PLASTIC WASTE DISPOSAL LEADS TO CONTAMINATION OF THE FOOD CHAIN

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AUTHORS

***Jindrich Petrlik*[1,2] – *Lee Bell*[1,3] – *Bjorn Beeler*[1] – *Marketa Møller*[2] – *Miroslava Jopkova*[2]**

Supporting Data
Prigi Arisandi[4] – Karolína Brabcová[1,2] – Maria Carcamo[5] – Sofía C. Chávez Arce[6] – Thony Dizon[7] – Yuyun Ismawati Drwiega[8] – Gilbert Kuepouo[9] – Silvani Mng'anya[10] – Griffins Ochieng Ochola[11] – Martin Skalsky[12]

Affiliation of the contributors
[1] IPEN, Gothenburg, Sweden
[2] Arnika – Toxics and Waste Programme, Prague, Czech Republic
[3] NTN – National Toxics Network, Australia
[4] Ecoton, Indonesia
[5] RAP-AL – Red de Acción en Plaguicidas y sus Alternativas para América Latina, Uruguay
[6] CasaCem, Mexico
[7] EcoWaste Coalition, Philippines
[8] Nexus for Health, Environment, and Development, Indonesia
[9] CREPD – Centre de Recherche et d'Education pour le Développement, Cameroon
[10] AGENDA for Environment and Responsible Development (AGENDA), Tanzania
[11] CEJAD – Centre for Environmental Justice and Development, Kenya
[12] Arnika - Citizens Support Centre, Prague, Czech Republic

CONTENTS

Praeksa, Thailand. Photo: Jindrich Petrlik, Arnika

EXECUTIVE SUMMARY

Waste generated from the use of plastics is a challenge for the whole of human society. Plastics are everywhere around us, and we can find tiny parts of plastics in even the most pristine places. Most plastics were invented by chemical scientists, and in order to make the plastic suitable for many different uses or to make them meet legislative requirements for fire safety, for example, they need chemical additives that make the plastic resistant, flexible, durable or less flammable. Many of these additives have not only been found to be toxic in themselves, but also lead to the creation of new chemicals, like brominated and chlorinated dioxins, when burned. These new chemicals can be even more toxic than the original additives.

MANY [PLASTIC] ADDITIVES HAVE NOT ONLY BEEN FOUND TO BE TOXIC IN THEMSELVES, BUT ALSO LEAD TO THE CREATION OF NEW CHEMICALS, LIKE BROMINATED AND CHLORINATED DIOXINS, WHEN BURNED.

The focus of this study has been on very toxic persistent organic pollutants (POPs) entering the food chain at locations where plastic waste is being recycled, burned, incinerated or dumped. Samples of free-range chicken eggs were analyzed for brominated and chlorinated dioxins (PBDD/Fs and PCDD/Fs), dioxin-like PCBs (dl-PCBs), BFRs, SCCPs, and PFASs. Free-range chicken eggs are sensitive indicators of POP contamination in soils/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 BFRs. Thirty-five pooled and one individual free-range eggs from twenty-five different locations worldwide were analyzed for selected POPs in accredited laboratories.

The levels of some POPs measured in the collection of free-range egg samples included in this study are among the ten highest levels ever measured globally:

- PCDD/Fs in four samples in this study
- PBDD/Fs in seven samples in this study
- PBDEs in four samples in this study and
- HBCD in six samples in this study.

In eggs from an e-waste scrap yard in Agbogbloshie, Ghana, the levels measured were the highest ever measured level of brominated dioxins, the second highest level of chlorinated dioxins, the fifth highest level of HBCD, and the eighth highest level of PBDEs.

In eggs from Tropodo, Indonesia, we found the second highest level of PDBEs ever measured, and the sixth highest level of chlorinated dioxins. In eggs from Pitarne, Czech Republic, we found the third highest level of HBCD ever measured as a result of the hens picking at polystyrene foam treated with HBCD.

Eggs from e-waste and plastic waste yards represent the most critically contaminated egg samples in this study.

Eight out of the thirty-six samples presented in this study have levels of dioxins above 20 pg TEQ g^{-1} fat (see Tables A1-2, A1-3 and A1-4 in Annex 1), which is ten times more than the EU limit for dioxins in eggs as food, and only one sample had a PCDD/Fs + dl-PCBs level below the EU limit for eggs as food (5 pg TEQ g^{-1} fat).

The EU limit for dioxins (2.5 pg TEQ g^{-1} fat) was exceeded in 31 (out of 35) samples by 1.5 – 264 times. In four samples the level was below the EU limit, but only one of them was also below the limit for PCDD/Fs and dl-PCBs combined (5 pg TEQ g^{-1} fat).

For eggs contaminated with highest levels of dioxins and dl-PCBs, an adult person weighing 70 kg can reach the Tolerable Daily Intake (TDI) limit, set by EFSA, by eating just 4 thousandths and one hundredth of an egg in the case of the samples from Agbogbloshie and Tropodo respectively. The same can be reached by eating 4 and 5 hundredths of an egg from Tangerang and Samut Sakhon or one of the samples from Aguado respectively.

In some cases, brominated dioxins contribute significantly to the total TEQ levels in the egg samples and also at the same time to the total dioxin exposure of the human body, in particular in egg samples from the sites

Samut Sakhon, Thailand. Photo: Jindrich Petrlik, Arnika

affected by e-waste, because those plastics have originally been treated with BFRs (Agbogbloshie, Wuhan, Tangerang, Samut Sakhon, Bagong Silang, and Guadal).

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 TDI for PFOS from 3 to almost 16 times.

The levels of POPs present in the free-range chicken egg samples show that current plastic waste sorting, dumping and open burning practices lead to serious contamination of the food chain in developing countries. Recycling of some kinds of plastics can also lead to serious contamination with POPs as shown in some of the examples included in this study. This applies to PVC and e-waste in particular.

The eggs represent only a segment of domestic animal products used for food that might be contaminated with POPs, as at some hot spots there were also, for example, cows or camels spotted foraging at the waste dumpsites or waste yards. The scale of the food contamination can therefore be much larger in some of the localities included in this study.

RECOMMENDATIONS

Stop increasing and start decreasing production and use of plastics is one major recommendation we can give. This report is focused only on certain segments of contaminants released from plastic waste burning, but the burning of plastics can release a much broader range of toxic chemicals, so plastic waste prevention is the most critical measure that must be taken in order to prevent further releases of POPs at sites like the ones presented in this study.

Stop plastic and electronic waste exports. There is a clear link between current global policy that allows uncontrolled movement of plastic waste or e-waste and toxic chemical contamination of the food chain where dumping occurs, such as Agbogbloshie, Tropodo, Tangerang and many other sites presented in this study. To stop plastic and e-waste exports to countries with inadequate capacities for their environmentally sound management, the regulatory measures of international conventions' must be strengthened: countries should adopt the Basel Convention Ban Amendment, a new amendment on plastic waste should be applied and more strict measures to control POPs in waste should be introduced.

The current provisional e-waste guidelines under the Basel Convention contain a loophole that allows for e-waste export under the guise of 'export for repair'. This industry-promoted loophole makes the guidelines contradictory to the Convention because electronic products at end-of-life are hazardous waste. This loophole should be closed to preserve the integrity of the treaty.

Strengthen Low POPs content levels. The hazardous waste limits in the Stockholm Convention should prevent the export of POPs waste, including plastic waste containing high levels of BFRs. These limits are currently too weak to be effective. Currently, the existing and proposed limits for POPs found in e-waste and generated by its 'recycling' in developing countries are far too weak and allows the trade to continue. This includes limits for chlorinated dioxins/furans, flame-retardant chemicals such as PBDEs and HBCD, and short-chain chlorinated paraffins. These stricter limits (defined as Low POP Content in the Stockholm Convention) should be 50 mg/kg for PBDEs, 100 mg/kg for HBCD and SCCPs and 1 ug TEQ/kg for PCDD/Fs (1 ppb) at a maximum. The Stockholm Convention could be further strengthened by listing brominated dioxins.

Some cases in this report demonstrate that ash residues used for construction, paving roads or simply dumped contribute significantly to the spread of pollution by dioxins and other POPs. Prohibition of the use of wastes and materials with concentrations of dioxins and dl-PCBs exceeding a level of 50 pg TEQ g-1 dw (0.05 ppb) on the soil surface should be applied in addition to other POPs waste limits, in order to prevent further contamination of the food chain.

Toxic additives in plastics should be banned without any exemptions, and existing exemptions such as for use of decaBDE, PFOS, PFOA and SCCPs should end as soon as possible. The weak regulatory level set for trace contamination with PBDEs in the EU that allows recycling of POPs in plastics should also end.

Prevent creation of dioxins from burning or incineration of plastic waste. Instead of trying to improve dioxin-producing technologies such as small medical waste incinerators, a strategy that prevents dioxin formation is desired. Non-combustion technologies that can be used for medical waste and POPs-containing waste treatment are available. PVC should be substituted in as many applications as possible in order to prevent dioxin releases from its burning or incineration.

Tropodo, Indonesia. Photo: Jindrich Petrlik

1. INTRODUCTION

We live in the age of plastics. Plastics are everywhere around us, and tiny parts of plastics can be found even in the most pristine environments. Most kinds of plastics have been invented by chemical scientists, and in order to make them suitable for many different uses or meet legislative requirements for fire safety, for example, they need chemical additives that make the plastic resistant, flexible, durable or less flammable. Many of these additives have been found to be toxic in themselves, but in addition, when burned they also lead to the creation of new chemicals that can be even more toxic than the original additives.

When burned, halogenated plastic such as polyvinyl chloride (PVC) leads to the formation of chlorinated dioxins[1] (Stockholm Convention on POPs 2008). The burning of non-halogenated plastics creates other toxics still as well as persistent chemicals such as polycyclic aromatic hydrocarbons (PAHs).

TOXIC CHEMICALS ARE RELEASED INTO THE ENVIRONMENT NOT ONLY DURING THE PRODUCTION AND THE USE OF PLASTICS, BUT ALSO DURING THEIR DISPOSAL.

It is well established that toxic chemicals are released into the environment not only during the production and the use of plastics, but also during their disposal (Hahladakis, Velis *et al.* 2018, Basel Convention Secretariat and Stockholm Convention Secretariat 2019), in particular when burning or incineration is involved (Blankenship, Chang *et al.* 1994, Thornton, McCally *et al.* 1996, Yasuhara, Katami *et al.* 2006, Stockholm Convention on POPs 2008).

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[1] Dioxins are used as a synonym for a group of 210 chemicals – polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs). We here use the terms chlorinated dioxins and brominated dioxins, as there is also a group of polybrominated dibenzo-p-dioxins and dibenzofurans (PBDD/Fs).

In brief: Waste generated from the use of plastics is a challenge for all of society.

Some developing countries, such as Ghana, Indonesia, Malaysia, Nigeria, the Philippines, Tanzania, Thailand and Vietnam have become destinations for waste exports, including plastic waste, paper for recycling and/or electronic waste (e-waste). These wastes may contain a whole range of POPs added intentionally to the products now present in the waste chain, including brominated flame retardants (BFRs), short-chain chlorinated paraffins (SCCPs), and per- and polyfluoroalkyl substances (PFASs).

The issue with plastic is not only a waste management problem, as claimed by the plastic industry (Dunn 2019). Instead, it is something that has to be solved by minimization of its production from the start. Less burned plastic will also decrease the generation of vast amounts of toxic chemicals, including POPs.

In this study, free-range chicken eggs have been used to investigate POPs contamination of the food chain in the vicinity of plastic waste disposal sites and facilities.

Free-range chicken eggs are sensitive indicators of POPs contamination in soils and dust and 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 can be used to reveal POPs contamination, particularly in areas impacted by dioxins (PCDD/Fs) and PCBs (Pless-Mulloli, Schilling *et al.* 2001a, Pirard, Focant *et al.* 2004, DiGangi and Petrlik 2005, Shelepchikov, Revich *et al.* 2006, Aslan, Kemal Korucu *et al.* 2010, Arkenbout 2014), as well as by BFRs (Blake 2005, Luo, Liu *et al.* 2009, Zhao, Zhou *et al.* 2009, Polder, Müller *et al.* 2016, Petrlik, Kalmykov *et al.* 2017, Hogarh, Petrlik *et al.* 2019).

We decided to include the following types of plastic waste management and disposal operations and facilities:

1. plastic and electronic waste yards;
2. waste dumpsites with significant amounts of plastic wastes;
3. recycling and shredder plants which deal with significant amounts of plastic waste;
4. waste incineration and co-incineration operations.

This study focused on POPs, whose releases are closely related to plastic wastes. The POPs include additives in the plastic as such, as well as unintentionally produced POPs (UPOPs) generated mostly by burning,

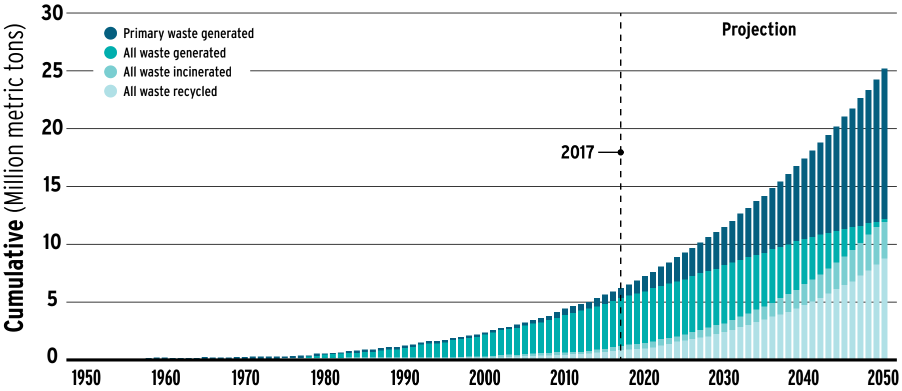

Figure 1: Historical data and projections to 2050 of plastic waste production and disposal. "Primary waste" is plastic becoming waste for the first time and doesn't include waste from plastic that has been recycled. Source: Geyer, Jambeck *et al.* 2017, Guglielmi 2017

incineration and/or other thermal treatment of plastics. We focused on POPs already listed in the Stockholm Convention, representing additives to plastic, and UPOPs.

There are specific risks in the burning of plastics treated with BFRs because of the creation of brominated dioxins, which exhibit a similar toxicity to that of chlorinated dioxins. They have been found in fish (Falandysz, Smith *et al.* 2020), and in eggs (Pajurek, Pietron *et al.* 2019). Brominated dioxins were also recently found to be responsible for high dioxin-like toxicity in toys made of black recycled plastic (Budin, Petrlik *et al.* 2020). Brominated dioxins were therefore included into the range of chemicals analyzed in the free-range chicken eggs from sites affected by plastic waste.

A more thorough description of the substances analyzed in the eggs in this study can be found in chapters 1.2 – 1.5. This report focused only on a select group of contaminants released from plastic waste burning, but the burning of plastics can release a much broader range of toxic chemicals than the ones mentioned in this study.

IPEN, in cooperation with its participating organizations; Arnika in the Czech Republic, the Center for Environmental Solutions in Belarus, CREPD in Cameroon, Green Beagle in China, Nexus3 Foundation and Ecoton in Indonesia, EcoMuseum Karaganda in Kazakhstan, CEJAD in Kenya, Casa Cem in Mexico, EcoWaste Coalition in the Philippines, EcoMuseum and EcoMangystau in Kazakhstan, Agenda in Tanzania, EARTH in Thailand, and RAPAL in Uruguay, conducted chemical analyses of free-range chicken egg samples collected by their experts and/or by local groups, in order to assess the potential presence of toxic substances in the environment of the people living in the vicinity of plastic waste handling and disposal facilities.

Some of the samples presented in this study had already been analysed for previous studies. We included them here if they came from a locality relevant for this study and where the analysis was not older than six years. Free-range chicken eggs from Gatovo (Belarus), Bangun, Tangerang, and Tropodo (all in Indonesia), Samut Sakhon and Praeksa (both in Thailand), Agbogbloshie, Accra and Kumasi (all in Ghana), Yaounde (Cameroon), and Wuhan (China) were either analyzed or their data used in previous studies focused on specific countries or regions like Africa (Petrlik, Adu-Kumi *et al.* 2019), Ghana (Hogarh, Petrlik *et al.* 2019), China (Petrlik 2016), Kazakhstan (Petrlík, Kalmykov *et al.* 2016) and/or Thailand (Mach, Petrlík *et al.* 2017, Petrlík, Dvorská *et al.* 2018, Teebthaisong, Petrlik *et al.* 2018). Eggs bought in supermarkets were chosen as reference samples for comparison with those from potentially contaminated sites. Samples from supermarkets in Accra, Bangkok, Beijing, Jakarta, and Karaganda were used as reference samples in some previous studies (Petrlik 2016, Petrlík, Kalmykov *et al.* 2016, Petrlík, Dvorská *et al.* 2018, Petrlik, Adu-Kumi *et al.* 2019, Petrlik, Ismawati *et al.* 2020). Their analysis was conducted for those previous studies and then also included in this study.

1.1 TOXIC ADDITIVES IN PLASTICS

Plastics and food packaging contain chemical contaminants from manufacturing along with many additives to make them inflammable (flame retardants), more flexible (plasticizers), grease-resistant (fluorinated chemicals known collectively as PFASs), sterile (biocides), and other substances to create many other properties. Many of these additives are toxic and they leak from products during use and can be released during recycling and 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 risk of allergy and asthma, and have an adverse impact on children's neurological development (Jurewicz and Hanke 2011). Many of the additives in plastics were found to last for a long time in the environment and accumulate in animals, and some


Baskuduk, Kazakhstan. Photo: Magda Slamova, Arnika

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). Other additives in plastics, such as BFRs, SCCPs and/or PFASs, also exhibit serious impacts on human health which are described in sub-chapters 1.2 – 1.4.

Substances of concern 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 referred to 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 more closely described in sub-chapter 1.5.4.

Some of the additives in plastics such as short-chain chlorinated paraffins (SCCPs), polybrominated diphenyl ethers (PBDEs), hexabromocyclododecane (HBCD) as well as by-products of their burning (PCDD/Fs, dioxin-like PCBs or hexachlorobenzene), are already regulated under the Stockholm Convention (Stockholm Convention 2010, Stockholm Convention 2017). In addition, 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 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 applying the precautionary principle or any restriction (DiGangi, Blum *et al.* 2010).

PBDEs are of primary interest to 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. There is only limited information 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 Hexabromocyclododecane (HBCD)

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

HBCD was listed in Annex A of the Stockholm Convention for global elimination with a five-year specific exemption for use in building insula-

Agbogbloshie, e Ghana. Photo: Martin Holzknecht, Arnika

tion that expired for most Parties in 2019 (Stockholm Convention 2013). This chemical also belongs among the substances of very high concern (SVHC) under the REACH legislation.

1.2.2 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, but are not chemically bound to the material and therefore leach into the environment. They have already been identified in breast milk in Indonesia, in research more than one decade ago, and *"the levels were in the same order as those in Japan and some European countries, but were one or two order 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 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 plastic waste and/or by its incineration all of the mentioned PBDEs were part of the main focus of our investigation.

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 additional additives in plastics which might be also 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) is 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 Statements 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 scientists called upon decision makers for regulatory assessment to address PFASs in 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 the lipid metabolism and 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 toxicological profiles of PFASs (Blum, Balan *et al.* 2015, ATSDR 2018, Ritscher, Wang *et al.* 2018, Fenton 2019).

The European Food Safety Authority (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

Yaounde, Cameroon. Photo: CREPD

substances in Indonesia found that they are unregulated and contaminate 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 to understand the difference in 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) was listed in the Stockholm Convention in 2009 together with its salts, and 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, in the semiconductor industry, in firefighting foams, ski waxes, paper packaging from 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 and it is 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 surfactant (foam formation for reduction

Libreville-Ozounge. Ministry of Forestry, Fisheries and Env., Gabon

of fuels fires) and surface protector (metal plating processes, consumer products such as carpets, textile, and in leather industry). It is one of the most persistant 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 also with the indoor dust inhalation or from consumer products containing PFHxS or its precursors (POP RC 2019). PFHxS can trigger hypersensitivity and suppression of immune system (asthma, allergic reactions), changes of lipids and protein metabolism pathways, changes in liver and thyroid functioning, and can also affect 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 countries in which the samples presented in this study were taken. Fourteen samples of free-range chicken eggs and one reference sample of eggs from a supermarket in this study were analyzed in the laboratory of the University of Chemistry and Technology in Prague, Department of Food Chemistry and Analysis. Analysis was performed for 17 PFASs, both individual substances and/or their groups.[2] We also refer to other scientific references on PFASs as a group or to other ones not described here (Blum, Balan *et al.* 2015, ATSDR 2018, Ritscher, Wang *et al.* 2018, Fenton 2019).

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), dioxin-like PCBs (dl-PCBs), polychlorinated dibenzo-p-dioxins (PCDD), polychlorinated dibenzofurans (PCDF), and polychlorinated naphthalenes. Analyses of eggs in this study covered PCDD/Fs and dl-PCBs. Polychlorinated naphthalenes were not analyzed. We also focused on brominated dioxins (PBDD/Fs) which are not listed in the Stockholm Convention yet.

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 are therefore 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 6, sometimes also 7, 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 experts panel in 2005 (van den Berg, Birnbaum *et al.* 2006). These TEFs were used to evaluate dioxin-like toxicity in pooled samples of chicken eggs 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

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[2] 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

Sampling of soot from medical waste incinerator in hospital at Kumasi, Ghana in 2018. Photo: Martin Holzknecht, Arnika

birth defects and stillbirths. Fish exposed to these substances died shortly after the exposure ended. Food (particularly from animals) is the major source of dioxin 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 US during the Vietnam war.[3] The production of 2,4,5 T pesticide as the 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 made workers sick 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).

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[3] 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 a further two million people have suffered cancers or other illnesses, which can be also 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.

1.5.2 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 of the presence of polybrominated dibenzo-p-dioxins and dibenzofurans (PBDD/Fs)[4] in the food chain has arisen, 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 to the sampled sites with e-waste and/or plastic waste which may contain brominated flame retardants, such as 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, so PBDD/Fs were therefore also analyzed in fifteen free-range chicken egg samples in this study.

PBDD/Fs have been known to be potential by-products of commercial PBDE mixtures since 1986 (Buser 1986). They were also found to be by-products of some novel BFRs like DBDPE (Brenner and Knies 1990) or BTBPE (Ren, Zeng *et al.* 2017, Zhan, Zhang *et al.* 2019). This is similar to the chlorinated dioxins which have been observed as impuritities in PCBs, and other chlorinated chemicals. PBDFs have also 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). 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).

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

> *"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 unregulated substances, there is less data about their presence in the environment. There is also very little information about their presence in food and/or consumer products, where they can have direct impacts on human health, including for vulnerable groups such as children and women of childbearing age. This applies in particular to developing countries, but there are also a large number of EU member states that do not control PBDD/Fs in food or waste incineration emissions, for example.

drej Petrlik

2. SAMPLING AND ANALYTICAL METHODS

The samples of free-range chicken eggs and reference eggs from Gabon, Indonesia, Kenya, Mexico, the Philippines, Tanzania and Uruguay were sampled during the period from April 2019 until January 2020. Samples from Pitarne in the Czech Republic were taken by the end of 2017 as part of the project focused on plastic waste recycling sites, and reference samples were collected in Prague in April 2018 and February 2019. One additional sample from Pitarne was taken in August 2020.

The analyses of recently sampled eggs were conducted in European laboratories between June 2019 and March 2020 closely following sampling campaigns in the above-mentioned countries.

Previously sampled eggs were analyzed in the same laboratories, but in previous years, and the description of their sampling and analyses is included in previous reports for Africa (Petrlik, Adu-Kumi *et al.* 2019), China (Petrlik 2016), Kazakhstan (Petrlík, Kalmykov *et al.* 2016), and Thailand (Mach, Petrlík *et al.* 2017, Petrlík, Dvorská *et al.* 2018).

Thirty-five pooled samples and one individual sample of free-range chicken eggs were collected at twenty-five hot spots in fourteen countries, over four continents. Just as in previous studies, six samples of eggs purchased in a supermarket or convenient stores (in Prague, Accra, Beijing, Jakarta, Karaganda, and Bangkok) served as background samples as they were not from free-range hens and therefore unlikely to be exposed to POPs chemicals in soil (DiGangi and Petrlik 2005). A basic description of these twenty-five localities 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. In one case it was not possible to obtain more eggs, so we then decided to analyze an individual egg (PH-E-7) and returned to potentially resample that site, which was successful (see sample PH-E-S-5/2). 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 month and year sampling occurred.

DR CALUX: Free-range chicken eggs from the seventeen pooled samples (for specific egg samples see Tables in Annex 1) and three pooled samples of commercial eggs (non free-range) were analyzed for polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs)[5] and 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% (v/v) FCS under standard conditions (37°C, 5% CO_2, 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 (analysed 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).

TABLE 1: OVERVIEW OF SAMPLES OF CHICKEN EGGS IN THIS STUDY.

Country	Activity	Locality	Sample ID	Matrix	Month/ year of sampling	Number of eggs in pooled sample	Fat content (%)
Belarus	RE	Gatovo	Gatovo	Eggs	06/2014	3	15.4
Cameroon	DU	Yaoundé - TKC Quart.	YA-1	Eggs	08/2018	6	19.6
Cameroon	WI	Yaoundé - hospital	YA-2	Eggs	08/2018	5	14.6
Cameroon	DU	Yaoundé - Etetar Quart.	YA-3	Eggs	08/2018	6	14.3
Czech Rep.	RE	Pitarne	N1-3	Eggs	11/2017	3	12
Czech Rep.	RE	Pitarne	S1-4	Eggs	11/2017	4	12.1

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

Country	Activity	Locality	Sample ID	Matrix	Month/ year of sampling	Number of eggs in pooled sample	Fat content (%)
Czech Rep.	RE	Pitarne	PIT03	Eggs	09/2017	3	13
Czech Rep.	RE	Pitarne	PIT 2/2020	Eggs	08/2020	5	10.3
Czech Rep.	Ref	Prague	PHA-1 and 2	Eggs	04/2018 02/2019	6 and 10	10.2
Gabon	WI	Nkoltang	GA-E-NKOL	Eggs	11/2019	5	13.6
Gabon	DU	Libreville - Owendo	GA-E-OWE	Eggs	11/2019	5	13.8
Gabon	DU	Libreville - Ozounge	GA-E-OZOU	Eggs	11/2019	5	11.2
Ghana	WY-E	Agbogbloshie	AGB-E	Eggs	12/2018	4	14.7
Ghana	Ref	Accra (supermarket)	ACC-M-E	Eggs	12/2018	6	8.8
Ghana	WI	Accra – hospital	KBI-E	Eggs	12/2018	6	12.3
Ghana	WI	Kumasi – hospital	KU-E	Eggs	12/2018	5	14.7
China	WI	Wuhan	Wuhan 2	Eggs	09/2014	3	12.5
China	WI	Wuhan	Wuhan 1	Eggs	03/2014	6	15.5
China	Ref	Beijing	Control	Eggs	10/2014	3	10.1
Indonesia	WY-E	Bangun	Bangun 1	Eggs	05/2019	3	13
Indonesia	WY-E	Bangun	BAN-E-1	Eggs	11/2019	3	9.5
Indonesia	WY-E	Tangerang	SEM-E-1	Eggs	11/2019	3	16.2
Indonesia	WY-E	Tangerang	TAN-ESIN-01	Eggs	11/2019	5	13.7
Indonesia	WI	Tropodo	Tropodo 1	Eggs	05/2019	3	15
Indonesia	WI	Tropodo	TROP-E-1	Eggs	10/2019	6	13.9
Indonesia	Ref	Jakarta	JAK-SUP	Eggs	11/2019	6	9.5
Kazakhstan	DU	Baskuduk	BAS 02	Eggs	10/2016	3	15.6
Kazakhstan	Ref	Karaganda	KAR-SU	Eggs	04/2015	6	14
Kenya	WI	Nairobi – Mirema	KE_001	Eggs	01/2020	5	14.0
Mexico	RE	Guadalajara	GUDAL-EGG1	Eggs	04/2019	5	14
Philippines	WY-E	Bagong Silang	PH-E-1-2	Eggs	09/2019	2	13.8

Country	Activity	Locality	Sample ID	Matrix	Month/ year of sampling	Number of eggs in pooled sample	Fat content (%)
Philippines	WI	Aguado	PH-E-3-4	Eggs	11/2019	4	16.1
Philippines	WI	Aguado	PH-E-5-6	Eggs	11/2019	3	13.0
Philippines	WI	Aguado	PH-E-7	Eggs	11/2019	1	14.4
Philippines	WI	Aguado	PH-E-S-5/2	Eggs	01/2020	4	12.4
Tanzania	DU	Pugu Kinyamwezi	TZ-PU-KI_EGG	Eggs	01/2020	9	18.0
Thailand	Ref	Bangkok	supermarket	Eggs	02/2016	6	11.6
Thailand	WY-E	Samut Sakhon	Samut Sakhon	Eggs	02/2015	3	11.6
Thailand	WY-E	Samut Sakhon	SMS 2-13	Eggs	02/2016	3	19.4
Thailand	DU	Praeksa	PKS-EGG-1	Eggs	11/2015	4	18.1
Uruguay	DU	Cerro de Montevideo	UR-CM-E	Eggs	09/2019	4	8.9
Uruguay	DU	Minas	UR-MIN-E	Eggs	09/2019	4	11.8

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, chlorinated and brominated polycyclic aromatic hydrocarbons, N-dioxins)6; however, for confirmation it is recommended to go for more specific PCDD/Fs and dl-PCBs congener analyses, which also allows examination of finger prints of dioxins (PCDD/F congener patterns), specific for different sources of pollution. Thirty-four pooled and one individual free-range egg samples as well as all six commercial eggs samples 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

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6 "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 may 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.

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 analysis for brominated dioxins. The sample from Praeksa, Thailand was analyzed in Axys Varilab laboratory in the Czech Republic, also by HRGC-HRMS. Only one sample from Yaounde, TKC Quarter, was not analyzed for PCDD/Fs and dl-PCBs by HRGC-HRMS.

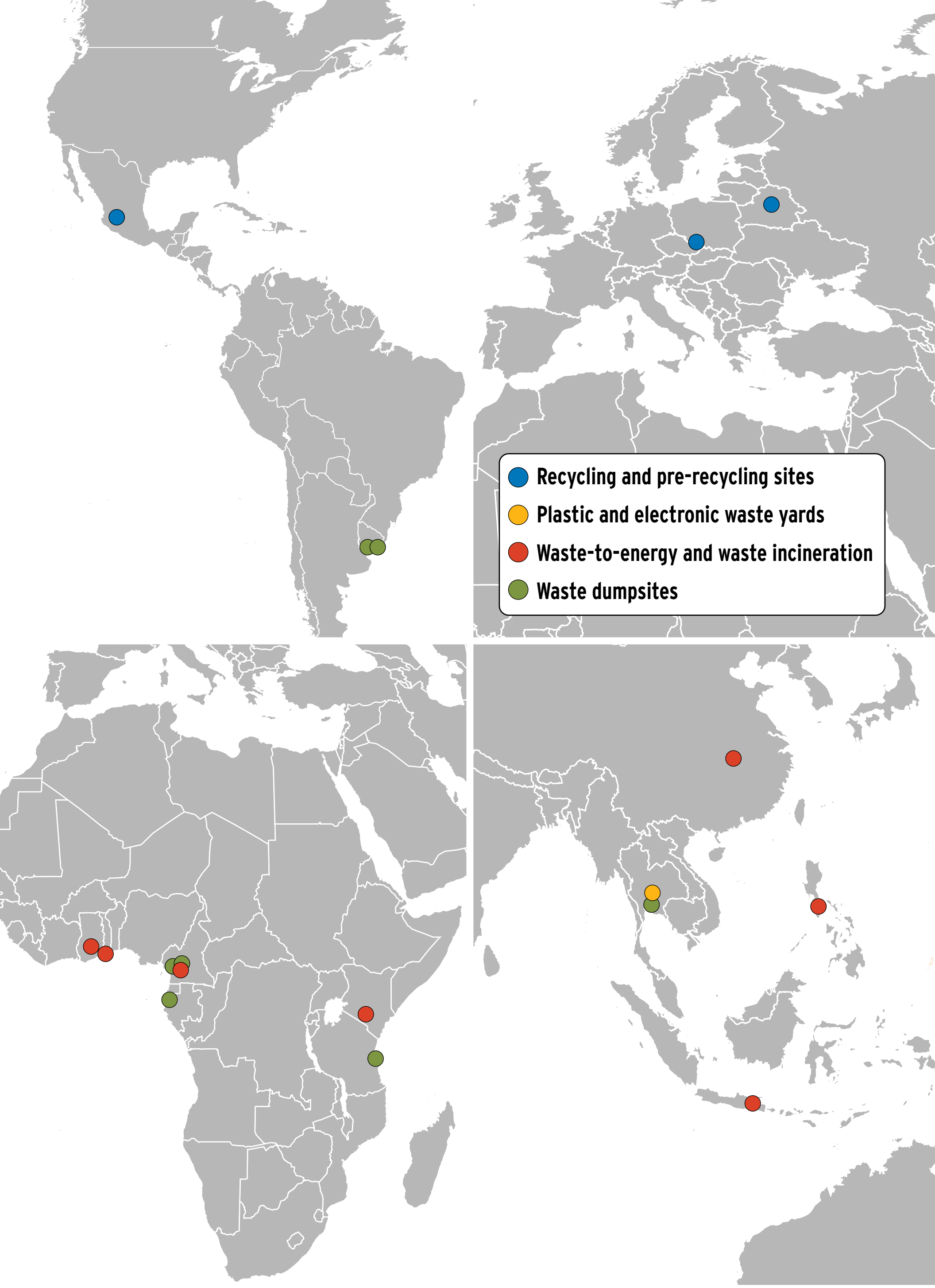
The twenty-seven free-range and all six reference egg samples (see Tables in Annex 1) were also analyzed for PBDEs and HBCD, and short-chain chlorinated paraffins (SCCPs). The thirteen free-range samples (see Tables in Annex 1 for their specification) and one reference sample (from Jakarta) were also analyzed for the range of 17 PFASs, including PFOA, PFOS and PFHxS. All of these analyses were conducted in a Czech certified laboratory (University of Chemistry and Technology in Prague, Department of Food Chemistry and Analysis).

Identification and quantification of PBDEs were performed using gas chromatography coupled with mass spectrometry in negative ion chemical ionization mode (GC-MS-NICI). Identification and quantification of HBCD isomers and selected PFASs 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 the 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).

Fifteen pooled samples of free-range chicken eggs and three samples from supermarkets (see Tables in Annex 1) were also analyzed 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 13C12-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).



Recycling and pre-recycling sites
Plastic and electronic waste yards
Waste-to-energy and waste incineration
Waste dumpsites

3. DESCRIPTION OF HOTSPOTS

The localities chosen for sampling in 2019 and 2020 and/or sites from previous research studies selected for inclusion in this study are 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 many of these sites as well. All of them represent places affected by current disposal or management of plastic waste as such.

Those sites are of four categories: 1) sites with plastic waste involved in some kind of recycling and/or pre-recycling processes, mostly related to e-waste or automotive industry 2) waste yards with open burning of waste, including plastic from electronic waste in some cases (rural dumpsites), 3) sites affected in some way by waste incineration, either with plastic waste used as fuel in tofu production/cooking or medical and municipal waste incineration, and 4) dumpsites with large volumes of plastic wastes. We aimed to choose locations in different parts of the world but with limited time and resources we were not able to cover all regions equally. The twenty-five hot spots presented in this study are distributed over different regions as follows: Latin America 3, Central Eastern Europe 2, South-East Asia 8, Central Asia 1, and Africa 11.

Eggs bought in supermarkets in Accra, Bangkok, Beijing, Jakarta, Karaganda, and Prague were chosen as reference samples for comparison with those from potentially contaminated sites.

Samples are further grouped according these categories as: RE – recycling + pre-recycling processes; WY-E – waste yards, large e-waste sites; WI – waste incineration, waste to energy; DU – dumpsites, and Ref – reference samples from supermarkets or convenient stores.

3.1 RECYCLING AND PRE-RECYCLING SITES

Three chosen localities for sampling and inclusion in this study can be characterized as either e-waste, plastic waste recycling or car shredder residue with significant amount of processed plastic waste.

3.1.1 Guadalajara, Mexico

In Guadalajara, we took egg samples in the close vicinity of a plastic e-waste shredder and recycling home workshop.

3.1.2 Pitarne, Czech Republic

There is a recycling plant for insulation of wires and similar types of PVC waste located in the village Pitarne close to the Polish border in the Czech Republic. The final product is sold as roof covers, and there has been some customer complaints about releases of VOCs, which were also found to be released when the roofing material is heated (Kosina 2016).[7] We have taken 4 pooled egg samples at different distances and directions from the factory.

3.1.3 Gatovo, Belarus

There is a large car shredder plant in a small town approximately 10 km to the south from Minsk. Plastic waste is quite often burned at the plant. A pooled egg sample was taken in one house nearby the car shredder plant. There are more factories in that area as well.

3.2 PLASTIC AND ELECTRONIC WASTE YARDS

Five of the sampled sites are either e-waste scrap yards or large plastic waste yards. E-waste sites handle high volumes of e-waste plastic, often burned at the site. There are also sites where mostly foreign plastic and other waste was imported and is handled as kind of plastic waste yard. Open burning is used to reduce volumes and clear the space for trucks bringing more plastic waste.

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[7] Major evaporating substances from recycled roofing materials produced as recycling product from PVC wire insulation: 2-ethylhexanol, acetaldehyde, acetone, methacrolein, benzene, styrene, phenol, aniline, and phthalates: DIBP and DnBP. Kosina, J. (2016). Protokol č. JK 31/16. Stanovení emisí těkavých organických sloučenin (VOC) uvolňujících se ze střešní krytiny CAPACCO SK-2 Velká šablona. (Chemical analysis protocol No JK 31/16; Determination of emissions of volatile organic compounds (VOC) released from roofing CAPACCO SK-2). VŠCHT - Centrální laboratoře, Laboratoř hmotnostní spektrometrie.

3.2.1 Bangun, Indonesia

One of the sites where a mixture of plastic and paper waste for additional sorting was imported from developed countries like Australia, Canada, Ireland, Italy, New Zealand, UK, and the USA (Ismawati Drwiega, Septiono *et al.* 2019) is in Bangun, next to a paper factory. We took two pooled egg samples from the area where residual plastic waste is dumped and burned (Petrlik, Ismawati *et al.* 2019, Petrlik, Ismawati *et al.* 2020).

3.2.2 Tangerang, Indonesia

Another destination of imported, mostly plastic waste is located in Tangerang. Waste is being sorted here, its residues are dumped and most of it burned. We took samples from two different locations. At one of these sampling sites, from where sample SEM-E-1 was taken, we have sighted refrigerator insulation plastic residues which might be treated with BFRs (Petrlik, Ismawati *et al.* 2020).

3.2.3 Bagong Silang, Philippines.

There is a site where e-waste is dismantled and partly dumped in Bagong Silang, just north of Manila. Eggs were sampled from chickens that can access the area where the e-waste is being dismantled.

3.2.4 Samut Sakhon, Thailand

Samut Sakhon is slightly different from the other sites in this group as it is a concentration of different “recycling” workshops, where e-waste is handled in some of them, but small smelters are often also present in the area as well. The egg sample with the ID “Samut Sakhon” was taken at a typical e-waste site where waste is burned to recover metal materials out of it. Another sample was taken in the middle of many different small facilities including metal smelters (Mach, Petrlík *et al.* 2017, Teebthaisong, Petrlik *et al.* 2018).

3.2.5 Agbogbloshie, Ghana

There is a very large e-waste and car wreck scrap yard located next to the Korle Bu lagoon in Accra, Ghana (Petrlik, Adu-Kumi *et al.* 2019), with a lot of burning of plastic from e-waste including smoldering of copper cables, listed as one of the major source categories of chlorinated dioxins and furans as UPOPs in the Annex C of the Stockholm Convention (Stockholm Convention 2010). The eggs were sampled from a man living right on the scrap yard. This sample was already part of a previous study focused on POPs in eggs from Ghana and Cameroon (Petrlik, Adu-Kumi *et al.* 2019).

3.3 WASTE-TO-ENERGY AND WASTE INCINERATION SITES

We also sampled free-range chicken eggs in seven different Asian and African countries at eight sites where plastic waste is or was either used as fuel or incinerated, often in combination with other waste.

3.3.1 Tropodo, Indonesia

There are 50 small-scale tofu makers that utilize unwanted plastic scraps as fuel to create steam to turn the soybean milk into tofu in Tropodo, which is located in the Sidoarjo Regency on Java Island. Five years ago, most tofu makers used wood to create hot steam. The combustion of mixed plastic scraps takes place all day long, from 6AM to 6PM, releasing thick black smoke. Free-range chicken egg samples were taken in two different households next to the tofu factories. The chickens also had access to ash residues from the burning of plastic waste (Petrlik, Ismawati *et al.* 2020).

3.3.2 Aguado, Philippines

The medical waste incinerator of IWMI (Integrated Waste Management, Inc.) is located in Barrangay Aguado, south-west of Manila. Next to the waste incinerator burning a lot of plastic medical waste lies WARM (Waste and Resources Management, Inc.), which produces bricks from waste incineration ash and other materials. Free-range chicken egg sampling was done here already in 2005 in close vicinity of the waste incinerator. A level of 13 pg TEQ g^{-1} fat was measured in eggs from that time (Calonzo, Petrlik *et al.* 2005). For this study, EcoWaste Coalition collected 4 pooled egg samples in some surrounding households of both facilities in Barrangay Aguado.

3.3.3 Wuhan, China

Wuhan is the capital of the Hubei Province, and with more than 10 million inhabitants it is the most populous city in Central China. There are two waste incinerators in Hanyang, Wuhan. One is a municipal waste incinerator (MWI) and the other is a medical waste incinerator. The former one burns 1,500 tons of waste per day with circulating fluidized bed technology. The latter one burns 50 tons of medical waste each day. The MWI started operating in December 2012. The medical waste incinerator started in 2013. The medical waste incinerator is next to the MWI,

Large municipal waste incinerator in Wuhan, China. Photo: Jindrich Petrlik, Arnika

to the north. The eggs were sampled at two sites, one (Wuhan 1) from the neighborhood of the waste incinerators in households that were later destroyed and the other one (Wuhan 2) at 2 km distance from the waste incinerators. The sampling was conducted in 2014 as part of a joint project between IPEN and its POs (Petrlik 2016).

3.3.4 Yaoundé – hospital, Cameroon

For the study focused on POPs in free-range chicken eggs from areas affected by waste disposal activities in Ghana and Cameroon, we chose a very small and low-cost incinerator operating a couple of days per week or so in Yaoundé, Cameroon. Categories of wastes incinerated include plastics and other materials containing syringes and needles, as well as biological residues. The ashes from the incinerators are buried in open pits close to the incinerator. The incinerator and the open pits are within the hospital premises with less than 100 metres to nearby homes (Petrlik, Adu-Kumi *et al.* 2019). Open burning also occurs in this area. Waste is also open burned at this place. It is necessary to note that the scenario of the sampling in this case was a bit different from the other samples as it is a pooled sample from five different households within a radius of 0.3 km in all directions from the hospital. The eggs were part of a previous study

(Petrlik, Adu-Kumi *et al.* 2019), along with some other African samples presented here as well.

3.3.5 Accra – hospital, Ghana.

The chosen hospital used a locally built small-scale DeMontfort type of medical waste incinerator. It had *"…an in-built drier that could dry wet waste very fast and a burning chamber for five tons of waste which could burn completely within three hours"* (Adama, Esena *et al.* 2016). Adjacent to the incinerator is the ash dumpsite where the bottom ash and some fly ash was disposed of after incineration. This waste incinerator started operating in 2004 and stopped working several years ago. However, the ash dumpsite next to the waste incinerator was left. There is a family living in a house next to the waste incinerator and raising chickens which have access to the whole area including the ash dumpsite. The area of the waste incinerator was included in two previous studies, one focused on heavy metals (Adama, Esena *et al.* 2016), the other one on POPs in eggs (Hogarh, Petrlik *et al.* 2019, Petrlik, Adu-Kumi *et al.* 2019).

3.3.6 Kumasi – hospital, Ghana

In Kumasi, the second largest city in Ghana, we have chosen one of the small medical waste incinerators and its neighborhood as the sampling site. The waste incinerator burns waste only from the hospital once per week. This waste incinerator does not store ash on the hospital grounds, but it is collected by a waste management company for disposal elsewhere. The waste incinerator does not have any air pollution abatement. It has a chimney approximately 10 m high. It has been in operation for eight years now. The eggs were sampled from a household close to the waste incinerator but outside of the fenced hospital area. They were also part of a previous study (Petrlik, Adu-Kumi *et al.* 2019).

3.3.7 Nkoltang – medical waste incinerator, Gabon

There is a small medical waste incinerator located in Nkoltang, a town east of City of Libreville, the capital of Gabon.

3.3.8 Nairobi – Mirema, Kenya

A school in Mirema, one of the north quarters of Nairobi, was one of the places where a so-called "community cooker" using waste as fuel was established. Community cookers are high heat stoves burning waste

Abandoned medical waste incinerator in hospital in Accra, Ghana.
Photo: Martin Holzknecht, Arnika

(Maarifa Centre 2018, Wikipedia 2020).[8] We could not find any information about filters being used as air pollution control systems in community cookers. Mirema School is an international school located in Mirema Estate, Zimmerman, and Nairobi County. The school is located approximately 400 m from the Zimmerman shopping center. Pooled egg samples were taken in a household approximately 300 m from the community cooker which is situated on the fenced-in school grounds. The community cooker in Mirema School was established in 2016.

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[8] *"The technology only uses combustible waste as fuel. The structure is made of firebrick to maintain as much heat as possible. The wastes are clumped together to form ball-like shapes which are fed in to the 1st chamber of the structure by the stove operator through a wide metal chute where they are heated to temperatures of about 1,000°C. The heat produced disseminates to the 2nd chamber that has cast iron metal plates (5-6) fitted at the top, which serve as the cooking surface. Temperatures in the 2nd chamber can rise up to 1,200°C."* Maarifa Centre. (2018, May-21-2018). "Community cooker." Retrieved 23-08-2020, 2020, from https://maarifa.cog.go.ke/51/community-cooker/.

3.4 WASTE DUMPSITES

Nine of the sampled localities are dumpsites and their surrounding neighborhoods.

3.4.1 Cerro de Montevideo, Uruguay

There are dumps alongside the road in Cerro de Montevideo with a lot of plastics, including some old electronics. The eggs were sampled from a family living on the opposite side of the road, but their chicken can freely walk to the dumpsite as well. Cerro de Montevideo is a quarter on the western edge of the City of Montevideo, the capital of Uruguay.

3.4.2 Minas, Uruguay

Minas is the capital of the Department Lavalleja in Uruguay. There is a dumpsite just across the road from the area where the chicken eggs were collected in Minas, Lavalleja, probably around 25 meters away. The landfill often catches fire, intentionally or naturally. Obsolete electronics have been spotted at the dumpsite. The place is also close to the local airport.

3.4.3 Yaoundé – TKC and Etetak Quarters, Cameroon

Based on the criteria of close proximity to homes where free-range chicken are raised, and the composite nature of the waste dumped (plastics, electronics, cables, tyres, organic matter, cardboard, etc…), two waste dumpsites subjected to regular open burning as a way to reduce the waste stockpile volume were selected in the city of Yaoundé, the political capital of Cameroon. The two sites are located in the Etetak and TKC quarters in Yaoundé. The eggs from these two sites were part of a previous study focused on POPs in eggs from sites in Ghana and Cameroon (Petrlik, Adu-Kumi *et al.* 2019).

3.4.4 Libreville – Owendo and Ozounge Quarters, Gabon

We selected two municipal waste dumpsites in the Gabonese capital Libreville for sampling. They are located in the quarters of Owendo and Ozounge. There are large quantities of plastic waste mixed with other kinds of wastes as well. Used electronics have been spotted at the Ozounge site as well. The dumpsite in Ozounge is also close to a fish-drying workshop while the dump in Owendo is close to a timber-processing plant. Eggs were taken from hens that can access the dumpsites freely.

Minas, Uruguay. Maria Carcamo, RAPAL

Baskuduk, Kazakhstan. Magda Slamova, Arnika

Cerro de Montevideo, Uruguay. Maria Carcamo

3.4.5 Pugu Kinyamwezi, Tanzania

There is a large municipal solid waste dumpsite called “Pugu Kinyamwezi City Solid Waste Dumpsite” on the south-western edge of Dar es Salaam, the capital city of Tanzania, in a municipality called Ilala. Free-range chicken eggs were collected from five households located just next to the dumpsite area which is fenced, however in some houses additional sorting of waste containing large amounts of plastic wastes occurs. Polluted air from the dumpsite where solid waste self-ignites causing open burning of waste as well as leachate from the dumpsite may affect the area where hens forage.

3.4.6 Praeksa, Thailand

Praeksa belongs to the Samut Prakan Province that lies at the mouth of the Chao Phraya River on the Gulf of Thailand. There is also a dumping site of 0.24 km^2 with a depth of 50 m that accumulates municipal solid waste and illegally even some industrial waste. It has been in operation for over 20 years. In 2014 there was a massive fire that lasted for almost a week, during which large amount of the dumped waste was uncontrollably burned (EARTH 2016). There are also many factories in the area that produce motor vehicles, car parts and other equipment, metal products, electronics, textiles, food products, chemicals, plastics, etc. Free-range chicken eggs were taken from a household located approximately 0.5 km north of the dumpsite in 2015 as part of a larger project by Arnika and EARTH. It was included in a previous study evaluating the results of that project (Petrlík, Dvorská *et al.* 2018).

3.4.7 Baskuduk, Kazakhstan

There is a quite large old dumpsite, partly fenced, on the north-western edge of Aktau, Baskuduk in the Mangystau region of Kazakhstan (Petrlík, Kalmykov *et al.* 2016). Relatively large amounts of domestic waste, including plastics, are dumped there. The dumpsite is almost constantly burning. The eggs were sampled from a household at approximately 200 meters distance from the landfill. The egg samples were part of previous studies focused on evaluation of POPs in eggs from Kazakhstan (Petrlík, Kalmykov *et al.* 2016, Petrlik, Kalmykov *et al.* 2017, Petrlik, Teebthaisong *et al.* 2018).

3.5 SUMMARIZED INFORMATION ABOUT THE SELECTED SITES

In Table 2 we have summarised the information about the sites included in this study.

TABLE 2: INFORMATION ABOUT THE SITES INCLUDED IN THIS STUDY, INCLUDING THEIR GROUPING INTO CATEGORIES OF SAMPLED SITES AS CHARACTERIZED IN THE INTRODUCTION TO CHAPTER 3.

Group	Locality (country)	Samples	Brief characteristics
Recycling and pre-recycling including shredder plants (RE)	Guadalajara (Mexico)	GUADAL-EGG1	Shredder of e-waste plastics and preparation of plastic recyclate
	Pitarne (Czech Republic)	N1-3	Recycling of PVC insulation of wires involving heat and pressure; production of roofing from recyclate
		S1-4	
		PIT03	
		PIT 2/2020	
	Gatovo (Belarus)	Gatovo	Car shredder plant
Plastic and electronic waste yards (WY-E)	Bangun (Indonesia)	Bangun 1	Mainly plastic waste yard, most waste imported from outside of Indonesia
		BAN-E-1	
	Tangerang (Indonesia)	SEM-E-1	Mainly plastic waste yard, most waste imported from outside of Indonesia
		TAN-ESIN-01	
	Bagong Silang (The Philippines)	PH-E-1 and 2	E-waste site (neighborhood dismantling area)
	Samut Sakhon (Thailand)	Samut Sakhon	Waste sorting workshop with regular open burning including e-waste, small metal smelters and waste sorting and recycling workshops in other parts
		SMS2-13	
	Agbogbloshie (Ghana)	AGB-E	One of the largest e-waste sites, regular open burning and smoldering of copper cables, car wreck dismantling

Group	Locality (country)	Samples	Brief characteristics
Waste-to-energy and waste incineration (WI)	Tropodo (Indonesia)	Tropodo 1	Local tofu factories using plastic waste as fuel
		TROP-E-1	
	Aguado (Philippines)	PH-E-3 and 4	Hazardous waste incinerator with related facility producing bricks from incineration ash.
		PH-E-5 and 6	
		PH-E-7	
		PH-E-S-5/2	
	Wuhan (China)	Wuhan 1	Large municipal solid waste incinerator and smaller medical waste incinerator
		Wuhan 2	
	Yaoundé - hospital (Cameroon)	YA-2	Small medical waste incinerator and open burning of plastic waste
	Accra - hospital (Ghana)	KBI-E	Abandoned medical waste incinerator with ash residues left within the hospital area
	Kumasi - hospital (Ghana)	KU-E	Small medical waste incinerator operating a limited amount of hours / week
	Nkoltang - medical waste incinerator (Gabon)	GA-E-NKOL	Small medical waste incinerator in a town east of the City of Libreville
	Nairobi - Mirema (Kenya)	KE_001	Community cooker with large stoves burning waste as fuel under high temperatures

Group	Locality (country)	Samples	Brief characteristics
Dumpsites (DU)	Cerro de Montevideo (Uruguay)	UR-CM-E	Dumpsite alongside the road with observed e-waste
	Minas (Uruguay)	UR-MIN-E	Dumpsite, often catching fire
	Yaoundé - TKC Quarter (Cameroon)	YA-1	One of the dumpsites in the City of Yaoundé, regular open burning
	Yaoundé - Etetak Quarter (Cameroon)	YA-3	One of the dumpsites in the City of Yaoundé, regular open burning
	Libreville - Owendo (Gabon)	GA-E-OWE	Large waste dumpsite in the City of Libreville, open burning occurs
	Libreville - Ozounge(Gabon)	GA-E-OZOU	Large waste dumpsite in the City of Libreville, open burning occurs; some electronic waste observed
	Pugu Kinyamwezi (Tanzania)	TZ-PU-KI_EGG	Large municipal solid waste dumpsite called "Pugu Kinyamwezi City Solid Waste Dumpsite" on the south-western edge of Dar es Salaam
	Praeksa (Thailand)	PKS-EGG-1	Partly abandoned dumpsite in Praeksa, in the Samut Prakan Province; large fire occured in 2014
	Baskuduk (Kazakhstan)	BAS 02	Large old dumpsite, partly fenced, on the north-western edge of Aktau, Baskuduk in the Mangystau region
Reference samples (Ref)	Prague (Czech Republic)	PHA-1(and2)	Sample PHA-1 bought in a supermarket in Prague; sample PHA-2, used only for SCCPs, also bought in a supermarket
	Jakarta (Indoesia)	JAK-SUP	Eggs bought in a supermarket in Jakarta
	Bangkok (Thailand)	Supermarket	Eggs bought in a supermarket in Bangkok
	Accra (Ghana)	ACC-M-E	Eggs bought in one of the major supermarkets in Accra
	Karaganda (Kazakhstan)	KAR-SU	Eggs bought in a convenient store in Karaganda

4. RESULTS AND DISCUSSION

Results from the chemical analyses of 35 pooled free-range chicken egg samples and 6[9] reference samples from supermarkets. The samples come from twenty-five hot spots in fourteen countries of Africa, Asia, Europe and Latin America. Basic information about the samples can be found in Tables 1 and 2 as well as in the text of chapters 2 and 3 of this report.

Details about the sampling and the sampled localities are provided in chapters 2 and 3. Their evaluation is discussed further in separate subchapters according to the natural groups of POPs. The sampled locations and hot spots were grouped according to major activity which is considered as a potentially major source of contamination with POPs, although in several cases there might be more different sources of contamination which are described under hot spots characteristics in chapter 3. Activities considered as potentially major sources of contamination and the abbreviations used for them are described in the introduction of chapter 3.

The results of the analyses for POPs are summarized according to the groups of potential contamination in Table 4. Detailed results for each sample are in the tables in Annex 1 to this study.

The measured levels of POPs in the chicken eggs were compared with legislative limits established in the European Union, although most of the measured chemicals in this study don't 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. Limit values for PCDD/Fs and dl-PCBs in eggs are summarized in Table 3. These limits are used for comparison with levels measured in food in many other studies, mainly in developing countries which do not have limits for dioxins and other POPs in food. We also included limit set in Indonesia for dioxins and dl-PCBs in eggs (Badan pengawas obat dan makanan Republik Indonesia 2018) as well as limit set in Russia and used in other neighboring countries for dioxins in eggs (Russian Federation 2008).

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[9] There were two reference samples bought in supermarkets in Prague which are merged here as one reference sample, as the second one (PHA-2) was analyzed for SCCPs only while the first one (PHA-1) was analyzed for most other POPs presented in this study excluding PBDD/Fs, DR CALUX dioxin activity, and PFASs. The result of the analysis of PHA-2 was part of a previously published study on SCCPs in eggs, Adu-Kumi, S., J. Petrlík, E. Akortia, M. Skalský, J. Pulkrabová, J. Tomáško, L. Bell, J. N. Hogarh, D. Kalmykov and A. Arkenbout (2019). "Short-chain chlorinated paraffins (SCCPs) in eggs from six countries." *Organohalogen Compounds* **81**(2019): 337-339.

TABLE 3: LIMIT CONCENTRATION VALUES FOR PCDD/Fs AND DL-PCBs IN TEQS IN CHICKEN EGGS.

			Hen eggs
	Indonesia	**Russian Federation**[2]	**EU ML**[3]
Unit	pg g^{-1} fat	pg g^{-1} fat	pg g^{-1} fat
WHO-PCDD/Fs TEQ	-	3.0	2.5
WHO-PCDD/Fs-dl-PCB TEQ	2.5[1]	-	5.0

Notes to the Table:

[1] *Limit is set in TEF that includes both PCDD/Fs and dl-PCBs (Badan pengawas obat dan makanan Republik Indonesia 2018)*

[2] *Current Russian* СанПиН *2.3.2. 2401-08 (Russian Federation 2008). Maximum Allowed Concentration (MAC) is in practice used also in other countries like Belarus or Kazakhstan.*

[3] *EU Regulation (EC) N°1881/2006. 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. (European Commission 2016)*

TABLE 4: OVERVIEW OF RESULTS OF CHEMICAL ANALYSES FOR POPs IN THIRTY-SIX FREE-RANGE CHICKEN EGG SAMPLES FROM PLASTIC WASTE HOT SPOTS, AND FIVE EGG SAMPLES FROM COMMERCIAL FARMS.

Locality	Recycling and pre-recycling sites (RE)	Plastic & electronic waste yards (WY-E)	Waste to energy and waste incineration (WI)	Dump-sites (DU)	Reference samples (Ref)	EU standard / limits
PCDD/Fs (pg TEQ g^{-1} fat)	1.6 - 15.4	6 - 661	1.7 - 200	2.16 - 26	0.0012 - 0.9	2.5
DL PCBs (pg TEQ g^{-1} fat)	2.3 - 16	3 - 195	0.9 - 32	3.41 - 18	0.001 - 0.34	-
Total PCDD/F + DL PCBs (pg TEQ g^{-1} fat)	5.8 - 32	12 - 856	2.6 - 232	9.6 - 35	0.0032 - 0.9	5.00
Total PCDD/Fs + DL PCBs - DR CALUX (pg BEQ g^{-1} fat)	8.1 - 37	13 - 840	5.2 - 560	12	<0.6 - 1.2	-
PBDD/Fs (pg TEQ g^{-1} fat)	<1.4 - 5.4	7 - 300	0.33 - 27	0.17 - 3	<1.8 - <21.3	-
SCCPs	235	97 - 2067	65 - 162	50 - 1950	25 - 136	-
sum HBCD	<LOQ - 4602	< LOQ - 1961	< LOQ - 379	5.2 - 314	< LOQ - 1036	-
sum of PBDEs	<LOQ - 230	3.1 - 1457	< LOQ - 27159	< LOQ - 164	< LOQ - 9.5	-
sum of N-BFRs	<LOQ - 6.2	< LOQ - 124	< LOQ - 2166	< LOQ - 6.2	< LOQ - 3.7	-
sum of PFASs (ng g^{-1} of fresh weight)	1.5	1.15 - 97	0.3 - 2.4	4.7 - 9.8	0.1 - 0.34	-
L-PFOS (ng g^{-1} of fresh weight)	0.86	0.36 - 76	0.11 - 1.2	2.3 - 7.3	<0.01	-

Samples were bought in supermarkets in different parts of the world in 2014 – 2020. For more details see Table 1. Levels of POPs are in ng g^{-1} fat if not specified otherwise. Samples are grouped according to classification of hot spots as specified in chapter 3. Detailed results for each sample can be found in Annex 1.

4.1 POLYCHLORINATED DIBENZO-P-DIOXINS AND DIBENZOFURANS (PCDD/Fs) AND DIOXIN-LIKE POLYCHLORINATED BIPHENYLS (DL-PCBs)

Eight out of thirty-five samples presented in this study and measured for PCDD/Fs had levels of dioxins above 20 pg TEQ g^{-1} fat (see Tables in Annex 1), which is ten times more than the EU limit for dioxins in eggs for food.

Four samples in this study had levels of dioxins below the EU limit for eggs as food (2.5 pg TEQ g^{-1} fat). These were two samples from the vicinity of a PVC recycling plant in the Czech Republic, one sample from the neighborhood of a small medical waste incinerator in Ghana, and one sample from the vicinity of a waste dumpsite in Kazakhstan. Only one of the samples from the vicinity of a small medical waste incinerator in Kumasi, Ghana had levels of PCDD/Fs plus dl-PCBs that were lower than the limit set by the EU, which means the other three had high levels of dl-PCBs and exceeded the acceptable standard for dioxin-like compounds as it is set in the EU.

The EU standard for dioxins (2.5 pg TEQ g^{-1} fat) was exceeded in 31 (out of 35) samples 1.5 – 264 times.

We compared the results of the analyses for dioxins in eggs from this study to maximum levels measured in free-range chicken eggs in other studies. This comparison can be found in the graph in Figure 2 and Table 5.

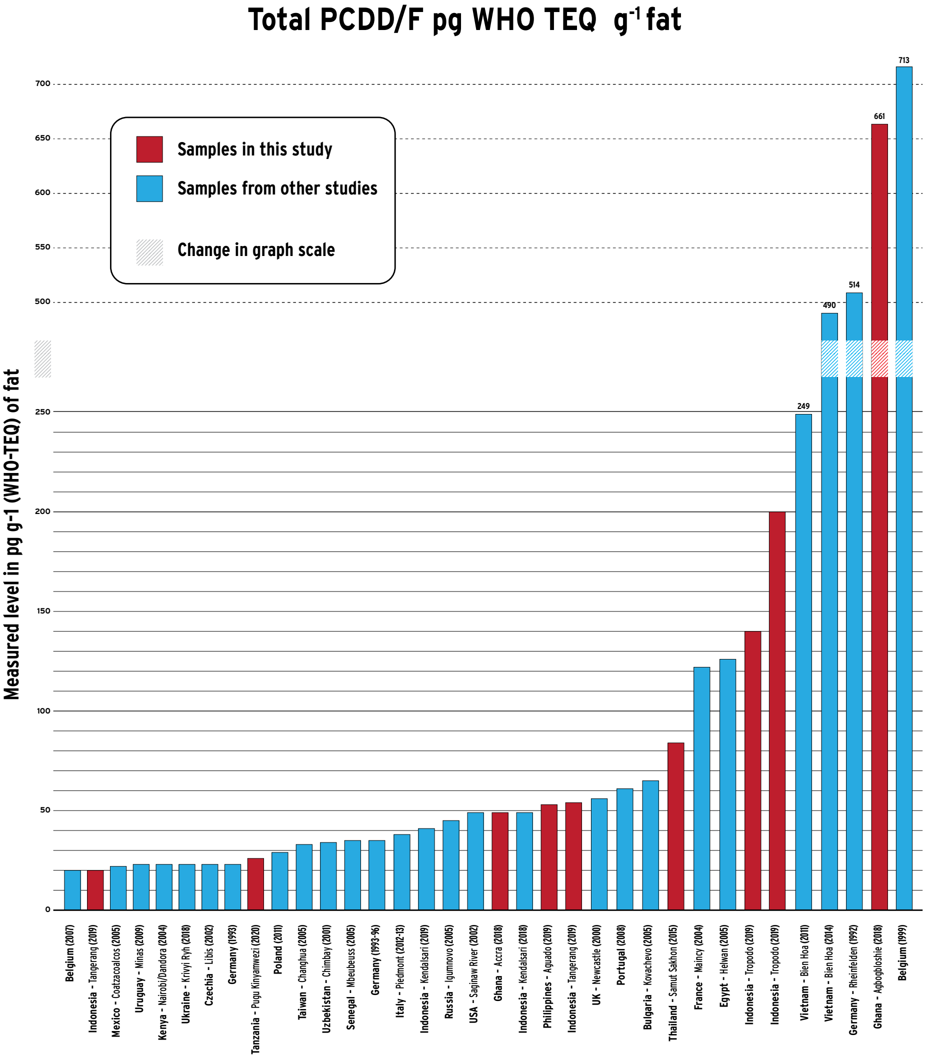

Figure 2: Graph showing maximum levels of PCDD/Fs measured in chicken eggs in different countries. Samples before 2006 are in WHO-TEQ 1998. Sources of information are listed in Table 5.

TABLE 5: 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 (Fürst, Fürst *et al.* 1993)
Tanzania	2020	Pugu Kinyamwezi	26	This study	Open burning of waste
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 aluminium smelter

Country	Year	Locality	PCDD/Fs pg WHO-TEQ g^{-1} fat	Source	Comments
Indonesia	2019	Kendalsari	41	Petrlik, Ismawati *et al.* 2020	Secondary aluminium smelters / contaminated ash
Russia	2005	Igumnovo	45	DiGangi and Petrlik 2005	Chlorine chemical industry area; Hazardous Waste Incinerator (HWI)
USA	2002	Saginaw River	49	MDEQ 2003	Floodplain downstream from chlorine chemical industry
Ghana	2018	Accra - hospital WI	49	This study	Medical waste incinerator ash
Indonesia	2018	Kendalsari	49	SVÚ Praha 2018	Secondary aluminium smelter
Philippines	2019	Aguado	53	This study	Medical waste incineration; incineration ash
Indonesia	2019	Tangerang	54	This study	Open burning of plastic waste and e-waste plastics
UK	2000	Newcastle	56	Pless-Mulloli, Schilling *et al.* 2001a	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	This study	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

Country	Year	Locality	PCDD/Fs pg WHO-TEQ g^{-1} fat	Source	Comments
Vietnam	2011	Bien Hoa	249	Traag, Hoang *et al.* 2012	Former US military base, dioxin-contaminated site
Vietnam	2014	Bien Hoa	490	Kudryavtseva, Shelepchikov *et al.* 2020	Former US military base, dioxin-contaminated site
Germany	1992	Rheinfelden	514	Malisch, Schmid *et al.* 1996	Waste from chlor-alkali chemical plant
Ghana	2018	Agbogbloshie	661	This study	E-waste and automobile scrapyard
Belgium	1999	Not specified	713	van Larebeke, Hens *et al.* 2001	Dioxin contamination of feed

Four samples from this study are among the ten highest ever measured levels of dioxins in chicken eggs globally, and they are second, sixth, seventh and tenth highest. The second highest ever measured level of dioxins in eggs globally was measured in the sample from the e-waste scrap yard in Agbogbloshie.

The graphs in Figures 3 and 4 demonstrate the balance between (the proportion of) PCDD/Fs and dl-PCBs in total TEQs, and the balance between PCDD and PCDF congeners measured in the free-range chicken eggs in this report. More detailed data are available in the tables in Annex 2. These basic characteristics help to identify potential sources of contamination of chicken eggs.

When there is electronic waste involved, eggs are contaminated with higher levels of dioxins and dioxin-like PCBs in total, but dioxins contribute to the overall toxicity more than dl-PCBs. From waste incineration sites, higher toxicity of samples were found at sites where the chickens have access to places where ash residues containing significant levels of dioxins are stored. PCDD/Fs contribute more to TEQ levels than dl-PCBs in the free-range chicken eggs taken from the vicinity of most of the waste incineration sites. Only the sample from the neighborhood of the hospital waste incinerator in Yaoundé is an exception where dl-PCBs prevail. It can be explained by the fact that open burning of medical and other waste occurs next to the small medical waste incinerator as well.

The tables in Annex 2 and the graph in Figure 3 show that sites where some kind of shredding or recycling occurs had lower total levels of

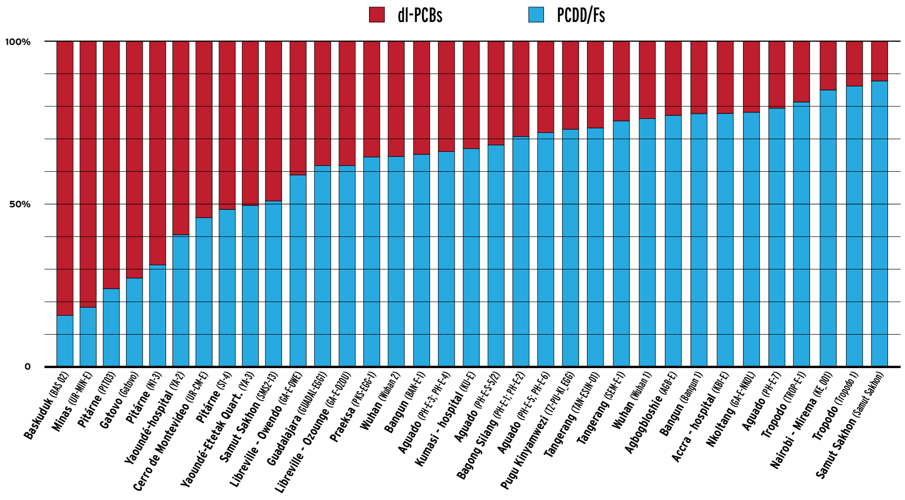

Figure 3: This graph shows the balance between PCDD/Fs and dl-PCBs in total TEQ levels measured in the free-range chicken eggs in this study.

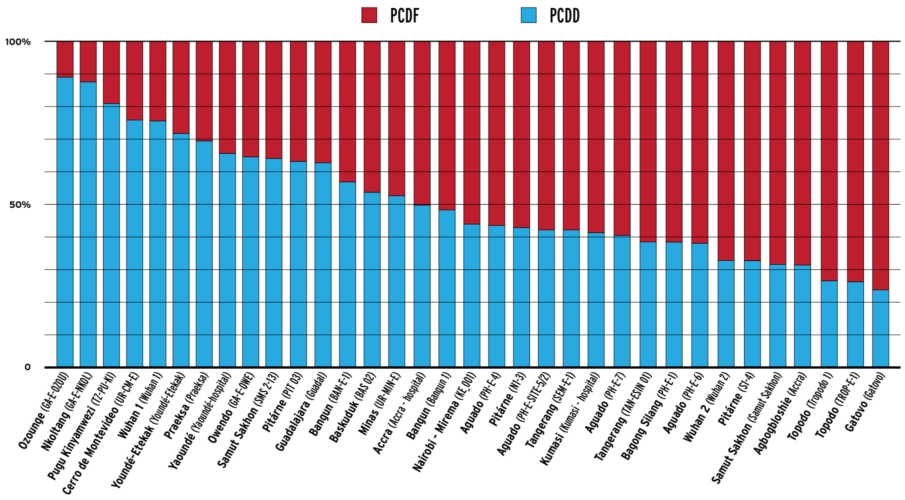

Figure 4: This graph shows the balance between PCDD and PCDF congeners in absolute levels in the samples of free-range chicken eggs in this study.

PCDD/Fs + dl-PCBs in comparison with other groups, while e-waste sites and waste incineration sites with accessible ash storages had the highest. Other groups like dumpsites, waste incineration sites and plastic waste yards had medium levels of total PCDD/Fs + dl-PCBs out of the studied groups of samples. Still, only one sample from Kumasi in Ghana had a total PCDD/Fs + dl-PCBs level below the EU limit for eggs (see tables in Annex 1).

Samples from dumpsites had higher levels of dl-PCBs among the studied group in general, although the highest measured levels were in the samples from Agbogbloshie and Tropodo, followed by the samples from a dumpsite in Minas – Lavalleja, Uruguay and a plastic waste yard in Tangerang, Indonesia. One sample from the vicinity of a PVC recycling plant in Pitarne, Czech Republic also had high levels of dl-PCBs.

Samples from dumpsites and from the vicinity of shredders and PVC recycling sites had higher levels of dl-PCBs in total TEQ in comparison with the other samples, although they were not always prevailing in total TEQ. They only prevailed in three of the samples from dumpsites (DU), and three of the samples from recycling and pre-recycling sites (RE) (see Figure 3 and Table A2-1 in Annex 2).

The balance between PCDD and PCDF congeners can be seen in the graph in Figure 4. PCDDs prevail in all samples from dumpsites in most of the cases in comparison with PCDFs when expressed in absolute levels (= not recalculated in TEQs). In samples taken from the surroundings of waste incineration activities the balance is the opposite.

"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). In our study, this rule seems to apply to waste incineration sites, waste yards and/or recycling and shredder operations where chlorinated or brominated compounds in plastics can be expected. In all these cases, PCDF congeners are prevailing in the free-range chicken egg samples presented in this study (see graph in Figure 4).

A similar dioxin pattern behavior was observed in our recent study from Indonesia too, partly but not only because some of the samples from that study have been included into this study as well. Prevailing PCDF congeners were also observed in samples from *"the group potentially influenced by kind of combustion sources in "closed" systems and/or ash residues from such processes,"* followed by *"the group of samples potentially contaminated by open burning of waste containing e-waste plastic"* in the study from Indonesia (Petrlik, Ismawati *et al.* 2020).

4.1.1 Comparison with older study from African dumpsites

Free-range chicken eggs were also sampled in the vicinity of two African dumpsites in 2004 and 2005, and the results of the analyses for PCDD/Fs, dl-PCBs and HCB were published in separate reports in March and April 2005 (DiGangi and Petrlik 2005, Petrlik, Diouf *et al.* 2005, Petrlik, Kamande *et al.* 2005).

The levels of dioxins and dl-PCBs measured in eggs from two African sites in Dandora, Kenya and Mbeubeuss, Senegal were either comparable (Dandora) or higher (Mbeubeuss) to what we have measured in eggs from dumpsites in this study, although in the case of Mbeubeuss there is the question of whether it should not rather be compared to a site like Agbogbloshie, as there were clear signs that the dumpsite also served as a hazardous waste dump.

When we compared the site from Nairobi, Kenya included in this study, where a community cooker burning waste is a potential source, with the sample from the Dandora landfill taken in 2004 we could see many differences - in the total level of PCDD/Fs and dl-PCBs (27 pg TEQ g-1 fat in eggs from Dandora and 14 pg TEQ g-1 fat in eggs from Nairobi – Mirema), in the balance between PCDD/Fs and dl-PCBs, as well as in the balance between PCDD and PCDFs (see graphs in Figures 3 and 4). In addition to this, the dioxin pattern was also different (see graph in Figure 5).

The sample from Dandora taken in 2004 has much more in common with the egg samples from the Pugu Kinyamwezi dumpsite in Tanzania included in this study, although the total level of PCDD/Fs and dl-PCBs is higher in the eggs from Pugu Kinyamwezi at 36 pg TEQ g-1 fat, which is the highest level among all egg samples from dumpsite areas in this study. Both the sample from Dandora taken in 2004 and the recent sample from Pugu Kinyamwezi have the same balance between PCDD and PCDF congeners (81:19), and the same PCDD/Fs and dl-PCBs proportion of TEQ (73:27). Their dioxin congener patterns are also very close to each other (see graph in Figure 6). Both samples were taken at dumpsites with large quantities of plastic waste where open burning occurs quite often. The influence on the dioxin levels from such practices in Dandora were also proven by passive air sampling. At the Dandora site “mean concentrations of 1041 pg sample-1“ were measured during a pilot passive air sampling project in Africa in 2005 – 2006 (Přibylová and Klánová 2013).

However, the level of dioxins in the eggs from Nairobi – Mirema, exceeding the EU limit value by more than fourfold, show that the burning of waste in community cookers is not a safe way of dealing with plastic waste.

4.2 POLYBROMINATED DIBENZO-P-DIOXINS AND DIBENZOFURANS (PBDD/Fs)

PBDD/Fs are not measured very often in the environment yet. It is obvious from studies made in China, Japan, Taiwan or Vietnam that PBDD/Fs are widely present in Asia (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).

We have found only one other study assessing PBDD/Fs in chicken eggs. 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 levels measured in free-range chicken egg samples from Wuhan or Samut Sakhon, and three orders of magnitude lower than in the samples from Agbogbloshie presented in this study and already published in previous studies (Weber, Watson *et al.* 2015, Teebthaisong, Petrlik *et al.* 2018, Hogarh, Petrlik *et al.* 2019). However, the levels of PBDD/Fs in egg samples from Tropodo and Yaoundé – Etetak Q. are similar to those measured in Ireland (See Tables A1-3 and A1-4 in Annex 1).

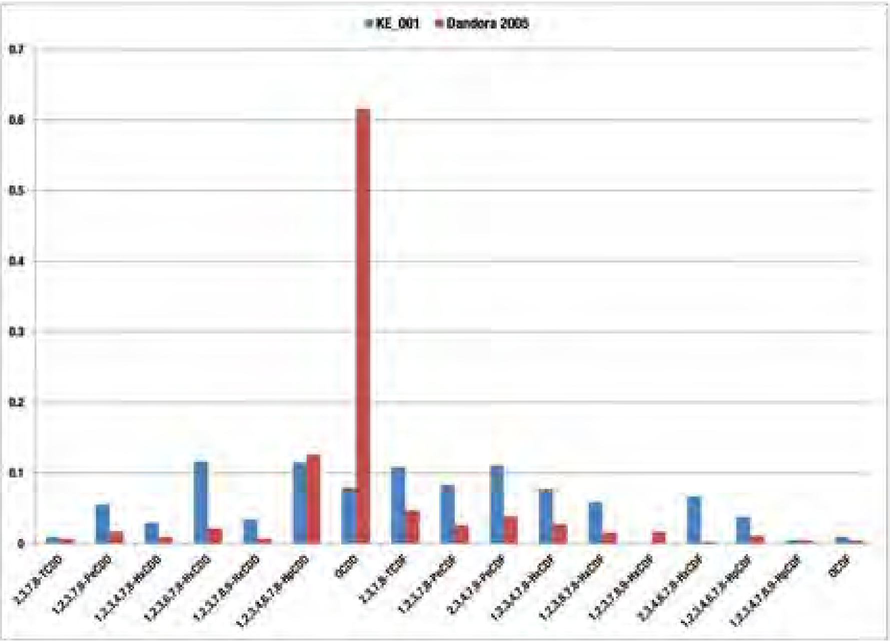

Figure 5: PCDD/F congener patterns in samples from Nairobi Mirema from 2020 and from Dandora dumpsite from 2004.

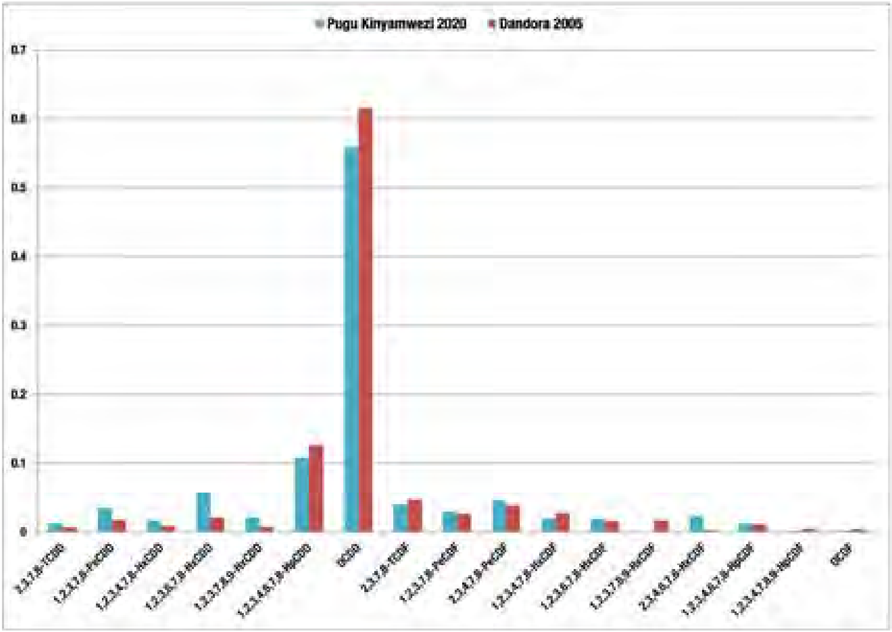

Figure 6: PCDD/F congener patterns in samples from Pugu Kinyamwezi from 2020 and Dandora dumpsite from 2004.

In samples from two dumpsites in Africa, Pugu Kinyamwezi in Tanzania and Libreville – Ozounge in Gabon, PBDD/Fs contributed to the overall dioxin toxicity expressed in TEQ levels by one tenth, proportionally similar to the sample from Tangerang in which, however, the dioxin level was generally higher (by two- to threefold) than in those two African samples (see Tables A1-4 and A1-2 in Annex 1).

The highest level of brominated dioxins was measured in eggs from Agbogbloshie followed by eggs from the vicinity of waste incinerators in Wuhan (27 pg TEQ g^{-1} fat). The following four high PBDD/Fs levels in eggs from Samut Sakhon (16 pg g^{-1} fat), Bagong Silang (11 pg g^{-1} fat), Tangerang (7 pg g^{-1} fat) and Guadalajara (5 pg g^{-1} fat) can be explained by e-waste plastics being handled and/or even burned at the sites. PBDD/Fs are already present in e-waste plastics as by-products in BFRs (Ren, Peng *et al.* 2011, Budin, Petrlik *et al.* 2020), and they are also released as a result of burning plastics treated with BFRs.

The eggs from Wuhan had higher levels of TEQs originating from brominated dioxins compared to from chlorinated dioxins and dl-PCBs.

The graph in Figure 7 shows levels of PBDD/Fs measured in the samples in this study exceeding a level of 2.5 pg TEQ g^{-1} fat of PBDD/Fs, which is equal to the limit value set for PCDD/Fs in eggs in the EU. There are seven out of fifteen samples of free-range chicken eggs that were analyzed for PBDD/Fs in this study. Two samples from supermarkets were also analyzed for PBDD/Fs and both of them had levels below the laboratory limit of quantitation (LOQ), which five other free-range egg samples also had (see Tables A1-1, A1-2, A1-3, A1-4 and A1-5 in Annex 1 for more details).

4.3 DIOXIN-LIKE ACTIVITY IN EGGS MEASURED BY USING BIOASSAY ANALYSES

Several bioanalytical tools are accepted by international standards[10] 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 it to evaluate contaminations 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). Four pooled egg samples in this study were analyzed using the DR CALUX® method. The highest levels of BEQs were measured in the samples from Agbogbloshie (840 pg BEQ g^{-1} fat) and Tropodo (560 pg BEQ g^{-1} fat) followed by samples from the sites affected by open burning and dumping of plastic waste in Tangerang (88 pg BEQ g^{-1} fat) or partly by e-waste in Samut Sakhon (100 pg BEQ g^{-1} fat).

As for PBDD/Fs, not all samples in this study were analyzed using the DR CALUX® method, but only 17 free-range egg samples and three reference samples.

There is a big difference between the total TEQ level (232 pg TEQ g^{-1} fat) and the BEQ level in the sample from Tropodo. The difference between the results from the DR CALUX® analysis and the chemical HRGC/HRMS analysis could potentially be explained by more chemicals showing dioxin-like activity than the ones included in any of the instrumental analyses in our study.[11] Part of that difference could also possibly be explained

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[10] 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.

[11] 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.

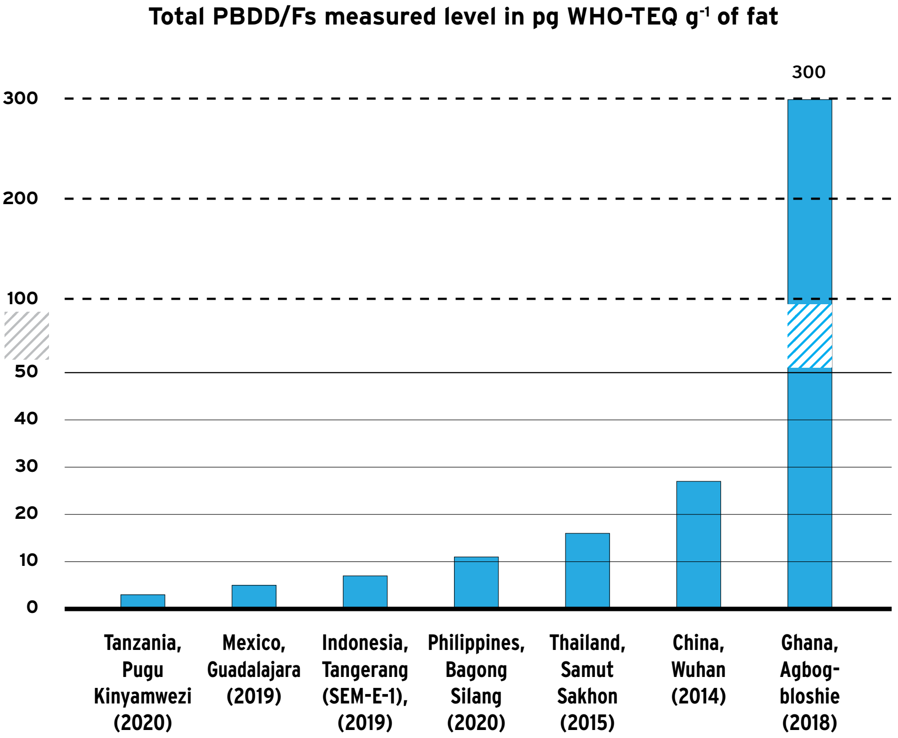

Figure 7: The graph shows levels of PBDD/Fs measured in eggs in this study exceeding 2.5 pg WHO TEQ g-1 fat.

by a variation in homogenicity within the samples, even though the same homogenate was used for all analyses. The chemical analyses of PCDD/Fs and dl-PCBs were done with a generally used certainty of ffl40%.

Bioassay analyses of eggs and other environmental samples could be a pathway to broader monitoring of dioxin contamination in food.

4.4 BROMINATED FLAME RETARDANTS (BFRs)

4.4.1 HBCD

Six egg samples in this study are among the ten highest levels of HBCD ever measured in poultry eggs globally, however not all six come from sites affected by plastic waste disposal or recycling. One sample is a reference egg sample from a convenience store in Karaganda in which a level of 1,036 ng g^{-1} fat of HBCD was measured, and that is the sixth highest level ever measured globally. The source of contamination in the eggs from the locality of Pitarne in the Czech Republic is most likely not the PVC recycling plant, but rather the polystyrene insulation foam used in the house where the chickens forage. The chickens probably consider the polystyrene marbles as feed and eat them. Similarly, polystyrene foam treated with HBCD could be the source of contamination for the eggs from the supermarket in Karaganda as well. So what we see is the influence of toxic additives used in plastics, both during their use and while being accessible to domestic animals raised for food.

The level of HBCD in the eggs from Pitarne (4,062 ng g^{-1} fat) is the third highest ever measured in free-range chicken eggs, right after samples from Shetpe, Kazakhstan (Petrlik, Kalmykov *et al.* 2017) and the Chinese e-waste site Guiyu (Zeng, Luo *et al.* 2016), see graph in Figure 8. The highest level of HBCD in the eggs from Shetpe of 18,321 ng g^{-1} fat can also be related to plastic waste from car wrecks, but the researched site in Kazakhstan was no typical dumpsite or waste yard, instead several car wrecks had just been left in the backyard where chickens foraged (Petrlík, Kalmykov *et al.* 2016).

The level of HBCD in the eggs from Agbogbloshie included in this study was also high, reaching almost 2,000 ng g^{-1} fat, and it was the fifth highest level ever measured globally. Free-range eggs from another e-waste site, Guiyu in China, had the second highest level of HBCD globally (7,600 ng g^{-1} fat).

Hexabromocyclododecane and SCCPs seem to contaminate the environment not only at sites affected directly by plastic waste disposal. We can also observe increased levels of SCCPs and very high levels of HBCD in eggs from large farms sold in supermarkets. HBCD, which is present in polystyrene insulations, contaminates the food chain of free-range chicken foraging for example next to houses where the polystyrene is being used as energy-saving insulation on the exterior of the buildings. This way it can cause contamination of chickens raised in large farms as well.

TABLE 6: LEVELS OF HBCD IN NG G^{-1} FAT MEASURED IN CHICKEN OR GOOSE EGGS IN DIFFERENT STUDIES WORLDWIDE ABOVE 50 NG G^{-1} FAT.

Country	Year	Locality	HBCD in ng g^{-1} fat	Source of information
Tanzania	2012	Arusha	63	(Polder, Müller *et al.* 2016)
Uruguay	2019	Cerro de Montevideo	86	This study
Uruguay	2004	Minas	89	(Blake 2005)
Slovakia	2004	Kokshov - Baksha	89	(Blake 2005)
Mexico	2004	Coatzacoalcos	91	(Blake 2005)
China	2013	Guiyu	110	(Zeng, Luo *et al.* 2016)
Cameroon	2018	Yaoundé - TKC Quarter	124	This study
South Africa	2008-2009	Vanderbijlpark	136	(Quinn 2010)
Thailand	2016	Samut Sakhon	159	This study
Kenya	2004	Dandora	160	(Blake 2005)
Thailand	2016	Koh Samui	165	(Petrlik, Teebthaisong *et al.* 2017)
Thailand	2016	Map Ta Phut	184	(Petrlik, Teebthaisong *et al.* 2017)
Kazakhstan	2016	Baskuduk	188	This study
Kazakhstan	2014	Balkhash - Rembaza	225	(Petrlik, Kalmykov *et al.* 2017)
Kenya	2020	Nairobi - Mirema	287	This study
Gabon	2019	Libreville - Owendo	314	This study
China	2010	South China	350	(Zheng, Wu *et al.* 2012)
Cameroon	2018	Yaoundé - hospital WI	379	This study
Kazakhstan	2016	Tauchik	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	1036	This study
Ghana	2018	Agbogbloshie	1961	This study
Germany	2007	Bavaria	2000	(Hiebl and Vetter 2007)
Czechia	2017	Pitárne	4602	This study
China	2013	Guiyu	7600	(Zeng, Luo *et al.* 2016)
Kazakhstan	2016	Shetpe	18321	(Petrlik, Kalmykov *et al.* 2017)

All other than free-range chicken egg samples are marked in parentheses after the name of the locality.

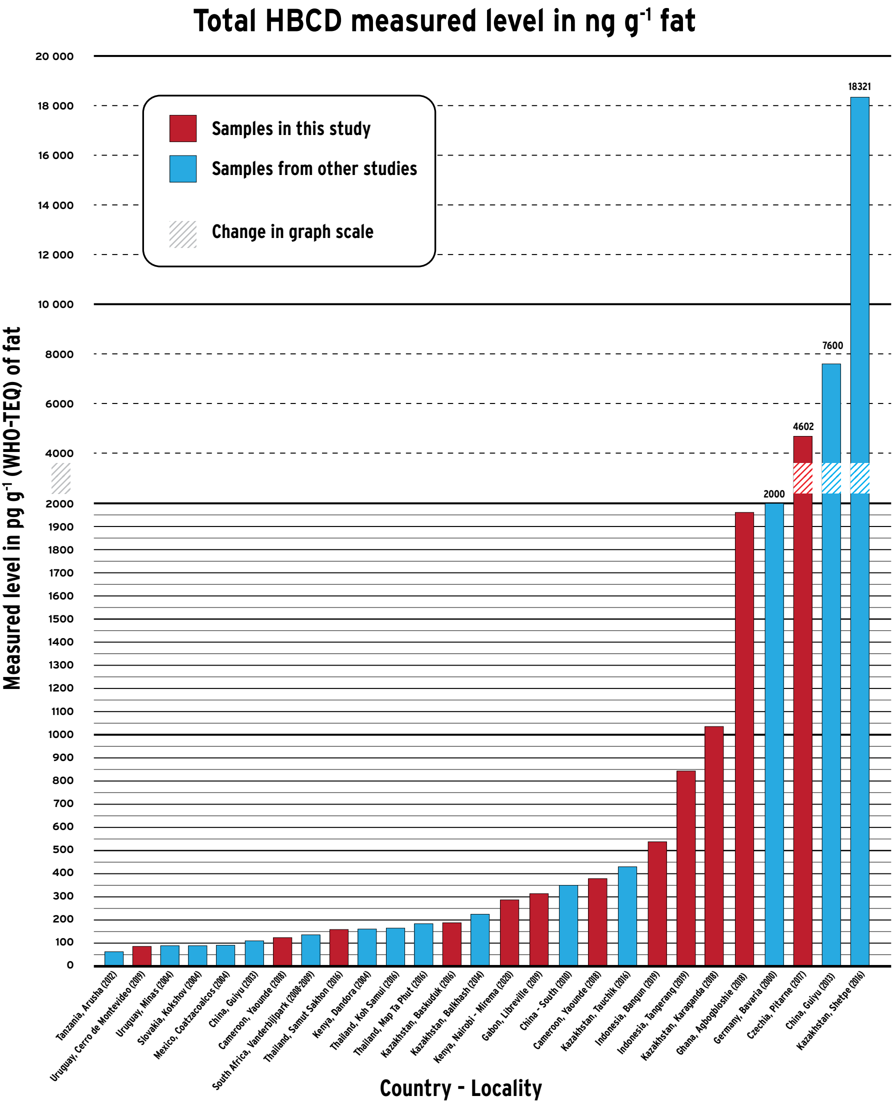

Figure 8: Highest levels of HBCD measured in eggs globally. Sources of information are listed in Table 6.

4.4.2 PBDEs

We summarized the information currently available in literature about levels of PBDEs measured in chicken eggs and compared it with the levels measured in the pooled egg samples included in this study. All samples with levels above 30 ng g^{-1} fat of PBDEs (including decaBDE) are summarized in Table 7 and the graph in Figure 9.

Four of the samples from this study rank among the ten highest levels of PBDEs ever measured in free-range eggs globally. The extremely high level 27,159 ng g^{-1} fat of PBDEs, and decaBDE in particular, was measured in the pooled egg sample from Tropodo taken in an area where plastic waste was being used as fuel in local tofu factories. The PBDE levels in three other samples in this study exceeded 1,000 ng g^{-1} fat: Two came from the waste yards in Bangun and Agbogbloshie with levels of 1,457 and 1,258 ng g^{-1} fat respectively, and one came from the vicinity of waste incinerators in Wuhan with a level of 1,054 ng g^{-1} fat. PBDE levels measured in samples from other sites in this study were mostly within the range below LOQ to 100 ng g^{-1} fat, only the samples from sites where plastics that have potentially been treated with PBDEs (e-waste or furniture) were handled had levels between 150 and 500 ng g^{-1} fat: Guadalajara (e-waste plastics recycling), Bagong Silang (e-waste site), Tangerang (furniture and e-waste plastics at a waste yard), Samut Sakhon (e-waste recycling and scrap yards), and Minas (where e-waste was spotted at the dumpsite).

The results from Tropodo highlight the big potential that using plastic as fuel can be a major source of contaminating the environment with PBDEs, which is in agreement with the findings of a previous study made in China which estimated that: *"... ΣPBDE release to the air and land from municipal solid waste (MSW) incineration plants in China in 2015 were 105 kg/year and 7124 kg/year"* (Ni, Lu *et al.* 2016). The same study came to the conclusion that PBDEs are mainly emitted to the air with airborne particles when incinerated, but a substantial part still stays in the ash.

TABLE 7: LEVELS OF PBDEs IN NG G^{-1} FAT MEASURED IN FREE-RANGE CHICKEN EGGS IN DIFFERENT STUDIES WORLDWIDE, ABOVE A SUM TOTAL OF 30 NG G^{-1} FAT OF PBDEs.

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)
Gabon (2019)	Libreville - Ozounge	36	this study
Tanzania (2020)	Pugu Kinyamwezi	50	this study

Country (year)	**Locality**	**PBDEs in ng g^{-1} fat**	**Source of information**
Uruguay (2019)	Cerro de Montevideo	50	this study
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
Uruguay (2019)	Minas	164	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)

Only samples that were also analyzed for decaBDE (congener BDE 209) have been included. There are two examples of wild birds from Antarctica and some duck or goose eggs included as well for comparison. All samples other than chicken eggs, are marked by specification of the bird species in parenthesis after the name of the locality.

The sample with extremely high level of PBDEs taken in October 2019 in Tropodo also contained a very high level of novel BFRs (nBFRs)[12] at 2,166 ng g^{-1} fat (Petrlik, Ismawati *et al.* 2020).

4.5 SHORT-CHAIN CHLORINATED PARAFFINS (SCCPs)

The highest levels of SCCPs found in this study were measured in eggs from Agbogbloshie, Baskuduk, Libreville – Obendo and Pugu Kinyamwezi in decreasing level order (see Tables A1-2 and A1-4 in Annex 1). The corresponding levels in the reference eggs were in the range of 25 – 136 (see Tables 4 and A1-5). Only eggs from the above-mentioned four locations exceeded a level of 500 ng g-1 fat. SCCPs in samples from other locations were within the range of 50 – 300 ng g-1 fat if they were analyzed for SCCPs. Dumpsites seem to be the most seriously contaminated with SCCPs among the sampled localities in this study, and locations with e-waste, in particular. High levels of SCCPs have also been measured in eggs from Chinese e-waste sites (Zeng, Luo *et al.* 2016, Zeng, Huang *et al.* 2018). Waste, and e-waste in particular, has been found to be a major source of contamination of free-range eggs with SCCPs in earlier studies (Zeng, Luo *et al.* 2016, Adu-Kumi, Petrlík *et al.* 2019) but the contamination of our reference samples is at a level that does not allow us to consider them as background levels of SCCPs. Identifying the sources, i.e. the pathways of SCCPs in the environment, including food, requires more studies as they probably somehow also contaminate commercial feed provided to chicken and/or the space where chicken are kept in large farms, which is somewhat similar to the case of HBCD, but in a different way.

4.6 PER- AND POLYFLUOROALKYL SUBSTANCES (PFAS)

Thirteen out of the thirty-six free-range chicken eggs pooled samples in this study and one reference sample from a supermarket in Jakarta were analyzed for a range of seventeen PFASs[13], including PFOA, PFOS and PFHxS. The results of the analyses are summarized in Annex 1 and in Table 4, but not all results for individual PFASs are presented there. The levels of PFPeA and PFHxA, two of these seventeen PFASs, were below LOQ of 0.01 ng g-1 of fresh weight (fw) in all samples.

The highest level of PFASs, and PFOS in particular, was measured in both samples of eggs from Bangun (see Table A1-2 in Annex 1). This can be

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

[13] 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

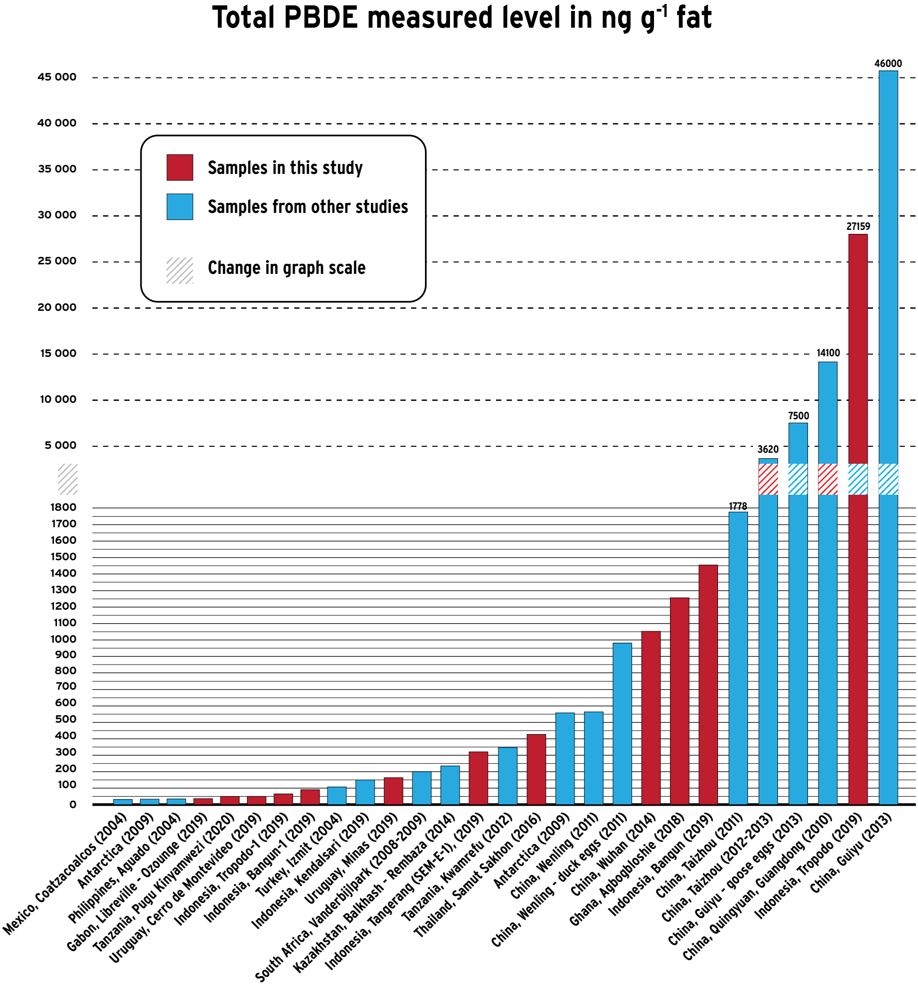

Figure 9: Graph showing the levels of PBDEs in ng g-1 fat measured in free-range chicken or duck eggs in different studies worldwide above a sum total of 30 ng g-1 fat of PBDEs. Only samples that were also analyzed for decaBDE (congener BDE 209) have been included. There are two examples of wild birds from Antarctica included as well for comparison. Specific data and sources of information can be found in Table 7.

explained by the presence of larger volumes of wastes containing these chemicals, including plastics and paper food packaging.

Much lower, but still high levels of PFASs (4.8 – 10 ppm) were measured in eggs from Tangerang, Cerro de Montevideo, Minas – Lavalleja, and Pugu Kinyamwezi (see Table A1-2 and A1-4). The eggs from Uruguay exhibit higher levels of PFOS compared to Tangerang, 4.8 and 9.3 ng g-1 fw in the eggs from Cerro de Montevideo and Minas – Lavalleja respectively. One of the samples from Tangerang (SEM-E-1) contained 2.5 ng g-1 fw of PFOS. The levels of PFOS were not as high at the other localities in this study.

4.7 BACKGROUND LEVELS OF POPs IN EGGS

The approach to establishing background levels of POPs in eggs differs in different studies. It is difficult in the world of today 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 the poultry do not have access to contaminated soil as background level samples (Malisch, Schmid *et al.* 1996, Dvorská 2015). We sampled chicken eggs from supermarkets in five countries and from one convenience store in Kazakhstan from chickens raised on a farm without access to open-air space, in order to obtain information about the background levels of POPs in chicken eggs. The results of the analyses for these samples are provided in Tables 4 and A1-5. The levels of POPs in these samples were mostly either below the level of quantification (LOQ) of the analytical methods used for most of 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 cases of HBCD (Petrlik, Kalmykov *et al.* 2017) and 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 HBCD level in the eggs from a convenience store in Karaganda is among the highest ever measured levels in eggs globally, and the third highest among the egg samples in this study. This is not the first time when such a high level has been measured in eggs from supermarkets or convenient stores. An even higher level of 2,000 ng g^{-1} fat of HBCD was measured in eggs bought in Bavaria, Germany in 2007, but that sample was traced to originate from a small farm where the hens could walk outside (Hiebl and Vetter 2007).

The level of PCDD/Fs measured in eggs from a supermarket in Jakarta was one or two magnitudes lower than in the egg samples from supermarkets used as reference in other countries or studies (Petrlik, Teebthaisong *et al.* 2018, Petrlik, Arkenbout *et al.* 2019). It is also visible from comparison with other reference egg samples in this study (see Table A1-5 in Annex 1).

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

The egg proportion of the total food consumption is different in each country and part of the world. For example, in Indonesia in 2007 it was close to 1% of the total food basket per day according to the World Atlas – Food Security data [14] (Knoema 2012), and is increased by approximately 1/4 of the total amount per day (12 g per person per day) every five years. It would mean that in 2017 consumption would be about 18 g per person per day if the trend remained the same. The assumption for 2016 was 470 g of eggs per person per month according to the World Food Programme, which means approximately 16 g of eggs per person per day (WFP 2017). If we count 35 - 40 g per one free-range chicken egg (the typical weight of free-range chicken eggs in rural areas of Asia and Africa where most sampled hotspots in this study are located) as the average weight it would mean consumption of half such an egg or a little bit less per person per day as the general consumption pattern for the Indonesian population these days. We have reached very similar figures for the population in Kazakhstan or Thailand as well (Petrlík, Kalmykov *et al.* 2016, Petrlík, Dvorská *et al.* 2018). The total consumption of eggs can be much lower in some African countries (Petrlik, Adu-Kumi *et al.* 2019). We have counted with consumption of half an egg as an indicative level for this report.

We have tried to calculate the dietary intake for the following groups of contaminants per day: 1) PCDD/Fs plus DL-PCBs; 2) PBDD/Fs, and 3) PBDEs from the pooled samples in this study. The calculation was made by using measured levels of certain chemicals per gram of fresh weight and the calculation of the daily intake by a presumed consumption of half an egg per day (18 grams of egg weight). An average body weight was taken from information about human body weights in different parts of the world available in literature or from Wikipedia (Walpole, Prieto-Merino *et al.* 2012, Wikipedia 2020c). Different average body weights of an adult person were applied for each of the continents: 60.7 for Africa, 57.7 kg for Asia, 70.8 for Europe and 67.9 for Latin America.

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[14] 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 is depending on the magnitude of wastage 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.

The results of the calculated dietary intake data for each group of hot spots according to contamination sources are summarized in Table 8. The results are also discussed in subchapters 4.8.1 – 4.8.3 for each of the evaluated POPs. The calculations for PCDD/Fs plus dl-PCBs, and for PFOS were also compared with the tolerable weekly intake (TWI) suggested by EFSA and/or WHO. For PBDEs no TWI has been established (JECFA 2006, WHO/FAO 2006).

TABLE 8: RESULTS OF THE CALCULATED DIETARY INTAKE OF POPs DATA FOR DIFFERENT LOCALITIES IN THIS STUDY ARE SHOWN IN THIS TABLE. WE HAVE INCLUDED ONLY PCDD/Fs, DL-PCBs AND PBDEs INTO THIS CALCULATION.

Activity	RE	WY-E	WI	DU	Ref
Total PCDD/F + DL PCBs (pg TEQ kg-1 bw - half an egg)	0.2 - 1	0.4 - 37	0.1 - 11	0.4 - 1.9	0.00009 - 0.04
PBDD/Fs (pg TEQ kg-1 bw - half an egg)	0.2	0.4 - 13	0 - 1.3	0 - 0.16	0
Sum of PBDEs (ng kg-1 bw - half an egg)	0.8 - 8.5	0.1 - 55	0.1 - 1177	0.03 - 5.1	0.01 - 0.4
Exceedance of tolerable intake for PCDD/Fs + dl PCBs for the EFSA 2018 level	0.7 - 4	1.7 - 149	0.5 - 43	1.6 - 7.5	0.0004 - 0.16
Exceedance of tolerable intake for PCDD/Fs + dl PCBs for the WHO 2005 level	0.1 - 0.5	0.2 - 19	0.06 - 5.4	0.2 - 0.9	0.00005 - 0.02
PCDD/Fs + DL PCBs (pg WHO-TEQ in one egg)	24-135	48 - 4400	13 - 1218	52 - 221	0.01 - 4
Number of eggs to reach 140 pg WHO-TEQ per day	1 - 5.7	0.03 - 2.9	0.1 - 10.4	0.6 - 2.7	31.8 - 13155
Number of eggs to reach 17,5 pg WHO-TEQ per day	0.13 - 0.72	0.004 - 0.4	0.01 - 1.3	0.08 - 0.34	4 - 1644

4.8.1 PCDD/Fs, dl-PCBs and PBDD/Fs

By eating half an egg, the tolerable dietary intake for PCDD/Fs and dl-PCBs would not be exceeded in four of the samples of free-range eggs presented in this study. Two of those samples came from Pitarne but in the third and the fourth samples from that location it would however be exceeded. It would also not be exceeded eating half an egg sampled in Kumasi and Guadalajara, however in the sample from Guadalajara it would be exceeded if we also include the brominated dioxins measured in the eggs in the total TEQ level.

On the other hand, by eating half an egg from most of the contaminated samples in Agbogbloshie, Tropodo, Tangerang (SEM-E-1), Samut Sakhon (from 2015), and Aguado (PH-E-7) an adult person of typical weight per continent can exceed the TDI as set by EFSA (EFSA CONTAM 2018a) by 149, 30 – 43, 15, 14, and 12 times respectively.

For most of the sampled eggs contaminated with dioxins and dl-PCBs, an adult person weighing 70 kg can reach the TDI set by EFSA by eating just 4 thousandths and one hundredth of an egg in the case of the samples from Agbogbloshie and Tropodo respectively. The same can be reached by eating 4 and 5 hundredths of an egg from Tangerang and Samut Sakhon or one of the samples from Aguado respectively.

Eating half an egg from the sample most contaminated by dioxins and dl-PCBs among those taken from the vicinity of dumpsites in Pugu Kinyamwezi, Tanzania would exceed EFSA's TDI by 7.5-fold. One egg from this locality can contain around 220 pg TEQ of dioxins and dl-PCBs, which is almost equivalent to the total TDI for 13 persons weighing 70 kg each. One egg from the sample in Agbogbloshie would be enough to reach the TDI for 251 persons weighing 70 kg each. As comparison, one egg from the sample taken in Nairobi – Mirema close to a community cooker would almost be equivalent to the TDI for 4 such persons.

The calculated intake of dioxins from consumption of the eggs with the lowest concentration of dioxins and dl-PCBs, from the vicinity of the medical waste incinerator in Kumasi, would exceed the reference value eight times. The eggs with the highest measured value from Agbogbloshie would exceed those of the reference samples 2,767 times.

Reaching the level of the maximum permissible intake of dioxins and dl-PCBs can be achieved in the monitored localities by consuming, on average, from tenth and a half (from waste yards and e-waste sites) up to almost half of the egg (see Table 8). For eggs from the reference samples, you would have to eat an average of 16 eggs and for eggs from the Jakarta supermarket even up to 1,644 eggs to meet the maximum recommended intake of dioxins and dl-PCBs, i.e. about 17.5 pg WHO-TEQ per day (for a person weighing 70 kg).

In some cases, brominated dioxins contribute significantly to the total TEQ levels in the egg samples and at the same time to the dioxin exposure of the human body, in particular for the egg samples from sites affected by e-waste, because those plastics have originally been treated with BFRs. This is mainly the case for the samples from Agbogbloshie, Wuhan, Tangerang, Samut Sakhon, Bagong Silang, and Guadalajara.

4.8.2 PBDEs

The highest intake of PBDEs was calculated in the pooled egg sample from Tropodo, taken in October 2019, which had extremely high levels of these BFRs. This sample also exhibits a very high ratio of decaBDE congener intake. The second highest intake was calculated for the sample from Agbogbloshie followed by the samples from Wuhan and Bangun (sample taken in November 2019). The calculated intakes from other egg samples from Samut Sakhon and Tangerang are also considerably high.

For the sake of comparison with other studies, we had to discount the decaBDE congener (BDE 209) contribution to the total 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 taken in Tropodo would be 110 ng kg^{-1} bw, according to our previous study from Indonesia (Petrlik, Ismawati *et al.* 2020). It is almost 28 times 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 (JECFA 2006, WHO/FAO 2006).

The calculated intake from eggs sampled in October 2019 in Tropodo or Bangun is even one or two magnitudes higher than that of those found in Chinese polluted areas just for decaBDE (Chen, Cao *et al.* 2014).

4.8.3 PFASs

We have not included calculation of daily intakes of PFASs, as more than half of the egg samples in this study were not analyzed for PFASs (see subchapter 4.6), but the PFOS daily intake was evaluated for eggs from some Indonesian sites in a previous study of ours (Petrlik, Ismawati *et al.* 2020). The highest intake of these chemicals was observed in a pooled egg sample from Bangun, taken in November 2019, with very high levels of n-PFOS as well as br-PFOS isomers.

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 times respectively.

The eggs from Tangerang exhibited the second highest level of PFOS among the sampled eggs from Indonesia in this study, and an adult eating just one egg from a free-range chicken in 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 (Haug, Salihovic *et al.* 2010, Noorlander, Leeuwen *et al.* 2011). The eggs from Uruguay exhibited even higher levels of PFOS compared to Tangerang. When recalculated according to the data for Latin America we reached similar dietary intake levels as for the eggs from Tangerang. By eating half an egg the exceedance levels are 1.4 and 2.7 for Cerro de Montevideo and Minas – Lavelleja respectively. The exceedance level for half an egg from the Tangerang and Pugu Kinyamwezi samples would reach 0.8 and 0.9 respectively, so the contamination of the food chains in all these four locations is at relatively similar levels, but is even more critical in the two localities sampled in Uruguay compared to the contamination in Tangerang. The measured levels of PFOS were not as high in other localities in this study.

Chicken in temple, Thailand. Photo: JOndrej Petrlik, 2016

5. CONCLUSIONS

The levels of POPs in free-range chicken egg samples show that the current plastic waste sorting, dumping and open burning practices lead to serious contamination of the food chain in developing countries. The recycling of some plastics can also lead to serious contamination with POPs as shown by some of the examples included in this study. This applies to PVC and e-waste in particular.

Toxic POPs additives in plastics leak out of them at disposal and recycling sites and contaminate the food chain as demonstrated by extremely high levels of brominated flame retardants and increased levels of PFASs in pooled free-range chicken egg samples in this study. 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 times respectively.

The eggs from large plastic and e-waste scrap yards, as well as eggs from areas where plastic waste is used as fuel or where it is incinerated, seem to be contaminated with extremely high levels of POPs. Agbogbloshie and Tropodo rank among the sites with the highest pollution by POPs globally, according to levels found in free-range chicken eggs and soil and ash samples (Petrlik, Adu-Kumi *et al.* 2019, Petrlik, Ismawati *et al.* 2020).

The maximum level of dioxin contamination in free-range chicken eggs from hot spots in this study is six times higher than the levels measured in a global report by IPEN in 2005.[15] Among results published in recent years only the dioxin levels in free-range chicken eggs from Bien Hoa, a former US military base in Vietnam (Traag, Hoang *et al.* 2012, Hoang, Traag *et al.* 2014, Kudryavtseva, Shelepchikov *et al.* 2020) are similar compared to those presented in this study (see graph in Figure 2).

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[15] A maximum level of 126 pg TEQ g^{-1} fat of PCDD/Fs was measured in eggs from Helwan, Egypt. The highest level of 661 pg TEQ g^{-1} fat of PCDD/Fs measured in this study was from Agbogbloshie, Ghana, but two more samples in this study coming from Tropodo, Indonesia, also exceeded the maximum level previously measured in Helwan.

The mixing of plastics and electronic waste creates an “ideal” combination for the creation of unintentionally produced POPs, such as chlorinated or brominated dioxins. Halogenated plastics like PVC or plastics treated with brominated flame retardants are donors of chlorine and bromine, while electronics contain a variety of metals, including copper which is a typical catalyst for the creation of polyhalogenated dioxins and furans (Olie, Addink *et al.* 1998, Tame, Dlugogorski *et al.* 2003). The burning of this kind of mixture occurring at e-waste sites very often leads to much more severe contamination with dioxins than other open burning of wastes at general dumpsites, and the results of this study confirm that.

Eating half an egg from the sample most contaminated by dioxins and dl-PCBs among those from the vicinity of the dumpsite in Pugu Kinyamwezi, Tanzania exceeds EFSA’s TDI by 7.5 times. One egg from this locality can contain around 220 pg TEQ of dioxins and dl-PCBs, which almost equals the TDI for 13 persons weighing 70 kg each. One egg from the sample taken in Agbogbloshie would be enough to reach the TDI for 251 persons of 70 kg each.

The maximum levels of PBDEs in egg samples in this study taken from sites affected by plastic waste are comparable only to the most seriously contaminated e-waste sites in China, such as for example Guiyu (see graph in Figure 9 and Table 7).

POPs accumulate not only in air releases, but they also bind to particulate matters and/or ash produced by burning plastic waste. The measured levels of dioxins, but also of other non-destroyed POPs, in the ash residues from Agbogbloshie, and the medical waste incinerators in Accra or Tropodo were many times higher than those measured in soil or sediments. These residues, accessible for domestic animals for food, can become a major source and/or important contributor for POPs contamination of the food chain, which then accumulates in dairy products, eggs and meat. This was also confirmed for several hot spots in this study, e.g. Tropodo, Accra – hospital, Samut Sakhon or Agbogbloshie (Petrlík, Dvorská *et al.* 2018, Petrlik, Adu-Kumi *et al.* 2019, Petrlik, Ismawati *et al.* 2020). The waste reprocessing plant in Aguado using waste incineration ash to make bricks (Calonzo, Petrlik *et al.* 2005) and their subsequent use by local residents afterwards also most likely contributes to the high dioxin levels in the free-range chicken eggs from this locality in the Philippines.

The thirty-six free-range chicken egg samples from twenty-five plastic waste hot spots in this study include some samples with the highest levels of POPs ever measured in poultry eggs:

1. Four samples from this study are among the ten highest ever measured levels of chlorinated dioxins in chicken eggs globally, and they are the second, sixth, seventh and tenth highest (see graph in Figure 2).
2. Seven samples have the highest levels of brominated dioxins ever measured in eggs (see graph in Figure 7), although, in general, PBDD/Fs are not measured in eggs very often.
3. Six egg samples in this study are among the ten highest ever measured levels of HBCD in poultry eggs globally, however, not all six come from sites affected by plastic waste disposal or recycling. One of the samples is a reference egg sample from a convenience store in Karaganda.
4. Four samples in this study are among the ten highest ever measured levels of PBDEs in free-range eggs globally. The extremely high level of 27,159 ng g^{-1} fat of PBDEs, and decaBDE in particular, was measured in a pooled egg sample from Tropodo, Indonesia.

The examples of eggs from the store in Karaganda and from Pitarne in the Czech Republic, where the chicken were most likely contaminated by foraging on the polystyrene insulation of the house, show that even the use of plastics with POPs additives can be a source of food chain contamination, which is why the use and disposal of such plastics should be strictly regulated.

E-waste scrap yards or the burning of plastic waste as fuel became obvious sources of serious POPs-contamination of food chains in different parts of the world, but even some recycling operations can be sources of contamination to their surroundings as demonstrated in the cases of hot spots in Mexico, the Czech Republic and Belarus in this study. The results of the contamination of chicken eggs demonstrated in this study show that although their contamination in these locations is not as high as in e-waste sites, it still exceeds the maximum levels suggested to be tolerable by public authorities controlling food contamination such as EFSA by up to six times. Plastics from electronic waste or cars seem to be most problematic even in these operations and should be addressed by stricter controls of additives like brominated flame retardants, as well as of their leakage from recycling plants.

The results of the analyses of free-range eggs from two sites where plastic waste was being used as fuel, either in tofu production or in so-called "community cooker" stoves, show that using plastic waste as fuel leads to serious contamination of the environment with dioxins. There is enough evidence that the burning of PVC is linked to the generation of dioxins, and this and other chlorine-containing plastics may also contribute to the formation of dioxins in Tropodo and Nairobi – Mirema. For example, the 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*[16] *(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 PVC burning (see Table 9).

TABLE 9. THE RELATION OF PCDD/F EMISSION FACTORS ON PVC CONTENT IN BURNED MATERIAL.

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

Source: (Stockholm Convention on POPs 2008)

This study reveals an increasing share of brominated dioxins in environmental contamination and there is no doubt that the cause of this should be sought in the treatment of plastics with brominated flame retardants. In some cases, brominated dioxins contribute significantly to the total TEQ levels in egg samples and at the same time to the total dioxin exposure of the human body. This is mainly the case for the samples from Agbogbloshie, Wuhan, Tangerang, Samut Sakhon, Bagong Silang, and Guadalajara. At almost all these sites e-waste plastics play a significant role.

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[16] The data from Table 7 in the referenced literature were converted into Table 9 in this study.

6. LIMITATIONS OF THE STUDY

The most important limitation of this study is the range of toxic contaminants that we were able to assess in the free-range chicken eggs. This report could focus only on a selected segment of contaminants released from plastic waste or its burning. Burning, or even dumping of plastic can by itself release a much broader scale of toxic chemicals (Simoneit, Medeiros *et al.* 2005, Hahladakis, Velis *et al.* 2018) and we could only focus on some of them within the scope of this report.

Potentially affected areas where plastic waste is handled or disposed of are often vast, and we could not map the situation to its full scale. In order to get a better picture of the level of contamination of the food chain we have chosen free-range chicken eggs as they are proven to be sensitive indicators of POPs contamination in soils/dust. They 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 were 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).

This study also includes some samples analyzed for previous studies, in which more information about the levels of pollution in soil or ash at the places where the eggs were sampled can be found as well. But in general, we must admit that our knowledge about the overall contamination in the different locations is often limited because of the number of samples being limited, and because we have no measured data about, for example, dioxin congener profiles of the air emissions at the sites. Our ability to follow the food chain contamination fully was restricted by the limited resources and time available for this study.

Although the eggs represent a good “sampler” of overall food chain contamination in selected hotspots, they definitely cannot give us a complete picture of the food chain contamination in the studied hotspots. Therefore, it would be very useful for any follow-up monitoring of the selected sites to focus on other types of locally grown food as well. There were cows observed foraging on the rural plastic waste dumpsite in Tangerang, for example (see Figure 10). It would be useful to take samples of the milk from these animals. Contamination of cow’s milk by various chemicals has previously been studied in relation to specific contaminated sites (Braga, Krauss *et al.* 2002, Esposito, Cavallo *et al.* 2009). From sites like Baskuduk in Kazakhstan there are available data about contamination of camel milk (Konuspayeva, Faye *et al.* 2011, Petrlik, Kalmykov *et al.* 2016) which can provide some light on another missing part of the food chain in this study.

Figure 10. Cows walking through the dumpsite in Tangerang, Indonesia. Ash residues after the burning of plastic wastes are visible in this picture as well. Photo: Jindrich Petrlik, Arnika

Figure 11. Camels foraging on waste near a Baskuduk dumpsite in Kazakhstan's Mangystau region. Photo: Martin Skalsky, Arnika

7. RECOMMENDATIONS

7.1 STOPPING WASTE EXPORTS AND POPs EXPOSURE

There is a clear link between current global policy that allows uncontrolled movement of plastic waste or e-waste and toxic chemical contamination of the food chain in places where dumping occurs, such as Agbogbloshie, Tropodo, Tangerang and many other sites presented in this study. The scrap yard in Ghana and the plastic waste yards in Indonesia are only a few examples of many similar sites in Africa, Asia, Latin America and other developing regions where plastic waste and e-waste from developed nations causes environmental and food chain contamination and human exposure. However, there are measures that can be taken to change this situation. These include:

- Set strict limits for POPs in waste. Banned chemicals should be kept out of waste streams and recycling. Materials that are defined as POPs waste must not be transported internationally and must be sequestered and destroyed according to strict protocol. The setting of strict hazardous waste limits for POPs waste is a critical tool for preventing their free movement across borders to developing countries, which are lacking technologies to destroy POPs in waste in an environmentally and health protective manner. These stricter limits (defined as Low POP Content in the Stockholm Convention) should be 50 mg/kg for PBDEs, 100 mg/kg for HBCD and SCCPs and 1 ug TEQ/kg for PCDD/Fs at a maximum.
- Transfer cleaner non-combustion techniques for destruction of POPs and help introduce environmentally sound management of electronic waste in developing countries.
- List brominated dioxins (PBDD/Fs) under the Stockholm Convention.
- Repair loopholes in the e-waste technical guidelines under the Basel Convention.
- Improve illegal shipment surveillance in developed countries’ jurisdictions through the use of intelligence sharing, GPS tracking devices,

and communication with customs agents in regions where the illegal imports are commonly targeted (Africa, Asia, Eastern Europe and Latin America).

- Impose harsh penalties on illegal shippers and brokers as a long-term deterrent.
- All countries around the world that have not already done so should ratify the Basel Ban Amendment which prohibits all exports of materials legally defined as hazardous wastes under the Basel Convention from countries that are members of the EU or the OECD to non-members. This would include used electronic products.

7.2 STOPPING THE FLOOD OF PLASTIC WASTE

This study links waste mismanagement and uncontrolled movement of plastic waste with contamination of the food chain in many different countries. 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 that would occur in emissions of burning such fuel.
4. Remediate sites contaminated with dioxins and other POPs to ensure that human health is protected and food chain contamination cannot occur.
5. Increase the monitoring of POPs chemicals in compliance with Stockholm Convention provisions along with other pollutants of concern.
6. 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.
7. Set better systems for sorting e-waste and prevent use of plastics from electronics as fuel or to be burned.

In May 2019, the Fourteenth Conference of the Parties to the Basel Convention (COP14) agreed by consensus to bring most plastic wastes under the control regime of the Basel Convention (BAN 2019, IPEN 2019). The decision takes effect on January 1, 2021 according to decision BC-14/12 of

POLIS

the Basel Convention (Basel Convention 2019) and is expected to have a major impact on global plastic waste flows and production.

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 indicate that this exemption should be ended. Currently, the Basel Convention Small Intersessional Working Group is examining this issue and will make recommendations to the 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 up front, 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 developing countries like Indonesia, Philippines, Ghana, Kenya or Tanzania. After 1 January 2021, they will have the power to refuse mixed or contaminated wastes through the prior informed consent procedure.

7.3 STRENGTHENING LOW POP CONTENT LEVELS TO STOP TOXIC TRADE

The Basel and Stockholm Conventions establish the Low POP Content Levels (LPCLs) that define 'POPs Waste' and the Stockholm Convention mandates the destruction of POPs wastes. Such waste cannot be exported to developing countries recognizing that they do not have the infrastructure and capacity to manage and destroy them.

Currently, the Low POPs Content levels for brominated flame retardants commonly found in e-waste plastics are very weak. These include polybrominated diphenyl ethers (PBDE), and hexabromocyclododecane (HBCD). The LPCL for plasticizer chemicals also found in plastic waste such as short-chained chlorinated paraffins (SCCP) are also very weak and subject to a proposal by the EU to enshrine the weakest LPCL in the history of the Basel and Stockholm Conventions at 10 000 mg/kg (PCBs and similar POPs have a LPCL of 50 mg/kg).

The deadliest of all POPs – polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDDs and PCDFs), commonly known as chlorinated dioxins and furans – currently has a LPCL that is so weak it allows free use of incineration ash residues and subsequent food chain contamination, particularly for open foraging food production poultry and dairy livestock.

To prevent the contamination of the food chain and stop human exposure to POPs through the e-waste trade and uncontrolled plastic waste incineration, IPEN has proposed the following LPCLs to be adopted at the next Basel and Stockholm Convention Conferences of the Parties – see Table 10.

TABLE 10: IPEN PROPOSAL FOR LOW POPs CONTENT LEVELS (LPCLS) IN COMPARISON WITH CURRENTLY USED ONES, MOSTLY PROMOTED BY THE EUROPEAN UNION, CANADA AND JAPAN.

Substance	IPEN proposal	Current limit
Dioxins and furans (PCDD/F) including dioxin-like PCBs	1 ppb (1 µg TEQ/kg)	15 ppb
Hexabromocyclododecane (HBCD)	100 mg/kg	1000 mg/kg Promoted and used by the EU and other developed countries
Polybrominated diphenylethers (PBDEs)	50 mg/kg as a sum of listed PBDEs. Includes: TetraBDE, PentaBDE, HexaBDE HeptaBDE DecaBDE	1000 mg/kg Promoted and used by the EU and other developed countries
Short-chain chlorinated paraffins (SCCP)	100 mg/kg	10,000 mg/kg Proposed by the EU

Some cases in this report demonstrated that ash residues used for building roads or as other construction materials contribute significantly to spreading pollution into locally grown food. In order to prevent further contamination of food by POPs more protective limits for these chemicals in wastes should be applied. Prohibition of the use of wastes and materials with concentrations of dioxins and dl-PCBs exceeding a level of 50 pg

TEQ g-1 dw (0.05 ppb) on the soil surface would also help, based on studies focused on POPs and ash residues (Weber, Watson *et al.* 2015, Katima, Bell *et al.* 2018, Petrlik, Adu-Kumi *et al.* 2019, Weber, Bell *et al.* 2019).

7.4 CONTROL TOXIC ADDITIVES IN PLASTICS AND SUBSTITUTE MOST HARMFUL PLASTICS

The sites presented in this study also reflect problems with plastic waste generated from domestic use and the POPs present in the food chain are a result of burning halogenated plastics such as PVC or plastics treated with brominated and chlorinated flame retardants. The small medical waste incinerators burn large quantities of medical equipment made of soft PVC. In order to address these problems, we suggest to:

- End all exemptions for the use of toxic additives in plastics, such as exemptions for the use of DecaBDE, SCCPs, PFOS and PFOA.
- Prevent further undermining of the Stockholm Convention's restrictions for POPs – stop any exemptions for newly listed POPs.
- Minimize the use of PVC in medical equipment and ban it for packaging, flooring and other uses where alternatives exist.
- Stop all kinds of recycling exemptions for POPs as additives, including very weak trace contamination limits set e.g. for PBDEs.

International guidelines and rules are still lacking in guidance for decision-makers on the 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;"*, and 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 wastes occurs, like Agbogbloshie, Yaoundé, Pugu Kinyamwezi, Bangun or Tangerang in this study, and many others, would benefit from a phase-out of PVC in the broadest possible sense. There is enough evidence that the burning of PVC is linked to generation of dioxins.

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. The links between medical waste incineration and dioxin formation in the US stimulated a resolution from the American Public Health Asssociation which, *"Urges all health care facilities to explore ways to reduce or eliminate their use of PVC plastics"* (American Public Health Association 1996).

7.5 PREVENT CREATION OF DIOXINS, USE NON-COMBUSTION ALTERNATIVES

Instead of trying to improve dioxin-producing technologies such as small medical waste incinerators, a strategy that prevents dioxin formation is the more cost effective and consistent with the objectives of the Stockholm Convention. This includes changing the hospital waste stream by moving away from PVC products, implementing robust waste segregation since most hospital waste is not infectious, and implementing the use of non-combustion methods such as autoclaves for infectious waste. The Stockholm Convention Guidelines on Best Available Techniques and Guidance on Best Environmental Practices describes the use of source reduction, segregation, recycling, training, and using autoclaves and other non-combustion methods (Stockholm Convention on POPs 2008). The Guidelines note that non-combustion techniques such as autoclaving, *"do not result in the formation and release of chemicals listed in Annex C and should therefore be given priority consideration for their ultimate elimination."* These methods have been implemented as described by WHO, Health Care Without Harm and others (Health Care Without Harm Europe 2004, Emmanuel 2012, UNDP 2015).

Work to implement sustainable healthcare waste management has been underway for some time in developing and transition countries. In Africa, this includes sustainable procurement (Tanzania, Zambia) (HCWH 2015), a non-combustion waste treatment pilot project (Tanzania) (Stringer, Kiama *et al.* 2010), and non-incineration healthcare waste management and mercury-free medical devices (Ghana, Madagascar, Tanzania, Zimbabwe) among others.

Using non-combustion alternative methods for treatment of hazardous waste, e.g. non-combustion technologies for POPs waste disposal (IPEN Dioxin PCBs and Waste Working Group 2010, Basel Convention 2017) or medical waste disposal (Emmanuel 2007, Stringer, Kiama *et al.* 2010, Emmanuel 2012, UNEP 2016) can prevent the creation of UPOPs caused by the incineration of such wastes.

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
na	not applicable
nBFRs	novel brominated flame retardants
ndl-PCBs	non-dioxin-like PCBs
NGO	non-govermental organization (civil society organization)
NIP	National Implementation Plan
NOAEL	no observed adverse effect level
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
PFOA	perfluorooctanic acid
PFOS	perfluorooctanesulfonic acid
PICs	products of incomplete combustion
POPs	persistent organic pollutants
PVC	polyvinyl chloride, one of the broadly used plastics
SC	Stockholm Convention on Persistent Organic Pollutants
SCCPs	short-chain chlorinated paraffins

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 WHO experts panel in 2005
WI	waste incinerator and/or waste incineration
ww	wet weight

ANNEX 1 – RESULTS OF ANALYSES OF FOURTY-ONE INDIVIDUAL POOLED EGG SAMPLES

TABLE A1

L-PFOS (ng g-1 fw)	sum of PFASs (ng g-1 fw)	sum of N-BFRs	sum of PBDEs	sum HBCD	SCCPs	PBDD/Fs (pg TEQ g-1 fat)	Total PCDD/Fs + DL PCBs – DR CALUX (pg BEQ g-1 fat)	Total PCDD/F + DL PCBs (pg TEQ g-1 fat)	DL PCBs (pg TEQ g-1 fat)	PCDD/Fs (pg TEQ g-1 fat)	Fat content (%)	Number of eggs in pooled sample	Date of sampling	Sample ID (eggs)	Locality
0.86	1.47	6.2	230	< LOQ	235	5.37	19	5.89	2.25	3.64	14	5	Apr-19	GUADAL-EGG1	**Guadalajara**
NA	NA	<LOQ	<LOQ	<LOQ	NA	NA	NA	5.82	4	1.82	12	3	Nov-17	N1-3	**Pitárne**
NA	NA	<LOQ	<LOQ	4.5	NA	NA	37	31.85	16.45	15.4	12.1	4	Nov-17	S1-4	**Pitárne**
NA	NA	<LOQ	25	4602	NA		NA	6.85	5.2	1.65	13	3	Sep-17	PIT03	**Pitárne**
NA	NA	<LOQ	<LOQ	6.2	78	NA	NA	9.65	7.58	2.07	10.3	5	Aug-20	PIT 2/2020	**Pitárne**
NA	NA	NA	NA	NA	NA	<1.4	8.1	15.58	11.33	4.25	15.4	3	Jun-14	Gatovo	**Gatovo**
15	26	NA	91	5.2	153	< 21.3	21	14	3.1	11	13	3	May-19	Bangun 1	**Bangun**
76	97	124	1457	538	97		13	15	5.1	9.5	9.5	3	Nov-19	BAN-E-1	**Bangun**
1.8	6.2	33	321	844	153	6.9	88	72	18	54	16.2	3	Nov-19	SEM-E-1	**Tangerang**
NA	NA	NA	NA	NA	NA	NA	NA	28	7.4	20	13.7	5	Nov-19	TAN-ESIN-01	**Tangerang**
0.4	1.1	34	177	< LOQ	160	11	64	20	6	14	13.8	2	Sep-19	PH-E-1 and 2	**Bagong Silang**

L-PFOS (ng g-1 fw)	sum of PFASs (ng g-1 fw)	sum of N-BFRs	sum of PBDEs	sum HBCD	SCCPs	PBDD/Fs (pg TEQ g-1 fat)	Total PCDD/Fs + DL PCBs – DR CALUX (pg BEQ g-1 fat)	Total PCDD/F + DL PCBs (pg TEQ g-1 fat)	DL PCBs (pg TEQ g-1 fat)	PCDD/Fs (pg TEQ g-1 fat)	Fat content (%)	Number of eggs in pooled sample	Date of sampling	Sample ID (eggs)	Locality
NA	NA	< LOQ	3.1	NA	NA	16	100	96	12	84	11.6		Feb-15	Samut Sakhon	**Samut Sakhon**
NA	NA	< LOQ	427	159	173	NA	NA	12	6	6.2	19.4		Feb-16	SMS2-13	**Samut Sakhon**
NA	NA	39	1258	1961	2067	300	840	856	195	661	14.7	4	Dec-18	AGB-E	**Accra - Agbogbloshie (e-waste)**
0.9	2.7	NA	65	0	65	< 21.3	560	232	32	200	15	3	May-19	Tropodo 1	**Tropodo**
0.1	0.3	2166	27159	0	97	0.3	NA	172	32	140	13.9	6	Oct-19	TROP-E-1	**Tropodo**
1.2	2.4	0.7	25	37	65		NA	22	7.5	15	16.1	4	Nov-19	PH-E-3; PH-E-4	**Aguado**
0.3	0.9	0.4	11	5.5	153		NA	7.4	2.1	5.3	13	3	Nov-19	PH-E-5; PH-E-6	**Aguado**
NA	NA	NA	NA	NA	NA	NA	NA	67	14	53	14.4	1	Nov-19	PH-E-7	**Aguado**
							66	23	7.4	16	12.43	4	Jan-20	PH-E-S-5/2	**Aguado**
NA	NA	NA	NA	NA	NA	<3.6	8.8	13	4.7	8.6	12.5	3	Sep-14	Wuhan 2	**Wuhan**
NA	NA	51	1054	NA	NA	27	35	16	3.8	12	15.5	6	Mar-14	Wuhan 1	**Wuhan**
NA	NA	NA	2.3	379	NA	NA	9.6	11	6.8	4.6	14.6	5	Aug-18	YA-2	**Yaoundé-hospital**

Locality	Sample ID (eggs)	Date of sampling	Number of eggs in pooled sample	Fat content (%)	PCDD/Fs (pg TEQ g-1 fat)	DL PCBs (pg TEQ g-1 fat)	Total PCDD/F + DL PCBs (pg TEQ g-1 fat)	Total PCDD/Fs + DL PCBs – DR CALUX (pg BEQ g-1 fat)	PBDD/Fs (pg TEQ g-1 fat)	SCCPs	sum HBCD	sum of PBDEs	sum of N-BFRs	sum of PFASs (ng g-1 fw)	L-PFOS (ng g-1 fw)
Accra - hospital	KBI-E	Dec-18	5	12.3	49	14	63	56	NA	NA	NA	NA	NA	NA	NA
Kumasi - hospital	KU-E	Dec-18	5	14.7	1.7	0.9	2.6	5.2	NA	NA	NA	NA	NA	NA	NA
Nkoltang (MWI)	GA-E-NKOL	Nov-19	5	13.6	12	3.2	15	NA	< LOQ	162	< LOQ	< LOQ	< LOQ	NA	NA
Nairobi - Mirema	KE_001	Jan-20	5	14	12	2.1	14	NA	NA	102	287	24	3.6	1.5	0.6
Cerro de Montevideo	UR-CM-E	Sep-19	4	8.9	7.6	9	17	NA	NA	50	86	50	6.2	5.5	4.3
Minas	UR-MIN-E	Sep-19	4	11.8	4.1	18	23	12	NA	97	57	164	0.6	9.8	7.3
Yaoundé-TKC Quart.	YA-1	Aug-18	6	19.6	NA	NA	NA	NA	NA	152	124	0.5	NA	NA	NA
Yaoundé-Etetak Quart.	YA-3	Aug-18	6	14.3	6.5	6.6	13	NA	0.2	149	25	2.8	NA	NA	NA
Libreville - Owendo	GA-E-OWE	Nov-19	5	13.8	9.7	6.8	16	NA	NA	631	314	< LOQ	< LOQ	NA	NA
Libreville - Ozounge	GA-E-OZOU	Nov-19	5	11.2	13	7.6	21	NA	2	299	5.2	36	< LOQ	NA	NA
Pugu Kinyamwezi	TZ-PU-KI_EGG	Jan-20	9	18	26	9.5	35	NA	3	599	30	50	< LOQ	4.7	2.3
Praeksa	PKS-EGG-1	Nov-15		18	6.2	3.4	9.6	NA	NA	NA	NA	NA	NA	NA	NA
BAS 02	LN 483/16	Oct-16	3	15.6	2.2	11	14	NA	NA	1950	188	28	3.5	NA	NA

Locality	**Prague**	**Jakarta**	**Bangkok**	**Beijing (supermarket)**	**Acrra -supermarket**	**Karaganda – convenient store**
Sample ID (eggs)	PHA-1 (and 2*)	JAK-SUP	supermarket	Beijing (superm.)	ACC-M-E	KAR-SU
Date of sampling	04/2018 (02/2019)	Nov-19	Feb-16	Oct-14	Nov-18	Apr-15
Number of eggs in pooled sample	6 (10*)	6	6	3	6	6
Fat content (%)	10.2	9.5	11.6	10.1	8.8	14
PCDD/Fs (pg TEQ g-1 fat)	0.032	0.0012	0.1	0.2	0.39	0.9
DL PCBs (pg TEQ g-1 fat)	0.337	0.002	0.001	0.28	0.17	0.0003
Total PCDD/F + DL PCBs (pg TEQ g-1 fat)	0.369	0.0032	0.1	0.48	0.56	0.9
Total PCDD/Fs + DL PCBs – DR CALUX (pg BEQ g-1 fat)	NA	LOQ <0.6	NA	1.2	1.2	NA
PBDD/Fs (pg TEQ g-1 fat)	NA	NA	< 21.3	<1.8	< LOQ	NA
SCCPs	25*	136	-	NA	62	NA
sum HBCD	< LOQ	< LOQ	< LOQ	NA	< LOQ	1036
sum of PBDEs	< LOQ	1.39	3.1	0.2	11.2	9.5
sum of N-BFRs	< LOQ	< LOQ	< LOQ	3.7	< LOQ	0.29
sum of PFASs (ng g-1 fw)	NA	0.1	NA	NA	NA	NA
L-PFOS (ng g-1 fw)	NA	<0.01	NA	NA	NA	NA

ANNEX 2 – COMPARISON OF BALANCE BETWEEN PCDD/Fs AND DL-PCBs AND BETWEEN PCDD AND PCDF CONGENERS

Table A2 over the next three pages shows the balance between PCDD/Fs and dl-PCBs and between PCDD and PCDF congeners in individual pooled egg samples from locations affected by plastic waste management presented in this study.

Color coding and explanation for groups of activities:

RE	recycling + pre-recycling processes
WY-E	waste yards, large e-waste sites
WI	waste incineration, waste to energy
DU	dumpsites, and Ref – reference samples from supermarkets or convenient stores.

Activity	DU	DU	RE	RE	RE	RE	WI	DU	RE	DU	WY-E	DU
Locality	Baskuduk	Minas	Pitárne	Pitárne	Gatovo	Pitárne	Yaoundé-hospital	Cerro de Montevideo	Pitárne	Yaoundé-Etetak Q.	Samut Sakhon	Libreville - Owendo
Sample ID	BAS 02	UR-MIN-E	PIT-2/2020	PIT03	Gatovo	N1-3	YA-2	UR-CM-E	S1-4	YA-3	SMS2-13	GA-E-OWE
PCDD/Fs (pg TEQ g^{-1} fat)	2.16	4.13	2.07	1.65	4.25	1.82	4.62	7.62	15.41	6.47	6.23	9.69
DL PCBs (pg TEQ g^{-1} fat)	11.48	18.39	7.58	5.21	11.33	4.00	6.76	9.00	16.44	6.58	6.00	6.75
PCDD/F + dl PCBs	13.64	22.52	9.65	6.86	15.583	5.82	11.38	16.62	31.85	13.05	12.23	16.44
PCDD/Fs	15.82%	18.33%	21.42%	24.04%	27.28%	31.31%	40.60%	45.83%	48.36%	49.58%	50.95%	58.93%
dl PCBs	84.18%	81.67%	78.58%	75.96%	72.72%	68.69%	59.40%	54.17%	51.64%	50.42%	49.05%	41.07%

RE	recycling + pre-recycling processes
WY-E	waste yards, large e-waste sites
WI	waste incineration, waste to energy
DU	dumpsites, and Ref – reference samples from supermarkets or convenient stores.

Activity	RE	DU	DU	WI	WY-E	WI	WI	WI	WY-E	WI	DU
Locality	Guadalajara	Libreville – Ozounge	Praeksa	Wuhan	Bangun	Aguado	Kumasi – hospital MWI	Aguado	Bagong Silang	Aguado	Pugu Kinyamwezi
Sample ID (eggs)	GUA-DAL-EGG1	GA-E-OZOU	PKS-EGG-1	Wuhan 2	BAN-E-1	PH-E-3; PH-E-4	KU-E	PH-E-S-5/2	PH-E-1; PH-E-2	PH-E-5; PH-E-6	TZ-PU-KI_EGG
PCDD/Fs (pg TEQ g^{-1} fat)	3.64	12.94	6.17	8.59	9.50	14.54	1.74	15.89	14.45	5.34	25.62
DL PCBs (pg TEQ g^{-1} fat)	2.25	7.65	3.41	4.70	5.06	7.46	0.86	7.42	5.98	2.09	9.50
PCDD/F + dl PCBs	5.89	20.59	9.58	13.29	14.56	22.00	2.60	23.31	20.43	7.428084	35.12
PCDD/Fs	61.80%	62.85%	64.44%	64.61%	65.26%	66.11%	67.01%	68.15%	70.72%	71.93%	72.96%
dl PCBs	38.20%	37.15%	35.56%	35.39%	34.74%	33.89%	32.99%	31.85%	29.28%	28.07%	27.04%

Activity	WY-E	WY-E	WI	WY-E	WY-E	WI	WI	WI	WI	WI	WI	WY-E
Locality	Tangerang	Tangerang	Wuhan	Agbogbloshie	Bangun	Accra - hospital	Nkoltang (MWI)	Aguado	Tropodo	Nairobi - Mirema	Tropodo	Samut Sakhon
Sample ID (eggs)	TAN-ESIN-01	SEM-E-1	Wuhan 1	AGB-E	Bangun 1	KBI-E	GA-E-NKOL	PH-E-7	TROP-E-1	KE_001	Tropodo 1	Samut Sakhon
PCDD/Fs (pg TEQ g^{-1} fat)	20.40	54.20	12.17	661.06	10.80	49.15	11.52	53.07	139.88	11.65	200.00	84.04
DL PCBs (pg TEQ g^{-1} fat)	7.41	17.55	3.79	194.75	3.10	14.00	3.21	13.75	32.10	2.05	32.00	11.67
PCDD/F + dl PCBs	27.81	71.75	15.96	855.81	13.90	63.15	14.73	66.82	171.98	13.70	232.00	95.71
PCDD/Fs	73.37%	75.54%	76.23%	77.24%	77.70%	77.83%	78.18%	79.42%	81.33%	85.03%	86.21%	87.80%
dl PCBs	26.63%	24.46%	23.77%	22.76%	22.30%	22.17%	21.82%	20.58%	18.67%	14.97%	13.79%	12.20%

RE	recycling + pre-recycling processes
WY-E	waste yards, large e-waste sites
WI	waste incineration, waste to energy
DU	dumpsites, and Ref – reference samples from supermarkets or convenient stores.

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@arnikaorg

for a toxics-free future

ipen.org

ipen@ipen.org

@ToxicsFree