HIDDEN EMISSIONS OF UPOPs: CASE STUDY OF A WASTE INCINERATOR IN THE NETHERLANDS

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Introduction

In 2013 ToxicoWatch found elevated levels of dioxins in eggs of backyard chicken in the vicinity of new built incinerator in the Netherlands[1,2], several programs of research were set up. One of these researches is a long-term sampling of emissions of the incinerator and analysis on PolyChlorinated DibenzoDioxine/Furans (PCDD/F) and dioxin-like PolyChlorinated Biphenyls (dl-PCBs). The results were presented earlier[2]. A parallel program is presented here on brominated and fluorinated unintentionally produced persistent organic pollutants (UPOPs): PolyBrominated Biphenyl (PBB) , PolyBrominated DibenzoDioxine/Furans (PBDD/F), PolyBrominated DiphenyEthers (PBDE), PolyBrominated DibenzoDioxine/Furans (PBDD/F), PerFluoroOctanoic Acid (PFOA), PerFluoroOctaneSulfonic acid (PFOS) and emissions of Polycyclic Aromatc Hydrocarbons (PAH). The efficiency of long-term sampling is analysed.

Materials and methods

The case study is the 'Waste to Energy' incinerator (*Reststoffen Energie Centrale)*, abbreviated as REC (2011), located along the coastline of the Unesco WaddenSea, 2 km from the centre of the harbour city Harlingen, the Netherlands (16.000 inhabitants). This installation is the most modern in the Netherlands, set up with modern concepts of sustainable waste disposal applying BAT/BEP, transforming thermic energy into electrical (17 MW).

Continuous sampling of flue gas is performed from August 2015 till December 2017, with one measurement during the annual maintenance stop in May 2018. Method of sampling is the **A**dsorption **ME**thod for **SA**mpling, abbreviated as AMESA[3] (Environnement). Analyses on PCDD/F/dl-PCBs, PBB, PBDE, PBDD/F, PFOS, PFOA and PAH in long-term sampling cartridges were performed by Eurofins, Hamburg, Germany. Dioxin emission measurement by short term sampling by Promonitoring, Deventer, the Netherlands. The program of short-term measurements of start-up events (2015, 2016 and 2017) by 'OmgevingsDienst Regio Arnhem' (ODRA), the Netherlands, and dioxins analysed with HR/GC/MS by Al-West, Deventer, the Netherlands and PAHs by Agrolab, Deventer.

Results and discussion

The results of 20,139 hours (offline hours included) of long-term sampling of dioxins are shown in Figure 1 (log PCDD/Fs). Long-term sampling of AMESA was 1,676 hours off-line (8,3 %), included 1,159 hours during the annual maintenance stop. Total off-line time long-term sampling, during start-ups, shutdowns and other transient stages, were found to be 517 hours (2,6 %). This could be traced back in the log files of the AMESA[4]. In figure 1 the circled peaks exceeding 0,1 ng TEQ/Nm3, but sampling was interrupted for hours. There's a correlation between elevated emissions of dioxins and interruptions in sampling. Even regular measurements are impaired, blocked or clipped in calamities of the REC. The notion of 'Out of Operation' is common during transient stages and data of parameters are lacking. What we know, however, is that different studies on the REC calamities show a cascade of incidents were multiple issues, sometimes resulting in uncontrolled shutdowns.

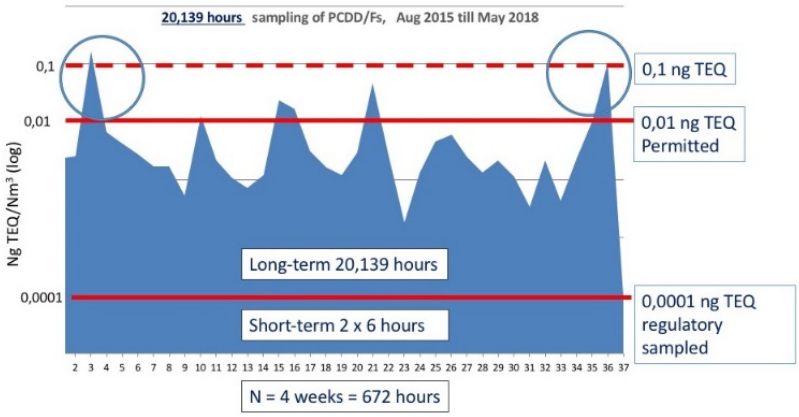

Figure 1: Results of 20,139 hours AMESA long-term sampling PCDD/Fs, REC Harlingen

It means not all emissions could be sampled on a correct way. And with this perspective the other UPOPs had to be considered. The other UPOPs are not regulated in the directive of Industrial Emissions.

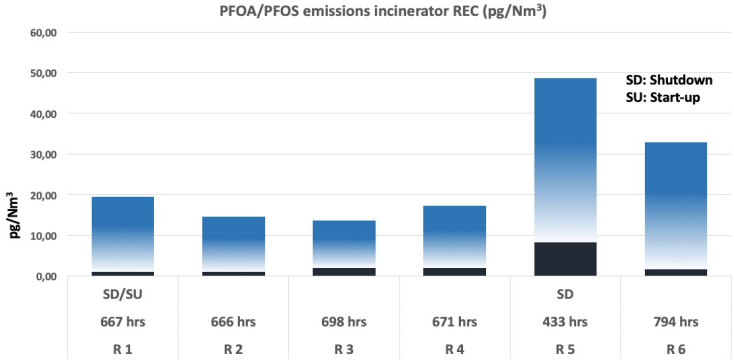

Figure 2: Longterm sampling of PFOS and PFOA

In Figure 2 is shown the results of 6 long-term sampling measurements of PFOA and PFOS. PFOA is recently prioritized in the Stockholm Convention May 2019, to be eliminate in production and use because of its extreme hazardous characteristics. PFOA as waste product, wasn't discussed, because of lack of data. This is the first data of PFOA in flue gases. in the range of 0,013 – 0,041 ng/Nm3 (n = 6, sampling time 3,929 hrs). The total sampling time was 3,929 hours. If the incinerator is a substantially source of PFOA more research is needed. The results of PFOA are indicative and are possible underestimated by sampling obstruction, as discussed before. PFOA are found nowadays everywhere and a recent research in backyard chicken eggs, show surprisingly a linear correlation with dioxins and dl-PCBs[5]. EFSA adjusted recently the toxicity of PFOA with a factor 1500 below. Every emission of PFOA needs to be considered.

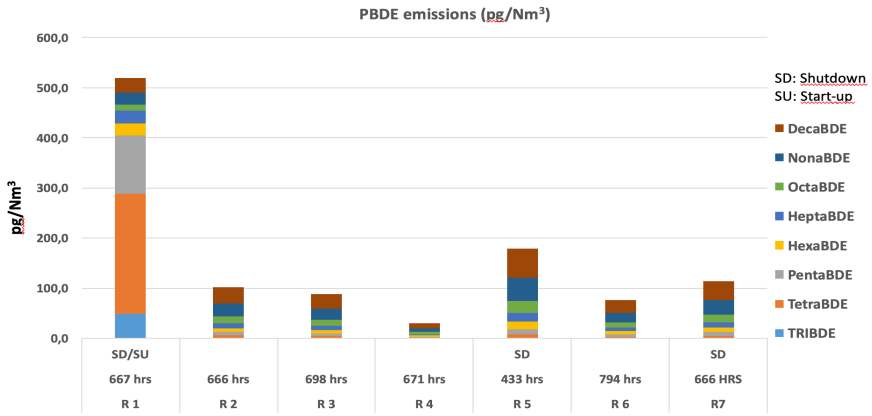

Figure 3: Long-term sampling of PBDE

In figure 3 the results of polybrominated diphenyl ethers (PBDEs) are shown in the flue gases of the incinerator. Most PBDEs are found in transient stages of start-ups and shutdowns. And what is mentioned before, all transient events are characterised with interruptions of sampling.

Polybrominated biphenyls (PBBs): 0.038 – 0.133 ng/Nm^3 (Figure 4). Normally, PBBs already decompose with temperature around 300 ^{0}C. This can be an indication, temperature requirements in the post combustion zone are not reached.

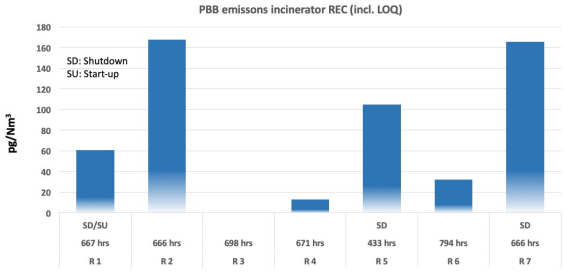

Figure 4: Long-term sampling of PBB

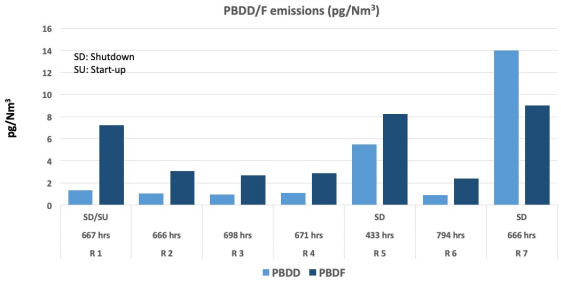

Figure 5: Long-term sampling of PBDD/F

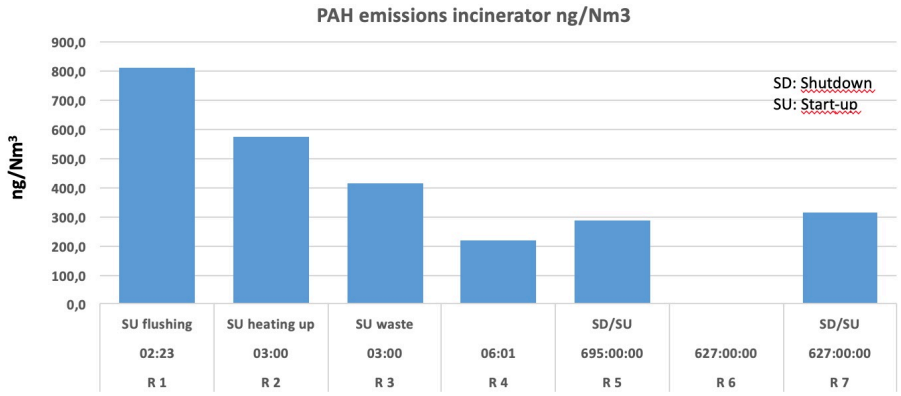

Figure 6: Short- and long-term sampling results PAH

In figure 5 sampling results of PBDD/F show elevated levels during the start-ups and shutdown. In figure 6 the results of long- and short-term sampling of PAH during start-up events are seen. The concentration of PAH is found between 2,4 – 808,8 ng/Nm^3. This is substantial lower than the permit for PAH emitting installation, 50 ug/Nm^3. However, the incinerator has no permit for PAH emission, because no emissions of PAH was expected. This sampling of PAH was done, because of findings of high levels of PAH in the environment, but no correlation with the emissions of the incinerator could be made.

Dioxin-like polychlorinated biphenyls (Dl-PCBs) are 8,5% of the total TEQ (n = 36 total 20,139 hours). This finding is in contrast with Sakurai et al[5], who found less than 3% contribution of dl-PCBs in the total TEQ in the flue gas. A research in 2017 (6 years after the start in 2011) by TÜV Rheinland Energy GmbH finds a lack of homogeneity of temperature and oxygen in the post-combustion zone[6,7], while this is required in the technical guidelines of proper combustion of waste with requirements of residence of 2 seconds above the 850^0 C in the post combustion zone. Too low temperatures can be a reason of incomplete destruction of PCBs.

Conclusions

The finding of a broad scale of UPOPs indicates an incompleteness of combustion. The question arises if modern waste combusting has to be performed at all time at 1100^0 C to a more effectively destruction of the chemical cocktail of the household/municipal waste? There's a pattern of elevated UPOPs during start-up or shutdown events. Together with the findings of impairment of sampling during this transient stages, the findings only can be interpreted as indicative, like we find more PCDD/F.

Sources of dioxins are controlled by governmental regulations only 0,1 % of operating time (2 times a year, 6- 8 hours per event, pre-announced). And in spite of the many improvements of combustion control, the regulation still is set for 0,1 ng TEQ/Nm3. The new state of the art incinerator in Harlingen, however, is given a more stringent of 0,01 ng TEQ/Nm3. Modern incinerator can use bypasses in the flue gas cleaning system, allowing to 'dump' dust without any regulation. Already in 1998 de Frey[8] reported the excess of dioxins during the start-ups. Still no regulation is developed to reduce this exceeding emissions of dioxin, because it 'posed no legal problem' for the facility. Norms are only stipulated to 'apply to steady state operation'. And so, modern incinerators can continue to use bypassing the flue gas cleaning system. In agreement with Cheruiyot et al.[9] this study show dioxins and other UPOPs emissions should be re-considered during start-up stages in waste incineration[10]. Measurement of a much broader spectrum of POPs in flue gases is strongly advised to meet the need for the elimination of all POPs in the environment, which is the major objective of the Stockholm Convention.

Attention needs to be paid to PFOA, which is recently at the Basel, Rotterdam Stockholm Conventions (COPs 9) of 2019 listed as an urgent contamination issue. PFOA as waste product is still not regulated, but finding now in the cleaned flue gas can be a threat for the environment. Taking also in consideration, sampling was not complete during start-up and shutdown events.

Another point of attention is the emission of dl-PCBs by incineration. The finding of a factor 3 more dl-PCBs in the emissions can be due a high PCB-input or level of temperature is too low to destroy PCBs effectively. It needs more research in order to eliminate PCB completely.

The technique of incineration of waste and of biomass is promoted as a sustainable green answer. But the question arises if we are not facing a very regrettable solution, because of producing extremely toxic substances. This report underlined the need to update the data reported by the Harlingen waste incinerator or at least thoroughly being controlled by engaged enforcements to be in line with the guidance documents of the Stockholm and Basel Conventions such as for example BAT/BEP Guidelines for technologies listed in Annex C[12].

Moreover, the results of the measurements in the REC incinerator raise important questions for future policy-making concerning what can be accepted as normal operating – and monitoring- conditions for incinerator plants concerning their potential effects on public health and the environment. The studies reviewed here show unequivocally that dioxins are still a serious issue, that measurement programs still show serious shortages, that the health of the population is still under threat and there is unfortunately still a long way to go to eliminate emissions of persistent organic pollutants to the environment.

Acknowledgements

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References

1. Arkenbout A, Esbensen KH, (2017), *Eighth World Conference On Sampling and Blending*. 117-124.
2. Arkenbout A, Olie K, Esbensen, KH (2018), Vol 80, 413-416
3. Reinmann, J, Weber, R and Haag, R, (2010); Science China Chemistry, 53(5):1017–1024.
4. Arkenbout A, (2018*), Hidden Emissions of an incinerator,* http://bit.ly/2Q2lRJS
5. Zafeiraki et al, 2016, *Chemosphere 144*, 2106–2112
6. Sakurai T et al, 2003, *Chemosphere 53*, 619–625
7. Measurement report REC, Harlingen, Netherlands, 21.08.2017, *TÜV Report No.: 936/21239402/A* Cologne
8. Arkenbout A, Sarolea HA, (2018), *poster Dioxin2018, http://bit.ly/2zZrBt5*
9. Cheruiyot, N. K. et al. (2016), *Aerosol Air Qual. Res 16*, 2965-2988.
10. Neuwahl, F. and G. Cusano (2018): *Waste Management 8***,** 15-24
11. De Fre, R. and M. Wevers (1998). "Underestimation in dioxin emision inventories." Organohalog Compd 36 (1998): 17-20
12. Stockholm Convention. (2008): *Guidelines on Best Available Techniques and Guidance on Best Environmental Practices Relevant to Article 5 and Annex C of the Stockholm Convention.*