Emission regimes of POPs of a Dutch incinerator: regulated, measured and hidden issues

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Introduction

In 2011 a 'state of the art' waste incinerator was built in Harlingen, The Netherlands, a strict permit of a maximum 0,01 ng TEQ/Nm3 for PCDD/Fs emissions was given. In 2013 NGO ToxicoWatch found high concentration PCDD/Fs/ dl-PCBs in eggs of backyard chicken in the surroundings of the incinerator. In August 2015 a continuous sampling program of flue gases for dioxin monitoring was implemented, but in December 2017 the admission permit for this long-term sampling program was terminated by plant management (for unstated reasons), neglecting the wish of both the Dutch government and the concerned population to continue publicly controlled monitoring. The present research shows ongoing underestimation of dioxins emissions.

Materials and Methods

The waste incinerator is a Waste-to-Energy installation, a *Reststoffen Energie Centrale* (abbreviated as REC). The location is at Harlingen, The Netherlands. A program of continuous sampling of flue gas was performed by Environnement, France with the AMESA (Adsorption MEthod for SAmpling of dioxins). The measuring technique is explained in an article of Reinmann (2006) [1]. Analyses of POPs on PCDD/F/dl-PCBs are performed by Eurofins, Hamburg, Germany. The official mandated dioxin emission control, a short term sampling (1 x 6 hours/year), are performed by Promonitoring, Deventer, The Netherlands. Start-up measurements by ODRA (OmgevingsDienst Regio Arnhem), The Netherlands, analyses by Al-West, Deventer, The Netherlands.

Results and discussion

The following is a summary of the findings reported in full in Arkenbout & Esbensen (2017) [2]. The incinerator REC received a very stringent emission limit of 0,01 ng TEQ/Nm3 (EU-norm is 0,1 ng TEQ/Nm3). The plant was found to exceed the limit of 0,1 ng TEQ/Nm3 six times in start-up events, but this 'posed no legal problem since the regulation are stipulated to apply only for steady state operations'. In October 2015, a conglomerate of events produced a significant emission 0.17 ng TEQ/Nm3, exceeding the general European standard of 0.1 ng TEQ/Nm3, indeed exceeding the local licensed emissions of 0.01 ng TEQ/Nm3 by a factor of 17. AMESA was shut down during this failure for more than 10 hours, instead only sampling the *tailing* of emissions.

Figure 1 shows the results of a **18,748** hours long-term sampling of PCDD/Fs from Augustus 2015 till December 2017, revealing that excess emissions are not exceptional ("outlier events"), but rather constitute a regular feature of the REC incineration operations.

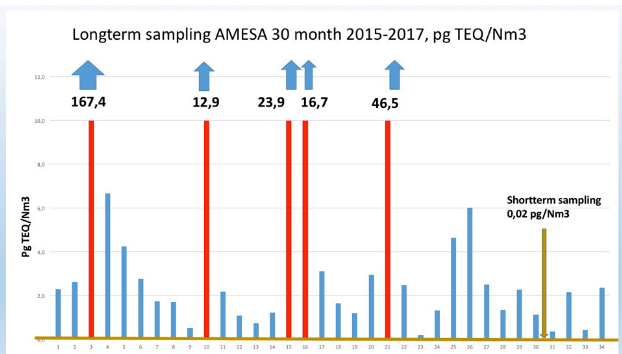

Figure 1: Results 30 month AMESA long-term sampling PCDD/Fs, REC Harlingen Note numerous excursions above the legal threshold limit. Horizontal line (yellow) shows the 0,01 ng TEQ/Nm3 emission limit.

Continuous sampling like the AMESA approach is not obligated, so recourse to short-term sampling was considered as an acceptable, much less demanding approach. However AMESA measurements were performed at the same time as the long-term sampling took place, so comparison is feasible (Table 1).

From Table 1 it is seen that short-term sampling is severely *underestimating* emission dioxin levels by factors between 460-1290 in TEQ per Nm3 flow. Sampling for official monitoring purposes must be *representative* (both technically as well as administratively, i.e. unannounced controls etc.). However, the current regimen of regulatory flue gas sampling is pre-announced.

Sampling	hours	ng TEQ/Nm3	Factor
Short-term, April 2016	**6**	**<0,00001**	
Long-term April 2016	**256**	**0,01290**	**1290**
Short-term, 8 March 2017	**6**	**0,00001**	
Long-term March 2017	**690**	**0,00460**	**460**

Table 1: Comparison of parallel short- and long-term measurements (assumed flow: 230.000 Nm3)

The current short-term sampling is only representing ~0.2 % of of the total yearly operating time. This can under no assumptions and conditions be considered representative for real dioxin emissions of the incinerator. Sampling in only 0,2% of the yearly operating periods makes for a likely enormous sampling error.

Many researchers (Reinmann 2009, Tejima 2007, Hung 2016, Wang, 2016, Li 2018) [1, 4,5, 9, 10] demonstrate high releases of dioxins during start-up and shut-down of incinerators. Estimations of the dioxin emissions range from 40% till 200% times the annual TEQ allowed, depending on the type of incinerator, by-pass use and dust emissions. Emissions of dust, most of the time unfiltered, are accepted during start-up procedures, for cleaning the installation before waste feed may start. In the literature this is known as a "filter bypass mode", "abatement bypass" or "dump stacks". The UK (2009) [7] regulations suggests that this is only acceptable once a year. Most of the time, bypassing (dump stacks) takes only a few minutes.

In the REC the procedure for bypassing is when the stack comes under the 140^0 or below the lower limit of gas flow, the bypass will be activated in order to protect the filter cloth. Bypassing will also be activated when the temperature goes above 210^0 for fire prevention. Concentration of PCDD/F in dust deposit is much higher than fly ash in steady state; 0,01 mg TEQ/kg dust (Tejima 2006) [4], this study 0,005 – 0,009 mg TEQ/kg. Many plants are currently operating during start-up with one or more bypasses flue gas cleaning devices or even of the entire system to avoid technical problems such as bonding of used sorbents at the fabric filter at low temperatures (Kriekouki 2018) [8]. After May 2017 the incinerator will not use bypasses anymore over bagfilter and $DeNO_{x,}$; they adapt the program of start-up.

Shutdown and start-ups are not exceptional occasions during annual maintenance stoppages, it's rather a *regular* feature of normal incineration procedures. In the US, start-ups and shutdown emissions, as well as bypass emissions, are known as 'excess emissions', a category of air pollution that has received little attention in the research literature (Zirogiannis 2018) [6]. In the program of long-term sampling, a total of 12 start-ups and shutdowns events were observed. Hung (2016) [5] reports a frequency of 4.75 start-ups/year per unit in Taiwan (290 start-ups were counted for 61 incineration lines). This underlines that start-ups and shutdown are simply part of normal operations. From the first start-up of the REC in 2011 there has been registered more than 55 start-ups and shutdowns.

Hung (2016) applies an emission factor of 9.32 mg I-TEQ for a start-up. A conservative estimation of a start-up emission in the REC is about 2 mg I-TEQ, but comparison is hard to make because start-up definitions differ. The process of start-up procedure in the REC incinerator is partitioned into 5 stages: pre-flushing, flushing/cleaning, pre-heating, waste feed and regular phase (Table 3). Tejima (2007) [3] starts measurements after closing the bypass with the ignition of the side burners and stops after 12 days.

In 2016 and 2017 start-ups after the annual maintenance stops were *not sampled* by AMESA. The reason for this were remarkable "incidents" (explosions) as well as unclear communications. Substitute short-term sampling by ODRA could be performed in four of the 5 stages of the start-ups. PCDD/F during cleaning actions could not be measured in gas phase because lack of isokinetic flow sampling. Gravimetrical measurements of particulate dioxins could only be performed indicative and results in 180-340 mg dust/Nm3. Analyse of the 180 mg filter results gives a concentration of 1,7 ng TEQ/Nm3.

Phase	Description	2016	2017
	Annual stop, AMESA off-time	408 hours	571 hours
Phase 1	**Pre-flushing (annual report)**	25-50 kg	?
	Pre-flushing 2 (ODRA)	Mention	25-50 kg
Phase 2	**Flushing**		
	Dust gravimetric (indicative) deviation EN 13284-1	180 – 270 mg/Nm3	340 mg/Nm3
	Flow (data REC)	335.000 Nm3	217.058 Nm3
	Dust ODRA (Dust REC Durag)	60,3 kilo (11,7 kilo)	73,8 kilo (1,8 kilo)
	Particulate bound PCDD/Fs (indicative)	1,6 ng TEQ/Nm3	1,7 ng TEQ/Nm3
	Gas phase PCDD/Fs (18 - 20 degrees)	--	--
Phase 3	**Heating up**		
	PCDD/F vapour/particulate	0,11 ng TEQ/Nm3	0,32 ng TEQ/Nm3
	Length of heating up	50 hours	32 hours
Phase 4	**Starting waste feed**		
		0,03 ng TEQ/Nm3	0,03 ng TEQ/Nm3
Phase 5	**Regular operation (after 3 days)**	0,007 ng TEQ/Nm^3 (6 hrs)	0,005 ng TEQ/Nm3 (672 hrs)

Table 2: Short-term measurements start-up after annual maintenance stoppage

Table 2 show difference between gravimetric measurement of 73,8 kilos and dust measurement by the incinerator of only 1,8 kilos. Emissions of dust occur as an acceptable part in start-up procedures for cleaning the installation before waste feed may start. In phase 3 (32-50 hours) the dioxins are exceeding 0,1 ng TEQ/Nm3, but this paradoxically poses "no legal problems" since "regulations only apply to steady state operations". It is very difficult to understand this kind of official reasoning, which certainly does not benefit the environment nor the population in the surrounding regions.

AMESA sampled for a continuous period of **18,748** hours, with an off-time of 1291 hours (6,8 %). Patterns of switching OFF of long-term sampling is found to match *systematically* start-up and shutdown procedures. When the ID-fan is shut-off and the velocity of the stack comes under the 1,5 m/s, the AMESA shut down lasts for several minutes. A leak test follows in the cartridge of the AMESA and restart after 3-4 minutes. Repetitive sessions of more than 10 terminations are noticed in start-up procedures and disable the AMESA for a long time and disrupt the continuous measurements significantly. It seems to be a *routine operation*, the patterns described above are also observed when no such events take place.

Interactions between industrial cleaning and monitoring sampling needs more research. From the start of the current studies, cooperation was agreed with the incinerator management, but still a lot of data are missing and some data could only be obtained through court or juridical procedures. This underlines the difficulty to do scientific work at incinerators, mainly because the huge economic interests and publicly sensitive information involved.

The present long-term sampling started in 2015 after findings of the highly polluted eggs in the environment of the REC incinerator. The ongoing study tries to find an answer at the question if there's a quantitative and qualitatively relationship between dioxins in the flue gas and in the eggs. A study of Hoogenboom (2015) [3] conclude dioxins pollution in eggs of backyard chicken are to be everywhere nowadays, but finding more 'clean' eggs (<2,5 pg TEQ /g) in location Rotterdam with far more heavy industries than in Harlingen points to the particularly impacting pollution potential of *uncontrolled* incinerators. All eggs sampled in Harlingen (within a radius of 2 km) were found to have concentrations above the 2,5 pg TEQ/g, while 50% of the eggs in Rotterdam (as well as in the remaining parts of all of the Netherlands) were below the limit of 2,5 pg TEQ/g [2].

Conclusions

Dioxin emissions are continuing to be underestimated, not infrequently involving very large excursions. Regulatory short-term measurements underestimate dioxin emissions structurally due to administrative and duration idiosyncrasies. The most modern 'state-of-the-art' incinerator of the Netherlands REC has to deal with many problems in relation to dioxin emission reduction approaches. On the positive side, dust-emissions are diminished since AMESA monitoring. However a serious remaining problem with incinerator management is communication and enforcement so currently there is no guarantee that bypasses will not be used, and intense cleaning will be continued.

The officially mandated dioxin emission *pre-announced* control of 6-8 hours (2 times a year) is a clear example of *grab sampling* in the time domain which must be replaced by an appropriate scheme of *long-term sampling*. Long-term sampling is a step ahead to apply as standard in all facilities where dioxin emissions are possible in order monitor, and subsequently to reduce/eliminate emission of these hazard POPs into the environment. AMESA demonstrated a significantly enhanced public trust. Special focus on interruptions is needed (AMESA). Several events can interfere with, or even block monitoring programs. This should be *avoided*, continuous measurements with the AMESA is a good help and must not blocked.

Eggs from backyard chickens have proved to be excellent biomarkers of dioxin pollution. In the North of the Netherland the smallest national incinerator, could emit dioxins *unfiltered* into the environment, no eggs from backyard chickens can be considered safe.

The current results raise important questions for future research on what can be accepted as *normal* operating – and monitoring- conditions for incinerator plants with respect to their potential effects on public health. The current studies show unambiguously that dioxins are *still* a serious issue, measurement programs show serious shortages, the health of the population is *still* under threat. There is unfortunately *still* a long way to eliminate dioxin emissions to the environment and to protect human and wildlife health.

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