











## ACCRA - YAOUNDÉ - GOTHENBURG - PRAGUE MARCH, 2019

# Persistent Organic

# Pollutants (POPS) in Eggs:

(FINAL DRAFT REPORT - DO NOT CITE)

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## **Executive Summary**

Incineration of medical waste and open burning of waste – including electronic waste – are potentially large sources of toxic chemicals known as persistent organic pollutants (POPs). These substances are slated for global reduction and elimination under the Stockholm Convention.

Medical waste incineration and open burning are common in developing countries and both are listed in the Stockholm Convention as source categories for unintentionally-produced POPs such as hexachlorobenzene (HCB), hexachlorobutadiene (HCBD), pentachlorobenzene (PeCB), PCBs, chlorinated dioxins/furans (PCDD/F), and polychlorinated naphthalenes. Municipal and electronic waste is also known to contain other chemicals listed in the Stockholm Convention such as short chain chlorinated paraffins (SCCPs), polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCD).

This study investigated POPs contamination at a total of six sites: the world's largest e-waste scrap yard in Agbogbloshie (Ghana); medical waste incinerators in Accra (Ghana), Kumasi (Ghana) and Yaoundé (Cameroon); and two open-burning waste dump sites in in Yaoundé (Cameroon). The study measured POPs in eggs because free-range chickens are "active samplers" of materials on the ground. Eggs also represent an important human exposure pathway through consumption. To our knowledge, this is the first study to measure POPs in free-range chicken eggs from hens foraging at the Agbogbloshie e-waste scrap yard, as well as in Yaoundé.

#### The key findings of this study are:

#### High levels of POPs were found at all six sites

The sampling revealed very high levels of chlorinated dioxins, brominated dioxins, PCBs, PBDEs, and SCCPs in the eggs of chickens that had foraged in areas at the e-waste scrap yard, open burning dump sites and medical waste incinerators.

## Some of the highest levels of POPs ever measured in eggs were found in samples collected at the Agbogbloshie e-waste scrap yard in Ghana

Eggs sampled at the Agbogbloshie scrap yard in Ghana contained the highest level of brominated dioxins ever measured in eggs and one of the highest ever measured levels of the flame retardant chemical, HBCD. These eggs also contained the second highest level of chlorinated dioxins ever measured in poultry eggs. An adult eating just one egg from a free-range chicken foraging in Agbogbloshie area would exceed the European Food Safety Authority (EFSA) tolerable daily intake (TDI) for chlorinated dioxins by 220-fold. Indicator PCBs in these eggs were four-fold higher than the EU standard and dioxins and dioxin-like PCBs were 171-fold higher than the standard. These eggs also contained very high levels of SCCPs and PBDEs and relatively high levels of other POPs such as PeCB and HCB.

#### Eggs sampled near medical waste incinerators exceeded EU dioxin standards

Eggs near the medical waste incinerator in Accra, Ghana exceeded the EU dioxin limit by 13-fold and eggs sampled near the facility in Yaoundé exceeded the limit by more than two-fold. PCBs did not exceed limits, but significant levels were also found. High levels of HBCD were also found in eggs from the vicinity of the Yaoundé waste incinerator and one of the dumpsites.

#### Stockholm and Basel Convention provisions need strengthening

Hazardous waste limits in the Stockholm Convention should prevent the export of POPs waste, including e-waste, but 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 Africa and other developing regions is 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 at a maximum. The Stockholm Convention could be further strengthened by listing brominated dioxins.

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.

## Greater attention is needed to fully implement sustainable healthcare waste management

The data obtained from egg samples near medical waste incinerators in this study reinforce concerns over the inadequate healthcare waste management including the use of small incinerators. None of the medical waste incinerators in this study could be considered to employ Best Available Techniques / Best Environmental Practices due to their design, operation, lack of pollution control and lack of waste management for the waste incineration residues. Changing the hospital waste stream by moving away from PVC products, source reduction, segregation, recycling, training, and use of autoclaves and other non-combustion methods should be prioritized. A hospital facility designed for healing should not pollute the food chain or cause adverse impacts on human health and the environment.

## 1. Introduction

POPs contamination in developing countries can include both domestic and foreign sources. Two potentially large sources are incineration of domestic medical waste and open burning of waste – including electronic waste (e-waste) that comes from developed countries. Both types of sources are listed in the Stockholm Convention as source categories for unintentionally-produced POPs. In addition, electronic waste is known to contain short chain chlorinated paraffins (SCCPs) and flame retardant chemicals listed in the treaty.

Medical waste incineration is a major dioxin source, primarily due to combustion of PVC plastic which is a dominant source of organically bound chlorine [1]. The health sector is also a source of mercury pollution due to improper disposal of mercury-containing thermometers and sphygmomanometers. The Stockholm Convention Guidelines on Best Available Techniques and Guidance on Best Environmental Practices note concerns over small hospital incinerators and that, "Due to the poor design, operation, equipment and monitoring of many existing small hospital incinerators these installations cannot be regarded as employing best available techniques" [2]. In developing countries, medical waste is often not segregated by type and polluting open pit and single chamber incinerators are common. Successful implementation of medical waste management and non-combustion techniques has been demonstrated in developing countries [3-6].

Global estimates of annual e-waste production exceed 40 million tons with an annual growth rate of 4 to 5 percent [7]. The export of electronic waste from developed countries to developing countries [8], under the guise of 'recycling', 'repair' and/or 'reuse', has effectively become a form of hazardous waste dumping that international agreements such as the Basel Convention or Stockholm Convention were created to prevent.

In this study, free-range chicken eggs were used to investigate POPs contamination in Cameroon and Ghana near medical waste incinerators and open burning dumpsites – including a large e-waste site. Free-range chicken eggs are sensitive indicators of POP contamination in soils/dust and represent an important human exposure pathway [9-11]. As "active samplers" they can be used to reveal POPs contamination, particularly in areas impacted by dioxins (PCDD/Fs) and PCBs [12-17].

This study investigated POPs contamination at the Agbogbloshie e-waste scrap yard (Ghana), medical waste incinerators in Accra (Ghana), Kumasi (Ghana) and Yaoundé (Cameroon), and two open-burning waste dump sites in in Yaoundé (Cameroon). To our knowledge, this is the first study to measure POPs in free-range chicken eggs from hens foraging at the Agbogbloshie e-waste scrap yard.

## 2. Sampling and analytical methods

The samples of free-range chicken eggs, soil and waste incineration residues discussed in this report were sampled during second half of 2018. Their analyses were conducted in European laboratories in the period between October 2018 and February 2019.

Six pooled samples of free-range chicken eggs were collected in three African cities: Yaounde, the capital of Cameroon; Accra, the capital of Ghana; and Kumasi, Ghana. Samples of soil, ash or soot were also sampled at sampling sites in Ghana. As performed in other studies, a sample of eggs purchased in a supermarket (in Accra) served as a background sample as it was from not free-range chickens [14]. Six localities in three cities were expected to be contaminated by POPs and particularly unintentionally produced POPs (UPOPs) to a certain level. A basic description of these six 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. Table 1 summarizes the basic data about the size of samples and the measured levels of fat content in each of the pooled samples. All samples were taken in 2018.

Free-range chicken eggs from the four pooled samples (one sample from Yaoundé and three samples from Ghana) and one pooled sample of commercial eggs (non free-range) from Accra were analyzed for polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs)¹ 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 [18]. Briefly, rat liver H4IIE cells stably transfected with an AhR-controlled luciferase reporter gene construct were cultured in an  $\alpha$ -MEM culture medium supplemented with 10% (v/v) FCS under standard conditions (37°C, 5% CO2, 100% humidity). Cells were exposed in triplicate on 96-well microtiter plates containing the standard 2,3,7,8-TCDD calibration range, a reference egg sample (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, soil and waste incineration residues from selected sites in Cameroon and Ghana.

No.	Sample	Locality	Matrix	Month/Year of sampling	Eggs in pooled samples; No. of points sampled (soil/ash)	Fat content (%)
1	YA-1	Yaoundé - TCK Quart.	Eggs	08/2018	6	19.6
2	YA-1	Yaoundé - hospital	Eggs	08/2018	5	14.6
3	YA-1	Yaoundé - Etetak Quart.	Eggs	08/2018	6	14.3
4	ACC-M-E	Accra (supermarket)	Eggs	12/2018	6	8.8
5	AGB-E	Accra - Agbogbloshie	Eggs	12/2018	4	14.7
6	AGB-S-1	Accra - Agbogbloshie	Soil	12/2018	4	-
7	KBI-E	Accra - hospital	Eggs	12/2018	6	12.3
8	KBI-A-1	Accra - hospital	Ash/soil	12/2018	5	-
9	KU-E	Kumasi - hospital	Eggs	12/2018	5	14.7
10	KU-A-1	Kumasi - hospital	Soot	12/2018	NA	-

The DR CALUX® bioassay method has been shown to be a cost-efficient semi-quantitative effect-based toxicity screening analyses for all kinds of stable dioxin-like compounds (PCDD/Fs, dl-PCBs, PBDD/Fs, PBBs, chlorinated and brominated polycyclic aromatic hydrocarbons, N-dioxins)²; however, for confirmation it is recommended to go for more specific PCDD/Fs and dl-PCBs congener specific analyses, which also allows examination of finger prints of dioxins (PCDD/F congener patterns), specific for different sources of pollution. One sample from a Yaoundé representative site was potentially influenced by both waste incineration and open burning of medical waste. All pooled egg samples from Ghana as well as samples of soil, ash and soot 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.

All samples were also analyzed for content of non-dioxin-like (indicator) PCBs (iPCBs), DDT and its metabolites, hexachlorocyclohexanes (HCHs), hexachlorobutadiene (HCBD), pentachlorobenzene (PeCB) and hexachlorobenzene (HCB) in a Czech certified laboratory (University of Chemistry and Technology in Prague, Department of Food Chemistry and Analysis). The analytes were extracted by a mixture of organic solvents hexane: dichloromethane (1:1). The extracts were cleaned by means of gel permeation chromatography (GPC). The identification and quantification of the analyte was conducted by gas chromatography coupled with tandem mass spectrometry detection in electron ionization mode.

The eggs from Yaoundé, Agbogbloshie and the Accra supermarket as well as soil/ash samples from Agbogbloshie were also analyzed for PBDEs and HBCD. Three samples (eggs from Agbogbloshie, Accra supermarket and soil/ash from Agbogbloshie) were also analyzed for novel BFRs³ (nBFRs), tetrabromobisphenol A (TBBPA) and short chain chlorinated paraffins (SCCPs). All of these analyses were conducted in a Czech certified laboratory (Institute of Chemical Technology, Department of Food Chemistry and Analysis).

Identification and quantification of PBDEs and nBFRs were performed using gas chromatography coupled with mass spectrometry in negative ion chemical ionization mode (GC-MS-NICI). Identification and quantification of HBCD isomers were performed by liquid chromatography interfaced with tandem mass spectrometry with electrospray ionization in negative mode (UHPLC-MS/MS-ESI).

The extract which was prepared same way as for the other analyses was transferred into cyclohexane and diluted. Identification and quantification of SCCPs was accessed via gas chromatography/time-of-flight high resolution mass spectrometry (GC/TOF-HRMS) in the mode of negative chemical ionization (NCI). The free-range chicken egg samples from the Agbogbloshie scrap yard, the control group chicken egg sample from supermarket in Accra and mixed soil/ash samples from Agbogbloshie, were also analysed for polybrominated dibenzo-p-dioxins and dibenzofurans (PBDD/Fs) in MAS laboratory, Muenster, Germany. 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 <sup>13</sup>C<sub>13</sub>-labelled PBDD/F internal standards to the sample extract
- » Multi-step chromatographic clean-up of the extract
- » Addition of <sup>13</sup>C<sub>12</sub>-labelled PBDD/F recovery standards
- » HRGC/HRMS analysis
- » Quantification via the internal labelled PBDD/F-standards (isotope dilution technique and internal standard technique).

<sup>&</sup>lt;sup>1</sup> Synonym "dioxins" is used for this group of chemicals as well, while "brominated dioxins" applies for PBDD/Fs, another group of polyhalogenated dibenzo-p-dioxins and dibenzofurans. We use both these shorter synonyms in this report.

<sup>&</sup>lt;sup>2</sup> "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 [19]. European Commission, 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, Editor. 2012: Official Journal of the European Communities. p. L 84, 23.3.2012, p. 1–22.

<sup>&</sup>lt;sup>3</sup> This group of chemicals is represented by following chemicals: 1,2-bis(2,4,6-tribromophenoxy) ethane (BTBPE), decabromodiphenyl ethane (DBDPE), hexabromobenzene (HBB), octabromo-1,3,3-trimethylpheny-1-indan (OBIND), 2,3,4,5,6-pentabromoethylbenzene (PBEB), and pentabromotoluene (PBT).

## 3. Description of hot spots

Localities chosen for sampling in Ghana and Cameroon were sites where higher exposure to unintentionally produced POPs, such as dioxins were expected due to the activities on or near the sites. Those sites were of two categories: 1) sites with open burning of waste, and electronic waste in particular (dumpsites and e-waste scrap yards), and 2) medical waste incinerators. Most of them were located in capitals of these two African countries. One site in Ghana was located in Kumasi which is the second largest city in the country. Each site is described more detailed in the following text.

#### 3.1 Ghana

#### 3.1.1 Accra - Agbogbloshie, e-waste scrap yard

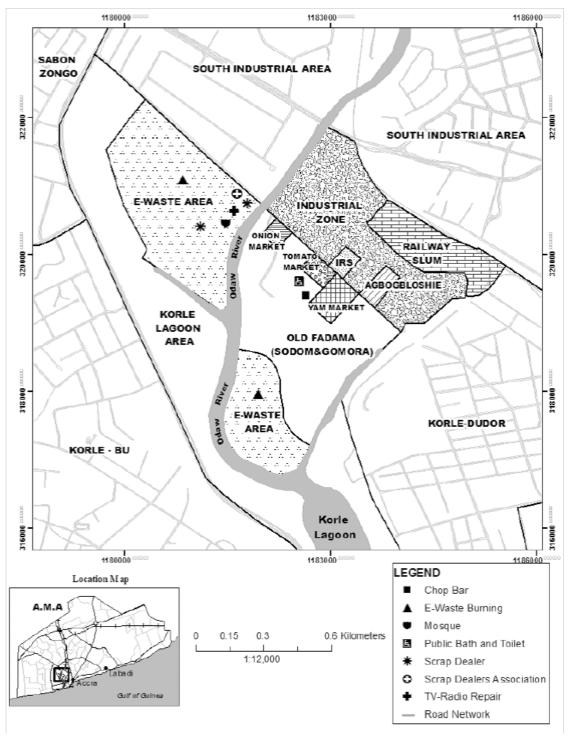
Agbogbloshie is part of Ghana's capital city Accra. It is the nickname of a commercial district on the Korle Lagoon of the Odaw River, near the city center. It became known as a destination for automobile and electronic scrap collected from Accra, but also from many other locations. Roughly 40,000 Ghanaians inhabited the area of Agbogbloshie according the estimates made between years 2009 - 2011, most of whom are migrants from rural areas [20]. In 2018, the population is estimated to be even higher. Oteng-Ababio et al. [21] claim that this settlement now serves over 80,000 people.

The Agbogbloshie market and scrap yard is situated on flat ground alongside the Densu River. During periods of heavy rainfall much of the site becomes flooded and, during these times, it is likely that surface dusts and soils, along with any chemical contaminant that may contain, are carried into the adjacent, lower-lying lagoons and the Densu river which ultimately flows into the ocean. For more details about the location, see Figure 1.

The Basel Action Network (BAN) has referred to Agbogbloshie as a "digital dumping ground". Its recent report confirmed Ghana as one of the destinations for used electronics (in fact e-waste) exported from European Union. "There have been numerous stories in the press over the years about Agbogbloshie, and with it much denial by some that the location is really a significant global dumpsite for imported electronic waste," states BAN in its recent report [8]. Prior studies have shown that Agbogbloshie recycling site received approximately 171,000 tonnes of e-waste in 2009, which were exclusively processed through informal practices [22].



Figure 1: Map of the Agbogbloshie market and scrap yard area. Source: [23],



At Agbogbloshie, the main electronic wastes being processed are obsolete computers, monitors and televisions. These are manually dismantled at numerous small workshops within the market. Certain materials, mainly plastic coated wires and cables, are subsequently taken to sites on the edge of the market where they are burned to enable the separation of metals from plastics. This work is commonly done by children or very young men.

Concerns remain over methods of waste processing at Agbogbloshie scrap yard, especially burning emitting toxic chemicals into the air, land and water. Exposure is especially hazardous to children, as toxic chemicals released due to activities at the site are known to inhibit the development of the reproductive system, the nervous system, and development of the brain. Concerns about human health and the environment of Agbogbloshie continue to be raised as the area remains heavily polluted [20].

A number of studies have focused on various consequences of this e-waste and car scrap yard in Accra as it raises not only concerns regarding the environmental pollution but also its social aspects [23-26] such as poverty of the community living at this site and related economic perspectives [27].

Previous studies found serious contamination of the air, soil, sediments or water by various contaminants including heavy metals [28, 29], polybrominated diphenyl ethers (PBDEs) [28, 30], polychlorinated biphenyls (PCBs) [31, 32], polychlorinated naphthalenes (PCNs) [33], chlorinated and brominated polycyclic aromatic hydrocarbons [34], and polychlorinated as well as polybrominated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs, PBDD/Fs) [31, 35, 36].

Other studies focused on levels of PCDD/Fs [37] and/or lead [38] in blood, PCBs in breast milk [39], and PAHs [40] or heavy metals in urine [41, 42] of workers at the Agbogbloshie scrap yard. Health aspects of living and working at Agbogbloshie was a topic discussed in separate studies [43-45]. Huang et al. [46] focused on effects of contamination on water organisms in Korle Lagoon.

Some reports looked at the situation in the Agbogbloshie scrap yard from broader perspective [24, 26, 47-51].

In addition to this major site in Accra itself, smaller e-waste recycling and disposal operations can be found in other cities. For example, a scrap yard in Korforidua, a smaller city to the north of Accra, is thought to be typical of these numerous small e-waste recycling operations within Ghana, engaged in similar activities to those at Agbogbloshie, but on a far smaller scale [47].

#### 3.1.2 Accra – medical waste incinerator

A small medical waste incinerator in Accra was second site chosen for sampling.

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" [52]. Adjacent to the incinerator is the ash dump site where the bottom ash and some fly ash was disposed of after incineration. This waste incinerator started operation in 2004 and stopped working several years ago. However, the ash dumpsite was left next to the waste incinerator. There is family living in a house next to the waste incinerator and raising chickens which have access to whole area including the ash dumpsite.

This waste incinerator was studied by Adama et al. [52]. Their study has focused on heavy metals in ash and soil in the area surrounding the waste incinerator. They anticipated: " … that continuous exposure to heavy metals in ash and soil may pose direct health risk to waste workers at the incinerator site and unauthorized persons who come to the waste incineration area and remotely through the consumption of exposed plants and animals that may have accumulated heavy metals in their tissues and water sources contaminated with heavy metals or by the inhalation of heavy metal laden dust from polluted soils or ash" [52].

The Stockholm Convention has identified waste incineration as a sector "for comparatively high formation and release" of persistent organic pollutants such as dioxins, furans, PCBs, hexachlorobenzene and pentachlorobenzene [53].









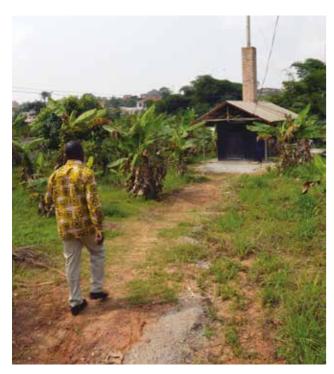






Figure 4: Photo shows small medical waste incinerator in Yaoundé, in which surrounding the eggs were sampled. Photo by CREPD.

#### 3.1.3 Kumasi – medical waste incinerator

In Kumasi 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 in the area of the hospital 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 operated for 8 years now.

#### 3.2 Cameroon

#### 3.2.1 Yaounde, medical waste incinerator

In order to address the negative impacts of improper disposal of medical wastes, especially contaminated sharps from the Cameroonian health care facilities, low-cost small-scale incinerators with very short chimenies are constructed and used in urban and rural hospitals. For this study, we choose the Mimboman health care centre located in the Eastern of the City of Yaoundé, the political capital of Cameroon. This healthcare centre is located in a densely populated residential area. The low cost incinerator operates with gasoline a couple of days in a week or so. Categories of wastes incinerated include plastics and other materials containing polyvinyl chloride, syringes and needles, and biological residues. Ashes from the incinerators are buried in open pits close to the incinerator. Both the incinerator and the open pits are within the hospital premises, less than 100 m to the nearby homes.

#### 3.2.2 Yaounde - waste dumpsites

Based on the criteria of close proximity with homes where free-range chicken are raised, the composite nature of the waste dumped (organic matter, cardboard, plastics, electronics, cables, tyres, etc...); two waste dumpsites subjected to regular open burning as 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 TCK quarters in Yaoundé.

## 4. Results and discussion

## 4.1 Free-range chicken eggs

Results of chemical analyses of six free-range chicken eggs samples from Yaoundé, Accra and Kumasi for various POPs are summarized in Table 2. Details about sampling and sampled localities are in chapters 2 and 3. Their evaluation is discussed further in separate subchapters according the natural groups of POPs. There is no special subchapter dedicated to organochlorine pesticides in these eggs, DDT and metabolites and HCHs as they were not found in very high levels in our samples in comparison with samples from some other locations in Africa, e.g. eggs from Vikuge, Tanzania sampled in 2005 with observed level of DDT at 7041 ng g<sup>-1</sup> fat [54].

Table 2: Overview of results of chemical analyses for POPs in six free-range chicken egg samples, and one egg sample from a commercial farm from two African countries, Cameroon and Ghana. Samples were taken in 2018. Levels of POPs are in ng g¹ fat if not specified otherwise.

Locality	Yaoundé- TCK Quart.	Yaoundé -hospital	Yaoundé-Ete- tak Quart.	Accra – Agbogbl.	Accra - hospital	Kumasi - hospi- tal	Accra-su- per-market	EU stand./
Sample	YA-1	YA-2	YA-3	AGB-E	KBI-E	KU-E	ACC-M-E	limits
Fat content (%)	19.6	14.6	14.3	14.7	12.3	14.7	8.8	
PCDD/Fs (pg TEQ g <sup>-1</sup> fat)	NA	4.6	NA	661	49	1.7	0.39	2.50
DL PCBs (pg TEQ g-1 fat)	NA	6.8	NA	195	14	0.86	0.17	
Total PCDD/F + DL PCBs (pg TEQ g <sup>-1</sup> fat)	NA	11.4	NA	856	63	2.6	0.56	5.00
PCDD/Fs DR CALUX (pg BEQ g <sup>-1</sup> fat)	NA	4.5	NA	NA	NA	NA	NA	
Total PCDD/ Fs + DL PCBs - DR CALUX (pg BEQ g <sup>-1</sup> fat)	NA	9.6	NA	840	56	5.2	1.2	
PBDD/Fs (pg TEQ g <sup>-1</sup> fat)	NA	NA	NA	300	NA	NA	< 8.5	
НСВ	1.5	1.4	7.1	25.1	3.63	0.76	< 0.2	-
PeCB	0.56	0.35	4.7	22.4	2.88	< 0.2	< 0.2	
HCBD	< 0.1	< 0.1	< 0.1	< 0.2	< 0.2	< 0.2	< 0.2	
7 PCB	28	32	36	286	7.8	< 1.4	< 1.4	-
6 PCB	27	30	34	168	7.8	< 1.2	< 1.2	40.00
PCNs *	NA	NA	NA	< 1.4	NA	NA	< 1.4	-
SCCPs	NA	NA	149	2067	NA	NA	62	
sum HCH	4.5	2.5	7.6	< 0.6	< 0.6	< 0.6	< 0.6	
sum DDT	39	22	36	9.7	79	0.82	< 1.2	

<sup>\*</sup>Seven PCN congeners were measured: PCN 52, 56, 66, 70, 73, 74 and 75.

Measured levels of POPs in chicken eggs were compared with legislative limits established in the European Union, although not all measured chemicals in this study have defined limits. For example, the European Union does not currently have a limit for brominated flame retardants or PBDD/Fs. In chicken eggs. Limit values for 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.

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Table 3: Limit concentration values for OCPs, PCBs and PCDD/Fs-TEQs in chicken eggs.

	Hen eggs			
	EU ML <sup>1</sup>	EU MRL <sup>2</sup>		
Unit	pg g⁻¹ fat	ng g⁻¹ fresh weight		
WHO-PCDD/Fs TEQ	2.5	-		
WHO-PCDD/Fs-dl-PCB TEQ	5.0	-		
PCBs <sup>3</sup>	40	-		
НСВ	-	20		
DDT total <sup>4</sup>	-	50		
γ-HCH (lindane)	-	10		
α-, β-HCH*	-	20, 10		

#### Notes to the Table:

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

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

3sum of PCB28, PCB52, PCB101, PCB138, PCB153 and PCB180

<sup>4</sup>sum of p,p´-DDT, o,p´-DDT, p,p´-DDE and p,p´-DDD

\*for each isomer is MRL set separately

#### 4.1.1 Dioxins (PCDD/Fs) and other unintentionally produced POPs

Annex C of the Stockholm Convention lists six unintentionally-produced POPs: HCB, hexachlorobutadiene (HCBD), pentachlorobenzene (PeCB), PCBs, PCDD/F, and polychlorinated naphthalenes. Eggs measured in this study contained HCB, HCBD, PeCB, PCBs, and PCDD/Fs. Polychlorinated naphthalenes were not measured only in two egg samples and one soil sample (see chapter 4.2). They were below LOQ in egg samples.

#### 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) [55]. 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 [56]. These new TEFs were used to evaluate dioxin-like toxicity in pooled samples of chicken eggs from two African countries as well as in sampled soil and waste incineration residues from Ghana (see Tables 2 and 8).

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 [57, 58]. Laboratory animals given dioxins suffered a variety of effects, including an increase in birth defects and stillbirths. Fish exposed to these substances died shortly after the exposure ended. Food (particularly from animals) is the major source of exposure for humans [59].

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.<sup>4</sup> The production of 2,4,5 T pesticide as basic ingredient for Agent Orange left one of the most seriously contaminated sites in Europe [61-63] and sick workers with many symptoms of exposure to most toxic of dioxin congeners 2,3,7,8-TCDD [64, 65].

#### PeCB and HCB

PeCB and HCB are primarily produced unintentionally during combustion, as well as thermal and industrial processes. In the past, they were produced intentionally as pesticides or technical substances and present as impurities in products such as solvents or pesticides. PeCB was used as a component in PCB products, in dyestuff carriers, as a fungicide, a flame retardant and as a chemical intermediate for the production of the pesticide, quintozene [66]. In high doses, HCB is lethal to some animals and, at lower levels, adversely affects their reproductive success. HCB has been found in food of all types [59]. Pentachlorobenzene is very toxic to aquatic organisms and may cause long-term adverse effects in the aquatic environment [67].

#### **HCBD**

HCBD occurs as a by-product during the the production of chlorinated hydrocarbons such as perchloroethylene, trichloroethylene and carbon tetrachloride. It is also formed unintentionally during incineration processes including incineration processes of acetylene and incineration of chlorine residues. Perchloroethylene is widely used in dry cleaning and trichloroethylene and carbone tetrachloride have been used extensively as degreasing agents and as a solvents for other chlorine-containing compounds. HCBD is very toxic aquatic organisms and and causes kidney damage and cancer in animal studies and chromosomal aberrations in occupationally-exposed humans [68]. Systemic toxicity following exposure via oral, inhalation, and dermal routes. Other effects may include fatty liver degeneration, epithelial necrotizing nephritis, central nervous system depression and cyanosis [59].

#### PBDD/Fs

There are also other unintentionally produced POPs that are not yet listed in the Stockholm Convention. The most relevant groups of unintentionally produced POPs to the sampled site in Agbogbloshie, Ghana are polybrominated dibenzo-p-dioxins and dibenzofurans (PBDD/Fs),<sup>5</sup> which were analyzed in samples from this site. Results for brominated dioxins are discussed in one subchapter together with BFRs (see 4.1.3) as they have been known to be potential by-products of commercial PBDE mixtures since 1986 [69]. 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 [70]. PBDD/Fs are similar to the PCDD/Fs however they have been studied less extensively than their chlorinated analogues.

PBDDs/F have been found to exhibit similar toxicity and health effects as their chlorinated analogues (PCDD/Fs) [71-75]. They can for example affect brain development, damage the immune system and fetus or induce carcinogenesis [75].

"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" [73].

In general, brominated dioxins are less regulated than chlorinated dioxins. For example, PBDD/Fs are not

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<sup>&</sup>lt;sup>4</sup> 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. 60. York, G. and H. Mick. Last ghost' of the Vietnam War. 2008 April 27, 2018 [cited 2018 19-11-2018]; <a href="https://www.theglobeandmail.com/incoming/last-ghost-of-the-vietnam-war/article1057457/?page=all">https://www.theglobeandmail.com/incoming/last-ghost-of-the-vietnam-war/article1057457/?page=all</a>.

<sup>&</sup>lt;sup>5</sup> Synonym "brominated dioxins" is used for this group of chemicals as well, while "dioxins" applies for PCDD/Fs. We use both these shorter synonyms in this report.

currently listed under the Stockholm Convention [53], 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" [76].

Because brominated dioxins tend to be less regulated, there is less data about their presence in the environment. There is also very little information about their presence in consumer products and food, where they can have direct impacts on human health, including in vulnerable groups such as children and women of childbearing age.

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

Several bioanalytical tools are accepted by international standards<sup>6</sup> for measuring dioxin-like activity of environmental and food the 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 such contaminations by dioxins and dioxin-like substances, e.g. for food [77-80]. Five pooled egg samples in this study were analyzed by the DR CALUX® method. The highest level in BEQs was measured in the sample from Agbogbloshie (840 pg BEQ  $g^{-1}$  fat) followed by a sample from the medical waste incinerator in Accra (56 pg BEQ  $g^{-1}$  fat). Also samples from the Yaoundé hospital site and the Kumasi hospital were suspected not to meet the EU limit for PCDD/Fs + dl-PCBs (5 pg TEQ  $g^{-1}$  fat).

All sample results measured by DR CALUX® method (see Table 2; total PCDD/F + dI-PCBs – DR CALUX in pg BEQ  $g^{-1}$  fat) were also in the same order of magnitude in the chemical HRGC/HRMS analysis for PCDD/F + dI-PCBs -TEQ (see Table 2; total PCDD/F + dI-PCBs in pg TEQ  $g^{-1}$  fat):

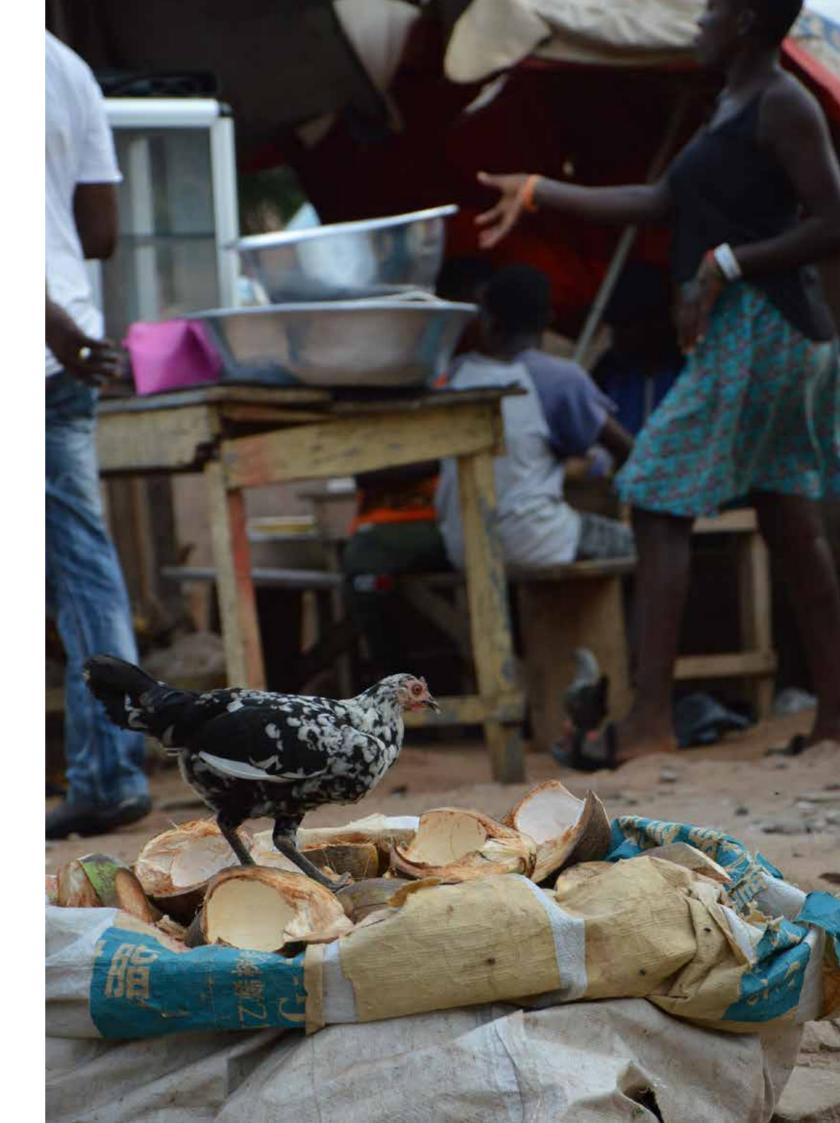
- » Yaoundé hospital (DR CALUX 9,6 vs chemical analysis 11,4; therefore non-compliant according EU guidelines);
- » Agbogbloshie (DR CALUX 840 vs chemical analysis 856; therefore non-compliant according EU guidelines);
- » Accra hospital (DR CALUX 56 vs chemical analysis 63; therefore non-compliant according EU guidelines);
- » Kumasi Hospital (DR CALUX 5,2 vs chemical analysis 2,6; therefore above action levels from EU guidelines) and finally the only in the EU compliant egg samples from the
- » Accra supermarket (DR CALUX 1,2 vs chemical analysis 0,56).

Another study conducted in Arusha, Tanzania found several composite egg samples suspected of exceeding the EU standard of 5 pg TEQ  $g^{-1}$  fat with the highest BEQ concentration found in a sample from Kwamrefu of 20.4 pg BEQ  $g^{-1}$  fat [77], which is higher than that measured in eggs from Yaoundé but lower than eggs from Accra - hospital, most likely influenced by waste incinerator ash left in the area where hens forage.

Positive DR CALUX® activities were measured in 82% of the twenty seven egg samples from Arusha in a study carried out in 2012 [77]. There are several small industries in Arusha and two large regional referral hospitals [1]. Emission from these and uncontrolled burning of waste close to or within the backyards where chickens are scavenging may be the main pathways of dioxin contamination for the chicken in Arusha [77]. This is situation is similar to the places sampled in Yaoundé.

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

<sup>&</sup>lt;sup>6</sup> 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.



#### 4.1.1.2 PCDD/Fs and dl-PCBs

Three out of four free-range chicken egg samples in this study analyzed for PCDD/Fs and dl-PCBs by instrumental analysis exceeded the EU maximum level (ML) of PCDD/Fs and sum of PCDD/Fs and dl-PCBs, expressed as WHO-TEQ (see Table 5) [55]. The background levels for PCDD/Fs and dl-PCBs measured in chicken eggs from a supermarket in Accra were 0.39 and 0.17 pg WHO-TEQ  $g^{-1}$  fat, respectively. The highest level of dioxins (661 pg WHO-TEQ  $g^{-1}$  fat) and dl-PCBs (195 pg WHO-TEQ  $g^{-1}$  fat), respectively, were measured in eggs from Agbogbloshie scrap yard, sampled in a slum close to Korle lagoon in the middle of the area.

The second highest levels of PCDD/Fs and dl-PCBs in this study, 49 and 14 WHO-TEQ g<sup>-1</sup> fat respectively, were measured in eggs sampled from the area close to the closed down medical waste incinerator in Accra. There is waste incineration ash left in that area accessible to foraging hens. However, the dioxin pattern in the eggs shows that other contamination sources might also be contributing dioxins (see graph at Figure 8). The intake of different dioxin congeners and their bioavailability and transfer into eggs may also differ according a recent study: "The data indicate that the bioaccumulation rate depended on the congener; that is, the lower chlorinated PCDDs/PCDFs congeners showed higher bioaccumulation than the higher chlorinated PCDDs/PCDFs congeners" [81].

The only sample from Yaoundé analyzed for PCDD/Fs + dl-PCBs from the vicinity of the small medical waste incinerator and open fire pit had levels of PCDD/Fs and total WHO-TEQ levels of 4.6 and 11.4 pg WHO-TEQ  $\rm g^{-1}$  fat respectively. Both levels exceeded the EU ML, by approximately two-fold. It is necessary to note that the scenario of the sampling in this case was a bit different from other samples as it is a pooled sample from six different households in radius of 0.3 km in all directions from the hospital. So, it reflects the overall situation in the area.

All samples were above the background level of WHO-TEQ measured in eggs from the supermarket by almost 5-fold (Kumasi hospital) to 1528-fold (Agbogbloshie sample). Dioxin levels in samples from Agbogbloshie and Accra hospital are among 15 egg samples with the highest ever measured levels of PCDD/Fs (see graph at Figure 5). Sample of eggs from Agbogbloshie with 661 pg WHO-TEQ  $g^1$  fat of PCDD/Fs is the second highest ever measured level of these chemicals in eggs globally. The highest level was found only in samples of poultry eggs during the dioxin scandal in Belgium in 1999 [82]. It is a level almost six-times higher than the highest concentration of PCDD/Fs measured in free range chicken eggs in IPEN study from 2005 in eggs sampled in Helwan, Egypt – a site with metallurgical plants and a coal-based chemical and cement industry (125 pg WHO-TEQ  $g^{-1}$  fat) [14].

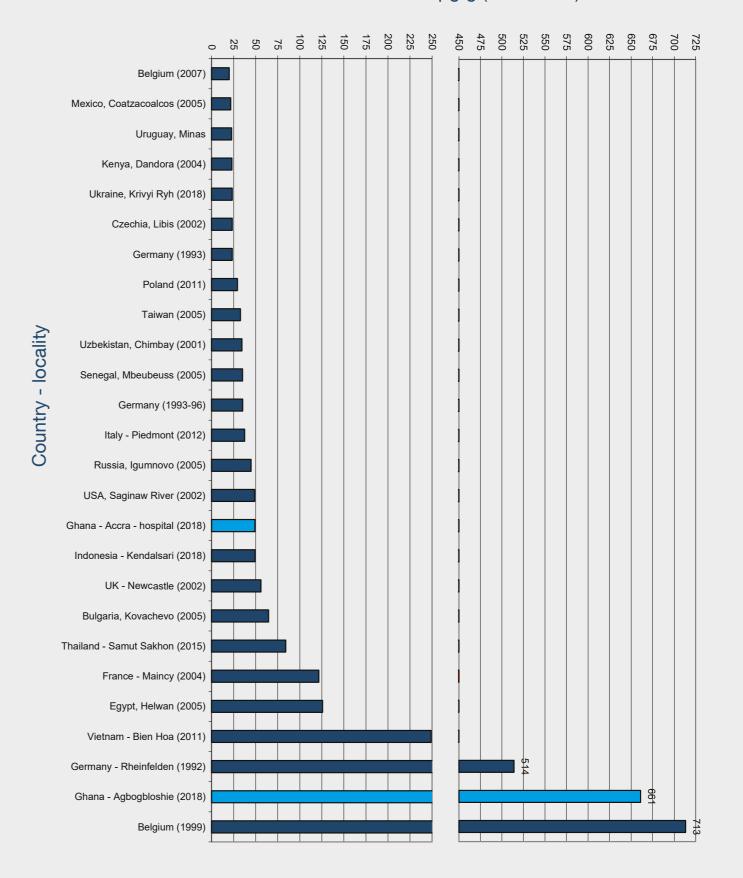
Almost 50 pg WHO-TEQ g<sup>-1</sup> fat of PCDD/Fs in eggs from the Accra hospital site is comparable with levels in another sample (unpublished data) from a site influenced by a secondary aluminium smelter in Indonesia (see Table 4), and also comparable to the highest level of PCDD/Fs in free-range eggs from Newcastle alotments in the area where the incineration ash from the Byker waste incinerator was used to pave the path between alotments in 2000 [16]. It is also close to the highest level of PCDD/Fs and dl-PCBs measured in BEQs in pooled egg samples from another site with impact from waste incineration fly ash in an UK farm in Bishops Cleeve [83] (see Table 9 in this report).

Dioxin-like PCBs were lower in comparison with PCDD/Fs in most of samples but Yaoundé – hospital sample, where dl-PCBs were prevailing, and contributed by almost 60% to total WHO-TEQ in eggs. In all other egg samples dl-PCBs contributed by  $\frac{1}{3}$  to  $\frac{1}{3}$  of the to total WHO-TEQ value. Highest level of dl-PCBs of 195 pg WHO-TEQ  $g^{-1}$  fat was measured in eggs from Agbogbloshie. The lowest dl-PCBs in free range eggs had sample from Kumasi – hospital site, 0.86 pg WHO-TEQ  $g^{-1}$  fat, which exceeded the background level in eggs from the supermarket by 5-fold.

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Figure 5: Graph showing selection of maximum levels of PCDD/Fs measured in chicken eggs in different countries. Samples before 2006 are in WHO-TEQ 1998. Sources of information are listed in Table 4.

#### Measured level in pg/g (WHO-TEQ) of fat



2007     Not specified     20     [82]       2005     Coatzacoalcos     22     [14]       2009     Minas     23     [84, 85]       2004     Nairobi - Dandora     23     [86]       2004     Nairobi - Dandora     23     [86]       2004     Krivyi Ryh     23     [88]       2002     Libis     23     [88]       y     1993     Not specified     23     [88]       2011     Not specified     29     [89]       2005     Changhua county     33     [90]       2005     Changhua county     33     [90]       3005     Mbeubeuss     35     [14]       4     2005     Changhua county     33     [90]       3     2005     Changhua county     33     [90]       4     2005     Mbeubeuss     35     [14]       4     2005     Rendalsari     49     this study       4     2018     Kendalsari     49     this study       8     2015     Kovachevo     65     [14]       9     2014     Maincy (near Melun)     122     [15]       10     2005     Rheinfelden - disposal     249     [93]       1	Country	Year	Locality	PCDD/Fspg WHO-TEQ g <sup>-1</sup> fat	Source	Comments
V         2005         Coatzacoalcos         22         [14]           y         2009         Minas         23         [84, 85]           y         2009         Minas         23         [84, 85]           y         2009         Minas         23         [84, 85]           y         2018         Kriryi Ryh         23         [84]           y         1993         Not specified         23         [87]           y         1993         Not specified         29         [89]           y         2005         Changhua county         33         [90]           y         1993-96         Not specified         35         [14]           y         1993-96         Not specified         35         [91]           y         1993-96         Not specified         35         [14]           y         1993-96         Not specified         35         [91]           y         1993-96         Not specified         49         [14]           y         2015         Kendalsari         49         [14]           y         2005         Kendalsari         49         [14]           y         201	Belgium	2007	Not specified		[82]	-
y         2009         Minas         23         [84, 85]           2004         Nairobi - Dandora         23         [14]           2004         Not specified         23         [86]           2002         Libis         23         [87]           2002         Libis         23         [87]           2002         Libis         23         [87]           2002         Libis         23         [88]           2011         Not specified         29         [89]           2005         Changhua county         33         [90]           19         2005         Mbeubeuss         35         [14]           19         2005         Mbeubeuss         35         [91]           19         2012-13         Piedmont region         38         [92]           19         2012-13         Piedmont region         49         [14]           2005         Igumnovo         49         [14]           49         201         (16]         [16]           49         201         [16]         [16]           49         201         [16]         [16]           40         201         Majory (	Mexico,	2005	Coatzacoalcos	22	[14]	Petrochemical complex; hazardous waste incinerator
2004 Nairobi - Dandora 23 [14] 2018 Krivyi Ryh 23 [86] 2018 Not specified 23 [86] 2002 Libis 23 [87] 2002 2002 2002 2003 2004 Specified 29 [89] 2005 Changhua county 33 3 [90] 2005 Changhua county 33 35 [91] 2005 Mbeubeuss 35 [14] 2005 Rot specified 35 [91] 2012 2012-13 Piedmont region 38 [92] 2012 2005 Igumnovo 45 [14] 2005 Igumnovo 45 [14] 2006 Newcastle 56 [14] 2007 Newcastle 56 [14] 2008 Newcastle 56 [14] 2009 Newcastle 56 [14] 2	Uruguay	2009	Minas	23	[84, 85]	Cement kiln co-incinerating PCBs
8         2018         Krivyi Ryh         23         [86]           197         2002         Libis         23         [87]           193         Not specified         23         [88]           2011         Not specified         29         [89]           2005         Changhua county         33         [90]           10         2005         Mbeubeuss         35         [14]           19         2005         Mbeubeuss         35         [91]           19         2012-13         Piedmont region         38         [92]           2012         2012-13         Piedmont region         38         [92]           8iia         2015         Kendalsari         49         this study           8iia         2018         Kendalsari         49         [14]           9         2005         Kovachevo         65         [14]           6         2005         Kovachevo         65         [14]           10         2004         Maincy (near Melun)         122         [15]           10         2011         Bien Hoa         249         [95]           10         201         Agbogbloshie         661	Kenya	2004	Nairobi - Dandora	23	[14]	Open burning at dumpsite
1902     Libis     23     [87]       194     1993     Not specified     23     [88]       2011     Not specified     29     [89]       2005     Changhua county     33     [90]       1     2005     Mbeubeuss     35     [14]       1     2005     Not specified     35     [91]       1     2012-13     Piedmont region     38     [92]       2005     Igumnovo     45     [92]       2018     Accra - hospital Wl     49     this study       3iia     2018     Kendalsari     49     this study       4     2005     Kovachevo     65     [14]       5     Chamt Sakhon     84     [93]       1     2005     Kovachevo     65     [14]       2     2005     Helwan     126     [14]       1     2005     Helwan     126     [14]       1     2011     Bien Hoa     249     [95]       1     2018     Agbogbloshie     514     this study       1     1999     Not specified     713     (97]	Ukraine	2018	Krivyi Ryh	23	[86]	Metallurgical and coke plants
ny         1993         Not specified         23         [88]           2011         Not specified         29         [89]           2005         Changhua county         33         [90]           1         2005         Changhua county         33         [90]           ny         1993-96         Not specified         35         [14]           ny         1993-96         Not specified         35         [91]           2005         Piedmont region         38         [92]           2012         2013         Piedmont region         38         [92]           2018         Accra - hospital WI         49         this study           siia         2018         Kendalsari         49         this study           siia         2005         Kovachevo         65         [14]           d         2015         Samut Sakhon         84         [93]           n         2005         Helwan         126         [14]           p         2005         Helwan         126         [14]           p         [14]         [95]         [95]           p         Site "Ziegasse"         514         this study	Czechia	2002	Libis	23	[87]	Chlor-alkali plant, dioxin contaminated site
2011     Not specified     29     [89]       1     2005     Changhua county     33     [90]       ny     1993-96     Not specified     35     [14]       ny     1993-96     Not specified     35     [91]       2012-13     Piedmont region     38     [92]       2005     Igumnovo     45     [14]       2005     Igumnovo     49     this study       siia     2018     Kendalsari     49     [93]       Kiingdom     2000     Newcastle     56     [16]       a     2005     Kovachevo     65     [16]       d     2015     Samut Sakhon     84     [94]       2005     Helwan     122     [15]       10     2005     Helwan     126     [14]       10     Rheinfelden - disposal     [14]     [95]       10     Agbogbloshie     661     this study	Germany	1993	Not specified	23	[88]	Either PVC burning or PCP - not clear from [88]
n     2005     Changhua county     33     [90]       al     2005     Mbeubeuss     35     [14]       any     1993-96     Not specified     35     [91]       any     1993-96     Not specified     35     [91]       any     2012     Piedmont region     38     [92]       2005     Igumnovo     45     [14]       2005     Igumnovo     49     [14]       4 Kingdom     2008     Newcastle     56     [16]       8     2005     Kovachevo     65     [14]       9     2015     Samut Sakhon     84     [94]       2015     Samut Sakhon     84     [94]       2016     Maincy (near Melun)     122     [15]       2017     Rheimfelden - disposal     [14]       101     Rheinfelden - disposal     [14]       102     Site "Zielgasse"     514     (15)       103     Helwan     661     (15)       104     Hois study	Poland	2011	Not specified	29	[89]	PCP treated wood
al     2005     Mbeubeuss     35     [14]       any     1993-96     Not specified     35     [91]       any     2012-13     Piedmont region     38     [92]       any     2005     Igumnovo     45     [14]       any     2018     Accra - hospital Wl     49     this study       any     2001     Newcastle     56     [16]       ia     2005     Kovachevo     65     [16]       ia     2005     Kovachevo     65     [14]       any     2005     Samut Sakhon     84     [94]       any     2005     Helwan     122     [15]       any     1992     Site "Zielgasse"     249     [95]       any     1992     Site "Zielgasse"     514     [96]       any     1999     Not specified     713     [97]	Taiwan	2005	Changhua county	33	[90]	Metallurgical plants (steelworks); (duck eggs)
any     1993-96     Not specified     35     [91]       any     2012-13     Piedmont region     38     [92]       any     2005     Igumnovo     45     [14]       any     2018     Accra - hospital WI     49     this study       any     2018     Kendalsari     49     [93]       any     2005     Kovachevo     65     [16]       any     2005     Kovachevo     84     [94]       any     2004     Maincy (near Melun)     122     [15]       any     2011     Bien Hoa     249     [14]       any     1992     Site "Zielgasse"     514     [96]       any     1999     Not specified     713     [97]	Senegal	2005	Mbeubeuss	35	[14]	Mixed waste dumpsite, potential PCP contamination
2012-13       Piedmont region       38       [92]         1       2005       Igumnovo       45       [14]         2018       Accra - hospital WI       49       this study         esia       2018       Kendalsari       49       [93]         ia       2000       Newcastle       56       [16]         ia       2005       Kovachevo       65       [14]         nd       2015       Samut Sakhon       84       [94]         p       2004       Maincy (near Melun)       122       [15]         pm       2005       Helwan       126       [14]         pm       2011       Bien Hoa       249       [95]         pa       Rheinfelden - disposal       [96]       [96]         pa       2018       Agbogbloshie       661       this study         p       1999       Not specified       713       [97]	Germany	1993-96	Not specified	35	[91]	Not specified (free range chicken eggs)
2005     Igumnovo     45     [14]       1     2018     Accra - hospital WI     49     this study       esia     2018     Kendalsari     49     [93]       d Kingdom     2000     Newcastle     56     [16]       ia     2005     Kovachevo     65     [14]       nd     2015     Samut Sakhon     84     [94]       e     2004     Maincy (near Melun)     122     [15]       m     2005     Helwan     126     [14]       m     2011     Bien Hoa     249     [95]       any     1992     site "Zielgasse"     514     [96]       any     1999     Not specified     713     this study	Italy	2012-13	Piedmont region	38	[92]	Secondary aluminium smelter
a       2018       Accra - hospital WI       49       this study         esia       2018       Kendalsari       49       [93]         d Kingdom       2000       Newcastle       56       [16]         ia       2005       Kovachevo       65       [14]         nd       2015       Samut Sakhon       84       [94]         p       2004       Maincy (near Melun)       122       [15]         pm       2005       Helwan       126       [14]         pm       2001       Bien Hoa       249       [95]         pany       1992       site "Zielgasse"       514       [96]         pany       1999       Not specified       713       this study	Russia	2005	Igumnovo	45	[14]	Chlorine chemical industry area; HWI
esia       2018       Kendalsari       49       [93]         ł Kingdom       2000       Newcastle       56       [16]         ł Kingdom       2000       Newcastle       56       [14]         ia       2005       Kovachevo       65       [14]         nd       2015       Samut Sakhon       84       [94]         2004       Maincy (near Melun)       122       [15]         126       Helwan       126       [14]         127       Rheinfelden - disposal       249       [95]         128       Agbogbloshie       514       [96]         129       Not specified       713       [97]	Ghana	2018	Accra - hospital WI	49	this study	Medical waste incinerator ash
4 Kingdom       2000       Newcastle       56       [16]         ia       2005       Kovachevo       65       [14]         nd       2015       Samut Sakhon       84       [94]         e       2004       Maincy (near Melun)       122       [15]         nm       2005       Helwan       126       [14]         nm       2011       Bien Hoa       249       [95]         any       1992       site "Zielgasse"       514       [96]         any       1999       Not specified       713       this study	Indonesia	2018	Kendalsari	49	[93]	Secondary aluminium smelter
iia       2005       Kovachevo       65       [14]         nd       2015       Samut Sakhon       84       [94]         p       2004       Maincy (near Melun)       122       [15]         p       2005       Helwan       126       [14]         p       2011       Bien Hoa       249       [95]         p       Rheinfelden - disposal       249       [95]         p       1992       site "Zielgasse"       514       [96]         p       1999       Not specified       713       [97]	<b>United Kingdom</b>	2000	Newcastle	56	[16]	Waste incineration ash
nd       2015       Samut Sakhon       84       [94]         2004       Maincy (near Melun)       122       [15]         2005       Helwan       126       [14]         nm       2011       Bien Hoa       249       [95]         any       1992       Rheinfelden - disposal site "Zielgasse"       514       [96]         any       2018       Agbogbloshie       661       this study         m (1999)       1999       Not specified       713       [97]	Bulgaria	2005	Kovachevo	65	[14]	Industrial area with coal burning power plants
2004 Maincy (near Melun) 122 [15]  2005 Helwan 126 [14]  m 2011 Bien Hoa 249 [95]  Rheinfelden - disposal 249  1992 site "Zielgasse" 514  2018 Agbogbloshie 661 this study  m (1999) 1999 Not specified 713	Thailand	2015	Samut Sakhon	84	[94]	Artisanal e-waste and general waste recycling; open burning
2005       Helwan       126       [14]         Im       2011       Bien Hoa       249       [95]         Rheinfelden - disposal       514       [96]         any       1992       site "Zielgasse"       514       [96]         am (1999)       1999       Not specified       713       [97]	France	2004	Maincy (near Melun)	122	[15]	Old waste incinerator operating between 1974-2002
2011       Bien Hoa       249       [95]         Rheinfelden - disposal       [96]         1992       site "Zielgasse"       514       [96]         2018       Agbogbloshie       661       this study         1999       Not specified       713       [97]	Egypt	2005	Helwan	126	[14]	Metallurgical workshops
Rheinfelden - disposal 1992 site "Zielgasse" 514 [96] 2018 Agbogbloshie 661 this study 1999 Not specified 713 [97]	Vietnam	2011	Bien Hoa	249	[95]	Former US military base, dioxin contaminated site
2018       Agbogbloshie       661       this study         1999       Not specified       713       [97]	Germany	1992	Rheinfelden - disposal site "Zielgasse"	514	[96]	Waste from chlor-alkali chemical plant
1999 Not specified 713 [97]	Ghana	2018	Agbogbloshie	661	this study	E-waste and automobile scrap yard
	Belgium (1999)	1999	Not specified	713	[97]	Dioxin contamination of feed

#### 4.1.1.3 Hexachlorobenzene, pentachlorobenzene and Hexachlorobutadiene

Among the six free-range egg samples in this study, the highest levels of PeCB and HCB were measured in eggs from Agbogbloshie. None of the samples were above LOQ for HCBD.

The highest level of HCB observed in this study in Agbogbloshie (25 ng g<sup>-1</sup> fat) is equal to the level found in Kwamrefu, Tanzania with a mean value of HCB (23 ng g<sup>-1</sup> fat) [77]. However, the study in Arusha found a much higher maximum level of 167 ng g<sup>-1</sup> fat in the pooled eggs sample from Kwamrefu. Lower levels of HCB in eggs from Yaoundé – TCK and Yaoundé - hospital are comparable to those measured in Mbeubeuss, Senegal, and the highest from Yaoundé – Etetak was comparable to levels observed in Eloor, India in an IPEN study from 2005 [14]. In general none of the observed levels was extremely high nor exceeded EU limit values. The same applies to PeCB in eggs from this study.

#### 4.1.2 Non-dioxin-like PCBs

Levels of 6 or 7 indicator PCB congeners represent a potential influence of technical mixtures of PCBs, which is likely not the outcome of unintentional generation, but intentional production and use. The EU limit for 6 i-PCB congeners in eggs is set at  $40 \text{ ng g}^1$  fat. The egg samples from Agbogbloshie were more than 4-times this limit value for i-PCBs. The reason might be that technical PCBs are present in oils from car wrecks and other WEEE which end up at the scrap yard.

All remaining eggs samples in this study were below 40 ng g<sup>-1</sup> fat. Levels of i-PCBs in eggs from Yaoundé were above half of the limit set in the EU.

#### 4.1.3 PBDD/Fs and BFRs in eggs

With the broad use of brominated flame retardants, the question of the presence of polybrominated dibenzo-p-dioxins and dibenzo-furans in the food chain has arisen, as they are found in different environmental compartments [75]. The WHO expert panel has concluded that polybrominated dibenzo-p-dioxins (PBDDs), dibenzo-furans (PBDFs) and some dioxin-like polybrominated biphenyls (dl-PBBs) may contribute significantly to daily human background exposure to the total dioxin toxic equivalencies (TEQs) [98].

PBDD/Fs are not measured very often in the environment yet, although there are some studies focused on their presence in the African environment. Several studies have focused on PBDD/Fs in environmental compartments at the Agbogbloshie scrap yard [31, 35, 36, 99]. This type of contamination is discussed in a more detailed way in subchapter 4.2 below. Other studies focused on the presence of PBDD/Fs in e-waste and plastic wastes in Nigeria [100, 101]. IPEN and Arnika recently found PBDD/Fs in toys from recycled e-waste plastic sold in Nigeria [102].

The e—waste dismantling process is a significant source of PBDD/Fs releases into the environment as demonstrated in several studies from e-waste dismantling sites in China [103, 104], Vietnam [105] and Thailand [106].

The results of analyses of free-range eggs from Cameroon and Ghana for BFRs and PBDD/Fs are summarized in Table 5. For comparison there are also results in eggs from Wuhan, China and Samut Sakhon, Thailand from which PBDD/Fs were found in high levels previously. The sample from Wuhan had highest ever measured level of PBDD/Fs until this study and analysis of eggs from Agbogbloshie.

Decabromodiphenyl ethane (DBDPE), octabromo-1,3,3-trimethylpheny-1-indan (OBIND), 2,3,4,5,6-pentabromoethylbenzene (PBEB), and pentabromotoluene (PBT) from the group of nBFRs and tetrabromobisphenol A (TBBPA) were measured below LOQ in all samples, and therefore they were not included in the Table 5. This finding is in agreement with measured levels in eggs from Arusha, where these nBFRs were not detected [77].

Table 5: Summarized results of analyses for different BFRs in free-range chicken eggs samples from Cameroon and Ghana (this study) in comparison with samples from Wuhan (China) and Samut Sakhon (Thailand). Also results for eggs from supermarkets (background) in Accra and Beijing respectively are included. The table also contains results of analyses for PBDD/Fs in addition to BFRs. Sources of data from China and Thailand: [94, 107, 108]

Chemicals	$\Sigma$ PBDE	$\Sigma$ HBCD	НВВ	ВТВРЕ	PBDD/Fs			
Units		ng g⁻¹ fat						
Yaoundé - TCK Quart.	0.5	0.5 124 NA NA						
Yaoundé - hospital	2.3	379	NA	NA	NA			
Yaoundé - Etetak Quart.	2.8	25	NA	NA	NA			
Agbogbloshie	1258	1961	1.1	38	300			
Wuhan	1054	NA	< 0.1*	51	27			
Samut Sakhon - SMS E	3.1	NA	< 0.1*	< 0.5*	16			
Samut Sakhon - SMS 2-13	1.3	159	NA	NA	NA			
Accra (supermarket)	11	< 12.6*	< 0.2*	< 0.3*	< 8.5*			
Beijing (supermarket)	0.2	NA	3.7	< 0.5*	< 1.8*			

<sup>\*</sup>below LOQ

We have found only one study assessing PBDD/Fs in chicken eggs in countries other than China and Thailand from which data are incorporated in the Table 5. A report from Ireland showed levels of 0.244 - 0.415 pg TEQ g<sup>-1</sup> fat [109]. It is two orders of magnitude lower than the level measured in free -range chicken eggs samples from Wuhan or Samut Sakhon, and three orders of magnitude lower than in samples from Agbogbloshie.

In the egg samples from Agbogbloshie, high levels of PBDEs and HBCD were also measured (1258 and 1961 ng  $g^{-1}$  fat respectively). The level of PBDEs is comparable to e-waste dismantling sites studied by Labunska et al. [110] as well as the municipal waste incinerator site in Wuhan (see Table 4). A significantly increased level of 51 ng  $g^{-1}$  fat was also measured in a sample from Wuhan for BTBPE<sup>7</sup>, which is comparable to the findings in eggs from Agbogbloshie in this study (38 ng  $g^{-1}$  fat). They exceeded the background samples by at least two orders magnitude (see Table 4). A similar study from Tanzania found 4-times lower levels of BTBPE in eggs from Arusha area. Relatively low levels of HBB were observed in eggs from Agbogbloshie and Arusha [77].

The level of HBCD in eggs from Agbogbloshie is one of the highest ever measured, comparable to very high levels measured in Germany (2000 ng g<sup>-1</sup> fat) [111]. However, in eggs from Shetpe in Kazakhstan the level of HBCD in samples was one order of magnitude higher than that measured in samples from Agbogbloshie. It was speculated that the source of contamination of eggs in Shetpe were chickens feeding among car wrecks and ingesting particles of brominated materials from the deteriorated car interiors that had entered the soil [112, 113]. This exposure pathway is somewhat similar for the eggs from Agbogbloshie. Another potential source of contamination might also be polystyrene foam used in obsolete electronic devices or in their packaging [114].

In two samples from Yaoundé (from TCK Quarter and from the area close to the medical waste incinerator) relatively high levels of HBCD exceeding background levels by 10 to 30-fold were measured. They are much lower in comparison with Agbogbloshie but comparable to levels in eggs from Samut Sakhon (Thailand), Balkhash (Kazakhstan) or Koh Samui (Thailand) [112]. All these sites had some waste related activities as well. HBCD in eggs from two sites in Yaoundé also exceeded levels found in Arusha [77]. HBCD in the third sample from Etetak Quarter is lower than highest levels found in Arusha. PBDEs were low in samples from Yaoundé, lower than some samples measured in Arusha, and close to the minimum levels there [77].

What is remarkable is the level of PBDEs found in pooled sample from the supermarket in Accra which exceeded levels found in several samples of free-range chicken eggs presented in Table 5 including those from Yaoundé.

#### 4.1.4 Short chain chlorinated paraffins (SCCPs)

Egg samples from Agbogbloshie and Accra supermarkets, are, to our knowledge, the first egg samples from Africa which have been analyzed for SCCPs. Measured levels in these samples were 2067, 149 and 62 ng  $g^{-1}$  fat in eggs from Agbogbloshie, Yaoundé – Etetak Q. and Accra – supermarket respectively (see also Table 2). By way of comparison, the EU limit for SCCPs in water is 0.4  $\mu$ g  $I^{-1}$  (~ng  $g^{-1}$ ) [115].

The total concentrations of SCCPs in eggs ranged from 477 to 111000 ng  $g^{-1}$  fat from an e-waste- polluted area in South China [116]. Level of SCCPs in eggs from Agbogbloshie (2067 ng  $g^{-1}$  fat) is higher than minimum level but it is also much lower than maximum level from the South China site.

#### 4.1.5 Background levels of POPs in eggs

The Agbogbloshie scrap yard has been the focus of numerous previous studies regarding its contamination by POPs and heavy metals. Most of them focused either on soil or human tissue contamination by different POPs, especially polyhalogenated dibenzo-p-dioxins and dibenzofurans. Our study is focused on free-range chicken eggs as part of the diet for people living on the scrap yard as well as part of local food chain. To our knowledge, this is the first study to measure POPs in free-range chicken eggs from hens foraging in the area of the Agbogbloshie scrap yard. We also analyzed one pooled soil sample from the closest area to the location where the chicken eggs were sampled.

The results of analyses for PCDD/Fs, PCBs, HCB, PeCB, HCBD, PBDD/Fs, SCCPs, some OCPs, PBDEs, HBCD, TBBPA and nBFRs are summarized in Table 6.

### 4.2 E-waste scrap yard

Agbogbloshie scrap yard was the focus of numerous previous studies regarding its contamination by POPs and heavy metals. Most of them focused either on soil or human tissue contamination by different POPs, and various groups of these chemicals, polyhalogenated dibenzo-p-dioxins and dibenzofurans in particular. Our study is focused on free-range chicken eggs as part of the diet for people living on the scrap yard as well as part of local food chain. This is first time to our knowledge where POPs were analyzed in free-range chicken eggs from hens foraging in the area of Agbogbloshie scrap yard. We also analyzed one pooled soil sample from the closest area to the location where the chicken eggs were sampled.

The results of analyses for PCDD/Fs, PCBs, HCB, PeCB, HCBD, PBDD/Fs, SCCPs, some OCPs, PBDEs, HBCD, TBBPA and nBFRs are summarized in Table 6.

Table 6: Summarized results of analyses for various POPs in two egg samples from Agbogbloshie scrap yard (analyzed for this study) compared with three egg samples from a small artisanal e-waste and other wastes recycling site in Samuth Sakhon, Thailand; source [108, 118].

Locality	Accra - Agbo	ogbloshie	Samut Sakh	on (Thailand)	)
PCDD/Fs (pg TEQ g <sup>-1</sup> fat)	661	4524	84	13	12
dl-PCBs (pg TEQ g <sup>-1</sup> fat)	195	399	12	0.001	1.5
Total PCDD/F + dl-PCBs (pg TEQ g <sup>-1</sup> fat)	856	4924	96	13	13.5
PBDD/Fs (pg TEQ g <sup>-1</sup> )	300	62	16	NA	NA
HCB (ng g <sup>-1</sup> fat)	25	91	4.2	1.4	<lod< td=""></lod<>
PeCB	22	181	NA	0.35	NA
HCBD	< 0.2	0.15	NA	NA	NA
7 iPCB	286	618	13	1.1	<lod< td=""></lod<>
6 iPCB (EU)	168	452	11	1.1	<lod< td=""></lod<>
PCNs*	< 1.4	4.3	NA	NA	NA
SCCPs	2067	311	NA	NA	NA
sum HCH	< 0.6	0.10	0.31	NA	<lod< td=""></lod<>
sum DDT	9.7	0.79	2.9	NA	<lod< td=""></lod<>

<sup>&</sup>lt;sup>7</sup> BTBPE stands for 1,2-bis(2,4,6-tribromo-fenoxy)ethane. It is one from the family of novel brominated flame retardants used e.g. in electronics where replaced PBDEs. Its accumulation in the eggs highlights the need of more detailed screening of new retardants used as alternatives replacing PBDEs for their potential properties similar to POPs, otherwise we will continue to repeat the same mistake and will use new POPs to replace other POPs which is not intention of the Stockholm Convention.

PBDEs	1258	765	3.1	NA	NA
sum HBCD	1961	9.8	NA	NA	NA
ВТВРЕ	38	20	<0.5	NA	NA
DBDPE	<3.3	35	NA	NA	NA
НВВ	1.1	1.6	<0.1	NA	NA
PBT	<0.2	0.15	<0.1	NA	NA
ТВВРА	<4.2	149	NA	NA	NA

dw = dry weight

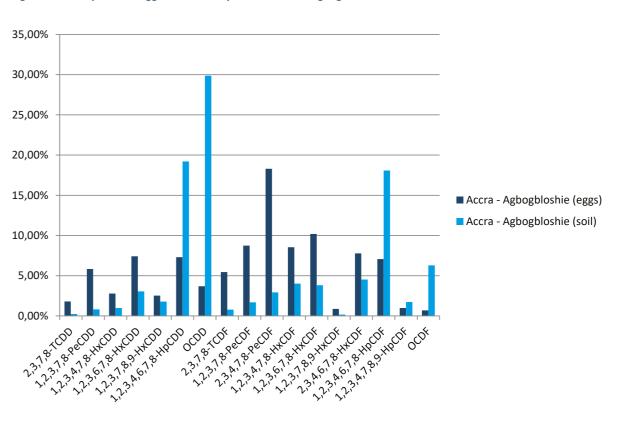
The most recent study by Tue et al. [99] summarized potential health effects of high contamination of soils by PCDD/Fs, PBDD/Fs and other dioxin-like chemicals: "The elevated TEQ concentrations in surface soils of the dismantling area suggest high risk of exposure to dioxin-like chemicals and potential dioxin-related health effects for e-waste dismantling workers and other people frequenting the area, especially for children who may accidentally ingest more soils (36) and are subjected to higher per-weight exposure doses than adults. Considering the dioxin exposure threshold of 1 pg TEQ/kg/day [119] and an average soil ingestion rate of 30 mg/day for 12 year old children[120] with an average body weight of 40 kg, TEQ concentrations in soils exceeding 1300 pg g<sup>-1</sup> can be considered as posing risk for children exposure. Chronic exposure to PCDD/Fs resulted in high concentrations in blood of workers in Agbogbloshie [37]. Meanwhile, despite the high contamination levels of PBDFs in Agbogbloshie, the contribution of these brominated dioxins to the total dioxin exposure risk is still unclear considering the potentially lower bioaccessibility and shorter half-life [121, 122]. Nevertheless, as significant dioxin-like activities were detected in the brominated dioxin fraction (rather than in the chlorinated dioxin fraction) of several randomly selected serum samples from Agbogbloshie e-waste workers, further studies on potential health effects of brominated dioxins in e-waste workers are warranted" [99].



PBDD/Fs measured in soil in this study are much lower than those found in samples from 2013 by Tue et al. [99]. However, PCDD/Fs in our sample are comparably high to the maximum level of 5.2 ng TEQ  $\rm g^{-1}$  dw found by Tue et al. [99]. This study did not focus on mapping different soil levels, but on free-range chicken eggs from different locations in Ghana and Cameroon, so we definitely cannot say that our soil sample is representative for Agbogbloshie. We wanted to obtain a better characterisation of the closest soil environment for chickens laying eggs for the household which provided us with a sample. Tue et al. [99] also highlighted "The concentrations of PCDD/Fs, PBDD/Fs and PBDEs in surface soil samples from the Agbogbloshie e-waste site varied widely, ranging over one to two orders of magnitude even within the same area (Table 1). The contamination levels of PCDD/Fs and PBDD/Fs were not significantly different between the open burning areas and the dismantling area (Wilcoxon's rank sum test, p > 0.05). However, the highest concentrations of total PCDD/Fs were found in the open burning areas (1.3–380, median 33 ng/g as opposed to 5.6–230, median 16 ng/g), whereas the highest concentrations of total PBDD/Fs were found in the dismantling area ...".

The dioxin congener pattern in the egg samples is different than that in the mixed soil sample. It appears that the hens, from which we obtained the eggs, forage in a much broader area (see Figure 7). In addition, the balance between PBDD/Fs and PCDD/Fs, expressed in WHO-TEQs, is quite different in the pooled eggs (1:99) compared to the composite soil sample (3:7).

Figure 7: Dioxin profile of eggs and soil sample from Accra - Agbogbloshie.



The POPs levels in free-range chicken eggs sampled in this study indicate that production of food at the scrap yard in Agbogbloshie might have serious health implications. Based on the results of analyses demonstrated in Table 6 and discussed in chapter 4.1, we can say that: An adult eating just one egg from Agbogbloshie would exceed the tolerable daily intake (TDI) as set by the European Food Safety Authority (EFSA) in 2018 for dioxins and dioxin-like PCBs by 220-fold [123]. One egg from Agbogbloshie would exceed the TDI suggested by the World Health Organization (WHO) by thirty-fold [124]. The typical daily egg consumption per person in Ghana is less than one egg a day, but even eating 2.5 grams of egg a day would exceed the EFSA TDI by more than 15-fold.

<sup>\*</sup>Seven PCN congeners were measured: PCN 52, 56, 66, 70, 73, 74 and 75.

## 4.3 Dump sites

CREPD took samples of free-range chicken eggs at two dump sites in Yaoundé, the capital city of Cameroon. These samples were analyzed for unintentionally produced POPs: HCB, PeCB and HCBD. We were not able to analyse them for dioxins due to the financial constrains of our project, however there are older egg samples from two dumpsites in African countries from previous studies, and another sample from Yaounde which was taken in the surroundings of a small medical waste incinerator combined with an open firing pit for wastes. So, the sample taken near the medical waste incinerator can be considered to reflect potential levels from open burning of wastes in Yaoundé. The total levels of PCDD/Fs and dl-PCBs in the medical waste incinerator sample was 11 pg TEQ g<sup>-1</sup> fat, which is lower in comparison with samples from two sampled dumpsites in Nairobi, (Dandora) and Dakar (Mbeubeuss) in 2005 where levels of 27 and 40 pg TEQ g<sup>-1</sup> fat respectively were found in free-range chicken eggs [14, 125, 126].

Table 7: Summarized results of analyses for various POPs in two samples from Yaoundé dumpsites (analyzed for this study) compared with two samples from earlier studies of chicken egg samples from two other African locations in Nairobi, Kenya and Dakar, Senegal and a more recent sample from a landfill site in Praeksa, Thailand. Sources [125-127].

Locality	Yaoundé-TCK Quart.	Yaoundé- Etetak Quart.	Nairobi – Dandora (2005)	Dakar – Mbeubeuss (2005)	Praeksa (Thailand)
Sample	YA-1	YA-3	Dandora	Malika	PKS-EGG
Units	(ng g <sup>-1</sup> fat)	(ng g <sup>-1</sup> fat)	(ng g <sup>-1</sup> fat)	(ng g <sup>-1</sup> fat)	(ng g <sup>-1</sup> fat)
PCDD/Fs (pg TEQ g <sup>-1</sup> fat)	NA	NA	20	36	6.5
dl-PCBs (pg TEQ g <sup>-1</sup> fat)	NA	NA	7.2	3.1	2.2
Total PCDD/F + dIPCBs (pg					
TEQ g <sup>-1</sup> fat)	NA	NA	27	40	8.7
HCB (ng g <sup>-1</sup> fat)	1.5	7.1	4.4	1.7	NA
PeCB	0.56	4.7	NA	NA	NA
HCBD	< 0.1	< 0.1	NA	NA	NA
7 iPCB	28	36	31	29	NA
6 iPCB (EU)	27	34	25	28	NA
sum HCH	4.5	7.6	2.5	6.0	NA
sum DDT	39	36	83	23	NA
PBDEs	0.50	2.8	29	NA	NA
sum HBCD	124	25	160	NA	NA

Indicator PCBs, HBCD and DDT seem to be major POPs contaminants in samples of eggs from the two dumpsites in Yaoundé. Similar levels of PCBs in eggs were observed at all the African dumpsites. Eggs near all four African dumpsites contained DDT and three of the sites contained HBCD and PBDEs.

Significant contamination by HBCD can be found in free-range chicken egg samples from the vicinity of dumpsites in general as observed in much higher levels at the Agbogbloshie scrap yard. This is most likely due to polystyrene or upholstery waste present at the dumpsites. Similar levels of HBCD to those found in eggs from dumpsites in Yaoundé were found in eggs from some places in Kazakhstan from sites near rather uncontrolled landfills or dumpsites (e.g. egg samples from Baskuduk or Balkhash – Rembaza [112]). Levels of PBDEs are lower in comparison with HBCD and also much lower than that observed at the scrap yard in Agbogbloshie where e-waste is major source of contamination by PBDEs. E-waste is not present in high quantities at general dumpsites like those sampled in Yaoundé.

#### 4.4 Waste incinerators

The surroundings of three small medical waste incinerators were sampled for this report. We took samples of free-range chicken eggs in close vicinity of all three of them and we also sampled waste incineration residues from two Ghana waste incinerators, ash in one case and soot in another hospital waste incinerator. All were pooled samples as described in chapter 2.

The results of analyses for PCDD/Fs, PCBs, HCB, PeCB, HCBD, PBDD/Fs, SCCPs, some OCPs (DDT and HCHs), PBDEs, and HBCD in samples from three waste incineration sites in Yaoundé, Accra and Kumasi are summarized in Table 8.

Table 8: Summarized results of analyses for various POPs in samples from medical waste incinerators areas and surrounding in Youndé. Accra and Kumasi in eggs, ash and soot.

Locality	Yaoundé- hospital	Accra - hospital	Kumasi - hospital	Accra - hospital	Kumasi - hospital
Sample	YA-2	KBI-E	KU-E	KBI-A-1	KU-A-1
Matrix	Eggs	Eggs	Eggs	Ash	Soot
Units	(ng g <sup>-1</sup> fat)	(ng g <sup>-1</sup> fat)	(ng g <sup>-1</sup> fat)	(ng g <sup>-1</sup> dw)	(ng g <sup>-1</sup> dw)
PCDD/Fs (pg TEQ g <sup>-1</sup> )	4.6	49	1.7	551	2315
dl-PCBs (pg TEQ g <sup>-1</sup> )	6.8	14	0.86	28	99.5
Total PCDD/F + dl-PCBs (pg TEQ g-1)	11.4	63.1	2.60	579	2414
НСВ	1.43	3.63	0.76	4.09	NA
PeCB	0.35	2.88	< 0.2	3.49	0.774
HCBD	< 0.1	< 0.2	< 0.2	< 0.02	< 0.02
7 iPCB	32.0	7.8	< 1.4	0.29	NA
6 iPCB (EU)	30.4	7.8	< 1.2	0.29	NA
sum HCH	2.50	< 0.6	< 0.6	0.10	NA
sum DDT	22.23	79.43	0.82	0.79	NA
PBDEs	2.31	NA	NA	NA	NA
sum HBCD	379.41	NA	NA	NA	NA

Figure 8: Dioxin congeners patterns in eggs and ash from Accra – hospital site.

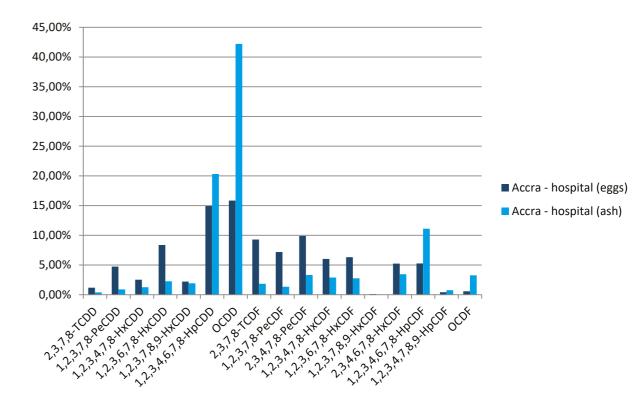




Table 9 summarizes results of analyses for dioxins in free-range chicken eggs from places influenced by ash from waste incinerators or other wastes containing PCDD/Fs in different locations around the world. The table is constructed to show the potential pathway of pollution by dioxins from wastes to soil (as career) and then to free-range chicken eggs as the receptor. They are also compared with background levels (reference) of PCDD/Fs in eggs from countries, mostly levels in eggs from larger farms where chickens are kept inside and do not have access to dioxin contamination sources.

Table 9. Summary of levels of PCDD/Fs (in TEQs and/or BEQs) observed at different sites influenced by fly ash and other waste contaminated by PCDD/Fs described in this study or in the literature.

	Year(s) of sampling	Fly ashes (waste)	Soil/sediment direct impact	Soil/sed. reference	Eggs	Eggs – reference <sup>1)</sup>		
Units		pg TEQ g <sup>-1</sup> dw				pg TEQ g⁻¹ fat		
Thailand (WI Phuket)	2010 - 2011	3,200 - 8,000	2,700**	na	6.1*	0.08[106]		
China (WI Wuhan)	2014 - 2015	779	Na	na	12.2	0.2 [112]		
UK (Bishops Cleeve)	2010 - 2011	2,500	6.5 – 11*	0.05 - 1.2	1.8; 21; 55*	0.2[5]		
UK (Newcastle) [5, 113]	2000	20 - 9,500	7 – 292	na	0.4 – 56	0.2 [5]		
Peru (Zapallal) [114]	2010	50 - 12,000	5 – 11	0.05 - 1.2	3.4 - 4.4	0.12 [114]		
Taiwan (eggs event) [115]	2005	Na	na	na	32.6	0.274 [116]		
Poland (henhouse) [80]	2015	3,922	16 – 47	0.1 - 0.8	12.5 - 29.3	0.44 [80]		
Ghana (Accra, hospital)	2018	551	na	na	49	0.39		

Notes: \*BEQs (total dioxin-like toxicity), \*\* sediment, Na – not available

In previous studies, processing/disposal of waste containing PCDD/Fs between 20 and 12,000 pg TEQ  $g^{-1}$  led to contamination of the food chain (eggs or poultry meat) up to levels >20-times higher than the suggested EU limit for PCDD/Fs in food (2.5 pg TEQ  $g^{-1}$  fat)19. Levels from reference sites (background levels) in freerange chicken eggs were exceeded up to 280-fold.

In Accra, hens have access to a site with stored ash, but also forage in a larger area which is probably not contaminated with such high levels of dioxins. However, like other studies, the results show that incinerator fly ash with a level of dioxins (551 pg TEQ  $\rm g^{-1}$  dw) well below the current internationally set provisional limit value for PCDD/Fs in wastes (15,000 pg TEQ  $\rm g^{-1}$ ) leads to contamination of eggs at a level (49 pg TEQ  $\rm g^{-1}$  fat) exceeding EU limit by almost 20-fold.

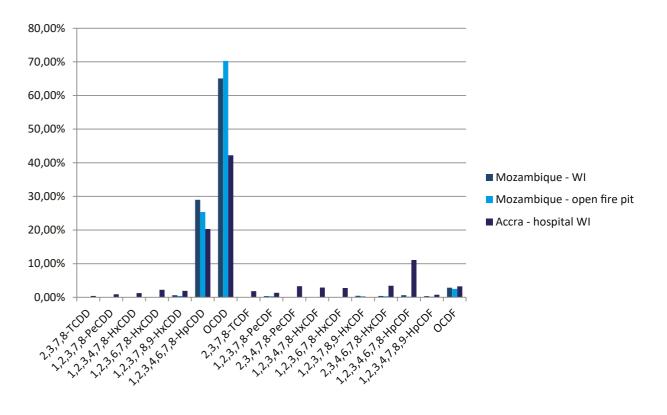
A Swedish EPA study demonstrated that PCDD/Fs levels of 30 pg TEQ g¹ fat in an egg will be exceeded at soil concentrations of approximately 4 to 75 ng TEQ kg¹ dw. Therefore, the European maximum level of 2.5 pg TEQ g¹ PCDD/F in fat [132] can be exceeded at levels that are ten times lower (i.e. 0.4 and 7 ng TEQ kg¹ dw). Based on the upper level of the range given in the Swedish EPA study and examples of a scenario with contaminated wood waste [130], it can be concluded that application of fly ash and other wastes containing levels of dioxin over 0.05 ppb in land-based applications can lead to unacceptable contamination of the local food chain. In some other studies, even lower levels of dioxins in soils led to contamination of free-range chicken eggs exceeding the EU standard for food [14, 15]. Free-range eggs can be impacted at critical levels, and in some cases revealing a more than 20-fold exceedance of current EU limits. Locally produced food is of great importance in developing countries and rural locations in developed countries therefore this exposure scenario is of particular concern.

Free-range chicken eggs from the Accra – hospital site also exceeded the EU suggested limit for PCDD/Fs for PCDD/Fs + dl-PCBs by more than almost 13-fold. This is most likely a consequence of access to untreated waste incineration residues. The waste incinerator has not been in operation for several years but discarded ash was found to still be contaminated with high levels of PCDD/Fs + dl-PCBs (see photo in Figure 9).



The level of PCDD/Fs in waste incineration ash from the Accra hospital was 551 pg WHO-TEQ g<sup>-1</sup>dw. This level is higher than what was measured in ash from a medical waste incinerator in Mozambique (346 pg WHO-TEQ g<sup>-1</sup> dw) for which the dioxin profile is presented in the graph at Figure 10 and compared with the profile from Accra hospital. Also, data for ash from an open fire pit in the Mozambique hospital were included. The dioxin congener pattern in ash from Accra is somewhat different from those observed in ashes from Mozambique.

Figure 10: Comparison of PCDD/Fs congener patterns in samples of ash from Mozambique and hospital waste incinerator ash from Accra, Ghana. In order to get equal comparison, the levels for each congener are in relative percentage from total PCDD/Fs value (in absolute real amount). Source of information about PCDD/Fs in ashes from Mozambique is Peter I. K. Mochungong's thesis [133].



In comparison with observed levels of PCDD/Fs in ashes from similar waste incinerators in Pakistan [134] or Thailand [135] the dioxin content in ash from Accra – hospital is 2 – 4 times lower. However, it can be caused by the age of ash at this site as some PCDD/Fs could be already diluted and released to the environment.

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## 5. Conclusions

Eggs sampled at the Agbogbloshie scrap yard in Ghana contained the highest level of brominated dioxins ever measured in eggs and one of the highest ever measured levels of the flame retardant chemical, HBCD. These eggs also contained the second highest level of chlorinated dioxins ever measured in eggs. An adult eating just one egg from a free-range chicken foraging in Agbogbloshie area would exceed the European Food Safety Authority (EFSA) tolerable daily intake (TDI) for chlorinated dioxins by 220-fold. Indicator PCBs in these eggs were four-fold higher than the EU standard and dioxins and dioxin-like PCBs were 171-fold higher than the standard. These eggs also contained very high levels of SCCPs and PBDEs and relatively high levels of other POPs such as PeCB and HCB. These findings raise further concerns about e-waste 'recycling' at Agbogbloshie and add further information to already published results of high levels of POPs measured in soil, sediments, water and human tissues.

Eggs near the medical waste incinerator in Accra, Ghana exceeded the EU dioxin limit by 13-fold and eggs sampled near the facility in Yaoundé exceeded the limit by more than two-fold. PCBs did not exceed limits, but significant levels were also found. High levels of HBCD were also found in eggs from the vicinity of the Yaoundé waste incinerator and one of the dumpsites

## 6. Discussion & Policy Implications:

#### 6.1 POPs waste and e-waste

There is a clear link between current global policy that allows uncontrolled movement of e-waste and the health and environmental crisis in areas where dumping occurs, such as Agbogbloshie. A recent report by Basel Action Network confirmed that Ghana is on the list of destinations for old, used electronic devices from Europe—devices that contain high levels of PBDEs in the plastic casings and wire insulation [8].

Recent research has demonstrated that brominated dioxins show up in products made from recycled e-waste plastics. A recent study conducted by IPEN, Arnika and HEAL [102], in cooperation with NGOs from different countries, found worrying levels of brominated dioxins in toys made of recycled plastics that originated in e-waste. Brominated dioxins, which are similarly toxic to chlorinated dioxins—one of the Stockholm Convention's original "dirty-dozen"—occur with some brominated flame retardants and are also formed when brominated flame retardants are burned. The findings of high levels of brominated dioxins in eggs in Agbogbloshie, the destination of vast amounts of used electronic products, is especially concerning, as it illustrates a pathway for some of the most dangerous chemicals into food sources.

Dioxins and dioxin-like PCBs have previously been found in high levels in egg samples from Egypt, Senegal, Kenya, and Tanzania connected to pollution hot spots in Africa, mostly related to waste [14]. This underscores the findings of this current study and highlights the need to improve waste management and to control dioxin flow in wastes.

To prevent further contamination of hot spots such as Agbogbloshie or the creation of new ones in Africa, governments must change international rules to:

- 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 maximum.
- » Transfer cleaner technologies for destruction of POPs and help to introduce environmentally sound management of electronic waste in developing countries.

- » Repair loopholes in e-waste technical guidelines under the Basel Convention.
- » List brominated dioxins (PBDD/Fs) under the Stockholm Convention.
- » Continuous monitoring by governments and taking appropriate measures to deal with releases e.g. through legislation and setting national standards.

Improvements to waste management that include technical assistance are needed. Such improvements will help establish sustainable sorting and recycling of waste and not move pollution back into the environment where it harms workers and communities around waste sites. Non-combustion technologies used for medical waste treatment, accompanied with sorting of waste, would prevent contamination of chicken eggs at hospitals such as the sites examined in this study. Additionally, the introduction of stricter limits for dioxins in wastes allowed to be used on surfaces without pre-treatment (at level 50 pg TEQ g<sup>-1</sup>= 0.05 ppb) will prevent cases such as those documented in places where waste incineration residues are not handled properly.

## **6.2 Healthcare waste management**

The data presented in this study verify long-standing concerns over the use of small medical waste incinerators in developing countries. The Stockholm Convention Guidelines on Best Available Techniques and Guidance on Best Environmental Practices (BAT/BEP) note concerns over small hospital incinerators, stating that, "Due to the poor design, operation, equipment and monitoring of many existing small hospital incinerators these installations cannot be regarded as employing best available techniques" [2]. None of the medical waste incinerators in this study could be considered to employ BAT/BEP due to their design, operation, lack of pollution control, and lack of waste management for the ash. The high POPs levels in eggs reveal the consequences of inadequate healthcare waste management. A hospital facility designed for healing should not pollute the food chain or cause adverse impacts on human health and the environment.

The high levels of POPs near medical waste incinerators are consistent with observations in other developing countries. A survey of small healthcare facilities in Tanzania showed only 30 – 40% of the incinerators were in good operating conditions and half of them had missing chimneys, ash pits, and other problems [136]. In Iran, 6 of 9 private hospitals that had incinerators had operational problems.<sup>8</sup> A study in Bangladesh of occupational health of waste workers did not find a single properly operating medical waste incinerator.<sup>9</sup>

Dioxin emissions from medical waste incineration have also raised concerns in developed countries. In the US EPA reassessment of dioxin sources in the 1990s, medical waste incinerators in the US were identified as one of the largest dioxin sources in the country [1]. Medical waste incineration is a major dioxin source, primarily due to 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 Association which, "Urges all health care facilities to explore ways to reduce or eliminate their use of PVC plastics" [137].

Instead of trying to improve dioxin-producing technologies such as small medical waste incinerators, a strategy that prevents dioxin formation is more cost effective and consistent with Stockholm Convention objectives. 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 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 use of source reduction, segregation, recycling, training, and use of autoclaves and other non-combustion methods [2]. The Guidelines note that

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<sup>8</sup> Askarian M, Vakili M, Kabir G (2004) Results of a hospital waste survey in private hospitals in Fars province, Iran. Waste Management 24:347-352

<sup>&</sup>lt;sup>9</sup> Patwary MA, O'Hare WT, Sarker MH (2011) 'Occupational accident: An example of fatalistic beliefs among medical waste workers in Bangladesh', Safety Science, 50: 76-82

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 [4, 138, 139].

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) [140], non-combustion waste treatment pilot project (Tanzania)[6], and non-incineration healthcare waste management and mercury-free medical devices (Ghana, Madagascar, Tanzania, Zimbabwe) among others.

### 6.3 Environmental, food and human monitoring

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

- We international standards for monitoring of dioxins in food (e.g. eggs) and mandatory number of analysis per year per critical food items.
- >> Use international accepted standards (such as as EC/644/2017); [141] for the analysis of dioxins/PCBs in food/feed using high-throughput screening tests (such as DR CALUX) as well as chemical confirmative analysis.
- » Use screening tests (such as DR CALUX) which allows an easier, cost-efficient and high capacity testing of not only polychlorinated dioxins, but also here for polybrominated dioxins which are high relevant to e-waste.
- » Recommend using high-through-put and cost efficient screening tests (such as DR CALUX® method) for blood and human milk for polyhalogenated (chlorinated and brominated) dioxins/furans for wildlife and humans [142, 143].<sup>10</sup>
- » It is immediately necessary to evaluate also the most toxic mode of actions such as the well described effets of endocrine disrupting chemicals, "hormone-like" of e.g. PBDEs (female hormone estrogen-like, inhibition male hormone-like), TBBPA (thyroid transport competitor) and therefor related risks of such e-waste dismantling sites for wildlife and humans.

## 7. Aknowledgements

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## 8. Abbreviations

BDS - BioDetection Systems (laboratory in Netherlands)

BEQ – bioanalytical equivalent

CALUX - chemically activated luciferase gene expression

BTBPE - 1,2-bis(2,4,6-tribromo-fenoxy) 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

EDCs – endocrine disrupting chemicals

EPA - Environment Protection Agency

EU - European Union

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 POPs 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

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

OBIND - octabromotrimethylfenylindane

<sup>&</sup>lt;sup>10</sup> It is important to notice that the DR CALUX method also includes already the brominated dioxins and biphenyls (PBDD/Fs and PBBs) without any further costs, while the chemical analyses needs here an additional expensive analysis. Globally, at the moment only a handfull chemical laboratories are available to perform this additional chemical analysis in routine for brominated dioxins and biphenyls (PBDD/Fs and PBBs), while many laboratories already perform the DR CALUX® method. Such easy, low cost and high-capacity analysis tools are urgently needed in such cases with a wide-spread contamination of brominated dioxins/biphenyls (PBDD/Fs/PBBs) in e-waste, products from recycled e-waste plastic, soil, feed/food and human biomonitoring (blood, mother milk).

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

POPs – persistent organic pollutants

SC – Stockholm Convention on Persistent Organic Pollutants

SCCPs – short chain chlorinated paraffins

SOP - standard operating procedures

TBBPA - tetrabromobisphenol A

TEF – toxic equivalency factor(-s)

TEQ – toxic equivalent

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

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