Evaluation of persistent organic pollutants (POPs) in samples of eggs from the vicinity of municipal waste incinerator Košice

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FOREWORD

Closed materials loop or in other words the transition from the linear to circular economy is one of the key elements of sustainability. Therefore, it is not surprising that the circular economy is also prominently featured in the European Green Deal

Currently the largest proportion of the municipal waste in Slovakia is being landfilled. Sadly, while there is Europe wide push for transition to circular economy, where the waste hierarchy is strictly implemented, in Slovakia we can observe push to shift from the worst to second worst waste management. From landfill to incineration. Instead of strong preference for solutions from the high end of the waste hierarchy. Waste reduction, reuse, repair, recycling.

One of the side effects of the incineration is the pollution with POPs. Persistent Organic Pollutants, especially dioxins. These cancer-causing chemicals, are banned under the Stockholm Convention and the Union is currently in the process of updating its own POPs regulation.

Analysing the free-range eggs from the vicinity of the Košice Municipal Waste Incinerator was a way to showcase the interlinkage of two issues – Circular Economy and Persistent Organic Pollutants. And while the results did not show any samples above the legal limit, the presence of these cancer-causing chemicals, is nevertheless reason for concern.

Minimising the waste incineration and creating proper legislative and financial framework for the transition to circular economy is one of the tasks that we have as Union. Making sure that the remaining incinerators are operated without emissions of POPs and within the Emissions Trading System another. As European Parliament we have now a unique chance to advance the progress on both fronts.

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1. INTRODUCTION

This brief report evaluates results of the analyses of free-range chicken eggs sampled in a vicinity of the municipal waste incinerator Kosit based in Košice, in Eastern Slovakia (further as MWI Košice). This evaluation was done in response to a request from the secretariat of Member of the European Parliament, Martin Hojsík.

Waste incineration is considered to be a significant source of various toxic chemicals partly released in air emissions (Jay 1995) and partly transferred in wastes produced by waste incinerators, in fly ashes and/or air pollution control residues in particular (Labunska, Stephenson et al. 2000, Petrlik and Ryder 2005, Wang, Hu et al. 2019). This includes also persistent organic pollutants (POPs) such as dioxins and furans (PCDD/Fs), dioxin-like polychlorinated biphenyls (dl PCBs), pentachlorobenzene (PeCB) and hexachlorobenzene (HCB), all listed as unintentionally produced POPs in Annex C of the Stockholm Convention (Stockholm Convention 2010).

There was done sampling in surrounding of the MWI Košice in 2005 already. Its results were published in separate report (Petrlik, Hegyi et al. 2005) as well as part of a global “The Egg Report” (DiGangi and Petrlik 2005). MWI was proven as the potentially significant source of contamination with PCDD/Fs and dl PCBs that time. Situation has changed since that time. MWI is better equipped with APC abatement, and waste incineration residues are landfilled at separate sites (SIŽP 2021).

We were able to get samples of free-range chicken eggs from the vicinity of MWI Košice only, so this report cannot evaluate potential pollution caused in surrounding of the sites where waste incineration residues are stored.

We were not able to analyze all POPs and **other toxic chemicals which might be released from waste incinerator because of time constrains and laboratories available for such analyses. So this is not an exhaustive research on all toxic chemicals among which polybrominated dibenzo-p-dioxins and dibenzofurans (PBDD/Fs) should be mentioned explicitly as they are often found in waste incineration releases** (Soderstrom and Marklund 2002, Wang, Hsi et al. 2010, Wang, Chen et al. 2010).

2. MATERIALS AND METHODS

2.1. Sampling

Sampling sites in the surrounding of the MWI Košice were chosen in the villages Kokšov – Bakša and Valalíky, in the parts potentially affected by a deposition of the emissions from the incinerator. They are marked in map at Figure 1. They are similar to areas of samples taken in 2005 (Petrlik, Hegyi et al. 2005). The estimation of the areas was based on data about prevailing winds obtained from Slovakian Hydro-meteorological Institute, Bratislava (Pecho 2021). Prevailing winds for the period of 2019 – 2020 are summarized in a graph at Figure 2.

Figure 1: Areas of sampling of free-range chicken eggs in the vicinity of MWI Košice in villages Valalíky (Oblast A), and Kokšov – Bakša (Oblast B).

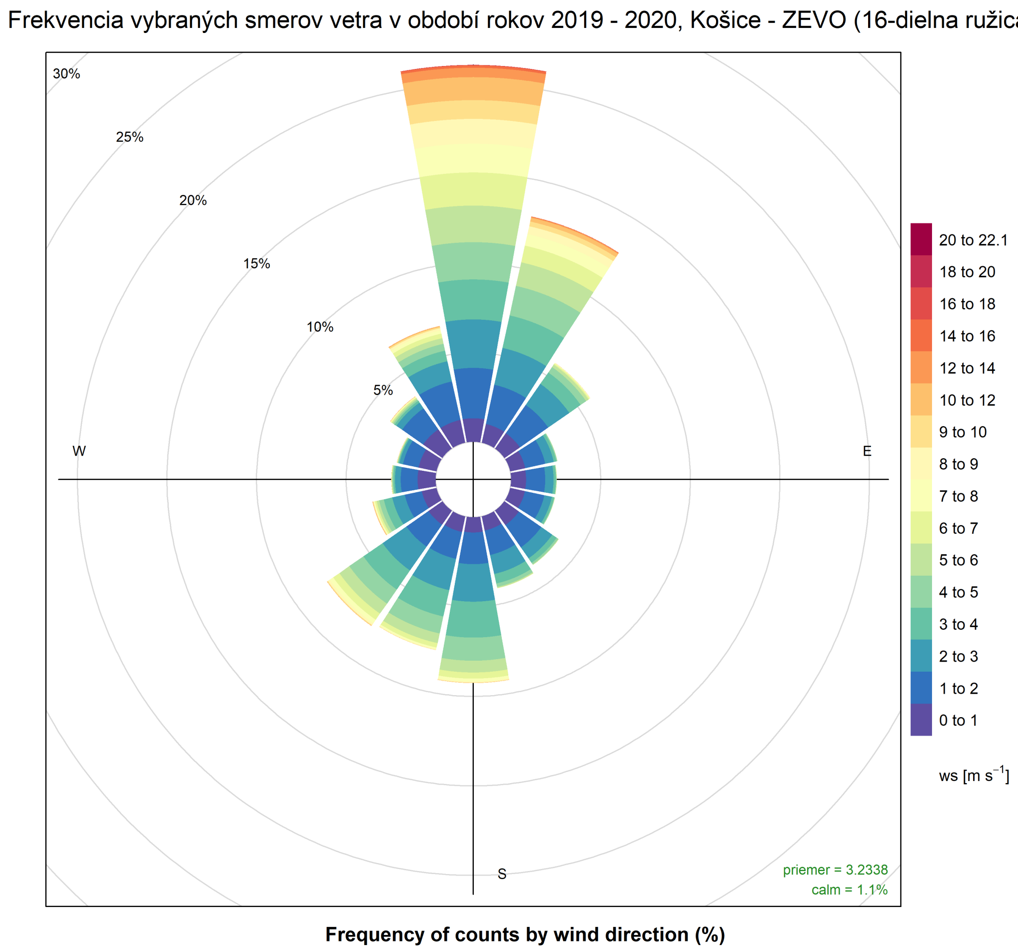

Figure 2: Windrose for MWI Košice. Source: (Pecho 2021).

Pooled egg samples were collected at each of the selected sampling sites Kokšov-Bakša and Valalíky in order to obtain representative samples. All sampled eggs origin from free-range chicken. For comparison we also include an older analysis of eggs bought in supermarket, in Prague, Czech Republic. Samples from 4 different houses/fanciers were collected in Kokšov-Bakša, 3 – 4 eggs from each, so 15 eggs in total. Eggs from 3 houses/fanciers were obtained in Valalíky, 4 eggs from each, so 12 eggs in total. The houses were close from each other, so we could mix them into two pooled samples representing selected areas.

Fanciers were investigated about conditions of housing, feeding, and others for hens, in order to avoid contamination of eggs from other sources than deposition of pollutants via air and dust in the environment. Treatment of the wood with pentachlorophenol is often found to be a significant source of PCDD/Fs contamination of eggs and poultry meat (Brambilla, Fochi et al. 2009, Piskorska-Pliszczynska, Strucinski et al. 2016). Open burning of waste can be another potential source of PCDD/Fs (Gullett, Wyrzykowska et al. 2010, Zhang, Buekens et al. 2017). We wanted to avoid such contamination therefore we asked fanciers whether their hens might be exposed by either of these pathways. Table 1 summarizes the basic data about the size of samples and measured levels of fat content in each of the pool samples. Two pool samples of free-range chicken eggs were analyzed in total, plus the one sample taken in Prague, where we bought chicken eggs in a supermarket, in May 2018. The last of the above-mentioned samples is used to exhibit background levels of POPs as suggested by Dvorská (2015). Samples of free-range chicken eggs were taken in October 2021.

Table 1: Overview of chicken egg samples presented in this report

No	Sample	Locality	Month/year of sampling	Eggs in pooled samples	Fat content
1	Kok-E-1, Kok-E-2, Kok-E-4, Kok-E-5	Kokšov – Bakša	10/2021	15	10.11
2	Val-E-1, Val-E-2, Val-E-3	Valalíky	10/2021	12	10.22
3	Praha - supermarket	Prague	05/2018	10	8.16

Sampled eggs were collected into typical plastic egg packaging. Eggs were boiled for approximately 10 minutes right after sampling. Cold eggs were then stored in refrigerator and kept in cold conditions also during the transport to the laboratory.

2.2. Chemical analysis

In the laboratory the edible part of eggs was homogenized and used for analyses. All samples were analyzed for content of individual PCDD/Fs and dioxin-like PCBs (dl PCBs) by GC/HRMS in ISO 17025 accredited laboratory at State Veterinary Institute, an accredited laboratory in Prague, Czech Republic at with a resolution >10,000 using ^{13}C isotope labelled standards. PCDD/F and dl-PCB analysis. PCDD/F and dl-PCB analysis followed the European Union's methods of analysis for the control of levels of PCDD/Fs and dl-PCBs for levels in certain foodstuffs in Commission Regulation (EC) No 252/2012 (European Commission 2012).

The samples were also analyzed for content of indicator congeners of PCBs (ndl PCBs), HCB, and PeCB in the same laboratory. The analytes were extracted by a mixture of organic solvents hexane: dichloromethane (1:1). The extracts were cleaned by means of gel permeation chromatography (GPC). The identification and quantification of the analyte was conducted by gas chromatography coupled with tandem mass spectrometry detection in electron ionization mode. Fat content in the eggs was measured as part the analyses as well.

3. THE EU LIMITS FOR POPS IN EGGS

Chicken eggs are common component of diet in almost each country of the world. There are limit values set for several POPs for chicken eggs in European Union. These levels are legally binding in all EU member states, including Slovakia. The limit values we used for free-range chicken eggs are summarized in Table 2.

Table 2: Limit concentration values for HCB, PCBs and PCDD/Fs in chicken eggs.

	EU ML[1]/MRL[2]	
Unit	pg /g fat	ng/ g fat
WHO-PCDD/Fs TEQ	2.5	-
WHO-PCDD/Fs - dl PCB TEQ	5.0	-
PCBs[3]	-	40
	ng/g fresh weight	
HCB[2]	20	

[1]EU Regulation (European Commission 2016) sets maximum levels for dioxins, dioxin-like PCBs and non dioxin-like PCBs in foodstuffs.

[2]Regulation (EC) N°149/2008 (European Commission 2008). Maximum residue level (MRL) means the upper-legal level of a concentration for a pesticide residue in or on food or feed set in accordance with the Regulation, based on good agricultural practice and the lowest consumer exposure necessary to protect vulnerable consumers.

[3]sum of PCB28, PCB52, PCB101, PCB138, PCB153 and PCB180

European Food Safety Agency (EFSA) has lowered tolerable weekly intake (TWI) of dioxins (PCDD/Fs) and dioxin-like PCBs (dl PCBs) to 2 pg TEQ/kg of body weight/week from previous 14 pg TEQ/kg of body weight/week (EFSA CONTAM 2018). There is idea to lower also limit values for PCDD/Fs and dl PCBs in food as logical follow up. It can be about 1 pg TEQ/g fat for chicken eggs (Malisch and Schaechtele 2019), however this step was not announced officially yet.

4. RESULTS

Pooled samples of eggs were analyzed for indicator PCBs congeners (ndl PCBs) and unintentionally produced POPs (UPOPs): hexachlorobenzene (HCB), pentachlorobenzene (PeCB), polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs), and dioxin-like PCBs (dl PCBs). The results are summarized in Table 3. We work with lower bound levels of PCDD/Fs and dl PCBs measured in this report. Zero is counted for congeners measured below LOQ in lower bounds. Upper bound levels can be found in brackets in Table 3.

Table 3: Summary of the results of the analyses of POPs measured in free-range eggs samples from the vicinity of MWI Košice and reference sample from Praha - supermarket.

Persistent organic pollutant	Units	Kokšov-Bakša	Valalíky	Praha - supermarket
hexachlorobenzene	ng/g	<1	<1	NA
PCB 28	ng/g fat	<2.000	<2.000	NA
PCB 52	ng/g fat	<1.000	<1.000	NA
PCB 101	ng/g fat	<1.000	<1.000	NA
PCB 153	ng/g fat	<2.000	<2.000	NA
PCB 138	ng/g fat	<2.000	<2.000	NA
PCB 180	ng/g fat	<1.000	<1.000	NA
ndl PCBs	**ng/g fat**	**<9.000**	**<9.000**	**NA**
pentachlorobenzene	ng/g	<1	<1	NA
2,3,7,8-TCDD	pg/g fat	<0.101	<0.101	<0.101
1,2,3,7,8-PeCDD	pg/g fat	<0.123	<0.123	<0.123
1,2,3,4,7,8-HxCDD	pg/g fat	0.191	<0.131	<0.131
1,2,3,6,7,8-HxCDD	pg/g fat	0.417	0.449	<0.109
1,2,3,7,8,9-HxCDD	pg/g fat	0.398	0.313	<0.115
1,2,3,4,6,7,8-HpCDD	pg/g fat	3.94	3.11	<0.265
OCDD	pg/g fat	5.92	6.21	0.757
2,3,7,8-TCDF	pg/g fat	0.395	0.601	0.321
1,2,3,7,8-PeCDF	pg/g fat	<0.134	0.24	<0.134
2,3,4,7,8-PeCDF	pg/g fat	0.288	0.532	<0.121
1,2,3,4,7,8-HxCDF	pg/g fat	<0.133	<0.133	<0.133
1,2,3,6,7,8-HxCDF	pg/g fat	<0.119	<0.119	<0.119
1,2,3,7,8,9-HxCDF	pg/g fat	<0.127	<0.127	<0.127
2,3,4,6,7,8-HxCDF	pg/g fat	<0.113	<0.113	<0.113
1,2,3,4,6,7,8-HpCDF	pg/g fat	0.54	<0.129	<0.129
1,2,3,4,7,8,9-HpCDF	pg/g fat	<0.125	<0.125	<0.546
OCDF	pg/g fat	<0.306	0.589	<0.306

Persistent organic pollutant	Units	Kokšov-Bakša	Valalíky	Praha - supermarket
WHO-PCDD/F-TEQ	**pg/g fat**	**0.273 (0.552)**	**0.336 (0.625)**	**0.032 (0.391)**
PCB 81	pg/g fat	1.65	3.56	<0.402
PCB 77	pg/g fat	19.9	32.8	4.74
PCB 123	pg/g fat	4.52	9.65	1.35
PCB 118	pg/g fat	400	1080	297
PCB 114	pg/g fat	7.71	17.5	3.24
PCB 105	pg/g fat	141	320	36.4
PCB 126	pg/g fat	4.79	12.1	1.74
PCB 167	pg/g fat	103	254	89.1
PCB 156	pg/g fat	209	486	218
PCB 157	pg/g fat	25.1	62.3	16.8
PCB 169	pg/g fat	0.656	1.13	<0.43
PCB 189	pg/g fat	41.8	88.9	51.1
WHO – dl PCB - TEQ	**pg/g fat**	**0.528**	**1.315**	**0.196 (0.208)**
WHO-PCDD/F + dl PCB-TEQ	**pg/g fat**	**0.801 (1.080)**	**1.651 (1.940)**	**0.228 (0.599)**

*upper bounds are in brackets. Full level of LOQ is included in calculation for congeners measured below LOQ in upper bounds.

4.1. Unintentionally produced POPs

4.1.1. Dioxins (PCDD/Fs) and PCBs

Dioxins belong to a group of 75 polychlorinated dibenzo-p-dioxin (PCDD) congeners and 135 polychlorinated dibenzofuran (PCDF) congeners, of which 17 are of toxicological concern. Polychlorinated biphenyls (PCBs) are a group of 209 different congeners which can be divided into two groups according to their toxicological properties: 12 congeners exhibit toxicological properties similar to dioxins, and are therefore often referred to as 'dioxin-like PCBs' (dl PCBs). The other PCBs do not exhibit dioxin-like toxicity but have a different toxicological profile and are referred to as 'non dioxin-like PCB' (ndl PCBs) (European Commission 2011). Levels of PCDD/Fs and dl PCBs are expressed in total WHO-TEQ calculated according toxic equivalency factors (TEFs) set by a WHO experts panel in 2005 (van den Berg, Birnbaum et al. 2006). These new TEFs were used to evaluate dioxin-like toxicity in samples of chicken eggs in this study.

Levels of PCDD/Fs and dl PCBs in free-range eggs from Kokšov-Bakša and Valalíky were below current limits set for eggs as food in the European Union at 2.5 pg TEQ/g fat and 5 pg TEQ/g fat for PCDD/Fs and PCDD/Fs + dl PCBs respectively (European Commission 2016). They would be close or above new potential ML of 1 pg TEQ/g fat as it is envisaged by scientists from the EU Reference Laboratory for halogenated POPs in feed and food (Malisch and Schaechtele 2019) if it would be established as follow up of more stringent TWI for dioxins and dioxin-like PCBs.

Levels of PCDD/Fs in eggs from Valalíky and Kokšov-Bakša exceed background level in eggs from supermarket by ten- and more than eight-folds respectively. They are ten-times lower than level measured in pooled egg sample from Kokšov-Bakša and Valalíky collected in 2005 (Petrlik, Hegyi et al. 2005).

4.1.2. PeCB and HCB

Pentachlorebenzene, hexachlorobenzene, and ndl PCBs were below LOQ in recent pooled egg samples from Kokšov-Bakša and Valalíky.

5. LIMITATIONS OF THE STUDY

Municipal waste incinerator Košice is considered to be potentially significant source of UPOPs. It obviously decreased levels of PCDD/Fs released in air emissions since 2005. Dioxins were at levels within the range of 0.002 – 0.024 ng TEQ/m^3 between the years 2015 and 2021 according the official reports published at MWI Košice website (KOSIT 2021), although these data are based on dis-continual measurements twice a year only. Data from another modern waste incinerator in Netherlands demonstrated that there might be large difference between the estimates of total dioxin releases based on such dis-continual measurements and semi-continual ones (Arkenbout 2019). Current limit for PCDD/Fs in air emissions is set at level of 0.1 ng TEQ/m^3 for waste incineration in EU, although new BAT – associated emission levels are more stringent and set them in the range < 0.01–0.06 ng TEQ/m^3 of PCDD/Fs for existing plants (European Commission 2019). It is well established that major flow of PCDD/Fs from modern municipal waste incinerators is rather via fly ashes (80% and more) than in air releases (single % units); (Zhang, Hai et al. 2012). Improper handling of waste incineration residues, and fly ashes in particular can lead to serious contamination of the food chain, including free-range chicken eggs (Katima, Bell et al. 2018). Recent estimation of total amount of dioxins transferred in waste incineration fly ashes revealed that it is underestimated flow (Petrlik, Kuepouo et al. 2021) of these toxic chemicals.

We were not able to monitor environmental levels of PCDD/Fs in the vicinity of the sites where fly ash from MWI Košice is transported and disposed of. We also do not know the levels of PCDD/Fs and other UPOPs in ashes produced by MWI Košice as this data is not publicly available if it exist at all. So we can consider this report as incomplete information about potential impact of the MWI Košice on PCDD/Fs levels in the environment only.

6. CONCLUSIONS

There were two pooled free-range chicken eggs samples collected in the vicinity of the MWI Košice and analyzed for unintentionally produced POPs, PCDD/Fs and dl PCBs in particular. Levels of PCDD/Fs + dl PCBs (0.801 and 1.651 pg TEQ/g fat) measured in these eggs did not exceed current EU maximum limits set for eggs as food. However this study cannot be considered as complete evaluation of potential impact of MWI Košice on levels of UPOPs in the environment because it was not possible to obtain environmental samples from the sites where waste incineration residues from MWI Košice are disposed of. All POPs and other toxic chemicals were not analyzed in sampled eggs because of time constrains and available laboratories capacities, in addition to previously mentioned limitations of this study.

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