

Chicken eggs as an indicator of POPs pollution in Thailand

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Supporting data: Akarapon Teebthaisong, Autthaporn Ritthichat
Bangkok, Prague, November 2017



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This report was prepared and published as a part of the project “Increasing Transparency in Industrial Pollution Management through Citizen Science” funded by the European Union (EU) and co-funded by the Ministry of Foreign Affairs of the Czech Republic within the Framework of the Transition Promotion Programme – a financial assistance programme supporting democracy and human rights using the Czech Republic’s experience with social transition and democratization. Production of this publication was possible also thanks to Government of Sweden and Global Greengrants Fund.

The Project was implemented by Arnika – Toxics and Waste Programme based in Prague, Czech Republic and Ecological Alert and Recovery - Thailand (EARTH) based in Bangkok, Thailand as part of the work of Dioxin, PCBs and Waste working group of International POPs Elimination Network (IPEN).

The content of this publication does not reflect the official opinion of the European Union, Ministry of Foreign Affairs of the Czech Republic, Government of Sweden or any of the institutions providing financial support. Responsibility for the content lies entirely with authors.

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Chicken Eggs as an Indicator of POPs Pollution in Thailand

Results of sampling conducted in 2015 – 2016

Prague – Bangkok – 2017

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An updated report based on the results of environmental sampling conducted in Thailand in 2015–2016 as a part of the project “Increasing Transparency in Industrial Pollution Management Through Citizen Science,” financially supported by the European Union and Ministry of Foreign Affairs of the Czech Republic. We would also like to acknowledge financial support from the Global Greengrants Fund and the Government of Sweden, both of which contributed to conducting the chemical analyses and preparation of the report through the grant to IPEN.

This report is published in English.

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This report does not reflect the attitude of the donors – the European Union, the Ministry of Foreign Affairs of the Czech Republic, and Government of Sweden. Its content is the sole responsibility of the authors.

Bangkok - Prague, 2017

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1 Introduction

In this study, we present the results of monitoring free-range chicken eggs from selected sites in Thailand which are contaminated by persistent organic pollutants (POPs). Free-range chicken eggs were used for monitoring levels of contamination by POPs in various locations in many previous studies (Pless-Mulloli, Schilling et al. 2001a, Pirard, Focant et al. 2004, DiGangi and Petrlik 2005, Shelepchikov, Revich et al. 2006, Aslan, Kemal Korucu et al. 2010, Arkenbout 2014). Eggs have been found to be sensitive indicators of POP contamination in soils or dust and are a significant exposure pathway from soil pollution to humans. Eggs from contaminated areas can readily lead to exposures which exceed thresholds for the protection of human health (Van Eijkeren, Zeilmaker et al. 2006, Hoogenboom, ten Dam et al. 2014, Piskorska-Pliszczynska, Mikolajczyk et al. 2014). Chickens and their eggs might

therefore be ideal “active samplers”: an indicator species for evaluation of contamination levels of sampled areas by POPs, particularly by dioxins (PCDD/Fs) and PCBs. Based on this assumption, we have chosen a sampling of free-range chicken eggs and their analyses for selected POPs as one of the monitoring tools within the project “Increasing Transparency in Industrial Pollution Management through Citizen Science” (further information about the project can be found at <http://english.arnika.org/thailand> or <http://earththailand.org/en/>).

The data and analyses of free-range chicken eggs discussed in this report were obtained during several field visits in 2015 – 2016 as a result of the two above-mentioned joint projects of Thai and Czech NGOs. A general description of samples and a list of sampled localities can be found in Chapter 2. Major localities are described in the broader report about POPs in four hotspot areas (Mach, Teebthaisong et al. 2017) and in the report on mercury levels in fish from Thai hotspots (Tremlova 2017). These reports are also complementary in overall evaluation of chosen hotspots.

1.1 Acknowledgements

The field survey, sampling, analysis, writing, designing and printing of this publication was conducted as a part of the project “Increasing Transparency in Industrial Pollution Management Through Citizen Science,” financed by EU AID (EuropeAid 2014/350-604). This project was co-financed by the Czech Ministry of Foreign Affairs, the Swedish government through IPEN, the Global Greengrants Fund, and individual donors of each of the participating organization in the project. We are also grateful for the cooperation of the laboratories for their expert advice and quality assistance on chemical analyses, which often required their lab technicians to work overtime. The authors would also like to give sincere thanks and appreciation to the many individuals who helped us in putting this publication together, and let us namely thank at least some of them: Penchom Saetang, Director of Ecological Alert and Recovery Thailand (EARTH), Vaclav Mach, Arnika’s Expert, Jitka Strakova, Eva Hladikova, and Petra Slovackova, all from Arnika – Toxics and Waste Programme, Marek Sir and Martin Bystriansky, Chemical Experts, who helped with sampling in Thailand, and Cheryl Bowes, who helped us keep this publication in understandable English.

1.2 Abbreviations

BDS – BioDetection Systems (laboratory in Netherlands)

BEQ – bioanalytical toxic equivalent

CALUX – chemically activated luciferase gene expression

CAS – chemical abstracts service registry number (a unique numerical identifier assigned to every chemical substance described in open scientific literature)

BTBPE – 1, 2-bis (2,4,6-tribromo-fenoxy)ethane

DDD – dichlorodiphenyldichloroethane (a metabolite of DDT)

DDE – dichlorodipenyldichloroethylene (a chemical compound formed by the loss of hydrogen chloride from DDT)

DDT – dichlorodiphenyltrichloroethane (pesticide)

DI – dietary intake

DL PCBs – dioxin-like PCBs

d.w. – dry weight

EFSA – European Food Safety Agency

EU – European Union
f.w. – fresh weight
GC – gas chromatography
GEF – Global Environment Facility
GPC - gel permeation chromatography
GPS - global positioning system
HBB - hexabromobenzene
HCB – hexachlorobenzene
HCBD - hexachlorobutadiene
HCHs – hexachlorocyclohexanes (pesticides and their metabolites)
HpCDD – heptachlorodibenzo-p-dioxin
HpCDF – heptachlorodibenzo-p-furan
HRGC-HRMS – high resolution gas chromatography – high resolution mass spectroscopy
HxCDD – hexachlorodibenzo-p-dioxin
HxCDF – hexachlorodibenzo-p-furan
IPEN – International POPs Elimination Network
IARC - International Agency for Research on Cancer
INC – Intergovernmental Negotiating Committee (normally set up for negotiations of new international convention)
LOD – limit of detection
LOQ – limit of quantification
MAC – maximum acceptable (allowable) concentration
ML – maximum level
MRL – maximum residue level
NA – not analyzed
NGO – non-governmental organization (civil society organization)
NIP – National Implementation Plan of the Stockholm Convention
OBIND – octabromotrimethylfenylindane
OCPs – organochlorinated pesticides
OCDD – octachlorodibenzo-p-dioxin
OCDF – octachlorodibenzo-p-furan
PBDD/Fs – polyfrominated dibenzo-p-dioxins and furans
PBEB – pentabromoethylbenzene
PBT – pentabromotoluen
PCBs – polychlorinated biphenyls
PCDD/Fs – polychlorinated dibenzo-p-dioxins and furans
PCDDs – polychlorinated dibenzo-p-dioxins
PCDFs – polychlorinated furans
PeCB - pentachlorobenzene
PeCDD – pentachlorodibenzo-p-dioxin
PeCDF – pentachlorodibenzo-p-furan
POPs – persistent organic pollutants
SC – Stockholm Convention on Persistent Organic Pollutants
TEQ – toxic equivalent
TCDD – tetrachlorodibenzo-p-dioxin
TCDF – tetrachlorodibenzo-p-furan
TDI – tolerable daily intake

TEQ – toxic equivalent
TWI – tolerable week intake
UNDP – United Nations Development Programme
UNECE – United Nations Economic Commission for Europe
US EPA – United States Environmental Protection Agency
WHO – World Health Organization
WHO-TEQ – toxic equivalent defined by WHO experts panel in 2005
w.w. – wet weight

2 Sampling

Samples of free-range chicken eggs were collected at six localities in Eastern, Central, and Southern Thailand. One sample was taken from a supermarket in the city of Bangkok, considered as a background sample for Thailand, as suggested e.g. by Dvorská (2015). Chosen localities were expected to be contaminated by POPs to a certain level. The larger areas of Map Ta Phut, Samut Sakhon, Saraburi, Tha Tum and Khon Kaen were expected to be polluted by POPs as industry estate parks contain different types of industries. High levels of dioxins in sediments were collected in the Khon Kaen area in 2006 (Reungsang, Takizawa et al. 2006). Koh Samui is a popular tourist island; however, there is a large municipal solid waste landfill and an abandoned waste incinerator, both of which are expected to be potential sources of POPs pollution. The assumption on potential contamination by POPs was also based on data from the Dioxin Sampling and Analysis Program held in Thailand in 1997 (Fiedler 2001), and the Thai National Implementation Plan for the Stockholm Convention (Government of the Kingdom of Thailand 2007), as selected industries are considered and proven to be sources of unintentionally produced POPs. More information about the selected sites can be found in the following subchapter 2.1 and a broader report on POPs at selected hotspots (Mach, Teebthaisong et al. 2017).

2.1 Sampling Sites

A detailed description of the sampled sites and information about samples taken are provided in the report focused on POPs from four hotspots: Map Ta Phut, Samut Sakho, Tha Tum and Khon Kaen (Mach, Teebthaisong et al. 2017). In addition to these hotspots, there were also samples taken at Koh Samui and Saraburi. These two additional hotspots are briefly described in further text.

2.1.1 Koh Samui

Koh Samui is one of the most famous of Thailand's numerous islands. However, there is a large municipal waste landfill hidden in mountainous forest terrain in the southeastern part of the island, approximately 1.6 km from the sea shoreline. Additionally, a large municipal waste incinerator was built in 1997 for 501 million Baht (\$13.5 million USD) through a joint Thai-Japanese venture (CAIN, Greenpeace et al. 2006). The incinerator was never used to its full capacity of burning 140 tons of garbage per day. Operation ceased in 2012 due to problems including corrosion of the boiler tank, which caused the incinerator to run at half of its capacity (Ade, Brown et al. 2016). The waste incinerator was left abandoned in the middle of a growing waste landfill. We have chosen an area 0.5 km downstream from both the waste landfill and abandoned waste incinerator for sampling. The ash sample from the waste incinerator was analyzed for heavy metals and other contaminants by Greenpeace in

1999 (Labunska, Stephenson et al. 2000). Waste incinerator was also suspected to be a source of the soil contamination by mercury downwind from the facility (Muenhor, Satayavivad et al. 2009).

2.1.2 Saraburi

Saraburi is the area with big concentration of cement kilns owned by three major companies:

1. Siam City Cement Public Co., Ltd.
2. Siam Cement Group (SCG) – The Siam Cement Public Co., Ltd.
3. TPI Polene Public Co., Ltd.

It is located in three different districts: Kaeng Khoi (94,555 inhabitants), Ban Mo (total population 42,409 inhabitants) and Phra Phutthabat (total population 63,611 inhabitants); (National Statistical Office 2011). Saraburi belongs to the cities with most severe pollution by PM_{2.5} (Rujivanarom 2017).

2.2 Sampling and Analytical Methods

To obtain representative samples, pooled samples of individual egg samples were collected at each of the selected sampling sites. Table 1 summarizes the basic data about sample size and measured levels of fat content in each of the pool samples. Eleven pool samples of free-range chicken eggs were taken in total, plus the one sample taken in Bangkok, where we bought chicken eggs in a supermarket. The last of the above-mentioned samples are used to exhibit background levels of POPs as suggested by Dvorská (2015). Two samples were taken in 2015, and nine in the following year (2016).

Table 1: Overview of Free-Range Chicken Egg Samples From Selected Sites in Thailand.

No	Sample	Locality	Month/year of sampling	Eggs in pooled samples	Fat content
1	Samut Sakhon	Samut Sakhon	02/2015	3	11.6
2	SMS2-13	Samut Sakhon	02/2016	3	19.4
3	Tha Tum	Tha Tum	02/2015	4	12.5
4	SAR1	Saraburi	02/2016	5	11.1
5	KK1	Khon Kaen	02/2016	5	13.0
5a	KK1/1	Khon Kaen	02/2016	2	16.3
5b	KK1/2	Khon Kaen	02/2016	3	14.1
6	MTP2-18	Map Ta Phut	02/2016	3	14.7
7	MTP2-19	Map Ta Phut	02/2016	3	18.5
8	MTP1-11	Map Ta Phut	02/2016	4	18.2
9	Samui 01	Koh Samui	02/2016	4	14.1
10	Samui 02	Koh Samui	02/2016	4	14.7
11	Control group, supermarket	Bangkok	02/2016	6	11.6

Most of the samples were analyzed for content of individual PCDD/Fs, an extended list of PCB congeners, and 16 PAHs by HRGC-HRMS at Axys Varila, an accredited laboratory in Vrane nad Vltavou, Czech Republic.

Egg samples were also analyzed for content of OCPs, PeCB, HCB and BFRs in a certified Czech laboratory (Institute of Chemical Technology, Department of Food Chemistry and Analysis). 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. The method of ultra-performance liquid chromatography, coupled with tandem mass spectrometry detection (UHPLC–MS/MS), was chosen for the analysis of isomers of HBCD. The other BFRs were analyzed via previously mentioned technique.

3 The Thai and EU Limits for POPs in Eggs

Chicken eggs are a major component of the human diet, and it is common practice for Thai people to raise their own chickens. However, there is a limited number of Thai chemical laboratories, and this fact is likely to limit the values/hygienic standards that are set for limited number of POPs. We have not found limits for PCDD/Fs and PCBs in Thai legislation, so we had to compare the results of analyses for POPs with EU standards. The limit values we used for free-range chicken eggs are summarized in Table 2.

Table 2: Limit Concentration Values for OCPs, PCBs and PCDD/Fs TEQs in Chicken Eggs.

	Hen eggs		
	Thai	EU ML ² /MRL ³	
Unit	ng g ⁻¹	pg g ⁻¹ fat	ng g ⁻¹ fat
WHO-PCDD/Fs TEQ	None	2.5	-
WHO-PCDD/Fs-dl-PCB TEQ	None	5.0	-
PCBs ⁵	None	-	40
	ng g ⁻¹ fresh weight		
DDT total ⁶	100 ⁷	-	50 (fresh)
γ-HCH (lindane)	<LOD ⁸	-	10 fresh
α-, β-HCH**	none; <LOD ⁸	-	20, 10
HCB	<LOD ⁸	-	20 (fresh)

²EU Regulation (EC) N°1259/2011 (European Commission 2011) sets maximum levels for dioxins, dioxin-like PCBs and non dioxin-like PCBs in foodstuffs.

³Regulation (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.

⁵sum of PCB28, PCB52, PCB101, PCB138, PCB153 and PCB180

⁶sum of p,p'-DDT, o,p'-DDT, p,p'-DDE and p,p'-DDD

⁷Thai: Pesticide Residues: Extraneous Maximum Residue Limits (EMRL), Thai Agricultural Standard (TAS 9003 – 2004)

⁸Thai: Pesticide Residues: Extraneous Maximum Residue Limits (EMRL), Thai Agricultural Standard (TAS 9002 – 2016)

**for each isomer is MRL set separately

4 Results

Eleven pooled samples of eggs were analyzed for PCDD/Fs and DL PCBs in total, and twelve samples for other POPs¹, nine for OCPs and PAHs, eight for HCB and PeCB, and six for BFRs. GCMS-HRMS analyses were chosen for confirmation of contamination by dioxins and dioxin-like PCBs of sampled chicken eggs. The same samples were also analyzed for other POPs and OCPs: hexachlorobenzene (HCB), hexachlorocyclohexanes (HCHs) and DDT and its metabolites. HCB is also considered to be unintentionally produced POP (U-POP) in the same processes as dioxins and DL PCBs (Stockholm Convention on POPs 2008), although it is commonly measured together with other OCPs. Also, two other U-POPs, pentachlorobenzene (PeCB) and hexachlorobutadiene (HCBT) were analyzed in some samples. The results for U-POPs and OCPs are summarized in Table 3, for PAHs in Table 4 and for BFRs in Table 5.

Discussion about results of the analyses for POPs will focus on dioxins and dioxin-like POPs, PAHs and brominated flame retardants in this study. Other POPs such as, for example, organochlorine pesticides (OCPs) were already discussed sufficiently in the report by Mach, Teebthaisong et al. (2017).

4.1 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 eleven pooled samples of chicken eggs from Thailand.

Four out of eleven samples from Thailand exceeded EU ML of PCDD/Fs and/or PCDD/Fs and DL PCBs, expressed as WHO TEQ in chicken eggs (compare Tables 3 and 2; see graph at Figure 1); (European Commission 2011). The background levels for PCDD/Fs and DL PCBs measured in chicken eggs from a supermarket in Bangkok were 0.08 and 0.001 pg WHO-TEQ g⁻¹ fat, respectively. The highest level of dioxins (84.04 pg WHO-TEQ g⁻¹ fat) and DL PCBs (11.67 pg WHO TEQ g⁻¹ fat), respectively, was measured in eggs from Samut Sakhon, sampled in an area of a small artisanal recycling facility where metals are reclaimed from wastes after they are burned. The same sample was analyzed by the bioassay DR CALUX

¹ Sample KK1 is a pooled sample from KK1/1 and KK1/2. It comes from two different fanciers living close to each other. It was handled as two separate samples for certain analyses, while the others were handled as one pooled sample.

method and a level of 100 pg BEQ g⁻¹ fat was measured. It also contained high levels of polybrominated dioxins (see subchapter 4.2).

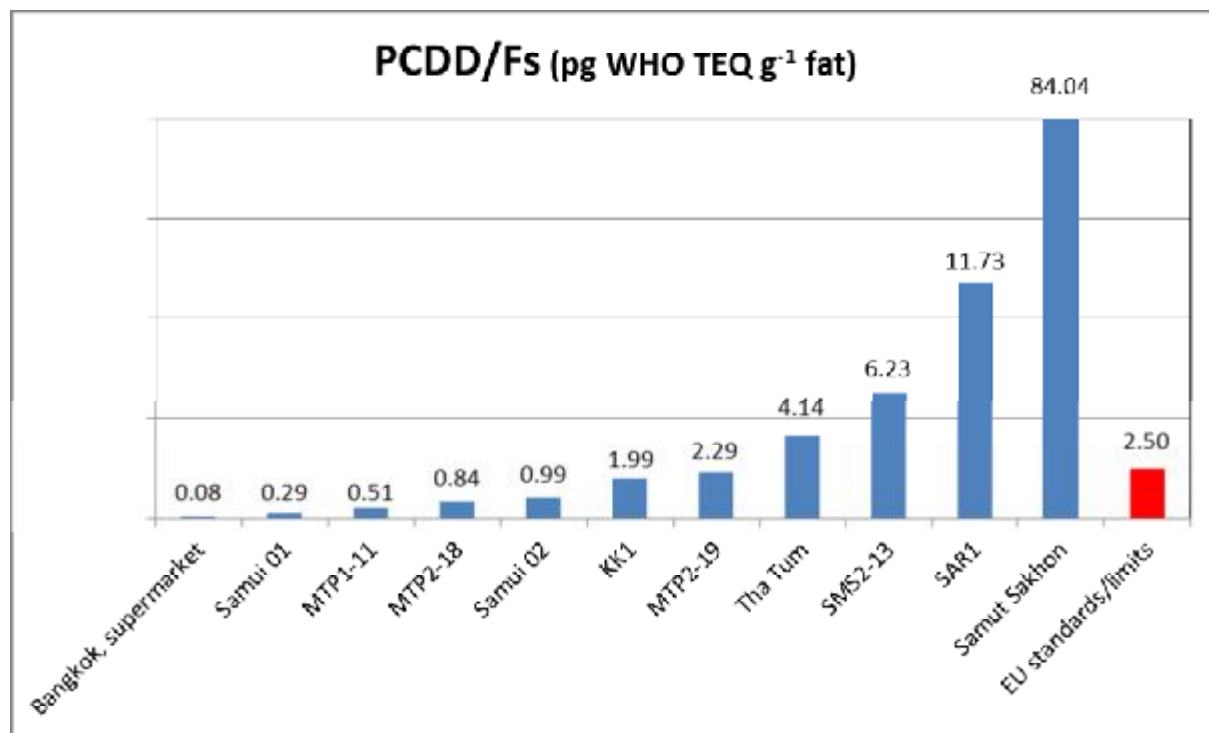


Figure 1: Graph of PCDD/Fs concentrations in chicken eggs samples from selected hot spots and the Bangkok supermarket in Thailand collected in 2015 – 2016 (in pg WHO-TEQ g⁻¹ fat)

Another egg sample from Samut Sakhon, located in an area close to “recycling” facilities that focused on secondary metal production, also exceeded the EU standard for dioxins in eggs.

From the collected egg samples exceeding the EU standard for dioxins, only those from an artisanal recycling facility area - as well as those from Tha Tum - were used for the chicken owners’ personal consumption. The others were meant to be used for chicken reproduction – for raising cockerels.

Total WHO-TEQ levels of PCDD/Fs and DL PCBs in samples from most Thai hotspots are lower than in selected samples from Chinese, Kazakhstani and Belorussian hotspots selected in other recent studies; however, the highest level of dioxins measured in eggs from Samut Sakhon belongs to the highest levels observed in free range chicken eggs during last years.

Places with higher egg contamination than found in Samut Sakhon were observed in Vietnam, in areas sprayed by herbicides contaminated by dioxins during Vietnam war (Hoang, Traag et al. 2014), and/or in Portugal, at a locality where pentachlorophenol was used as a wood preservative (Cardo, Castel-Branco et al. 2014). The dioxin level in the eggs sample from Samut Sakhon is comparable with levels observed in Pontypool, UK, near a hazardous waste incinerator (Lovett, Foxall et al. 1998), Kovachevo, Bulgaria (IPEN Dioxin PCBs and Waste Working Group, Za Zemiata et al. 2005) or Oroville, California, in an area where accidental fires in a pentachlorophenol wood treatment facility occurred (Chang, Hayward et al. 1989, Goldman, Harnly et al. 2000).

Table 3: Summarized results of analyses for POPs for eleven pooled free-range chicken eggs samples collected at selected localities in Thailand in a two-year period (2015 - 2016), plus a background sample from a supermarket in Bangkok, sampled in 2016.

Locality	Samut Sakhorn	Samut Sakhorn	Tha Thum	Saraburi	Khon Kaen	Khon Kaen	Khon Kaen	Map Ta Phut	Map Ta Phut	Map Ta Phut	Koh Samui	Koh Samui	Bangkok	EU
Sample	Samut Sakhorn	SMS2-13	Tha Thum	SAR1	KK1	KK1/1	KK1/2	MTP2-18	MTP2-19	MTP1-11	Samui 01	Samui 02	Control gr., superm.	standards /limits
Fat content	11.6	19.4	12.5	11.1	13	16.3	14.1	14.7	18.5	18.2	14.1	14.7	11.6	
PCDD/Fs (pg WHO TEQ g ⁻¹ fat)	84.04	6.23	4.14	11.73	1.99	NA	NA	0.84	2.29	0.51	0.29	0.99	0.08	2.50
DL PCBs (pg WHO TEQ g ⁻¹ fat)	11.67	6.00	3.94	6.71	0.83	NA	NA	0.73	0.95	1.61	0.01	0.001	0.001	
Total PCDD/F + DL PCBs (pg WHO TEQ g ⁻¹ fat)	95.71	12.23	8.09	18.44	2.82	NA	NA	1.57	3.24	2.12	0.30	0.99	0.08	5.00
HCB (ng g ⁻¹ fat)	4.21	NA	1.51	NA	NA	5.52	4.54	3.62	6.81	4.79	1.84	NA	<0.18	-
PeCB (ng g ⁻¹ fat)	NA	1.49	NA	NA	NA	<0.31	<0.35	<0.34	<0.27	<0.27	<0.35	NA	<0.43	
HCBD (ng g ⁻¹ fat)	NA	<0.26	NA	NA	NA	<0.31	<0.35	<0.34	<0.27	<0.27	<0.35	NA	<0.43	
7 PCB (ng g ⁻¹ fat)	12.97	8.07	0.39	2.68	1.16	NA	NA	1.11	4.46	0.64	0.91	1.27	0.22	-
6 PCB (ng g ⁻¹ fat)	11.40	7.08	0.39	2.23	1.00	NA	NA	0.95	3.54	0.50	0.72	1.07	0.22	40.00
Sum of HCH (ng g ⁻¹ fat)	0.31	NA	0.23	NA	NA	1.53	2.34	1.77	1.68	6.04	2.55	NA	0.52	
Sum of DDT (ng g ⁻¹ fat)	2.85	NA	0.83	NA	NA	10.06	20.43	19.25	8.43	8.08	37.80	NA	<LOQ***	

*** Levels of quantification for individual DDT metabolites varied between 0.25 – 2.15 ng/g fat.

Levels observed in one third of the samples were lower than the median level found in chicken eggs in a recent Chinese dietary intake study focused on PCDD/Fs and DL PCBs (Shen, Guan et al. 2017). The lowest levels of PCDD/Fs 0.29 and 0.99 pg WHO TEQ g⁻¹ fat, respectively, were observed in free-range chicken eggs collected at Koh Samui, even though these samples were collected in an area near a municipal waste landfill and abandoned waste incinerator covered by forest. All samples of free-range chicken eggs had levels of PCDD/Fs and DL PCBs, respectively higher than those observed in the pool sample of eggs bought in a Bangkok supermarket, which is used as a control sample showing background levels of PCDD/Fs and DL PCBs in chicken eggs from Thailand for this study. On this topic, see also discussion about background levels in other studies focused on POPs in free-range chicken eggs (DiGangi and Petrlik 2005, Petrlik, Kalmykov et al. 2017).

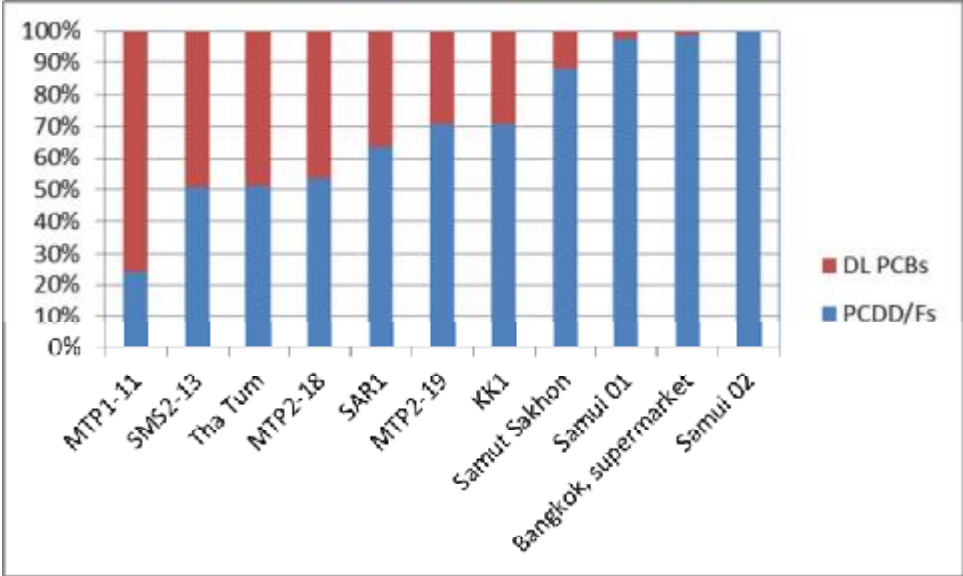


Figure 2: Graph showing balance between DL PCBs and PCDD/Fs expressed in WHO-TEQ levels in analyzed pooled egg samples from Thailand.

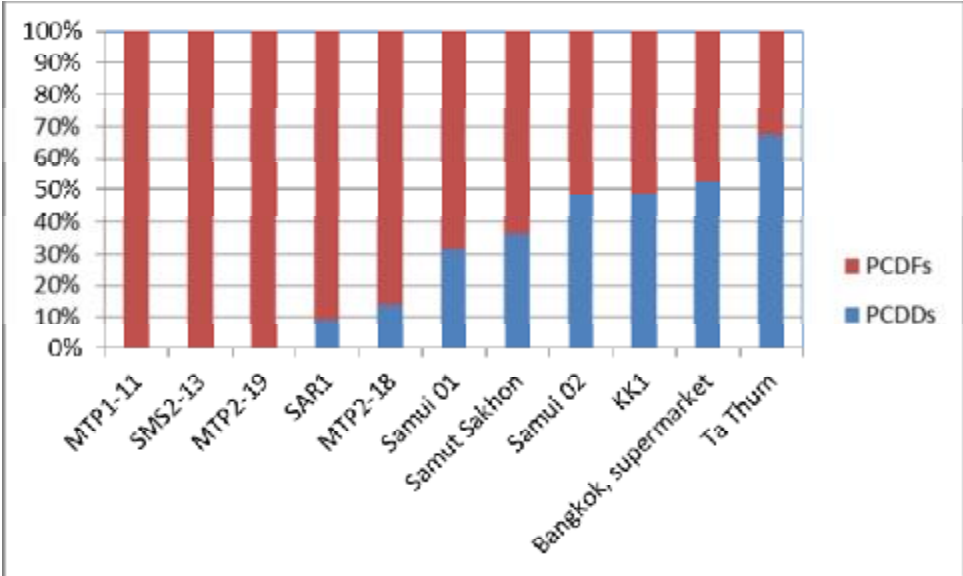


Figure 3: Graph showing balance between PCDDs and PCDFs from total WHO-TEQ levels of PCDD/Fs in analyzed pooled egg samples from Thailand.

PCDD/Fs cause a substantial part of the overall toxicity of chicken eggs in seven out of eleven samples analyzed in this study, and PCDFs prevail in the same portion of samples over PCDDs (see graphs at Figures 2 and 3). This is a completely different situation in comparison with the samples e.g. from Kazakhstan (Petrlík, Kalmykov et al. 2017). It reflects the fact that Thai environment, overall, is not so seriously contaminated by PCBs as it is in Kazakhstan and other countries, e.g. Czech Republic or Slovakia. Also indicator PCB congeners are not presented in high levels in almost all egg samples, except one from Samut Sakhon, in which the level of indicator congener PCBs reached one fourth of EU ML which is 40 ng g^{-1} fat (European Commission 2011), so it is still well below the EU limit for PCBs in eggs.

4.2 Polybrominated dioxins (PBDD/Fs)

Polybrominated dioxins and furans (PBDD/PBDFs) are formed from the brominated flame retardants present in e-waste plastic (Weber and Kuch 2003). Pollution by these dioxins can be expected at sites with e-waste plastic or cable burning, and likely result in the contamination of grazing animals at these sites. It has recently been established in a UK food survey that polybrominated dibenzo-p-dioxin and dibenzofurans can also contribute significantly to total dioxin exposure for the UK population (Mortimer, Acheampong et al. 2013). They are also formed as U-POPs in BFRs production as well as in waste incineration (Preud'Homme and Potin-Gautier 2002) and other combustion processes (Wang, Wang et al. 2010, Conesa, Ortuño et al. 2016) with the presence of brominated compounds.

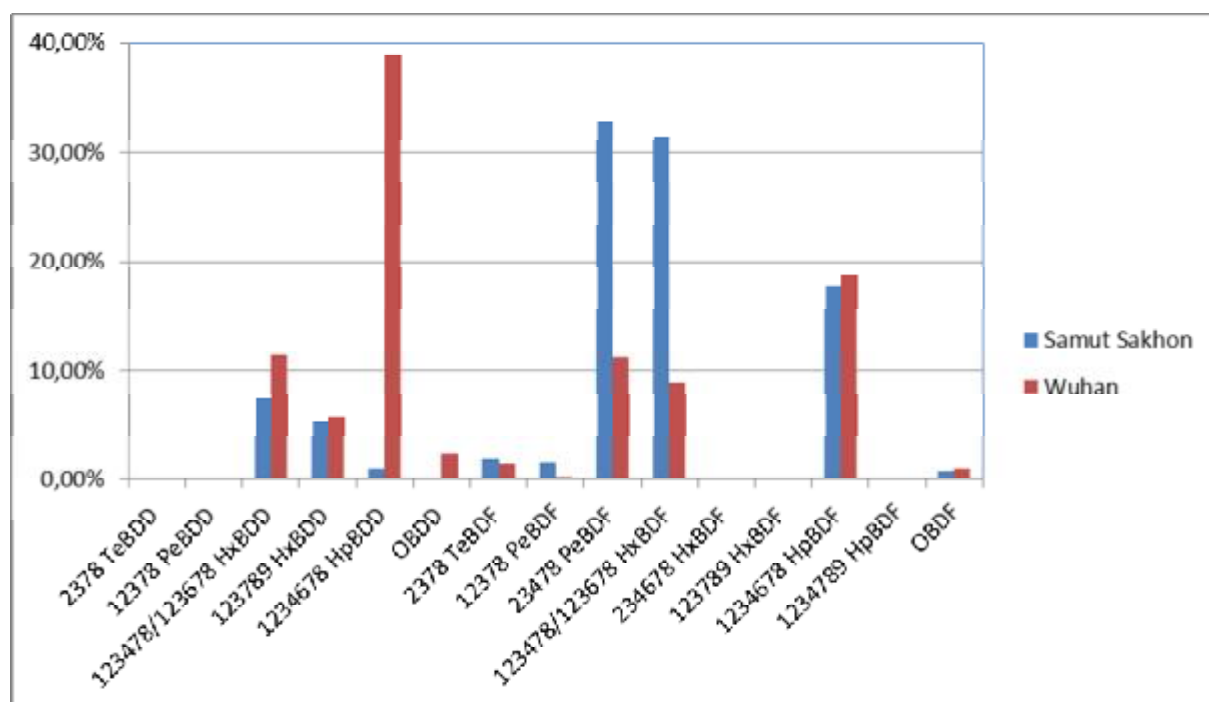


Figure 4: PBDD/Fs congeners profile in egg samples from Samut Sakhon, Thailand and Wuhan, China. Percentage calculation was made from balance of congeners on total WHO-TEQ level.

The eggs sampled in Samut Sakhon in February 2015 were also analyzed for PBDD/Fs. The level of these chemicals measured in these eggs at 15.8 – 22.9 pg WHO-TEQ g⁻¹ fat was the

second highest level ever measured in chicken eggs globally. A higher level of 27.3 – 29.2 was measured in free-range chicken eggs sampled in Wuhan, China in a neighborhood that contained a municipal waste incinerator (Petrlik 2015, Weber, Watson et al. 2015). The Figure 4 Graph shows the difference between brominated dioxin congeners profiles for the eggs sample from both Samut Sakhon and Wuhan. The presence of individually-detected congeners is the same, however, their balance is quite different.

4.3 Polycyclic aromatic hydrocarbons (PAHs)

Polycyclic aromatic hydrocarbons (PAHs) were analyzed in only nine samples of the total group of samples. Results of these analyses are summarized in Table 4, and a comparison of total sums of 16 PAHs between samples is in graph in Figure 5. Naphthalene and phenanthrene were the most dominant homologues from 16 measured PAHs in analyzed samples. Balance between these two PAHs in individual samples is shown in the graph in Figure 6.

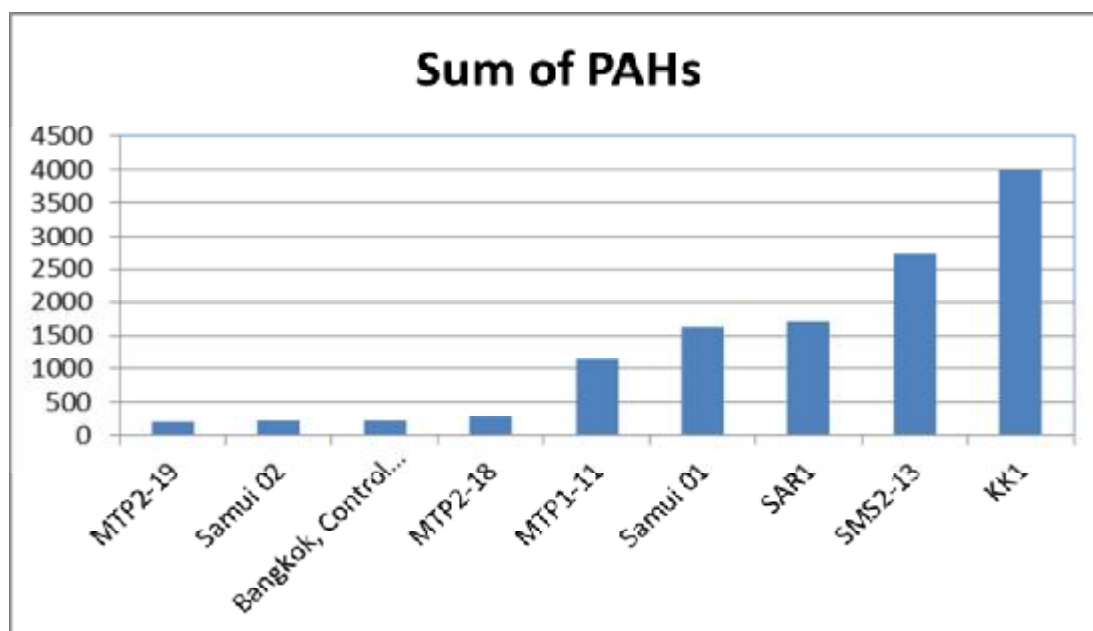


Figure 5: Comparison of total sums of 16 PAHs between chicken eggs samples from Thailand.

The highest level of PAHs was measured in eggs sample from Khon Kaen (3985 ng g⁻¹ fat) followed by the sample from Samut Sakhon (2730 ng g⁻¹ fat). PAHs in two samples from Map Ta Phut (MTP 2-18 and 2-19), and one sample from Koh Samui (Samui 02) were below or slightly above level of PAHs in eggs from the Bangkok supermarket. Apart from those samples, the levels of PAHs in other samples from selected localities in Thailand were much higher than in free-range chicken eggs from Mangystau Region, Kazakhstan, and some of them were also well above the level of PAHs in eggs from Likeng, China, sampled near a municipal waste incinerator.

Table 4: Summarized results of analyses for 16 PAHs in free -range chicken eggs samples from Thailand collected in 2015 – 2016 (in ng g⁻¹ fat).

Sample	SMS2-13	SAR1	KK1	MTP2-18	MTP2-19	MTP1-11	Bangkok, supermarket	Samui 01	Samui 02
Locality	Samut Sakhon	Saraburi	Khon Kaen	Map Ta Phut	Map Ta Phut	Map Ta Phut	Bangkok	Koh Samui	Koh Samui
Naphthalene	690	1300	3100	170	110	86	140	220	140
Acenaphthylene	< 30	62	42	< 30	< 30	< 20	< 30	33	< 30
Acenaphthene	< 30	75	< 30	< 30	< 30	< 20	< 30	< 30	< 30
Fluorene	50	130	270	< 30	< 30	< 20	< 30	41	< 30
Phenanthrene	1200	150	360	110	88	700	93	160	83
Anthracene	460	< 40	110	< 30	< 30	190	< 30	72	< 30
Fluoranthene	220	< 40	47	< 30	< 30	120	< 30	110	< 30
Pyrene	110	< 40	56	< 30	< 30	56	< 30	110	< 30
Benzo(a)anthracene	< 30	< 40	< 30	< 30	< 30	< 20	< 30	110	< 30
Chrysene	< 30	< 40	< 30	< 30	< 30	< 20	< 30	110	< 30
Benzo(b)fluoranthene	< 30	< 40	< 30	< 30	< 30	< 20	< 30	110	< 30
Benzo(k)fluoranthene	< 30	< 40	< 30	< 30	< 30	< 20	< 30	150	< 30
Benzo(a)pyrene	< 30	< 40	< 30	< 30	< 30	< 20	< 30	94	< 30
Indeno(1,2,3-cd)pyrene	< 30	< 40	< 30	< 30	< 30	< 20	< 30	100	< 30
Benzo(ghi)perylene	< 30	< 40	< 30	< 30	< 30	< 20	< 30	100	< 30
Dibenzo(a,h)anthracene	< 30	< 40	< 30	< 30	< 30	< 20	< 30	100	< 30
Sum of PAHs	2730	1717	3985	280	198	1152	233	1620	223

The potential source of egg contamination in Khon Kaen can be attributed to fly ash, which was distributed to the local people as fertilizer by Phoenix Pulp and Paper Plc. Ltd. Samples of chicken eggs were collected from fanciers where chickens had access to this ash; either spread out on gardens or in piles in front of their homes. Chickens would pick at their feed near these potential sources of contamination. Most likely, the source of egg contamination in Samut Sakhon is either secondary production of metals, or open-air burning of waste in the area that hens use to breed. It is necessary to note that the eggs sampled in that location in Samut Sakhon were not used for food consumption, but for raising new chickens – roosters in particular.

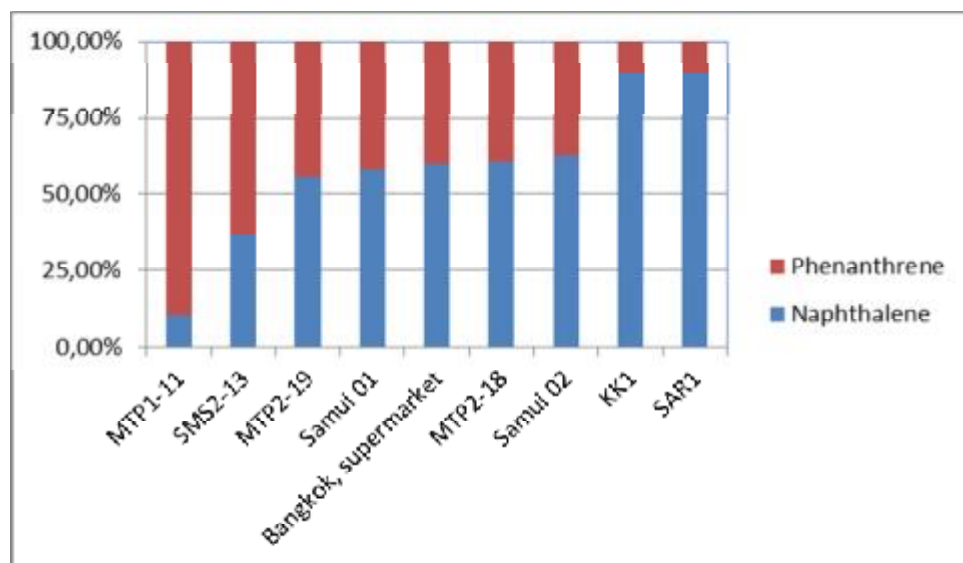


Figure 6: Balance between phenanthrene and naphthalene in chicken eggs samples from Thailand.

4.4 BFRs and polybrominated dioxins (PBDD/Fs) in eggs

Broad family of brominated flame retardants, including those used as alternatives to older ones such as polybrominated diphenyl ethers (PBDEs) and/or hexabromocyclododecane (HBCD), were analyzed in free-range chicken eggs from Samut Sakhon, Map Ta Phut, Koh Samui, as well as in a sample of eggs from the Bangkok supermarket. Results can be found in Table 5.

The highest level of PBDEs was measured in eggs sample from Samut Sakhon. The source of the contamination can be attributed to recycling facilities in the surrounding area where the eggs were sampled.

In an IPEN report focused on different hotspots, the maximum level of HBCD ($160 \text{ ng g}^{-1} \text{ fat}$) in free-range chicken eggs was observed in sample from the Dandora landfill in Nairobi. The highest level of PBDEs ($107 \text{ ng ng g}^{-1} \text{ fat}$) in that report was observed in a sample from the vicinity of a hazardous waste incinerator in Turkey (Blake 2005). The sample from Samut Sakhon had PBDE levels four times higher than the maximum levels found in the IPEN study; however, the study is more than ten years old. The level of HBCD in samples from several

sites is comparable to what was found at the Dandora landfill, but again, this is in comparison with a situation from ten years ago.

Table 5: Summarized results of analyses for different BFRs in chicken eggs samples from Central Kazakhstan, in comparison with selected samples from Thailand (in ng g⁻¹ fat). The levels below LOQ were counted as "0" for calculation of sums of the groups of BFRs.

Sample	SMS2-13	MTP2-18	MTP2-19	MTP1-11	Samui 01	Bangkok, supermarket
Locality	Samut Sakhon	Map Ta Phut	Map Ta Phut	Map Ta Phut	Samui	Bangkok
HBB	<0.26*	<0.20*	<0.16*	0.16	<0.21*	<0.26*
Sum of PBDEs **	426.89	1.09	3.30	44.95	0.92	3.10
PBEB	<0.26*	<0.20*	<0.16*	<0.16*	<0.21*	<0.26*
α-HBCD	159.18	38.91	165.41	184.12	<0.92*	<1.12*
β-HBCD	<1.28*	<0.88*	<0.70*	<0.71*	<0.92*	<1.12*
γ-HBCD	<1.28*	<0.88*	<0.70*	<0.71*	<0.92*	<1.12*
Suma HBCD	159.18	38.91	165.41	184.12	<LOQ	<LOQ
BTBPE	<0.26*	0.34	0.27	0.60	<0.21*	<0.26*
OBIND	<2.55*	<1.70*	<1.35*	<1.37*	<1.77*	<2.16*
DBDPE	<5.10*	<3.40*	<2.70*	<2.74*	<3.54*	<4.31*
Sum of new BFRs	<LOQ	0.34	0.27	0.60	<LOQ	<LOQ

*below LOQ

**Following congeners were analyzed: BDE 28, BDE 47, BDE 49, BDE 66, BDE 85, BDE 99, BDE 100, BDE 153, BDE 154, BDE 183, BDE 196, BDE 197, BDE 203, BDE 206, BDE 207 and BDE 209

In more than two occasions, poultry eggs in the vicinity of Chinese municipal solid waste incinerator (Petrlik 2015) and e-waste recycling site in Eastern China (Labunska, Harrad et al. 2013) registered PBDEs levels four times higher than those found in eggs from Samut Sakhon.

5 Discussion about potential exposure to POPs from chicken eggs

At some of the locations we received information that the eggs were not intended to be consumed as food, but for raising new chickens - roosters. So instead, we used the eggs for comparison of potential exposure, as they are commonly used for biomonitoring. The eggs collected as samples Samut Sakhon, Tha Tum, MTP 1-11, KK 1/1, KK 1/2, Samui 01 and Samui 02 were intended for food consumption.

Table 6: Summarized results of calculation of dietary intake of selected POPs by eating daily portion of eggs (26 g) from chickens raised at some Thai hotspots or eggs bought in a supermarket in Bangkok from chickens raised at a commercial farm. Twenty six grams of egg is the approximate current average consumption per person per day in Thailand based on calculation from available data (Knoema 2012). Total daily intakes of selected POPs from literature are given for comparison (see explanation below table).

Sample	Samut Sakhon	SMS2-13	Tha Thum	SAR1	KK1	MTP2-18	MTP2-19	MTP1-11	Samui 01	Samui 02	Bangkok, supermarket	Suggested levels
Data about samples of free range chicken eggs												
Fat content	11.6	12.5	19.4	11.1	13	14.7	18.5	18.2	11.6	14.1	14.7	-
PCDD/Fs + DL PCBs (pg WHO-TEQ g ⁻¹)	95.71	12.23	8.09	18.44	2.82	1.57	3.24	2.12	0.30	0.99	0.08	5*
PBDEs (ng g ⁻¹)	NA	426.89	NA	NA	NA	1.09	3.30	44.95	0.92	NA	3.10	-
16 PAHs (ng g ⁻¹)	NA	1890	NA	1450	3460	280	198	786	804	223	233	-
Daily intake of toxic chemicals from eggs per kg body weight by adults (DI _{adult})												
PCDD/Fs + DL PCBs (pg kg ⁻¹ bw)	4.12	0.88	0.38	0.76	0.14	0.09	0.22	0.14	0.02	0.05	0.00	2**
PBDEs (ng kg ⁻¹ bw)	NA	30.76	NA	NA	NA	0.06	0.23	3.04	0.05	NA	0.13	1.88***
16 PAHs (ng kg ⁻¹ bw)	NA	136	NA	60	167	15	14	53	42	12	10	-
Daily intake of toxic chemicals from eggs per kg body weight by approx. 10 years old child (body weight 35 kg); (DI _{child})												
PCDD/Fs + DL PCBs (pg kg ⁻¹ bw)	8.25	1.76	0.75	1.52	0.27	0.17	0.44	0.29	0.03	0.11	0.01	2**
PBDEs (ng kg ⁻¹ bw)	NA	61.52	NA	NA	NA	0.12	0.45	6.08	0.10	NA	0.27	1.88***
16 PAHs (ng kg ⁻¹ bw)	NA	272	NA	120	334	31	27	106	84	24	20	-

* EU Regulation (EC) N°1259/2011 (European Commission 2011) sets maximum levels for dioxins, dioxin-like PCBs and non dioxin-like PCBs in foodstuffs

** DI suggested by EFSA (European Commission 2001, Gies, Neumeier et al. 2007).

*** DI in Sweden (Lind, Aune et al. 2002)

The egg share in total food consumption in Thailand in 2007 was close to 1.5% of total food basket per day, according World Atlas – Food Security data ² (Knoema 2012), and its share changes were not constantly increasing or decreasing. It would mean that 2017 consumption could remain approximately the same, about 26 g per person per day if the trend continued. If we count 50 g per one chicken egg as average weight, it would mean consumption of ½ egg per person per day as general consumption pattern for the current Thai population.

We tried to calculate dietary intake for the following groups of contaminants per day: 1) PCDD/Fs plus DL PCBs; 2) PBDEs and 3) 16 PAHs. Calculation of daily intake levels was made by using following formula:

$$DI_{\text{adult}} = (((C \cdot F\%)/100) \cdot 26)/70;$$

$$DI_{\text{child}} = (((C \cdot F\%)/100) \cdot 26)/35,$$

where DI = daily intake; C = concentration of certain group of chemicals (PCDD/Fs, DL PCBs etc.), and F% = fat content in sample. Results are summarized in Table 6.

Results were then compared with available information about daily intake of evaluated chemicals: 1) PCDD/Fs + DL PCBs: Calculations for PCDD/Fs and DL PCBs were compared with TWI suggested by Scientific Committee on Food (SCF) at a level of 14 pg WHO-TEQ/kg of body weight per week = 2 pg WHO-TEQ/kg of b. w. per day (European Commission 2001, Gies, Neumeier et al. 2007). 2) PBDEs: There is an estimated available of daily intake of these two groups of BFRs in Sweden at the beginning of this century (Lind, Aune et al. 2002). We compared our data with those available in the Swedish study for egg consumption. 3) For 16 PAHs no DI value was found and used. See the comparison in Table 6.

Among samples intended to be used for consumption as food, the most critical levels of POPs were found in the sample from Samut Sakhon. Levels of PCDD/Fs, DL PCBs and PBDD/Fs were far too high in comparison with most of the samples collected in other countries during a new round of free-range chicken eggs sampling, organized by IPEN and Arnika (Petrlík 2015, Petrlík and Behnisch 2015, Petrlík, Kalmykov et al. 2015, Petrlík, Kalmykov et al. 2017). Just by eating approximately half an egg from the sample collected in an artisanal recycling facility area, one would exceed the daily intake of dioxins and DL PCBs by at least double, and this is without the inclusion of comparably toxic PBDD/Fs factored into this calculation. It is too much when we take into account that dioxins and DL PCBs are contained in other foods as well (Mach, Teebthaisong et al. 2017).

Also, intake of PCDD/Fs and DL PCBs from samples SMS 2-13 (Samut Sakhon), Tha Tum, and SAR1 (Saraburi) is at a troubling level.

The intake of PBDEs in eggs from SMS 2-13 (Samut Sakhon) would be very high, as well as from MTP 1-11, although it is ten times lower. By eating half an egg that was laid at both of

² The food consumption refers to the amount of food available for human consumption as estimated by the FAO Food Balance Sheets. However the actual food consumption may be lower than the quantity shown as food availability depending on the magnitude of wastage and losses of food in the household. Food consumption per person is the amount of food, in terms of quantity, for each individual in the total population. Food from eggs relates to the quantity of eggs used also for preparation the food such as bakery products.

these sites, one would exceed the daily intake by eating eggs in Sweden in 1998 – 99, as researched by Lind, Aune et al. (2002); (see Table 6).³

The highest dietary intake of PAHs can be obtained from eating eggs sampled in Khon Kaen (KK 1) and Samut Sakhon (SMS 2-13) followed by the sample from Saraburi (SAR 1).

6 Conclusions and recommendations

This study demonstrated some emerging problems of pollution by POPs in relation to the growing industry in Thailand. Some sites with a large concentration of industrial facilities, although there were many limitations⁴, create a broad and exhaustive picture of POPs in free-range chicken eggs in such a large country.

This study has discovered serious contamination within food chain by various POPs in Samut Sakhon, with a dense concentration of secondary metal production from artisanal recycling facilities, which quite often burn waste, including e-wastes, for reclamation of metals. These facilities are common sources of U-POPs (Stockholm Convention on POPs 2008) as well as BFRs. To prevent the release of these chemicals, Samut Sakhon would require better organization and regulation of such facilities. Existing facilities should be replaced by more appropriate recycling facilities that utilize clean technologies. It would require assistance from the authorities and potentially the state. It could be included among other actions into the updated NIP for the Stockholm Convention on POPs.

The eggs sampled in Samut Sakhon in February 2015 were also analyzed for PBDD/Fs. The level of these chemicals measured in the eggs was the second highest level ever measured in chicken eggs globally.

Relatively low levels of indicator PCBs in free-range chicken eggs show very low contamination levels by technical PCBs used in transformer oils, which confirms findings in NIP (Government of the Kingdom of Thailand 2007).

MTP 1-11 was more contaminated by PAHs and BFRs than the other two samples from Map Ta Phut area. Additional analyses from the same sampling location should highlight whether contamination was brought via canal from the industrial area located north of this site, and whether it is located on the seashore or not. Mainly this sample in addition with significant levels of PCDD/Fs and PAHs in another sample confirms that the concentration of the chemical industry brings more pollution by POPs in the area as well. There are many chemical factories producing, for example, PVC or specialized chemical products that were recognized to be significant sources of U-POPs (Stockholm Convention on POPs 2008, UNEP

³ The study by Törnkvist et al. (2011) shows that the levels, and intake, of different POPs (including HBCD) from food of animal origin, in the Swedish market basket of 2005 seem to have decreased since the market basket study in 1999. Törnkvist, A., A. Glynn, M. Aune, P. O. Darnerud and E. H. Ankarberg (2011). "PCDD/F, PCB, PBDE, HBCD and chlorinated pesticides in a Swedish market basket from 2005 – Levels and dietary intake estimations." *Chemosphere* 83(2): 193-199..

⁴ Limitations of the study are discussed in chapter 7.

and Stockholm Convention 2013). Three samples of free-range chicken eggs cannot give us the full picture of potential pollution of an area by POPs.

Soil additive containing ash from the power plant in Khon Kaen is most likely the source of high level of PAHs measured in free-range chicken eggs collected there. The eggs contained the highest level of PAHs among all samples from Thailand. This practice cannot be considered as environmentally sound management of coal burning power plant residue.

Chicken eggs are an important part of the human diet. The eggs from localities polluted by POPs can significantly burden the body, as demonstrated in Chapter 6 of this report, although eggs are a less common in food for the Thai population, in comparison with some other countries. The solution is not to discourage the public from eating chicken eggs or punishing the chicken farmers, but to prevent further contamination of the food chain at certain hotspots by addressing the issues of POPs being released into the environment.

We used chicken eggs, as they are a proven indicator of potential contamination within the food chain. We didn't sample meat but results of some other studies demonstrated simultaneous contamination of chicken eggs and meat from contaminated sites (Chang, Hayward et al. 1989, Lovett, Foxall et al. 1998).

7 Limitations of the study

The major limitations of the study were the limited financial, temporal and personal resources. Therefore, only a limited number of chicken egg samples could be taken, and only a limited scale of analyses could be conducted. We could not repeat whole ranges of analyses for each sample, which created some loopholes in the data available for our final evaluation.

We also lack data about the level of total dietary intake of different contaminants in Thailand, and for some even globally, e.g. PAHs. We worked with the limited information available instead. Still, an impression of the situation, including the identification of major issues in relation to potential pathways of contaminants into food chain (represented by free range chicken eggs in this study) in Central, Eastern and Southern Thailand, was obtained. However, future investigations in this field are still necessary. The results presented here cannot be considered exhaustive; rather, expressing the need for extended research in future.

The comparison of pollutant concentration levels found in the samples with legal standards also has its limitations. Each of the legal standards is defined in a different way, and for a different purpose. In addition, there are no existing legal standards for some of the pollutants and some legal limits or TDI levels might be outdated. The estimation of a potential risk to humans and the environment cannot be conducted by consulting legal standards only; an extensive risk analysis based on a sufficient number of samples and detailed description of the state of the area and the potential risk receivers is crucial. We tried to draw a basic evaluation of the health risk expressed as the daily intake of some crucial pollutants through consumption of eggs from free range-chickens raised at selected hotspots, in order to give at least a basic idea about the level of human exposure to different pollutants.

We believe that it is of the utmost importance to begin to address the overall pollution by such contaminants as PCDD/Fs, BFRs as well as by PAHs in Thailand.

8 Photos



Photos 1 and 2: Samut Sakhon. Characteristics of the place where most contaminated free range chicken eggs were collected. Children are most vulnerable members of community living in Samut Sakhon. They play and live right next to „recycling“ facilities, where workers with almost no protective suits sort metals out of burned wastes. Photos by: Jitka Strakova and Jindrich Petrlik, Arnika, 2015.



Photo 3: Saraburi, nearest cement kiln to free range chicken eggs sampling site in Saraburi. Photo by Ondřej Petrlik, Feb-2016.



Photos 4 - 7: Saraburi. Sampling of free range chicken eggs in Saraburi. Photos by: Jindřich Petrlík, Ondřej Petrlík and Karnt Thassanaphak.



Photo 8: Typical picture from Samut Sakhon with many small and medium size metal smelters and artisanal recycling facilities. Photo by Ondřej Petrlík, Feb-2016.



Photo 9: Night in Map Ta Phut. Photo by Ondřej Petrlík, Feb-2016.



Photo 10: Petrochemical complex in Map Ta Phut. Photo by Ondřej Petrlík, Feb-2016.



Photos 11 – 12: Khon Kaen. Sampling at site where fly ash was used as soil additive. As his visible as a pile in front of the house, where free range chicken eggs were collected. Photos by Jindřich Petrlík, Feb-2016.



Photo 13: Industrial landscape in Khon Kaen, and fly ash again. Photo by Jindřich Petrlík, Feb-2016.

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This publication has been produced with the support of the European Union (EU) as a part of the project "Increasing Transparency in Industrial Pollution Management through Citizen Science". The content of this publication does not reflect the official opinion of the European Union. Responsibility for the information and views expressed in the publication lies entirely with the authors.