PCDD/Fs in Waste Incineration Fly Ash

Petrlik J^{1, 2}, Bell L^{2, 3},

- ¹Arnika Toxics and Waste Programme, Prague, Czech Republic, CZ-17000
- ² IPEN, Gothenburg, Sweden, SE-40235
- ³ National Toxics Network, Perth, Western Australia, Australia, 6054

Introduction

This study was undertaken to address the hypothesis that the open use fly ash and other residuals from waste incineration facilities, which are contaminated with persistent organic pollutant (POPs), may be leading to accumulation of polychlorinated dibenzodioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) and other POPs in the environment at levels which may be of concern to human health and ecological values despite existing regulatory limits. The PCDD/F which has been entrained in the ash residues (and other wastes) is subject to regulation in the form of Low POPs Content Levels (LPCL) established under the Basel Convention General Technical Guidelines [1]. The current provisional LPCL of PCDD/F used to classify wastes as POPs wastes under the Basel Convention is 15 µg TEQ/kg (15 parts per billion) [1]. To test our hypothesis, we reviewed available literature on incinerator residues used in scenarios which can lead to POPs leaching from the ash. We then determined transfer rates of PCDD/F from residues to soil under current regulatory controls and assessed whether this led to food contamination exceeding current threshold limits for human consumption. The global production of fly ash and dioxin content were also calculated.

Materials and methods

To assess the total annual dioxin in residues we assessed the current number of global operational incineration facilities, their throughput capacity, operational capacity and waste treatment type (hazardous waste, medical waste or municipal waste). It was then possible to apply the release/emission factors (in the case of wastes the term "transfer" is also used in PRTR databases) by category included in the updated 2013 UNEP Dioxin Toolkit. By multiplying the facility numbers (according to category) by release factors and incorporating operational capacity estimates it was possible to obtain an estimate of total PCDD/F loading in annual arisings of incinerator residues. Incinerator residue uses were identified in the scientific literature and supported by reports from government, industry and civil society including National Implementation Plans (NIP) submitted to the Stockholm Convention. Several of these studies quantified concentrations of PCDD/F in fly ash and the subsequent impacts on biota used for human consumption. In this case, the commonly used biomarker was chicken eggs which efficiently concentrate PCDD/F in their lipid content. Previous studies had established the relationship between PCDD/F in residues and other wastes and their release to soils and subsequent concentration in chicken eggs for human consumption [2-6]. Citing existing literature, it was possible to determine if current EU regulatory limits for PCDD/F in eggs has been exceeded and extrapolate this data to determine if tolerable daily intake (TDI) levels for humans would be exceeded.

Results and discussion

The amount of dioxin contained in annual production of waste incineration residues is highly underestimated. Bottom ash, can reach 20 - 30% by mass of the original waste on a wet basis. The fly ash component, in the form of APC residues, are in the order of 1-3% and total APC residues account for 2-5% of the waste input mass on a wet basis [7]. Estimates in this study suggest that total volumes of fly ash generated globally per

annum are in the region of several million tonnes. This study estimates that, in total, approximately 7 kg I-TEQ - 10 kg TEQ/year of PCDD/Fs are released into waste incineration residues, which is 3-10 times higher than previous estimates, and that this quantum may be exceeded considering that not all MSWI are in Category 4 of the UNEP Dioxin Toolkit classes [8]. Fly ash is reused for a variety of different purposes on a broad scale [9-13] including applications such as construction material and paving which are eventually demolished and can lead to remobilisation of POPs. This is occurring on a large scale via incinerator ash distribution, trade and reuse. The most difficult to control uses are residues employed as food additives for poultry [14], fertilizer, soil amendment for agricultural use, road base, embankment construction and as pavement foundations or on surfaces in areas with locally grown food [15-18].

A recent Swedish EPA study demonstrated that PCDD/Fs levels of 30 pg TEQ g-1 fat in an egg will be exceeded at soil concentrations of approximately 4 to 75 ng TEQ kg-1 d.m. Therefore, the European maximum level of 3 pg TEQ g-1 PCDD/F in fat in eggs can be exceeded at levels that are ten times lower (i.e. 0.4 and 7 ng TEQ kg-1 d.w.). Based on the upper level of the range given in a recent Swedish EPA study and an example of a scenario with contaminated wood waste, [19] it can be concluded that application of fly ash and other wastes containing levels of dioxin over 0.05 ppb in agriculture (and other land based application) can lead to 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 (2.5 pg WHO-TEQ g⁻¹ of fat) [20-22]. Free range poultry eggs can be impacted at critical levels exceeding currently used safety limits (2.5 pg WHO-TEQ g-1 fat) by severalfold, with some cases revealing a 10-fold exceedance. Locally produced food is of great importance in developing countries and rural locations in developed countries therefore this exposure scenario is of particular concern. Weber et al. [23] suggested that contamination levels in soil used for the production of free-range eggs should ideally be less than 2 ng TEQ kg-1 d.m. for the sum of PCDD/Fs and DL PCBs (and certainly less than 5 ng TEQ kg-1 d.m.) based on conclusions of the recent report on POPs in free range chicken eggs in the Netherlands [24]. Levels higher than this will present a risk of exceeding tolerable daily intakes for PCDD/Fs and DL PCB in humans from chicken eggs [22, 23]. Chicken eggs concentrate DL PCBs more efficiently than PCDD/F and therefore must be taken into consideration in the total dioxin load present in an egg as well as in the transfer mechanism model from residue to soil and then accumulation to eggs [19].

Even the lowest proposed Low POPs Content Level of 1 ppb for adoption at the Basel Convention [25] underestimates the risk associated with POPs contaminated incineration residues as it does not include dioxin -like PCBs (DL PCBs) in modelling and overlooks cases where lower levels of dioxin in soil can lead to serious exceedances of the EU standard for eggs [19]. The range of observed dioxin levels in fly ash is from below the level of quantification (virtual zero) to 96,000 ppb. Alternative waste management practices and waste disposal technologies and techniques that can prevent formation of dioxin should be adopted as a priority [26] as an alternative to waste incineration. While the EU and other jurisdictions use the provisional Low POPs Content level of 15ppb to define POPs waste², far more stringent levels for PCDD/Fs control in waste and/or

¹ "Empirical data show that chickens that are exposed to wood containing 50 ng WHO-TEQ kg-1 (0.05 ppb) may produce eggs with concentrations significantly exceeding the maximum limits (88 pg WHO-TEQ g-1 fat or 8.8 pg WHO-TEQ g-1 whole egg).

² According to Article 6 d) (ii) of the Stockholm Convention, waste defined as POPs waste must be "disposed of in such a way that the persistent organic pollutant content is destroyed or irreversibly transformed so that they do not exhibit the characteristics of persistent organic pollutants".

contaminated soil are used in some other countries which are below the current provisional LPCL value of 15 ppb, e.g. 1 ppb or 3 ppb [27-29] suggesting an awareness of the environmental impacts caused at higher levels and the need to reduce the 15 ppb level.

Recommendations

Parties to the Stockholm Convention and Parties to the Basel Convention should adopt the more stringent value for Low POP Content Level for PCDD/F of 1 ppb to avoid the risk of transboundary movement of incinerator residues and loss of control of the fate and transport of large volumes of POPs contaminated residues. Authorities should restrict the use and application of wastes to soil or on terrain surface (without stabilization) with a level of PCDD/Fs and DL PCBs above 0.05 ppb to prevent food chain contamination. Dioxin-like PCBs should be considered in the evaluation of LPCL, so the level of 1 ppb will be applicable for both PCDD/Fs and DL PCBs as a total expressed in WHO-TEQ. Technologies, which do not generate UPOPs, should be used for destruction of dioxins in waste incineration fly ash above 1 ppb with a destruction efficiency (DE) above 99.999% measured as total TEQ level. Guidelines for BAT and BEP should be developed and the Basel Convention Technical Guidelines on POPs Wastes should include more of these technologies. An updated inventory of PCDD/Fs and other U-POPs in waste incineration residues should also be conducted.

Acknowledgements

IPEN, Arnika and NTN gratefully acknowledge the financial support provided by the Government of Sweden, Global Greengrants Fund, and other donors, which make the production of this document possible. The views herein shall not necessarily be taken to reflect the official opinion of any of the institutions providing financial support. We also thank to all participating organizations of IPEN which helped to collect information and/or conduct sampling at selected locations, and to thank Center of Environmental Solutions – CES (Belarus), Nature University (China), EARTH (Thailand), Taiwan Watch Institute (Taiwan), and Public Interest Consultants (UK) in particular.

References

- 1. Basel Convention, General technical guidelines for the environmentally sound management of wastes consisting of, containing or contaminated with persistent organic pollutants, in Technical Guidelines. 2017: Geneva.
- 2. Hoogenboom, R., et al., *The German bakery waste incident; use of a combined approach of screening and confirmation for dioxins in feed and food.* Talanta, 2004. **63**(5): p. 1249-1253.
- 3. Hoogenboom, R., et al., *Increased levels of dioxins in Irish pig meat: the Dutch connection*. Organohalogen Compd, 2009. **71**: p. 2182-2186.
- 4. Malisch, R., *Increase of the PCDD/F-contamination of milk, butter and meat samples by use of contaminated citrus pulp.* Chemosphere, 2000. **40**(9–11): p. 1041-1053.
- 5. Bernard, A., et al., *The Belgian PCB/Dioxin Incident: Analysis of the Food Chain Contamination and Health Risk Evaluation*. Environmental Research, 2002. **88**(1): p. 1-18.
- 6. Llerena, J., et al., *An episode of dioxin contamination in feeding stuff: the choline chloride case.* Chemosphere, 2003. **53**(6): p. 679-683.
- 7. Sabbas, T., et al., *Management of municipal solid waste incineration residues*. Waste Management, 2003. **23**(1): p. 61-88.
- 8. UNEP and Stockholm Convention, *Toolkit for Identification and Quantification of Releases of Dioxins, Furans and Other Unintentional POPs under Article 5 of the Stockholm Convention*. 2013, United Nations

- Environment Programme & Stockholm Convention Secretariat: Geneva. p. 445.
- 9. Astrup, T., Management of APC Residues from W-t-E Plants An overview of management options and treatment methods Second edition, October 2008. 2008, ISWA. p. 116.
- 10. Ferreira, C., A. Ribeiro, and L. Ottosen, *Possible applications for municipal solid waste fly ash.* Journal of Hazardous Materials, 2003. **96**(2–3): p. 201-216.
- 11. Sun, X., et al., A Review on the Management of Municipal Solid Waste Fly Ash in American. Procedia Environmental Sciences, 2016. **31**: p. 535-540.
- 12. Lam, C.H., et al., Use of incineration MSW ash: a review. Sustainability, 2010. 2(7): p. 1943-1968.
- 13. Lichtfouse, E., J. Schwarzbauer, and D. Robert, *Pollutant Diseases, Remediation and Recycling*. Environmental Chemistry for a Sustainable World. Vol. 4. 2013.
- 14. Lee, W.-J., et al., Assessment of polychlorinated dibenzo-p-dioxins and dibenzofurans contribution from different media to surrounding duck farms. Journal of Hazardous Materials, 2009. **163**(2-3): p. 1185-1193.
- 15. Pless-Mulloli, T., et al., Executive Summary: PCDD/F and heavy metals in soil and egg samples from Newcastle allotments: Assessment of the role of ash from Byker incinerator. 2001. p. 50.
- 16. Watson, A., Comments on the "Report on the analysis of PCDD/PCDF and Heavy Metals in Soil and Egg samples related to the Byker incinerator". 2001.
- 17. Pless-Mulloli, T., Air, V., Schilling, B., Päpke, O. and Foster, K., *Follow-up Assessment of PCDD/F in Eggs form Newcastle Allotments*. 2003, University of Newcastle upon Tyne, Newcastle City Council: Newcastle. p. 39.
- 18. Pless-Mulloli, T., et al., Report on the analysis of PCDD/PCDF and heavy metals in foodpaths and soil samples related to the Byker incinerator. 2000, University of Newcastle upon Tyne, Newcastle City Council: Newcastle. p. 35.
- 19. Swedish EPA, *Low POP Content Limit OF PCDD/F in Waste. Evaluation of human health risks*. 2011: Swedish Environmental Protection Agency, Stockholm. p. 145.
- 20. Pirard, C., et al., Assessment of the impact of an old MSWI. Part 1: Level of PCDD/Fs and PCBs in surrounding soils and eggs. Organohalogen Compounds, 2004. 66: p. 2085-2090.
- 21. Harnly, M., et al., *Polychlorinated Dibenzo-p-dioxin and Polychlorinated Dibenzofuran Contamination in Soil and Home-Produced Chicken Eggs Near Pentachlorophenol Sources*. Environ. Sci. Technol., 2000. **34**(7): p. 1143-1149.
- 22. DiGangi, J. and J. Petrlik, *The Egg Report Contamination of chicken eggs from 17 countries by dioxins, PCBs and hexachlorobenzene.* 2005.
- 23. Weber, R., et al., *High levels of PCDD/F, PBDD/F and PCB in eggs around pollution sources demonstrates the need to review standards.* Organohalog Compd, 2015. **77**(2015): p. 615-618.
- 24. Hoogenboom, R., et al., *Dioxines en PCB's in eieren van particuliere kippenhouders*. 2014, RIKILT (University & Research centre): Wageningen. p. 25.
- 25. 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.
- 26. Petrlik, J. and L. Bell, *Toxic Ash Poisons Our Food Chain*. 2017. p. 108.
- 27. Government of Japan, *The National Implementation Plan of Japan under the Stockholm Convention on Persistent Organic Pollutants*. 2006: Tokyo. p. 99.
- 28. Government of Japan, *Dioxins. Information brochure*. 2012: Tokyo. p. 28.
- 29. Government of Republic of Korea, *The National Implementation Plan of the Republic of Korea under the Stockholm Convention on Persistent Organic Pollutants*. 2009: Seoul. p. 59.