TOXICO WATCH

Biomonitoring research on persistent organic pollutants in the environment of the Waste Incinerator REC, Harlingen, The Netherlands

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Report

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Thank you to Zero Waste Europe for enabling this research on persistent organic pollutants (POPs). We extend our gratitude to all the participants from the surrounding villages of Harlingen for their confidence in this research, allowing us to analyse their backyard chicken eggs, vegetation, fruit and sheep wool.

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Introduction

In 2013 ToxicoWatch (TW) initiated biomonitoring studies on dioxins in eggs of backyard chickens owned by private chicken coop owners in the vicinity of the recently built the REC Waste-to-Energy (WtE) incinerator in Harlingen in the Netherlands (2011). Equipped with a dry flue gas cleaning system, the incinerator was designed to emit no visible plumes except in cases of some operational disruptions. However, **TW analyses in 2013 revealed high dioxin levels in chicken eggs sampled within a 3 km radius of the waste incinerato**r. Upon validation of the results by the National Health Service of the Netherlands (NL), *Rijksinstituut voor Volksgezondheid en Milieu* (RIVM), a joint committee comprising the Dutch Health Service, management of the waste incinerator, and local governmental stakeholders was established. ToxicoWatch (A. Arkenbout) an independent toxicologist specialising in persistent organic pollutants (POPs), joined this committee in its full two years (2014-2016) of existence.

Figure 1 - The REC incinerator in Harlingen, the Netherlands

This committee approved the installation of AMESA for semi-continuous measurement of flue gases inside the chimney of the waste incinerator for a period of 2,5 years (2014 - April 2017). A list of concerns and issues were identified, including preventing emissions of uncleaned flue gases by closing bypass pipes structurally and implementing additional alarm systems for emergencies. However, after two years (2014 until the beginning of 2017), **when a major violation involving a dioxin eruption occurred once again, the management of the incinerator ceased cooperation without providing an end-evaluation report on the results of the semi-continuous measurements. Structural problems leading to frequent start-ups, temperature issues, excessive HCL emissions, and wet sewage sludge remained unsolved.** Recently, permits have been issued to burn more waste with significantly higher hydrochloric acid (HCl) emissions. Surprisingly, the 'state-of-the-art' REC WtE incinerator was even nominated as the greenest industry in the Netherlands. Since 2017, no semi-continuous measurements have been conducted, making a gap of 7 years. **Residents in Harlingen and the surrounding region, all located near the UNESCO Wadden Sea, rely on local enforcement for assurance.**

This report presents the initial results on dioxins, PFAS and PAH in biomatrices such as eggs of backyard chickens, vegetables, and fruits. In 2024-2025 research on these Substances of Very High Concern (SVHC) will be extended by ToxicoWatch to the environment around the REC WtE incinerator in Harlingen, the Netherlands.

Sampling biomonitoring method

The biomonitoring research incorporates biomarkers such as eggs of backyard chickens, eggshells, and a selection of vegetables like kale and cauliflower, as well as fruit - namely apples. Additionally, matrices of water, soil, roof dust, and sheep wool will be included in this research (2024-2025).

Regarding vegetation, the focus is on collecting evergreen leaves from trees and shrubs, as well as mosses (*Bryophyta*), from both private gardens and public nature domains. The research area for 2024-2025, will encompass the environment of six (6) surrounding villages within a 3 km radius centred on the REC waste incinerator . Samples will be taken at locations in Harlingen, Wijnaldum, Midlum, Sexbierum, Herbaijum and Kimswerd. Reference locations will be sampled in the northern province of *Friesland*, specifically in the villages Tzummarum, (9,5 km away, North), Zurich (8 km away, South), Winsum (15 km East) and in the province of *South Holland* (Warmond, South-West, 150 km away).

In this first analysis report of biomonitoring 2023-2024 Harlingen, the Netherlands, samples already analysed are taken from:

Eggs
In October 2023, TW sampled eggs of backyard chickens at three (3) private locations. Tzummarum, located 9,5 km from the incinerator, is a reference location in the area. Additionally, in the 'green heart' of the South Holland province, specifically in the village of Warmond at a historic estate with two private gardens, eggs were sampled from two locations to serve as a second reference point due to historical soil conditions. The results from these locations are still pending and are not included in this report. For egg samples Wijnaldum, Tzummarum, and Warmond, TW collected 6-10 fresh eggs from each location (November 2023 - February 2024), combined the contents (egg yolk and white) and stored them in an HDPE lab container in a freezer until lab they were analysed. The TW team conducted a questionnaire and a location inspection at each backyard chicken egg location to identify any confounding factors.

Fruit and vegetables
Fresh vegetables (kale and cauliflower) and fruit (apples) were harvested in autumn (October-November) 2023, weighing between 200–300 grams/sample, and placed in special HDPE-lab bags and stored in a cool, dry environment until lab analysis.

Vegetation
Mosses from reference location Warmond (150 km away in the Province of *South Holland*) were collected in February 2024. About 250 grams of fresh moss (*Bryophyta)* was stored directly in HDPE bags, and placed in a cool, dark, and dry environment. Vegetation samples, 200–300 grams of fresh pine needles and evergreen leaves will be collected in June-September 2024, as well as 200-300-gram fresh mosses (*Bryophyta*) from the roofs of sheds (May-June 2024) at the same locations as the egg sampling. Additionally, mosses (*Bryophyta*) growing in a rural open field near the UNESCO Wadden See dyke, will be

collected in May-June 2024. All vegetation samples will be placed in HDPE lab bags, and stored in a cool, dark, and dry environment until lab analysis.

Sheep wool
Sheep wool, weighing 200 grams, was collected from *Fries Melkschaap* sheep located less than 1,5 km from the waste incinerator (northeast). The wool was obtained by a professional sheep shearer in July 2023 and stored in untreated jute bags in a dry, cool environment. Samples from these bags were taken in March 2024 and stored in HDPE bags for laboratory analysis. A sheep wool sample of reference location in Tzummarum was collected and stored in February 2024. Results are pending and not included in this report.

Analysis methods

The collected samples undergo analyses for persistent organic pollutants (POPs) using both bioassays (CALUX) and chemical analysis methods. The targeted substances are PCDD/F/dl-PCB (dioxins), per- and poly-fluoroalkyl Substances (PFAS), Polycyclic Aromatic Hydrocarbons (PAH), and 6-14 heavy metals: Arsenic, Cadmium, Cobalt, Chromium, Lead, Nickel, Aluminium, Barium, Copper, Manganese, Mercury, Silver, Tin, and Zinc.

In this research, bioassay analysis methods are DR CALUX® for dioxins/furans (PCDD/F) and dioxin-like PCBs (dL-PCBs), PAH CALUX for PAH substances, and FITC-T4 for the PFAS. When results of DR CALUX exceed the EU Limits for eggs (1.7 pg BEQ/g fat for PCDD/F and 3.3. pg BEQ/g fat for the sum of dioxins (PCDD/F/dl-PCB), the analysis will be extended by chemical analyses for dioxins on which EU limits are 2.5 pg TEQ/g fat for PCDD/F and 5.0 pg TEQ/g fat for the sum of dioxins (PCDD/F/dl-PCB). The lab analyses are conducted by BioDetection Systems, Amsterdam, the Netherlands (NL), which is accredited under RvA L401.

Chemical analysis for PAH, PFAS and heavy metals is performed by Normec, Groen Agro Control, Delft, the Netherlands (NL), an accredited laboratory. PFAS chemical analyses employ LC-MS/MS for 24 PFAS, while heavy metals are analysed using ICP-MS.

Results

Eggs analysis results: dioxins

The results of **dioxin analyses with the DR CALUX bioassay at location Wijnaldum**, situated 2 km from the REC incinerator is **9.8 pg BEQ/g fat**. This level **exceeds the permissible EU limit for dioxins** (PCDD/F/dl-PCB) analysis results with the DR CALUX of 3.3 pg BEQ/g fat, **by a factor of nearly 300%**. In 2013 measurement by TW at this location recorded a value of 1.7 pg BEQ/g fat. The chemical analysis required for commercial eggs after surpassing the DR CALUX limit reveals a value of 4.4 pg TEQ/g fat for the sum 29 congeners of chlorinated dioxins (PCDD/F/dl-PCB), excluding the contribution of brominated

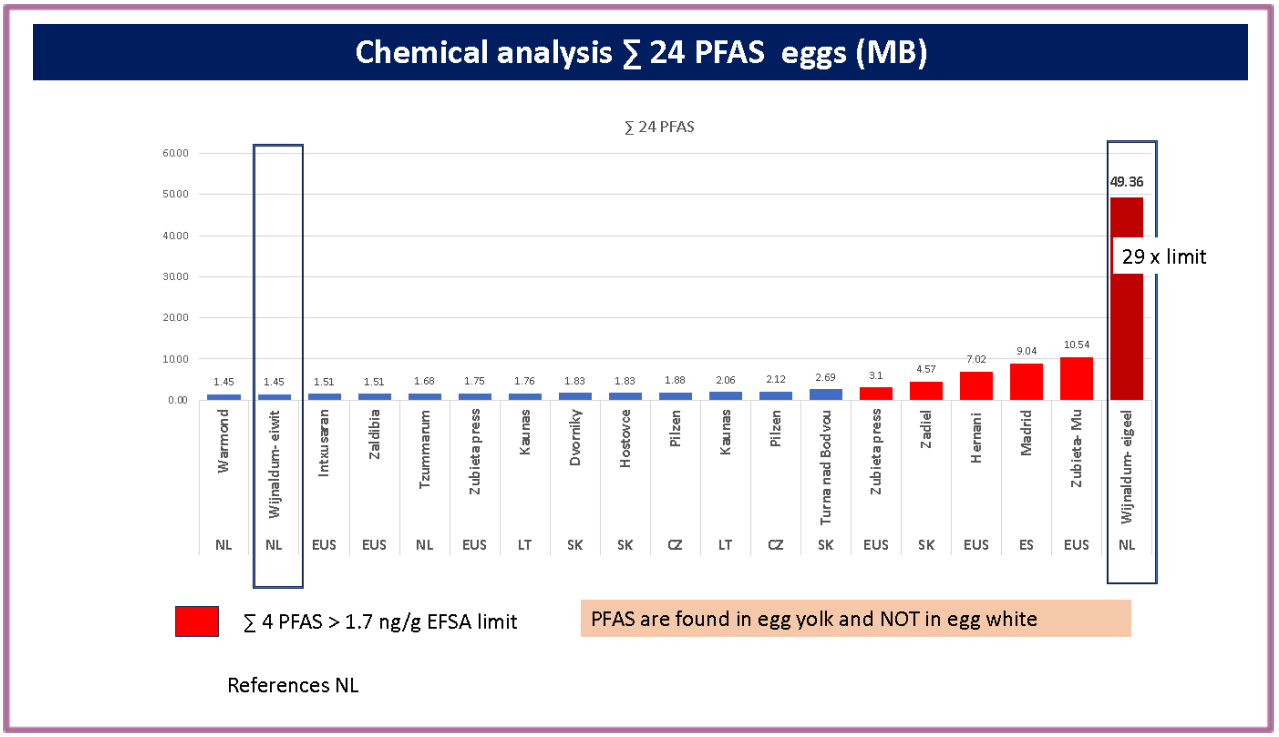
dioxins, which are not (yet) included in the mandatory analysis program for food and feed in the EU. **The result for the dl-PCB is 2.0 pg TEQ, exceeding the EU action limit for dl-PCB of 1.7 pg TEQ/g fat.**

Eggs analysis results: PFAS

In Wijnaldum, located just 2 km from the waste incinerator the **recorded PFOS level in 2023, of 38.4 ng/g fat, exceeds the EU limit for PFOS of 1 ng/g by a factor of 38. This result surpasses even the highest PFOS concentration found in eggs among 64 locations in the Netherlands** (Zafeiriki, maximum 24.8 ng/g, 2016)[1]. Further investigation is necessary to identify possible other confounding factors.

The concentration of PFAS measured in the egg yolk at location Wijnaldum is comparable to the high values recorded in the vicinity of the fluorochemical plant Antwerp, as well as the results obtained from the environment surrounding a fluorochemical plant in Wuhan (Wang et al. 2010).[2] In Wijnaldum, **11 different PFAS congeners were detected**, including PFOS, PFOA, PFNA, PFHxS, PFDA, PFUnDA, PFDoA, PFTrDA, PFTeDA, PFHpS, 6:2FTS.
At the reference sites in the Netherlands, specifically in Tzummarum, a village located 9,5 km from the incinerator, PFOS levels were measured at a relatively low amount of 0.28 ng/g. Conversely, at the reference location in Warmond (province SH), situated in the green heart of the Netherlands and devoid of waste incinerators in its vicinity, no PFAS was detected in eggs.

Figure 2 -Chemical analysis ∑ 24 PFAS eggs (MB)



[1] *Zafeiraki, E. et al. (2016). Perfluoroalkylated substances (PFASs) in home and commercially produced chicken eggs from The Netherlands and Greece. Chemosphere 144, 2106–2112.*
[2] *Wang, Y., et al., 2010. Distribution of perfluorooctane sulfonate and other perfluorochemicals in the ambient environment around a manufacturing facility in China. Environ. Sci. Technol. 21, 8062–8067.*

Fluorochemicals such as PFAS are notoriously resistant to destruction, even at temperatures ranging from 900 to 1100°C, which are typical in the Post Combustion Zone (PZC) of specialized hazardous waste incinerators, not for municipal waste incinerators, like most are, as well as the REC in Harlingen, with a performance of a maximum of 850 degrees°C. Accurately measuring temperatures in the PCZ remains a difficult task, with the evaluation status still questioned by the technical committee as of 2014 for the REC incinerator in Harlingen.[3] A review conducted by the National Institute for Public Health and the Environment in 2020 revealed that temperatures applied in the REC incinerator are still unknown.[4] **In other biomonitoring research conducted around waste incinerators, the trend of increasing PFAS concentrations towards the installations is concerning and warrants further research.**[5]

Fruit
No PFAS were detected above the limit of detection (> LOD) in fruits and vegetables at location Wijnaldum. However, this does not imply the absence of other PFAS compounds, as current analytical tools are not sensitive enough to detect the majority of PFAS. No dioxins could be detected above the limit of detection (>LOD) in fruit and cauliflower. Only, dioxin-like PCB (dl-PCB) with a concentration of 0.1 pg TCDD eq./g (DR CALUX), wet weight (ww) was found in fresh white cabbage at location Wijnaldum. The PAH concentration of a pooled sample of apples at this location was 1.70 ng Benzo(a)pyrene equivalent/gram. In cauliflower 3.10 ng Benzo(a)pyrene equivalent/gram product was found.

Heavy metals
Heavy metal analyses in eggshells reveal a high level of aluminium at location Wijnaldum. At the reference location in Warmond (province SH), lead (PB) and nickel (Ni) levels were found to be higher than those at the location in Wijnaldum. Additional analyses in soil samples will be performed to determine the possible deposition of heavy metals in this environment. The results will be compared with accredited research on persistent organic pollutants (POPs) and heavy metals from 2011, just before the incinerators started production.

Conclusion

The results of **dioxin analyses with the DR CALUX bioassay on eggs of backyard chicken, at location Wijnaldum**, 2 km distance from the REC incinerator , are **9.8 pg BEQ/g fat**. **This level exceeds the permissible EU limit** of 3.3 pg BEQ/g fat for dioxins (PCDD/F/dl-PCB) in eggs with the DR CALUX by a factor **of nearly 300%.** Furthermore, the result of PFAS concentration in egg yolk at the same location in Wijnaldum **is 38.4 ng/g fat** was comparable to those found near one of the biggest fluorochemical plants in other parts of the world. The **result of PFOS in this egg analysis (as a part of the EFSA 4 by the**

[3] *Arkenbout A.,Bouman KJAM. (2020). Hidden temperatures, Case study ToxicoWatch 2020.*
[4] *Bakker J. et al (2020). Per- and polyfluorinated substances in waste incinerator flue gases, RIVM report 2021-0143*
[5] *Arkenbout A., Bouman KJAM. (2022). The True Toxic Toll: Biomonitoring research on dioxins (PCDD/F and DL-PCB), PFAS and PAH.*

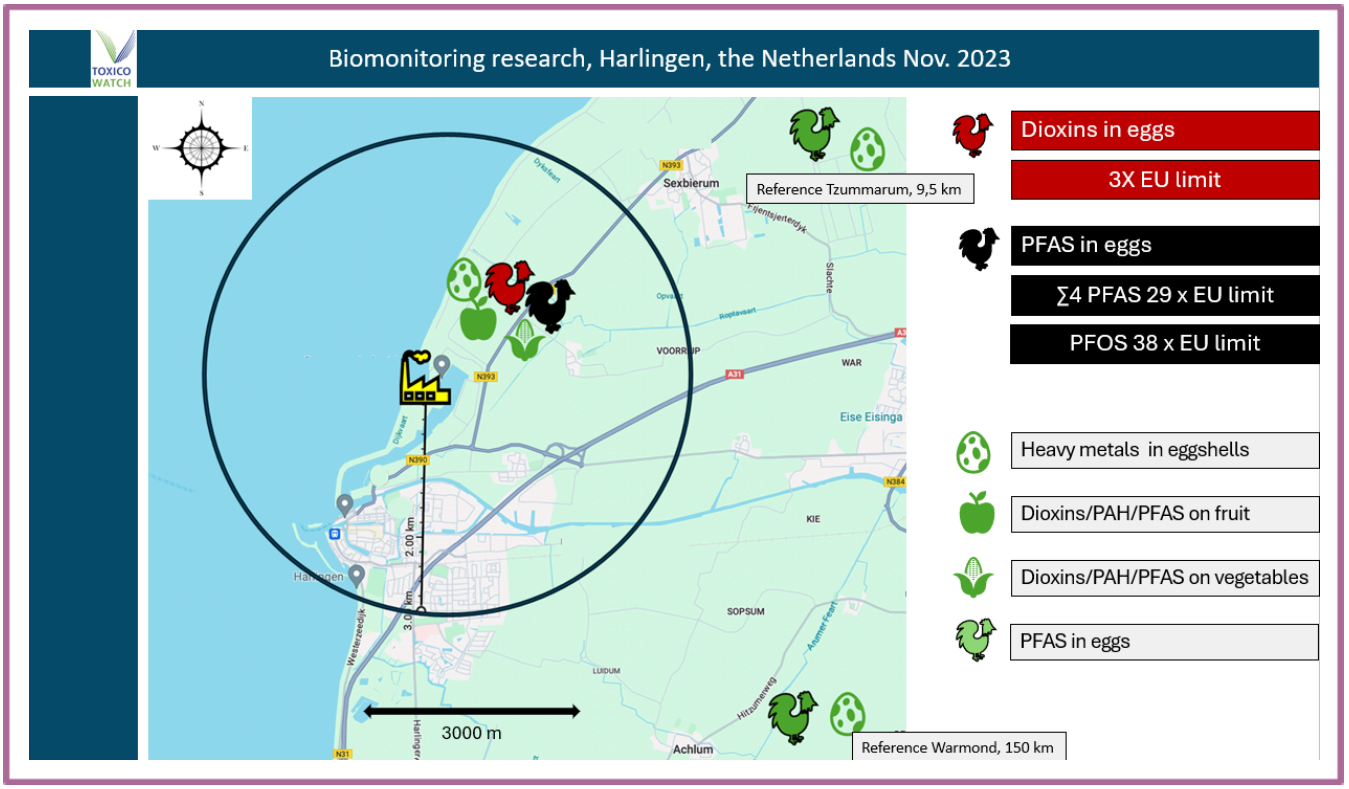
EU-regulated PFAS) exceeded the EU limit for PFOS of 1 ng/g, by a factor of more than 38. The Reference site of Tzummarrum shows PFOS 0.28 ng/g. and at the reference site Warmond, no PFOS could be detected. The PFAS result at Location Wijnaldum, highlights the impact of proximity to the incinerator. Further research is needed to identify additional contributing factors.

Unlike in the eggs, no PFAS could be detected with chemical analysis in fruits and vegetables sampled near the incinerator at the location of Wijnaldum. Because of the potential presence of undetected PFAS compounds, the next step will be the application of PFAS bioassays. Bioassay analyses on PAH, dioxins (PCDD/F/dl-PCB), could be detected in fruits and vegetables at levels below the EU limits for food.

Heavy metals found in the samples need to be researched in a broader context to understand the levels found in eggshells, mosses, vegetation, and fruit. Soil samples from reference locations will be compared with historical data to assess changes over time, providing insight into the impact of the waste incinerator on soil contamination of dioxins (PCDD/F/dl-PCB), and heavy metals.

Overall, these findings underscore the complex interplay between waste incineration, environmental contamination, and human health, necessitating continued research and monitoring efforts in 2024-2025.

Figure 3 -Biomonitoring research results, Harlingen, the Netherlands 2023



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