GLOBAL CONTROL OF DIOXIN IN WASTES IS INADEQUATE: A WASTE INCINERATION CASE STUDY

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Introduction

Solid waste incineration (WI)[a] is listed as one of the largest sources of dioxins (PCDD/Fs) in Annex C to the Stockholm Convention (SC) as WI releases dioxins in air emissions but also in fly ashes and other residues from the air pollution control (APC) system. In total, the residues range between 25% and 35% of the original weight of waste input[1,2]. The larger volume is bottom ash, which can reach 20 – 30% by mass of the original waste on a wet basis. APC fly ash accounts for 1–3% of all residue and total APC residues account for 2 – 5% of the waste input mass on a wet basis[3]. Fly ash and APC residues contain, in general, higher concentrations of PCDD/Fs[4,5].
The objective of the SC is *"…to protect human health and the environment from persistent organic pollutants"*[6]. It applies to releases of both intentionally produced persistent organic pollutants (POPs) as well as to unintentionally produced POPs (UPOPs) such as PCDD/Fs or dioxin-like PCBs (dl-PCBs). The Stockholm Convention's Article 6 addresses measures to reduce or eliminate releases from stockpiles and wastes. This requires a definition of POPs waste, which is called Low POP Content level (LPCL) in the Convention. The level at which the LPCL is set is a crucial decision because it defines the volumes of wastes which must be specially treated so that POPs content is *"destroyed or irreversibly transformed"*[6]. POPs waste is prohibited from *"… recovery, recycling, reclamation, direct reuse or alternative uses*". It cannot be *"… transported across international boundaries…"* to developing countries with no or low capacity for environmentally sound management (ESM)[6] of the waste. The Basel Convention's (BC) experts are tasked with proposing LPCL for POPs within the *General Technical Guidelines for the ESM of POPs wastes* (The Guidelines) and only the Conferences of Parties to the BC and SC can approve them[7]. Negotiations and decisions on LPCL should be based on the protection of human health and the environment but political and economic factors often interfere, resulting in weak, unprotective LPCL.
Current LPCL for PCDD/Fs has two provisional values in The Guidelines, either 1 ppb and/or 15 ppb. However, the LPCL of 15 ppb[8] is more commonly used. This definition was established using the primary criteria of avoiding special treatment for large volumes of wastes in 2005[9]. Its revision proposes a new LPCL in the EU at 5 or 10 ppb[10].
EU at the crossroads: A study by Ramboll[10] prepared for the European Commission, expressed the concerns of the Confederation of the European Waste-to-Energy Plants (CEWEP): *"Fly ashes from municipal solid waste incineration are considered an important mass flow in this respect. Available data indicate that 11% and 3% of such fly ashes would exceed an LPCL of 5 μg TEQ/kg and 10 μg TEQ/kg respectively. This indicates that if the LPCL for PCDD/Fs would be lowered correspondingly, the majority of the fly ash can still be used according to current treatment. Stakeholders, however, claim when the LPCL of PCDD/Fs is lowered, the safe recycling (e.g. as filler in asphalt) could be hampered … For PCDD/Fs, based on the results of this study a lower value in the range for example of 5-10 μg/kg TEQ should be considered"*[10]. This shows that the main criteria LPCL is not human health protection but protecting the interests of waste-to-energy plants (W-t-E) to maintain "recycling" of PCDD/Fs contaminated fly ash in various applications. This undermines the requirements of the SC as "recycling" of POPs waste is prohibited.
Gaps in reporting about PCDD/Fs in residues/wastes: Recent estimates of total PCDD/Fs releases globally were 101.4 kg TEQ year[11]. It was also based on earlier summary of PCDD/Fs national inventories[12]. Out of 86 countries, 8 did not report about PCDD/Fs transfers in residues in these inventories, including Australia, Austria, Bulgaria, Finland, France, Germany, Russia and Switzerland[12]. The other group of countries with major W-t-E capacity such as Denmark, Netherlands, Spain, UK, and Japan were not included. Austria, Finland, France, Germany and Switzerland had 277 W-t-E plants with capacity of 47.4 million tons / year which counted for 20.77% of overall global capacity according the database from 2013[13]. Denmark, Netherlands, Spain and UK had 112 W-t-E with capacity of 22.2 million tons / year amounting to an additional 9.7% of total global capacity, and finally Japan had 826 W-t-E with capacity of 60.1 million tons / year, or 26.33% of global capacity. In total there is missing data about PCDD/Fs in residues from countries which held 56.8% of world capacity of W-t-E[13]. According previous estimate W-t-E generates between 3.4 and 45.6 kg TEQ PCDD/Fs per year in fly ash[14]. It

[a] Municipal solid waste incineration (MSWI) is also described as Waste-to-Energy plants (W-t-E) in some parts of this study.

means that several kg TEQ/annum of PCDD/Fs in waste incineration residues is missing from the primary global PCDD/Fs inventory[11]. This study aims to fill this gap by using available data about PCDD/Fs in WI residues.

Materials and methods

Estimation of total PCDD/Fs in WI fly ashes requires data on global capacity of municipal solid waste incinerators (MSWI) and/or W-t-E and hazardous waste incinerators (HazWI) and medical waste incinerators (MedWI). It also requires emission factors for PCDD/Fs flow into their fly ashes. We used databases about global capacity of W-t-E for MSWI[13,15,16] and emission factors for PCDD/Fs in MSWI fly ashes from the Dioxin Toolkit 2013 version[5]. There was no data on global capacity of HazWI and MedWI, so we extrapolated from national data available about their releases/transfers of PCDD/Fs in ashes and extrapolated the data.

We analysed discrepancies between estimations of PCDD/Fs releases and transfers in MedWI and HazWI residues in Central European EU member states and data available in state reporting systems. We used data from national Pollutants Release and Transfer Registers (PRTRs) where chemically specific reporting about waste transfers is available[17]. In other states we used data from their National implementation plans (NIPs) for the SC double checked with data available in inventories provided to SC[18]. However from 139 countries, 32 countries (23%) did not report PCDD/Fs transfers in residues (wastes)[19] in their official inventories.

Results and discussion

Estimation of PCDD/Fs in MSWI fly ashes: The total volume of fly ash produced by WI globally is not easy to calculate as basic data is only available for a fraction of all WI. Total capacity of MSWI/W-t-E globally was estimated at over 228.24 million tpa in 2013[13]. Estimates in 2017 were around 250 to 258.4[b] million tpa[15,16].

If we calculate that 3% of fly ash is created from the total weight of burnt waste, the result is the production of 7.75 million tons of fly ash per year for the 2017 capacity estimate of 258.4 million tons[15]. W-t-E mostly use more sophisticated APC systems so they belong to the class 3 and 4 MSWI classification of the Dioxin Toolkit[5]. Classes 3 and 4 have emission factors for fly ash of 200 and 15 μg TEQ/t MSW incinerated respectively. This calculation leads us to estimates of 3.9 kg TEQ and 51.7 kg I-TEQ PCDD/Fs releases/transfers per year (in fly ash wastes) for class 3 and class 4 municipal waste incinerators respectively. This estimate assumes that W-t-E plants use 100% of their installed capacity which is mostly not the case but there is no data about the actual capacity used every year by W-t-E plants. Assuming 90% of their capacity is used per annum. The total amount of PCDD/F released in WI residues should be adjusted accordingly.

Class 4 of MSWI is assigned a PCDD/F concentration value for fly ash at 1 ng TEQ/g and for bottom ash at 5 pg TEQ/g[5]. Class 3 has levels approximately half of those established for class 2, defined by 30 ng TEQ/g in fly ash and 100 pg TEQ/g in bottom ash[5]. Levels of PCDD/Fs in fly ashes are not measured regularly in WI but were in the range of 0.2 – 23.9 ng TEQ/g in 35 W-t-E units from the EU presented in Table 85 of the Ramboll study[10]. Median and average levels in these 35 W-t-E were 1.2 and 2.5 ng TEQ/g respectively. One plant had level of 9.9 and one had a level of 23.9 ng TEQ/g[10]. In MSWI in South Korea the levels of PCDD/Fs in fly ash were 0.244, 2.548 and 24.786 ng TEQ/g respectively[19]. Levels in the range of 0.034 – 2.5 ng WHO-TEQ/g were measured in fly ash from waste incinerators in China[20]. It seems there are not many W-t-E with levels of PCDD/Fs as class 3 (approx. 15 ng TEQ/g) in fly ash so we estimate they should be 10% of all W-t-E.

After correction of the estimate from available data (10% of class 3 W-t-E and 90% of class 4 W-t-E) corrected for 10% unused capacity of W-t-E, we calculate a final estimate of PCDD/Fs in fly ashes from global MSWI as ***7.8 kg TEQ/annum***.

Estimation of PCDD/Fs in hazardous waste incineration residues: We are not able to conduct the same calculation for HazWI and MedWI as information on their global capacity is not available. There are some indications of the scale of PCDD/Fs releases in fly ash from these waste incinerators in the NIPs from a few countries, and there is additional data from other information sources. They are summarized in Table 1. Large HazWI are mostly operated only in developed countries while in developing countries MedWI are often used with a small annual capacity up to several thousand tons. They also produce bottom ash mixed partly with fly ash with relatively high content of PCDD/Fs of 500 ng TEQ/kg and more[21,22] which is often dumped next to MedWI[23] so we include this in the total calculation of PCDD/Fs produced in “fly ash” globally.

There are estimates about total content of PCDD/Fs in waste incineration residues (mainly fly ash) derived from country reports and in NIPs submitted to the Secretariat of the SC from 86 countries. Based on this information PCDD/Fs releases in waste incineration residues are almost 800 g I-TEQ per annum[18], however, when we look closer we can see that countries with the highest capacity of WI (e.g. Germany, China, Japan) did not report any PCDD/Fs in waste incineration residues and some others were not included, such as Ukraine which reported PCDD/Fs releases in residues of 156.5 g I-TEQ/annum for 2002[24].

Estimations based on PRTRs: We can also use data about PCDD/Fs transfers in wastes from national PRTR if available. We calculated and average of PCDD/Fs reported in WI residues by waste incineration companies to

[b] Calculation based on information that W-t-E plants burn daily approximately 700.000 metric tons of waste[15].

the Czech PRTR system in 2012 – 2019. Average of 15 g TEQ/year and 20.7 g TEQ/year was reported for WI ashes from MSWI and HazWI (including MedWI) respectively. It is more than estimated previously for HazWI and MedWI from the Czech Republic, and it is obviously more than estimated in a collective inventory from 2004 which estimated total releases in waste incineration residues from HazWI and MedWI to be 5 g I-TEQ and 28 g I-TEQ respectively per year for 13 EU candidate countries[25]. In 2006 the Hungarian hazardous waste incinerators released more than 11.5 g I-TEQ/annum[26] PCDD/Fs into waste residues. So only 2 of 13 former EU candidate states count for the total level estimated for all 13 countries.
Also Japan based its reporting of 1,514 g TEQ of PCDD/Fs in wastes transferred or buried, such as particulates and burnt residues on data from PRTR for 2018[27].
These examples show that more emphasis should be given to chemically specific reporting about POPs listed under the SC in waste flows (transfers) in PRTR systems. The obligation to report about PCDD/Fs in waste into the European PRTR could fill the data gap for many EU states about PCDD/Fs in WI residues[18].
PCDD/Fs in waste incineration fly ashes globally: For the estimation of the transfers of PCDD/Fs in HazWI and MedWI we had to use data available in NIPs, PRTRs and similar sources (see Table 1).

Table 1. PCDD/F releases in g TEQ/annum. Source Petrlik, Bell (2017) [14] if not specified otherwise.

Country[(source)]	Argentina	Brazil	China[28]	Czechia[a)]	EU[b) 9]	Hungary[d) 26]	India[e)]
HazWI	27	20.72	186	20.7	61.8	11.53	3,965.8
MedWI	-	-	748.9		29.1	-	-
Year	2006	2014	2004	2015	2005	2006	2010
Country[(source)]	Indonesia[f)]	Japan[27]	Kenya[g)]	Lithuania	Nigeria	South Africa[29]	USA[h) 5,30]
HazWI	58	1,514	10.15	0.64	0	12.22	93 – 1,395
MedWI	-		-	0.5	15.851	-	NA
Year	2001	2018	2006	2004	2004	2012	2005

Notes:; a) Calculation based on 8 years reporting in PRTR, includes also MedWI; b) MedWI calculated for 10 EU member states only c) Industrial waste and sewage sludge incineration; EU + Switzerland and Norway; d) Calculated from data in Annex 6; e) This figure is for all waste incineration plants in India (including MedWI), however there was only one W-t-E plant in operation in India with capacity 54,000 tonnes/annum[13]; f) Not very clear whether all comes from hazardous waste incinerators; g) Both HazWI and MedWI h) Calculated by using Dioxin Toolkit[5].

Based on available data, the overall calculation for PCDD/Fs in fly ash and other APC residues produced by HazWI and MedWI globally, might be within a similar scope as they are for MSWI when calculated according to the Dioxin Toolkit emission factors, what means approximately ***7 kg TEQ/annum***, as total for selection of countries in Table 1 only it is more than 6 kg TEQ/annum.
It means that total releases from waste incineration residues annually could amount to approximately ***14 - 15 kg TEQ of PCDD/Fs***. This seems to be a bigger share of total PCDD/Fs releases into the environment than estimated from inventories obtained by SC Secretariat from individual countries[18]. We also estimate that 10% of PCDD/Fs in fly ash produced by current MSWI are in the ash with levels above current provisional LPCL 15 ppb. Data about levels of PCDD/Fs in fly ash from HazWI and MedWI indicate that their percentage above 15 ng TEQ/g will be higher but it is much harder to estimate proportion among this group.
How big part of these PCDD/Fs is meant to be under control according the Stockholm Convention rules? It was estimated in 2005 that approximately 86,000 tons of POPs waste in EU would be above LPCL of 15 ng TEQ/g while 2,255,000 tons would be above level of 1 ng TEQ/g[9]. By establishing LPCL at 15 ppb level EU decided to take responsibility for at least 1.3 kg PCDD/Fs (86 x 15g TEQ) in wastes while approximately more than 2 kg of PCDD/Fs (2250 x 1g TEQ) were left without control. We estimate that at least half of total amount of 14 – 15 kg TEQ PCDD/Fs in WI fly ashes but most likely much bigger portion of them is suggested to be left in wastes without control under currently used LPCL 15 ng TEQ/g.
Comparison with food safety standards: European Food Safety Authority (EFSA) set stricter value of 2 pg TEQ/kg body weight for tolerable weekly intake (TWI) of PCDD/Fs and dl-PCBs recently. It means that one person of 70 kg weight should not have intake of more than 7280 pg TEQ PCDD/Fs/dl-PCBs per year. For population of planet Earth which is now 7.7 billion people it is 56.056 g TEQ of PCDD/Fs/annum. Half of the amount of PCDD/Fs in WI fly ash which is suggested to be left without control is equal to the tolerable dose of human population of 133 planets Earth. If we use TWI of 14 pg TEQ/kg body weight we get to the amount equal to the tolerable dose for 19 planets Earth. Fortunately not all PCDD/Fs reach our food chain. There were documented examples of contamination of food chain at sites with unsafe disposal of WI ash or other industrial ash contaminated with PCDD/Fs at levels of 500 pg TEQ/g dw and more[31,32]. Also proposed most “contaminated coral reef in the world” lies next to a seafill in Bermuda where cement-stabilized WI ash was used[33]. These cases might also happen because of weak LPCL for PCDD/Fs is used globally.

Conclusions

The documented gaps in the inventory of PCDD/Fs in waste incineration residues reveal the extent to which the potential threat from uncontrolled transfers of POPs in wastes to the food chain and human health is underestimated. It is a consequence of weak limits for PCDD/Fs and dl-PCBs in wastes which do not require industry to monitor their concentrations and allow 'recycling' as the option to manage this waste globally despite its high POPs content. We estimated that ***14 - 15 kg TEQ of PCDD/Fs is in waste incineration fly ash*** and/or mixed fly and bottom ash residues globally. Open reared animals can access often sites contaminated with POPs including PCDD/Fs and waste incineration ashes contaminating food product for human consumption. The LPCL is one of the critical control measures for POPs waste at a global level that could be used to address this problem if it were strengthened. Data collected from some countries demonstrate that PRTR can help with more precise inventory of PCDD/Fs transfers in wastes if chemically specific reporting for wastes is introduced.

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