

Biomonitoring research Paris Ivry-sur-Seine, 2021





ToxicoWatch Foundation - 2021



Biomonitoring research Paris France, 2021

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www.toxicowatch.org

Abbreviations

Abbreviation	Meaning
APCD	Air Pollution Control Devices
BAT	Best Available Techniques
BEP	Best Environmental Practice
BEQ	Biological Equivalents
BMI	Body Mass Index
dl-PCB	Dioxin-Like Polychlorinated Biphenyls
DR CALUX®	Dioxin Responsive Chemical-Activated LUciferase gene eXpression
dw	Dry Weight
EFSA	European Food and Safety Authority
FITC-T4	Fluorescein IsoThioCyanate L-Thyroxine (T4)
GC-MS	Gas Chromatography Mass Spectrometry GC-MS
GenX	Group of fluorochemicals related to of hexafluoropropylene oxide dimer acid (HFPO-DA)
i-PCB	Indicator Polychlorinated Biphenyl
LB	Lower Bound; results under detection limit are set to zero
LOD	Limit of Detection
LOQ	Limit of Quantification
MB	Middle Bound; values are set as half the detection limit values
MWI	Municipal Waste Incineration
ndl-PCB	Non-Dioxin-Like Polychlorinated Biphenyl (Non-Dioxin-Like PCB)
ng	Nanogram; 10 ⁻⁹ gram
OTNOC	Other Than Normal Operating Conditions
РАН	Polycyclic Aromatic Hydrocarbons
РСВ	Polychlorinated Biphenyl
PCDD	Polychlorinated Dibenzodioxins
PCDF	Polychlorinated Dibenzofurans
PFAS	Per- and PolyFluoroAlkyl Substances
pg	Picogram; 10 ⁻¹² gram
РОР	Persistent Organic Pollutants
RPF	Relative Potency Factors
RvA	Dutch Accreditation Council
SVHC	Substances of Very High Concern
SWI	Solid Waste Incineration
TCDD	2,3,7,8-tetrachloordibenzo-p-dioxine
TDI	Tolerable Daily Intake
TEF	Toxic Equivalency Factor
TEQ	Toxic Equivalents
TOF	Total Organic Fluorine
тw	ToxicoWatch
TWI	Tolerable Weekly Intake
UB µg	Upper Bound (ub), results under detection limit are set as detection limit values. Microgram 10 ⁻³ gram
WtE	Waste to Energy (waste incinerator)
	Maste to Energy (Waste memeratory

Abbreviation	Dioxins, furans (PCDD/F) and dioxin-like PCBs	Toxic equivalency factor
	Congeners	TEF
Dioxins (n=7)		
TCDD	2,3,7,8-Tetrachlorodibenzo-p-dioxin	1
PCDD	1,2,3,7,8-Pentachlorodibenzo-p-dioxin	1
HxCDD1	1,2,3,4,7,8-Hexachlorodibenzo-p-dioxin	0,1
HxCDD2	1,2,3,6,7,8-Hexachlorodibenzo-p-dioxin	0,1
HxCDD3	1,2,3,7,8,9-Hexachlorodibenzo-p-dioxin	0,1
HpCDD	1,2,3,4,6,7,8-Heptachlorodibenzo-p-dioxin	0,01
OCDD	Octachlorodibenzo-p-dioxin	0,0003
Furans (n=10)	
TCDF	2,3,7,8-Tetrachlorodibenzofuran	0,1
PCDF1	1,2,3,7,8-Pentachlorodibenzofuran	0,03
PCDF2	2,3,4,7,8-Pentachlorodibenzofuran	0,3
HxCDF1	1,2,3,4,7,8-Hexachlorodibenzofuran	0,1
HxCDF2	1,2,3,6,7,8-Hexachlorodibenzofuran	0,1
HxCDF3	1,2,3,7,8,9-Hexachlorodibenzofuran	0,1
HxCDF4	2,3,4,6,7,8-Hexachlorodibenzofuran	0,1
HPCDF1	1,2,3,4,6,7,8-Heptachlorodibenzofuran	0,01
HPCDF2	1,2,3,4,7,8,9-Heptachlorodibenzofuran	0,01
OCDF	Octachlorodibenzofuran	0,0003
Polychlorinate	d biphenyl (n=12)	
PCB77	3,3',4,4'-Tetrachlorobiphenyl (#77)	0,0001
PCB81	3,4,4',5-Tetrachlorobiphenyl (#81)	0,0003
PCB126	3,3',4,4',5-Pentachlorobiphenyl (#126)	0,1
PCB169	3,3',4,4',5,5'-Hexachlorobiphenyl (#169)	0,03
PCB105	2,3,3',4,4'-Pentachlorobiphenyl (#105)	0,00003
PCB114	2,3,4,4',5-Pentachlorobiphenyl (#114)	0,00003
PCB118	2,3',4,4',5-Pentachlorobiphenyl (#118)	0,00003
PCB123	2,3,4,4',5-Pentachlorobiphenyl (#123)	0,00003
PCB156	2,3,3',4,4',5-Hexachlorobiphenyl (#156)	0,00003
PCB157	2,3,3',4,4',5'-Hexachlorobiphenyl (#157)	0,00003
PCB167	2,3',4,4',5,5'-Hexachlorobiphenyl (#167)	0,00003
PCB189	2,3,3',4,4',5,5'-Heptachlorobiphenyl (#189)	0,00003

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Key results

- DR CALUX analyses show high dioxin levels in 83% of eggs from backyard chickens.
- Chemical GC-MS analyses confirm breaches of EU limits for safe egg consumption.
- High amounts of dioxins are found in pine needles and mosses in the vicinity of the incinerator.
- The results of the analyses of dioxins in both eggs and vegetation are, according to ToxicoWatch biomonitoring studies, among the highest values found in Europe.

Introduction

The complexity of the chemical content of today's household and industrial waste presents a challenge for turning modern waste into energy in (WtE) waste incinerators. Even with the application of the most developed air pollution control devices (APCD), it is still a huge challenge to eliminate the multitude of persistent organic pollutants (POPs) in waste incinerator residues and flue gases. The dynamics of combustion processes and the inevitable emissions of toxic substances of very high concern (SVHC) into the environment is the main topic of ongoing research worldwide. Even in the most remote areas of the world, such as the Arctic (marine environment), toxic chemicals are found, which have been transported huge distances from industry in other parts of the world. Because of the transboundary behavior of persistent organic pollutants, international treaties are required to regulate, mitigate or even eliminate toxic chemical emissions. Loopholes still exist in national and international regulations, resulting in an underestimated registration of POPs.

Mandatory measurements take place twice a year during periods of 6 to 8 hours, according to European legislation (Directive 2010/75/EU) on emissions of waste incinerators. The maximum emission limit is 0,1 ng TEQ/nm3 for PCDD/F and it is the average of a sampling period of a minimum of 6 and a maximum of 8 hours. This regulation is based on chemical analyses of only a few chlorinated dioxins and furans, while many other persistent organic pollutants (POPs) remain outside the scope, such as dl-PCBs, brominated dioxins and perfluoroalkyl compounds (PFAS). The measurements in the chimney of waste incinerators are pre-announced and are only performed under optimal operating conditions. The explicit exclusion of OTNOC means these regulations hardly can reduce the emission of dioxins into the environment.

The limitations of the chemical GC-MS analyses could be overcome with the application of bioassays for measuring POPs even in the flue gases of an incinerator. Continuous monitoring of dioxins and other substances of very high concern in the chimney gives a far more accurate picture of the emission from combustion, especially when it is measured in the event of incomplete combustion as in exceptional operating conditions such as shutdown or start-up.

All over the world, there is growing public awareness and concern over the potentially toxic effects of POPs on human health and the environment. In particular, people living near waste incinerators need to be reassured about their health risks, short- and long-term exposure to incineration emissions, the safety of such combustion facilities, and compliance with regulations – not only under normal conditions, but also in other than normal operating conditions (OTNOC), such as shut-downs, start-ups, and failures.

ToxicoWatch (TW) aims to function as a bridge between people, science, and government when it comes to dioxins, POPs, and waste incineration. TW performs research on dioxins with a focus on a possible sources like waste incineration emissions by carefully selecting biomarker samples in an area. A sampling with focused matrices like distance, sample location and collecting information about the research area needs to be performed according to the theory of sampling (TOS) with references in the interest of the research. The biomatrices for this study are primarily backyard chicken eggs, pine needles, and mosses. The chemical analyses are expanded with innovative bioassays to investigate a broader spectrum of POPs such as dioxin-like PCBs, other (mixed) halogenated dioxins, PAHs, and PFAS.

WtE Waste incinerator lvry / Paris XIII

The lvry or lvry / Paris XIII incinerator is owned by the Syctom (agence métropolitaine des déchets ménagers, the Île-de-France metropolitan agency for household waste, located partly on the territory of the 13th arrondissement of Paris and in the municipality of lvry-sur-Seine. It is bordered by the railway line of the Paris-Austerlitz station, rue François-Mitterrand, and rue Victor-Hugo. An administrative entrance is on rue Bruneseau. Built in 1969, subsequently modernized in 1995 and 2005, and nowadays 2021 the largest waste incinerator in Europe. Ivry Paris XIII is located in a densely populated area in the capital city of France, Paris. Since the end of 2018, the entire site has undergone renovation work scheduled until 2023. The centre is managed by Syctom and operated by the Suez company.

With an authorized treatment capacity of 700,000 t / year, lvry-Paris XIII is the largest multi-sector treatment center in the region of Paris and the largest waste incinerator in Europe. It receives residual household waste from more than 14 municipalities in and around Paris. The heat produced by incineration is mainly used in the local urban heating network (owned by the Compagnie Parisienne du Chauffage Urbain - CPCU).



The lvry incinerator burns 100 tonnes of waste per hour by installing innovative technologies in the waste treatment sector. lvry-Paris XIII thus has a reception pit of 9,000 m³, equipped with two overhead cranes with grapples, two groups of furnace-boilers with a capacity of 50 tons/hour, in which waste is incinerated at a temperature of 900°C.

Figure 1: Ivry / Paris XIII incinerator Paris (Google Earth)

The current plant is now reaching the end of its life, Figure 1. Its operating life, around 40 years, cannot be extended beyond 2023. The aim is to transform the current, ageing center into a state-of-the-art facility. By 2023, the existing incinerator will be replaced by a new incinerator with a capacity reduced by half: 350,000 t / year against 700,000 t / year currently. This new unit, whose commissioning is scheduled for 2023, the expected end of operation date for the current equipment, is built on land adjacent to the current incinerator, which remains in operation until receipt of the new one.

Wind direction and depositions



Figure 2: Wind rose of Paris, France

The annual average wind direction in Paris is shown in Figure 2¹. This can be used as a model predict possible depositions by the to incinerator. The dominant wind direction is mainly South-West. The use of a wind rose for deposition modelling emissions from incineration processes is limited. Figure 3 shows on the left the dominant wind direction in Harlingen, the Netherlands (NL), South-West wind from the North Sea. On October 1st 2015, a major malfunction occurred at the WtE waste incineration plant, which was accompanied by prolonged emissions of black clouds that blew (North-East) in the direction of the UNESCO Wadden Sea on that particular day, Figure 3b. The city and region of Harlingen (NL) escaped

being hit by an enormous toxic cloud of dioxins. This example of a calamity in a waste incineration process illustrates the limitations of using annual average wind direction "safety models" to determine the load of emission depositions. Dense clouds of emission-loaded dust can and will occur during OTNOC situations like failures, shutdowns, and start-ups. TW research has proven that in just a few hours (during an incident, for instance), the amount of dioxins escaping from the incinerator can be much higher than the theorical amount calculated by the regulatory 12 hours (2x 6 hours/year, preannounced) measurement during normal operating conditions based on real emission data and not by provided re-calculated information data. Assuming the emission of dioxins is a discontinued process, calculation with average wind direction and speed is of little importance as large emissions can occur in a very short time frame. Figure 3c shows dioxin-contaminated eggs of a TW research around the WtE waste incinerator in Harlingen (NL).

Wind direction is an indication, but the deposition of emissions can differ completely when OTNOC and other parameters like coastline fumigation along seashores, or mountain ridges and valleys are included, as they should be. In a very short time, in a few hours or even in a few minutes, extremely polluted POP clouds of loaded dust can be emitted in whichever wind direction is dominant at that moment. This relativizes the use of average dominant wind directions in calculation models for POP emissions



Figure 3: Wind rose Harlingen (a), dioxin cloud during calamity, 2015 (b), contaminated eggs Harlingen (c)

¹ https://world-weather.info/archive/france/paris/

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Dioxins

Dioxins and furans are classified as highly toxic chemicals that have a serious effect on human health, causing cancer, diabetes, neurotoxicity, immunotoxicity, and chloracne. The emission of dioxins by incinerators was discovered in 1977 in the Netherlands². Although dioxins also can be formed by volcanic eruptions, forest fires, or other natural events, anthropogenic emissions are a much more common source of dioxins. Major sources of atmospheric dioxins (PCDD/Fs) include stationary emissions, especially from various types of incinerators, including secondary aluminum smelters, sinter plants, small-scale municipal solid waste incinerators (MSWI), medical waste incinerators (MWI), electric-arc furnaces, industrial waste incinerators, cement kilns, and crematoria. At the Stockholm Convention in 2004, 184 nations agreed to do their utmost to reduce the emissions of dioxins and other unintentionally produced organic pollutants. To achieve the goal of the Convention, Parties are required to implement the Best Available Techniques (BAT) and apply the Best Environmental Practices (BEP)³.

The term 'dioxin' refers to three groups of substances: polychlorinated dibenzo-p-dioxins, (PCDDs), polychlorinated dibenzofurans (PCDFs), and dioxin-like polychlorinated biphenyls (dI-PCBs). Figure 4 provides a schematic view where the black balls represent carbon atoms, the red oxygen, and the orange chlorine atoms (these can be substituted by other halogenated elements, like bromine, fluorine and iodine to form dioxins). The possible combinations with chlorine atoms (congeners) are 75 for dioxins (PCDDs), 135 for furans (PCDFs), and 217 PCBs congeners. Of these chlorinated congeners, 29 are found to be toxic and therefore regulated in EU; 7 PCDDs, 10 PCDFs, and 12 dl-PCBs. Only chlorinated dioxins and furans (PCDD/F) are regulated by EU for emissions of persistent organic pollutants (POPs) from waste incinerators. Dioxin-like polychlorinated biphenyls, brominated and mixed halogenated dioxins, all substances with dioxin-like properties, are (still) not regulated in the EU⁴.



Figure 4: Schematic overview of dioxins (PCDD/F/dl-PCB), © ToxicoWatch

² Olie K., Vermeulen P.L., Hutzinger O. (1977). Chemosphere 8, po 455 - 459

³ 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 (2008). Stockholm Convention on Persistent Organic Pollutants.

⁴ C. Budin et al. (2020). Chemosphere 251, 126579

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The EU sets limits of 2.5 pg TEQ/g fat for dioxins (PCDD/F) and of 5.0 pg TEQ/g fat for the sum of dioxin (PCDD/F/dl-PCB) for eggs. An EU action limit is set on 1.75 pg TEQ/g fat for PCDD/F and dl-PCB in eggs, see Figure 5. For bioassay DR CALUX the EU limits are 1.7 pg BEQ/g fat (eggs) for dioxins (PCDD/F) and 3.3. pg BEQ/g fat (eggs) for the sum of dioxins (PCDD/F/dl-PCB), see Figure 5.



Figure 5: EU regulations for dioxins (PCDD/F/dl-PCB), ©ToxicoWatch

Figure 6 displays the difference between the chemical analysis with GC-MS and the bioassay DR CALUX. GC-MS analyse specific compounds, while DR CALUX measures the total toxic effect of a mixture of dioxin-like activity.



Figure 6: Chemical GC-MS analysis of dioxins (PCDD/F/dl-PCB) vs bioassay DR CALUX analysis, ©ToxicoWatch

Emissions of waste incineration

In this biomonitoring research the focus will be on persistent organic pollutants (POPs) like PCDD/F, PXDD/F, PAH and PFAS. See red clouds in Figure 7. A central question in this research is whether waste incineration is a solution for waste disposal and energy production, when there is an unintentionally production and emissions of POPs, such as dioxins (PCDD/F/dl-PCB). Figure 7 shows the quantities of emissions per 100,000 tons of waste. This figure, is made up the configuration of the WtE waste incinerator REC in Harlingen, the Netherlands with the specific configuration of Air Pollution Control Devices (APCD) and specific waste input. A big difference in volume of mega-tonnage CO₂ and the relative tiny amount of the extreme toxic of dioxins, expressed in milligrams.

Although this research is mainly focus on the emissions of substances by air, which is only a small amount of the toxic substances, the main output are the incinerator residues, like fly and bottom ash. The processing, storage and sustainable application of toxic incineration residues is an environmental risk⁵. For more sustainability and a healthy environment the focus need to be on more recycling of waste materials. Important in this context, the production of non-toxic material in order to prevent (unknown) toxic recycling and with that to prevent a possible toxic greenwashed recycling waste tsunami in the future.



What are the real emissions & residues of a modern waste (WtE) incineration plant ?

Figure 7: What are the real emissions of WtE incineration? © TW

⁵ ToxicoWatch (2020). The hidden impacts of incineration residues, Zero Waste Europe

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Polychlorinated biphenyl (PCB)

Polychlorinated biphenyls (PCBs) are chemicals that were widely used in industrial processes from the 1930s until the late 1970s. PCBs were used extensively in many industrial applications, including fireresistant transformers and insulating condensers. The substances were used as heat exchanger fluids, and in aluminum, copper, iron, and steel manufacturing processing. PCBs were also used as plasticizers, in natural and synthetic rubber products, as adhesives, insulating materials, flame retardant, lubricants in the treatment of wood, clothes, paper, and asbestos, chemical stabilizers in paints, pigments, and as dispersing agents in formulations of aluminum oxide. PCBs were added in small quantities to inks, plastics, paints, sealants, adhesives, and dye solvents for carbonless paper. Although their production ended in 1979, huge amounts of PCBs are still in the environment⁶.

From a toxicological point of view, there is a significant difference between dioxin-like PCBs and nondioxin-like PCBs. Polychlorinated biphenyl congeners without chlorines in the ortho positions are called "coplanar" because the two phenyl rings can assume a planar state. This subgroup of 12 PCB congeners (non-ortho or mono-ortho chlorine substituted) with at least four chlorine substituents easily adopt a coplanar structure with toxicological properties similar to 2,3,7,8-Tetrachlorodibenzop-dioxin (TCDD), see Figure 8. This subgroup is termed dioxin-like PCBs (dl-PCBs) and are referred to as the 12 dioxin-like PCBs, see also Figure 5, 6. Due to their lipophilic properties and poor degradation, PCDD/Fs and dl-PCBs accumulate in the food chain and are persistent in the environment. Prevention or reduction of human exposure is best performed by source-directed measures, i.e., strict control of industrial processes to reduce the formation of dioxins. The greatest uncertainty with PCB and incinerator emissions lies in the composition of waste content and the distribution of PCB between air and waste. In a TW research conducted in the Netherlands, 10 % of the emissions in flue gases of the chimney of an incinerator were found to be dioxin-like PCBs⁷. However, in biomatrices around the incinerator, including eggs, milk and vegetation, the contribution of the TEQ of dl-PCB is often more than 50%⁸. More research is needed to confirm a direct relation to the emissions from a waste incinerator. PCB 126 was particularly dominant in all biomatrix samples.



Figure 8: dioxin-like PCB (dl-PCB) congeners

⁶ Petrlík J., Arkenbout A. (2019) Dioxins – The old dirty (dozen) guys are still with us www.researchgate.net/publication/332877688

⁷ Toxicowatch (November 2018). Hidden Emissions: A story from the Netherlands, a case study, Zero Waste Europe, https://zerowasteeurope.eu/wp-content/uploads/2018/11/NetherlandsCS-FNL.pdf

⁸ Arkenbout A, Esbensen K H. (2017) Sampling, monitoring and source tracking of Dioxins in the environment of an incinerator in the Netherlands, Proceedings Eighth World Conference On Sampling and Blending / Perth

Polycyclic aromatic hydrocarbon (PAH)

Polycyclic aromatic hydrocarbon (PAH) represent a class of ubiquitously occurring environmental compounds that are implicated in a wide range of toxicological effects. This class of compounds is known by their carcinogenic, mutagenic, and teratogenic properties. PAH leads to the development of a variety of disorders affecting all body systems as well as causing skin cancer and other skin diseases in animals and humans.

The PAHs with more than four (4) benzene rings have the most carcinogenic activity. PAH is able to reduce the effectiveness of measles vaccination through immunotoxicity to innate and adaptive immune cells⁹. Routine measurement of PAH contamination generally involves chemical analytical analysis of a selected group of representatives. The United States Environmental Protection Agency (EPA) and the European Commission (EU) classify 16 PAHs as priority pollutants (EPA-16): naphthalene, acenaphthylene, acenaphthene, fluorene, anthracene, phenanthrene, fluoranthene, pyrene, chrysene, benz[a]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene (B[a]P), indeno[1,2,3-c,d]pyrene, benzo[g,h,i]perylene, and dibenz[a,h]anthracene, see Figure 9. However, this will result in an underestimation of the PAH in a sample¹⁰. PAHs form a very large group of several tens of thousands (>10.000) of compounds when taking into account the attaching with halogens, hydroxyl or when a nitrogen atom can be in the place of a carbon atom in the ring. In this research a bioassay (PAH CALUX) analysis method is used to measure the total toxic effect of all toxic PAH in a sample. When measuring with a chemical (GC-MS) analysis on a pure sample with known PAH individual congeners, like benzo[a]pyrene, the results with a bioassay (PAH CALUX) analysis, are the same in measured values if the Relative Potency Factor (RPF) are taken into account. In environmental samples, like in this research, high levels of PAH are found, because the bioassay measures the total toxic effect of all present PAH in the sample. The results of a PAH CALUX analysis will be expressed in equivalent benzo[a]pyrene, a class 1B carcinogen.



Figure 9: Molecular structures of the most common PAHs (Hussain 2018)

 ⁹ Ruri Vivian Nilamsari et al. 2020. Polycyclic Aromatic Hydrocarbons (PAH) Reduces the Effectiveness of Measles Vaccination Through Immunotoxicity to Innate and Adaptive Immune Cells. Research J. Pharm. and Tech. 2020; 13(12):6128-6131.
¹⁰ Andersson J.T., Achten C. (2015). Time to Say Goodbye to the 16 EPA PAHs? Toward an Up-to-Date Use of PACs for Environmental Purposes - Polycyclic Aromatic Compounds, 35:330–354

PFAS

Per- and PolyFluoroAlkyl Substances (PFAS) are a class of man-made chemicals with a wide range of industrial and commercial applications, which has resulted in their ubiquitous presence in the environment. The consolidated PFAS list of EPA contains 6330 PFAS CAS-name substances, of which 5264 are represented with a defined chemical structure resulting in increasingly complex mixtures entering the environment. PFAS possess thermal, chemical, and biological stability, non-flammability, and surface-active properties. Their high applicability combined with chemical stability has led to an inevitable accumulation of PFASs in the environment and as a result to their detection in environmental matrices (air, sewage, rivers, and dust) in food products and food packaging, in drinking water, and also in human samples (breast milk, blood) PFAS are associated with adverse human health effects on thyroid function, metabolism (including overweight/obesity, diabetes, insulin resistance, and high cholesterol, foetal development, and the immune system¹¹. The risk of immunotoxicity for humans and wildlife cannot be discounted¹².



Figure 10: Overview figure of EU Commission Staff Working document on PFAS, October, 2020

According to the EU Commission Staff Working Document on Poly- and perfluoroalkyl substances (PFAS), October 2020, SWD(2020) 249 final, see Figure 10, "A recent opinion from the European Food Safety Agency (EFSA) concluded that both PFOS and PFOA are associated with reduced antibody response to vaccination. PFOS also causes a reduced resistance to infection". EFSA concluded that parts of the European population exceeds the tolerable weekly intake (TWI) from food of four PFAS.¹³

However, analysis techniques for PFAS are only available for a limited number of PFAS substances. Chemical (GC-MS) analysis are not capable to detect the currently known > 8000 PFAS congeners. Some substances are known to be present, these are called known unknowns, the substances that are not known to be present are called the unknown unknowns.

¹¹ Young, A.S. et al., (2021). Env. Health Perspect. 129 (4), 047010-1 to 047010-13.

¹² Corsini, E., et al. (2014). Perfluorinated compounds: Emerging POPs with potential immunotoxicity. Toxicol. Lett.

¹³ <u>https://ec.europa.eu/environment/pdf/chemicals/2020/10/SWD_PFAS.pdf</u>

It is a struggle for quality for laboratories to produce consistent data in PFAS analysis. Laboratories may suffer from multiple difficulties, which hinder clear identification of the error sources. The lack of analytical standards, the distinctive physical-chemical properties of the PFCs, and matrix effects, at every step of the analysis from sampling to detection is a common problem¹⁴. Therefore, in this biomonitoring study, a different analysis methodology is chosen to measure the PFAS in the biomarkers around a waste incinerator.

The used analysis method in this research is based on the competition between thyroid hormone (T4) and PFAS for T4-binding site on the blood-protein transthyretin (TTR). The analysis methods are the bioassays FITC-T4 and PFAS CALUX. The **Relative Potency Factor (RPF)** for 12 different PFAS congeners are expressed in PFOA equivalency (Table 1, Zeilmaker 2018¹⁵), see Table 1.

Overview of PFAS exposure pathways to the human population and the environment, see Figure 11, (Sunderland et al. 2019).¹⁶ "PFAS are man-made substances that do not naturally occur in the environment. Examples of PFAS are GenX, PFOA perfluoro octanoic acid and PFOS perfluorooctane sulfonates. PFASs are used in many products. As a result, and due to emissions and incidents, these substances have ended up in the environment and are now found in, among other things, soil, dredging spoil and surface water."¹⁷

Congener	RPF
Perfluorobutanesulfonate (PFBS, C4)	0.001
Perfluorohexanesulfonate (PFHxS, C6)	0.6
Perfluorooctanesulfonate (PFOS, C8)	2
	0.05
Perfluorobutanoic acid (PFBA, C4)	0.05
Perfluoropentanoic acid (PFHxA, C6)	0.01
Perfluorooctanoic acid (PFOA, C8)	1
Perfluorononaoic acid (PFNA, C9)	10
Perfluoroundecanoic acid (PFUnDA, C11)	4
Perfluorododecanoic acid (PFDoDA, C12)	3
Perfluorotetradecanoic acid (PFTeDA, C14)	0.3
Perfluorohexadecanoic acid (PFHxDA, C16)	0.02
Perfluorooctadecanoic acid (PFODA, C18)	0.02

Table 1: Relative Potency Factor (RPF) for 12 PFAS expressed in PFOA equivalency (RIVM, Zeilmaker 2018)



Figure 11: Overview of PFAS exposure pathways to the human population and the environment (Sunderland et al. 2019)

¹⁴ Van Leeuwen SPJ, Kärrman A, Van Bavel B, De Boer J and Lindstrom G, (2006). Struggle for quality in determination of perfluorinated contaminants in environmental and human samples. Environmental Science and Technology, 40, 7854–7860. ¹⁵ M.J. Zeilmaker et al 2018. Mixture exposure to PFAS: A Relative Potency Factor approach, National Institute for Public Health and the Environment, RIVM Report 2018-0070.

¹⁶ Sunderland EM. (2019). Journal of Exposure Science & Environmental Epidemiology (2019) 29:131–147

¹⁷ <u>https://www.rivm.nl/en/pfas</u>

Bioassays

DR CALUX

The bioassay DR CALUX[®] (Dioxin Responsive Chemical Activated LUciferase gene eXpression) is used for quantification of dioxins/furans (PCDD/F) and dioxin-like PCBs (dl-PCBs). The results in this research with DR CALUX[®] for analyses on dioxins (PCDD/F/dl-PCBs) on eggs are expressed in **Bioassay Equivalent, BEQ (pg BEQ/g fat)**. The term "**BEQ**" is used for food elements to distinguish between the **TEQ** (Toxic Equivalence) derived from chemical analyses (Gas Chromatography-Mass Spectrometry GC-MS, pg TEQ/g fat). For non-food biomatrices like mosses or pine needles, the results with the DR CALUX will be expressed in **TCDD eq./g product** or abbreviated as **pg TEQ/g product**. TCDD stands for 2,3,7,8-Tetrachlorodibenzo-p-dioxin, the most toxic dioxin congener.

COMMISSION REGULATION (EU) **2017/644 of 5 April 2017¹⁸**, concerning laying down methods of sampling and analysis for the control of levels of dioxins, dioxin-like PCBs and non-dioxin-like PCBs in certain foodstuffs, is the latest and in force. **Regulation EU 1881/2006¹⁹** is included. The regulation sets maximum levels for dioxins (PCDD/F/dI-PCB) in food products. The food products which are listed should not be placed on the commercial market if a contaminant exceeds the maximum level set out in the Annex of the EU documents.

The limits set in legislation are expressed in pg TEQ/g, based on GC-MS measurements. The GC-MS analysis concerns 7 dioxins (PCDDs), 10 furans (PCDFs), 12 dioxin-like polychlorinated biphenyls (dl-PCBs), and 6 indicator polychlorinated biphenyls (i-PCB).

The results of the chemical analyses with GC-MS of dioxins (PCDD/F/dI-PCBs) will be calculated with a specific Toxic Equivalency Factor (TEF) towards a TEQ value (see page 4 Abbreviation and TEF for dioxins, and dI-PCBs). The sum of the TEQ will be measured with upper bound values, meaning calculation with the value of the limit of detection (LOD) of a specific congener. These GC-MS **limit values** for chicken eggs are 2.5 pg TEQ/g fat for dioxins (PCDD/F) and for the sum of dioxins and dioxin-like PCBs (PCDD/F/dI-PCBs), the GC-MS limit value is set at 5 pg TEQ/gram fat. When exceeding these GC-MS limit values, chicken eggs are not allowed to be on the commercial market, (see Figure 5 and 6).

Recommendations 2013/711/EU²⁰, update 2017/644 sets out the cut-off values of the DR CALUX analysis determined. If the analysis exceeds the 70% value of dioxins (PCDD/F), i.e. 1.7 pg BEQ/g fat and/or 70% of the limit of the sum of dioxins (PCDD/F/dI-PCB) i.e. 3.3 pg BEQ/g fat a GC-MS analysis of the egg sample is recommended to establish the results with the GC-MS chemical analysis, where **EU 1881/2006** can be applied.

2013/711/EU²¹ includes the **action levels GC-MS** for both dioxins (PCDD/F) and dioxin-like PCBs (dl-PCBs) in chicken eggs set at 1.75 pg TEQ/g fat, see Figure 5. These action levels are a tool for competent authorities and operators to highlight cases where it is appropriate to identify a source of contamination and to take measures for its reduction or elimination.

¹⁸ <u>https://eur-lex.europa.eu/legal-content/en/ALL/?uri=CELEX:32017R0644</u>

¹⁹ https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:02006R1881-20210919&from=EN

²⁰ <u>https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32014R0709&from=EN</u>

²¹ <u>https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32013H0711&from=EN</u>

PAH CALUX®

High molecular weight PAHs are known ligands of the aryl hydrocarbon receptor (AhR), a nuclear receptor that mediates toxic effects related to these compounds. The PAH CALUX assay uses a mammalian, H4IIE- cell-based reporter assay for the hazard identification of total PAH mixtures. The PAH CALUX reporter cell line allows for specific, rapid (4-hour exposure time) and reliable quantification of AhR-induced luciferase induction relative to benzo[a]pyrene (BaP). BaP is a compound with five benzene rings and a class 1B carcinogen and is used here as an toxicity indicator of PAH exposure^{22,23}.

PFAS CALUX®

The chemical analyses on individual PFAS congeners are very limited, depending on the lab, only 8 - 55 substances are currently analysed in routine laboratories. Practically, this means that only 0.1- 1% can be determined with the chemical analyses, compared with the value of the Total Organic Fluorine $(TOF)^{24}$. The bioassay of PFAS CALUX comprises human bone marrow cell lines (U2OS), incorporating the firefly luciferase gene coupled to Thyroid Responsive Elements (TREs) as a reporter gene for the presence of thyroid hormone-like inhibiting compounds. It is based on the TTR-binding of PFAS in combination with the TR β CALUX detection. The presence of increasing concentrations of PFAS capable of competing with T4 for TTR-binding sites will result in a decreased amount of TTR-bound T4. Disruption of T4-TTR binding is benchmarked against the reference compound Perfluorooctanoic acid (PFOA), which value is set to one (1), just like TCDD in the TEQ calculation²⁵. See table 1 for relative potency factors of other PFAS. The analysis results of the PFAS CALUX are expressed in: μg PFOA equivalent/g product.

FITC-T4 assay

In the FITC-T4 binding bioassay, sample extracts, suspected to be contaminated with PFAS, are tested for the potency of binding with the thyroid hormone thyroxine (T4) to the plasma transport protein Transthyretin (TTR). The fluorescent-labelled thyroxine (FITC-T4) consisting of Fluorescein isothiocyanate (FITC) and L-thyroxine (T4) are used in this assay (Smith, 1977, Hamers 2020)^{26,27}. The thyroid hormone homeostasis can be disrupted by environmental chemicals at different points of interaction in the thyroid pathway, including during transport of the hormone through the blood. Some chemicals are known to bind to the transport protein TTR thereby replacing the endogenous ligand T4. PFAS are such chemicals known for their capability to bind TTR thereby replacing T4. The measurement is based on the difference in fluorescence between bound and non-bound FITC-T4 to the TTR-binding site. Bound FITC-T4 will result in a higher fluorescence than non-bound. The analysis results of the FITC-T4 will be expressed in: μ**g PFOA equivalent/g product**.

The DR CALUX[®], PFAS CALUX[®], FITC-T4, and GC-MS-analysis were performed by BioDetection Systems, Amsterdam, the Netherlands. BDS is accredited under RvA L401.

²⁷ Hamers T. (2020). Transthyretin-Binding Activity of Complex Mixtures Representing the Composition of Thyroid-Hormone Disrupting Contaminants in House Dust and Human Serum, Environmental Health Perspectives 017015-1 128(1)

²² Category 1B carcinogen according to Annex VI to the CLP Regulation (EC) No 1272/2008 of the European Parliament, and is classified as a Substance of Very High Concern by the POP Regulation EC No 850/2004.

²³ Pieterse B, Felzel E, Winter R, van der Burg B, Brouwer A. (2013). PAH-CALUX, an optimized bioassay for AhR-mediated hazard identification of polycyclic aromatic hydrocarbons (PAHs) as individual compounds and in complex mixtures. Environ Sci Technol. 2013 Oct 15;47(20):11651-9. doi: 10.1021/es403810w. Epub 2013 Sep 25. PMID: 23987121.

²⁴ Straková, J., Schneider, J., Cingotti, N. et al., 2021. Throwaway Packaging, Forever Chemicals: European wide survey of PFAS in disposable food packaging and tableware. 54 p.

²⁵ P.A. Behnisch et al.(2021). Developing potency factors for thyroid hormone disruption by PFASs using TTR-TRB CALUX® bioassay and assessment of PFASs mixtures in technical products, Environment International 157, 106791 26 Smith, D.S., (1977). FEBS Lett. 77, 25-27.

Backyard chicken eggs

Backyard chicken eggs are used for biomonitoring levels of contamination by POPs in various studies. Eggs are sensitive indicators of POP contamination in soil and dust and are a significant exposure pathway from soil pollution to humans. Eggs from contaminated areas can readily lead to exposures that exceed thresholds for the protection of human health. Chickens and their eggs might, therefore, be ideal "active samplers": an indicator species for the evaluation of contamination levels of sampled areas by POPs, particularly by dioxins (PCDD/Fs) and dioxin-like-PCBs (dl-PCBs)^{28,29}.

When chickens are free to forage on natural uncovered soil in the open air without roofing, they are in optimal contact with the environment. Eggs can reflect the chemical situation of soil biota related to the atmospheric deposition of hazardous chemical particles from industrial emissions, such as car shredding, metallurgy, coal-fired power plants, foundries, the PVC industry, cement kilns, the paper industry, and waste incineration. Chickens forage on and in the soil, eating insects, invertebrates, vegetation even grass (Figure 12). As a result, persistent organic pollutants (POPs) like dioxins (PCDD/F/dl-PCB)can be found in the fatty egg yolk and act as a biomarker for the environment. The chicken excretes the toxic compounds like dioxins into the fatty yolk when producing the eggs (dioxins are fat related). The older the chicken is, the more toxic compounds can be collected in the body, a process called bioaccumulation. Biotransformation refers to the capability of an organism to break down certain substances. Xenobiotic metabolism refers to the metabolism or breakdown of foreign substances not belonging to the substances of an organism of an ecological system.



Figure 12: Biomonitoring of backyard chicken eggs in natural environment

²⁸ Arkenbout A, Esbensen K H. (2017) Sampling, monitoring and source tracking of Dioxins in the environment of an incinerator in the Netherlands, Proceedings Eighth World Conference On Sampling and Blending / Perth ²⁹ Petrlík J. (2015). Persistent Organic Pollutants (POPs) in Chicken Eggs from Hot Spots in China.Beijing-Gothenburg-Prague, Arnika - Toxics and Waste Programme,

European Food Safety Authority (EFSA)

Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) and dioxin-like polychlorinated biphenyls (dl-PCBs) are important contaminants in the food chain. In 2018 the European Food Safety Authority (EFSA), the official adviser of European decision-makers, advised to reduce the tolerable weekly intake (TWI) from 14 to 2 pg TEQ (Toxic Equivalents)/kg body weight per week, based on extended scientific reviews conducted on humans and animals (EFSA, 2018)³⁰, see Figure 13. It demonstrates the present exposure to dioxins for most consumers in the EU exceeds the TWI. The European Commission, Council and Parliament have until now decided not to take into account this strong scientific advise to reduce these limit values by a factor of 7, as it would have huge implications on the economy, probably implying to withdraw important amounts of products from the European market. The actual dioxin limit value for eggs is 2.5 pg TEQ PCDD/g fat and 5.0 pg TEQ/g fat PCDD/F/dl-PCB. A reduction of these limit values with a factor of 7 will have enormous implications see Figure 13. The actual EU limits (Figure 5 and 6), based on pre EFSA advise, before 2018, and can been seen as more the result of political economic rather than preliminary on behalf of human health arguments.



Figure 13: Tolerable Weekly Intake of dioxins revision for adults and children (EFSA 2018), graphs by TW[©].

Public concern about ongoing contamination of POPs in human bodies has increased since several of these substances of very high concern have been identified as hormone disrupters and immune depressors. There are many risks and effects of having these chemicals in our environment and, as far as dioxins are concerned, they are of no benefit. Pollutants like dioxins contaminate the environment, persist for decades, and cause problems such as cancer, birth defects, learning disabilities, immunological deficiency, behavioral, neurological, and reproductive discrepancies in human and other animal species.

For PFOS and PFOA the EFSA established a **tolerable weekly intake (TWI) of 13 ng/kg body weight per week (PFOS) and 6 ng/kg body weight per week (PFOA) respectively**³¹. For both compounds, the exposure of a considerable proportion of the population exceeds the proposed TWI. A safe daily dose of **GenX or HFPO-DA is 3 ng/kg of body weight**, according to the EPA.

³⁰ EFSA CONTAM Panel (EFSA Panel on Contaminants in the Food Chain), Knutsen HK et al. 2018. Scientific Opinion on the risk for animal and human health related to the presence of dioxins and dioxin-like PCBs in feed and food. EFSA Journal 2018;16(11):5333, 331 pp.

³¹ EFSA CONTAM Panel (EFSA Panel on Contaminants in the Food Chain), Knutsen HK et al, 2018. Scientific Opinion on the risk to human health related to the presence of perfluorooctane sulfonic acid and perfluorooctanoic acid in food. EFSA Journal 2018;16(12):5194, 284 pp.

Sampling

The sampling for this research is performed by a team of Collectif 3R (réduire, réutiliser, recycler) in Paris, lvry-sur-Seine, Alfortville, Charenton, Paris 12th and 13th arrondissement. At first an exploration was undertaken, by the sampling team of Collectif 3R (réduire, réutiliser, recycler), for the possibilities of biomonitoring in the region around the waste incineration on biomarkers as eggs of backyard chicken, and vegetation (pine needles, leaves and mosses). The first set-up of the initial sample plan for biomonitoring eggs is given in Figure 14 and vegetation around the waste incinerator lvry XIII in Figure 15. This biomonitoring research in Paris on vegetation of mosses, pine needles (*Pinus sylvestris* and *Cedrus atlantica*), foliage of evergreen tree *Cupressus arizonica* and leaves of Olive tree - *Olea europaea* is concentrated in the center of the inner circle of 1 km.

	TW-REF-NR	Distance (m)	
	lvry-egg1	866	
	lvry-egg3	1070	pooled
	lvry-egg5	978	
Paris	lv-egg2	1640	
	lv-egg4	1115	
	alf-egg1	2810	
	alf-egg2	2160	
	Paris-egg7	2230	
Supermarket	lv-egg9	reference	

Sampling plan eggs, Paris Ivry - 2021



Figure 14: Sample plan eggs for backyard chicken eggs - Paris 2021

		Sample	e plan vege
	Paris 2021 Vege	tation sampling plar	n
Sample number		Specie	Distance incinerator (m)
V1	IVRY_VEG-02	Cedrus atlantica	487
V2	CHAR_VEG-9a	Cuppressus arizonica	724
V3	PARIS-13_VEG-24a	Pinus sylvestris	1060
V4	IVRY_VEG-23	Pinus sylvestris	525
V5	IVRY VEG-5	Olea europaea	427

Figure 15: Sample plan vegetation - Paris 2021

Questionnaire

All the chicken coop owners of the eight (8) participating egg locations were asked to fill a questionnaire provided by TW. The summary of the answers on the questions in the questionnaire about keeping chickens, like number hens, rooster, breed, foraging area, and possible confounders are in given in Table 2. Five chicken coop owners give permissions for using pictures in this biomonitoring report. Egg location lvry_egg1, lvry_egg3 and lvry-egg5, in this research pooled as one (1) egg location, because they have in total 4 laying hens and therefore analysed together to minimize to individual variability.

				Paris lvry	2021				
TW-REF-NR	IVRY_EGG1	IVRY_EGG3	IVRY_EGG5	IVRY_EGG2	IVRY-EGG4	ALF_EGG1	ALF_EGG2	PARIS-EGG7	IVRY-EGG9
Distance (m)	866	1070	978	1640	1115	2810	2160	2230	
Pics permissions		yes	yes						
Chicken breed	red hooded			red hooded		Silk,	black	Maran	
						wyandotte	red	Susex	
Hens (n)	1	1	2	2 x 5	5 (3-2)	12	2	4	
Rooster (n)						3			
Age (month)	48	13	24	24	6 - 24		10	24-60	
Eggs/day				7-10	2			variable	
Eggs/week	2	6			14		6		
Eggs/month	8	24		210	56	variable	24		
Foraging area (m2)	6	9		20	400	25	150	75	
Housing (m2)	20	1		5 (2)	5	2	12	6	
Terrain	soil	soil	soil	soil	soil	soil	soil	soil	
					trees	trees	grass	fruit trees	
					grass	grass	trees	mirabellier	
					herbes		herbes	cerisier	
Feed	grain	grain		grain		mais	grain	grain var.	
	wet bread	peelings		food rests		grain	earthworms	food rests	
	cheese coating	no				food rests	snails		
Outdoor fireplace	1x a year	no		no	no	no	no	no	
Housing material	plastic	plastic		straw	plastic	straw	straw	wood	
		lino		wood	lino	wood	wood	plastic	
Barbeque		sometimes							
All purpose burner	non	no		moderate	moderate			no	
Pesticides use	no	not known		no	unknown	no	no	no	
Industry nearby	no	nearby		no	yes		yeast factory		
Highway nearby	road	road		road	yes		Alfortville	Rue de Reuilly	

Table 2: Answers of chicken coop owners on the questions of the Egg questionary Paris - 2021, provided by TW

Results biomonitoring

Eggs

The bioassay DR CALUX analysis on dioxins (PCDD/F and dl-PCBs) is performed on the eggs of backyard chicken. This method has a EU regulatory cut off value of 1.7 and 3.3 pg BEQ/g fat for PCDD/F and respectively PCDD/F/dl-PCB. These limits are 2/3 of maximum level of a GC-MS analysis, to determine if a sample is compliant to the EU regulations or, when exceeding the cut off values, is suspected. All egg samples except sample IV-egg2 exceed the maximal level for the sum of dioxins (PCDD/F/dl-PCB) and exceed the maximal level for dioxins (PCDD/F) according to EU regulation 1881/2006³². See Table 3.

	Results dioxons in eg	ggs samples P	aris-Ivry - 20	21		
DR CALUX	lvry-egg1/3/5	lvry-egg2	lvry-egg4	Alf-egg1	Alf-egg2	Paris-egg7
pg BEQ/g fat	WI 9-688 1/ 5/ 5	1419-6552	1419-6554	AII-CEEL	AII-6852	ганз-едд/
PCDD/F	4.63	0.84	7.43	4.3	7.1	2.8
dI-PCB	2.87	0.36	3.22	4.2	7.1	3.1
PCDD/F/dl-PCB	7.5	1.2	10.65	8.5	14.2	5.9

Table 3: Results for dioxins in Eggs with DR CALUX, Paris - 2021

OR CALUX Eggs

lvry-egg1

lvry-egg3 lvry-egg5

lv-egg2 lv-egg4

alf-egg1

alf-egg2

Paris-egg7

lv-egg9

EU limit

Paris

Supermarke

DR CALUX

TW-REF-NR PCDD/F/dl-PCB

(ng BEO /g /fat)

7.5

1.2

10.7

8.5

14.2

bove the EU limit of the sum of PCDD/F/dl-PCBs

PCDD/F

4.63

0.8

7.4

4.3

7.1

2.8

Figure 16 presents the DR CALUX analyse results for the sum of dioxins (PCDD/F/dI-PCB) in eggs of backyard chicken. The results of this analyses method are according to the EU requirements as indicated in Commission Regulation (EU) 2017/644 of 5 April 2017, and when exceeding the (cut-off) values for DR CALUX, a chemical (GC-MS) analysis is needed for control of dioxins (PCDD/F), dioxin-like PCBs (dI-PCB) levels and non-dioxin-like PCBs in eggs. See Figure 5 and 6.



Results dioxin PCDD/F/dl-PCB analyses eggs DR CALUX Paris - 2021

Figure 16: Results of dioxins PCDD/F/dl-PCB analyses in eggs, Paris - 2021

³² https://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:02006R1881-20210919&from=EN Biomonitoring research Paris, France - 2021

The DR CALUX results (pg BEQ/g fat) for only dioxins (PCDD/F) in the egg samples in this biomonitoring research are presented in Figure 17. Five (5) of the six (6) egg samples exceeding levels for safe consumption according to the EU regulations.



Results dioxin PCDD/F analyses eggs DR CALUX Paris - 2021

Figure 17: Results of dioxins (PCDD/F) analyses in eggs, Paris - 2021

For dioxin-like PCBs (dl-PCBs) in the egg samples the DR CALUX results show rather low values comparing to the results of dioxins (PCDD/F), which are more combustion related, inside the circle of 2 km around the waste incinerator. The eggs samples on further distance, outside the 2 km circle, like egg location alf-egg2 shows much higher dl-PCB contamination (Figure 18).



Results dioxin dl-PCB analyses DR CALUX eggs Paris - 2021

Figure 18: Results dioxin like PCB (dl-PCB) in eggs, Paris - 2021

GC-MS analyses

A chemical GC-MS analysis was necessary to be performed for verification of the DR CALUX results to have recommendations and obligations in the framework of the European food safety. The food safety regulation, with the limits for dioxins and dioxin-like PCB, is applied to the commercial egg market. However, some backyard chicken coops can have a production up to 300 eggs a month, not only consumed by the owner of the chicken coop, but also to a certain community (family, friends, neighbors i.e.).

The GC-MS analyses confirms the by the DR CALUX suspected samples. Egg location Alf-egg 2 exceed the EU limit of safe food consumption of eggs nearly with a factor 6, with a result of 29.00 pg TEQ/g fat, Figure 19. Egg location lvry-egg4 measures in the GC-MS analyses > 4 x times more dl-PCB than with the DR CALUX. See Table 6 for the percentage of PCB 126 in all the egg samples. This PCB congener has a TEF of 0,1 and gives therefore a high TEQ value. Figure 20, results on a TW indicative scale.



Results GCMS sum of dioxins (PCDD/F/dl-PCB) eggs - PARIS 2021

Figure 19: GC-MS Results for the sum of dioxins (PCDD/F/dl-PCBs) in eggs GC-MS eggs, Paris - 2021

The results of these analyses are exceptionally high. To clarify the results, a comparative color scale has been created to give more profoundness to the results, see Figure 20.



Results GC-MS sum of dioxins (PCDD/F/dl-PCB) in eggs - PARIS 2021

Figure 20: TW indicative scale of GC-MS analysis of sum of dioxins(PCDD/F/dl-PCBs) in food.

All except egg location Iv-Egg2 exceeding the limit for dioxins (PCDD/F). The congeners of dioxins (PCDD/F) are the most associated with uncomplete combustion. The highest one is measured in Iv-egg4, with 8.6 pg TEQ/g fat, the place also with a high dl-PCB contamination of 16.0 pg TEQ/g fat, Figure 21.



Results GC-MS dioxins (PCDD/F) in eggs - PARIS 2021

Figure 21: Results GC-MS analyses eggs, Paris - 2021

The highest level of dl-PCB in this measurement series is measured at egg location alf-egg2 with a value of 21.0 pg TEQ/g fat. This is exceptional high, compared with other measurement of dl-PCB of ToxicoWatch biomonitoring researches (see also TW comparative scale page 38).



Results GC-MS dioxin-like PCB (dl-PCB) in eggs - PARIS 2021

Figure 22: Results GC-MS analyses on dioxin-like PCBs, Paris - 2021

Congeners

In Table 4 the 17 dioxin (XCDD) and furan (XCDF) congeners are shown as a percentage of the total TEQ PCDD/F and in Table 5 a percentage of the total concentration PCDD/F at the different egg locations is presented. The dominant congeners are marked in dark brown boxes: TCDD, PCDD, PCDF2 and to some extent TCDF.

The most highest value are marked black, ranking the second and third in grey with white and black letters respectively. The congeners of the supermarket eggs (Ivry-EGG9) are mainly based on upper bound values, 16 dioxin and furan congeners were all below the detection limit, except for HpCDD, , only 1% in TEQ (Table 4) and 9% in concentration (Table 5). What can be tentatively deduced is the observation of some difference of Paris-egg7 and Ivry-egg4 with the other locations. A dominant presence of HxCDD2 and a low value of PCDF2 is observed in the TEQ profiles of Ivry-egg4. The dominant presence of HxCDF2 indicates a specific yet unclarified dioxin source.

	% TEQ ir	ndividual co	ongeners P	CDD/F egg	s - Paris 20	21	
	IVRY 1/3/5	IVRY_EGG2	IVRY-EGG4	ALF_EGG1	ALF_EGG2	PARIS-EGG7	IVRY-EGG9
TCDD	9%	20%	6%	8%	8%	6%	33%
PCDD	30%	23%	23%	36%	26%	22%	33%
HxCDD1	1%	2%	3%	1%	3%	1%	3%
HxCDD2	5%	5%	15%	7%	7%	10%	3%
HxCDD3	3%	2%	6%	2%	5%	2%	3%
HpCDD	3%	3%	13%	2%	8%	3%	1%
OCDD	0%	0%	1%	0%	1%	0%	0%
TCDF	12%	9%	6%	10%	12%	6%	3%
PCDF1	1%	1%	1%	2%	1%	1%	1%
PCDF2	26%	15%	10%	16%	19%	13%	10%
HxCDF1	4%	5%	3%	4%	4%	5%	3%
HxCDF2	4%	6%	11%	6%	5%	18%	3%
HxCDF3	1%	2%	0%	0%	0%	0%	3%
HxCDF4	3%	4%	2%	3%	2%	3%	3%
HPCDF1	0%	0%	0%	2%	0%	9%	0%
HPCDF2	0%	0%	0%	0%	0%	0%	0%
OCDF	0%	0%	0%	0%	0%	0%	0%

Table 4: Fraction TEQ (%) of dioxin (PCDD/F) congeners in eggs, Paris - 2021

		con	c % PCDD/F	eggs Paris 2	021		
	IVRY 1/3/5	IVRY-egg2	IVRY-egg4	ALF_egg1	Alf_egg2	Paris-Eegg7	IVRY-EGG9
TCDD	1%	1%	0%	1%	0%	0%	6%
PCDD	3%	2%	1%	3%	1%	1%	6%
HxCDD1	1%	1%	1%	1%	1%	1%	6%
HxCDD2	4%	3%	3%	6%	2%	5%	6%
HxCDD3	2%	1%	1%	2%	1%	1%	6%
HpCDD	23%	18%	28%	19%	25%	14%	9%
OCDD	32%	43%	58%	22%	58%	9%	6%
TCDF	10%	7%	1%	8%	4%	3%	6%
PCDF1	4%	3%	1%	5%	1%	2%	6%
PCDF2	7%	4%	1%	4%	2%	2%	6%
HxCDF1	4%	3%	1%	4%	1%	3%	6%
HxCDF2	3%	4%	2%	5%	2%	9%	6%
HxCDF3	0%	1%	0%	0%	0%	0%	6%
HxCDF4	2%	3%	1%	3%	1%	1%	6%
HPCDF1	0%	1%	0%	15%	0%	47%	6%
HPCDF2	0%	1%	0%	1%	0%	0%	6%
OCDF	3%	1%	2%	1%	1%	1%	6%

Table 5: Fraction of dioxins (PCDD/F) congeners concentration (%) in eggs, Paris - 2021

Figure 23 can be seen in the right square mark that egg location Alf_EGG1 and egg location Paris-EGG7 have similar pattern for HpCDF1 and show a clearly different pattern compare with other egg locations in Paris 2021.



Figure 23: Congeners distribution in eggs, Paris - 2021

Egg locations

Egg location lvry_egg1/3/5

In the next chapters the dioxin congeners of each individual egg location are compared in fraction concentration (%) and fraction TEQ (%) with the patterns of a WtE incinerator in Harlingen, the Netherlands (NL). These NL profiles are the results of real measured data inside the chimney of this WtE incinerator in NL which have been performed for > 20,000 of hours continuous measurements, under mainly normal conditions and more or less limited measurements during shutdown and start-up conditions.

The three following egg locations (1,3 and 5) are pooled together for analyses, (Figure 24 and 25). In total 4 hens are involved at these three locations. The presence of 1,2,3,4,6,7,8-Heptachlorodibenzo-p-dioxin and Octachlorodibenzo-p-dioxin in this analyses are typical for waste Incineration (red bars in the graph). Usually the ratio of these two congeners is two to one, but here the ratio is more like 3 to 2. Typically is the presence of 2,3,7,8-Tetrachlorodibenzofuran (TCDF) in this profile.



Egg location IVRY_EGG 1, 3 & 5 – Paris 2021

Figure 24: Overview egg location Ivry-egg 1/3/5, Paris - 2021



Figure 25: Data and comparison combustion congener patterns egg pooled location Ivry_egg1/3/5, Paris - 2021

Egg location lvry_egg2

2,3 1,2 1,2 1,2 1,2 1,2 2,3 1,2 2,3 1,2 1,2 1,2 1,2 1,2 1,2 1,2

Oct

Egg location lvry_egg2 seems to be preserved from dioxin contamination in these series of measurements. In Figure 26 some pictures are shown of this location, with a very "clean" results. Photos of the housing and the large roof of the chicken enclosure, maybe a factor in protecting the hens from dioxins contamination by air. It is quite possible that cleaned soil has been used in the chicken coop.

IVRY-egg2		
PCDD/F congeners	TEF	pg/g
3,7,8-Tetrachlorodibenzo-p-dioxin	1	< 0,2
2,3,7,8-Pentachlorodibenzo-p-dioxin	1	0,23
2,3,4,7,8-Hexachlorodibenzo-p-dioxin	0,1	< 0,2
2,3,6,7,8-Hexachlorodibenzo-p-dioxin	0,1	0,48
2,3,7,8,9-Hexachlorodibenzo-p-dioxin	0,1	< 0,2
2,3,4,6,7,8-Heptachlorodibenzo-p-dioxin	0,01	2,50
ctachlorodibenzo-p-dioxin	0,0003	6,00
3,7,8-Tetrachlorodibenzofuran	0,1	0,91
2,3,7,8-Pentachlorodibenzofuran	0,03	0,45
3,4,7,8-Pentachlorodibenzofuran	0,3	0,51
2,3,4,7,8-Hexachlorodibenzofuran	0,1	0,47
2,3,6,7,8-Hexachlorodibenzofuran	0,1	0,56
2,3,7,8,9-Hexachlorodibenzofuran	0,1	< 0,2
3,4,6,7,8-Hexachlorodibenzofuran	0,1	0,43
2,3,4,6,7,8-Heptachlorodibenzofuran	0,01	< 0,2
2,3,4,7,8,9-Heptachlorodibenzofuran	0,01	< 0,2
ctachlorodibenzofuran	0,0003	< 0,2

Egg location IVRY_EGG2 - Paris 2021

Figure 26: Egg location Ivry_egg2 - Housing and foraging area, Paris - 2021

Although this is egg location comply with the EU limits, dominant presence of the typical incineration congeners 1,2,3,4,7,8,9-Heptachlorodibenzofuran, HpCDD, and Octachloro-dibenzofuran, OCDD, are visible in the congener patterns found in these eggs, see Figure 27.



Egg location IVRY_EGG2 - Paris 2021

Figure 27: Data egg location Ivry_egg2, Paris - 2021

Egg location lvry_egg4

On location lvry_egg4 a high level of dioxins (PCDD/F) and dl-PCBs is measured. In Figure 28 are the results of the PCDD/F results in eggs demonstrated. In Figure 29, table left the results in BEQ and TEQ are demonstrating these eggs do not comply the EU regulations, neither for the bioassay nor for the GC-MS. The sum of PCDD/F/dl-PCB exceed the EU limit, and the action limits for PCDD/F, and for dl-PCB are exceeded. In the concentration fraction (%) HpCDD and OCDD are the dominant congeners. In the TEQ pattern are beside PCDD and PCDF2, three other congeners obvious present: HxCDD HpCDD and HxCDF2. This pattern is only observed at this location. On this location the highest dl-PCB concentration is measured of TW researches in eggs see comparison scale at Figure 39.

lvry-egg4			SE ARR	20E ANR	Montread	-			
PCDD/F congeners	TEF	pg/g				a series		AC II	ALT .
2,3,7,8-Tetrachlorodibenzo-p-dioxin	1	0,54	A Manual Contraction	Constant		and a second	de des Patits suis	e al	A STATE OF
1,2,3,7,8-Pentachlorodibenzo-p-dioxin	1	2,00				IDI			COUNCIL PERSON
1,2,3,4,7,8-Hexachlorodibenzo-p-dioxin	0,1	2,50				and the second	34	C. 4.	A Second
1,2,3,6,7,8-Hexachlorodibenzo-p-dioxin	0,1	13,00				11	D154A	ALL AND	Sever 5
1,2,3,7,8,9-Hexachlorodibenzo-p-dioxin	0,1	5,20	600			1 1 1	310		3
1,2,3,4,6,7,8-Heptachlorodibenzo-p-dioxin	0,01	110,00			the colum			· Mu	C LAN
Octachlorodibenzo-p-dioxin	0,0003	230,00	hry-egg4	S D.		79/ 2	0-10 3	C. C. Y.	Decteur Bouaved N
2,3,7,8-Tetrachlorodibenzofuran	0,1	5,20					()-) ×		Copernants
1,2,3,7,8-Pentachlorodibenzofuran	0,03	2,40					D15	AA	
2,3,4,7,8-Pentachlorodibenzofuran	0,3	2,90							
1,2,3,4,7,8-Hexachlorodibenzofuran	0,1	2,20	Dioxins and furans distribution in eggs Paris 2021						
1,2,3,6,7,8-Hexachlorodibenzofuran	0,1	9,20		IVRY 1/3/5	IVRY_EGG2	IVRY-EGG4	ALF_EGG1	ALF_EGG2	PARIS-EGG7
1,2,3,7,8,9-Hexachlorodibenzofuran	0,1	0,24	PCDD	31,0	9,8	363,2	39,6	229,0	37,2
2,3,4,6,7,8-Hexachlorodibenzofuran	0,1	2,10	PCDF	16,5	4,1	31,9	33,8	28,7	83,6
1,2,3,4,6,7,8-Heptachlorodibenzofuran	0,01	< 0,2	PCDD/F	47,5	13,9	395,1	. 73,4	257,7	120,7
1,2,3,4,7,8,9-Heptachlorodibenzofuran	0,01	0,56	PCDF%	35%	30%	8%	46%	11%	69%
Octachlorodibenzofuran	0,0003	6,90	PCDD %	65%	70%	92%	54%	89%	31%

Figure 29: Data egg location Ivry_egg4, Paris - 2021



Figure 28: Data egg location Ivry_egg4, Paris - 2021

Egg location ALF_EGG1

Egg location ALF_EGG1, in Alfortville, shows a high level of for dioxins. The results do not comply with the EU-limit for dioxins (PCDD/F) and the sum of dioxins (PCDD/F/dI-PCB), and they are exceeding the EU action limit for dI-PCB. Action is needed for elimination or reduction of the source of these dioxins and dioxin-like PCBs. In the graph of the concentration patterns the dominant presence of furans are observed, see Figure 30.

		.0			
Alf-egg1					
PCDD/F congeners	TEF	pg/g			
2,3,7,8-Tetrachlorodibenzo-p-dioxin	1	0,49			
1,2,3,7,8-Pentachlorodibenzo-p-dioxin	1	2,20			
1,2,3,4,7,8-Hexachlorodibenzo-p-dioxin	0,1	0,89			
1,2,3,6,7,8-Hexachlorodibenzo-p-dioxin	0,1	4,60			
1,2,3,7,8,9-Hexachlorodibenzo-p-dioxin	0,1	1,40			
1,2,3,4,6,7,8-Heptachlorodibenzo-p-dioxin	0,01	14,00			
Octachlorodibenzo-p-dioxin	0,0003	16,00			
2,3,7,8-Tetrachlorodibenzofuran	0,1	6,00			
1,2,3,7,8-Pentachlorodibenzofuran	0,03	3,40			
2,3,4,7,8-Pentachlorodibenzofuran	0,3	3,30			
1,2,3,4,7,8-Hexachlorodibenzofuran	0,1	2,60			
1,2,3,6,7,8-Hexachlorodibenzofuran	0,1	3,80			
1,2,3,7,8,9-Hexachlorodibenzofuran	0,1	< 0,2			
2,3,4,6,7,8-Hexachlorodibenzofuran	0,1	1,90			
1,2,3,4,6,7,8-Heptachlorodibenzofuran	0,01	11,00			
1,2,3,4,7,8,9-Heptachlorodibenzofuran	0,01	0,49			
Octachlorodibenzofuran	0.0003	1 10			

Egg location ALF_EGG1 – Paris 2021





Figure 30: Data egg location Alf-egg1, Paris - 2021



Egg location ALF_EGG1 – Paris 2021

Figure 31: Data egg location Alf_egg1, Paris - 2021

Egg location ALF EGG2

Egg location ALF_EGG2, also in Alfortville, shows a high level of the sum of dioxins (PCDD/F/dI-PCB) of 29.0 pg TEQ/g fat. This value is nearly an exceeding of a factor 6 of the EU limit of safety of eggs. The action limit for dioxins (PCDD/F) is exceeded with a factor 3 and the action limit for dioxin-like PCB (dl-PCB) more than 12 times, (Figure 32). Action is very needed to reduce this extreme toxic contamination. The level of PCDD/F/dI-PCB and dI-PCB is the highest found in all TW researches as presented in Figure 38.

	Eg	gg loca	ation ALF_EGG2 – Paris 2021
Alf-egg2			THE RATE OF THE PARTY OF THE PA
PCDD/F congeners	TEF	pg/g	THE REAL PLANT IN THE AREA
2,3,7,8-Tetrachlorodibenzo-p-dioxin	1	0,61	
1,2,3,7,8-Pentachlorodibenzo-p-dioxin	1	2,10	
1,2,3,4,7,8-Hexachlorodibenzo-p-dioxin	0,1	2,10	
1,2,3,6,7,8-Hexachlorodibenzo-p-dioxin	0,1	5,50	
1,2,3,7,8,9-Hexachlorodibenzo-p-dioxin	0,1	3,70	
1,2,3,4,6,7,8-Heptachlorodibenzo-p-dioxin	0,01	65,00	
Octachlorodibenzo-p-dioxin	0,0003	150,00	Alfeg2
2,3,7,8-Tetrachlorodibenzofuran	0,1	9,90	
1,2,3,7,8-Pentachlorodibenzofuran	0,03	3,00	
2,3,4,7,8-Pentachlorodibenzofuran	0,3	5,00	
1,2,3,4,7,8-Hexachlorodibenzofuran	0,1	2,80	
1,2,3,6,7,8-Hexachlorodibenzofuran	0,1	4,3	
1,2,3,7,8,9-Hexachlorodibenzofuran	0,1	0,18	
2,3,4,6,7,8-Hexachlorodibenzofuran	0,1	1,40	
1,2,3,4,6,7,8-Heptachlorodibenzofuran	0,01	< 0,2	
1,2,3,4,7,8,9-Heptachlorodibenzofuran	0,01	0,24	
Octachlorodibenzofuran	0,0003	1,70	

Figure 32: Data and pictures of egg location Alf-egg2, Paris - 2021

The concentration patterns show a resemblance at combustion congener patterns. The TEQ patterns reveals a presence of TCDD and TCDF, see Figure 33.



Egg location ALF_EGG2 – Paris 2021

Figure 33: Data egg location Alf-egg2, Paris - 2021

Egg location Paris-egg7

The henhouse is a closed part of a shared garden and belongs to inhabitants of the surrounded flat buildings. This 'backyard' chicken experience was initiated and is still managed by 15 families, living in the apartment buildings, (no public access). The chicken enclosure is cleaned once a week (changing papers and cardboards on the floor). There are now 4 hens, one of race "Marans" (brown), Figure 34. Hens are eating seeds and food waste. Their droppings are managed separately because of potential issue. Despite the efforts at this location to keep it clean, the eggs are not complying the EU-limit of dioxins (PCDD/F) and the sum of dioxins (PCDD/F/dl-PCBs), as well the action limit of dl-PCB is exceeded. In Figure 35 the dominant presence of HpCDF1 congener in the concentration and TEQ patterns is observed. The congener HpCDF1 indicate more contamination sources can be involved.

Egg location PARIS-EGG7 – Paris 2021

Paris-egg7					
PCDD/F congeners	TEF	pg/g			
2,3,7,8-Tetrachlorodibenzo-p-dioxin	1	0,35			
1,2,3,7,8-Pentachlorodibenzo-p-dioxin	1	1,30			
1,2,3,4,7,8-Hexachlorodibenzo-p-dioxin	0,1	0,70			
1,2,3,6,7,8-Hexachlorodibenzo-p-dioxin	0,1	5,90			
1,2,3,7,8,9-Hexachlorodibenzo-p-dioxin	0,1	0,94			
1,2,3,4,6,7,8-Heptachlorodibenzo-p-dioxin	0,01	17,00			
Octachlorodibenzo-p-dioxin	0,0003	11,00			
2,3,7,8-Tetrachlorodibenzofuran	0,1	3,80			
1,2,3,7,8-Pentachlorodibenzofuran	0,03	2,00			
2,3,4,7,8-Pentachlorodibenzofuran	0,3	2,70			
1,2,3,4,7,8-Hexachlorodibenzofuran	0,1	3,10			
1,2,3,6,7,8-Hexachlorodibenzofuran	0,1	11,00			
1,2,3,7,8,9-Hexachlorodibenzofuran	0,1	0,21			
2,3,4,6,7,8-Hexachlorodibenzofuran	0,1	1,80			
1,2,3,4,6,7,8-Heptachlorodibenzofuran	0,01	57,00			
1,2,3,4,7,8,9-Heptachlorodibenzofuran	0,01	0,54			
Octachlorodibenzofuran	0,0003	1,40			



Figure 34: Data egg location Paris-egg7, Paris - 2021



Egg location PARIS-EGG7 – Paris 2021

Figure 35: Data egg location Paris-egg7, Paris - 2021
lvry-egg9

Egg sample Ivry-egg9 are eggs from the supermarket, taken as reference in the analysis of this biomonitoring research. All the congeners, except HpCDD, measures are under the limit of detection 0.1 pg/g with the chemical analysis of GC-MS. Only HpCDD was measured with a levels of 0,17 pg TEQ/g fat, see Figure 36.

Egg sample Ivry-egg9 (supermarket) – Paris 2021

lvry-egg9 (supermark	et) Paris
Congener concentration	pg/g fat
TCDD	< 0,1
PCDD	< 0,1
HxCDD1	< 0,1
HxCDD2	< 0,1
HxCDD3	< 0,1
HpCDD	0,17
OCDD	< 0,1
TCDF	< 0,1
PCDF1	< 0,1
PCDF2	< 0,1
HxCDF1	< 0,1
HxCDF2	< 0,1
HxCDF3	< 0,1
HxCDF4	< 0,1
HPCDF1	< 0,1
HPCDF2	< 0,1
OCDF	< 0,1



Figure 36: Ivry-egg9: Eggs from the supermarket, Paris - 2021

PCB contamination

PCBs are ubiquitous found in this research. The proportion of the different congeners is for PCB 118 58%-70%, PCB 156 5%-9% and PCB 105 from 18%-28%. Location IVRY-EGG4 is considerable contaminated by PCB 118 with a level of 120000 pg/g, counting for 22% of the dl-PCB TEQ. Location Alf_egg2 showed a high level of PCB 126 with 200 pg/g, 93% of the dl-PCB TEQ of 21 pg TEQ/g. Alf-egg2 have a different source of this highly toxic dl-PCB, (Table 6). Ivry-egg9 are the supermarket eggs, in which only PCB 118 could be measured above the limit of detection.

	dl-PCB in eggs (pg/g fat) - Paris 2021													
pg/g	IVRY 1/3/5	IVRY_EGG2	IVRY-EGG4	ALF_EGG1	ALF_EGG2	PARIS-EGG7	IVRY-EGG9							
PCB77	580,0	26,0	220,0	420,0	350,0	190,0	< 1							
PCB81	13,0	<2	6,0	4,2	6,4	4,5	< 1							
PCB126	56,0	12,0	110,0	110,0	200,0	64,0	< 1							
PCB169	4,5	1,2	5,7	6,7	6,7	5,2	< 1							
PCB105	9500,0	520,0	30000,0	12000,0	8300,0	5300,0	< 1							
PCB114	320,0	23,0	1300,0	550,0	250,0	300,0	< 1							
PCB118	21000,0	1700,0	120000,0	25000,0	25000,0	12000,0	3,7							
PCB123	250,0	33,0	850,0	340,0	290,0	190,0	< 1							
PCB156	3100,0	250,0	11000,0	2000,0	3200,0	1500,0	< 1							
PCB157	450,0	51,0	2200,0	610,0	660,0	380,0	< 1							
PCB167	660,0	120,0	4700,0	1100,0	780,0	700,0	< 1							
PCB189	170,0	30,0	340,0	260,0	190,0	220,0	< 1							
Sum (LB)	36103,5	2766,2	170731,7	42400,9	39233,1	20853,7	3,7							

Table 6: dl-PCB in eggs, Paris - 2021

Table 7 shows the high contribution of PCB 126 to the Total TEQ, which might come due to a too high assigned WHO-TEF value. This may explain that DR CALUX results for dI-PCB are usually lower than GC-MS TEQ results. The analysis results of the supermarket eggs are also included, although almost all congeners, with the exception of PCB 118, are below the detection limit. In a continue measurements of the emissions of the incinerator 10% of the TEQ found to be related to dioxin-like PCBs³³. A remark has to be made, that semi-continuous measurements are by far the best way in measuring emissions of dioxins during normal operation. However, measuring emissions during transient phases, such as start-up and shutdown, requires a different methodology due to changing conditions such as temperature and gas velocity. The amount of data is limited, however a study by Li from 2018 indicates formation of dioxin like PCBs during transient phases such as start-ups and shutdowns is very likely³⁴.

	% T	EQ dl-PCB	congeners	eggs Paris,	France 2	021	
	IVRY 1/3/5	IVRY_EGG2	IVRY-EGG4	ALF_EGG1	ALF_EGG2	PARIS-EGG7	IVRY-EGG9
PCB105	4%	1%	6%	3%	1%	2%	0%
PCB118	9%	4%	22%	6%	4%	5%	0%
PCB126	82%	91%	67%	88%	93%	89%	77%
PCB169	2%	3%	1%	2%	1%	2%	23%

Table 7: % TEQ dl-PCB congeners eggs, Paris - 2021

³³ Arkenbout A. (2018). Hidden Emissions: A story from the Netherlands, a case study, November 2018, Zero Waste Europe, <u>www.zerowasteeurope.eu</u>

³⁴ Li M, Wang C, Cen K, Ni M, Li X. (2018). Emission characteristics and vapour/particulate phase distributions of PCDD/F in a hazardous waste incinerator under transient conditions. R. Soc. open sci. 5: 171079.

Biomonitoring research Paris, France - 2021

Comparison with AIRPARIF study (2017)

In a study of Airparif³⁵ in 2017 with the active air sampler DA80 sampler 25290 m3 air was collected for analysing chlorinated and brominated dioxins. The measurement site near the Ivry/Paris XIII waste incineration plant is located in Charenton-le-Pont. In Figure 37 a comparison is made with the congener patterns found in eggs. Dioxin (PCDD) patterns looks similar (green and blue, resp. HpCDD and OCDD), while the difference with the egg locations are more seen in the diversity of furans. Furans can be originated by backyard burning or incidental fire, although a recently study by Chen dominating furans can be more traced back to 'fresh' produced dioxins by incineration. Clear to see that egg location Iv-Egg4 has a dominant PCDD fraction (92%) and the location of Paris-egg7 (right) has a dominant PCDF fraction (69%). The dominant present of HpCDF of Paris-egg 7 can indicate another source, although a study of Chen³⁶ showed that the 1,2,3,4,7,8,9-Heptachlorodibenzofuran (HpCDF1) congener is an indicator of incineration. Perhaps a different configuration of the incinerator or a different waste input is to be held responsible for this particular emission, different from the patterns of the REC in the Netherlands.



Figure 37: : Comparison of the PCDD/F congeners found in eggs and in air with the DA80 sampler

³⁶ Chen P. et al. (2017). Chemosphere 181, 360 - 367

³⁵ Etude des dioxines dans l'air ambiant | Août 2018, Airparif

Biomonitoring research Paris, France - 2021

Comparative scale of dioxins

Comparison GC-MS analysis of TW biomonitoring researches of dioxins (PCDD/F) and dioxin-like PCBs (dl-PCB) on eggs in the environment of a waste incinerator in Europe between 2019-2021. In the TW biomonitoring researches, Belgium, Spain, Lithuania, Czech Rep., and France, egg samples are taken within a distance of 5 km from a waste incinerator. The results of the egg locations in Paris Ivry have the highest level measured by TW for the sum of dioxins (PCDD/F/dl-PCB) and the dioxin-like PCB (dl-PCB). The figures 38 and 39 show the results of six (6) TW biomonitoring projects. A TW comparative scale is made with color marks to specify the degree of exceeding the EU limit.



TW comparative scale dioxins (TEQ PCDD/F/dl-PCB)in eggs around incinerators Europe

Figure 38: TW Comparative scale for sum of dioxins (PCDD/F/Dl-PCB) in eggs





Figure 39: Comparative scale dl-PCB concentrations in eggs

Other Than Normal Operating Conditions (OTNOC)

Dioxin-like PCBs form a substantial part of dioxin pollution in Paris (see also Figure 39). A study by Li (Zhejiang University, Hangzhou, China, 2018) demonstrates high emissions of dioxin like PCBs during transient phases of start-ups and shutdowns³⁷. Continuous measurements in the stack/chimney of a waste incinerator, also during OTNOC can measure the contribution of dI-PCB in the flue gases. However, measuring dioxins during the transient phases is extremely difficult due to the extreme physical conditions of speed, temperature and dust emissions.

In the official yearly report for 2020 of the Ivry-Paris XIII incinerator published by SUEZ company, DIP - Bilan Annuel 2020 ³⁸ results of measurements of dioxins (PCDD/F) and dl-PCB are given during the transient stage (figure 23, page 113). In a study of the Dutch Government (ODRA) and ToxicoWatch emissions of dioxins are measured in start-ups of the waste incinerator REC, The Netherlands. Five (5) different stages could be differentiated in the start-up process: Cold phase (SU1), Flushing (SU2), Heating up (SU3), Start waste feed (SU4), and normal incineration (SU5). All these stages have different dynamics, with different degrees of difficulty to perform dioxin measurements in particulate and gaseous phases. One problem in transient phases is the variance in velocity of flue gases. When the velocity of flue gases comes under the 1.5 m/s, sampling of dioxins is not possible. Sampling of dust, particulate phase, faces also much complications, conventional dust meters are not applicated to measure high levels of dust emitted during start-ups or shutdowns³⁹. Measurement can hardly be performed during highly fluctuating circumstances of shutdown or start-up conditions. The very low results in the DIP - Bilan Annuel in Figure 40 suggest measurements are performed when the conditions of velocity and temperature are stabilized. In our study this is the last stage of the start-up, 32-50 hours after the start of heating up the incinerator.

> Tableau de synthèse des moyennes des campagnes de mesures lors des phases transitoires d'arrêts et démarrages :



Phases transitoires de démarrages :

Figure 40: Measurement results incinerator during transient stages (page 113 DIP - Bilan Annuel 2020)

³⁷ Li M, Wang C, Cen K, Ni M, Li X. 2018 Emission characteristics and vapour/particulate phase distributions of PCDD/F in a hazardous waste incinerator under transient conditions. R. Soc. open sci. 5: 171079.

³⁸ Usine d'incineration d'ordures menageres D'ivry-Paris XIII, dossier d'information du public bilan annuel 2020 ³⁹ Arkenbout, A , Olie K , Esbensen, KH (2018). Emission regimes of POPs of a Dutch incinerator: regulated, measured and hidden issues, Dioxin Conference Krakow, http://dioxin20xx.org/wp-content/uploads/pdfs/2018/461.pdf

An example of emissions during an Other Than Normal Operation Conditions (OTNOC) situation in a TW research on a WtE incinerator in the Netherlands is shown in Figure 41. It shows clearly that in the five (5) stages of a start-up, the first stages gives high emissions but are (still) not included in the EU regulations. In this figure is demonstrated the exceeding dioxins during this stages of start-ups. The Y-axis scale is in log pg TEQ/Nm3. Measurements are performed by the Dutch government, the processing of the results is done by ToxicoWatch and presented on the dioxin conference in Krakow, Poland⁴⁰.



Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) emissions (18 months)

Figure 41: Dioxin emissions during transient stages of incineration - TW research

The topic of exceeding dioxin emissions during OTNOC is still being researched and placed on the agenda of the Basel Conference to be implemented structurally in the guidelines for incinerators. The problem of unintentional formed dioxins and dioxin-like PCBs, besides the many other POPs, is still not solved, but important enough to mention in this report.

Li (2018) found high levels of dl-PCB formation during the start-up and shutdown (OTNOC)⁴¹. It could be also here a factor in the relative high amount of dl-PCB found in Paris. Dioxin-like PCBs had a great application in all kind of construction materials and paints, and therefore could be in all kinds of products of demolition and dumping. Therefore, emissions during OTNOC is an issue that certainly needs to be investigated further in waste incineration plants

⁴⁰ Arkenbout, A, Olie K, Esbensen, KH (2018). Emission regimes of POPs of a Dutch incinerator: regulated, measured and hidden issues, <u>Conference paper Dioxin</u>

⁴¹ Li M, Wang C, Cen K, Ni M, Li X. (2018) Emission characteristics and vapour/particulate phase distributions of PCDD/F in a hazardous waste incinerator under transient conditions. R. Soc. open sci. 5: 171079.

Brominated and mixed halogenated dioxins

In general most of the results in eggs surrounding this incinerator by biological DR CALUX are higher than the follow-up by chemical GC-MS analysis. This is an indication that brominated and mixed halogenated dioxins are part of the content of a sample analysed on dioxins. Based on TW researches with a TW database of 104 analyse results of egg samples with DR CALUX and GC-MS, only 3% of the results give a higher GC-MS value. In our here presented research, five (5) of the six (6) DR CALUX results are slightly lower than the GC-MS. See Figure 42. This could be conflicting by the research done in air samples by Airparif Paris, finding high levels of brominated dioxins, even at TEQ level, with active air sampling. Therefore, we recommend to design here a more suitable follow-up study by using biological and chemical analysis in combination for egg and emission gas samples.

TW have setup the analyses in two different serie, with both series giving the same results with lower DR CALUX results. In our database of backyard chicken eggs measured with DR CALUX and GC-MS, there is an incidence of 3% of higher values of the GC-MS. This measurements serie of Paris counts 83% (5 out of 6) and that is remarkable. Interesting are the ones in our database falling apart of this negative TEQ/BEQ relation. The eggs were sampled within the influence area of two incinerators and one big chemical industry. One in Harlingen, REC, The Netherlands and one in Rotterdam, near the largest incinerator of The Netherlands (AVR). This makes the results suspect of another parameter influence the results. The bioassay is based on living cells, probably there's some interaction patterns, we still not understand. But the fact that it happens just in area where just complex mixtures of chemical compounds can be found. Could it be that some substances, probably brominated or mixed brominated dioxins or other halogenated POPs have counteractivity on the AhR receptor? This finding of reduced DR CALUX values at certain suspected locations for brominated dioxins should be further investigated.



A brominated question of DR CALUX

Figure 42: A brominated question: Chemical congener-specific GC-MS vs biological sum of all kinds of dioxin-like compounds by DR CALUX

Biomonitoring of evergreen trees

At first an inventory was made of all possible vegetations sample locations in the environment around the waste incinerator lvry/Paris XIII, lvry-sur-Seine, Paris 13^e arrondissement and Charenton-le-Pont (see Annex 3). The final sample plan implemented and collected on June 28th, 2021 is shown in Table 8 and Figure 43 present the vegetation locations on the map. In Table 8 the column with the distance refers to the distance in meters to the waste incinerator.

			Dioxins Veg	getation	s Paris	
Sample date	Specie	Sample	Ref. Nr.	Distance (m)	coordinates	address
28/06/2021	Cedrus atlantica	V1	IVRY_VEG-02	487	48.8182, 2.3933	Jardin Elisabeth
28/06/2021	Cuppressus arizonica	V2	CHAR_VEG-9a	724	48.8251, 2.3960	Near 3 Jardin du Cardinal de Richelieu,
28/06/2021	Pinus sylvestris	V3	PARIS-13_VEG-24a	1060	48.8299, 2.3801	Jardin Abbé Pierre
28/06/2021	Pinus sylvestris	V4	IVRY_VEG-23	525	48.8214, 2.3946	lvry- 3-11 Quai Jean Compagnon
28/06/2021	Olea europaea	V5	IVRY_VEG-5	427	48.8193, 2.3920	Allée Chanteclair

Table 8: Sample plan vegetation around waste incineration Ivry/Paris XIII, Paris 2021

Sample plan vegetation Paris Ivry 2021 Paris 2021 Vegetation sampling plan Sample Reference Specie Distance Number number incinerator (m) IVRY_VEG-02 Cedrus atlantica 487 V1 V2 CHAR_VEG-9a Cuppressus arizonica 724 V3 PARIS-13_VEG-24a Pinus sylvestris 1060 V4 IVRY VEG-23 Pinus sylvestris 525 IVRY_VEG-5 V5 Olea europaea 427



Figure 43: Sample plan of vegetation in Paris Ivry in 2021

Figures 44 shows the results with the DR CALUX of dioxins in pine needles, foliage and leaves of vegetation. There are very few studies using the bioassay to measure persistent organic pollutants in vegetation for comparison. Most of the researches uses the chemical GC-MS analyses for dioxin analysis. In this report a comparative indicative scale with results of TW data in other European researches is presented. Based on these results TW applicate an indicative (color) scale to interpret the divers DR CALUX results in the context.



Figure 44: Results of dioxin analyses in evergreen trees in our sampling campaign in Paris Ivry in 2021

Vegetation Locations, Paris Ivry 2021

In the following five figures an overview of each vegetation location in more detail, see Figure 45-49.

Vegetation V1

Vegetation location V1 in Ivry-sur-Seine, is a public garden, probably from the 1980s. It is called "Jardin de la rue Elisabeth". The resinous tree, Atlas cedar - *Cedrus atlantica*, is a tree of 15 meter. The needles are taken 2 meter from the ground. The sample place is 200 meter from a recycling metal industry. The results of the sum of dioxins (PCDD/F/dI-PCB) are quite high with a level of 1.7 pg TCDD eq./g dry product. The result of DR CALUX for dioxins (PCDD/F) is 1.1 pg TCDD eq./g and 35% of this value is due to the dioxin-like (dI-PCB) fraction with a level of 0.6 pg TCDD eq./g dry product (Figure 45).



Figure 45: Vegetation location 1, Paris - 2021

Vegetation location 2 in Charenton-le-Pont is near Jardin du Cardinal de Richelieu. The garden is open for public and is situated between apartment blocks, see Figure 46. The sample of needles is taken from *Cupressus arizonica* from 1,5 – 2 meter from the ground. The results of the bioassay DR CALUX on this needles is found to be extremely high with 4.4 pg TCDD eq./g product for the sum of dioxins (PCDD/F/dl-PCB). Compared to other TW studies, these levels are the same as the dioxins levels found 200 meter from a large municipal waste incinerator Valdimingómez in Madrid. Remarkable is the fraction of dioxins (PCDD/F) of 93% with a value of 4.1 pg TCDD eq./g product. PCDD/Fs are combustion related.

Cuppressus arizonica Near Jardin du Cardinal de Richelieu

Figure 46: Vegetation location 2, Paris - 2021

Vegetation location 3 is located in Paris 13e arrondissement 1060 meter North-West from the incinerator. It is public garden, named 'Jardin Abbé Pierre'. The tree, where pine needle samples were taken is most probably a *Pinus sylvestris*. The results with the bioassay are the lowest in this measurement, compared to other results slightly elevated. The fraction dioxins (PCDD/F) is less than the dioxin-like PCB (dl-PCB) fraction with a level of 0.24 pg TCDD eq./g product, respectively 0.32 pg TCDD eq./g product, Figure 47.

V3: PARIS-13 VEG-24a

		Diox	ins Vegetation	s Paris	s		
Sample date	Specie	Sample	Ref. Nr.	Distance	PCDD/F/dl-PCB	PCDD/F	PCB
				m		pg TCDD eq./g	
28/06/2021	Cedrus atlantica	V1	IVRY_VEG-02	487	1.70	1.10	0.60
28/06/2021	Cuppressus arizonica	V2	CHAR_VEG-9a	724	4.39	4.10	0.29
28/06/2021	Pinus sylvestris	V3	PARIS-13_VEG-24a	1060	0.56	0.24	0.32
28/06/2021	Pinus sylvestris	V4	IVRY_VEG-23	525	1.64	0.87	0.77
28/06/2021	Olea europaea	V5	IVRY_VEG-5	427	3.12	2.7	0.42



Figure 47: Vegetation location 3, Paris - 2021

Vegetation location 4 is located on the quayside of the Seine in Ivry-sur-Seine. The sample location V4 overlooks the quay, above the quay, in between the Seine river and the North-East of Ivry-Port. Pine needles were sampled from *Pinus sylvestris* which seems in good shape. The needles were sampled 2 meter from the ground. The result with the DR CALUX is 1.64 pg TCDD eq./g product for the sum of dioxins (PCDD/F/dI-PCB). The fraction for dioxins (PCDD/F) is 53% with a measured value of 0.87 pg TCDD eq./g product, Figure 48.

			V4: IV	RY_VE	G-23			
		Diox	ins Vegetation	s Paris	;			
Sample date	Specie	Sample	Ref. Nr.	Distance	PCDD/F/dl-PCB	PCDD/F	PCB	
				m		pg TCDD eq./g		
28/06/2021	Cedrus atlantica	V1	IVRY_VEG-02	487	1.70	1.10	0.60	
28/06/2021	Cuppressus arizonica	V2	CHAR_VEG-9a	724	4.39	4.10	0.29	
28/06/2021	Pinus sylvestris	V3	PARIS-13_VEG-24a	1060	0.56	0.24	0.32	
28/06/2021	Pinus sylvestris	V4	IVRY_VEG-23	525	1.64	0.87	0.77	
28/06/2021	Olea europaea	V5	IVRY_VEG-5	427	3.12	2.7	0.42	



Figure 48: Vegetation location 4, Paris - 2021

Vegetation sampling location 5 is the public garden Chanteclair in Ivry-sur-Seine. It is a small open place leading to the Dulcie September primary and pre-school school. Old olive trees – *Olea europaea* have been planted in 2011 -2016. The sample place is 427 meter away near from the incinerator. The fraction of dioxins (PCDD/F) is 84% with 2.7 pg TCDD eq./g product from the total sum of dioxins (PCDD/F/dl-PCB) of 3.12 pg TCDD eq./g product. As to see in Figure 49 location V5 and V2 belong to highest measured values in the European according to TW-researches on evergreen trees (Figure 50).

Figure 49: Vegetation location 5, Paris - 2021

Comparation with other TW-biomonitoring researches of evergreen trees

On the comparative scale of evergreen trees, Paris ranking high values. The TW indicative scale colored orange, red and dark red, meaning a serious load of dioxins (PCDD/F/dI-PCB) of the samples used in TW researches 2021, see Figure 50.



Figure 50: TW Indicative scale PCDD/F/dl-PCB evergreen trees

Remarkable at all five (5) vegetation locations in Paris, dioxin-like PCB (dl-PCBs) are among the highest values measured by TW in biomonitoring researches in Europe 2019-2021 (Figure 51, TW indicative scale dl-PCB).



Figure 51: TW Indicative scale dl-PCB evergreen trees

Mosses

Bryophytes are the non-vascular autotrophic cryptogams with second highest conglomeration among land plants after the angiosperms, and nearly about 25,000 species were present worldwide (Mishra et al. 2016). Mosses belong to the kingdom Plantae, and division Bryophyta. Mosses are a vegetation group that have 'rhizoids', small 'hairlike' structures with the main function of anchoring the plant to the ground, rock, bark or substrate, instead of a root system like plants and trees have for uptake of water, minerals, and possible contamination by (toxic) chemicals in the soil.

Dreyer (2018)⁴² find PCDD/F TEQ concentrations ranged from 0.024 pg TEQ to 0.81 pg TEQ. Caraballeira⁴³ (2006) et al. reported PCDD/F TEQ concentrations of 0.3 pg TEQ/g (in woodlands), 2,5 pg TEQ in relation to an incinerator. Most of the mosses are < 1 pg TEQ/g. Danielsson⁴⁴ et al.(2016) observed PCDD/F concentrations in Swedish moss samples (*Pleurozium schreberi* or *Hylocomium splendens*) from 0.0001 to 0.57 pg TEQ/g. Dioxin-like PCB (dl-PCB) in the study of Dreyer is below 0,5 pg TEQ/g. Generally, the concentrations of the analysed substances are low, often close to or below the quantification limits (LOQ) with the chemical analysis. A significant correlation is observed between PAHs, dioxin/furans, and dioxin-like PCBs in mosses and the distance to the closest industry.

In figure 52 it is shown the sample plan of mosses in the area < 1 km around the waste incinerator Ivry/Paris XIII. After sampling, transport and shipping to the Netherlands, the samples of mosses were for 24 hours dried at the air. For more reference results of mosses in remote areas used by TW biomonitoring researches in Europe 2019-2021, please see figure 51.



Figure 52: Sample plan mosses for analysis of dioxins (PCDD/F/dl-PCBs) - Paris 2021

⁴⁴ Danielsson et al. (2016). Persistant organic pollutants in Swedish mosses, IVL-report C 188

⁴² Dreyer et al. Environ Sci Eur (2018) 30:43 https://doi.org/10.1186/s12302-018-0172-y

⁴³ Carballeira A, Angel Fernandez J, Aboal JR, Real C, Couto JA (2006) Moss: a powerful tool for dioxin monitoring. Atmos Environ 40(30):5776–5786

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Results dioxin analysis mosses

In Figure 53 the analysis results for dioxins (PCDD/F) and dioxin-like PCBs (dl-PCB) are presented with the use of indicative colors according to TW indicative legenda. In this Figure 53 the results of the dioxin analyses on mosses with DR CALUX have been placed on the map, separated for the sum of dioxins (PCDD/F/dl-PCB), dioxins (PCDD/F), and dioxin-like PCB (dl-PCB). The results for dioxins in mosses are high, compare with other results of TW biomonitoring researches (see Figure 58-59). The dioxin values are between 1.17 - 4.90 pg TCDD eq./g product.



Figure 53: Results of DR CALUX analyses in mosses in Paris in 2021

Moss location 1

Moss location 1 is located near rue Westermeyer and rue JJ Rousseau and 620 meter from the incinerator. On this location a very high level of dioxins is measured of 4.9 pg TCDD eq./g product. More than 20% is accounted for dl PCB with 1.1 pg TCDD eq./g, (Figure 54)

		Мо	ss location 1			тохісо
	Mosses	Paris 202	21			1000 meter
Sample	TW-REF-NR	Distance	PCDD/F/dl-PCB	PCDD/F	PCB	
Sample	IW-REF-INR	m pg BEQ/g product				
M1	IVRY_Moss-01 + IVRY_Moss-02	620	4.90	3.80	1.10	
M2	IVRY_MOSS-04 + IVRY_MOSS-05	825	1.41	1.10	0.31	
M3	CHAR_MOSS-06a + CHAR_MOSS-06b	720	2.81	2.20	0.61	
M4	PARIS-13_MOSS-09 + PARIS-13_MOSS-10	920	1.17	0.88	0.29	



Figure 54: Moss location 1 – Paris 2021

Moss location 2

Moss location 2 is located in Parc Cormailles, 825 meter from the incinerator (Figure 55).

		Moss	location 2			
	Mosses	Paris 202	1			1000 meter
Sample	TW-REF-NR	Distance	PCDD/F/dl-PCB	PCDD/F	РСВ	
Sample	I W-REF-INK	m	pg B	EQ/g product		
M1	IVRY_Moss-01 + IVRY_Moss-02	620	4.90	3.80	1.10	
M2	IVRY_MOSS-04 + IVRY_MOSS-05	825	1.41	1.10	0.31	
M3	CHAR_MOSS-06a + CHAR_MOSS-06b	720	2.81	2.20	0.61	M2
M4	PARIS-13_MOSS-09 + PARIS-13_MOSS-10	920	1.17	0.88	0.29	



Figure 55: Moss location 2 - Paris 2021

Moss location 3

Moss location 3 is located at the Jardin du Cardinal Richelieu and 720 meters east of the waste incinerator. Just like the results on vegetation on this location a high level of dioxins is found, 2.8 pg TCDD eq./g product for the sum of dioxins (PCDD/F/dI-PCB). The dI-PCB is relative low with 0.3 pg TCDD eq./g product, Figure 56.

		Мс	oss location 3	3		
	Mosses	Paris 202	1			1000 meter
Comula	TW-REF-NR	Distance	PCDD/F/dl-PCB	PCDD/F	РСВ	
Sample	I W-REF-INR	m	pg B	EQ/g product		
M1	IVRY_Moss-01 + IVRY_Moss-02	620	4.90	3.80	1.10	
M2	IVRY_MOSS-04 + IVRY_MOSS-05	825	1.41	1.10	0.31	
М3	CHAR_MOSS-06a + CHAR_MOSS-06b	720	2.81	2.20	0.61	
M4	PARIS-13 MOSS-09 + PARIS-13 MOSS-10	920	1.17	0.88	0.29	



Figure 56: Moss location 3, Paris - 2021

Moss location 4

Moss location 4 is located at the Jardin Abbé Pierre, 920 North from the incinerator. At this location the lowest value in these measurements with 1.17 pg BEQ/g product for the sum of dioxins (PCDD/F/dl-PCB), Figure 57. However still high compared to other results in other biomonitoring projects of TW in Europe, see Figure 58 and 59.

		Moss	location 4				
	Mosses	Paris 202	21			1000 meter	
Sample	TW-REF-NR	Distance	PCDD/F/dl-PCB	PCDD/F	PCB		1
Sample	TW-KEP-WK	m	pg Bl	EQ/g product			
M1	IVRY_Moss-01 + IVRY_Moss-02	620	4.90	3.80	1.10		~
M2	IVRY_MOSS-04 + IVRY_MOSS-05	825	1.41	1.10	0.31		
М3	CHAR_MOSS-06a + CHAR_MOSS-06b	720	2.81	2.20	0.61		1
M4	PARIS-13_MOSS-09 + PARIS-13_MOSS-10	920	1.17	0.88	0.29		
M							No all and a second
Figure 5	7: Moss location 4, Paris - 2021	Ja	rdin Abbé Pierre			Constant of the second s	

Comparison with other biomonitoring studies on mosses

Figure 58 and 59 are TW indicative scales to compare and interpret DR CALUX results on mosses on data of TW researches on mosses in Europe 2019-2021.



Figure 58: TW Indicative scale mosses PCDD/F/dl-PCB with DR CALUX





Figure 59: Indicative scale of dl-PCB levels with DR CALUX in mosses

Comparison with SYCTOM research

The results of dioxin analyses in mosses is presented in the official yearly report for 2020 published by SUEZ. SUEZ's subcontractor conducted biomonitoring research on mosses locations further away from the Paris Ivry XIII incinerator than the moss locations used in this TW biomonitoring study 2021, namely within a circle of 1 km from this incinerator. The research on mosses conducted by the SYCTOM is being performed since 2016 (page 65 and page 124)⁴⁵. The results are summarized in Figure 60.

A number of remarkable things can be deduced from this report. First of all, it is noticeable that all the moss locations have almost the same dioxin contamination. Secondly, the range of dioxins in moss between 0.3 - 1.2 pg TEQ/g. Thirdly, the reference value for moss set at 0.6 pg TEQ.

This TW biomonitoring research shows higher dioxin levels at the four (4) moss locations in the vicinity of the incinerator. The dioxin values are between 1.2 - 4.9 pg TCDD eq./g product within a circle of 1000 meter around the incinerator. A difference of more than 400% or a factor 4 between the results of SUEZ and TW. The variation in the presented data of SUEZ demonstrate a low variation compared to the results of this biomonitoring study. In the incinerator report is stated that mosses have an ubiquitous value of 0.60 pg WHO-TEQ/g dry matter. However, no reference is attached and this value is contrary with the literature^{46,47,48}. In unpolluted areas, dioxins can't be measured above the limit of detection (LOD).

The threshold value set by DIP_IPXIII at 2 pg WHO-TEQ/g dry matter is the same as the limit value in the TW indicative scale for serious contamination. The differences with the TW scale is that the levels between 0.5 - 1.0 TCDD eq./g product are marked as elevated and not as normal values confirm the references 46-48.

The conclusion of SUEZ report is that "no activity by the waste incinerator can be detected". This TW biomonitoring research demonstrate the opposite by measuring increased levels of dioxins (PCDD/F) and dioxin-like PCBs (dI-PCB) in the environment of the incinerator. One explanation could be that with DR CALUX is more sensible for other POPs than the chemical analysis on only 17 PCDD/F congeners. In contrast, the results on the biomarkers of eggs demonstrated that the bioassay DR CALUX results in Paris showed lower levels than the chemical analysis. The reason is unknown, but it could be the result of antagonistic activity of a mixture of persistent organic pollutants, not good homogenized samples or too high analysed levels by chemical analysis. Therefore we recommend here to analyse again in parallel samples by chemical and biological analysis. Hypothetical the results could be more higher. The results of the mosses in the report of the waste incinerator have values between 0.5 -1.0 pg TEQ, while this TW research found much higher values of 1.2- 4.9 pg TCDD eq./g.

This TW research find large differences between moss locations in a circle of 1 kilometer from the incinerator. The conclusion of the report of SUEZ that no significant impact of the activity of the waste incinerator on the environment can be found, can't be verified. No support of reports of analyses are attached, therefore is no verification possible of these results, neither concerning the calculation of TEQ (table), neither the sampling program, pictures and even how the mosses are cleaned-up for the analyses.

 ⁴⁵ Usine d'incineration d'ordures menageres D'ivry-Paris XIII, dossier d'information du public bilan annuel 2020
 ⁴⁶ Danielsson H. et al. (2016). Persistant organic pollutants in Swedish mosses, IVL Swedish Environmental Research Institute 2016, report nr. C 188

⁴⁷ Carballeira A, Angel Fernandez J, Aboal JR, Real C, Couto JA (2006) Moss: a powerful tool for dioxin monitoring. Atmos Environ 40(30):5776–5786

⁴⁸ Dreyer et al. Environ Sci Eur (2018) 30:43 https://doi.org/10.1186/s12302-018-0172-y

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Comparison with France study - Paris 2021

> Résultats de mesure des mousses



 Distribution des teneurs en dioxines/furanes (pg OMS-TEQ/g de matière sèche) dans les mousses prélevées depuis 2016.

Figure 60: Comparison with study incinerator mosses

Conclusion

TW conducted a biomonitoring study, commissioned by Collectif 3R (réduire, réutiliser, recycler), on biomatrices eggs of backyard chicken, vegetation of evergreen trees and mosses in Paris/lvry-sur-Seine. High dioxin values are found in all three biomatrices. The eggs of backyard chicken, a sensitive biomarker of pollution of substances of very high concern in the environment, demonstrate with the bioassay of DR CALUX analyses 83% of the eggs (5 of the 6 samples) are exceeding the action limits for safe food consumption as regulated in the EU Directive 2017/644. The chemical GC-MS analyses of the eggs verify these results. Five (5) out of six (6) locations exceeding the EU limits for safe egg consumption according to EU Regulation 1881/2006. The chemical GC-MS analyses of the eggs resulted in five (5) locations exceeding the action limit for dioxin-like PCBs according to EU Regulation 1881/2006. If these eggs were produced for the commercial market, they should have to be withdrawn.

These EU limit values for food, and in this particular case for eggs, are commercially motivated. From a human health perspective, according to The European Food and Safety Authority (EFSA), these limits should be drastically modified. In 2018, EFSA re-evaluated the tolerable weekly intake (TWI) of dioxins and concluded that this TWI should be set 7 times lower to protect human health. The production of eggs from hobby hens can amount to 200 eggs per month and pose a substantial risk to communities.



Figure 61: Conclusion biomonitoring dioxins (PCDD/F/dl-PCB) in eggs, Paris Ivry - 2021

The congeners of the dioxins (PCDD/F) show identical patterns in concentration as in TEQ fractions with the incinerator patterns of the WtE incinerator (REC) in Harlingen, The Netherlands. Figure 61 shows an environment in Paris Ivry under stress of dioxins and dioxin-like PCBs. Dioxins can be produced by many sources, but incineration is an important source, still.

The analyse results of vegetation, pine needles and mosses, demonstrate high dioxin levels in the vicinity of the incinerator. Figure 62 demonstrate dioxins in mosses of 1.2 - 4.9 pg TCDD eq./g product and 1.6 - 4.4 pg TCDD eq./g product in pine needles. Fraction of PCDD/F dominates over dl-PCB.



Figure 62: Conclusion biomonitoring vegetation (evergreen trees and mosses), Paris Ivry - 2021

Based on the limited data of this biomonitoring study, contamination with dioxins is identified in Ivry Paris. More research is needed to eliminate or minimize this contamination of extremely toxic substances, which poses a risk for human health. The found results of the dioxin analyses (PCDD/F/dI-PCB) in eggs and vegetation are among the highest values in ToxicoWatch biomonitoring studies in Europe.

Further actions

- Based on these limited analysis results, a clear signal was found that further research is needed in Ivry Paris to advocate for reducing dioxins from the environment.
- More research is needed on biomarkers, such as eggs from backyard chickens, vegetation, like mosses, pine needles, leaves and foliage of evergreen trees. Analyses in humans may also be considered.
- More research is needed on the contribution of the incinerator to the levels of dioxins found in Ivry Paris.

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