight of free-range chicken eggs in Indonesia) as the average weight, it would mean that the consumption of half of such an egg, or a little bit less than that, per person per day, is the general consumption pattern for the Indonesian population these days.

We calculated the dietary intake for the following groups of contaminants per day: 1) PCDD/Fs plus dl-PCBs; 2) PBDD/Fs; 3) PBDEs, and 4) PFOS. The calculation was made by using measured levels of certain chemicals per gram of fresh weight egg and a calculation of the daily intake through consumption of half an egg per day (18 grams of egg weight). An average body weight was taken from information about average human body weight in different parts of the world, available from Wikipedia (Walpole, Prieto-Merino et al. 2012, Wikipedia 2020c). The average body weight of 58 kg for an adult person in Asia was applied. The results are summarized in Table 8.

The results were then compared with available information about daily intake of the evaluated chemicals. They are discussed in subchapters 4.2.1 – 4.2.3 for each of the evaluated POPs. Calculations for PCDD/Fs plus dl-PCBs, and for PFOS, were also compared with tolerable weekly intake (TWI) suggested by EFSA and/or WHO. For PBDEs no TWI was established (JECFA 2006, WHO/FAO 2006).

4.2.1 PCDD/Fs AND DL-PCBs

An adult eating just one egg from a free-range chicken foraging in the vicinity of the tofu factory in Tropodo, at the plastic waste yard in Tangerang, and in the neighborhood of aluminum smelters in Kendalsari, would exceed their tolerable daily intake (TDI) for dioxins by 57- to 84-fold, 28-fold and 21- to 56-fold respectively (EFSA CONTAM 2018a). The typical daily egg consumption per person in Indonesia is less than one egg per day. The calculations for the real dietary intake are presented in Tables 8 and 9.

Table 9: *The total number of eggs, based on the concentrations of PCDD/Fs and dl-PCBs, needed to be consumed in order to reach the tolerable daily dose derived from TWI, suggested by the WHO at a level of 14 pg WHO-TEQ kg^{-1} bw (European Commission 2001, van den Berg, Birnbaum et al. 2006, Gies, Neumeier et al. 2007) and/or by the revised EFSA document from 2018, a level of 2 pg WHO-TEQ kg-1 bw per week (EFSA CONTAM 2018a). The calculation was made for average adult person in Asia, with a weight of 58 kg (Wikipedia 2020c).*

Locality	**Bangun**		**Tropodo**		**Sumber-wuluh**
Sample	**Bangun 1**	**BAN-E-1**	**Tropodo 1**	**TROP-E-1**	**SUM-E-1 and 2**
PCDD/Fs + DL-PCBs (pg WHO-TEQ in one egg)	63	48	1218	836	64
Number of eggs to reach 116 pg WHO-TEQ per day	1.83	2.41	0.10	0.14	1.81
Number of eggs to reach 14.5 pg WHO-TEQ per day	0.23	0.30	0.01	0.02	0.23
Locality	Kendalsari		Tangerang		Jakarta
Sample	**KEN 01**	**KEN-E-1/19**	**SEM-E-1**	**TAN-ESIN-01**	**JAK-SUP**
PCDD/Fs + dl-PCBs (pg WHO-TEQ in one egg)	810	303	407	133	0.011
Number of eggs to reach 116 pg WHO-TEQ per day	0.14	0.38	0.28	0.87	10,900
Number of eggs to reach 14.5 pg WHO-TEQ per day	0.02	0.05	0.04	0.11	1,363

From the dietary intake of dioxins (PCDD/Fs) and dl-PCBs in half an egg for the pooled egg samples, from different Javanese localities (Table 8), it is evident that the situation is critical in all sampled localities. By the average egg consumption calculated as half an egg (18 g of egg) per day for an adult person weighing 58 kg, people eating free-range eggs would exceed the European Food Safety Authority (EFSA) TDI for chlorinated dioxins by 1.5 to 43-fold. Even the older and less strict TDI of 2 pg TEQ kg^{-1} body weight (European Commission 2001, van den Berg, Birnbaum et al. 2006, Gies, Neumeier et al. 2007) would be exceeded in the case of five samples of eggs by 1.3 to 5.4-fold. The most serious situation was in 2019 in Tropodo, Kendalsari and Tangerang.

According to the calculated results shown in Table 9, the tolerable daily intake for PCDD/Fs and dl-PCBs can be reached by eating 0.01 to 0.02 parts of a free-range egg in Tropodo, or one-quarter of an egg in Bangun or Sumberwuluh, where contamination by dioxins and dl-PCBs is lower than in Tropodo, Tangerang and Kendalsari. In comparison, it would be necessary to eat more than 1,350 eggs from the supermarket in Jakarta to reach the tolerable daily intake for dioxins and dl-PCBs.

Brominated dioxins contribute significantly to the daily intake of dioxin-like acting chemicals in a sample of eggs from Tangerang, which reached one-tenth of the total intake from PCDD/Fs and dl-PCBs (see data in Table 8). PBDD/Fs were also measured at levels above LOQ in eggs from Tropodo and Kendalsari, but their levels were not high, so their contribution to the daily intake is low — ten or more times lower than in Tangerang.

4.2.2 PBDEs

The daily intake of PBDEs from eating eggs from samples taken in Javanese hot spots is shown in the same table as for dioxins (see Table 8). The highest intake of these chemicals was observed in the pooled egg sample from Tropodo, taken in October 2019, with extremely high levels of these BFRs. It also exhibits a very high ratio of decaBDE congener intake.

The second-highest intake was calculated for a sample taken in November 2019 in Bangun, again with a very high contribution of the decaBDE congener. Intakes from other egg samples from Bangun (earlier sample from May 2019), Tropodo (earlier sample from May 2019), Kendalsari and Tangerang also are considerably high, and some even without including the decaBDE congener contribution, like e.g. the sample from Tangerang, which also had the highest contribution of the "older" seven congeners[24] of PBDEs, mostly counted in earlier studies.

For the sake of comparison with other studies, we had to discount the decaBDE congener (BDE 209) contribution to the intake from eggs as those studies were done ten or more years ago and did not include that congener. The intake from the egg sample in Tropodo would be 110 ng kg^{-1} bw. It is almost 28-fold higher than the average total daily intake from the average food basket as calculated by the joint committee of WHO and FAO in 2006, at a level of 4 ng kg^{-1} bw (JECFA 2006, WHO/FAO 2006).

We have to take into account the different PBDE congeners included in these calculations and also the difference in time by more than a decade. Daily intakes of different groups of PBDE congeners from eating free-range chicken eggs from the various studied locations are compared in Table 10. PBDEs intakes were in many studies calculated just for seven different PBDE congeners. The intake of these congeners from the eggs sampled in this study is estimated in Table 11, where there are also results from other studies for comparison.

Another study from China concluded that: *"PBDE concentrations varied among different areas, among which the contamination in Guangdong Province was most serious. Daily intake of ΣPBDEs was 225.1-446.0 ng/d for adults in the Pearl River Delta, which was higher than the intake for those living in the Yangtze River Delta (148.9-369.8 ng/d)"* (Chen, Cao et al. 2014). The estimates in that study include decaBDE (BDE 209) and two other PBDE congeners (BDE 47 and BDE 99) levels in food. The intake from eggs sampled in October 2019 in Tropodo or Bangun were even one or two magnitudes higher than those found in Chinese polluted areas just for decaBDE.

The daily intake of PBDEs at the studied locations in 2019 ranged from equal to more than ten times higher than the total daily intake of PBDEs in Finland, Sweden or Canada more than fifteen years ago. In this comparison, decaBDE and eight other PBDE congeners were not included, in order to make it more comparable to calculations of PBDE intake done between 2001 and 2004 (Ryan and Patry 2001, Lind, Aune et al. 2002, Kiviranta, Ovaskainen et al. 2004).

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[24] Those seven congeners are BDE 28, BDE 47, BDE 99, BDE 100, BDE 153, BDE 154, and BDE 183

***Table 10:** Data show the daily intake of various combinations of PBDE congeners from eating an average amount of eggs in the diet of Indonesians (18 g) from locations in this study in ng per day. Calculation is based on PBDEs levels and fat content measured in eggs (see Table 3)*

Locality	**Bangun**		**Tropodo**		**Sumberwuluh**
Sample	**Bangun 1**	**BAN-E-1**	**Tropodo 1**	**TROP-E-1**	**SUM-E-1 and 2**
Daily intake of 16 PBDEs[25] congeners by eating 18 g eggs	213	2,478	176	67,903	21
Daily intake of decaBDE congener by eating 18 g eggs	127	2,153	11	61,532	18
Daily intake of 7 PBDEs congeners by eating 18 g eggs	45	54	140	356	2.5
Locality	**Kendalsari**		**Tangerang**		**Jakarta**
Sample	**KEN 01**	**KEN-E-1/19**	**SEM-E-1**	**TAN-ESIN-01**	**JAK-SUP**
Daily intake of 16 PBDEs[25] congeners by eating 18 g eggs	31	385	937	N/A	2.4
Daily intake of decaBDE congener by eating 18 g eggs	na	192	224	N/A	N/A
Daily intake of 7 PBDEs congeners by eating 18 g eggs	31	115	529	N/A	2.4

The highest intake of seven PBDE congeners was estimated for eggs sampled in Tangerang at a level of 529 ng per day, which exceeds the general PBDE intake of 90.5 ng per day in the UK estimated in 2002 (Wijesekera, Halliwell et al. 2002), and the highest estimated intake of 61.2 ng per day in China (Li, Zhang et al. 2010) in 2010 by almost 6 and 9-fold, respectively. It has to be taken into account that this comparison is just of eggs from the studied locations, with a total diet or food basket in selected countries. People living in communities at the selected locations most likely also consume other food containing PBDEs. We have also take into account that the limited number of egg samples was taken at studied sites.

***Table 11:** Daily intake of seven PBDE congeners from eating an average amount of eggs in the diet of Indonesians (18 g) from locations in this study (in year 2019), in ng per day, compared to the total daily intakes of PBDEs in various countries at the beginning of this century. The calculation is based on PBDEs levels and fat content measured in eggs (see Table 3).*

Locality	**Bangun**	**Tropodo**	**Kendalsari**	**Tangerang**	**Sumberwuluh**	**Jakarta**	
Daily diet. intake of 7 PBDE congeners from eggs ($ng\ day^{-1}$)	45 / 54	140 / 356	31 / 115	529	2.5	2.4	
Country[1]	Finland	Sweden	Belgium	Canada	UK	ND	China
Year	2004	2002	2007	2001	2002	2003	2010
Total daily intake of PBDEs ($ng\ day^{-1}$)	43	31	23 - 48	44	90.5	13-185*	3.7-61.2[2]

ND = The Netherlands;

[1] Sources of information for country data: : (Ryan and Patry 2001, Lind, Aune et al. 2002, Wijesekera, Halliwell et al. 2002, de Winter-Sorkina, Bakker et al. 2003, Kiviranta, Ovaskainen et al. 2004, Li, Zhang et al. 2010)

[2] Range of daily intakes in different provinces of China;

** big lower and upper bound level; the difference is so high because many PBDE congeners measured below LOQ in food.*

The comparison with PBDEs intake from food raised at certain polluted hotspots looks different of course. For example, for an e-waste site in Zheijang, the total daily intake of seven PBDEs was estimated at a level of 195.9 ng per day. The highest intake of seven PBDEs calculated for the Tangerang sample exceed this intake by 2.7-fold, while the intake of seven PBDEs from eggs in Bangun is one quarter of that level.

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[25] These are all 16 congeners of PBDEs measured in this study: BDE 28, BDE 47, BDE 49, BDE 66, BDE 85, BDE 99, BDE 100, BDE 153, BDE 154, BDE 183, BDE 196, BDE 197, BDE 203, BDE 206, BDE 207 and BDE 209

The daily intake of seven PBDEs from the egg sample taken in Tangerang would reach 9 ng kg^{-1} bw, which is still twice as much as the total dietary intake calculated as the global average by the WHO/FAO in 2006 (WHO/FAO 2006). The eggs sampled in October 2019 in Tropodo would be the contribution of seven PBDEs to the total intake from eggs equal to the global WHO/FAO total intake level; it means 4 ng kg^{-1} bw (JECFA 2006, WHO/FAO 2006). The PBDE intake from dietary sources for adults in the U.S., Spain and the UK was estimated to be 0.9–1.2, 1.2–1.4 and 1.5 ng kg^{-1} bw day–1, respectively (Bocio, Llobet et al. 2003, Harrad, Wijesekera et al. 2004, Schecter, Päpke et al. 2006). A more recent study from China estimated that dietary intakes of PBDEs were 0.76 ng kg^{-1} bw day^{-1} (Zhang, Li et al. 2013).

4.2.3 PFOS

The daily intake of PFOS by eating eggs from samples taken at Javanese hotspots is shown in the same table as for dioxins (see Table 8). The highest intake of these chemicals was observed in the pooled egg sample from Bangun, taken in November 2019 with very high levels of n-PFOS as well as br-PFOS isomers (see Table 3).

An adult eating half an egg per day from a free-range chicken foraging in the vicinity of the Bangun dumpsite would exceed the proposed tolerable daily intake (TDI) of PFOS (EFSA CONTAM 2018b) by 3- and almost 16-fold respectively.

The eggs from Tangerang exhibited the second-highest intake of PFOS among the sampled eggs from Java in this study, and an adult eating one egg from a free-range chicken from the Tangerang plastic waste yard would almost reach the TDI for PFOS, but in reality people are exposed to PFOS from a much wider range of foods and drinks than just eggs (Haug, Salihovic et al. 2010, Noorlander, Leeuwen et al. 2011).

4.3 ASH, SOIL AND OTHER SAMPLES

We also sampled ash, soil or dust at most of the researched sites in order to be able to get a better idea about the overall POPs contamination of the environment. In Sidokampir, we also took samples of a rice crop from a field next to the road built from residual ash from aluminum smelters. Sidokampir is a neighbour community to the Kendalsari village where many small aluminum smelters are located (see 3.2.1). We also took soil samples in the Mbeji Forest, which is a relatively distant and cleaner area of West Java (see description in 3.2.5), to use as a reference sample of soil for comparison with samples taken at hot spots exposed to POPs-releasing activities.

Almost all the ash, dust and soil, as well as the rice crop samples were mixed/pooled samples from several individual ones taken in a way that should give a more representative picture of the contamination level at a certain site (see Table 1). We are aware of the limited number of those samples that we were able to analyze within our study with the resources available to us.

The soil was mostly taken after the removal of a surface layer with roots (approx. 2 cm), then an ~5 cm layer in a square of 5 x 5 cm was taken, if the conditions at the site allowed. Stones and roots were removed, as well as plastic waste parts. The cover layer was also removed from ash piles when the ash was sampled from such sites. A layer of approximately 10 cm was taken from several places.

Pooled samples were taken either in the corners and at the center of the square area or in a row, e.g. in the case of the road. The distance between sampling points varied from one to four meters. Mostly it was two meters if sampled in a row, and four meters if sampled in a square. The number of individual (point) samples presented in each pool is specified in Table 1.

The point samples were mixed and homogenized in a bowl made from stainless steel, and quartation was applied if needed to limit the size of

Table 12: *Overview of results of chemical analyses for POPs in 12 samples of different environmental matrices from Javanese hotspots. Samples were taken in 2018-2019. The levels of POPs are stated in ng* g^{-1} *dry weight if not specified otherwise.*

Locality	Tropodo		Kendalsari		Sidokampir					Bangun	Tangerang	Mbeji Forest
Sample ID	**TROP-A-1**	**TROP-A-2**	**KEN-A-1**	**KEN-AD-1**	**SA1, SA2, SA3**	**SID-D-1**	**SID 01**	**SID-S-1/19**	**RICE 01**	**BAN-S-1**	**TAN-EBUT-01**	**MBEJI-S**
Matrix	**ash**	**ash**	**ash/soil**	**ash/dust**	**ash**	**dust**	**soil**	**soil**	**rice crop**	**soil/ash**	**soil/ash**	**soil**
PCDD/Fs (pg TEQ g^{-1} dw)	1,246	122	17.3	57	484	33	1.07	2.7	0.84	205	14.4	0.71
dl-PCBs (pg TEQ g^{-1} dw)	119	12.2	0.70	4.4	13.1	2.2	0.18	0.47	0.39	88	2.1	0.07
Total PCDD/F + dl-PCBs (pg TEQ g^{-1} dw)	1,365	134	18	61	497	35	1.25	3.2	1.23	293	16.5	0.78
Total PCDD/Fs + dl-PCBs - DR CALUX® (pg BEQ g^{-1} dw)	NA	NA	NA	NA	NA	NA	NA	NA	NA	86	NA	0.5
HCB	34	2.1	4.3	7.9	19.50	2.5	NA	1.12	NA	13.4	1.05	0.08
PeCB	52	4.7	2.0	6.9	NA	1.4	NA	0.80	NA	21.6	1.15	0.16
HCBD	0.04	<0.02	<0.02	<0.02	NA	<0.02	NA	<0.02	NA	0.13	0.08	<0.02
7 PCB	0.66	1.33	0.18	0.51	0.69	0.20	NA	0.02	NA	18.8	0.88	<LOQ
SCCPs	NA	15.4	NA	NA	NA	NA	NA	NA	NA	3247	196	2.4
sum HCH	0.03	<LOQ	0.02	0.03	<LOQ	<LOQ	NA	0.06	NA	0.10	0.35	0.02
sum DDT	0.26	0.20	0.31	0.32	0.88	0.10	NA	0.11	NA	1.89	0.29	0.09
sum HBCD	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	0.00	NA	<LOQ	NA	99	<LOQ	<LOQ
sum of PBDEs	1.13	0.05	0.08	0.35	0.23	0.75	NA	0.57	NA	745	25	0.04
BDE 209 (decaBDE)	<5	<5	<5	<5	<5	<5	NA	<5	NA	701	23	<5
7 BDE congeners	0.54	0.05	0.06	0.28	0.23	0.58	NA	0.45	NA	5.9	0.34	0.04
sum of nBFRs	0.31	0.02	0.03	0.44	0.00	19.5	NA	0.02	NA	899	187	0.07
sum of PFASs	NA	<LOQ	NA	NA	NA	NA	NA	NA	NA	21	<LOQ	<LOQ
L-PFOS	NA	<0.3	NA	NA	NA	NA	NA	NA	NA	5.0	<0.3	<0.3

the pooled sample. The sample size was mostly 500 ml. Samples were kept in dark and cool conditions before their delivery to the laboratories. They were analyzed in the same laboratories and for the same or similar scale of chemicals as the chicken eggs (see 4.1).

The results of the chemical analyses are summarized in Table 12. Their evaluation is in subchapters 4.4.1 – 4.4.3, organized according to the type of pollution hot spots, where a comparison with the levels measured in eggs also can be found. A tofu sample was taken in one of the factories in Tropodo and analyzed for POPs as well. A reference sample of tofu was obtained in a Prague store of bio-quality food (organic certification). Results of their analyses are in Table 15.

4.4 POPs HOTSPOTS CATEGORIES

4.4.1 PLASTIC WASTE YARDS

Indonesia, and Java in particular, have become a destination for plastic waste exports from developed countries, as described in previous studies (GAIA 2019, Ismawati Drwiega, Septiono et al. 2019, Petrlik, Ismawati et al. 2019). In December 2019, we published the results of the analysis of pooled egg samples taken in Bangun at one of the major waste yards, where local community residents sort imported waste next to their houses. It showed high levels of certain POPs measured in samples taken in May 2019 (Petrlik, Ismawati et al. 2019) and led us to further investigations in Bangun and two other sites in Tangerang, located close to the Jakarta airport (see 3.1.1).

The additional pooled free-range chicken eggs were taken at all investigated sites, and two soil samples were taken at sites where plastic waste is sorted, and very often also burned and its residues buried. The soil was partly mixed with the residues of burning, and in the case of the sample from Tangerang, it was almost completely ash. These samples were taken from areas accessible by hens, from which we obtained the egg samples, although they do not necessarily represent whole area where the chickens foraged.

The results of the analyses for PCDD/Fs, PCBs, HCB, PeCB, HCBD, PBDD/Fs, SCCPs, PBDEs, HBCD, nBFRs, and PFASs in the samples from Bangun and Tangerang are summarized in Table 13. They are compared with the reference samples of eggs from the Jakarta supermarket, and of the soil from the Mbeji Forest.

Photo 28: *Chickens foraging on a plastic waste yard in Bangun.*
Photo: Jindrich Petrlik, November 2, 2019.

There were significant levels of a whole range of POPs measured in the free-range chicken eggs from plastic waste scrapyards/dumps in Bangun (see Photo 28) and Tangerang, in particular PCDD/Fs, dl-PCBs and BFRs (HBCD, PBDEs and nBFRs). The highest level of PBDD/Fs measured in eggs from Java were found in sample SEM-E-1 from Tangerang, while the eggs from Bangun had the highest levels of PFASs measured among the eggs from Java. Also, the levels of PeCB, HCB, and ndl-PCBs were one magnitude higher than those observed in the reference sample from the Jakarta supermarket. Only the levels of HCBD were below LOQ, and the levels of SCCPs were either below or the same as in the reference sample.

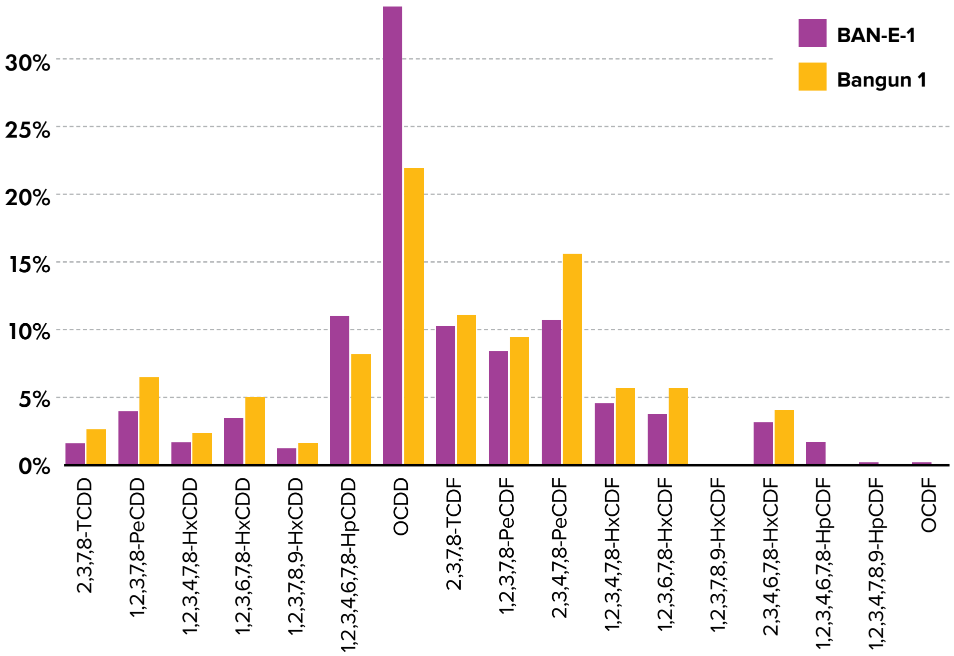
Emissions and ash from the open burning of plastic waste could be the source of dioxin contamination in the eggs from Tangerang, as both egg samples contained levels of PCDD/Fs and dl-PCBs of 72 and 28 pg WHO-TEQ g^{-1} fat, respectively. These levels exceed the EU limit value for eggs (5 pg WHO-TEQ g^{-1} fat) by almost 6- to more than 14-fold and the Indonesian limit value by 12- to 28-fold.

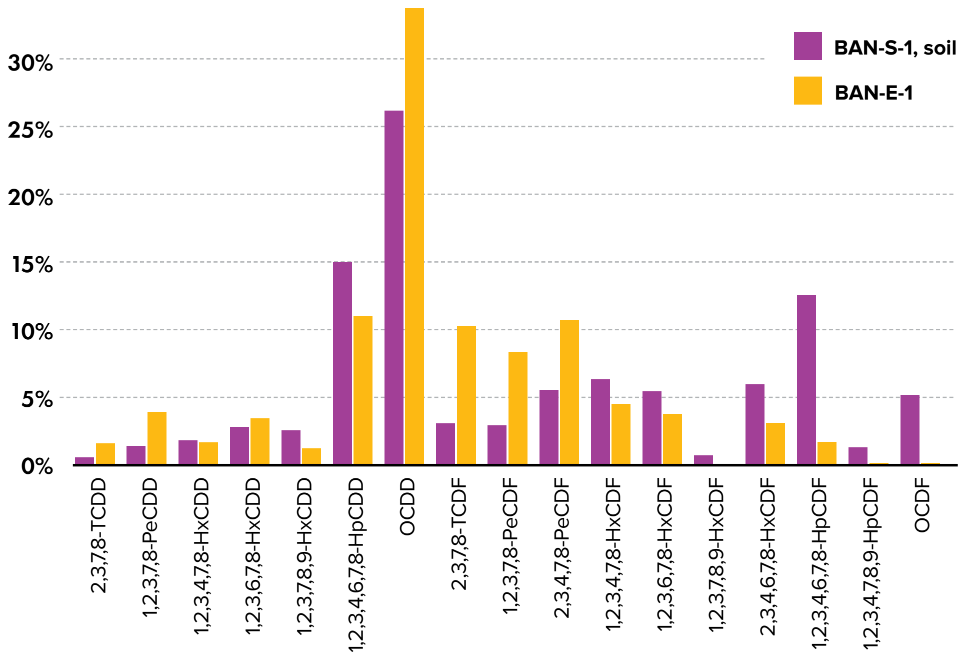
Table 13: *Summarized results of analyses for various POPs in six samples from Bangun and Tangerang.*

Locality	**Bangun**		**Tangerang**		**Jakarta/ref. sample**	**Bangun**	**Tangerang**	**Mbeji Forest - reference site**
Sample ID (eggs)	**Bangun 1**	**BAN-E-1**	**SEM-E-1**	**TAN-ESIN-01**	**JAK-SUP**	**BAN-S-1**	**TAN-EBUT-01**	**MBEJI-S**
Matrix	eggs	eggs	eggs	eggs	Eggs	soil/ash	soil/ash	soil
Fat content (%)	13	9.5	16.2	13.7	9.5	NA	NA	NA
PCDD/Fs (pg TEQ g^{-1} fat/dw)	10.8	9.5	54	20.41	0.0012	205	14.4	0.71
dl-PCBs (pg TEQ g^{-1} fat/dw)	3.1	5.1	17.6	7.41	0.0020	88	2.1	0.07
Total PCDD/F + dl-PCBs (pg TEQ g^{-1} fat/dw)	13.9	14.6	72	28	0.0032	293	16.5	0.78
PBDD/Fs (pg TEQ g^{-1} fat)	< 21.3	NA	6.9	NA	NA	NA	NA	<11.1
HCB	2.7	3.6	6.1	NA	<0.1	13.4	1.1	0.08
PeCB	1.1	2.23	3.63	NA	<0.1	21.6	1.2	0.16
HCBD	< 0.1	<0.1	<0.1	NA	<0.1	0.13	0.08	<0.02
7 PCB	15.4	16.9	3.4	NA	< LOQ	18.8	0.88	< LOQ
6 PCB	12.3	14.0	3.4	NA	< LOQ	NC	NC	NC
SCCPs	153	97	153	NA	136	3,247	196	2.41
Sum HBCD	5.2	538	844	NA	< LOQ	98.97	< LOQ	< LOQ
Sum of PBDEs	91	1,457	321	NA	1.4	745	25	0.04
209-BDE (decaBDE)	54.4	1,265	77	NA	<1.0	701	23	<5
7 BDE congeners	19.4	32	181	NA	1.39	5.9	0.34	0.04
Sum of nBFRs	NA	124	33	NA	< LOQ	899	187	0.07
Sum of PFASs (ng g^{-1} of fresh weight)	26	97	6.2	NA	0.1	21	< LOQ	< LOQ
L-PFOS (ng g^{-1} of fresh weight)	15.4	76	1.8	NA	<0.01	4.99	<0.3	<0.3

dw = dry weight; NA = not analyzed; NC = not calculated; < LOQ = below level of quantification

Figures 24 – 26: *Graphs showing dioxin congener patterns in the pooled egg samples and the soil/ash sample from Bangun.*





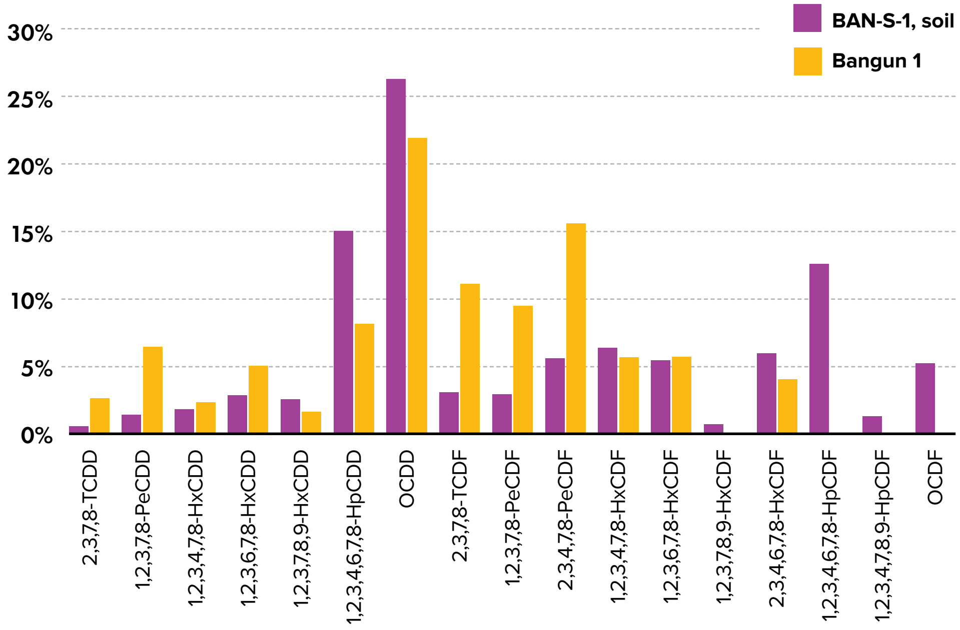
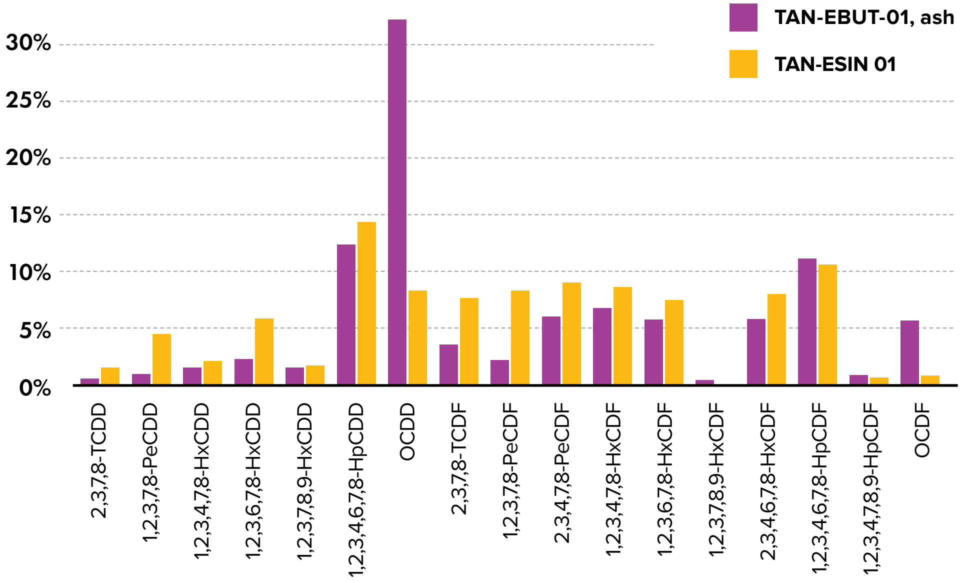


Figure 27: *PCDD/Fs congener patterns in the free-range egg sample and the soil/ash sample from one of the sampled sites in Tangerang.*



The egg samples from Bangun contained, in both cases, levels of PCDD/Fs and dl-PCBs that exceeded the EU standard by almost threefold and the Indonesian standard by sixfold. This is comparable to similar sites with open burning of plastic and other wastes in Yaounde, Cameroon (Petrlik, Adu-Kumi et al. 2019) and Bolshoi Trostenec, Belarus (Petrlik, Lobanov et al. 2005).

The level of 54 pg WHO-TEQ g^{-1} fat of PCDD/Fs measured in the eggs from Tangerang (sample SEM-E-1) is the sixth highest in egg samples from Asia, after eggs from Samut Sakhon, Thailand (Petrlik, Teebthaisong et al. 2017).

At both sites mixed soil and ash samples were also taken. The levels of dioxins and dl-PCBs in soil/ash samples from Tangerang and Bangun exceeded the level measured in the reference soil sample by 21- and 377-fold respectively. These levels explain the accumulation of dioxins and dl-PCBs in free-range chicken eggs, although there is a question mark about whether picking at soil/ash at the sites affected by open burning of plastic waste was only the source of eggs' contamination.

The graphs in Figures 24-26 show dioxin congener patterns in the egg and soil/ash samples from Bangun. They are slightly different from each other even between the individual egg samples, that came from the same chicken owner but possibly from different hens. However, they definitely represent different seasons of the same year (see Table 1). The main differences are in the ratios of OCDD/1,2,3,4,6,7,8 HpCDD /2,3,4,7,8 PeCDF/1,2,3,4,6,7,8 HpCDF congeners between the eggs, as well as in comparison with the soil/ash sample.

The OCDD congener is predominant in a sample of eggs taken in November 2019. The congener's lowest percentage among compared samples from Bangun is present in the egg sample from May 2019. Soil/ash samples from Bangun also exhibit higher levels of HpCDF and OCDF congeners in comparison with eggs. OCDD is the prevalent congener in the soil/ash sample from Tangerang, while there are higher ratios of less chlorinated PCDD congeners in the egg sample. The findings with regards to PCDF congeners is very similar (see graph in Figure 27).

All of these findings are consistent with findings of decreasing bioavailability of dioxin congeners with increased chlorination level (Stephens, Petreas et al. 1995, Kang, Yamamuro et al. 2002), although the same theory does not apply to OCDD which had the highest presence in the egg sample BAN-E-1. It seems that the explanation for the difference between the samples from Bangun also could lie in the potentially unidentified difference between levels of dioxin congeners in the whole foraging area for chicken at the plastic waste yard, and the changed composition of the waste in May as compared to November of 2019. The general profile of dioxin congeners between the soil/ash sample and egg sample shows that contamination of the plastic waste yard in Bangun is most likely the major source of dioxins in the eggs.

The graph in Figure 28 shows different dioxin congener patterns in free-range eggs from two different sites in Tangerang, which might be caused by the difference in the plastic wastes collected and burned at these sites. The site where the SEM-E-1 sample was taken had more electronic waste, in particular visible refrigerator insulation. This sample also exhibited the highest level of HBCD measured in eggs from sampled localities at Java island in 2019. It was also one of the highest levels of this brominated retardant ever measured in eggs globally.

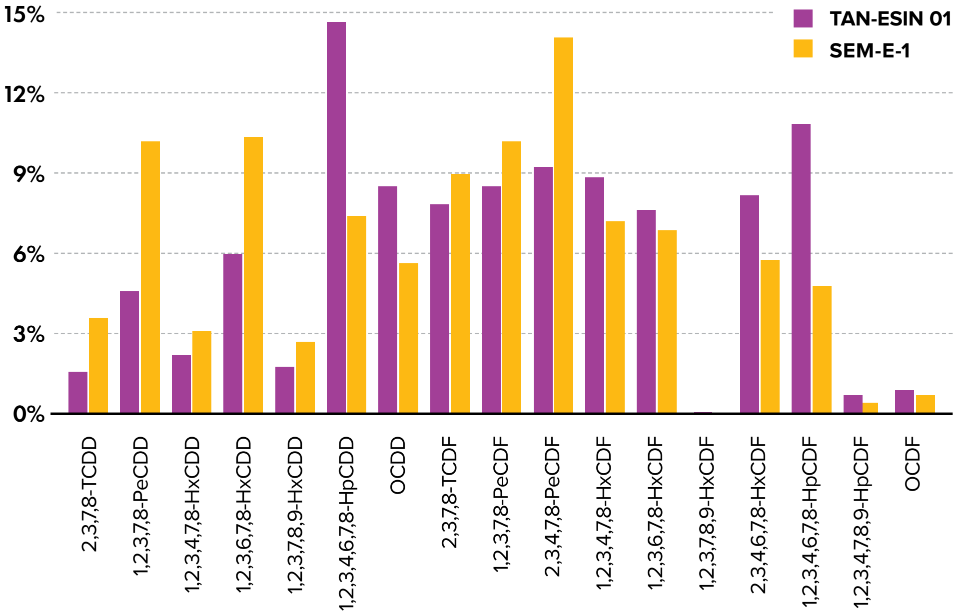
SCCPs were found at a higher level of 3,247 ng g^{-1} dw in the soil/ash sample from Bangun, while in the eggs it was 97 and 153 ng g^{-1} fat, respectively, from that site. It is the opposite situation to what was found in Agbogbloshie, where a higher level of SCCPs was measured in the free-range eggs in comparison with a soil/ash sample from the e-waste scrapyard (Petrlik, Adu-Kumi et al. 2019).

The total sum of nBFRs measured in soil/ash samples from Bangun and Tangerang, at levels of 899 and 187 ng g^{-1} dw, respectively, seem to be considerably high in comparison with the findings of a new study from Melbourne, Australia, where nBFRs *"were detected in 24/30 soil samples with Σ5NBFR concentrations ranging from nd-385 ng/g dw"* (McGrath, Morrison et al. 2017).

DBDPE was measured at a level of 835 ng g^{-1} dw in the soil/ash sample from Bangun. DBDPE also had the highest level of 124 ng g^{-1} fat in the egg sample from Bangun, in comparison with other nBFRs, but still not so high as in the eggs from Tropodo sampled in late autumn 2019 as well.

The accummulation of the decaBDE congener 209 in eggs from Bangun has been shown to be higher (701:1265) than DBDPE (835:106) in comparison with levels in the soil/ash sample. There is not enough information to compare this finding with any other research. Ezechias et al. (2014) in their review of available research about novel BFRs, concluded that *"The data reviewed here document that the mechanisms through NBFRs exhibit their ecotoxicity and the processes leading to their biotransformation in the environment are still poorly understood"* (Ezechiáš, Covino et al. 2014). Further, PFOS were found in higher concentration in eggs, compared to the soil/ash sample in Bangun (see Table 13).

Figure 28: *PCDD/Fs congener patterns in two different free-range egg samples from Tangerang.*



4.4.2 ALUMINUM SMELTERS

The results of analyses for PCDD/Fs, PCBs, HCB, PeCB, HCBD, PBDD/Fs, SCCPs, PBDEs, HBCD, nBFRs, and PFASs in samples from Kendalsari and the neighboring village of Sidokampir are summarized in Table 14. They are compared with reference samples of eggs from the Jakarta supermarket, and of soil from the Mbeji Forest.

Dioxin and dl-PCBs levels in free-range chicken egg samples from the Kendalsari aluminum smelters area (85 and 60 pg WHO-TEQ g^{-1} fat) are among the five highest levels observed in eggs in this study. The whole village of Kendalsari is paved with aluminum ash residues, which can

Figure 29: *The transfer of dl-PCBs from soil to egg is more efficient than the transfer of PCDD/Fs; dl-PCBs can therefore constitute a significant risk for food-chain transfer, even at low environmental concentrations. Source: Swedish EPA (2011).*

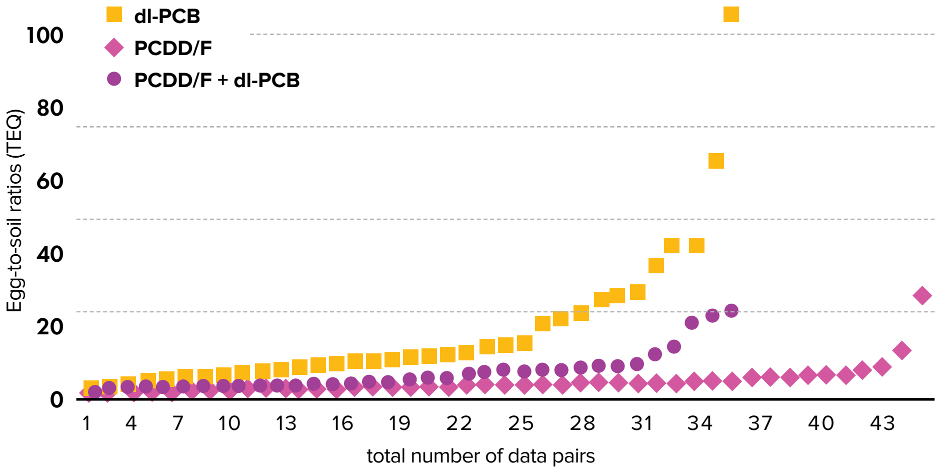


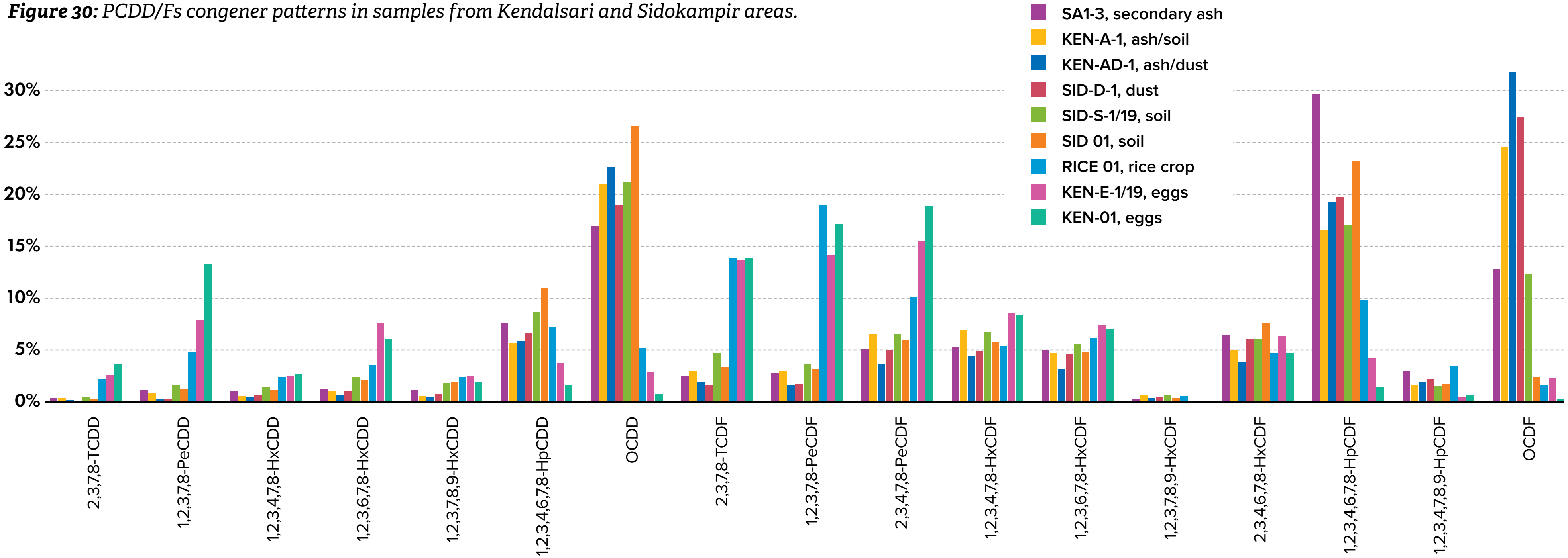
Table 14: *Summarized results of analyses for various POPs in nine samples from the Jombang regency.*

Locality	**Kendalsari**		**Jakarta**	**Kendalsari**		**Sidokampir**				**Mbeji Forest – ref. site**	**Sidokampir**
Sample ID (eggs)	**KEN 01**	**KEN-E-1/19**	**JAK-SUP**	**KEN-A-1**	**KEN-AD-1**	**SA1-3**	**SID-D-1**	**SID 01**	**SID-S-1/19**	**MBEJI-S**	**RICE 01**
Matrix	eggs			ash/soil	ash/dust	ash	dust	soil			rice crop
Fat content (%)	27.4	14.3	9.5								
PCDD/Fs (pg TEQ g^{-1} fat/dw)	49	41	0.0012	17.3	57	484	33	1.1	2.7	0.71	0.84
dl-PCBs (pg TEQ g^{-1} fat/dw)	35	20	0.0020	0.70	4.4	13.1	2.2	0.18	0.47	0.07	0.39
Total PCDD/F + dl-PCBs (pg TEQ g^{-1} fat/dw)	85	61	0.0032	18	61	497	35	1.2	3.2	0.78	1.2
PBDD/Fs (pg TEQ g^{-1} fat/dw)	NA	0.57	NA	NA	NA	NA	NA	NA	NA	<11.1	NA
HCB	1.5	2.5	<0.1	4.3	7.9	19.5	2.5	NA	1.1	0.08	NA
PeCB	1.07	1.3	<0.1	2.0	6.9	NA	1.4	NA	0.80	0.16	NA
HCBD	<0.1	<0.1	<0.1	<0.02	<0.02	NA	<0.02	NA	<0.02	<0.02	NA
7 PCB	7.0	3.7	0	0.18	0.51	0.69	0.20	NA	0.02	<LOQ	NA
6 PCB	5.1	2.9	< LOQ	NC	NC	NC	NC	NA	NC	NC	NA
SCCPs	NA	160	136	NA	NA	NA	NA	NA	NA	2.4	NA
Sum HBCD	< LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	NA	<LOQ	<LOQ	NA
Sum of PBDEs	6.2	150	1.4	0.08	0.35	0.23	0.75	NA	0.57	0.04	NA
209-BDE (decaBDE)	< 2	75	<1.0	<5	<5	<5	<5	NA	<5	<5	NA
7 BDE congeners	6.19	45	1.39	0.06	0.28	0.229	0.58	NA	0.45	0.04	NA
Sum of nBFRs	< LOQ	12.2	< LOQ	0.03	0.44	0.00	19.5	NA	0.02	0.07	NA
Sum of PFASs (ng g^{-1} fw)	NA	0.35	0.1	NA	NA	NA	NA	NA	NA	<LOQ	NA
L-PFOS (ng g^{-1} of fw)	NA	0.12	<0.01	NA	NA	NA	NA	NA	NA	<0.3	NA

fw = fresh weight; dw = dry weight; NA = not analyzed; NC = not calculated; < LOQ = below level of quantification

contain significant levels of PCDD/Fs; however, dl-PCBs have bigger TEQ values in eggs compared to both ash and soil samples from the area of Kendalsari and the neighboring Sidokampir. This can be explained by the more efficient transfer of dl-PCBs from the soil to the egg than the transfer of PCDD/Fs. Fs (Swedish EPA 2011). This process can be better understood from the graph in Figure 29.

Figure 30: *PCDD/Fs congener patterns in samples from Kendalsari and Sidokampir areas.*



The level of 49 pg WHO-TEQ g^{-1} fat of PCDD/Fs in eggs from the vicinity of a secondary aluminum smelter in Kendalsari is comparable with levels in another sample from an Accra hospital site (Petrlik, Adu-Kumi et al. 2019) influenced by residual ash from a small medical waste incinerator and is also comparable to the highest level of PCDD/Fs in free-range eggs from Newcastle (see data in Table 4).[26]

The level of PBDEs is also substantially increased in the egg sample taken in November from the same site and the same chicken owner as the previous sample taken in April 2018. However, that level does not reflect a similar magnitude of contamination of both the ash or soil samples in which the measured levels of PBDEs were much lower compared to eggs. This situation seems to be somewhat similar to dl-PCBs. PBDEs might also be in the emissions from secondary aluminum smelters as they also use aluminum waste for aluminum production, which can contain PBDEs as well. Also, nBFRs were increased in the egg sample from Kendalsari, as well as in the dust sample from the road in Sidokampir, both taken in November 2019.

The dioxin congener patterns measured in all samples from Kendalsari and Sidokamplir is shown in the graph in Figure 30, and the balance between PCDDs and PCDFs in the samples is shown in the graph in Figure 31. Samples seem to be grouped (close to each other) according to their origin: abiotic samples like ashes, dust and soil are close to each

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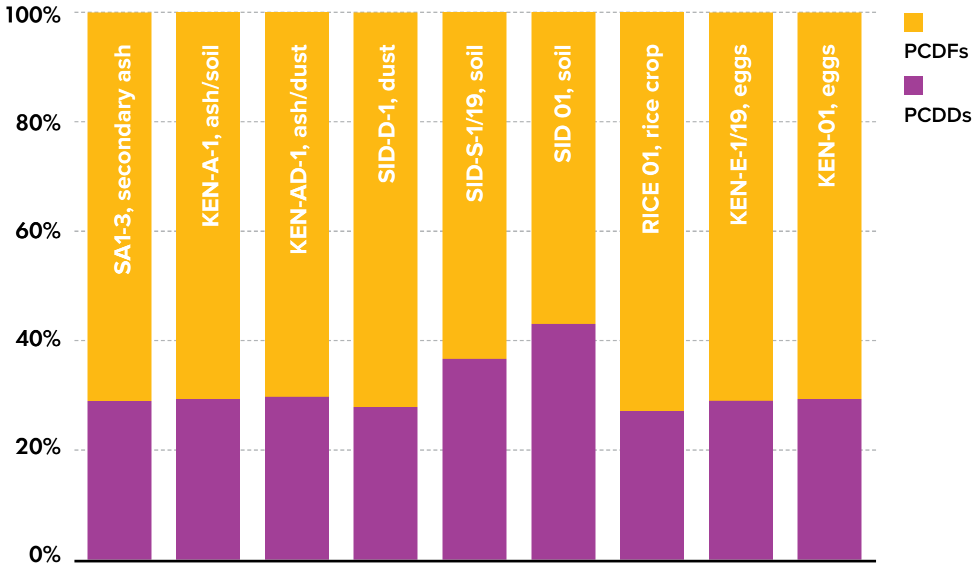
[26] Potential influence of industrial processes in the form of ash residues either from waste incineration or small metallurgical facilities is what all these sites have in common (see Table 16 in this report).

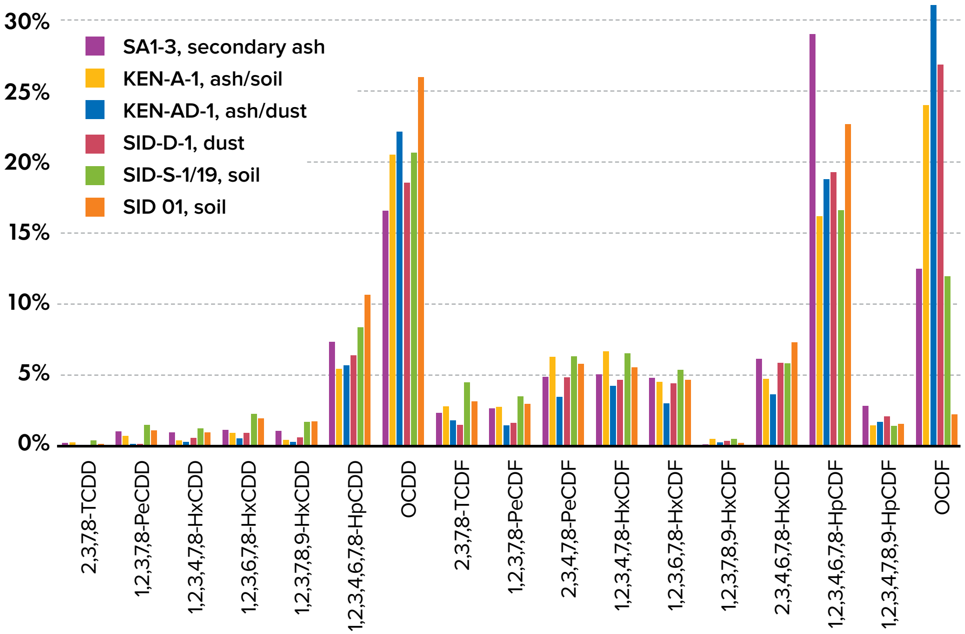
Figure 31: *Balance of PCDD and PCDF congeners in total in the samples from Kendalsari and Sidokampir.*



other (see also graph in Figure 32) and biotic samples like rice and eggs are closer to each other (see also graph in Figure 33). There are exceptions in two cases: the first is the mixed secondary ash sample SA 1-3, and the second is the point soil sample taken from a 20 cm depth level in a rice crop field. Another soil sample, SID-S-1/19, from the same rice field near Sidokampir consisted of six point samples of soil from a layer 2-7 cm below the field surface, taken in November 2019. The situation at the site is visible in Photo 29.

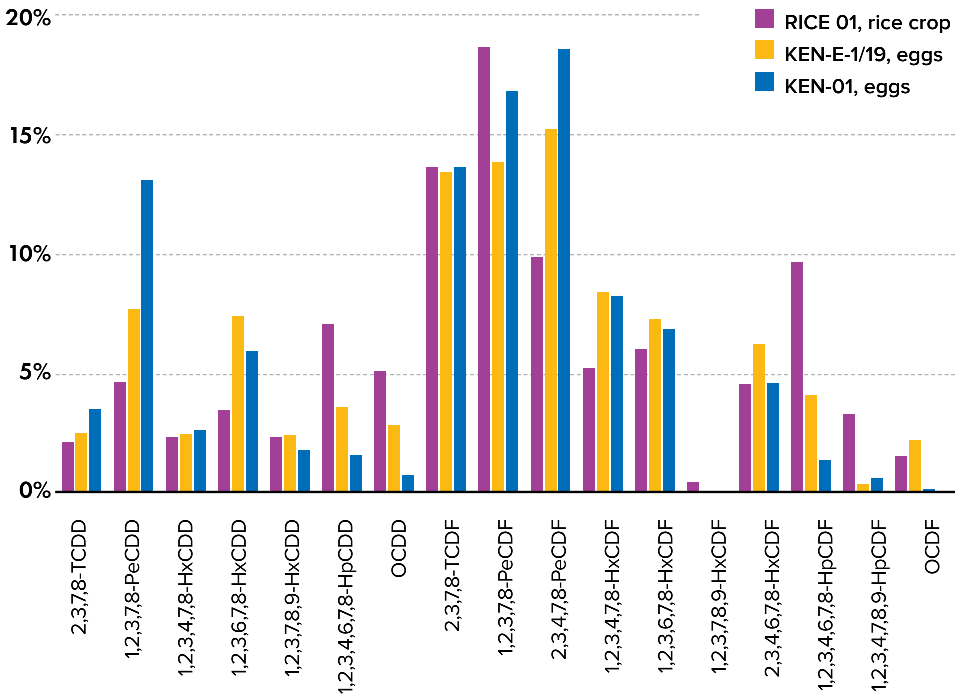
The secondary ash sample SA 1-3 consisted of three samples from residues of secondary aluminum production. The ash was produced by smelters in Jombang. The secondary smelters mix aluminum dross, waste aluminum scraps, cans, aluminum foil and sachet packaging, and add a flux in the process. The flux commonly mixes chloride and fluoride salts used to reduce melt oxidation, minimize penetration of the atmospheric hydrogen, and absorb non-metallic inclusions suspended in the melt. The secondary ash pooled sample was taken at the dumpsites in the Sidokampir and Kendalsari hamlets.

Figure 32: *Dioxin congeners patterns in abiotic samples from Kendalsari and Sidokampir.*



In the samples of eggs and rice, we can see lower accumulation of octa-congeners, of both dibenzo-p-dioxins and dibenzofurans, in comparison with samples of abiotic compartments. This finding is in agreement with decreasing bioavailability of dioxin congeners with increased chlorination level, as suggested by some studies on PCDD/Fs in eggs (Stephens, Petreas et al. 1995, Kang, Yamamuro et al. 2002). Husk and grains locally produced in the fields (in the neighborhood of roads built by using ash in big bags) are used to feed hens in the house where we obtained the egg samples. This fact may have contributed to their contamination also via additional feed provided to the chickens.

***Figure 33:** Dioxin congener patterns in samples of rice crop and eggs from Kendalsari and Sidokampir.*



Levels of PCDD/Fs and dl-PCBs in abiotic samples were higher in ash, mixed ash, and soil and dust samples, while they were ten times lower in soil samples from the crop field. We also compared the difference between the background level of POPs in a reference sample from the Mbeji Forest, with the soil samples from the crop field. The levels in the crop soil in Sidokampir exceeded the background level in the Mbeji Forest by 4- and 14-fold for the groups of PCDD/Fs, dl-PCBs and PeCB, HCB plus PBDEs, respectively. It also shows the scale of contamination caused by secondary aluminum smelters and the reuse of ash produced by a large number of these facilities accumulated in the area of Kendalsari and Sidokampir (see description in subchapter 3.2.1). They increased not only the levels of dioxins and dioxin-like PCBs in the environment, but also other unintentionally produced POPs, like HCB and PeCB. Contamination by PBDEs and novel BFRs is most likely linked to their activity, even though these substances were not found in high levels in the ash sample from secondary aluminum production.

Although the ash containing 0.48 ppb (ng TEQ g^{-1} dw) of PCDD/Fs does not reach current provisional levels of limits for the definition of POPs waste (LPCL),[27] its use contributes largely to serious contamination of the environment and locally grown food in the Kendalsari and Sidokampir villages.

4.4.3 WASTE INCINERATION

Residues of plastic waste sorted in Bangun were transported to Tropodo and used as fuel for local tofu factories. We consider this process as a kind of co-incineration of waste or a very simple waste-to-energy operation. Plastic waste replaced the originally used wood fuel in Tropodo. The whole process is described in subchapter 3.2.3 and/or in previous studies (GAIA 2019, Petrlik, Ismawati et al. 2019).

The samples taken in Tropodo came from the immediate neighbourhood of tofu factories burning waste, as well as from areas directly affected by

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[27] This limit is called "low POPs content" in Article 6 of the Stockholm Convention; Stockholm Convention (2010). Stockholm Convention on Persistent Organic Pollutants (POPs) as amended in 2009. Text and Annexes. Geneva: 64. It is established at levels of 1 or 15 ppb (ng TEQ g^{-1} dw) for PCDD/Fs in the last update of the General Technical Guidelines for POPs Waste; Basel Convention (2017). General technical guidelines for the environmentally sound management of wastes consisting of, containing or contaminated with persistent organic pollutants. Technical Guidelines. Geneva. EU uses the level of 15 ppb of PCDD/Fs in waste, as set in its last update of the POPs Regulation; European Parliament and Council (2019). "Regulation (EU) 2019/1021 of the European Parliament and of the Council of 20 June 2019 on persistent organic pollutants (Text with EEA relevance.)." Official Journal of the European Union: 33.

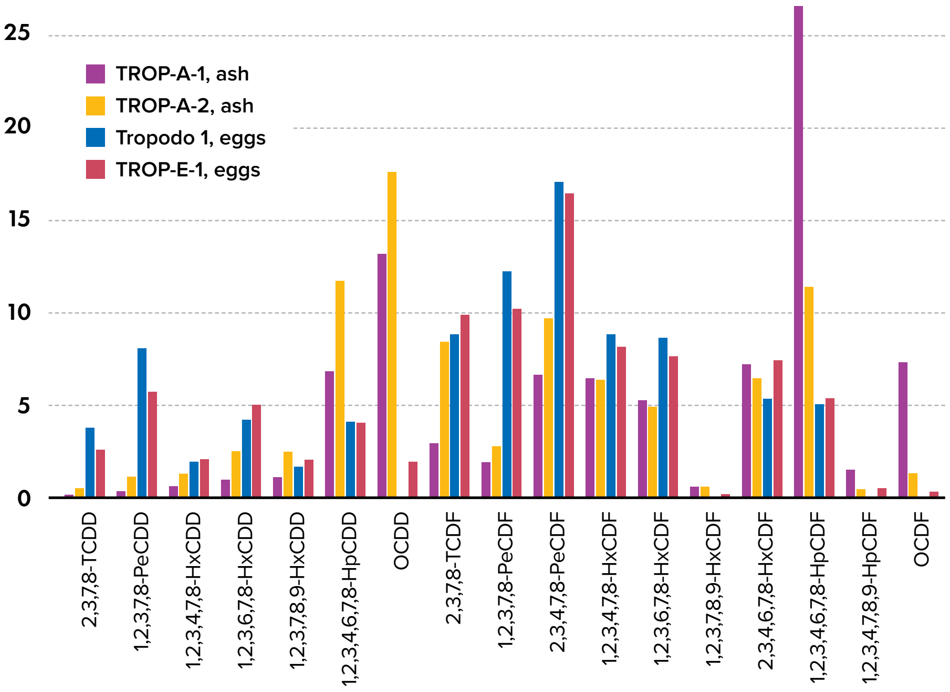
Photo 29: *Rice crop field near Sidokampir, where all samples of soil, dust from the road and rice crop were taken in the years 2018 and 2019 (see Table 1). The road was built from ash in big bags, as is the usual practice in Kendalsari and Sidokampir. Photo: Jindrich Petrlik, November 1, 2019.*

stored ash residues from them. The pooled egg sample from the area of Lakardowo comes from a further distance from the potential industrial source of contamination, which is a hazardous waste incinerator (see 3.2.4), but it could also be affected by local burning of agricultural waste, mostly wood, straw, etc.[28] However, according to research focused on the open burning of biomass, these sources do not create as much dioxins as waste rich in halogens and metals (Zhang, Buekens et al. 2017).

The results of analyses for PCDD/Fs, PCBs, HCB, PeCB, HCBD, PBDD/Fs, SCCPs, some OCPs, PBDEs, HBCD, nBFRs, and PFASs in samples from Tropodo and Sumberwuluh, located near Lakardowo, are summarized in Table 15. They are compared with reference samples of eggs from the Jakarta supermarket, of soil from the Mbeji Forest, and of tofu from the organic food store in Prague.

Extremely high levels of PCDD/Fs were measured in both egg samples from Tropodo, and extremely high level of PBDEs were measured in the egg samples from October 2019 as well. Dioxins and dioxin-like PCBs in the eggs from Sumberwuluh exceeded the level set as the standard for eggs as food in Indonesia and the EU by almost 6- and 3-fold, respectively, which is less than in eggs from Tropodo but still at a considerably high level.

Figure 34: *PCDD/Fs congener patterns as they were measured in ash and egg samples from Tropodo.*



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[28] The agricultural waste may also be contaminated by airborne deposition from incineration which becomes resuspended when burned , airborne as particulate and drops out to the soil.

Table 15: *Summarized results of analyses for various POPs in six samples from Tropodo and Sumberwuluh, compared with reference samples of eggs, soil and tofu.*

Locality	**Tropodo**		**Sumberwuluh**	**Jakarta**	**Tropodo**		**Mbeji Forest – ref. site**	**Tropodo**	**Prague - reference**
Sample ID (eggs)	**Tropodo 1**	**TROP-E-1**	**SUM-E-1 and 2**	**JAK-SUP**	**TROP-A-1**	**TROP-A-2**	**MBEJI-S**	**TROP-TOFU**	**PRAHA_BIO**
Matrix	eggs				ash		soil	tofu	
Fat content (%)	15	13.9	14.1	9.5	NA	NA	NA	7.0	9.6
PCDD/Fs (pg TEQ g^{-1} fat/dw)	200	140	11.0	0.0012	1,246	122	0.71	0.021	< LOQ
dl-PCBs (pg TEQ g^{-1} fat/dw)	32	32	2.0	0.0020	119	12.2	0.07	0.0011	0.0033
Total PCDD/F + dl-PCBs (pg TEQ g^{-1} fat/dw)	232	172	13.0	0.0032	1,365	134	0.78	0.022	0.0033
Total PCDD/Fs + dl-PCBs - DR CALUX® (pg BEQ g^{-1} fat/dw)	560	NA	NA	<LOQ (0.6)	NA	NA	0.5	<LOQ (0.1)	NA
PBDD/Fs (pg TEQ g^{-1} fat/dw)	< 21.3	0.33	NA	NA	NA	NA	<11.1	NA	NA
HCB	5.5	4.1	0.58	<0.1	34	2.1	0.08	<0.1	<0.1
PeCB	1.9	1.7	0.26	<0.1	52.48	4.73	0.16	<0.1	0.29
HCBD	< 0.1	<0.1	<0.1	<0.1	0.04	<0.02	<0.02	<0.1	<0.1
7 PCB	5.3	2.9	<LOQ	<LOQ	0.66	1.3	<LOQ	< LOQ	< LOQ
6 PCB	4.4	2.9	<LOQ	<LOQ	NC	NC	NC	< LOQ	< LOQ
SCCPs	65	97	50	136	NA	15.4	2.4	<50	249
Sum HBCD	<LOQ	<LOQ	4.5	<LOQ	<LOQ	<LOQ	<LOQ	< LOQ	< LOQ
Sum of PBDEs	65	27,159	8.2	1.4	1.1	0.05	0.04	0.79	< LOQ
209-BDE (decaBDE)	4.1	24,611	7.0	<1.0	<5	<5	<5	< LOQ	< LOQ
7 BDE congeners	52	143	0.97	1.4	0.54	0.05	0.04	0.79	< LOQ
Sum of nBFRs	NA	2,166	0.87	< LOQ	0.31	0.02	0.07	< LOQ	< LOQ
Sum of PFASs (ng g^{-1} of fresh weight)	2.7	0.30	0.46	0.1	NA	<LOQ	<LOQ	< LOQ	0.054
L-PFOS (ng g^{-1} of fresh weight)	0.9	0.11	0.22	<0.01	NA	<0.3	<0.3	<0.01	<0.01

fw = fresh weight; dw = dry weight; NA = not analyzed; NC = not calculated; < LOQ = below level of quantification

The congener profile for the Tropodo/Sumberwuluh samples is close to profiles from "closed" facilities, e.g. waste incinerators or smelters, as was already discussed in subchapter 4.1.1.3. They in fact come from the vicinity of facilities that incinerate wastes. Both of these sites exhibit contamination of the food chain as consequence of waste incineration activities.

PCDD/Fs congener patterns in two ash samples and two free-range chicken egg samples from Tropodo are shown in the graph in Figure 34. There is a difference between the respective patterns of eggs and ash.

It is also clear from the graph in Figure 35 that even between two different samples of ash, there is a difference in the scale of dominance of certain congeners. The two ash samples from Tropodo might differ because one was a very fresh sample, partly still hot ash from one tofu factory (TROP-A-1), while the other one (TROP-A-2) was several days or weeks old, taken from the area where it was dumped at the backyard of another tofu factory as shown in Photo 30. The incinerated waste may have differed in composition between these two facilities as well.

Hens in Tropodo could walk to the sites where the ash from tofu factories was dumped in their backyards (see Photo 30), although there were natural barriers that probably discouraged hens to go there, as we didn't see them there during sampling. Ash in big bags was used at the yards where hens walked, but we didn't take any samples from there due to limited resources for such extensive sampling.

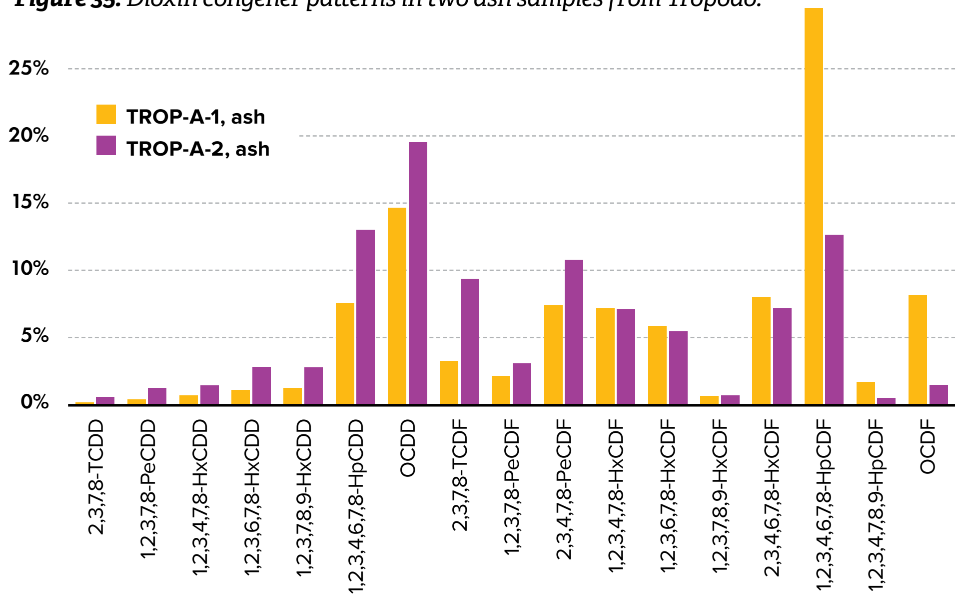
None of the ash samples contained PBDEs at a level corresponding to that found in eggs. This fact points to the apparently high proportion of hen's egg contamination associated with the deposition of dust from tofu factory emissions (see Photo 31). We didn't take any samples of dust in Tropodo, which might have shed a light on this potential pathway of contamination of the eggs.

Photo 30: *Pile of ash from burned plastic waste dumped in the backyard of one of the tofu factories in Tropodo, where the ash sample TROP-A-2 was taken. Photo: Jindrich Petrlik, Arnika.*

The very different levels of contamination of eggs with PBDEs indicate a different composition of the incinerated waste between May and October 2019, which was already reflected at the plastic waste scrapyard in Bangun from where the residual waste was transported to Tropodo (see subchapter 4.4.1). PBDEs substantially increased in eggs between the May and autumn samples in Bangun, as it was in Tropodo, although the level of PBDEs measured in Bangun was not as high as in the Tropodo eggs. This can be explained by the intense burning of waste, followed by its transport with air emissions and particulate matters deposited in the area of the Tropodo village.

We also sampled tofu during our October 2019 mission in Tropodo. Pooled samples of tofu were taken in one of the tofu factories, the same one where the ash sample TROP-A-1 was also taken (see Photo 32). This

***Figure 35:** Dioxin congener patterns in two ash samples from Tropodo.*



factory is located in a neighborhood very near the sampling place of both pooled egg samples.

The results of the analyses are summarized in Table 15 together with the results of the analysis of tofu bought in Prague, Czech Republic, which had declared organic quality.

There were slightly increased levels of PCDD/Fs and PBDEs in the tofu sample from Tropodo of 0.021 pg TEQ g^{-1} fat and 0.79 ng g^{-1} fat, respectively, compared to levels below LOQ in the tofu from Prague.

The tofu samples from Prague had a little bit higher level of dl-PCBs, which can result from the overall higher levels of PCBs in the Czech environment as a consequence of their wide use in the country (Holoubek, Klánová et al. 2005, van den Berg, Kypke et al. 2017). The concentrations of dioxin-like PCBs were one magnitude lower in the tofu sample compared to dioxins (see Table 15).

The level of 0.021 pg TEQ g^{-1} fat measured in tofu from Tropodo is well below the limit of 0.75 pg TEQ g^{-1} of PCDD/Fs in fat set for vegetable fats in the EU (European Commission 2016).

The above summarized analytical results of POPs in tofu samples from Tropodo show a potential trace contamination of the tofu produced in Tropodo due to the practice of burning plastic waste. High contamination of tofu was not expected, as it is not produced from locally grown soybeans, and also because POPs do not accumulate in the water in which tofu is boiled in the factories. The potential pathway can therefore only be dust that gets into the water, but POPs are not soluble in water as such. It is rather local food of animal origin rich in animal fats that is contaminated by the practice of plastic waste incineration in tofu factories.

4.5 USE OF RESIDUAL ASH FROM METALLURGY AND WASTE INCINERATION

Many industrial processes including waste incineration, power plants or metallurgy produce ash residues as waste. It is also common that this waste is used as construction material (Ferreira, Ribeiro et al. 2003, Calonzo, Petrlik et al. 2005, Petrlik and Ryder 2005, Sun, Li et al. 2016, Petrlik and Bell 2017, Arkenbout 2019, Assi, Bilo et al. 2020), as it would otherwise require large areas to store it in a safe way. In addition, incineration of plastic waste in Tropodo, hazardous waste in Lakardowo, and secondary aluminum smelters in Jombang produce large volumes of ash. We must ask whether the use of ash from waste incineration and metallurgical facilities in Java is safe for the environment and human health.

Currently, no general consensus appears to exist regarding residue disposal and solutions for use on a worldwide level, although the BAT/BEP Guidelines of the Stockholm Convention contain advice on how to avoid POPs release due to improper handling of APC residue.

Photo 31: *Particulate matter deposition from plastic waste incineration in tofu factories in Tropodo contributes to the contamination of locally grown food. Photo: Jindrich Petrlik, October 31, 2019.*

Some of the suggestions of the BAT/BEP Guidelines include: "*Fly ash from electrostatic precipitators and residues from air pollution equipment almost certainly contain significant amounts of chemicals listed in Annex C of the Convention, so these wastes have to be disposed of in a controlled way. .... Fly ashes should never be used as soil amendment in agricultural or similar applications*" (Stockholm Convention on POPs 2008).

It has been estimated that due to uncontrolled use of fly ash from waste incineration, we potentially lose control of up to 7-10 kg TEQ of dioxins annually (Petrlik and Bell 2017a). This estimate does not include ash and other residue produced by metallurgical processes such as we observed in Kendalsari and Sidokampir.

Photo 32: *Inside one of the tofu factories in Tropodo. The soybeans used for production of tofu are imported. Photo: Jindrich Petrlik, October 31, 2019.*

Table 16: *Summary of levels of PCDD/Fs (in TEQs and/or BEQs) observed at different sites influenced by fly ash and other waste contaminated by PCDD/Fs described in this study or in the literature.*

	Year(s) of sampling	Fly ash (waste)	Soil/sediment direct impact	Soil/sed. – refer.	Eggs	Eggs – reference[1]
Units		pg TEQ g^{-1} dw			pg TEQ g^{-1} fat	
Indonesia (Tropodo, tofu factories burning plastic waste)	2019	122; 1,246	N/A	0.71	140**	0.0012
Indonesia (Kendalsari, aluminum smelters)	2019	484	17.3	0.71	41; 49	0.0012
Thailand (WI Phuket) [a]	2010 - 2011	3,200 - 8,000	2,700***	N/A	6.1*	0.08 [a]
China (WI Wuhan) [b]	2014 - 2015	779	N/A	N/A	12.2	0.2 [b]
UK (Bishops Cleeve) [c]	2010 - 2011	2,500	6.5 – 11*	0.05 - 1.2	1.8; 21; 55*	0.2 [d]
UK (Newcastle) [d], [e]	2000	20 - 9,500	7 – 292	N/A	0.4 – 56	0.2 [d]
Peru (Zapallal) [f]	2010	50 - 12,000	5 – 11	0.05 - 1.2	3.4 - 4.4	0.12 [f]
Taiwan (eggs event) [g]	2005	N/A	N/A	N/A	32.6	0.274 [h]
Poland (henhouse) [i]	2015	3,922	16 – 47	0.1 - 0.8	12.5 - 29.3	0.44 [i]
Ghana (Accra, hospital) [j]	2018	551	N/A	2****; [k]	49	0.39

*Notes: [1] eggs from supermarkets are mostly used as a reference level for the whole country, if it is not otherwise stated in the literature referenced in this table; N/A – not available, *BEQs (total dioxin-like toxicity), **ash contributes to overall contamination of eggs, *** sediment, **** dl-PCBs + PCDD/Fs (site in Accra)*

References for Table 16: [a] (Petrlik, Teebthaisong et al. 2017); [b] (Petrlik 2015); [c] (Katima, Bell et al. 2018); [d] (Pless-Mulloli, Schilling et al. 2001); e) (Watson 2001); [f] (Swedish EPA 2011); [g] (The Epoch Times 2005); [h] (Hsu, Chen et al. 2010); [i] (Piskorska-Pliszczynska, Strucinski et al. 2016); [j] (Petrlik, Adu-Kumi et al. 2019); [k] (Tue, Goto et al. 2016)

As our research in Indonesia focused on POPs, we will not address the problem with heavy metals in ash. Instead, we will focus on a brief assessment of POPs and dioxins in particular in ashes. We will also try to put the use of ash in Tropodo, Kendalsari and Sidokampir in a broader context.

Table 16 summarizes the results of analyses of dioxins in free-range chicken eggs from places influenced by ash from waste incinerators, metallurgical operations or other wastes containing PCDD/Fs in different locations around the world. The data in the table map the potential pathways of pollution by dioxins from wastes to soil (as carrier) and then to free-range chicken eggs as the receptor. They are also compared with background levels (for reference) of PCDD/Fs in eggs from several different countries, mostly levels in eggs from larger farms where the chickens are kept inside and do not have access to dioxin-contamination sources.

In previous studies, the processing and disposal of waste containing PCDD/Fs between 20 and 12,000 pg TEQ g^{-1} led to contamination of the food chain (eggs or poultry meat) up to levels >20-times higher than the suggested EU limit for PCDD/Fs in food (2.5 pg TEQ g^{-1} fat). Levels from reference sites (background levels) in free-range chicken eggs were exceeded up to 280-fold. Used ash from aluminum smelters in Kendalsari contaminated the foraging area of hens (see Photo 33) up to the measured level of 17.3 pg TEQ g^{-1} dw of PCDD/Fs in the garden. Their eggs were found to be contaminated up to 49 pg TEQ g^{-1} fat (see also subchapter 4.4.2), which exceeded the EU limit for eggs by almost 20-fold, although the level of dioxins in secondary ash from aluminum smelters reached "only" 0.5 ppb. This demonstrates the ability of dioxin to bioaccumulate in eggs while starting from low levels in soil.

We also demonstrated that the use of ash from aluminum smelters leads to increased levels of dioxins in the soil in crop fields next to where roads were built from ash in big bags (see Photos 29 and 34, and subchapter 4.4.2). The use of rice or other crops as feed for animals (chicken, cows, pigs, etc.) can lead to further contamination of the food locally grown by the villagers.

Photo 33: *Chickens forage in a garden with ash and slag residue mixed with soil at the Kendalsari egg sampling site. Photo: Jindrich Petrlik, November 1, 2019.*

Photo 34: *This photo shows how the ash from secondary aluminum smelters is used for building roads almost everywhere in Kendalsari and Sidokampir. Photo: Jindrich Petrlik, November 1, 2019.*

Photo 35: *Waste incinerator in Phuket, Thailand. Waste incineration ash was storedright on the shoreline to a mangrove wetland. Photo: Jindrich Petrlik, December 11, 2010.*

Photo 36: *Sampling of ash from plastic waste incineration in Tropodo. On the right-hand side, there is a view into a backyard where chicken eggs were sampled in October 2019. Photo: Ecoton, October 31, 2019.*

A similar situation was demonstrated also regarding fish and other marine animals in the vicinity of the Phuket waste incinerator in Thailand where residual ash was stored next to a mangrove wetland (Petrlik 2011, Katima, Bell et al. 2018) (see Photo 35). The potential influence on the water ecosystems in the surrounding areas of Kendalsari, Sidokampir and Tropodo should also be tracked in a follow-up of this study, as river sediments and animals living in water ecosystems also could be affected by the large-scale use of ash residue in this part of Java.

A level of 140 pg TEQ g^{-1} fat was measured in the pooled egg sample from the site in Tropodo, where hens had access to ash piles and dumps in the backyards of tofu factories (see Photo 36). It was also found that ash is not the only source of contamination of eggs in Tropodo, based on PCDD/Fs congener analyses. The dust deposition of the emissions from burning plastic waste in tofu factories most likely contributed substantially to the contamination of the food chain in this village. The level of dioxins in ash was measured to be 0.1 and 1.2 ppb, respectively, in two samples taken from tofu factories and their backyards (see subchapter 4.4.3).

A Swedish EPA study demonstrated that PCDD/Fs levels of 30 pg TEQ g^{-1} fat in an egg will be exceeded at soil concentrations of approximately 4 to 75 ng TEQ kg^{-1} dw (=0.004 to 0.075 ppb). Therefore, the European maximum level of 2.5 pg TEQ g-1 PCDD/F in fat (European Commission 2006) can be exceeded at levels that are ten times lower (i.e. 0.4 and 7 ng TEQ kg^{-1} dw). Based on the upper end of the range given in the Swedish EPA study and examples of a scenario with contaminated wood waste (Swedish EPA 2011), it can be concluded that application of fly ash and other wastes containing levels of dioxin over 0.05 ppb in land-based applications can lead to unacceptable contamination of the local food chain. In some other studies, even lower levels of dioxins in soils led to contamination of free-range chicken eggs exceeding the EU standard for food (Pirard, Focant et al. 2004, DiGangi and Petrlik 2005). Free-range eggs can be impacted at critical levels, in some cases exceeding the current EU limits by more than 20-fold.

Locally produced food is of great importance in developing countries, and in rural locations in developed countries; therefore, this exposure scenario is of particular concern. The two case studies from the sites in Indonesia underscore the need for stricter control of POPs in wastes that are allowed to be used freely, in places in direct or indirect contact with agricultural or rural areas where local food is produced, and/or with residential areas. The demonstrated cases also show that waste with a dioxin content even below 1 ppb should be restricted from direct use in such areas.

5. Conclusions

5.1 PLASTIC WASTE DUMPSITES

The analyses performed in our study have shown that plastic waste dumpsites on the island of Java are not only a waste problem, but they are also a source of environmental contamination from a wide range of persistent organic pollutants (POPs). Many of them are already contained in the plastics themselves as additives, but others are created by burning waste to clear space for new waste brought in for sorting.

The level of POPs contamination caused by dumping, incineration and open burning of plastic waste ranks some sites on the island of Java among the most contaminated in the world, alongside sites heavily affected by industrial production or sites contaminated as a consequence of war.

The dioxin levels in the eggs from plastic waste dumpsites exceeded the EU regulatory limit by 4- to 22-fold. Eggs from Tangerang also had high levels of brominated dioxins and HBCD. Bangun egg samples had high levels of PBDEs, HBCD and PFOS.

5.2 E-WASTE

It was most likely plastics from e-waste that contributed significantly to the contamination of the food chain in Bangun, Tropodo and Tangerang during the October/November 2019 round of sampling. This was reflected in the high concentration of brominated flame retardants found in free-range chicken eggs.

Plastics from electronic waste contribute significantly to the contamination of the food chain in Bangun, Tropodo and Tangerang. This was reflected in the high concentrations of brominated flame retardants found in free-range hens' eggs, and in the case of Tangerang we also found a significant contribution of brominated dioxins to the overall toxicity of the egg sample from the site with plastic residues from refrigerator insulation.

Not only were high concentrations of flame retardants such as PBDEs and HBCD, which are already regulated by the Stockholm Convention, analyzed in the eggs, but also DBDPE, i.e. substances belonging to the novel-BFRs group. DBDPE replaced DecaBDE in electrical cable insulation and polystyrene. Although DecaBDE reached the highest concentrations in samples from Tropodo and Bangun compared to other congeners of PBDEs, other congeners forming previously banned commercial PBDE mixtures were also found in the eggs in high concentrations.

5.3 WASTE INCINERATION

In the vicinity of waste incinerators, we found contamination of hens' eggs, mainly dioxins and dioxin-like PCBs. In Tropodo, where plastic brought from Bangun is burned, we also found high concentrations of PBDEs in eggs. The combustion in the tofu ovens clearly does not reach such temperatures that PBDEs decompose. These instead accumulate in the dust and enter the food chain. Although the levels of dioxins and dl-PCBs were a little bit higher in the tofu alone compared to the reference sample, it was not a significant contamination.

The analytical results of POPs in the tofu samples from Tropodo showed potential trace contamination of the tofu produced in Tropodo due to the practice of burning plastic waste. The high contamination of tofu was not expected as it is not produced from locally grown soybeans, and also because POPs do not accumulate in the water in which the tofu is being boiled in the factories. The potential pathway can therefore only be dust that gets into the water, but POPs are not soluble in water as such. It is rather local food of animal origin rich in animal fats that is contaminated by the practice of plastic waste incineration in the tofu factories.

The level of contamination of the food chain in Tropodo has reached the level of sites such as the Bien Hoa, a former U.S. Army base in Vietnam, for dioxins, or the infamous e-waste scrap processing site in Guiyu, China, where it is contaminated with brominated flame retardants, especially PBDEs.

In a pooled sample of free-range chicken eggs, we found outrageous levels of 200 and 140 pg TEQ g^{-1} fat of dioxins respectively. These are the third- and the fourth-highest levels of dioxins in eggs from Asia ever measured, and the sixth- and seventh-highest levels of dioxins in eggs found globally. The level of PBDEs in an egg sample from November 2019 from Tropodo was the second-highest ever measured level of these flame retardants in eggs globally.

5.4 SECONDARY ALUMINUM SMELTERS

Secondary aluminum smelters in the Jombang Regency are significant sources of the release of dioxins, dl-PCBs and possibly PBDEs into the environment. This was demonstrated by analyses of hens' eggs, rice crop, soil, ash and dust from the villages of Kendalsari and Sidokampir. The level of dioxin contamination of eggs from free-range hens ranked the Kendalsari samples as the seventh- and eighth-highest among the samples analyzed from Asia so far and as the 15th- and 19th-highest among the samples taken worldwide. Taking into account also the very high levels of dl-PCBs, PCDD/Fs and dl-PCBs in the egg samples from Kendalsari, these exceeded the Indonesian acceptable limit for eggs for human consumption by 24- and 34-fold respectively.

The use of ash from secondary aluminum production as a building material to strengthen roads, flood defenses and building foundations has been shown to be a significant route to environmental contamination in the Jombang regency.

5.5 ASH RESIDUES

The situation found in Jombang regency around villages where secondary aluminum smelters are located, as well as in Tropodo, documents that dioxin-containing ash as a result of combustion processes causes, or significantly contributes to, the contamination of food chains with POPs.

The ash from the combustion processes in both Tropodo (plastic combustion) and the Jombang regency (secondary aluminum smelters) contains dioxin concentrations well below the now practically used provisional Low POPs Content Level of 15 ng g^{-1} dw (= 15 ppb)[29].

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[29] The limit called "low POPs content" in Article 6 of the Stockholm Convention defines when the waste is considered to be POPs waste, which has to be managed in special ways defined in Article 6; Stockholm Convention (2010). Stockholm Convention on Persistent Organic Pollutants (POPs) as amended in 2009. Text and Annexes. Geneva: 64. It is established at levels of 1 or 15 ppb (ng TEQ g^{-1} dw) for PCDD/Fs in the last update of the General Technical Guidelines for POPs Waste; Basel Convention (2017). General technical guidelines for the environmentally sound management of wastes consisting of, containing or contaminated with persistent organic pollutants. Technical Guidelines. Geneva. The EU uses a level of 15 ppb of PCDD/Fs in waste, as set in its last update of the POPs Regulation; European Parliament and Council (2019). "Regulation (EU) 2019/1021 of the European Parliament and of the Council of 20 June 2019 on persistent organic pollutants (Text with EEA relevance.)." Official Journal of the European Union: 33.

But the dioxin levels observed in eggs exceed the recommended food standards for their consumption by manyfold. A similar situation has been mapped in several cases in different locations around the world. Setting limits for dioxins in waste too loosely leads to uncontrolled handling of residual ashes from different combustion sources.

Although we monitored the concentrations of dioxins and other POPs only in the eggs of free-range hens, the milk and meat of cows and other cattle consumed by the locals may be similarly contaminated.

Locally produced food is of great importance in developing countries and rural locations in developed countries, therefore the exposure scenario when ash containing dioxins contaminates the food chain is of particular concern. Two case studies from the sites in Indonesia underscore the need for stricter control of POPs in wastes that are allowed to be used freely at places in direct or indirect contact with agricultural or rural areas where the local food is produced and/or with residential areas. The demonstrated cases also show that waste with a dioxin content even below 1 ppb should be restricted from direct use in such areas.

5.6 POPs IN EGGS

5.6.1 UPOPs

Analyses of nine pooled samples of free-range chicken eggs have shown overall contamination by PCDD/Fs and dl-PCBs at all the selected hot spots. All free-range chicken egg samples in this study exceeded the Indonesian and EU maximum levels (ML) of PCDD/Fs, and for the sum of PCDD/Fs and dl-PCBs. All samples also were above the background level of WHO-TEQ measured in eggs from the supermarket by more than 4,060-fold (sample from Sumberwuluh) to 72,500-fold (sample from Tropodo).[30] Brominated dioxins (PBDD/Fs) contributed significantly to the overall toxicity of free-range eggs from the site in Tangerang affected by plastics from e-waste.

Dioxin levels in five samples from this study are among the 20 egg samples with the highest-ever measured levels of dioxin globally.

None of the egg samples contained levels of HCBD above LOQ. Levels of HCB and PeCB were relatively low and did not exceeded the established EU limit for eggs.

5.6.2 PBDEs AND OTHER BFRs

There was a large difference in levels of PBDEs in the eggs collected in Bangun and Tropodo in May and in November 2019. The eggs from November contained an order-of-magnitude higher levels of these brominated flame retardants. This is most likely the consequence of a different composition of the plastic waste brought to Bangun and burned in Tropodo in the autumn of 2019, in which more plastics most likely had their origins in e-waste. The level of PBDEs measured in the pooled egg sample from Tropodo is the second-highest ever measured in chicken eggs globally. There was a very high level of DBDPE in the same sample of eggs. DBDPE is a representative of the group of nBFRs, which in the products replaced PBDEs and HBCD, regulated already by the Stockholm Convention on POPs, it replaced mainly DecaBDE in wire coatings and polystyrene products.

In addition, the levels of 844 and 538 ng g^{-1} fat in eggs from Tangerang and Bangun, respectively, are among the ten highest levels of HBCD observed in eggs globally.

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[30] In this regard it has to be noted that the level of PCDD/Fs measured in eggs from the supermarket in Jakarta was one to two magnitudes lower than in the egg samples from supermarkets used as a reference in other countries or studies.

5.6.3 PFASs

The highest levels of PFASs were measured in eggs from Bangun, in May and November samples; the sample from November was measured at a five-times higher level than the sample from May. These levels are comparable to those found in eggs from areas affected by industry in Europe. The levels in the samples from other sites on the island of Java were not so high.

5.7 TOLERABLE DAILY INTAKE (TDI) OF SELECTED POPs

We calculated the dietary intake for the PCDD/Fs, dl-PCBs, PBDD/Fs, PBDEs, and PFOS.

By average egg consumption calculated as half an egg (18 g of egg) per day for an adult person weighing 58 kg people eating free-range eggs would exceed the European Food Safety Authority (EFSA) tolerable daily intake (TDI) level for dioxins and dioxin-like PCBs by 1.5- to 43-fold. The most serious situation was in 2019 in Tropodo, Kendalsari and Tangerang.

The tolerable daily intake (TDI) of PCDD/Fs and dl-PCBs can be reached by eating only 0.01 to 0.02 of a free-range egg in Tropodo, 0.02 to 0.1 of an egg in Kendalsari and Tangerang, or one quarter of an egg in Bangun or Sumberwuluh, where contamination by dioxins and dl-PCBs is lower than in Tropodo. In comparison, it would be necessary to eat more than 1,350 eggs from the supermarket in Jakarta to reach the tolerable daily intake for dioxins and dl-PCBs. This example shows the enormous difference between background/reference contamination by PCDD/Fs and dl-PCBs and contamination at localities affected by improper handling of plastic waste, secondary aluminum smelters or waste incineration and waste-for-energy operations.

There is a significant contribution of brominated dioxins to the daily intake of dioxin-like acting chemicals in samples of eggs from Tangerang, which reached one-tenth of the total intake from PCDD/Fs and dl-PCBs in eggs.

The intake of PBDEs from an egg sample in Tropodo (110 ng kg^{-1} bw) is almost 28-fold higher than the average total daily intake from the food basket calculated by a joint committee of WHO and FAO in 2006, at level of 4 ng kg^{-1} bw. The daily intake of PBDEs at the studied locations was in 2019 equal to more than ten times higher than the total daily intake of PBDEs was in Finland, Sweden or Canada 15 or more years ago.[31]

An adult eating half an egg per day from a free-range chicken foraging in the vicinity of the Bangun dump site would exceed the proposed tolerable daily intake (TDI) of PFOS by 3- and almost 16 -fold respectively. The eggs from Tangerang exhibited the second-highest intake of PFOS among the sampled eggs from Java in this study, and an adult eating one egg from a free-range chicken in the Tangerang plastic waste yard would almost reach a TDI of PFOS, but in reality people get exposed to PFOS from a much wider range of foods and drinks.

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[31] In this comparison decaBDE and eight other PBDE congeners were not included in order to make it more comparable to calculations of PBDE intake done between 2001 and 2004. The highest intake of PBDEs was observed in a pooled egg sample from Tropodo, taken in October 2019 with an extremely high level of these BFRs. It also exhibits a very high ratio of decaBDE congener intake. The second-highest intake was calculated for a sample taken in November 2019 in Bangun, again with a very high contribution of decaBDE congener.

6. Limitations of the study

This study focused on three important sources of toxic pollution on the island of Java, and a broad range of POPs as well. As the potentially affected areas are large, we could not map the situation at its full scale. In order to get better picture of the level of contamination of the food chain, we have chosen free-range chicken eggs as proven sensitive indicators of POPs contamination in soils and dust.

Eggs also represent an important human exposure pathway (Van Eijkeren, Zeilmaker et al. 2006, Hoogenboom, ten Dam et al. 2014, Piskorska-Pliszczynska, Mikolajczyk et al. 2014). As "active samplers," they have been used to reveal POPs contamination in many other studies already (Papadopoulos, Vassiliadou et al. 2004, DiGangi and Petrlik 2005, Soerensen S 2011, Bouwman, Bornman et al. 2015, Weber, Watson et al. 2015, Adu-Kumi, Petrlík et al. 2019, Pajurek, Pietron et al. 2019, Petrlik, Behnisch et al. 2019, Kudryavtseva, Shelepchikov et al. 2020).

We also tried to take indicative pooled samples of soil or ash at places where we found them to be accessible for hens from which we received sampled eggs, however we acknowledge that our information about the overall contamination at each location is limited because of the limited number of samples and having no measured data about dioxin congener profiles of air emissions at the sites. Our ability to follow the food chain contamination fully was limited by the resources available for this study, which has to be considered as pilot information.

Photo 37: *Cows walking through the dumpsite in Tangerang. Ash residues after burning of plastic waste are visible in this picture as well. Photo: Jindrich Petrlik, November 3, 2019.*

The whole transfer and fate of dioxin congeners has not yet been fully discovered. We must keep this limitation of knowledge in mind when looking for sources of pollution. The similarity of the dioxin patterns from combustion sources also plays a significant role. We can try to find the most likely sources of egg contamination, bearing the aforementioned limitations of such a search in mind.

Although the eggs represent a good "sampler" of the overall food chain contamination at selected hotspots, they definitely cannot give us a complete picture about food chain contamination. This study was not designed to cover POPs contamination of the full food basket. It would therefore be very useful in follow-up monitoring to focus on other types of locally grown food.

For example, there were cows observed to forage on a rural plastic waste dumpsite in Tangerang (see Photo 37). It would be useful to take samples of milk from these animals. Contamination of cow's milk by various chemicals has been studied before in relation to specific contaminated sites (Braga, Krauss et al. 2002), as well as particular pollution sources (Liem, Hoogerbrugge et al. 1990, Riss, Hagenmaier et al. 1990, Grova, Feidt et al. 2002, Andre, Marchand et al. 2004, Diletti, Ceci et al. 2008, Esposito, Cavallo et al. 2009).

Because of the short time period for sampling, we did not include passive air sampling at the selected sites, which might be another potential avenue for future research. Passive air sampling is widely used as a monitoring tool to evaluate the Stockholm Convention's effectiveness (Arataki, Nagai et al. 2008, Lammel, Dobrovolný et al. 2009, Stockholm Convention 2013a).

7. International Conventions' measures

7.1 CONTROLS OVER PLASTIC WASTE AND E-WASTE EXPORTS IN THE BASEL CONVENTION

The toxic consequences of plastic waste imports into Indonesia demonstrated in this study provide strong justification for action under the Basel Convention. In May 2019, the Fourteenth Conference of Parties to the Basel Convention (COP14) agreed by consensus to bring most plastic wastes under the control regime of the Basel Convention (BAN 2019a, IPEN 2019a). The decision takes effect on January 1, 2021, according to decision BC-14/12 of the Basel Convention (Basel Convention 2019) and is expected to have a major impact on global plastic waste flows and production. Indonesia ratified the Basel Convention in 1993 (Basel Convention 2019a).

First, governments created a listing for hazardous plastic waste, which is subject to all treaty control procedures. Second, export of mixed or contaminated plastic wastes will now require prior informed consent, granting the importing country the right to refuse the shipment. Only a few narrow exemptions for non-hazardous, non-PVC, clean unmixed and uncontaminated plastic wastes can be exported freely, and only for recycling – not burning or landfilling (Basel Convention 2019). However, these exemptions include fluorinated polymers made with PFASs. The data in this study showing contamination of eggs with PFASs indicates that this exemption should be ended. Currently, the Basel Convention Small Intersessional Working Group is examining this issue and will make recommendations to Basel COP15 on the matter.

A second major decision at COP14 addresses actions governments should take on plastics. These decisions can be used to address both production and the numerous toxic chemicals used in plastics. Governments agreed that managing plastic waste begins upfront, noting the importance of more sustainable production. They also agreed on the importance of reducing single-use plastics and replacing them with environmentally friendly alternatives. Finally, governments agreed that actions on plastics should include removing or reducing the hazardous chemicals that are included in their production and at any subsequent stage of their life cycle.

The Basel Convention decisions at COP14 should have a positive impact on reducing and eliminating uncontrolled plastic waste imports into Indonesia. After January 1, 2021, Indonesia will have the power to refuse mixed or contaminated wastes through the prior informed consent procedure. Burning or landfilling of plastic imported wastes will not be permitted. Indonesia also can consider banning all plastic waste imports, as was done previously by China. Enforcement will be a key measure for either option.

Indonesia also should benefit from the entry into force of the Basel Convention Ban Amendment, which happened on 5 December 2019 (Basel Convention 2019b). Indonesia has ratified this amendment, which prohibits the member states of the Organization for Economic Cooperation and Development (OECD), the European Union (EU), and Liechtenstein from exporting hazardous wastes as defined by the Convention (Basel Convention 2014) to other countries – primarily developing countries, or countries with economies in transition. The Ban Amendment includes most Persistent Organic Pollutants (POPs), most electronic wastes, most obsolete ships, most flammable liquids, and most toxic heavy metals. It will include plastic or paper waste if contaminated with hazardous waste. Indonesia ratified the Ban Amendment in 2005 (Basel Convention 2019c) and played a key role in establishing it within the treaty.

It is a question mark whether e-waste plastics contributing to contamination of eggs in Bangun, Tropodo and Tangerang with high levels of BFRs were imported to Indonesia with end-of-life electronics, or if it came from locally produced e-waste. These plastics also led to an increased level of PBDD/Fs in eggs from Tangerang as consequence of burning plastics with high content of BFRs. Indonesia is one of the suspected destinations for e-waste exports from developed countries as well (WorldLoop 2013), and such imports to Indonesia have been identified in the past years (Agustina 2007).

Indonesia, just as other developing countries that can be destinations for e-waste exports, need better definitions between repairable and non-repairable electronics, which is in fact hazardous waste often exported as a second-hand product. As it is written in the Basel Convention's Technical guidelines on e-waste, *"The lack of clarity in defining when used equipment is waste and when it is not has led to a number of situations where such equipment is exported to, in particular, developing countries ostensibly for reuse but where a large percentage of the exported equipment is in fact not suitable for further use or is not marketable and must be disposed of as waste in recipient countries*". (Basel Convention 2015).

The problem is that there is still not a sufficient definition of the distinction between waste and non-waste with regard to used electronics. The "repairable loophole" in the current provisional guidelines should be rejected in order to have precise definition of e-waste that will protect developing countries better (Petrlik, Puckett et al. 2019). It will help them also to address their domestic e-waste.

7.2 STRENGTHENING THE STOCKHOLM CONVENTION WASTE PROVISIONS

This study demonstrates the presence of POPs substances in chicken eggs that are regulated under the Stockholm Convention such as PCDD/Fs, PCBs, PBDEs, HBCD, HCB, PeCB, SCCPs, PFOS, and PFOA.

Unintentional production of dioxins and PCBs should be addressed under the treaty by preventing uncontrolled combustion. The Stockholm Convention requires minimization and, where possible, elimination of these substances produced unintentionally. The treaty has identified dioxin sources including uncontrolled burning (illustrated at the Tropodo and Bangun sites) and waste incinerators (Stockholm Convention on POPs 2008, UNEP and Stockholm Convention 2013).

Parties to the Convention are obliged to develop an action plan to address these sources and advance toward the minimization and elimination goal, and they are obliged to implement the plan according the Article 5 of the Convention (Stockholm Convention 2010). Open burning as well as waste incineration are not preferable options of waste management according to the Stockholm Convention and its BAT/BEP Guidelines (Stockholm Convention on POPs 2008) in particular.

Provisions under the Stockholm Convention may also allow control of POPs present in plastic and paper waste such as SCCPs, PBDEs, PFOS and PFOA by using stricter limit values to define POPs waste (known as low POPs content levels). Waste with levels of these substances over the limit must be destroyed and not exported. However, the threshold values are currently weak, and therefore allow exports of large volumes of POPs in waste across borders from developed countries (IPEN 2019).

Two case studies from the sites in Indonesia, Tropodo and Jombang Regency, underline the need for more strict control of POPs in waste that are allowed to be used freely at places in direct or indirect contact with agricultural or rural areas where local food is produced and/or within residential areas. The demonstrated cases show also that waste with a dioxin content even below 1 ppb should be restricted from direct use in such areas. In addition, the application of fly ash and other wastes containing levels of dioxin over 0.05 ppb in land-based applications can lead to unacceptable contamination of the local food chain.

In other studies, even lower levels of dioxins in soils led to contamination of free-range chicken eggs exceeding the EU standard for food. In agreement with a previous analysis done by the Swedish EPA, we therefore suggest restrictions to use any waste containing PCDD/Fs and dl-PCBs above 50 pg g^{-1} dw (0.05 ppb) on the surface without its stabilization or other pre-treatment that will prevent dioxin releases into the environment. The suggested values for LPCL and the additional limit for the use of waste on the surface are based on more cases and studies around the world (Weber, Watson et al. 2015, Katima, Bell et al. 2018, Petrlik, Adu-Kumi et al. 2019, Weber, Bell et al. 2019).

7.3 IMPLEMENTATION OF THE STOCKHOLM CONVENTION'S PROVISIONS RELATED TO UNINTENTIONALLY PRODUCED POPs

The levels of contamination of food chains represented by the results of analyses of free-range chicken eggs for PCDD/Fs and dl-PCBs presented in this study demonstrate the need to focus on actions to dramatically reduce generation of UPOPs, and particularly dioxins. All practices of waste management of plastic wastes in the observed cases in Bangun, Tangerang and Tropodo in this study, show a lack of implementation of best environmental practice (BEP) to avoid burning of plastic waste that can lead to creation of vast volumes of dioxins and dl-PCBs.

International guidelines and rules are still lacking in guidance for the decisionmakers on steps toward substitution of such materials as PVC or plastics containing brominated compounds, although it is suggested in the Article 5 c) of the Stockholm Convention: *"Promote the development and, where it deems appropriate, require the use of substitute or modified materials, products and processes to prevent the formation and release of the chemicals listed in Annex C, taking into consideration the general guidance on prevention and release reduction measures in Annex C and guidelines to be adopted by decision of the Conference of the Parties."*

In addition, Annex C, Part V adds: *"Priority should be given to the consideration of approaches to prevent the formation and release of the chemicals listed in Part I. Useful measures could include: (d) Replacement of feed materials which are persistent organic pollutants or where there is a direct link between the materials and releases of persistent organic pollutants from the source"* (Stockholm Convention 2010).

Places where open burning of plastic waste occurs, like Bangun or Tangerang in this study and/or African sites like Agbogbloshie, dumpsites in Yaoundé (Petrlik, Adu-Kumi et al. 2019) and many others, would benefit from a phase-out of PVC on the broadest possible scale. There is enough evidence that burning of PVC is linked to the generation of dioxins, and this and other chlorine containing plastics can lead also to formation of dioxins in Tropodo. For example, BAT/BEP Guidelines of the Stockholm Convention for residential combustion sources suggest: *"Many studies show that combustion of chlorine containing waste such as PVC, leads to increased formation of unintentional persistent organic pollutants as shown in Table 7*[32] *(Gullett, Lemieux et al. 1999) A regulation specifying standard fuels could be implemented"* (Stockholm Convention on POPs 2008). This suggestion is followed by a table showing the results of burning PVC (see Table 17).

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[32] Data from Table 7 in referenced literature were converted into Table 17 in this study.

The BAT/BEP Guidelines of the Stockholm Convention also discourage use of artisanal secondary aluminum production: ”*Artisanal and other small-scale aluminum recovery processes are used in a number of countries. Achievable performance limits are not applicable to artisanal and small-scale aluminum recovery processes as the processes used cannot be considered best available techniques or best environmental practices and ideally would not be practised at all*“ (Stockholm Convention on POPs 2008).

If it is used anyway, the guidelines suggest: ”*However, where artisanal and other small-scale aluminum recovery processes are practised, certain measures can be put in place in order to reduce the amount of pollutants released into the environment. Measures to reduce emissions of persistent organic pollutants and other pollutants from artisanal processes include pre-sorting of scrap material, selecting a better fuel supply (oil or gas fuels instead of coal), adequate ventilation, filtration of exhaust gases, proper management of wastes and proper choice of degasifiers*“ (Stockholm Convention on POPs 2008). It is also not suggested to use the slags produced by aluminum production on land as it ”*can have an environmental impact*“ (Stockholm Convention on POPs 2008)

Table 17: *The relation of PCDD/F emission factors on PVC content in burned material. Source: (Stockholm Convention on POPs 2008)*

PVC content [%]	0	0.2	1	7.5
Average Emission factor in ng I-TEQ kg^{-1}	14	80	200	4,900
Range n ng I-TEQ kg^{-1}	2 - 28	9 - 150	180 - 240	3,500 – 6,700

Globally, steps taken by governments toward substitution of problematic materials such as PVC within the frameworks of the Stockholm Convention and Basel Convention would improve the situation at sites like those documented in this study. The BAT/BEP Guidelines were developed in order to assist countries in the implementation of obligations under Article 5 of the Stockholm Convention and prevent further formation and releases of dioxins and other UPOPs. These guidelines should be better applied to national policies and for the regulation of the pollution sources such as secondary aluminum smelters in Jombang, or waste incineration as it is practiced in Tropodo or Lakardowo.

8. Recommendations

Plastic and paper waste imports to Bangun and Tangerang result in complex POPs contamination. Waste present in Bangun and Tangerang obviously includes plastics from dismantled electronics. The use of the discarded plastic waste as fuel for production in tofu factories in Tropodo led to the creation of one of the most seriously POPs-contaminated sites in the world. This study links waste mismanagement and uncontrolled movement of plastic waste with contamination of the food chain in Indonesia. Bangun, Tangerang and Tropodo are just three examples of many similar sites in Southeast Asia.

Measures to address this issue include:

1. Prohibit combustion as a disposal option for plastic waste or as an example of the "circular economy." It should not be accepted as a best practice for plastic waste management.
2. Prohibit the combustion of plastics as a fuel for industrial operations due to the dioxin and other halogenated pollution generated in emissions and ash.
3. Restrict the use of halogen-containing synthetic fuels derived from plastics due to the persistent organic pollutants (POPs) that occur in emissions when burning such fuel.
4. Remediate sites contaminated with dioxins and other POPs to ensure that human health is protected and food chain contamination cannot continue to occur.
5. Increase monitoring of POPs chemicals in compliance with Stockholm Convention provisions, along with other pollutants of concern.
6. Update the Indonesian Stockholm Convention National Implementation Plan to evaluate the effectiveness of preventive measures and control of POPs in Indonesia.
7. Strictly apply the new provisions of the Basel Convention to block hazardous waste imports and control transboundary movement of plastic wastes, or enact a ban on plastic waste imports.
8. Enact a stronger international Beyond 2020 chemicals framework that includes work to reduce and eliminate PFASs as a class.
9. Reduce and minimize plastic production and use, and avoid the use of halogenated plastics or the addition of halogenated compounds in plastic production such as bromine, chlorine and fluorine.
10. Stricter control of potential imports of e-waste or end-of-life electronics to Indonesia.
11. Set a better system for sorting e-waste and prevent the use of plastics from electronics as fuel, or to be burned.
12. Substitution should be applied for plastics for which disposal, including burning, leads to the creation of UPOPs. This relates mainly to PVC, PVDC or plastics treated with any brominated compounds.

Secondary aluminum production is a major source of POPs contamination in Kendalsari, and potentially in a larger part of the Jombang regency. The Lakardowo hazardous waste incinerator is the most likely source of dioxin pollution in the nearby village of Sumberwuluh and/or it can be one of its important sources.

The following steps might improve the situation:

13. Implement the BAT/BEP guidelines of the Stockholm Convention for secondary aluminum production.

14. Prepare and/or update an action plan that would address pollution sources of UPOPs, one of the suggested measures, for which *"Each Party shall at a minimum take … to reduce the total release"* of these chemicals[33].
15. Reduce the use of aluminum and do not promote it as a replacement of plastic packaging for beverages or food.
16. The Lakardowo incinerator should be inspected for dioxin emissions as well as for waste management of residues (bottom, fly and boiler ash) after waste incineration.
17. Avoid halogenated compounds in inputs into thermal and combustion processes.
18. Use non-combustion alternative methods for treatment of hazardous waste, e.g. non-combustion technologies for POPs waste (IPEN Dioxin PCBs and Waste Working Group 2010, Basel Convention 2017) or medical waste (Emmanuel 2007, Stringer, Kiama et al. 2010, Emmanuel 2012, UNEP 2016) disposal.

Two case studies in this report demonstrated that ash and slag residues used for the building of roads, flood defenses and building foundations contributes significantly to the spread of pollution into locally grown food.

In order to prevent further contamination of food by POPs:

19. Remove ash contaminated with POPs used as building material, or at least prevent leachate and dust dispersion.
20. Introduce stricter, more protective limits for POPs in wastes in the frameworks of both the Stockholm and Basel Conventions.
21. Prohibit the use of wastes and materials with a concentration of dioxins and dl-PCBs exceeding a level of 50 pg TEQ g^{-1} dw (0.05 ppb) on the surface.

This and previous studies also show on gaps in monitoring of POPs and/or EDCs in environmental, food, wildlife, and human tissue samples in general. This leads us to the following suggestions:

22. Use international standards for monitoring dioxins in food (e.g. eggs) and undertake a mandatory number of analyses per year related to critical food items.
23. Use internationally accepted standards (such as as EC/644/2017); (European Commission 2017) for the analysis of dioxins/PCBs in food/feed using high-throughput screening tests (such as DR CALUX®) as well as chemical confirmative analysis.
24. Use screening tests (such as DR CALUX®), which allows easier, cost-efficient and high-capacity testing of not only PCDD/Fs and dl-PCBs, but also of PBDD/Fs[34], which are highly relevant to e-waste.
25. Immediately evaluate the most toxic mode of actions such as the well-described effects of endocrine-disrupting chemicals, "hormone-like" e.g. PBDEs (female hormone estrogen-like, inhibition male hormone androgen-like), TBBPA (thyroid transport competitor) and the related risks of sites contaminated with BFRs containing these EDCs.

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[33] "Develop an action plan or, where appropriate, a regional or subregional action plan within two years of the date of entry into force of this Convention for it, and subsequently implement it as part of its implementation plan specified in Article 7 …" Article 5 Stockholm Convention (2010). Stockholm Convention on Persistent Organic Pollutants (POPs) as amended in 2009. Text and Annexes. Geneva: 64.

[34] It is important to notice that the DR CALUX® method also includes already the brominated dioxins and biphenyls (PBDD/Fs and PBBs) without any further costs, while the chemical analyses need an additional expensive analysis. Globally, at the moment only a handful chemical laboratories are available to perform this additional chemical analysis routinely for brominated dioxins and biphenyls (PBDD/Fs and PBBs), while many laboratories already perform the DR CALUX® method. Such easy, low cost and high-capacity analysis tools are urgently needed in cases like these, with widespread contamination of brominated dioxins/biphenyls (PBDD/Fs/PBBs) in e-waste, products from recycled e-waste plastic, soil, feed/food and human biomonitoring (blood, mother milk).

ACKNOWLEDGEMENTS

We would like to acknowledge the financial support from the Government of Sweden, which made the chemical analyses and preparation of this report possible through a grant to IPEN, as well as the support received from other donors who supported our work on this study, the sampling in Indonesia and the analyses of the samples, in particular to the Global Greengrants Fund, and Plastic Solutions Fund. We would also like to thank all collaborators in Indonesia during the sampling process, and the personnel of the laboratories based in the Czech Republic, Germany and the Netherlands for the analyses, which often required their extra time.

ABBREVIATIONS

BDS – BioDetection Systems (laboratory in Netherlands)
BEQ – bioanalytical equivalent
BFRs – brominated flame retardants
bw – body weight
CALUX - chemically activated luciferase gene expression
br-PFOS – branched PFOS
BTBPE – 1,2-bis(2,4,6-tribromo-fenoxy) ethane
DBDPE – decabromodiphenyl ethane
DDD – dichlorodiphenyldichloroethane (a metabolite of DDT)
DDE – dichlorodiphenyldichloroethylene (a chemical compound formed by the loss of hydrogen chloride from DDT)
DDT – dichlorodiphenyltricholoroethane (pesticide)
dl-PCBs – dioxin-like PCBs
dw – dry weight
ECF – electrochemical fluorination
EDCs – endocrine-disrupting chemicals
EFSA – European Food Safety Authority
EPA – Environment Protection Agency
EU – European Union
fw – fresh weight
GC – gas chromatography
GPC – gel permeation chromatography
GPS – global positioning system
HBB – hexabromobenzene
HBCD – hexabromocyclododecane
HCB – hexachlorobenzene
HCBD – hexachlorobutadiene
HCHs – hexachlorocyclohexanes (pesticides and their metabolites)
HRGC-HRMS – high resolution gas chromatography – high resolution mass spectroscopy
IARC – International Agency for Research on Cancer
i-PCBs – indicator PCB congeners
IPEN – International Pollutants Elimination Network
LOD – limit of detection
LOQ – limit of quantification
MAC – maximum acceptable (allowable) concentration
ML – maximum level
MRL – maximum residue level
NA – not analyzed
N/A – not applicable
NC - not calculated
nBFRs – novel brominated flame retardants
ndl-PCBs – non-dioxin-like PCBs
NGO – nongovernmental organization (civil society organization)
NIP – National Implementation Plan
n-PFOS – linear PFOS
OBIND – octabromotrimethylfenylindane
OCPs – organochlorinated pesticides

PBDD/Fs – polybrominated dibenzo-p-dioxins and dibenzofurans
PBDEs – polybrominated diphenyl ethers
PBEB – pentabromoethylbenzene
PBT – pentabromotoluene
PCBs – polychlorinated biphenyls
PCDD/Fs – polychlorinated dibenzo-p-dioxins and dibenzofurans
PCDDs – polychlorinated dibenzo-p-dioxins
PCDFs – polychlorinated furans
PeCB – pentachlorobenzene
PFASs – per- and polyfluoroalkyl substances
PFDA - perfluorodecane acid
PFDoA - perfluorododecanoic acid
PFHxS - perfluorohexane sulfonate
PFOA – perfluorooctanic acid
PFOS – perfluorooctanesulfonic acid
PICs – products of incomplete combustion
POPs – persistent organic pollutants
PVC – polyvinyl chloride, one kind of broadly used plastic
PVDC - polyvinylidene chloride or polyvinylidene dichloride
SC – Stockholm Convention on Persistent Organic Pollutants
SCCPs – short-chain chlorinated paraffins
SMEs – small-medium enterprises
TBBPA – tetrabromobisphenol A
TDI – tolerable daily intake
TDS – total diet study
TEF – toxic equivalency factor(-s)
TEQ – toxic equivalent
TWI – tolerable weekly intake
UNDP – United Nations Development Programme
UNEP – United Nations Environment Programme
UPOPs – unintentionally produced POPs
US EPA – United States Environmental Protection Agency
WHO-TEQ – toxic equivalent defined by a WHO experts panel in 2005
WI – waste incinerator and/or waste incineration
ww – wet weight

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This publication has been produced with the support of the Government of Sweden, Global Greengrants Fund, Plastic Solutions Fund and International Pollutants Elimination Network (IPEN).

Project was implemented by Arnika – Toxics and Waste Programme, Nexus3 Foundation and Ecoton in cooperation with International Pollutants Elimination Network.

ISBN 978-80-87651-78-0