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PFASs and Waste Incineration – New Data on Residues and Free-range Chicken Eggs

Nikola Jelinek[1,2], Many Calonzo[3], Penchom Saetang[4], *Jindrich Petrlik[1,2], Lee Bell[2], Aileen Lucero[3], Tomas Gramblicka[5], Jana Pulkrabova[5], Yuyun Ismawati[6], Annisa Maharani[6], Sarah Ozanova[1], Karolina Brabcova[1]

[1]Arnika – Toxics and Waste Programme, Seifertova 85, 130 00 Praha, Czech Republic jindrich.petrlik@arnika.org
[2] International Pollutants Elimination Network (IPEN), PO Box 7256 SE-402 35, Göteborg, Sweden
[3] EcoWaste Coalition, 78a Masigla Ext, Diliman, Quezon City, Metro Manila, Philippines
[4] Ecological Alert and Recovery Thailand (EARTH), 211/2, Ngamwongwan Rd. 31, Nonthaburi 11000, Thailand
[5] University of Chemistry and Technology, Prague, Faculty of Food and Biochemical Technology, Department of Food Analysis and Nutrition, Technicka 3, 166 28 Prague 6, Czech Republic
[6] Nexus3 Foundation, Mandalawangi No. 5, Denpasar 80223, Indonesia

1 Introduction

The term PFASs refers to per- and polyfluoroalkyl substances used both in industrial processes and household products, which have been manufactured since the 1950s. This group, comprising around 10,000 compounds[1], is primarily known for its use in Teflon or Gore-Tex and in paper food packaging, outdoor clothing, or carpets.[2,3] PFASs are continuously released into the environment and subsequently bioaccumulate in living organisms, most of them are either persistent themselves or act as precursors to other persistent compounds[4-7]. They can also be produced unintentionally in various industrial processes.[8] Some PFASs are considered suspected human carcinogens[9] and are linked to the reduction of women's fertility[10], preeclampsia (placental disease), or lower birth weights of newborns[11].

Municipal waste can contain materials contaminated with PFASs and/or other fluorinated compounds, leading to PFASs emissions and release during incineration. PFOS and PFOA were measured in air emissions from the WtE plant in Harlingen, Netherlands[12]; they were also detected in biological analyses of moss and needles near municipal solid waste incinerators (MSWIs)[13], as well as in soil in the vicinity of a hazardous waste incinerator (HWI) in East Liverpool, Ohio.[14] Other studies have concluded that the flue gas could be a significant source of PFASs emissions from WIs[15]. In recent years, several detailed studies have focused on PFASs in connection with waste incineration, demonstrating their presence in air emissions and waste incineration residues (WIRs).[16-18] One of the studies concluded that: *"PFAS are not fully degraded by the high temperatures during WtE conversion and can be emitted from the plant via ash, gypsum, treated process water, and flue gas"*[19].

This study aims to contribute to the mosaic of data on PFASs in WIRs, their detection around WI facilities, and the potential contamination of food chains from these sources.

2 Materials and Methods

Our study focused on analyses of bottom ash (BA), fly ash (FA), and environmental samples from the vicinity of their disposal or handling from previous studies[20-22] and supplemented them with new analyses of samples from Frydlant (Czechia) and Katwijk (Netherlands). We also collected previously published analyses of free-range chicken eggs for PFAS from the vicinity of WI facilities[20,21,23] and added new samples. We then compared the PFAS analyses in the eggs with a reference sample of free-range eggs from Bystricna in clean forest area in the Czech Republic and another sample from a supermarket in Jakarta, used as reference in previous studies[22-24].

Localities chosen for this study are a HWI in Aguado (Philippines), MSWIs in Phuket (Thailand), MSWI Merah Putih in Bantar Gebang (Indonesia), sites where BA and/or mixed BA and FA from MSWIs were used as construction materials for back-filling (Frydlant, Czechia) and/or road embankments (Katwijk, Netherlands). One site for free-range chicken egg sampling was identical to those chosen for the sampling of WIRs (Aguado, Phuket). However, the locations of Dumbrava, near used tires pyrolysis in Vatra (Moldova)[25], Sumberwuluh near HWI Lakardowo (Indonesia) and Vapenny Podol near a cement kiln co-incinerating alternative fuel made of mainly plastic municipal waste based in Prachovice (Czechia) were also added.

Pooled samples of 3 – 6 individual egg samples from free-range hens were collected from each of the sampled sites to obtain more representative samples. We also used a pooled egg sample from a supermarket to exhibit background levels of PFAS, following previous studies[26,27]. The eggs were collected into plastic egg packaging and boiled for 7 minutes. The homogenates from the edible parts of the eggs were analyzed.

Samples from residual ash, usually dumped near waste incinerators, and a sediment sample from a BA impacted area (specifically in Katwijk, Netherlands) were taken. These were also pooled samples from 5-point samples. Sediment was taken at a depth of 2 to 10 cm from the surface by a stainless-steel shovel. Samples were homogenized in a stainless-steel bowl. They were transported either in 250 ml PE plastic containers or in PE zip-lock bags to the laboratory and kept in cool conditions during storage and transportation. Also, building materials such as bricks were taken directly from the waste incinerator and/or users of these materials (villagers in Aguado) when available. A water sample was taken directly into a 500 ml PE plastic container from the water channel next to the site with WI BA in Katwijk.

A BA sample and a brick made of it, from an HWI burning mainly medical waste, were taken from Aguado[20], BA, and/or mixed BA and FA samples were taken from their dumping sites next to MSWIs in Phuket (three pooled samples in total)[21]. FA from inside the Merah Putih MSWI and a brick made of it were taken in Bantar Gebang[22]. Processed BA from Dutch

MSWIs used as construction material for roads and back-filling together with sediment and water samples were taken in Katwijk. Pooled egg samples were taken in Dumbrava village near a used tire pyrolysis plant in Vatra (Moldova)[25], two pooled egg samples from Aguado (Phillippines)[20], one sample from Phuket (Thailand)[21,28], one pooled egg sample from Sumberwuluh (Indonesia)[22,24], and two samples from the vicinity of a cement kiln in Prachovice (Czechia). Pooled eggs sample from supermarket in Jakarta was the only reference for which PFAS analyses were available in our previous studies.

Ultra high-performance liquid chromatography coupled to tandem mass spectrometry (UHPLC–MS/MS) was used to identify and quantify 16 and 31 PFASs in presented samples, respectively. The extraction of samples and analytical methods are described elsewhere.[29,30] The analyses performed before 2024 were for 16 PFAS.[1] This number was enlarged to 31 PFAS[2] in studies conducted in 2024.

3 Results

The results of the analyses of PFASs for WIRs, sediment, and water from a site affected by the use of BA in Katwijk are summarized in Table 1. Table 2 summarizes the results of the analyses for pooled egg samples. PFASs found only in levels below the limit of quantification (LOQ) and/or were not analyzed were excluded from the tables. They are listed in the notes below the tables.

The highest concentrations of PFASs at 67.75 and 24.08 ng/g dm, respectively, were found in WIR samples from the HWI in Aguado. A high concentration was also found in a water sample from Katwijk. Four samples of WIRs (PHU-1-OLD, BEK-ASH-01/23, BEK-WASTE-01/22 and KAT-A-1) had concentrations below LOQ.

The graphs in Figures 1 and 2 demonstrate the percentage representation of individual PFAS in the analyzed samples of WI residues, sediment, and water (Figure 1) or eggs (Figure 2). PFOS is the predominant PFAS in egg samples, but also, for example, in one of the BA samples from Katwijk (KAT-A-2). In WIRs, PFBS was also a significant representative, completely predominating in samples from Aguado but also relatively high in the ashes from incinerators in Phuket. PFHxA and PFPeA have a relatively high representation in WIR samples from Phuket (see Figure 1). Ash samples from Frydlant (FV-01) and Katwijk (KAT-24/1-A) had a similar composition of PFASs measured above LOQ, however their percentage of total PFASs was different. Short-chain PFASs prevailed in samples of WIRs from Aguado and Phuket but not in samples of WIRs from Frydlant and Katwijk. However, they prevailed in water samples from Katwijk mainly because of high levels of PFBA, PFHxA, and PFPeA.

In free-range chicken eggs from areas surrounding waste incineration facilities, PFASs values were 4 to 27 times higher than a reference sample from Jakarta (0.1 ng/g ww), ranging from 0.38 to 2.69 ng/g ww. The highest levels of 2.69 and 2.38 ng/g ww were measured in samples from Phuket (PHU-EGG) and Aguado (PH-E-3_4), respectively. The lowest level of 0.38 ng/g ww was in free-range eggs from Dumbrava. PFOS is the predominant substance in egg samples, except for a sample from Dumbrava in which PFBA was dominant (see Figure 2).

Table 1. Summarized results of the analyses for PFASs of the samples from Aguado, Phuket, Bantar Gebang and Katwijk. Results are in ng/g dry matter for ash and sediment samples and in ng/l for water samples.

Locality (country)	Aguado (Philippines)		Phuket (Thailand		Frydlant (Czechia)	Katwijk (Netherlands)			
Sample ID	PH-A-AB-1	PH-A-1	PHU-2-FA	PHU-3-BA	FV-01	KAT-A-2	KAT-24/1-A	KAT-24/3-SED	KAT-24/2-W
Matrix	brick from ash	BA	ash	ash	BA/FA	BA (modified)		sediment	water
Units	ng/g dm								ng/l
PFBA	0.72	<0.3	0.06	0.02	<0.3	<0.3	<0.3	<0.3	7.71
PFPeA	0.18	<0.3	0.10	0.03	0.03	<0.02	<0.02	<0.02	5.42
PFHxA	0.29	<0.3	0.11	0.04	0.04	<0.02	<0.02	<0.02	8.64
PFHpA	0.05	<0.3	0.04	<0.02	0.06	<0.02	<0.02	<0.02	3.32
PFOA	0.11	1.01	0.04	<0.02	0.23	<0.02	0.04	0.06	8.55
PFNA	0.04	<0.3	<0.02	<0.02	0.04	<0.02	0.06	<0.02	0.44
PFDA	0.05	<0.3	<0.02	<0.02	0.12	<0.02	0.30	0.06	0.24
PFUnDA	<0.02	<0.3	<0.02	<0.02	0.07	<0.02	0.08	0.04	<0.02
PFDoDA	<0.02	<0.3	<0.02	<0.02	0.25	<0.02	0.04	0.10	<0.02
PFTrDA	<0.02	<0.3	<0.02	<0.02	0.22	<0.02	<0.02	0.05	<0.2
PFTeDA	<0.02	<0.3	<0.02	<0.02	0.23	<0.02	<0.02	0.03	<0.2

[1] PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, PFBS, PFHxS, PFOS, PFDS, PFOSA

[2] Additional 15 PFAS to originally measured 16 are the following ones: PFHxDA, PFODA, PFPrS, PFPeS, PFHpS, PFNS, PFUnDS, PFDoDS, PFTrDS, N-MeFOSA, N-EtFOSA, HFPO-DA, ADONA, 9Cl-PF3ONS, 11Cl-PF3OUdS

PFPrS	0.06	NA	NA	NA	<0.02	<0.02	<0.02	<0.02	<0.02
PFBS	66.25	23.07	0.08	0.04	<0.02	<0.02	<0.02	<0.02	4.34
PFPeS	<0.02	NA	NA	NA	<0.02	<0.02	<0.02	<0.02	0.33
PFHxS	<0.02	<0.3	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	1.63
∑PFOS	<0.02	<0.3	<0.02	<0.02	0.11	0.02	0.10	0.08	4.91
PFOSA	<0.02	<0.3	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	NA
Total	67.75	24.08	0.43	0.13	1.40	0.02	0.62	0.43	45.58

Notes: Following PFAS were either <LOQ and/or not analyzed: PFHxDA, PFODA, PFHpS, PFNS, PFDS, PFUnDS, PFDoDS, PFTrDS, N-MeFOSA, N-EtFOSA, HFPO-DA, ADONA, 9Cl-PF3ONS, 11Cl-PF3OUdS. Samples PHU-1-OLD (Phuket), BEK-ASH-01/23 (FA), BEK-WASTE-01/22 (brick from FA; both Bantar Gebang) and KAT-A-1 (BA, Katwijk) had found PFASs <LOQ, and were not included in this summarizing table.

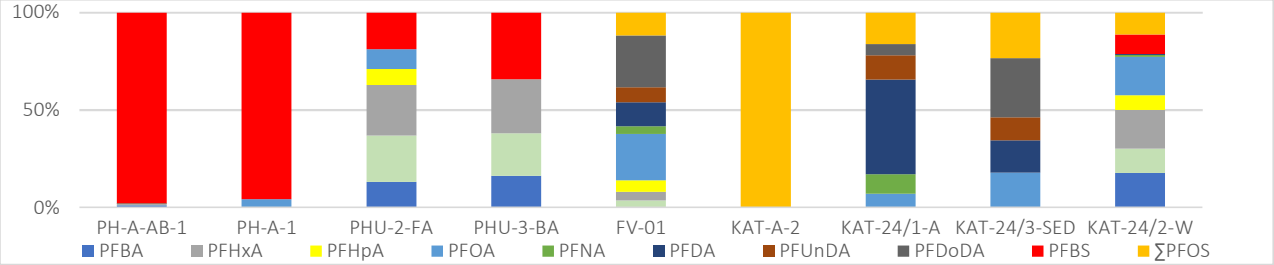

Figure 1: PFASs in WI residues from various locations and sediment and water sample from areas affected by use of WI BA for roads and embankments construction in Katwijk.

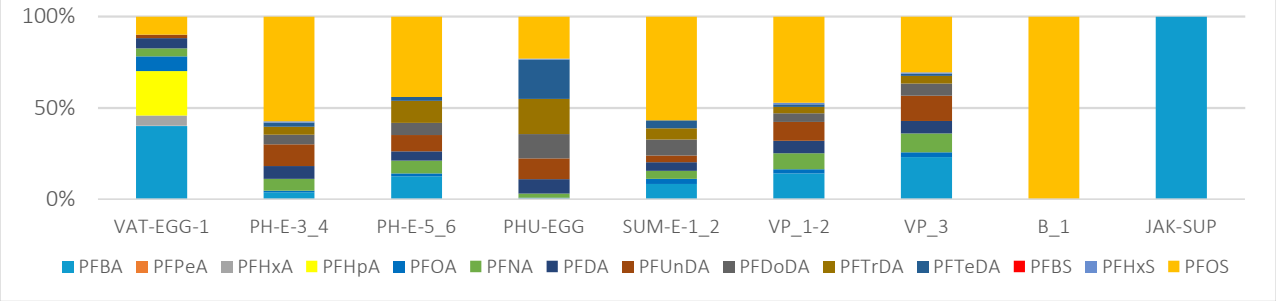

Figure 2: PFAS in free-range chicken egg samples from various locations near the sources of waste incineration and/or pyrolysis (Dumbrava, Moldova) in comparison with two reference samples.

Table 2. Summarized results of the analyses for PFASs in pooled free-range chicken egg samples from Dumbrava, Aguado, Phuket, Sumberwuluh and Vapenny Podol, plus reference samples of eggs from Bystrice and the supermarket in Jakarta (from hens kept inside enclosed farm buildings). Some of them were published in previous technical reports.[20,21,25]

Locality (country)	Dumbrava (Moldova)	Aguado (Philippines)		Phuket (Thailand)	Sumber-wuluh (Indonesia)	Vapenny Podol (Czechia)		Bystrice Ref.	Jakarta Ref.
Sample ID	VAT-EGG-1	PH-E-3_4	PH-E-5_6	PHU-EGG	SUM-E-1_2	VP_1-2	VP_3	B_1	JAK-SUP
PFOA	0.03	0.02	0.02	0.01	0.01	0.03	0.02	<0.006	<0.01
PFNA	0.02	0.15	0.06	0.06	0.02	0.13	0.10	<0.006	<0.01
PFHxS	<0.006	0.02	<0.01	0.02	<0.01	0.01	0.01	<0.006	<0.01
∑PFOS	0.04	1.37	0.41	0.61	0.26	0.70	0.28	0.065	<0.01
Total PFASs	0.38	2.38	0.94	2.69	0.46	1.48	0.93	0.065	0.10

In our study, PFBA levels of 0.21 and 0.15 ng/g ww were highest in eggs from Vapenny Podol and Dumbrava, respectively. Thus, PFBA was the predominant PFAS in the free-range egg sample from Dumbrava, followed by PFHpA, and PFOS was the third highest, which is different from the other samples presented in this study, where PFOS was dominant (see graph at Figure 2).

4 Discussion

Data from three studies on PFASs in WIRs are summarized in Table 3 and compared with data in this study. PFAS were studied in WIRs from three MSWI plants in southern China.[31] PFASs were found in all residual streams in a WtE plant in northern Sweden. Eight individual PFASs were detected, and short-chain PFCAs, mainly PFBA and PFHxA, were dominant in samples from the WtE plant.[19]