Dioxins – The old dirty (dozen) guys are still with us

Dioxins are a group of chemicals incorporating 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) [1]. Technical mixtures of PCBs are characterized by 6, sometimes 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 expert panel in 2005 [2]. Limits for dioxins in food and/or their daily intake are mostly set as a common level for both PCDD/Fs plus dl-PCBs.

Polychlorinated dioxins and furans, as well as PCBs, as unintentionally produced POPs (UPOPs) were listed under Annex C to the Stockholm Convention as part of the so-called 'dirty dozen' POPs at the very beginning of the Convention. In order to assess how well the convention has worked, these original POPs should also be monitored in the environment and the results of such monitoring are part of the effectiveness evaluation of the Stockholm Convention and regional reports published for each UN region [3-7].

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 [8, 9]. 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. Diet (particularly food derived from animals) is the major source of exposure fo

Dioxins in free-range eggs

IPEN, an international network of more than 500 public interest NGOs from more than 100 countries, looked at levels of unintentionally produced POPs, including dioxins and dl-PCBs, in free-range chicken eggs near potential sources of pollution by these chemicals at 20 sites in 17 countries in 2005. Results were published in The Egg Report at the First Conference of the Parties to Stockholm Convention. The lowest levels of dioxins in these egg samples were more than two times higher than the background levels of dioxins in eggs from foraging chickens that have been reported in several studies in Europe and North America. Seventy percent of the samples exceeded the European Union (EU) limit for dioxins in eggs. Samples collected near a metallurgical facility in Egypt, a thermal power plant in Bulgaria, and chlor- alkali facilities in Russia yielded dioxin levels that range from 44 – 126 pg/g (WHO-TEQ) of fat and from 63 – 138 pg/g (WHO-TEQ) of fat for PCDD/Fs and total WHO-TEQ (PCDD/Fs + dl-PCBs) respectively [11].

Since 2010 Arnika[1] and IPEN, in cooperation with many public interest NGOs, have undertaken new and broader sampling of free-range chicken eggs in several countries from three continents. The sampling focused on different industrial sites and sites influenced by waste management or contaminated sites. Their results were published in several studies focused on Armenia [12], Czechia, Kazakhstan [13], China [14], Thailand [15], Bosnia and Herzegovina, Montenegro, Serbia [16], Ukraine [17], Cameroon, Ghana [18] and sites specifically affected by waste incineration residues in Asia and Europe [18, 19]. In addition, we have conducted egg sampling and analyses for POPs in Belarus, UK and Indonesia. Those results were published in different reports focused on other countries or specific topics. In all countries we found levels of PCDD/Fs and/or total WHO-TEQ levels of PCDD/Fs + dl-PCBs exceeding EU standards for eggs as food by up to 264-times and 171-times respectively. In addition to these results we reviewed literature about the results of chemical and/or bioassay analyses of free-range poultry eggs available for samples since 2005.

Maximum levels of PCDD/Fs and total WHO-TEQ levels (PCDD/Fs + dl-PCBs) observed at different hot spots from reviewed literature (including those in above mentioned Arnika/IPEN studies) are summarized and sorted by UN regions in Table 1 and Table 2 respectively.

Table 1: PCDD/Fs in free range chicken eggs samples from different regions – maximum levels measured in 2005 – 2018 (in pg WHO-TEQ/g of fat)Table 2: PCDD/Fs + dl-PCBs in free range chicken eggs samples from different regions – maximum levels measured in 2005 – 2018 (in pg WHO-TEQ/g of fat if not specified otherwise)

Country	Region	Date/ year	Locality	Measured level of PCDD/Fs	Exceed-ance of EU stan-dard	Potential source(-s)	Source of in-forma-tion
Ghana - Agbogbloshie (2018)	Africa	2018	Agbogbloshie	661	264	e-waste site	[18]
Ghana - Accra - hospital (2018)	Africa	2018	Accra - hospital WI	49	20	waste incineration	[18]
Cameroon - Yaounde (2018)	Africa	2018	Yaounde - hospital WI	4.6	2	waste incineration; open burning	[18]
Egypt (2010-2014)	Africa	2010-2014	not specified	4.5	2	metallurgical industry	[20]
Vietnam - Bien Hoa (2011)	Asia	2011	Bien Hoa	248	99	contaminated site (Agent Orange)	[21]
Thailand - Samut Sakhon (2015)	Asia	2015	Samut Sakhon	84	34	e-waste site and open burning	[22]
Indonesia - Kendalsari (2018)	Asia	2018	Kendalsari	49	20	secondary aluminium smelter	[23]
Taiwan - Chang-Hua County (2004-2005)*	Asia	2004-2005	Chang-Hua County	15.0	6	ash from metallurgical plant	[24]
China - Wuhan (2014)	Asia	2014	Wuhan	12.2	5	municipal waste incinerator	[25]
Turkey - Dilovasi (2008)	Asia	2008	Dilovasi, Kocaeli region	10.9	4	metallurgical industry	[26]
Kazakhstan - Balkhash (2013)	Asia	2013	Balkhash - west	9.8	4	car wrecks; metallurgical industry	[18]
Kazakhstan - Shabanbai BI (2014)	Asia	2014	Shabanbai Bi	9.3	4	PCBs oil contamination	[18]
Poland (2011)	CEE	2011	Not specified	29	12	PCP treated wood	[27]
Ukraine, Krivyi Ryh (2018)	CEE	2018	Krivyi Ryh	23	9	metallurgical industry	[17]
Czechia - Pitárne (2017)	CEE	2017	Pitarne	15.4	6	PVC recycling plant	[28]
Serbia - Grabovac (2015)	CEE	2015	Grabovac	11.1	4	chemical contamination	[16]
Poland - Malopolska (2017)	CEE	2017	Malopolska region	9.5	4	air pollution (general)	[29]
Armenia - Alaverdi (2018)	CEE	2018	Alaverdi	7.5	3	copper smelter	[12]
Bosnia and Herzegovina - Zenica (2015)	CEE	2015	Zenica	5.6	2	metallurgical industry	[16]
Czechia - Lhenice (2015)	CEE	2015	Lhenice	5.3	2	PCB contaminated site	[30]
Belarus - Gatovo (2014)	CEE	2014	Gatovo	4.3	2	car shredder	[31]
Portugal (2008)	Europe	2008	Not specified	61	25	PCP treated wood	[32]
Italy - Piedmont (2012)	Europe	2012-2013	Piedmont region	38	15	metallurgical industry	[33]
Belgium (2007)	Europe	2007	Not specified	20	8	not specified	[34]
Germany - Teningen (2014)	Europe	2014	Teningen	11.4	5	former PCB capacitors production (contaminated site)	[35]
Netherlands - Friesland (2014)	Europe	2014	Eastern part of Friesland	9.6	4	not clear	[36, 37]
Netherlands - Rijnmond (2014)	Europe	2014	Rijnmond and Rotterdam	9.6	4	industrialized area of Netherlands	[37]
Italy - Caserta	Europe	2014-2015	Caserta, Campania	6.2	2	open burning of waste	[38]
Netherlands - Harlingen (2013)	Europe	2013	Midlum, Harlingen	4.8	2	municipal waste incinerator	[39]
Uruguay, Minas	GRULAC	2009	Minas	23	9	PCBs burning cement kiln	[40, 41]
Brazil - Vespasiano (2014)	GRULAC	2014	Vespasiano, Bello Horizonte	7.4	3	fire in cement kiln (used tires burnt)	[42]
Peru - Zapallal (2010)	GRULAC	2010	Zapallal	4.4	2	ash from metallurgical workshops	[43]
Canada (2005-2006)	North America	2005-2006	not specified	10.6	4	PCP treated wood	[44]

1 Arnika is IPEN participating organization and coordinated previous sampling in 2004 – 2005.

Table 2: PCDD/Fs + dl-PCBs in free range chicken eggs samples from different regions – maximum levels measured in 2005 – 2018 (in pg WHO-TEQ/g of fat if not specified otherwise)

Country	Region	Date/ year	Locality	Levels of PCDD/ Fs + dl-PCBs	Exceed-ance of EU stan-dard	Potential source(-s)	Source of in-forma-tion
Ghana - Agbogbloshie (2018)	Africa	2018	Agbogbloshie	856	171	e-waste site	[18]
Ghana - Accra - hospital (2018)	Africa	2018	Accra - hospital WI	63	13	waste incineration	[18]
Tanzania - Arusha (2012)	Africa	2012	Arusha	20*	4	open burning of waste	[45]
Cameroon - Yaounde (2018)	Africa	2018	Yaounde - hospital WI	11.4	2	waste incineration, open burning	[18]
South Africa - Vanderbijlpark (2008-2009)	Africa	2008-2009	Vanderbijlpark	6.4	1	metallurgical industry	[46]
Vietnam - Bien Hoa (2011)	Asia	2011	Bien Hoa	249	50	contaminated site (Agent Orange)	[21]
Kazakhstan - Shabanbai BI (2015)	Asia	2015	Shabanbai Bi	155	31	PCBs oil contamination	[18]
Kazakhstan - Balkhash (2013)	Asia	2013	Balkhash - north	101*	20	metallurgical industry	[25]
Thailand - Samut Sakhon (2015)	Asia	2015	Samut Sakhon	96	19	e-waste site and open burning	[22]
Indonesia - Kendalsari (2018)	Asia	2018	Kendalsari	85	17	secondary aluminium smelter	[23]
China - Zheijang (2006-2015)	Asia	2006-2015	Zhejiang	37	7	municipal waste incineration, e-waste site	[47]
China - Beihai (2014)	Asia	2014	Beihai	37*	7	metallurgical industry	[14]
China - Wuhan (2014)	Asia	2014	Wuhan	16.0	3	municipal waste incinerator	[25]
Kazakhstan - Shetpe (2016)	Asia	2016	Shetpe	6.4	1	car wrecks, waste, cement kiln	[18]
Poland - Silesia (2018)	CEE	2018	Silesia	43	9	co-burning of PVC waste in household heating	[48]
Armenia - Nubarashen (2010)	CEE	2010	Nubarashen	37*	7	contaminated site	[49]
Ukraine, Krivyi Ryh (2018)	CEE	2018	Krivyi Ryh	36	7	metallurgical industry	[17]
Czechia - Pitárne (2017)	CEE	2017	Pitarne	32	6	PVC recycling plant	[28]
Poland (2011)	CEE	2011	Not specified	30	6	PCP treated wood	[27]
Armenia - Alaverdi (2018)	CEE	2018	Alaverdi	27	5	copper smelter	[12]
Belarus - Gatovo (2014)	CEE	2014	Gatovo	15.6	3	car shreder	[31]
Serbia - Grabovac (2015)	CEE	2015	Grabovac	13.5	3	chemical contamination	[16]
Czechia - Lhenice (2015)	CEE	2015	Lhenice	9.1	2	PCB contaminated site	[30]
Bosnia and Herzegovina - Zenica (2015)	CEE	2015	Zenica	8.7	2	metallurgical industry	[16]
Italy - Piedmont (2012)	Europe	2012-2013	Piedmont region	113	23	metallurgical industry	[33]
Belgium (2007)	Europe	2007	Not specified	95	19	not specified	[34]
Italy - Lombardia (2010)	Europe	2010	Lombardia	90	18	industrialized areas of Lombardia	[50, 51]
Netherlands (2012)	Europe	2012	not specified	80	16	asbestos fiber plates roof	[52]
UK - Bishop's Cleeve (2010)	Europe	2010	Bishop's Cleeve	55*	11	waste incineration ash	[25]
Germany - Teningen (2014)	Europe	2014	Teningen	36	7	former PCB capacitors production (contaminated site)	[35]
Netherlands - Friesland (2014)	Europe	2014	Eastern part of Friesland	18.9	4	not clear	[36, 37]
Italy - Naples	Europe	2014-2015	Naples, Campania	17.2	3	open burning of waste	[38]
Netherlands - Rijnmond (2014)	Europe	2014	Rijnmond and Rotterdam	14.2	3	industrialized area of Netherlands	[37]
Italy (2013-2015)	Europe	2013-2015	not specified	12.7	3	not clear	[53]
Germany - State of Hesse (2013)	Europe	2013	State of Hesse	11.8	2	asbestos fiber plates roof	[54]
Netherlands - Harlingen (2013)	Europe	2013	Midlum, Harlingen	10.9	2	municipal waste incinerator	[39]
Germany - Eyller Berg (2014)	Europe	2014	Eyller Berg (near Kamp-Lintfort)	10.4	2	hazardous waste landfill	[35]
Italy - Caserta	Europe	2014-2015	Caserta, Campania	9.7	2	open burning of waste	[38]
Brazil - Vespasiano (2014)	GRULAC	2014	Vespasiano, Bello Horizonte	49	10	fire in cement kiln (used tires burnt)	[42]
Uruguay, Minas	GRULAC	2009	Minas	25	5	PCBs burning cement kiln	[40, 41]
Canada (2005-2006)	North America	2005-2006	not specified	12.8	3	PCP treated wood	[44]

*Mesured by DR CALUX® and expressed in pg BEQ (bioanalytical equivalent)/g of fat

If we compare the data obtained from IPEN and its members' research, much higher levels of PCDD/Fs and total WHO-TEQ were observed at studied hot spots, although the selection of the countries and the sites is not comparable to those studied in previous research in 2004/2005. The highest ever measured level of dioxins (661 pg WHO-TEQ/g of fat) in free-range chicken eggs was recently measured in eggs from an e-waste scrap yard in Agbogbloshie, Accra, Ghana. The level is six times higher than the highest previous level found in a 2005 study in Helwan, Egypt (126 pg WHO-TEQ/g of fat).

The results presented in Table 1 demonstrate that in many places dioxins are not under control due to several reasons. This provokes the question as to whether this group of chemicals is properly addressed by the Stockholm Convention and its implementation in these and other countries.

Putative or proven sources of contamination of free-range chicken eggs by dioxins or dl-PCBs, in cases demonstrated in Table 1, are often metallurgical plants, waste incineration residues and other industrial wastes. The sources also include dioxin contaminated sites as a result of previous chemical activity(-ies), including hazardous waste landfills, improper treatment of POPs wastes (e.g. PCBs burned in cement kilns), wood treated by pentachlorophenol (PCP) or other products contaminated by dioxins and dl-PCBs as well as open burning of wastes (mostly plastics). We can see that many of these activities relate to waste management or implementation of BAT/BEP for Annex C listed categories of dioxin sources. It leads us again to the question whether adequate measures are being implemented to address dioxins in all their routes to environment, including products and wastes as required by the Stockholm Convention which states that;

„In order to ensure that stockpiles consisting of or containing chemicals listed either in Annex A or Annex B and wastes, including products and articles upon becoming wastes, consisting of, containing or contaminated with a chemical listed in Annex A, B or C, are managed in a manner protective of human health and the environment, each Party shall:

(a) Develop appropriate strategies for identifying:

....

(ii) Products and articles in use and wastes consisting of, containing or contaminated with a chemical listed in Annex A, B or C; :“ [55]. Dioxin containing wastes should be destroyed or irreversibly transformed as other POPs waste. Setting the rules for POPs waste management including the limit for their definition is left with Basel Convention's experts.

Dioxins in wastes

Identification of dioxins in wastes should be a part of national inventories which should then be identified in National Implementation Plans (NIPs). Dioxin inventories are also the basis for establishing a targeted *„action plan … to identify, characterize and address the release of the chemicals listed in Annex C“* as stated in Article 5 of the Stockholm Convention [55]. If the country does not make an inventory of dioxins and other unintentionally produced POPs in waste, it **fails to address them or even recognize them.** It may be the reason why some parties support keeping the Low POPs Content level for dioxins as flexible and weak as possible.

From 86 inventories presented as basis for the evaluation of basic inventories, 9 did not include data about dioxin transfers in residues [56].

Of 139 countries, 32 did not include dioxin release data in residues, so 23% of countries which have sent their reports did not include data about wastes. From the EU only 5 countries reported on dioxin releases into residues. Among countries whose dioxin release data is missing are, for example, Austria, Denmark, Finland, France, Germany and Switzerland, the same countries with a high percentage of waste incineration and therefore residues containing dioxin. The updated inventory is also missing information about dioxins in residues in the report from Canada [57].

However there is estimation of the total global amount of PCDD/Fs produced in waste incineration fly ashes per year ranging from 7 to 10 kg WHO-TEQ/year but closer to 10 kg WHO-TEQ [58].

Dioxins: Effectiveness evaluation

Evaluation of how the Stockholm Convention addresses dioxin pollution is somewhat ambivalent. A report on the effectiveness evaluation of the Stockholm Convention released in 2017 [59] about PCDD/Fs in human tissues stated that: ***„In regions with sufficient data to evaluate changes over time, levels of legacy POPs such as polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/PCDF), polychlorinated biphenyls(PCB), and DDT/DDE, including their transformation products, have generally declined in human tissues,…"***

In another part of the report it states: ***„93. Time series data confirming trends over time in releases of unintentionally produced POPs are limited, particularly for developing country Parties, but some initial results showing decreases over time have been obtained to date."***

Addressing POPs in wastes, the Report stated that:" ***97. While some Parties have made progress in terms of developing strategies, measures and actions in the area of management of stockpiles and wastes, to identify stockpiles, products and articles in use, and wastes containing POPs, only a limited number of Parties report having such measures in place and even more limited information is available regarding the type of the measures or on the identification and disposal of wastes containing POPs"*** [59]

After checking available results for monitoring of dioxins in the Asia – Pacific region we recognized that data on PCDD/Fs and dl-PCBs in human breast milk were available for Pacific Islands, Japan and China but are missing for other countries in the region. The same applies to concentrations in ambient air [6].

Some scientists, including those who participated in implementation of the Global Monitoring Plan, are critical about even the most developed part of dioxin monitoring, their levels in ambient air: „We conclude that a decade of air monitoring data has not been sufficient for detecting general and statistically significant effects of the Stockholm Convention. Based on these lessons, we present recommendations for the future operation of existing monitoring programs and advocate for a stricter enforcement of the provisions of the Stockholm Convention, in the current absence of proof for its effectiveness," [60].

Dioxins in wastes need better control and stricter Low POPs Content level

Looking at all the obvious or hidden gaps regarding evaluation of the levels of dioxins and dl-PCBs in the environment, we are deeply concerned about the very weak measures currently in place with regards to control of dioxin flows into wastes.

A report by IPEN/Arnika/NTN followed dioxin levels in waste incineration residues and the way they are handled

worldwide. As the report demonstrated and follow up studies have proven, the current situation leads to loss of control over approximately 7 – 10 kg WHO-TEQ of dioxins every year released into waste incineration fly ashes which are not considered to be POPs waste, so they can freely move across borders and be used as e.g. construction materials, landscaping filler, paving and ashphalt mix product and landfill cover.

In order to better demonstrate the amount of toxic dioxins released every year, we can compare the amount of dioxins we lose control over in fly ashes and other air pollution control residues from waste incinerators every year, with the provisional tolerable intake as it was established by WHO [61], which is 70 pg WHO-TEQ/kg body weight/month. A volume of 7 -10 kg WHO-TEQ of dioxins is equal to tolerable intake for the entire populations of 17 – 25 planet Earths. Waste incineration residues are only one fraction of wastes containing dioxins that is out of control because of the very weak limits defined as „Low POP Content“ levels in Article 6 of the Stockholm Convention that are supposed to control these highly toxic chemicals. An expert group working on the Basel Convention's Technical Guidelines on POPs Wastes are responsible for researching and proposing these levels and Stockholm Convention parties can adopt or reject the expert group's suggested definition of POPs wastes. The Low POPs content levels, should be established using a precautionary approach to protect human health and the environment based on sound science. However, decisions about the levels are often based on political and commercial considerations as a priority.

It was very clear from the reviewed literature that current definition of Low POPs Content for PCDD/Fs at level of 15 ng TEQ/g is not strict enough to control much waste. The level is mainly decided by developed countries, and the EU in particular, who suggests to keep the LPCL weak in order to avoid much of their waste from being defined as POPs waste. These weak levels are driven and dictated by the waste incineration industry and presented by its EU based association as is obvious from the latest consultants' report prepared for the European Commission by Ramboll, a company very closely cooperating with the waste incineration industry.

Ramboll's report conclusion on setting the LPCL for dioxins is heavily influenced by the opinion of the waste incineration industry: ***"If the LPCL would be below typical fly ash contamination levels, the management option of using fly ash as filler in asphalt would be hampered. According to the recent data provided by CEWEP 11% and 3% of the fly ash would exceed an LPCL of 5 µg/kg TEQ and 10 µg/kg TEQ respectively."***

Industry influence is even more visible in another part of the report which says: ***"[CEWEP Sub. 2018b] voiced their concern that setting a LPCL close to the operating levels of the plants will increase the costs of monitoring and the no additional environmental benefit would be achieved. They stated that a LPCL close to the operating levels would hamper with the safe recycling[1] of fly ashes and thus resource efficiency,"***[63].

Was the Stockholm Convention established to prevent industry from additional costs or is *"the objective of this Convention to protect human health and the environment from persistent organic pollutants"*?

2 *It is necessary note that recycling of fly ashes as asphalt filler was not studied in real scenarios in Netherlands. Data about levels of dioxins in surrounding of the roads with fly ash as asphalt filler were never published and the BiPRO report from 2005 cited in the current study also included a statement of uncertainties "*It has to be noted that uncertainty remains with respect to superficial mechanical abrasion. Additional information will be needed in the future," 62. BiPRO, *Study to facilitate the implementation of certain waste related provisions of the Regulation on Persistent Organic Pollutants (POPs)*. 2005, European Commission: Brussels. p. 469. No additional information has been published since that time.

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