

Hidden emissions: A story from the Netherlands

Case Study

November 2018 – ToxicoWatch

Although presented as state of the art, the youngest incinerator in the Netherlands is far from a clean: long-term tests reveal emissions of dioxin, furan and persistent organic pollutants far beyond the limits.

The case of the REC plant raises important questions for future policy-making concerning waste incineration and its potential effects on public health and the environment.

The youngest of Dutch incinerators: Reststoffen Energie Centrale

Out of the 13 waste incinerators currently in operation in the Netherlands, the Reststoffen Energie Centrale (REC) is the most recent one. The so-called waste-to-energy plant is located in Harlingen, bordering the UNESCO Wadden Sea coastline in the North of the Netherlands. When it was built in 2011, it was proudly announced by the Dutch Ministry of Economic Affairs as 'a state of the art' installation, the best in Western Europe. However, long-term testing revealed the plant emits dioxin, furans and toxic pollutants far beyond the limits set by EU laws.

Initially, in order to deliver energy to the nearby salt industry plant, the REC incinerator was only supposed to burn Frisian household waste. However, nowadays the waste input comes from everywhere in the Netherlands. Besides household waste, the REC waste input includes also industrial waste, digestate¹ and sewage sludge. Chemical analyses to check the waste input were first undertaken at the start in 2011. It is debatable whether this installation with a post combustion temperature of 850⁰ Celsius is actually capable of combusting the chemical complexity of current 'household' and industrial waste.

Environmental biomarkers and toxic eggs

In 2013, a study by ToxicoWatch found high concentration of dioxins and furans² in eggs of backyard chickens in the surroundings of the REC incinerator³ ⁴. Eggs of backyard chickens are sensitive environmental biomarkers for persistent organic pollutants (POPs) like dioxins⁵. All eggs of backyard chickens in Harlingen, sampled within a radius of 2 km from the REC incinerator, showed a much higher concentration of dioxine than allowed by the EU⁶. Notably, the concentration exceeded 1.7 BEQ/gram fat (Bioanalytical EQuivalent)⁷, and the 2.5 picogram TEQ/gram fat⁸ limit set by EU law.

² Polychlorinated dibenzo-p-dioxins and dibenzofurans, PCDD/Fs.



¹ Digestate is the material remaining after the anaerobic digestion of a biodegradable feedstock.

³Arkenbout, A, 2014. Biomonitoring of dioxins/dl-PCBs in the north of the Netherlands; eggs of backyard chickens, cow and goat milk and soil as indicators of pollution. Organohalogen Compd. 76, 1407–1410

⁴ Arkenbout, A, Esbensen KH, 2017. Biomonitoring and source tracking of dioxins in the Netherlands, Eighth World Conference On Sampling and Blending / Perth, Wa, 9–11 May 2017, 117-124

 ⁵ Witteveen en Bos, Dioxine emissie oktober 2015 – Verspreidingsberekeningen, 2015, rapport LW217-12/16-002.590
 ⁶ See n=6, Figure 1 black spot

⁷ The values are expressed in Figure 1 in BEQ because analyses are performed with the bioassay of DR CALUX.

⁸ TEQ stands for Toxic EQuivalent, picogram is a millionth of a millionth of a gram or 10⁻¹² gram

This means that potentially highly toxic dioxins exceed the maximum limit for consumption of eggs in the environment of Harlingen.

A subsequent national survey⁹ found 50 % of the backyard chicken eggs in the Netherlands were below the maximum limit for dioxins in eggs. However, around the incinerator (Figure 1) all eggs are exceeding the limit for dioxins of 2.5 picogram TEQ/gram fat¹⁰.

A study of dioxin depositions on grass in the direct surroundings of the REC incinerator (see Figure 2) confirms elevated values of dioxins. Moreover, the fingerprints of these dioxins found on grass comply with the congeners found in the flue gases of the incinerator¹¹, tracking the source of dioxin contamination to the emissions of the incinerator.

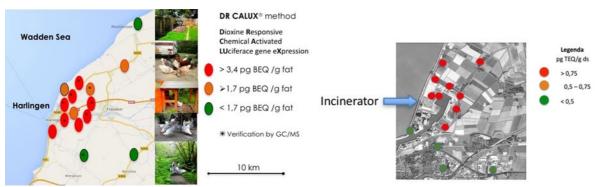


Figure 1: Results dioxins eggs backyard chickens



Dioxine emissions: long-term sampling reveal breaches

Long-term sampling is not mandatory for waste incineration facilities, that mostly rely on preannounced short-term sampling of 6-8 hours twice a year. After the alarming findings of dioxins in eggs of backyard chickens in the ToxicoWatch study, the local government decided, for the first time in the Netherlands, to perform long-term sampling of flue gases in the REC with the AMESA technique, which stands for **A**dsorption **ME**thod for **SA**mpling of dioxins¹². When short- and longterm sampling are carried out in the same period, remarkable differences become visible (Table 1). The results show that short-term sampling seriously underestimates actual dioxin emission levels by factors of 460 - > 1290 (Table 1). The current short-term sampling only represents ~0.2 % of the total yearly operating time, so short-term sampling cannot be considered representative for real dioxin emissions of the REC incinerator¹³.



 ⁹ Hoogenboom, RL, Ten Dam, G, van Bruggen, M, Jeurissen, SM, van Leeuwen, SP, Theelen, RM, Zeilmaker, MJ, 2016. Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) and biphenyls (PCBs) in home-produced eggs, Chemosphere, 150, 311–319
 ¹⁰ Arkenbout, A, Esbensen KH, 2017. Biomonitoring and source tracking of dioxins in the Netherlands, Eighth World Conference On

Arkenbout, A, Esbensen KH, 2017. Biomonitoring and source tracking of dioxins in the Netherlands, Eighth World Conference On Sampling and Blending / Perth, Wa, 9–11 May 2017, 117-124
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¹¹ Arkenbout, A, Esbensen KH, 2017. Biomonitoring and source tracking of dioxins in the Netherlands, Eighth World Conference On Sampling and Blending / Perth, Wa, 9–11 May 2017, 117-124

¹² Tejima H, Nishigaki M, Fujita Y, Matsumoto A, Takeda N, Takaoka M, 2007. Characteristics of dioxin emissions at startup and shutdown of MSW incinerators, Chemosphere 66, 1123–1130

¹³ Arkenbout, A, Olie K, Esbensen KH, 2018. Emission regimes of POPs of a Dutch incinerator: regulated, measured and hidden issues, abstract, http://bit.ly/2QQCmW1

Sampling	Hours	ng TEQ/Nm ³	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)

Figure 3 shows the results of a **20,139** hours long-term sampling of dioxins (PCDD/Fs) from August 2015 until December 2017, revealing that excess emissions ("outlier events") are not exceptional, but rather constitute a regular feature of the REC incineration operation. The results of long-term sampling clearly show the shortcomings of regulatory short-term measurements¹⁴.

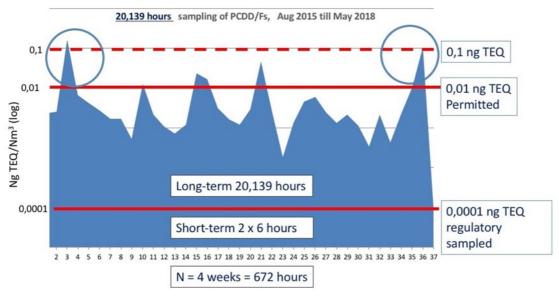


Figure 3: Results of 20,139 hours AMESA long-term sampling PCDD/Fs, REC Harlingen

Announced and presented as "State of the art" and applying with Best Available Techniques /Best Environmental Practices¹⁵, the REC incinerator has a very stringent emission limit of 0.01 ng TEQ/Nm³¹⁶. In Figure 3, a number of excursions above the legal threshold limit can be noted. The horizontal lines indicate from bottom to top the short-term measurements, emission limits set for the REC in the environmental permit, as well as in the permit by the Integrated Pollution Prevention and Control (which is now the IED, International Emission Directive¹⁷). A total number of 12 start-up and shutdown events occurred in the measuring period. The permitted limit of 0.01 ng TEQ/Nm³ was exceeded seven times, and the IED standard of 0.1 ng TEQ/Nm³ twice. As the exceeding of dioxin



¹⁴ Idem

¹⁵ Guidelines on Best Available Techniques and provisional guidance on Best Environmental Practices, relevant to article 5 and Annex C of the Stockholm Convention on Persistent Organic Pollutants, 2007, United Nations Environment Programme

¹⁶ The EU-norm is 0.1 ng TEQ/Nm3

¹⁷ In Dutch: RIE, Richtlijn Industriële Emissies

emissions occurred mostly during start-ups, this 'posed no legal problem' for the facility because the norms are stipulated to 'apply only to steady state operation'. From the very first start-up of the incinerator in Harlingen in 2011, more than 60 start-ups and shutdowns have been (officially) registered. In August 2015 the continuous sampling programme of flue gases for dioxin monitoring AMESA was implemented, but in December 2017 the plant management terminated this long-term sampling program for unstated reasons. With this decision, the management ignored the wish of both the local government and the concerned population to continue AMESA monitoring.

Hidden emissions

One of the reason why the REC incinerator exceeds the dioxins permit levels is the use of bypasses during transient phases, which means that the incinerator emits *without filtering* (Figure 4). In the technical literature this is known as a 'filter bypass mode', 'abatement bypass' or 'dump stacks'. The bypass mode is *structurally* programmed whenever elevated dust emissions occur. Although the plant management had recently promised to stop using bypasses, data don't confirm this has actually happened.

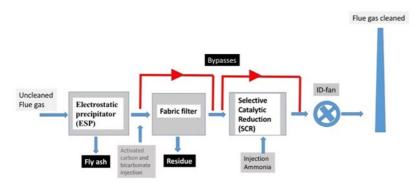


Figure 4: Block diagram flue gas cleaning REC Harlingen with bypasses

Unfortunately, even AMESA cannot perform continuous sampling during transient phases. In all breaches of the permit emission limit (see Figure 3) the long-term sampling by the AMESA was found to be incomplete. During the first outlier event in Figure 3 (exceeding 0.1 ng TEQ/Nm³), the long-term sampling was interrupted for 10 hours, and for more than 200 hours during the last outlier event. During the 20,139 hours of long-term sampling of the REC incinerator, AMESA was off-line for 1,496 hours¹⁸. While AMESA is mostly on-line (93% of the time), dust emissions especially occur when AMESA is off-line. During start-ups the ID-fan (see Figure 4) is regularly turned off, which results in a shut down and a restart of the AMESA, suspending the test for 3 minutes. When this process is repeated, the long-term sampling will be disabled for a certain time.



¹⁸ Arkenbout, A, Bouman KJAM, 2018. Emissions of dl-PCB, PBB, PBDD/F, PBDE, PFOS, PFOA, and PAH from a waste incinerator, poster Dioxin2018, http://bit.ly/2RZJe3j

Start-ups with no filter

Most studies of 'start-ups', including the AMESA long-term sampling, begin to measure when the waste feed is started (see Table 2, phase 4). Data of dioxin emissions before the waste feed starts are less prominent in the literature, but all show elevated dioxin emissions during phases where no waste is burnt ¹⁹ ²⁰ ²¹ ²² ²³ ²⁴ ²⁵.

In this study, gravimetrical and short-term measurements were performed in the phases before waste combustion in Phase 4 starts. The measurements in Phases 2 (flushing) and 3 (heating up) were performed by the governmental organisation ODRA in 2016, 2017 and 2018. The results show some remarkable elevated dioxin emissions in Phase 1, 2 and 3 of the start-up process.

Table 2 describes the different phases of a cold start-up. This means the installation is already several days inactive and stabilized at room temperature. In phase 3, lasting between 32 and 50 hours, the system is heating up from $15-25^{\circ}$ to 850° Celsius, which is the legal binding temperature at which waste can be put on the grate. In this phase, short-term measurements of 4 to 6 hours show all dioxin emissions in access of the IED limit of 0.1 ng TEQ/Nm³.

Phase 1	Pre-flushing	- Hereit
Phase 2	Flushing (cold)	315/_
Phase 3	Heating up	
Phase 3B	Flushing (hot)	
Phase 4	Starting waste feed	
Phase 5	Regular operation	

Table 2: Phases of start-up

Figure 5: Emission of dust (dumpstack)

In Phase 2, no short-term measurements are possible, and dust can only be measured by gravimetric methods. Figure 6 shows how an indicative dust load of 73 kg was found in 83 minutes' measure-time, while the incinerator only declared 2 kg dust during this period. Figure 6 clearly shows that the dust emission lasts only 3 minutes. The dust meter of the incinerator is unable to

hazardous waste incinerator under transient conditions, R. Soc. open sci. 5: 171079



¹⁹ Tejima H, Nishigaki M, Fujita Y, Matsumoto A, Takeda N, Takaoka M, 2007. Characteristics of dioxin emissions at startup and shutdown of MSW incinerators, Chemosphere 66, 1123–1130

²⁰ Hung PC, Chang SH, Buekens A, Chang MB, 2016. Continuous sampling of MSWI dioxins, Chemosphere 145, 119-124

²¹ Wang L-C, His HC, Chang JE, Yang XY, Chang-Chien GP, Lee WS, 2007. Influence of start-up on PCDD/F emission of incinerators, Chemosphere 67, 1346–1353

²² Chen CK, Lin C, Lin YC, Wang LC, Chang-Chien GP, 2008. Polychlorinated dibenzo-p-dioxins/dibenzofuran mass distribution in both start-up and normal condition in the whole municipal solid waste incinerator, Journal of Hazardous Materials 160, 37–44
²³ Li M, Wang C, Cen K, Ni M, Li X. 2018, Emission characteristics and vapour/particulate phase distributions of PCDD/F in a

²⁴ Zirogiannis N, Hollingsworth AJ, and Konisky DM, 2018. Understanding Excess Emissions from Industrial Facilities: Evidence from Texas, Environ. Sci. Technol., 52 (5), pp 2482–2490

²⁵ Witteveen en Bos, Dioxine emissie oktober 2015 – Verspreidingsberekeningen, 2015, rapport LW217-12/16-002.590

record excessive flows of dust in a short time. In Phase 1 the REC incinerator estimates the amount of dust emission to be 25-50 kg, but due the incapability of dust emissions meters (only 2% in Phase 2), the real quantity of dust emissions will be much higher. Dust emissions during start-ups without burning waste are structurally emitted without filtering. This has an economical reason: changing of filters, especially the bag or fabric filter is an expensive operation. Although emitting without filtering is prohibited, this practice occurs as a standard. As regards enforcement, penalising breaches is difficult, because emissions are only measured when waste is actually on the grate, and the bypassing system is still being applied (see Figure 5, which likely indicates emissions of dust saturated with dioxines and polycyclic aromatic hydrocarbon (PAHS)).

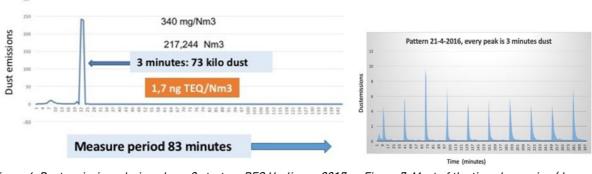


Figure 6: Dust emissions during phase 2 start-up REC Harlingen 2017

Figure 7: Most of the time, bypassing (dump stacks) takes only a few minutes

Although the AMESA test was prepared for operation during the start-up after the annual maintenance stop, it was blocked three times for unstated reasons. According to documents of the REC incinerator, several cleaning operations (flushing) have taken place in Phase 1, but without filtering. Sometimes this cleaning and dust emissions was visible (Figure 5), but most of the time these dump stacks took place at night. In Figure 7, regular patterns of 3-minute dust emissions are shown, just as a result of opening and closing the bypasses.

An exact number of dioxin emissions during start-ups is hard to give, but estimates are 5-10 mg dioxins for one cold start-up event. Annual emissions of the REC incinerator are estimated at around 5 mg dioxins during normal condition²⁶. More often start-ups occur without cooling down. An example is the first calamity in Figure 3, with an uncontrolled combustion of 19 tons of undefined waste, during which AMESA was off-line for more than 10 hours. An official conservative estimate of dioxin emissions is 33 mg²⁷, but this figure is probably much higher, since the waste was wet²⁸ and likely to have a Polyvinyl Chloride, PVC, content above 2% because of an impossibility of preseparation of PVC). Hot start-ups occur more often than cold start-ups, and these are also being sampled incompletely by AMESA, simply because the cartridge comes in a reset loop and interrupts the sampling. Problems with uncontrolled combustion happened several times in 2018, even the local fire brigades had to intervene, and the plant management seems not to be in control.



²⁶ Witteveen en Bos, Dioxine emissie oktober 2015 – Verspreidingsberekeningen, 2015, rapport LW217-12/16-002.590

²⁷ Idem

 $^{^{\}rm 28}$ Information provided by an internal source

Nonetheless, the REC incinerator would be able to defend these emissions during transient stages in courts, since regulations 'only apply to steady state operations' and exclude failure events. It is very difficult to understand this kind of official reasoning of enforcement, which certainly does not benefit the environment or the local population's health.

Breaches in the post-combustion zone

The IED, Directive 2010/75/EU²⁹, requires that the flue gases of a waste incinerator have a residence time of 2 seconds at 850°C in the post combustion zone under homogeneous conditions. Measurements in 2017 (6 years after the start in 2011) by TÜV Rheinland Energy Gmbh³⁰ indicate that **the REC incinerator in Harlingen does not comply with this requirement of homogeneity of temperature and oxygen in the post-combustion zone**³¹. The enforcement of these conditions should be more stringent, to ensure the requirements are fulfilled according to guidelines of the Best Available Techniques (BAT) and the Best Environmental Practices (BEP) ³². Moreover, the management of the REC plant violates the guidelines in article 5, Annex C of the Stockholm Convention^{33 34} on persistent organic pollutants, and notably the measures to reduce or eliminate releases of unintentional production. Moreover, the management of REC incinerator also acts in violation of article 10 of the Stockolm Convention³⁵, concerning public information, awareness and education, by refusing to disclose data on combustion temperatures, thus raising questions about the capacity of sufficient destruction of unintentionally produced persistent organic pollutants (POPs).



²⁹ DIRECTIVE 2010/75/EU OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 24 November 2010 on industrial emissions (integrated pollution prevention and control), p. 41

³⁰ Measurement report REC, Harlingen, Netherlands, 21.08.2017, TÜV Report No.: 936/21239402/A Cologne

³¹ Arkenbout, A, Sarolea, HA, 2018. Temperature and Oxygen levels in the post-combustion zone of a Waste-to-Energy Incinerator, poster Dioxin2018, http://bit.ly/2zZrBt5

 ³² Guidelines on Best Available Techniques and provisional guidance on Best Environmental Practices, relevant to article 5 and Annex C of the Stockholm Convention on Persistent Organic Pollutants, 2007, United Nations Environment Programme
 ³³ REGULATION (EC) No 850/2004 OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL, 29 April 2004 on persistent organic pollutants and amending Directive 79/117/EEC

³⁴ Stockholm Convention on Persistent Organic Pollutants (POPs) as amended in 2009,

https://www.env.go.jp/chemi/pops/treaty/treaty_en2009.pdf

Unintentionally produced persistent organic pollutants (UPOPs)

As the long-term sampling programme AMESA was extended to analyse other UPOPs^{36 37} in the flue gases, results pointed strongly to incomplete combustion in the REC incinerator. Notably:

- Near the incinerator, dioxin-like polychlorinated biphenyls (DI-PCBs) were dominantly found in eggs, milk, grass and soil, especially PCB 126. The coplanar dI-PCBs were prominent in the emissions of the incinerator, 8,5% of the total TEQ (n = 36, 20,139 hours), while other incinerators emit 3 times less dI-PCBs³⁸.
- Polybrominated diphenyl ethers (PBDEs) were detected during start-ups and shutdowns. In October 2015, 0,434 ng PBDE/Nm³ were measured when the waste supply was blocked and the waste ignited. In 2018, several similar fire calamities took place, but no data of UPOPs exist because AMESA measurements were stopped.
- Brominated dioxins (polybrominated dibenzodioxines and furans, PBDD/Fs) were detected during start-ups and shutdowns: 5,4 – 8,9 picogram PBDE/Nm³, indicating incomplete combustion of brominated flame retardants³⁹.
- 4. Polybrominated biphenyls (PBBs) were detected during steady state conditions with concentrations of 0,038 0,133 ng/Nm³. Normally these compounds should decompose above 300⁰ Celsius, and the presence of these substances indicate incomplete combustion.
- 5. Near the incinerator, the rain is regularly polluted with black particles. A CALUX screening test shows high concentrations of benzo(a)pyrene in black deposits on windows and roofs. Although the incinerator should not emit PAH at all (and the REC incinerator has, therefore, no PAH emission licence), all samples (n = 3), during steady state condition, were found to be positive with a PAH concentration of 2,4 314,8 ng/Nm³ in the flue gases⁴⁰.
- 6. Fluorinated compounds as perfluorooctanoic acid (PFOA) was detected in all (n = 6) samples (433 794 hours, total 3,929 hours)⁴¹. PFOA should not be detectable at all in modern waste incineration processes. Finding of PFOA in the stack can be an indicator of uncomplete combustion, i.e. not complying with a minimum 2 seconds residence time at 850 °C.

While these facts provide a conservative estimate of UPOPs-related pollution in the area, the actual impact would be much higher, as sampling is interrupted when dust emissions occur.

The finding of such a broad scale of UPOPs signals incomplete combustion, probably caused by insufficient homogeneous temperatures and oxygen levels in the after combustion zone and improper use of the bypasses.



³⁶ Arkenbout, A, Esbensen KH, 2017. Biomonitoring and source tracking of dioxins in the Netherlands, Eighth World Conference On Sampling and Blending / Perth, Wa, 9–11 May 2017, 117-124

³⁷ Arkenbout, A, Bouman KJAM, 2018. Emissions of dl-PCB, PBB, PBDD/F, PBDE, PFOS, PFOA, and PAH from a waste incinerator, poster Dioxin2018, http://bit.ly/2RZJe3j

³⁸ Sakurai, T, Weber, R, Ueno, S, Nishino, J & Tanaka, M, 2003. Relevance of coplanar PCBs for TEQ emission of fluidized bed incineration and impact of emission control devices. Chemosphere 53, 619–625

³⁹ Bjurlid F, Polybrominated dibenzo-p-dioxins and furans: from source of emission to human exposure, Örebro University, Repro 12/2017, ISBN 978-91-7529-221-2

⁴⁰ Arkenbout, A, Behnisch P, 2017. PAHs depositions in the environment of a waste incinerator, http://bit.ly/2Tot84Y

⁴¹ Arkenbout A, 2018. Long-term sampling emission of PFOS and PFOA of a Waste-to-Energy incinerator, http://bit.ly/2FtsEro

Further research is needed to clarify the real impact of emissions of incomplete combustion. Also, whether the change of waste input could lead to an increased change for the occurring of calamities is an aspect that must be considered.

Conclusions and recommendations

The dioxin emissions of the so called 'state of the art' REC incinerator Harlingen continue to be underestimated, and frequently go far beyond the limits set by the environmental permit (0,01 ng TEQ/Nm³). On top of that, the regulatory short-term measurements structurally underestimate dioxin emissions.

The mandatory *pre-announced* controls of dioxin emissions must be replaced by an appropriate scheme of *long-term sampling*. When using approaches like AMESA for long-term sampling, special attention should be paid to interruptions in the monitoring, as it is key for valid long-term sampling to be continuous.

The broad scale of UPOPs emitted by the REC incinerator signals incomplete combustion, probably caused by insufficient homogeneous temperatures and oxygen levels in the after combustion zone, and improper use of bypasses.

In order to reduce emissions of UPOPs in the environment, a more stringent application and a better enforcement of the Stockholm Convention is highly recommendable. The temperature and the oxygen levels in the after-combustion zone should be monitored on-line and duly enforced during normal operation, and this also under the most unfavourable incineration conditions, as mentioned by the Stockholm Convention papers and the IED.

Dioxin emissions during transient stages of start-up and shutdown easily exceed annual emissions during steady state. All dioxin emissions should be taken into account, not only emissions during the ideal steady state operation. Also, excluding emissions that occur during transient stages from monitoring regulations should be stopped immediately.

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, with respect to 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 totally eliminate dioxin emissions to the environment.

ROPE

Acknowledgements

This case study is written by Abel Arkenbout, ToxicoWatch

ToxicoWatch is funded by citizens concerned about industrial pollution in their environment.

The Dutch government funded long-term sampling with the AMESA technique of the waste incinerator Harlingen, national egg monitoring, grass-soil research and TÜV temperature research REC.

ToxicoWatch

NGO ToxicoWatch⁴² is an organization dedicated to creating a safer and healthier world by advancing the science of toxicology and raising awareness about toxic hazards in the environment. ToxicoWatch researches persistent organic pollutants as dioxins, PCBs, PAHs and brominated/fluorinated compounds. Source tracking of POPs in the environment and advising on regional and national level between government and industry. NGO ToxicoWatch is a member of International POPs Elimination Network, IPEN⁴³.

Zero Waste Europe was created to empower communities to rethink their relationship with the resources. In a growing number of regions, local groups of individuals, businesses and city officials are taking significant steps towards eliminating waste in our society.

Case study by Abel Arkenbout Editors: Roberta Arbinolo, Janek Vähk and Yianna Sigalou Zero Waste Europe, 2018



Zero Waste Europe gratefully acknowledges financial assistance from the European Union. The sole responsibility for the content of this event materials lies with Zero Waste Europe. It does not necessarily reflect the opinion of the funder mentioned above. The funder cannot be held responsible for any use that may be made of the information contained therein.

⁴² Toxicowatch website: www.toxicowatch.org



⁴³ IPEN; www.ipen.org