Persistent Organic Pollutants (POPs) in Chicken Eggs from Alaverdi, Armenia

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Yerevan - Prague – 2018

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Results of sampling conducted in 2018

Yerevan – Prague – 2018

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This report is based on the results of environmental sampling conducted in Armenia in July 2018 as a part of the project "Involvement of Civil Society in the Decision-making Process on Mining in Armenia", financially supported by the Ministry of Foreign Affairs of the Czech Republic and the Global Greengrants Fund.

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Contents

1 Introduction

In this study, we present the results of the monitoring of free-range hens' eggs from Alaverdi, a town in the north of Armenia, close to the border with Georgia, which was considered to be potentially contaminated by persistent organic pollutants (POPs). Free-range hens' eggs were used for monitoring levels of contamination by POPs in various locations in many previous studies [1-6]. 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 exposure which exceeds thresholds for the protection of human health [7-9]. Hens and their eggs might therefore be ideal "active samplers": an indicator species for the evaluation of levels of contamination of sampled areas by POPs, particularly by dioxins (PCDD/Fs) and PCBs. On the basis of this assumption, we have chosen a sampling of free-range hens' eggs and their analyses for selected POPs as one of the monitoring tools within the project "Involvement of Civil Society in the Decision-making Process on Mining in Armenia" (further information about the project can be found at http://english.arnika.org/armenia,http://www.awhhe.am/, https://www.ecolur.org/.).

The data and analyses of free-range hens' eggs discussed in this report were obtained during field visits in July 2018 as a result of the above-mentioned joint project of Armenian and Czech NGOs. A general description of the samples and of the locality they were taken in can be found in Chapter 2.

1.1 Acknowledgements

The field survey, sampling, analysis, writing, designing, and printing of this publication was conducted as a part of the project "Involvement of Civil Society in the Decision-making Process on Mining in Armenia" financed by the Czech Ministry of Foreign Affairs, and co-financed by IPEN, the Global Greengrants Fund, and individual donors of each of the organisations participating in the project. We are also grateful for the cooperation of the laboratories in 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 thank at least some of them by name: Kristina Žulkovská from Arnika – Toxics and Waste Programme, Marek Sir and Martin Bystriansky, Chemical Experts, who helped with the sampling in Armenia, and Simon Gill, who helped us keep this publication in understandable English.

1.2 Abbreviations

BDS – BioDetection Systems (laboratory in the 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 the open scientific literature)
DDD – dichlorodiphenyldichloroethane (a metabolite of DDT)
DDE – dichlorodiphenyldichloroethylene (a chemical compound formed by the loss of hydrogen chloride from DDT)
DDT – dichlorodiphenyltricholoroethane (a 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
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)
iPCBs – indicator PCBs (this mostly covers six PCB congeners (PCB28, PCB52, PCB101, PCB138, PCB153, and PCB180)
LOD – limit of detection
LOQ – limit of quantification
MAC – maximum acceptable (allowable) concentration
ML – maximum level
MRL – maximum residue level
NA – not analysed
NGO – non-governmental organisation (civil society organization)
NIP – National Implementation Plan of the Stockholm Convention
OCPs – organochlorinated pesticides
OCDD – octachlorodibenzo-p-dioxin
OCDF – octachlorodibenzo-p-furan
PBDD/Fs – polyfrominated dibenzo-p-dioxins and furans
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
U-POPs – unintentionally produced POPs (by-products of different processes including incineration and/or burning of halogenated materials)
US EPA – United States Environmental Protection Agency
WHO – World Health Organisation
WHO-TEQ – toxic equivalent defined by a panel of WHO experts in 2005
w.w. – wet weight

2 Sampling

Samples of free-range hens' eggs were collected at two localities in Alaverdi, a town located on the northern borders of Armenia. One sample was taken from a supermarket in the city of Yerevan, considered as a background sample for Armenia, as suggested e.g. by Dvorská [10]. The localities that were chosen were expected to be influenced by pollution from a copper smelter as they are within the range of its common plume area. Primary copper production is listed as one of the potential sources of dioxins (PCDD/Fs) and dioxin-like PCBs [11-13].

2.1 Description of the locality

According to the Armenian National Implementation Plan for the Stockholm Convention on persistent organic pollutants (POPs): *"Major mining, mining and processing, and metallurgical enterprises were located in the towns of Alaverdi and Zangezur. Non-ferrous metallurgy was one of the most important branches of industrial development. In the early 1970s the development of ferrous metallurgy began in Armenia."*

Armenian Copper Programme, CJSC, is a metallurgical plant from the union of the Vallex group. It is situated in the town of Alaverdi, which is in the Lori region in the north-east of Armenia. The town is situated at the bottom of the Debed river gorge. The town has an approximate population of 11,000 (2016). Since the end of the 18th century, the town has been home to a copper smelting plant.

The Alaverdi copper smelter is able to produce about 12,000 tonnes of blister copper annually. The peak of production was achieved in the 1980s, when nearly 55,000 tons of refined copper were produced annually. The company has more than 500 employees.

A dominant feature of the factory is a chimney that smokes non-stop on the hill above the factory. The smoke from the smokestack covers a big part of the town of Alaverdi and the surrounding villages. The smelter is a potential producer and emitter of heavy metals.

Several studies [14-16] shown higher concetrations of lead, arsenic, copper, and zinc in the soil, water, and even human tissue samples in Alaverdi. It was also chosen as one of the toxic hot spots studied in a joint project of Arnika and Armenian Women for Health and a Healthy Environment (AWHHE), conducted in 2010 and 2011 [17].

2.2 Sampling and Analytical Methods

To obtain representative samples, pooled egg samples were collected at each of the selected sampling sites. All the eggs that were sampled originate from free-range hens, with the exception of the eggs bought in a supermarket. Table 1 summarises the basic data about sample size and the measured levels of fat content in each of the pooled samples. Two pool samples of free-range hens' eggs were taken and analysed in total, plus one sample taken in Yerevan, where we bought hens' eggs in a supermarket. The last of the above-mentioned samples is used to exhibit background levels of POPs, as suggested by Dvorská [10]. All the samples were taken in July 2018.

Table 1: Overview of hens' egg samples from selected sites in Armenia.

No	Sample	Locality	Month/year of sampling	Eggs in pooled samples	Fat content
1	Alaverdi 1	Alaverdi	07/2018	3	14.3
2	Alaverdi 3	Alaverdi	07/2018	4	13.1
3	Yerevan – supermarket	Yerevan	07/2018	4	8.7

The Alaverdi 1 sampling site is located 0.9 km to the south-east of the copper smelter, in the valley. Alaverdi 3 is in the town of Alaverdi, in a new part, on top of the hill, 1.3 km to the south of the copper smelter. The Alaverdi 3 sample was chosen because the prevailing winds blow exactly in the direction of that sampling site (see Figure 1).

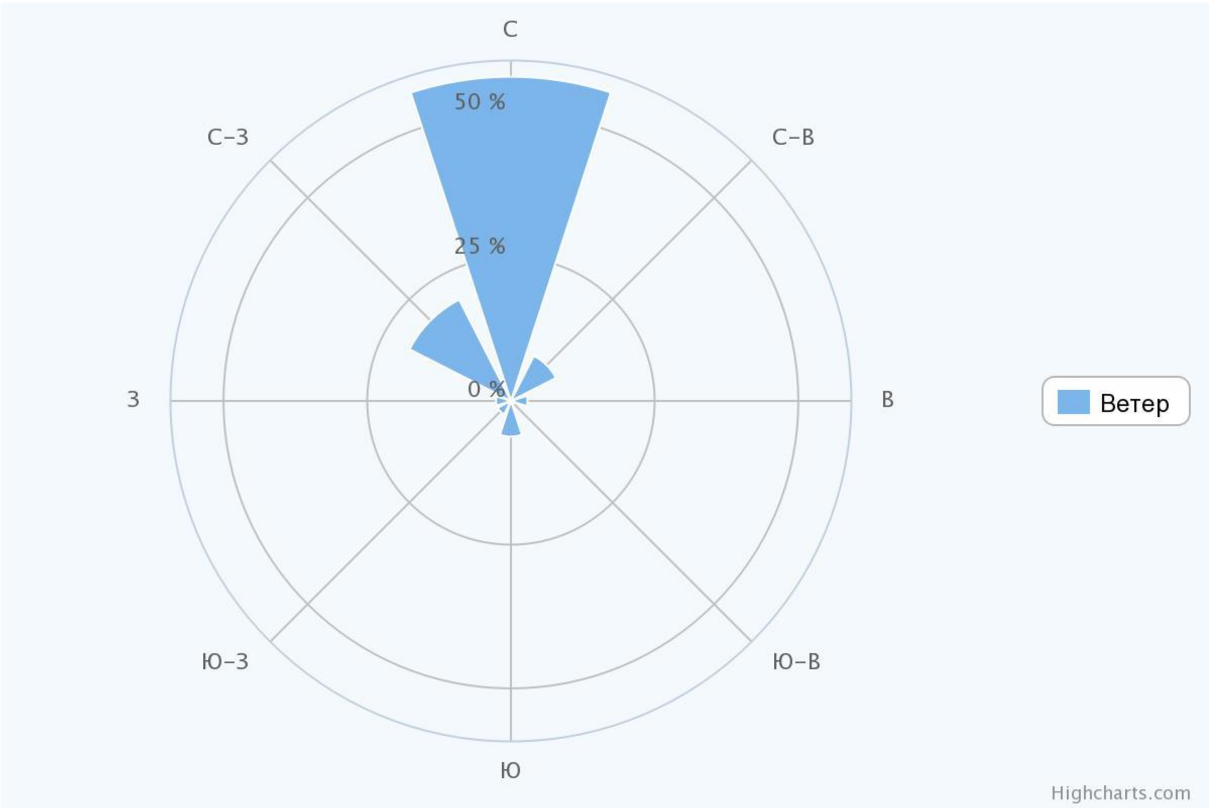


Figure 1: Windrose for Alaverdi. Source: [18].

The sampled eggs were collected into typical plastic egg packaging. They were boiled for approximately 7 minutes right after sampling. The cold eggs were then stored in a refrigerator and also kept in cold conditions during their journey to the laboratory. In the first laboratory the edible part of the eggs was homogenised and the same homogenised egg pool sample was used for the analyses in both laboratories.

All samples were analysed for their content of individual PCDD/Fs and dioxin-like PCBs (DL PCBs) by GC/HRMS in an ISO 17025 accredited laboratory at the State Veterinary Institute, an accredited laboratory in Prague, Czech Republic, with a resolution >10,000 using ^{13}C isotope labelled standards. 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 [19].

The samples were also analysed for their content of indicator congeners of PCBs (iPCBs), OCPs, PeCB, and HCBD in a certified Czech laboratory (Institute of Chemical Technology, Department of Food Chemistry and Analysis). The analytes were extracted with 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 ionisation mode.

3 The Armenian, Russian, and EU Limits for POPs in Eggs

Hens' eggs are a common component of the diet in almost every country of the world. Their proportion within the dietary basket differs from country to country. Armenia belongs among the countries with a lower level of egg consumption; however, their share in the diet has increased since the beginning of this century [20]. It is also common practice for Armenian people to raise their own hens.

There are limit values set for several POPs for hens' eggs in Armenia. We also used limits set either in Russia as Armenia is one of the countries in the Euroasian Economic Union (EAEU), in which the Russian Federation is a leading country. In addition to that, we also compared the results of analyses with limits set in the European Union (EU) because we also use the tolerable weekly intake set in the EU for the evaluation of exposure to PCDD/Fs and DL PCBs contained in eggs from Alaverdi (see Chapter 5). The limit values we used for free-range hens' eggs are summarised in Table 2.

Table 2: Limit concentration values for OCPs, PCBs, and PCDD/Fs TEQs in hens' eggs.

	Hen eggs			
	Armenian[1]	**Russian[2,3]**	**EU ML[4]/MRL[5]**	
Unit	$pg\ g^{-1}$ fat	$pg\ g^{-1}$ fat	$pg\ g^{-1}$ fat	$ng\ g^{-1}$ fat
WHO-PCDD/Fs TEQ	3.0	3.0	2.5	-
WHO-PCDD/Fs-dl-PCB TEQ	None	None	5.0	-
PCBs[6]	None	None	-	40
	$ng\ g^{-1}$ fresh weight			
DDT[7]	100	100 [8]	-	50 (fresh) [9]
γ-HCH (lindane)	None	100	-	10 fresh
α-, β-HCH**	None	100	-	20, 10
HCH[10]	100	None		None
HCB	None	None	-	20 (fresh)

[1]Hygienic Requirements for Food Raw Material and Food Value: Hygienic Guidelines N 2-III-4.9-01-2010 [21].
[2]Current Russian СанПиН 2.3.2. 2401-08 Hygienic safety and nutritional value requirements for food. Sanitary-epidemiological rules and norms (СанПиН 2.3.2. 2401-08 Гигиенические требования безопасности и пищевой ценности пищевых продуктов Санитарно-эпидемиологические правила и нормативы)
[3]Russian Federation GN 1.2.2701-10 Hygienic norms (standards) for concentrations of pesticides in environmental media (ГН 1.2.2701-10 "Гигиенические нормативы содержания пестицидов в объектах окружающей среды")
[4]EU Regulation (EC) N°1259/2011 [22] sets maximum levels for dioxins, dioxin-like PCBs, and non-dioxin-like PCBs in foodstuffs.

[5]Regulation (EC) N°149/2008 [23]. The 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.
[6]The sum of PCB28, PCB52, PCB101, PCB138, PCB153, and PCB180.
[7] The sum of p,p´-DDT, o,p´-DDT, p,p´-DDE, o,p´-DDE, p,p´-DDD, and o,p´-DDD.
[8]p,p´-DDT.
[9] The sum of p,p´-DDT, o,p´-DDT, p,p´-DDE, and p,p´-DDD.
[10] The sum of HCH-alpha, HCH-beta, HCH-gamma, and HCH-delta.
**For each isomer MRL is set separately

4 Results

The pooled samples of eggs were analysed for OCPs and U-POPs. GCMS-HRMS analyses were chosen for the confirmation of contamination of the sampled hens' eggs by dioxins and dioxin-like PCBs. The same samples were also analysed for other POPs (including indicator PCBs) and OCPs: hexachlorobenzene (HCB), hexachlorocyclohexanes (HCHs), and DDT and its metabolites. HCB is also considered to be an unintentionally produced POP (U-POP) in the same processes as dioxins and DL PCBs [24], although it is commonly measured together with other OCPs. Additionally, two other U-POPs, pentachlorobenzene (PeCB) and hexachlorobutadiene (HCBD), were analysed in all the samples. The results for U-POPs and OCPs are summarised in Table 3.

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 PCBs' (NDL PCBs) [22]. Levels of PCDD/Fs and DL PCBs are expressed in total WHO-TEQ calculated according to toxic equivalency factors (TEFs) set by a WHO expert panel in 2005 [25]. These new TEFs were used to evaluate dioxin-like toxicity in samples of hens' eggs from Armenia in this study.

Table 3: Summarised results of analyses for POPs for three pooled hens' egg samples from Armenia, sampled in July 2018.

Locality	Alaverdi	Alaverdi	Yerevan	Armenian/EU standards/ limits
Sample	Alaverdi 1	Alaverdi 3	Yerevan – supermarket	
Fat content (%)	14.4	13.1	8.7	-
PCDD/Fs (pg WHO TEQ g^{-1} fat)	7.54	4.45	0.20	3.0/2.50
DL PCBs (pg WHO TEQ g^{-1} fat)	19.31	9.05	0.14	-
Total PCDD/F + DL PCBs (pg WHO TEQ g^{-1} fat)	26.85	13.50	0.34	ND/5.00
HCB (ng g^{-1} fat)	0.9	1.7	< 0.1	-
PeCB (ng g^{-1} fat)	< 0.1	< 0.1	< 0.1	-
HCBD (ng g^{-1} fat)	< 0.1	< 0.1	< 0.1	-

7 PCB (ng g^{-1} fat)	24.10	13.20	0.50	-
6 PCB (ng g^{-1} fat)	16.20	8.30	0.50	ND/40.00
Sum of HCH (ng g^{-1} fat)	54.50	10.90	2.00	-
Sum of DDT (ng g^{-1} fat)	599.00	77.80	3.80	-

ND – not defined

Both the samples of free-range hens' eggs from Alaverdi exceeded the Armenian limit, as well as the EU ML for PCDD/Fs and/or PCDD/Fs and DL PCBs, expressed as WHO TEQ in hens' eggs (see Table 3); [22]. The background levels for PCDD/Fs and DL PCBs measured in hens' eggs from a supermarket in Yerevan were 0.20 and 0.14 pg WHO-TEQ g^{-1} fat, respectively. The highest levels of dioxins (7.54 pg WHO-TEQ g^{-1} fat) and DL PCBs (19.31 pg WHO TEQ g^{-1} fat), respectively, were measured in eggs from the Alaverdi 1 site, where they were sampled in a valley south-east of the copper smelter.

The total WHO-TEQ levels of PCDD/Fs and DL PCBs in the samples from Alaverdi are comparable with those observed in the vicinity of metallurgical plants in Balkhash or Temirtau, both in Kazakhstan [26, 27], and/or Beihai in China [28, 29]. The higher level of almost 27 pg WHO-TEQ/g fat represents a rather higher level in comparison with the results presented in IPEN's global Egg Report [3], and can be compared with levels observed at such hot spots as Gorbatovka, Russia [30], a site near a medical waste incinerator in Lucknow, India [31], and near a petrochemical complex in Coatzacoalcos, Mexico [32], with levels of 22, 29, and 26 pg WHO-TEQ/g fat respectively. It is also close to the level observed recently in free-range hens' eggs at a site seriously contaminated by wood treated with pentachlorophenol in Poland [33]. PCDD/Fs occurred as by-products in pentachlorophenol protection and then also contaminated treated wood [33, 34].

Both samples of free-range hens' eggs from Alaverdi had levels of PCDD/Fs and DL PCBs, respectively, higher than those observed in the pooled sample of eggs bought in a Yerevan supermarket, which was used as a control sample showing background levels of PCDD/Fs and DL PCBs in hens' eggs from Armenia for this study. On this topic, see also the discussion about background levels in other studies focused on POPs in free-range hens' eggs [3, 27].

Arnika and AWHHE had already sampled and analysed free range hens' eggs for POPs in 2010 at different locations, those where obsolete pesticides were stored in particular, but Alaverdi was also included as one of the POP hot spots in the report from 2011 [17]. The eggs were analysed for total dioxin activity by DR CALUX analysis, the results of which can be compared with the results of the GC-HRMS analyses in this report. The levels of total PCDD/Fs and content of DL PCBs in BEQ (= bioanalytical toxic equivalent) observed in eggs sampled in Armenia in 2010 are summarised in Table 4. The level of 11.9 pg BEQ/g fat was measured in eggs from Alaverdi in 2010, which is lower than the levels of dioxins and dioxin-like PCBs currently found in samples from this town. The sample was taken 1 km further to the south-east of the smelter than Alaverdi 1 in this study, but it was still located in the valley influenced by smelter emissions, during periods of climatic inversion in particular.

Table 4: Summary of the DR CALUX analyses results showing levels of PCDD/Fs in free-range hens' eggs sampled in 2010 at several Armenian toxic hot spots. Source: [17].

Locality	Year (month) of sampling	Number of eggs	Fat content	PCDD/Fs and DL PCB (DR CALUX) in pg BEQ/g fat
Alaverdi	2010 (XI)	5	12.6	11.9
Echmiadzin – Beriutyun	2010 (XI)	5	12.7	20.9
Griboedov	2010 (XI)	5	11.7	24.8
Kobayr village	2010 (XI)	5	15.5	7.3
Mushavan	2010 (XI)	5	12.9	<LOQ (0.79)
Nubarashen – cottages	2010 (XI)	5	13.7	37

Hexachlorobenzene (HCB), pentachlorobenzene (PeCB), and hexachlorobutadiene (HCBD) are three further chemicals listed as unintentionally produced POPs under Annex C to the Stockholm Convention. They are also listed under Annex A to the Convention as they were also produced intentionally and used mostly as pesticides in the past [35]. Only HCB was measured at levels above LOQ in the pooled egg samples from Alaverdi among these three additional U-POPs (see Table 3). The observed levels are discussed in Chapter 4.3, Organochlorinated pesticides.

4.1.2 Potential source(s) of dioxins and dioxin-like PCBs in eggs from Alaverdi

PCDD/Fs have a specific congener pattern that can reveal their most likely source. Their pattern in the two pooled egg samples from Alaverdi is visible in the graphs in Figures 2 and 3. It is very similar in both samples but not identical. It differs in terms of the proportions of 2,3,7,8 TCDD and 1,2,3,7,8 PeCDD, and of the balance between PCDD and PCDF. Dibenzo-p-dioxin congeners prevail in the sample from the Alaverdi 3 location, while in the eggs from Alaverdi 1 dibenzofuran congeners represent a bigger portion of TEQ. DL PCBs also have a slightly bigger portion of the total TEQ level in the eggs from Alaverdi 1 in comparison with Alaverdi 3 at 72% and 67% respectively.

Although we may consider that the major source of dioxin contamination is the same, there might be additional sources of contamination and those might influence the PCDD/Fs and DL PCBs levels in the eggs from Alaverdi. A much higher proportion in the total WHO-TEQ level is represented by DL PCBs in both samples and their source remains unknown as we do not know the profile of the pattern of the dioxin and dioxin-like PCBs of the copper smelter, as the most likely major source of dioxin contamination in Alaverdi. Not very much is known about emissions of PCDD/Fs and DL PCBs and other releases from primary copper production in general. For example, the UNEP Dioxin Toolkit states: "*For this class, the 'pure' primary copper smelters, there are no emission factors presently available.*" [36].

The influence of the differences between the uptake of different PCDD/F and DL PCB congeners by hens and their xxxx should also be taken into account: "*the biotransfer of dl-PCBs is much higher than for PCDD/F. This implies that they may constitute a significant food chain risk even if the environmental TEQ levels are lower than for PCDD/Fs. In the data set,* *dl-PCBs contributed on average to 47% of the total TEQ in eggs, while the average TEQ contribution in soil was 17%.*" [37] The bioavailability of dioxin congeners is "*chlorination-dependent, ranging from 80% for tetrachlorinated to less than 10% for octachlorinated congeners,*" according to a study by Stephens et al. [38]. It means that the dioxin congener patterns in eggs do not necessarily have to be the same as is shown in the contamination source.

We have to take all these limitations into account. However, there is no other obvious source of dioxin and dioxin-like PCBs than the copper smelter in Alaverdi. Potentially, it can be the open burning of waste or its burning in household stoves, which was not observed at the locations of the sampling of eggs in Alaverdi but cannot be excluded.

PCDD/F congener patterns for copper (and secondary metal production in general), as was observed in metallurgical plants in South Korea [39], is shown in the graph in Figure 4, while the patterns of the open burning of waste as observed in Eastern China [40] are obvious from the graph in Figure 5. The patterns of PCDD/Fs in the eggs from Alaverdi are closer to the copper metal industry pattern from South Korea than to the open burning of waste. It must be noted that this comparison has certain limitations, among which we also have to take into account potential regional differences between countries, as well as the influence of different fuels used in smelters, etc.

The BAT/BEP Guidelines for Annex C, sources of unintentionally produced POPs, of the Stockholm Convention noted: "*As copper is the most efficient metal to catalyse PCDD/PCDF formation, copper smelting is a particular concern.*" [24]

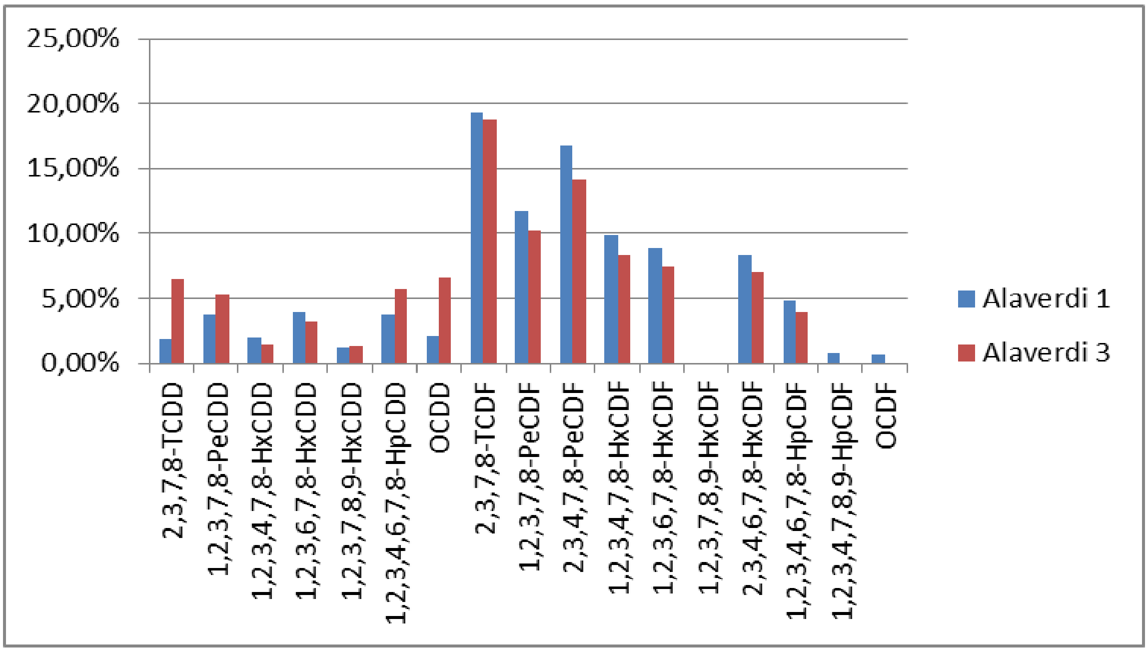


Figure 2: Congener patterns of PCDD/Fs in the egg samples from Alaverdi, expressed in absolute levels.

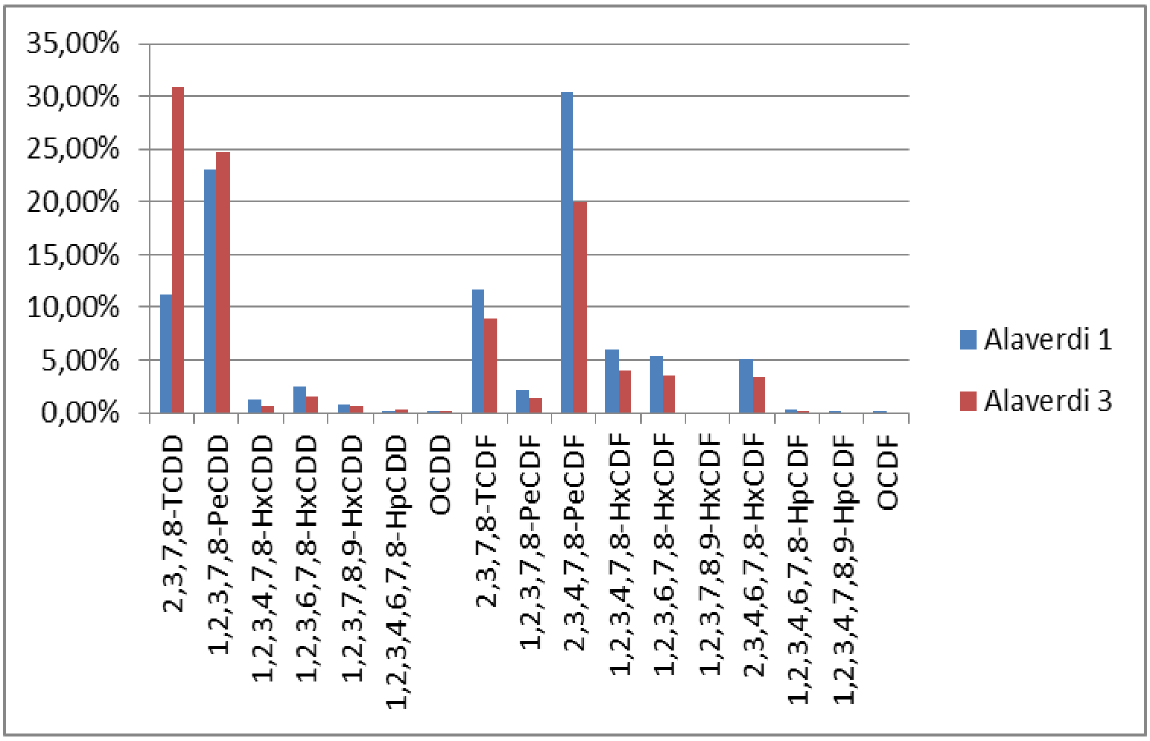


Figure 3: PCDD/F congener patterns in the egg samples from Alaverdi, expressed in TEQ levels.

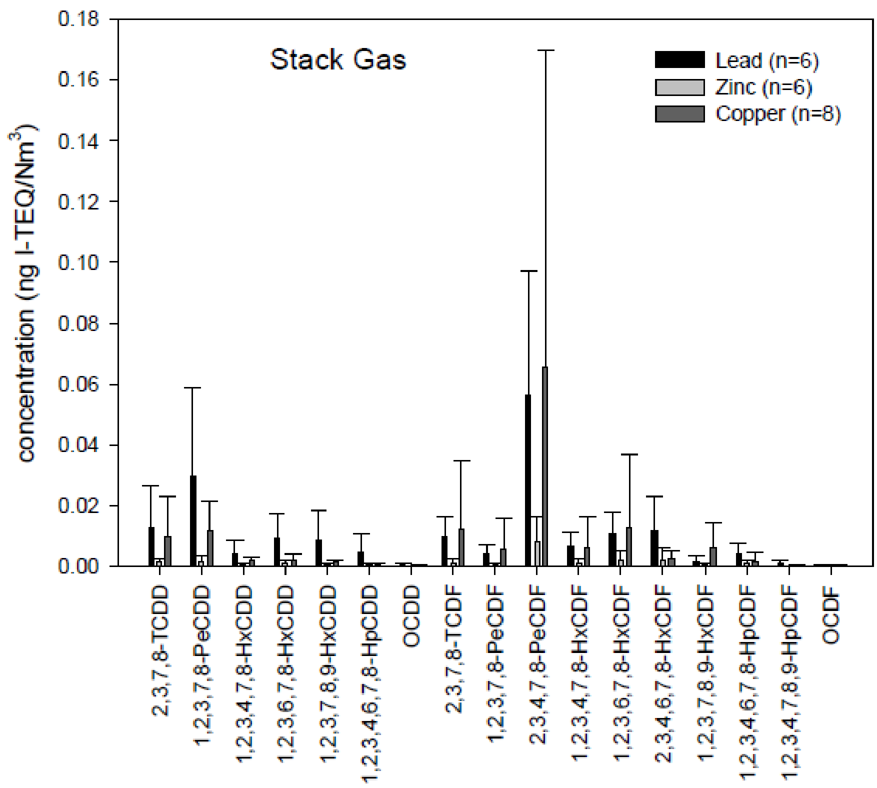


Figure 4: PCDD/F congener distribution patterns of stack gas and fly ash samples of non-ferrous metals. Expressed in ng TEQ/m^3. Source: [39]

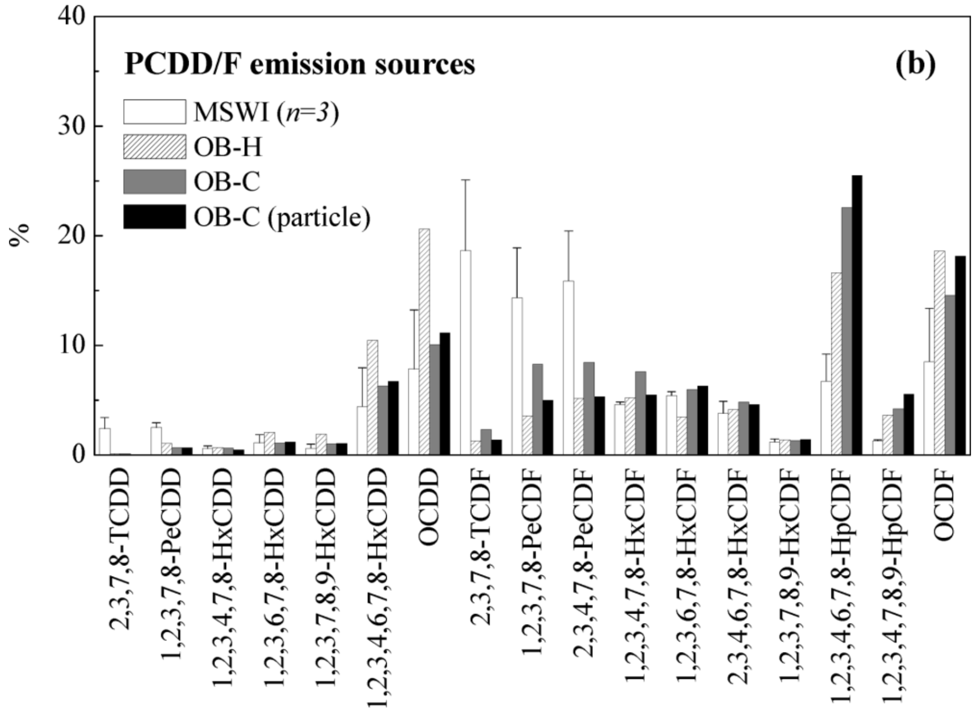


Figure 5: Open burning of waste and municipal solid waste incineration patterns as observed in Eastern China. Here the percentage of congeners on the TEQ level of dioxins (PCDD/Fs) are expressed. Source: [40]

4.2 Polychlorinated biphenyls (PCBs)

A basic description of PCBs is given in Chapter 4.1.1. They are listed under the Stockholm Convention in two different annexes, because this group includes both unintentionally produced chemicals (12 PCB congeners) and those produced and used intentionally in a variety of applications, among which those which are best known are PCBs containing oils used in transformers and capacitors. Unintentionally produced PCBs are listed under Annex C to the Stockholm Convention, and intentionally produced PCBs under Annex A, and there is a prohibition on their being newly produced and used in new applications and appliances [35]. Contamination by intentionally produced and used PCBs is represented by the level of swhat are termed indicator congeners of PCBs or NDL PCBs (see Chapter 4.1.1 for an explanation).

Indicator PCB congeners are present on levels higher than those observed, for example, at industrial sites in Thailand, but are comparable with the levels at some industrial sites in Kazakhstan [26] in the eggs from the sites sampled in Alaverdi. The level of indicator congeners of PCBs was higher in the samples of eggs from Alaverdi 1, and reached more than one third of the EU ML, which is 40 ng g^{-1} fat [22], so it is still well below the EU limit for PCBs in eggs.

4.3 Organochlorinated pesticides

Dichlorodiphenyltrichloroethane (DDT) and its metabolites, HCB, and the hexachlorocyklohexane (HCH) isomers α-HCH, β-HCH, and γ-HCH were chemicals from the group of organochlorinated pesticides in the pooled egg samples that were analysed in this study. The results are summarised in Tables 3 (expressed per gram of fat) and 5 (expressed per gram of fresh weight of eggs). The pooled egg sample from Alaverdi 1 nearly exceeded the Armenian limit for eggs, and exceeded the EU limit for the suggested level of DDT in eggs. The level of β-HCH almost reached the EU limit value in the same egg sample, but it is well below the Armenian limit for the sum of HCH.

Table 5: Summarised results of analyses for OCPs for three pooled egg samples from Armenia. Comparison with EU [23] and Armenian limit values. These results are expressed in ng g^{-1} fresh weight because the EU and Armenian limits are set for fresh weight for OCPs.

Locality	Alaverdi	Alaverdi	Yerevan	EU*/Armenian standard
Sample	Alaverdi 1	Alaverdi 3	Yerevan – supermarket	
HCB	0.1	0.2	0.0	20.0*
α-HCH	0.0	0.0	0.0	20.0*
γ-HCH	0.1	0.1	0.1	10.0*
β-HCH	7.7	1.3	0.1	10.0*
sum-4DDT (EU)	**86.0**	10.0	0.3	50.0*
sum HCH	7.8	1.4	0.2	100.0
sum DDT	86.0	10.2	0.3	100.0

DDT had a long history in former Soviet Union countries, including Armenia. It was produced in Armenia until 1962 in a limited amount of 50 tons per year according to the Armenian NIP [12]: "From *1970, the application of DDT was prohibited in the former USSR and in Armenia as well. However, there is evidence of the illegal/illicit use of POP pesticides by the owners of small and medium-sized farms in Armenia. Despite the preventive measures taken, residual amounts of DDT continue to be revealed in environmental media (soil, surface water, the water of Lake Sevan), foodstuffs, and the human organism. According to the data of the Monitoring, the frequency of the determination of POPs (Lindane, DDE) came to 87-97% in samples of breast milk taken from feeding mothers in rural regions of Armenia*." [12].

DDT and other OCPs were monitored in hens' eggs in Armenia in 2002-2003. The results were published in NIP, and are copied here in Table 6. As is obvious from the data presented in Table 5, all the pesticides that were investigated were present in the eggs. The findings also testified to the fresh application of DDT and HCH. [12]

Table 6: Results of the monitoring of several POPs in hens' eggs in Armenia in 2002 and 2003. Source: [12]

Average POPs content in eggs, 2002 – 2003, mcg/kg

Sampling site	∑ Heptachlor	∑ HCh	∑ DDT	HCB	PCB
Syunik, Megri district	NR*	1.72	6.03	0.26	NS**
Lori, Alaverdi district	0.04	0.12	0.48	1.47	2.78
Lori, Gugark district	0.006	0.07	0.75	0.41	1.61
Ararat, Artashat district	NR	1.53	2.29	0.04	NS
Armavir, Echmiadzin district	NR	0.71	2.29	4.53	NS
Aragatsotn, Ashtarak district	NR	1.01	2.46	NR	NS

*NR – not revealed; **NS – not studied

The total levels of DDT in the samples from Alaverdi are lower than those measured in the pooled sample from Alaverdi (91 ng/g fw) in 2010, and it is much lower than the highest level of 730 ng/g fw analysed in eggs from Echmiadzin-Berriutyun, sampled in the neighbourhood of a former obsolete DDT storage in 2010. However, the levels of the sum of DDT and its metabolites in the eggs from Alaverdi 1 and 3 are higher than those observed by the state authorities and published in the Armenian NIP (see Table 6). The sum of HCH in the Alaverdi 1 sample exceeded the residues of this pesticide found by state authorities in egg samples from Armenia several-fold.

It seems that there should be some source of continuing use of DDT in the Lori region, and feed for chickens might be contaminated by this obsolete pesticide which has been banned for many years.

HCB is another OCP followed in eggs by the state authorities in Armenia [12]. There were lower levels of HCB in the eggs from this study than in free-range hens' eggs collected from different sites in Armenia in 2010. HCB was found at levels of 0.3 ng/g and 2.4 ng/g fat, respectively, in eggs sampled in Alaverdi in 2010, while in the more recent samples presented in this study we observed levels of 0.1; 0.2 ng/g fw (see Table 5) and 0.9; 1.7 ng/g fat (see Table3) respectively. These levels are below all the samples from 2010, which were within the range 0.3-1.4 ng/g fw and 2.4-10.2 ng/g fat, respectively. The levels of HCB in the eggs from Alaverdi 1 and 3 are also lower than in most of the samples presented in the Armenian NIP (see Table 6); [12].

5 Discussion about potential exposure to dioxins and dioxin-like PCBs from hens' eggs

The share of eggs in total food consumption in Armenia in 2007 was close to 0.8% of the total food basket per day, according to the World Atlas – Food Security data[1] [20], and changes in its share had not increased greatly since 2002. It would mean that the 2017 consumption level could remain approximately the same, about 16 g per person per day if the further increasing trend stopped, and it could be around 20 g per person per day if the increasing egg consumption trend continued. If we count 50 g per hen's egg as the average weight, it would mean the consumption of one third of an egg per person per day as the general consumption pattern for the current Armenian population. The consumption of eggs within families who raise hens at the sampled sites is in agreement with this assumption, as they confirmed during our sampling.

We tried to calculate the dietary intake for the group of PCDD/Fs plus DL PCBs contaminants per day for the two pooled samples of free-range hens' eggs from Alaverdi and compare it

[1] 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 also relates to the quantity of eggs used in the preparation of food such as bakery products.

with the eggs from the Yerevan supermarket considered as the background country level in Armenia. The calculation of daily intake levels was performed by using the following formula:

DI_{adult} = (((C . F%)/100) . 18)/70;

DI_{child} = (((C . F%)/100) . 18)/35,

where DI = daily intake; C = concentration of certain groups of chemicals (PCDD/Fs, DL PCBs, etc.), and F% = fat content in the sample. The results are summarised in Table 7.

Table 7: Summarised results of calculation of dietary intake of selected POPs by eating a daily portion of eggs (18 g) from chickens raised at two sites in Alaverdi or eggs bought in a supermarket in Yerevan from hens raised on a commercial farm. Eighteen grams of egg is the approximate current average consumption per person per day in Armenia, on the basis of calculations from the available data [20].

Sample	Alaverdi 1	Alaverdi 3	Yerevan – supermarket	Suggested levels
Fat content	14.4	13.1	8.7	-
PCDD/Fs + DL PCBs (pg WHO-TEQ g^{-1})	26.85	13.50	0.34	5*
PCDD/Fs + DL PCBs (pg kg^{-1} bw) – adult	0.99	0.45	0.008	0.29**
PCDD/Fs + DL PCBs (pg kg^{-1} bw) – child	1.98	0.91	0.015	0.29**
Percentage of derived TDI	347%	159%	2.7%	-
Percentage of derived TDI	694%	318%	5.4%	-

* EU Regulation (EC) N°No 1881/2006 as amended by later regulations [41] sets maximum levels for dioxins, dioxin-like PCBs, and non-dioxin-like PCBs in foodstuffs.

** TDI derived from TWI suggested by EFSA [42] – calculated as one seventh of 2 pg WHO-TEQ/kg body weight/week, although in the text of the EFSA opinion 0.25 pg WHO-TEQ pg/kg bw/day is suggested.

The results were then compared with the tolerable daily intake derived from the tolerable weekly intake newly established in the EU by EFSA CONTAM at a level of 2 pg WHO-TEQ/kg bw/week [42]. The consumption of dioxins and dioxin-like PCBs in both egg samples leads to the suggested TWI being exceeded by their consumers. This is a very alarming result if we consider that PCDD/Fs and DL PCBs are certainly also present in other food consumed by the inhabitants of Alaverdi. Sources of PCDD/Fs and DL PCBs should be addressed in this town and its neighbourhood.

The health risk posed by the consumption of eggs contaminated by DDT and its metabolites from Armenian localities was calculated in a previous study based on broader sampling in 2010 by Dvorská et al. [43].

6 Conclusions and recommendations

This study discovered serious contamination of free-range hens' egg samples from two sites in Alaverdi with dioxins and dioxin-like PCBs. It can be expected that this contamination may also occur in other home-grown food sources in the area of Alaverdi and it can pose a serious risk to the health of the population in this town. Of course, this assumption has to be confirmed by much broader monitoring of PCDD/Fs and DL PCBs in local food.

The copper smelter seems to be the most likely source of this pollution; however, this hypothesis should be confirmed by the monitoring of dioxins in emissions and fugitive sources of dust from this production. Some basic suggestions for improvement of the technology should also be followed. The best available techniques and best environmental practices in the metallurgy sector are described in the BAT/BEP Guidelines for Annex C major dioxin sources [24].

There is a question about the potential source of DL PCBs, as primary copper production has not been greatly researched with regard to the origin of this group of PCBs. However, there might be a vast amount of obsolete equipment with leaking PCB oils in the area of the facility or in other appliances in the town which might contribute to toxic contamination of free-range hens' eggs. This area should be researched further.

The levels of DDT in one of the egg samples from Alaverdi from this year, as well as a sample from Alaverdi from 2010 [17], show the potential continuous contamination of feed or the area where hens live by this long-prohibited pesticide, and this source of contamination should be discovered as it adds to the overall burden imposed by home-grown food in the town and its surroundings.

Other POPs, such as PeCB, HCB, HCBD, or HCH, analysed in hens' eggs from Alaverdi were not found in levels raising serious health concerns for the local population.

We used hens' eggs as they are a proven indicator of potential contamination within the food chain. We did not sample meat but the results of some other studies demonstrated the simultaneous contamination of hens' eggs and meat from contaminated sites [44, 45].

7 Limitations of the study

The major limitations of the study were the limited financial, temporal, and personnel resources. Therefore, only a limited number of hens' egg samples could be taken, and only analyses on a limited scale could be conducted. We could not include more localities in this study, for example. We preferred to focus on one locality, and there to take at least two pooled egg samples.

The comparison of the 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.

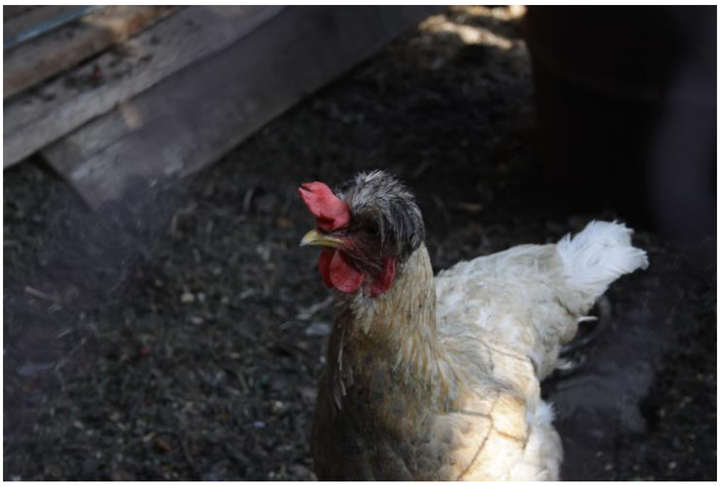
The TDI levels for PCDD/Fs and DL PCBs in the EU have changed recently; however, they are not established in Armenia or the neighbouring countries. The standards set for certain types of food did not necessarily conform to this new concept set by EFSA CONTAM during the time when we worked on this study and published it in November 2018 [42].

The estimation of the 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 PCDD/Fs and DL PCBs through the consumption of free-range hens' eggs from two sites in

Alaverdi, 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 or PCBs in Armenia.

8 Pictures from sampling



9 References

1. Arkenbout, A., *Biomonitoring of Dioxins/dl-PCBs in the north of the Netherlands; eggs of backyard chickens, cow and goat milk and soil as indicators of pollution.* Organohalog Compd, 2014. **76**: p. 1407-1410.
2. Aslan, S., et al., *Levels of PCDD/Fs in local and non-local food samples collected from a highly polluted area in Turkey.* Chemosphere, 2010. **80**(10): p. 1213–1219.
3. DiGangi, J. and J. Petrlik, *The Egg Report – Contamination of chicken eggs from 17 countries by dioxins, PCBs and hexachlorobenzene*. 2005: Available at: http://english.arnika.org/publications/the-egg-report.
4. Pirard, C., et al., *Assessment of the impact of an old MSWI. Part 1: Level of PCDD/Fs and PCBs in surrounding soils and eggs.* Organohalogen Compounds, 2004. **66**: p. 2085-2090.
5. Pless-Mulloli, T., et al., *Transfer of PCDD/F and heavy metals from incinerator ash on footpaths in allotments into soil and eggs.* Organohalogen Compounds, 2001. **51**: p. 48-52.
6. Shelepchikov, A., et al., *Contamination of chicken eggs from different Russian regions by PCBs and chlorinated pesticides.* Organohalogen Compounds, 2006. **68**: p. 1959-1962.
7. Hoogenboom, R., et al., *Dioxines en PCB´s in eieren van particuliere kippenhouders*. 2014, RIKILT (University & Research centre): Wageningen. p. 25.
8. Piskorska-Pliszczynska, J., et al., *Soil as a source of dioxin contamination in eggs from free-range hens on a Polish farm.* Science of The Total Environment, 2014. **466–467**(0): p. 447-454.
9. Van Eijkeren, J., et al., *A toxicokinetic model for the carry-over of dioxins and PCBs from feed and soil to eggs.* Food Additives & Contaminants: Part A, 2006. **23**(5): p. 509-517.
10. Dvorská, A., *Persistent Organic Pollutants in Ekibastuz, Balkhash and Temirtau. Final report on the results of environmental sampling conducted in Kazakhstan in 2013 and 2014 as a part of the project "Empowering the civil society in Kazakhstan in improvement of chemical safety"*, in *Toxic Hot Spots in Kazakhstan. Monitoring Reports*. 2015, Arnika – Toxics and Waste Programme: Prague-Karaganda.
11. Yu, B.-W., et al., *Emission of PCDD/Fs and dioxin-like PCBs from metallurgy industries in S. Korea.* Chemosphere, 2006. **62**(3): p. 494-501.
12. Republic of Armenia, *National Implementation Plan for the Stockholm Convention on Persistent Organic Pollutants*. 2005: Yerevan. p. 104.
13. UNEP Chemicals, *Toolkit for Identification and Quantification of Mercury Releases. Version 1.2*. 2013, Geneva, Switzerland: UNEP Chemicals Branch.
14. Grigoryan, R., et al., *Risk factors for children's blood lead levels in metal mining and smelting communities in Armenia: a cross-sectional study.* BMC Public Health, 2016. **16**(1): p. 945.
15. Suvaryan, Y., V. Sargsyan, and A. Sargsyan. *The Problem of Heavy Metal Pollution in the Republic of Armenia: Overview and Strategies of Balancing Socioeconomic and Ecological Development*. 2011. Dordrecht: Springer Netherlands.
16. Gevorgyan, G.A., et al., *Environmental Risk Assessment of Heavy Metal Pollution in Armenian River Ecosystems: Case Study of Lake Sevan and Debed River Catchment Basins.* Polish Journal of Environmental Studies, 2016. **25**(6).
17. Dvorská, A., et al., *Toxic Hot Spots in Armenia. Monitoring and Sampling Reports*, ed. J. Petrlík (editor). 2011, Prague – Yerevan. Available at: http://english.arnika.org/publications/toxic-hot-spots-in-armenia: Arnika – Toxics and Waste Programme, Armenian Women for Health and a Healthy Environment (AWHHE). 44.
18. World Weather. *Архив погоды в Алаверди - Роза ветров в Алаверди (Weather Archive for Alaverdi - Wind rose)*. 2018 [cited 2018 30-07-2018]; Available from: https://world-weather.ru/archive/armenia/alaverdi/.
19. European Commission, *Commission Regulation (EU) No 252/2012 of 21 March 2012 laying down methods of sampling and analysis for the official control of levels of dioxins, dioxin-like PCBs and non-dioxin-like PCBs in certain foodstuffs and repealing Regulation (EC) No*

1883/2006 Text with EEA relevance. European Commission, Editor. 2012: Official Journal of the European Communities. p. L 84, 23.3.2012, p. 1-22.

20. Knoema. *World Data Atlas Armenia Topics Food Security (in 2007)*. 2012 June 2012 31-10-2018]; Available from: https://knoema.com/atlas/Armenia/topics/Food-Security/Food-Consumption/Eggs-consumption.
21. MoH, *Hygienic Requirements for Food Raw Material and Food Value: Hygienic Guidelines N 2-III-4.9-01-2010*, (approved by the Order N 06N of 10.03.2010 of the RA Minister of Health), Editor. 2010.
22. European Commission, *Commission Regulation (EU) No 1259/2011 of 2 December 2011 amending Regulation (EC) No 1881/2006 as regards maximum levels for dioxins, dioxin-like PCBs and non-dioxin-like PCBs in foodstuffs (Text with EEA relevance)*. European Commission, Editor. 2011: Official Journal of the European Union. p. 18-23.
23. European Commission, *Commission Regulation (EC) No 149/2008 of 29 January 2008 amending Regulation (EC) No 396/2005 of the European Parliament and of the Council by establishing Annexes II, III and IV setting maximum residue levels for products covered by Annex I thereto. Text with EEA relevance.* European Commission, Editor. 2008: Official Journal of the European Communities. p. L 58, 1.3.2008, p. 1-398.
24. Stockholm Convention on POPs, *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, Secretariat of the Stockholm Convention on POPs: Geneva.
25. van den Berg, M., et al., *The 2005 World Health Organization reevaluation of human and Mammalian toxic equivalency factors for dioxins and dioxin-like compounds.* Toxicol Sci, 2006. **93**(2): p. 223-41.
26. Petrlik, J., et al., *PCDD/Fs and PCBs in eggs – data from China, Kazakhstan and* in *Abstracts Book of the Dioxin 2018: 38th International Symposium on Halogenated Persistent Organic Pollutants & 10th International PCB Workshop*. 2018: Kraków, Poland. p. 794-798.
27. Petrlík, J., et al., *Hens' eggs as an indicator of the pollution of the environment in Kazakhstan. Results of sampling conducted in 2013-2016 (Использование яиц кур свободного содержания в качестве индикатора загрязнения в Казахстане. Результаты опробования, проведенного в период в 2013 по 2016 гг.)*. 2017, Arnika – Citizens Support Centre, EcoMuseum Karaganda, Eco Mangystau: Prague – Karaganda – Aktau. p. 46.
28. Petrlik, J., P.A. Behnisch, and A. Teebthaisong, *Dioxin Contamination of Free Range Chicken Eggs from Selected Hot Spots – Based on CALUX Bioassay Analyses*, in *11th BioDetectors Conference 2018*. 2018: Aachen. p. 14-15.
29. Petrlik, J., *Persistent Organic Pollutants (POPs) in Chicken Eggs from Hot Spots in China*. 2015, Arnika – Toxics and Waste Programme, IPEN and Green Beagle: Beijing – Gothenburg – Prague. p. 25.
30. IPEN Dioxin PCBs and Waste Working Group, et al., *Contamination of chicken eggs from the Dzerzhinsk region, Russia by dioxins, PCBs and hexachlorobenzene*, in *Keep the Promise, Eliminate POPs Reports*. 2005, IPEN, Arnika Association, Spolocnost priatelov Zeme: Dzerzhinsk, Moscow, Prague. p. 30.
31. IPEN Dioxin PCBs and Waste Working Group, Toxics Link, and Arnika Association, *Contamination of chicken eggs near the Queen Mary's Hospital, Lucknow medical waste incinerator in Uttar Pradesh (India) by dioxins, PCBs and hexachlorobenzene*, in *Keep the Promise, Eliminate POPs Reports*. 2005, IPEN, Arnika Association, Toxics Link: New Delhi, Prague. p. 25.
32. IPEN Dioxin PCBs and Waste Working Group, RAPAM, and Arnika Association, *Contamination of chicken eggs near the Pajaritos Petrochemical Complex in Coatzacoalcos, Veracruz, Mexico by dioxins, PCBs and hexachlorobenzene*, in *Keep the Promise, Eliminate POPs Reports*. 2005, IPEN, Arnika Association, Secretariat, Red de Acción Sobre Plaguicidas y Alternativas en México – RAPAM: Mexico City, Prague. p. 29.

33. Piskorska-Pliszczynska, J., et al., *Pentachlorophenol from an old henhouse as a dioxin source in eggs and related human exposure.* Environmental Pollution, 2016. **208, Part B**: p. 404-412.
34. Fries, G.F., et al., *Treated wood in livestock facilities: relationships among residues of pentachlorophenol, dioxins, and furans in wood and beef.* Environmental Pollution, 2002. **116**(2): p. 301-307.
35. Stockholm Convention, *Stockholm Convention on Persistent Organic Pollutants (POPs) as amended in 2009. Text and Annexes*. 2010: Geneva. p. 64.
36. UNEP and Stockholm Convention, *Toolkit for Identification and Quantification of Releases of Dioxins, Furans and Other Unintentional POPs under Article 5 of the Stockholm Convention*. 2013, United Nations Environment Programme & Stockholm Convention Secretariat: Geneva. p. 445.
37. Watanabe, M., et al., *Bioaccumulation of organochlorines in crows from an Indian open waste dumping site: evidence for direct transfer of dioxin-like congeners from the contaminated soil.* Environ Sci Technol, 2005. **39**(12): p. 4421-30.
38. Stephens, R., M. Petreas, and D. Hayward, *Biotransfer and bioaccumulation of dioxins and furans from soil: chickens as a model for foraging animals.* Sci Total Environ, 1995. **175**(3): p. 253-73.
39. Yoon-Seok, C., Byeong-Woon, Y., Young-Hoon, M., Min-Kwan, K., Jong-Dai, K., *Inventory Study of PCDD/Fs for Metal Industries in South Korea.* Organohalog Compd, 2003. **63**: p. 94-97.
40. Xu, M., et al., *Concentrations, profiles and sources of atmospheric PCDD/Fs near a municipal solid waste incinerator in Eastern China.* Environ Sci Technol, 2009. **43**(4): p. 1023-1029.
41. European Commission, *Commission Regulation (EC) No 1881/2006 of 19 December 2006 setting maximum levels for certain contaminants in foodstuffs (Text with EEA relevance) (OJ L 364, 20.12.2006, p. 5)*. 2016: Official Journal. p. 1-40.
42. EFSA CONTAM, *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): p. 331.
43. Dvorská, A., et al., *Obsolete pesticide storage sites and their POP release into the environment – an Armenian case study.* Environmental Science and Pollution Research, 2012. **19**(6): p. 1944-1952.
44. Lovett, A., et al., *PCB and PCDD/DF concentrations in egg and poultry meat samples from known urban and rural locations in Wales and England.* Chemosphere, 1998. **37**(9-12): p. 1671-1685.
45. Chang, R., et al., *Foraging farm animals as biomonitors for dioxin contamination.* Chemosphere, 1989. **19**(1-6): p. 481-486.

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