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Alaverdi - Yerevan - Prague, December 2021

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MAPPING THE DISTRIBUTION OF HEAVY METALS AND PERSISTENT ORGANIC POLLUTANTS IN THE MINING AND INDUSTRIAL AREAS OF THE TUMANYAN AND STEPANAVAN REGIONS OF LORI PROVINCE, REPUBLIC OF ARMENIA, 2018-2021

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Content

1 Introduction

This study summarizes all the results of sampling campaigns carried out between 2018 and 2021 in 13 settlements of the Tumanyan and Stepanavan regions of Lori Province. The survey was conducted with the cooperation of three non-profit organizations: Arnika, Centre for Community Mobilization and Support (CCMS), and EcoLur. The individual surveys generated a large dataset covering a wide range of sampled matrices including environmental samples of soil, sediment, and household dust, and food samples such as domestic chicken eggs, fish, homemade cheese, and crops grown by local private farmers were collected from these localities. Biological samples such as urine, hair, and nails were also taken from local residents.

The Tumanyan and Stepanavan regions are characterized by rich mineral reserves. For many years, copper, molybdenum, gold, silver, and other precious and non-ferrous metals have been mined and continue to be mined here, which implies their increased content in environmental components. However, in addition to the natural high background, these elements are actively distributed in the environment by the mining and metallurgical industries. Such industrial practices affect the surrounding environment and the living conditions of local people who are directly exposed to metals for a long time. As a consequence, the adverse change in the living environment is felt by people either directly on their health or on the quality of the farming environment. As part of this study, analyses were carried out for the presence of heavy metals such as copper (Cu), zinc (Zn), lead (Pb), arsenic (As), cadmium (Cd), and others, as well as for the presence of persistent organic pollutants (POPs). An overview of heavy metal profiles is annexed to this study (Chapter 8.1.).

The aim of this study was to map the occurrence and distribution of heavy metals and POPs in components in the vicinity of the potential polluting points of the Tumanyan and Stepanavan regions, and to determine the burden of contaminants on the local population in a territory with a population of 40,000.

2 Description of the region and scope of the study

The determination of heavy metal content was carried out in the Tumanyan and Stepanavan regions of Lori Province, located in the northern part of Armenia. This study includes a review of analyses conducted in 13 settlements in 11 communities in the Tumanyan region and in two communities of the Stepanavan region in the years 2018-2021 (see Table 1). Samples were collected in the communities of Alaverdi, Akori, Akhtala, Shamlugh, Shnogh, Teghut, Pokr Ayrum, Mets Ayrum, Chochkan, Haghpat, Armanis, and Mghart, as well as in the Armanis suburb of the extended community of Stepanavan and its surroundings (Figure 1).

Table 1: *Overview of locations, population, and distance from pollution hotspots*

Location	Population	Year	Pollution hotspot	Distance (m)
Alaverdi	12,543	2020	Alaverdi copper smelting factory	50-2000
Odzun	4880	2020	Alaverdi copper smelting factory	6000-7000
Akori	2531	2020	Alaverdi copper smelting factory	2000-4000
Mets Ayrum	233	2020	Naatak tailings dam	50-500
Haghpat	791	2020	Alaverdi copper smelting factory	4500-6000
Chochkan	1976	2020	Naatak tailings dam	200-1500
Akhtala	1970	2020	Akhtala copper mine, Akhtala copper processing plant, Nazik tailings dam	50-5000
Shamlugh	606	2020	Akhtala copper mine	50-1500
Teghut	765	2020	Teghut copper mine, Teghut tailing dam	500-3000
Shnogh	2940	2020	Teghut copper mine, Teghut tailings dam	3000-6000
Armanis	344	2020	Armanis polymetallic gold mine	50-1000
Mghart	327	2020	Mghart polymetallic gold mine	200-1500

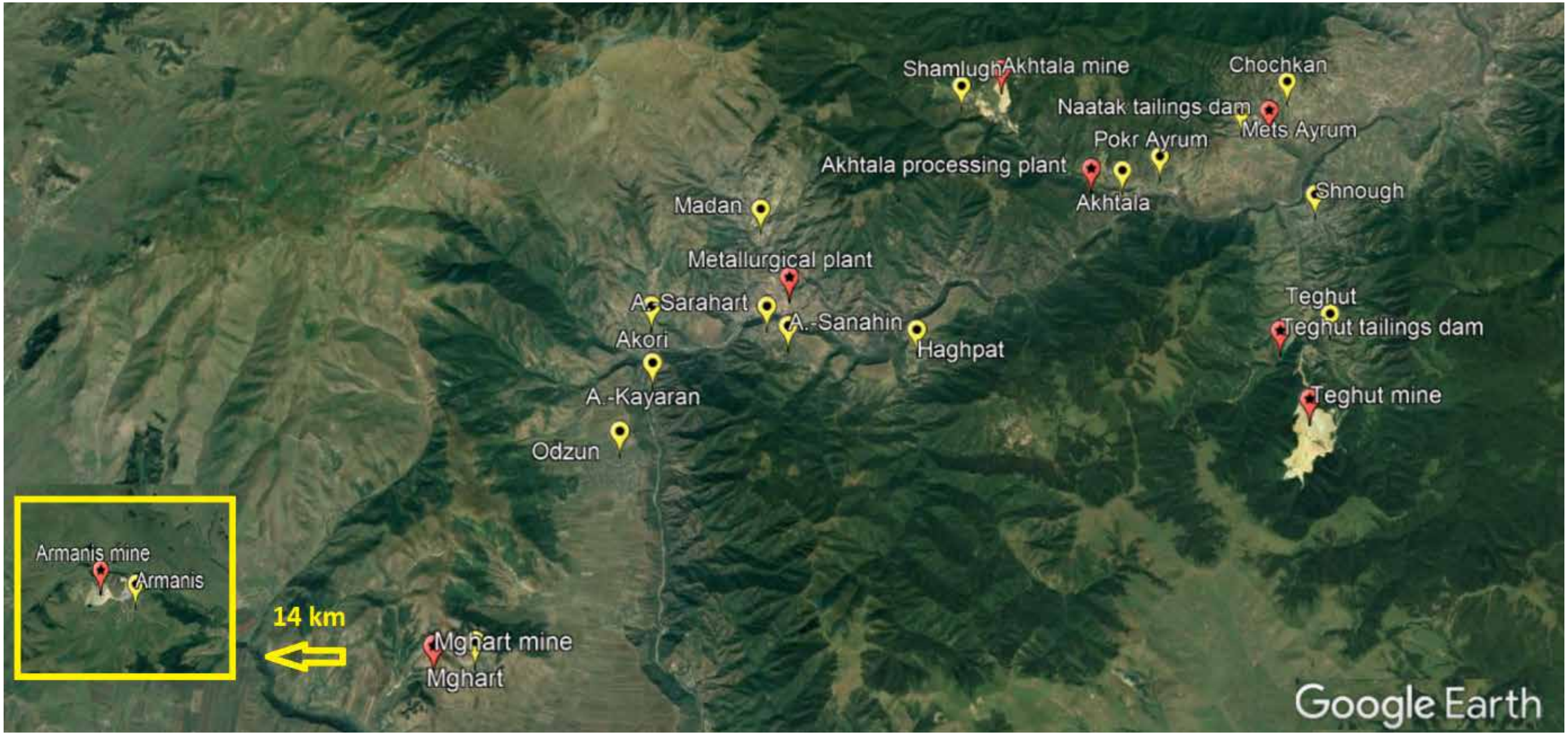

Figure 1 *Area of interest in the Tumanyan and Stepanavan regions (red marks – hotspots, yellow – communities)*

In the enlarged community of Alaverdi the residential areas, such as Sanahin, Alaverdi-Kayaran, Alaverdi-Sarahart, Lenanker (Madan), and the settlements of Haghpat and Akori were explored. The enlarged community of Odzun is located west of Alaverdi. Their geographical distance from the smelting factory is shown in Figure 2. The dominant wind direction is northerly, followed by northwesterly, southerly, and north-easterly winds [1]. Figure 3 shows the enlarged communities of Teghut and Akhtala.

Enlarged community of Alaverdi

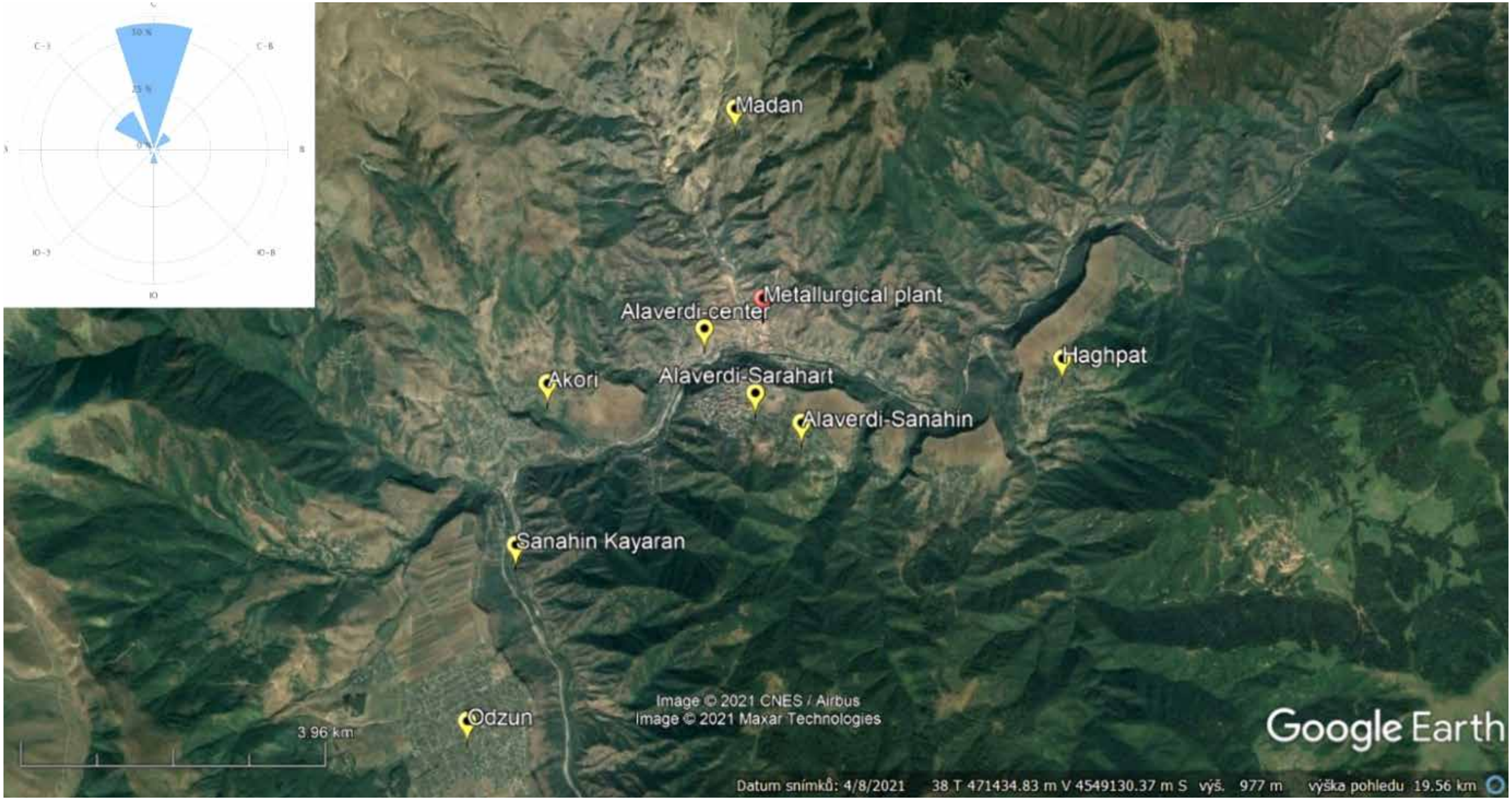

Figure 2 *Enlarged community of Alaverdi, where samples were taken (red marks – hotspots, yellow – communities)*

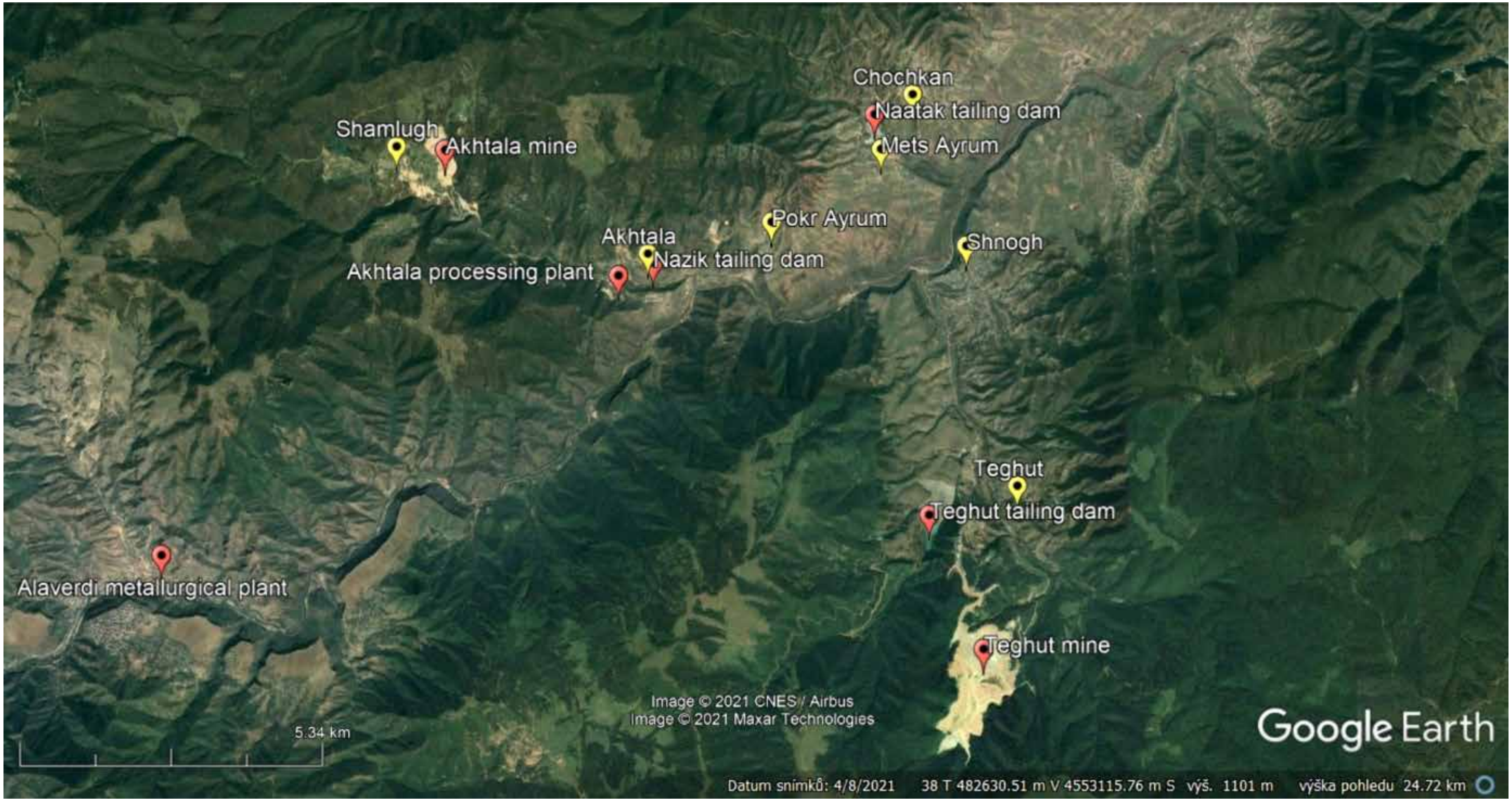

Figure 3 *Enlarged communities of Akhtala and Shnogh, where the samples were taken (red marks – hotspots, yellow – communities)*

For centuries, the Tumanyan and Stepanavan regions were well known for their developed mining and metallurgical industries. During this period, an extensive mining infrastructure in the form of mines, tailing ponds, and a copper smelter was developed. At the present time, the technologies used are outdated and require modernization. Moreover, already reclaimed sites do not always meet safety standards, contributing to leakages into the environment and thus endangering residents and the environmental situation of the region. All of these sites are potential sources of heavy metals in the environment. The main emissions from the mines are acidic mine drainage water and dust distribution. The Alaverdi copper smelting plant has polluted the environment for years by releasing emissions through chimneys, sewage, and solid waste such as slag. Leaks from functioning and closed tailings ponds are allowed, or industrial tailings waste is deliberately discharged into nearby watercourses.

Because of the above factors, mining areas become vulnerable to pollution. Metals and persistent organic matter are typical pollutants of the mining and metallurgical industry, spreading in the environment and accumulating there for decades as a result of their extremely low biotransformation and biodegradation.

3 Description of pollution hotspots

3.1 Alaverdi copper smelting plant

(GPS 41.102087, 44.661965)

The Alaverdi copper smelting plant is located in the town of Alaverdi. The town is situated in the lower part of the gorge of the Debed River. The plant was founded at the end of the 18th century. Production peaked in the 1980s, when nearly 55,000 tons of rough copper were produced annually. Emissions from the plant through the chimney covered most of the town of Alaverdi and the surrounding villages. Then the chimney was removed from the town and moved to a higher point, which only expanded the area of the impact of emissions. Over the years, during the plant's operation, the levels of sulphur and nitrogen oxides exceeded the permissible standards. To this were added emissions of heavy metals and other pollutants that were not analysed by the environmental impact monitoring centre but were detected in scientific and other specialized studies. According to the application of the Armenian Copper Programme (Vallex Group) to the European Bank for Reconstruction and Development, the level of sulphuric anhydride emissions until 2011 amounted to 25,000 tons.

The Alaverdi copper smelting plant was shut down in October 2018 because of financial difficulties, shortages of raw materials, and high fines resulting from poor compliance with emission regulations.

3.2 Akhtala mountain enrichment combine

(GPS 41.146378, 44.761768)

Copper ore from the Shamlugh copper deposit is processed at the Akhtala mountain enrichment combine, where ore concentrate is obtained. The Akhtala MEC is one of the oldest mining and processing enterprises in the Caucasus, founded at the end of the 18th century. The facility is a potential pollution hotspot because of the spread of dust from dumps and the frequent discharge of liquid residues ("tails") directly into water systems. Akhtala is a historic town located 15 km north-east of the city of Alaverdi, downstream of the Debed River. The town is located on the Akhtala River, which is a tributary of the Debed River.

Currently, the expanded community of Akhtala unites other settlements that are influenced by the infrastructure of the Akhtala MEC – the town of Shamlugh and the villages of Chochkan, Mets Ayrum, and Pokr Ayrum.

3.3 Ore mines

3.3.1 Teghut copper and molybdenum mine

(GPS 41.084309, 44.839015)

In the vicinity of the village of Teghut in the expanded community of Shnogh is located the Teghut copper-molybdenum deposit, which began to be exploited in 2015. In 2018, the operation of the field was stopped as a result of the bankruptcy process of the Vallex group of companies and the transfer of the property to the Russian VTB Bank, as well as the result of a negative assessment of the state of the Teghut tailings dump, which was recognised as unsafe. Currently, ore mining has resumed. The community of Shnogh is located in Lori Province, 70 km from the regional centre, the city of Vanadzor. The Shnogh River is a tributary of the transboundary Debed River. The villages of Shnogh and Teghut have the official status of impact of the Teghut programme for the exploitation of the deposit and the processing of raw materials.

3.3.2 Armanis gold-polymetallic mine

(GPS 41.010963, 44.315972)
The Armanis gold-polymetallic deposit and processing plant is located 1.5 km from the village of Armanis, in the expanded community of Stepanavan. The nearest watercourse is the Chqnagh River, with its tributary the Armanis. The main minerals extracted in the mine are gold, silver, copper, lead, and zinc. The mine started to be developed in 2011, but since 2015 production has been stopped. According to representatives of the company, active preparations are under way to resume ore mining.
The village of Armanis is attached to the town of Stepanavan and is part of its administrative territory. The expanded community of Stepanavan also includes the village of Urasar, which also has the official status of a “community impacted by mining” as a result of the exploitation of the Armanis gold-polymetallic deposit. Stepanavan is a well-known resort centre and is located on the right and left banks of the Dzoraget River in Lori Province, 36 km from the regional centre, Vanadzor. The Dzoraget River, in turn, is a tr0ibutary of the transboundary Debed River.

3.3.3 Mghart gold-polymetallic mine

(GPS 40.994621, 44.571805)
The Mghart gold-polymetallic deposit is located at a distance of 0.5 km west of the village of Mghart, between the Debed and Dzoraget Rivers in Lori Province. The village of Mghart is part of the expanded community of Odzun, with the official status of a “community impacted by mining” as a result of the Mghart deposit exploitation programme. The deposit has been exploited since 2005, with interruptions lasting from several months to several years.

Work on the rehabilitation of the territory has not been carried out during this time.
The technical parameters of the above-mentioned metal deposits of the Tumanyan and Stepanavan districts, where the samples were taken, are given in Table 2.

Table 2 *Overview of mines located in the Tumanyan and Stepanavan regions (* - ore content %)*

Location	Year of opening	Ore contents (g/ton)	Area (ha)	Extraction volume (tons/yr)	Main product	Status
Akhtala (Shamlugh)	18th century	Cu, Mo	55	Cu-300	Cu (metal and concentrate)	Active
Teghut	2015	Cu, Mo	240	Cu-100 Mo-1	Concentrated Cu, Mo	Non-active
Mghart	2005	Au-8.22 Ag-9.20	6.0	50	Au	Active
Armanis	2011	Au-2.2 Ag-14.76 Cu-0.79* Zn-1.92*, Hg-1.87*	7.2	300	Cu, Pb, Zn concentrates	Non-active

Table 3 summarizes the parameters and geographical locations of the tailings ponds that relate to the above-mentioned mine facilities.

Table 3 *Overview of tailings ponds (* - million tons)*

Location	Owner	Tailings content	Area (ha)	Volume (million m^3)	Status	GPS
Mets Ayrum	Akhtala Mining and Processing Combine	Cu, Pb, Zn, Au, Ag, Se, Te	20.0	3.75*	Active	41.172135, 44.820829
Shamlugh	Akhtala Mining and Processing Combine	Cu, Pb, Zn	2.0	0.50	Non-active	41.150402, 44.765622
Pokr Ayrum	Akhtala Mining and Processing Combine	Cu, Pb, Zn	3.6	0.45	Non-active	41.157842, 44.784367
Teghut	CJSC Teghut	Cu, Mo	185.7	213	Active	41.108688, 44.833328
Mghart	Multi Group LLC	Au	0.5	0.02	Full	40.989407, 44.556876
Mghart	Multi Group LLC	Au	1.2	0.48	Active	40.990509, 44.558434

4 Methodology

In total, during 2018-2021, a wide range of samples were collected in the Tumanyan and Stepanavan regions of Lori Province which characterize the mapping and movement of pollutants in this region. In total, about 400 samples were collected and analysed (Table 6). For the analysis of heavy metals, 10 elements were selected – arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), nickel (Ni), molybdenum (Mo), zinc (Zn), iron (Fe), and mercury (Hg). Samples of eggs and fish were analyzed for dibenzo-p-dioxins and benzofurans (PCDD/F) and dioxin-like polychlorinated biphenyls (dl-PCBs), indicator PCB congeners, organochlorine pesticides (OCP), pentachlorobenzene (PeCB), and hexachlorobutadiene (HCBD). The environmental samples included soil, river sediment, and indoor domestic dust samples. The foodstuff samples included free-range chicken eggs, fish from the local rivers, plants, and homemade cheese. For the biological data set hair, nails, and urine samples were collected from the residents.

The control samples (presumably without industrial influence) were taken in Tavush Province and elsewhere in Lori Province, outside the zone of influence of the mining and metallurgical industry. The control samples were used for comparative analysis with the samples taken in the affected areas.

The sampling was conducted according to a sampling plan covering areas in the spatial gradient from potential contamination hotspots using a combination of results from previous studies, the Google Earth system, and reports from local activists. The samples were taken from public sites, the yards of kindergartens or schools, and private gardens.

All analyses were performed in the laboratories of the Czech Republic at the Institute of Chemical Technology, Prague (Department of Food Chemistry and Analysis; Department of Environmental Chemistry), the Health Institute in Ústí nad Labem, and the State Veterinary Institute, Prague.

4.1 Environmental samples

Soil samples

In this study 54 samples of soils from private agricultural gardens and public places were taken in the Tumanyan and Stepanavan regions. The heavy metal distribution was compared with the directions of the prevailing winds. Two control samples of soil were collected from the Dilijan National Park (located in Tavush Province). In the control samples a low HM content was expected as a result of the absence of industry or other polluting activities. The aim of the soil sampling was to understand the transfer of heavy metals into the environment.

Sediment samples

In this study 31 samples of sediments were taken from various water sources in the Tumanyan and Stepanavan regions – the Rivers Debed, Akhtala, Shnogh, Chqnagh, and Madan and other small rivers. Each sample point is represented by one sediment sample. The Debed River is a transboundary river that begins in Armenia. It is part of the Kura-Araks river basin. Rivers are used to irrigate agricultural fields and private gardens, and in some cases are used as places for rafting and recreation.

The main objective of the sediment sampling was to identify downstream pollution distribution in the potential hotspot areas, with a comparison of the composition of sediments from uncontaminated locations without mining and metallurgical activities. The collected data was compared to the results obtained from two control samples taken from the Aghstev River, flowing through the Dilijan National Park and the territory of Haghartzin (Tavush Province).

Slag samples

Under this study three samples of slag were collected in 2018 in the towns of Alaverdi and Akhtala to determine a potentially additional source of pollutants in the environment.

House dust samples

Dust, created because of mining activities, especially those conducted in an open-cast way, is transported through the atmosphere and deposited around the mines. To understand the potential exposure of local residents of the surrounding villages to heavy metals transported through dust, 40 samples of dust were taken. One sample was taken from the village of Yagdan, located 6 km west of Mghart, which was supposed to be a control site. However, the heavy metal levels at the control location were significant, a fact that warrants further more thorough research at that site.

Plant samples

Under this study 28 samples of local agricultural products were collected from private households in the Tumanyan region to understand human exposure via agricultural products. A total of 15 plant species, including eight species of fruits, one species of pods, one species of bulb vegetables, three species of root and tuber vegetables, and two species of fresh herbs or leafy vegetables were sampled.

Sampling and analytical methods for environmental samples

For each location the soil and sediment samples were taken as mixed samples formed of several partial subsamples. The plant samples were sampled from private gardens and orchards on the farms that were investigated. Several subsamples of the same plant species were randomly taken to form composite samples and ensure their representativeness.

The samples were initially stored at low temperatures. The methodology used for the environmental samples is presented in Table 4.

The results are present in mg/kg dry matter for the soil and sediment samples, in µg/sample for the indoor dust samples, and mg/kg fresh matter in the plant samples.

Table 4 *Sampling methods and analytical techniques used for heavy metal detection in the environmental samples*

Sample	Sampling method	Year	Mineralization	Analytical technique	Standard method
Soil, sediment	Soil: sampling with a steel trowel (5 subsamples) Sediment: sampling with a steel trowel or a plexi-glass core sampler from the shore Removing vegetal cover- homogenization- quartation – storage in plastic containers/zip-lock bags	2018-2019	HNO_3, HCl	(AAS) with microwave Plasma Atomic Emission Spectrometer	-
		2021	HNO_3, HF, $H2O_2$	ICP-OES	SOP 040 SOP 201.01 (B)
Indoor dust	Sampling on a wet paper napkin from a specific area Storage in zip-lock plastic bags	2020-2021	Acids mix+ microwave	ICP-MS	SOP 201.01 (D) SOP 201.03
Plants	Random sampling of subsamples of the same plant Storage in zip-lock plastic bags	2019	HNO_3, HCl	ICP-MS AMA-254 Single-Purpose Atomic Absorption Spectrometer	-

4.2 Human food samples

Sampling sites for chicken eggs, cheese, and fish were selected taking into account the impact of emissions from the Alaverdi copper smelting plant and other industrial facilities.

Fish samples

Six samples of freshwater fish were taken from the Debed River, in areas potentially burdened by mining, as well as the activities of the Alaverdi copper smelting plant and its wastes.

Cheese samples

Five samples of homemade cow's-milk cheese were collected in the communities of Mets Ayrum, Akori, and Mghart.

Free-range chicken egg samples

In the Tumanyan region, six free-range chicken eggs were taken from local households in 2018-2020. One mixed sample of chicken eggs was purchased from a supermarket in Yerevan as a control sample.

Analytical methods for human food samples

All the egg samples and one pooled fish sample were analysed for their content of individual PCDD/Fs and dioxin-like PCBs (DL PCBs) in an ISO 17025-accredited laboratory at the State Veterinary Institute, an accredited laboratory in Prague, Czech Republic, with a resolution >10,000 using 13C 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 [2]. The identification and quantification of the analyte was conducted by gas chromatography coupled with high-resolution mass spectrometry detection (GC/HRMS).

The samples were also analysed for their content of indicator congeners of PCBs (iPCBs), oorganochlorine pesticides (OCPs), pentachlorobenzene (PeCB), and hexachlorobutadiene (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 identification and quantification of the analyte were conducted by gas chromatography coupled with tandem mass spectrometry detection in electron ionization mode (GC/MS/MS).

The results are presented in pg WHO TEQ g^{-1} of fat in the eggs and in wet weight (ww) in the fish samples if not specified otherwise. TEFs (Toxic Equivalency Factor) defined in 2005 [3] were used to evaluate dioxin toxicity in the samples.

The results were compared with the tolerable daily intake (TDI) newly established by the European Food Safety Authority at a level of 0.25 pg WHO-TEQ kg^{-1} bw day^{-1} [4].

4.3 Biological samples

Biological samples were taken from non-smokers who had been living for a long time in the area of interest. As part of each sampling protocol, there was a questionnaire designed to carry information about possible sources of exposure to heavy metals and other clarifying information (age and gender of the participant, occupation, presence of a smoker in the family, frequency of fish consumption, etc.). Samples of nails from hands and feet were collected in a total amount of 0.2 g. For urine samples fresh morning urine (10 ml) was put into sterile containers. All the samples were put into zip-lock plastic bags. The urine samples were kept at low temperatures during storage and transportation.

Hair samples

A total of 41 hair samples were collected from local inhabitants of the affected area during the years 2018-2021. Our dataset consists of 34 (83%) female and 7 (17%) male hair samples (Figure 4).

The metals As, Cu, Pb, and Hg were measured in all samples. In the hair samples taken in 2019 and 2021 Cd, Mo, and Ni were additionally determined. For the hair samples approximately 30 strands of hair were cut from the occipital region of the head, as close to the scalp as possible.

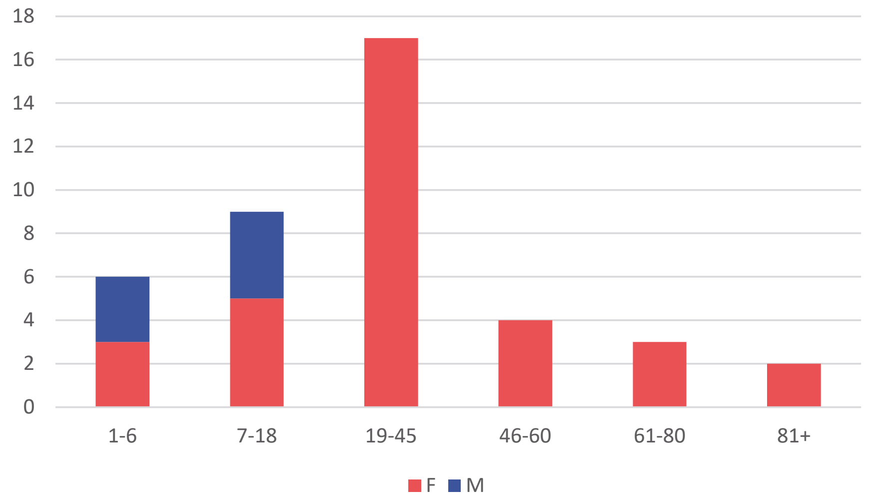

Figure 4 *Distribution of female and male hair samples taken from different age groups (years) (F – pink, M – blue)*

Nail samples

As an additional matrix to determine the impact of heavy metals on the local population, 25 samples of fingernails and toenails were collected from residents of three communities (Chochkan, Mets Ayrum, and Pokr Ayrum) and residents of the control community, Achajur (Tavush Province).

In the event of an insufficient number of nail samples from the upper and lower extremities, a mixed sample of the nails of the hands and feet of the same person was evaluated. 17 samples from the adult population were analysed, including 12 samples from women and five samples from men (Figure 5).

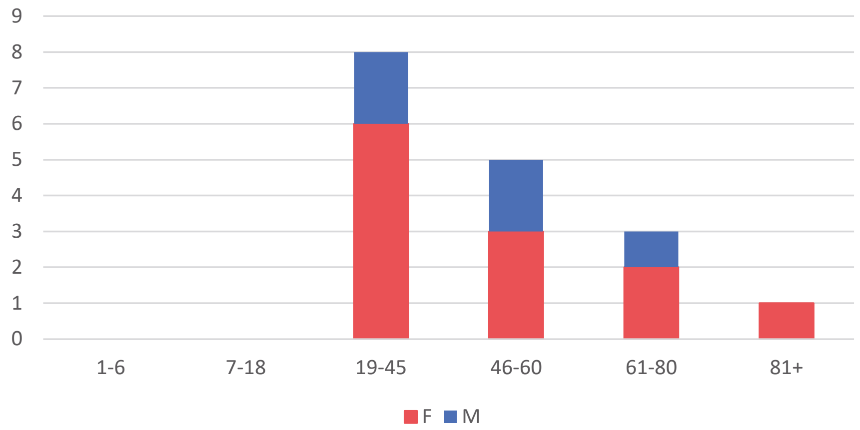

Figure 5 *Distribution of nail samples taken from women and men of different age groups (years) (F – pink, M – blue)*

Urine samples

To monitor the content of heavy metals in the urine of residents of the study area, five elements (As, Cd, Cu, Ni, and Pb) were selected. During 2020-2021, a total of 138 urine samples were collected, of which 82 were from women, 26 per cent of these from children below 18 years old, and 56 from men, 55 per cent of them children below 18 years of age (Figure 6). Ten urine samples were collected from the control area, Tavush Province, in the village of Achajur.

The content of heavy metals in urine is given in units of µg/g creatinine. Eighteen samples with creatinine levels below 0.3 g and above 3.0 g were excluded from the total evaluation.

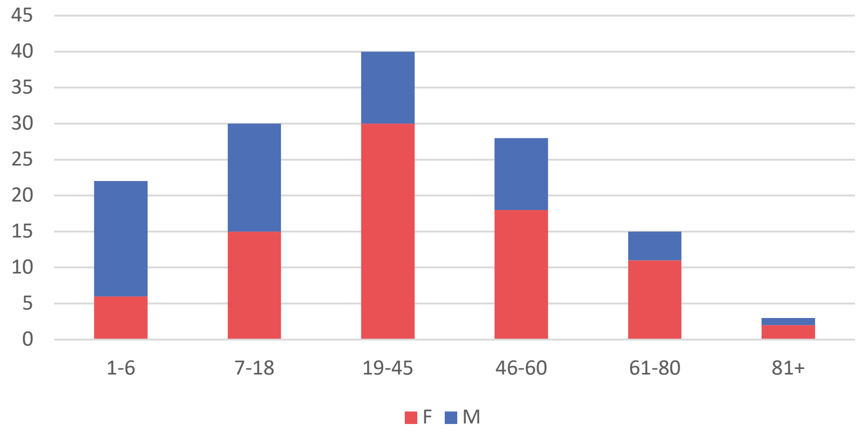

Figure 6 *Distribution of urine samples taken from women and men of different age groups (years) (F – pink, M – blue)*

Analytical methods for biological samples

Below (in Table 5) the analytical methods that were applied to the biological samples are summarized.

Table 5 *Analytical techniques used for detection of heavy metals in biological samples*

Sample	Year	Mineralization	Analytical technique	Standard method
Hair	2018–2019	HNO_3 HCl	(AAS) with microwave Plasma Atomic Emission Spectrometer	-
	2021	Acids mix+ microwave	ICP-MS	SOP 201.03 SOP 200.03 (C)
Nails	2020	Acids mix+ microwave	ICP-MS	SOP 201.03
Urine	2020–2021	-	ICP-MS	SOP 201.03 SOP 503

4.3.1 RISC evaluation and health risk assessment

Risk-Integrated Software for Cleanups (RISC) is a software package developed to assess risks to human health in contaminated areas. It can integrate up to fourteen possible exposure pathways, and calculates carcinogenic and non-carcinogenic risks.

The health risk assessment is based on the assumption that under certain specified conditions there is a risk of damage to human health, while the risk rate from zero to maximum is determined by the type of activity, length of stay in the location, and the environmental conditions. A zero health risk is not really possible; however, the risk of damage must be minimized to an acceptable level in terms of health and environmental risks. To determine the risk, it is necessary to clarify the most important transport routes and then specify exposure scenarios for potentially threatened recipients. There are two approaches to the evaluation of the dose effects – for substances with a threshold (non-carcinogenic) and non-threshold effect.

Soil samples for the carcinogenic and non-carcinogenic risks were evaluated in Risk-Integrated Software for Cleanups (RISC). This evaluation included assessing exposure to metals by ingestion of soil (including dust ingestion), dermal contact, and the consumption of crops grown in the soil.

A more detailed description of the health risk assessment methodology used for soils and foodstuffs is provided in previous reports.[1,2]

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[1] https://arnika.org/en/publications/heavy-metals-in-lori-region

[2] https://arnika.org/en/publications/heavy-metals-in-soils-foodstuffs-and-human-hair-in-the-mining-and-metallurgical-communities-of-alaverdi-and-akthala-lori- Province-of-armenia

Table 6 *Overview of samples collected in the Tumanyan and Stepanavan regions from 2018 to 2021*

Sample matrix	Alaverdi	Akori	Haghpat	Akhtala	Shamlugh	Pokr Ayrum	Mets Ayrum	Chochkan	Teghut	Shnogh	Armanis	Mghart	Reference	TOTAL
Soil	20	6	5	2	1	2	2	1	8	2	3	2	2	**56**
Sediment	8	-	-	9	-	-	1	3	8	-	2	-	2	**33**
Slag	1	-	-	1	-	-	-	-	-	-	-	-	-	**2**
Indoor dust	6	2	1	1	2	1	13	5	3	2	1	2	1	**40**
Products	6	3	4	4	2	-	6	3	-	-	-	-	-	**28**
Cheese	-	2	-	-	-	-	1	-	-	-	-	2	-	**5**
Eggs	5	-	-	-	-	-	1	-	-	-	-	-	1	**7**
Fish	6	-	-	-	-	-	-	-	-	-	-	-	-	**6**
Hair	18	4	6	2	2	-	3	-	3	2	-	1	-	**41**
Urine	19	8	8	7	5	12	41	10	11	10	2	5	10	**148**
Nails	-	-	-	-	-	1	10	11	-	-	-	-	3	**25**
TOTAL	**90**	**25**	**24**	**26**	**12**	**16**	**78**	**33**	**33**	**16**	**8**	**12**	**19**	**392**

5 Results and Discussion

5.1 Environmental samples

5.1.1 Heavy metals in soil

The soils in the metallurgically impacted Tumanyan and Stepanavan regions have higher levels of heavy metals (HM) than those in the area without industrial activity. The Dilijan National Park in the Tavush Province was chosen as a control zone without industrial activity; two samples were taken there with control concentrations of HM in the soil (in Table 7 – mean ref).

Table 7 below provides basic statistics on the presence of HM in 54 soil samples collected in the Tumanyan region.

Table 7 *Statistics on HM presence in soil samples (mg/kg dm), <LOQ-under limit of quantification*

	As	Cd	Cr	Cu	Mo	Ni	Pb	Zn
mg/kg dm								
min	3.1	0.1	8.4	47.0	0.3	5.7	6.8	77.4
max	147	12.9	108	7 737	72.7	74.0	206	1 823
mean	32.7	1.3	41.3	726	7.5	32.6	65.4	326
mean ref	5.7	0.1	27.2	29.3	<LOQ	17.6	9.7	70.1

The concentration of HM decreases with an increase in the distance from the source of pollution and is more massive in the direction of the prevailing wind. Within the town of Alaverdi, the distribution of metals in parts of the town gradually decreases in the direction from the nearest spot to the smelting plant (the city centre) to the furthest part (Sanahin) (Figure 2). The highest levels of HM taken in Alaverdi were observed in the town centre. The lowest concentrations were present in Sanahin (Table 8). This suggests that the source of pollution is the Alaverdi copper smelting plant.

As expected, high levels of Cu were not observed in the vicinity of the gold mining sites (Armanis, Mghart), because copper is not present in large amounts in the ores located in this area. On the other hand, high levels of Cr (76 mg/kg dm) were found at those sites, which is very worrying for these communities.

Table 8 *Average HM concentrations in communities of interest in the Tumanyan and Stepanavan regions (mg/kg dm), (* - location with only one sample)*

	As	Cd	Cr	Cu	Mo	Ni	Pb	Zn
mg/kg dm								
Alaverdi	**39.8**	**1.2**	**35.9**	**1 000**	**11.0**	**28.7**	**78.1**	**389**
Alaverdi-centre	71.0	2.2	42.1	3 161	13.0	31.2	136	846
Alaverdi-Sarahart	42.4	0.5	32.5	353	28.6	24.1	66.2	389
Alaverdi-Sanahin	27.1	0.7	48.3	216	3.2	36.8	38.0	199
Akori	23.2	1.4	43.7	397	2.7	34.6	70.7	237
Alaverdi-Lenanker (Madan)	68.2	2.0	10.2	874	2.3	8.2	128	341
Odzun	13.3	0.3	26.7	122	0.9	30.5	25.6	113
Alaverdi-Kayaran*	31.3	0.19	11.4	121	4.96	12.5	14.5	158
Haghpat	18.9	0.8	41.1	307	1.5	30.1	49.6	379
Teghut	**12.5**	**0.4**	**25.6**	**470**	**9.6**	**21.7**	**21.9**	**162**
Shnogh	8.0	0.4	40.2	123	3.6	32.5	20.5	164
Akhtala	**46.5**	**6.8**	**38.5**	**1 021**	**2.4**	**32.3**	**118**	**260**
Shamlugh*	41.0	0.9	67.6	435	2.0	58.4	115	-
Mets Ayrum	30.0	0.6	30.7	125	1.2	27.7	50.8	-
Pokr Ayrum	42.8	2.0	89.5	636	3.8	73.0	64.0	246
Chochkan*	147	3.9	39.2	787	5.9	52.5	122	-
Armanis	11.8	0.6	75.3	79.3	2.1	51.0	54.0	232
Mghart	14.0	0.7	76.4	92.0	3.1	49.9	53.0	321

Among other communities belonging to the enlarged community of Alaverdi (Odzun, Alaverdi-Kayaran, Haghpat, Akori, and Madan), the presence of HM was, on average, at a lower level than in the centre of Alaverdi. The highest concentrations of HM in soil were observed in the communities of Madan and Akori, located in the direction of the main winds blowing from Alaverdi. The lowest concentrations were found in the communities of Odzun and Alaverdi-Kayaran, which are located about 5-7 km southwest of Alaverdi and where the main winds do not blow. There was a significant difference from the centre of Alaverdi in terms of the presence of HM for Odzun and Alaverdi-Kayaran (26 times lower for Cu, five and nine times lower for Zn and Pb, and more than five and seven times lower for Pb in Odzun and Alaverdi-Kayaran respectively).

High levels of Cu, Zn, Pb, Cr, and As were detected in the soil samples in the town of Akhtala and neighbourhood locations close to the open-cast mine or tailing dumps (Table 8). Moreover, considerably high levels of HM were observed in a private agricultural garden located in Chochkan, watered with water taken from the River Debed.

In general, the results vary in terms of their location, orientation, and exposure to pollution. The main sources of soil pollution are emissions of pollutants into the air and the spread of dust from the mining and processing industries. Another significant contributor is the irrigation of soil with polluted water. Especially in the case of Cu, As, Cr, and Pb, the highest concentrations were found in samples located near copper mines and smelting factories in the towns of Alaverdi and Akhtala and in neighbouring villages located near factories or other industrial facilities.

Comparison with legislation

All of the soil samples (54 samples) fail to meet the Armenian limits of Order No. 01-N of 25 January 2010 of the Minister of Health of the Republic of Armenia “On Approving Sanitary Rules and Norms N 2.1.7.003-10 for Sanitary Requirements for Land Quality” for at least one metal. In comparison with the national limits in other countries, in the samples that most often exceeded the values for arsenic and copper, they did not meet the French, Dutch, and Czech soil standards, which relate to the protection of the quality of agricultural soil (Table 9). Various legal criteria or reference levels of HM for soils are presented in the Annex (Table 16).

Table 9 *Number and proportions (%) of 54 soil samples that exceed any of the mentioned legal standards for each heavy metal*

	As	Cd	Cu	Mo	Ni	Pb	Cr
Armenian soil standards	53 (98%)	-	54 (100%)	-	54 (100%)	36 (67%)	54 (100%)
French soil standards	14 (26%)	0 (0%)	31(57%)	-	-	0 (0%)	-
Dutch soil standards	15 (28%)	11 (20%)	54 (100%)	0 (0%)	16 (30%)	0 (0%)	1 (2%)
Czech soil pollution indication	13 (24%)	0 (0%)	28 (52%)	-	0 (0%)	0 (0%)	-
Levels of pollution limits – industrial areas (US EPA)	53 (98%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	-
Levels of pollution limits – other areas (US EPA)	53 (98%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	0 (0%)	-

Comparison of HM in soil with other studies

Our data correspond to the findings reported in previous reports of heavy metal pollution in Alaverdi, Akhtala and their surroundings with high levels of As, Pb, and other HM [5, 6].

The results collected in this study exceed the soil pollution near the Kapan copper-molybdenum combine and the Geghanush tailing dump for As, Cd, Cr, Cu, Pb, and Zn, by from 4 to 14 times [7]. Also, the levels of Pb and Zn were higher than in soils analysed in the surroundings of the Zangezur copper-molybdenum combine [8]. The mean concentration of HM as far as As, Cu, Pb, and Mo are concerned exceeds worldwide averages and concentrations in different soil types from other countries [9].

Health risks related to heavy metal pollution

Health risks to the local population were assessed for arsenic, cadmium, nickel, and lead.

A possible pathway for arsenic-related carcinogenic risks is the ingestion of crops grown on the soils under study, but for children, it is the ingestion of soil that is a potentially dangerous pathway. Out of all 54 soil samples in 36 soil samples (67%) for children and in 17 soil samples (30%) for adults, arsenic was present at levels which may cause adverse effects to occur in the future. In two samples taken from Alaverdi and one soil sample from Chochkan unacceptable levels of arsenic meaning that serious measures must be taken immediately were found. Non-carcinogenic risks posed by arsenic are significant for children in nine soil samples from Alaverdi, Akhtala, Chochkan, and Pokr Ayrum. According to the RISC evaluation of our soil samples, arsenic is the most problematic heavy metal for human health in the hotspot area located in the Tumanyan region.

The non-carcinogenic risks to local residents posed by cadmium, nickel, and lead were also assessed. Unacceptable risks to children posed by cadmium were identified in one soil sample collected in Akhtala (12.9 mg/kg). The problematic exposure pathway for cadmium is the ingestion of crops grown on the sampled soil. This result makes cadmium the second most risky heavy metal for human health in the hotspot area. The hazard quotients for nickel and lead do not exceed the value of hazard and therefore these metals do not represent unacceptable non-carcinogenic risks.

5.1.2 Heavy metals in sediments

Heavy metals in the Debed River

Higher concentrations of HM were found downstream in Akhtala than in sediment taken from the Debed River above Alaverdi.

Figure 7 shows the distribution of HM upstream and downstream of the Alaverdi pollution hotspot. Sediments collected in the town of Alaverdi and other spots downstream showed higher concentrations of HM along the river. There are several potential sources of pollution on the Debed River. The composition of the sediment in Alaverdi could be influenced by the Alaverdi copper smelting plant, ores and stockpiles of arsenic. Near Akhtala, the

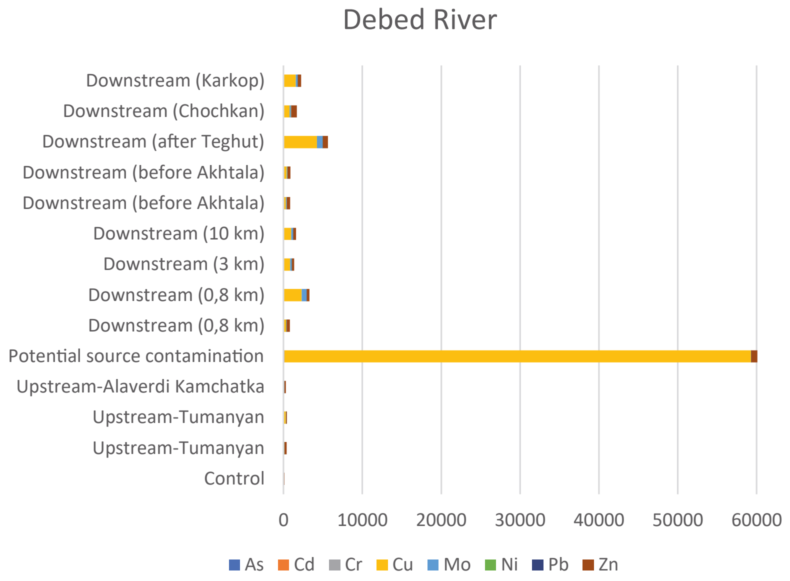

Figure 7 *Distribution of heavy metals in sediment samples taken from the Debed River (mg/kg dm)*

composition of the sediments of the Debed River may be influenced by the Akhtala River, which is a tributary of the Debed River. The Akhtala River flows through an area of open pits, a copper processing plant, and reclaimed tailings. According to local residents, in the area of Mets Ayrum, Chochkan, Teghut, and Shnogh there were numerous leaks from tailings dumps, which could be a source of pollution of the Debed River. A significant increase along the flow was observed for Cu (max 4160 mg/kg dm), Mo (max 603 mg/kg dm), Zn (max 728 mg/kg dm), and Cr (max 56 mg/kg dm). An overview of the sediment results is presented in the Annex (Table 17).

Heavy metals in the Akhtala River

On the basis of the collected data, the Akhtala River is significantly affected by industrial activity.

HM concentrations in the industrially unaffected sections of the Akhtala River, located in forests near na Shamlugh, are significantly lower than in the lower reaches, in the area of the town of Akhtala, in particular for Cu (an increase from 34 to 4555 mg/kg) and Zn (an increase from 79 up to 2310 mg/kg) (Figure 8).

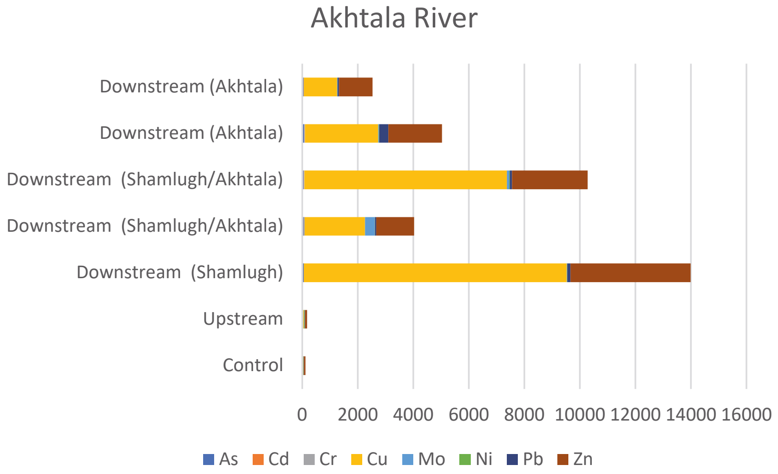

Figure 8 *HM distribution in sediments taken from the Akhtala River (mg/kg dm)*

Heavy metals in the Chqnagh River

Two sediment samples were collected from the Chqnagh River. Both samples had higher concentrations of the HM that were studied. The major HM detected in the sediments were Zn, Cr, Cu, and Ni. The section of the Chqnagh River that was investigated is located near the Armanis gold-polymetallic deposit and industrial waste dump, on the territory of the expanded community of Stepanavan, at a distance of approximately 5 km. Pollutants can spread through the air and enter the river, as well as penetrate with groundwater and rain. There is a direct source of pollution – an industrial waste dump, located in the downstream part of the river (Figure 9).

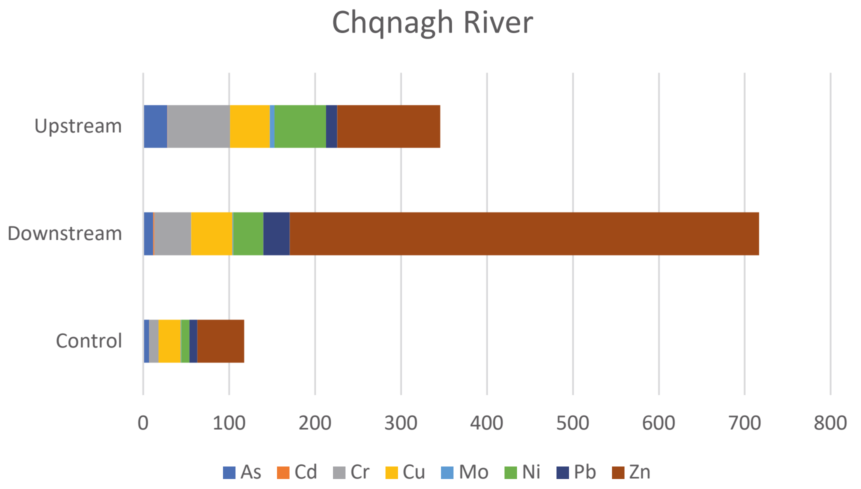

Figure 9 *HM distribution in sediments taken from the Chqnagh River (mg/kg dm)*

HM in the Shnogh River

There is no clear difference in the composition of the Shnogh River sediment samples and no visible trend in the change of the heavy metal content in the flow of the water stream (Figure 10). All samples are characterized by high Cu, Zn, and Mo contents. In addition, the composition of the tailings (a potential source of contamination) did not show significant differences from the samples collected from the Shnogh River. The higher Cu (617 mg/kg dm) and Mo (88 mg/kg dm) contents in the Upstream 1 sample are probably related to the Teghut copper-molybdenum mine, which is located upstream.

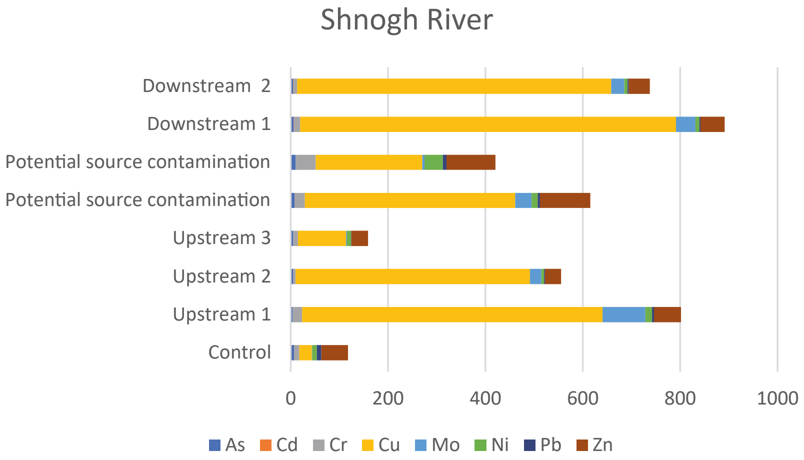

Figure 10 *HM distribution in sediments taken from the Shnogh River (mg/kg dm)*

Comparison of heavy metals in sediments of the Debed River with other studies

The sediment composition of the Debed River corresponds to the results from previous heavy metal research on the Debed River, in which alarming concentrations of Mn, Pb, Cu, Zn, and Cd were found [10]. Our results are comparable with the results obtained in studies of HM in the sediments of the Voghji River, which flows through the territory of the industrial towns of Kajaran and Kapan, in Syunik Province in Armenia. The increased content of HM in this region is associated with mining activities and mining facilities that affect the environment [11].

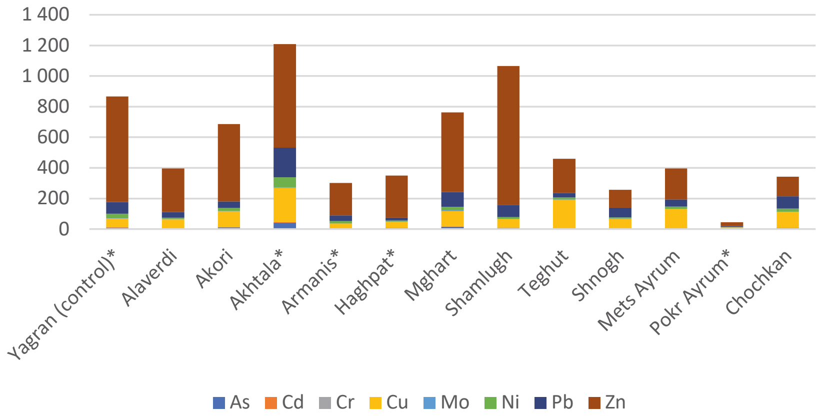

Figure 11 *HM distribution in indoor dust samples (µg/sample),*
(- location only with one sample)*

5.1.3 Heavy metals in indoor dust

Figure 11 shows the distribution of HM in indoor dust at each site that was studied. In addition to the eight listed HM, the presence of iron (Fe) in dust was also studied under this study.

The major elements in dust were Fe (5285 µg/sample), followed by Zn (270 µg/sample), Cu (102 µg/sample), and Pb (55 µg/sample), which were present in household dust in high levels. The predominant part of the sample composition was Fe; however, since Fe is not supposed to be related to the potential hotspots, Fe concentrations are not included in the graph below. Complete data for the indoor dust samples are presented in the Annex (Table 18).

A higher presence of Cu was observed in communities located near the opencast mines or ore processing plants (Akhtala, Teghut). The same elements were found in large quantities in soil samples taken in the Tumanyan and Stepanavan regions. Cd and Mo were present in the soil samples at low levels. Monitoring of indoor and outdoor dust in locations where the mining industry is active should be conducted regularly.

Comparison of HM in indoor dust with other studies

Taking methodological limitations (using µg/sample units) into consideration, it is not possible to compare the results collected under this study with other scientific papers. However, it might help with the identification of the elements with the most potential risks in affected areas.

Inhalation and other ways of ingesting dust pose serious risks associated with exposure to heavy metals in an indoor environment. In mining regions, dust is known to be an important source of several respiratory illnesses and can lead to carcinogenic effects [12].

5.1.4 Heavy metals in slag

The heaps of slag from which the samples were taken probably served as winter road cover, as they were located along the roads. The slag heaps have been exposed and therefore contaminants can be released into the environment, for example through airborne propagation. Concentrations of HM in the slag samples are given in Table 10.

Table 10 *HM concentrations in slag samples (mg/kg dm)*

	As	Cd	Cr	Cu	Mo	Ni	Pb	Zn
	mg/kg dm							
Alaverdi	30.9	0.18	204	7619	1486	4.87	8.75	839
Akhtala	54.5	1.26	6.28	545	8.51	2.25	37.2	473

5.2 Foodstuff samples

5.2.1 Heavy metals in vegetable matter

HM in food products are presented in the Annex (Table 19). Two leaf vegetable samples (basil and malva) contained higher concentrations of some risky HM, such as Hg (0.001 mg/kg fm) for the Basil and Malva plants and Pb (0.22 mg/kg fm) in the Malva plant.

The maximum levels of Cd and Pb in foodstuffs set by FAO/WHO and the European Union were met. In the case of the food safety requirements set by an Order of the Minister of Healthcare of Armenia, one sample of a leafy vegetable (Malva) exceeds the maximum permissible level of Cd (0.03 mg/kg fm).

In comparison to the recent study from the Alaverdi locality, generally, we found lower levels of Hg, Cu, Ni, and Pb in different species of vegetables and fruits except one sample of a leafy vegetable (Malva). On the other hand, in some samples we found slightly higher values of As levels in different species of fruits and vegetables [13].

5.2.2 Dioxins and dioxin-like PCBs in eggs and fish

5.2.2.1 Dioxins and dioxin-like PCBs in eggs

Table 11 summarizes the findings for PCDD/Fs and dl PCBs in the products that were collected. Three samples out of a total of six pooled free-range egg samples from Alaverdi and adjacent areas exceeded the Armenian and Russian limits for the sum of PCDD/Fs (3.0 pg WHO TEQ g^{-1} fat) [14, 15], as well as the EU ML for PCDD/Fs (2.5 pg WHO TEQ g^{-1} fat) and for PCDD/Fs and dl PCBs (5.0 pg WHO TEQ g^{-1} fat) respectively[16]. All the samples of free-range chicken eggs from Alaverdi and neighbouring locations that were collected exceeded the levels of PCDD/Fs and dl PCBs measured in the reference sample of eggs from Yerevan by two-73 and 11-182 times respectively, showing Alaverdi and its surroundings as a source of pollution with POPs.

Table 11 *Summarized results of analyses of PCDD/Fs and dl PCBs in egg and fish samples from Armenia.*

Year	Matrix	Name of the site	Eggs/fish in pooled sample	Fat	PCDD/Fs	dl PCBs	PCDD/Fs + dl PCBs
			number	%	pg WHO TEQ g^{-1} fat		
2018	**Eggs**	Alaverdi 1	3	14.4	7.5	19.3	26.8
2019		Alaverdi 2	4	12.0	14.6	25.5	40.1
2018		Alaverdi 3	4	13.1	4.4	9.1	13.5
2020		Alaverdi 4	3	12.3	1.7	3.3	5.0
2020		Sanahin (Alaverdi 5)	2	11.2	2.0	21.5	23.5
2020		Mets Ayrum	1	12.1	0.41	1.57	1.99
2018		Yerevan-Supermarket	4	8.7	0.20	0.14	0.34
	Fish (chub)				**pg WHO TEQ g^{-1} ww**		
2020		Alaverdi	6	4.8	0.10	1.25	1.35
					pg WHO TEQ g^{-1} fat		
2020		Alaverdi	6	4.8	2.0	26.1	28.1

The data of POPs collected in Alaverdi [17] in 2010 showed lower concentrations of POPs in chicken eggs (11.9 pg BEQ g^{-1} fat) in comparison to the levels of PCDD/Fs and dl PCBs in most of the samples from 2018-2020, except Alaverdi 4.

On the map below (Figure 12) it can be observed that the levels of PCDD/Fs/dl PCBs in eggs decrease with the distance from the copper smelter and that they are also quite clearly in agreement with the prevailing wind directions carrying the pollution from the copper smelting plant. This finding suggests that the copper smelting plant is the source of pollution of the environment with POPs. The lowest level of PCDD/Fs/dl PCBs (1.99 pg WHO-TEQ g^{-1} fat) was measured in the eggs from Mets Ayrum, 14 km from Alaverdi and affected rather by mining and tailing ponds which are not sources of PCDD/Fs and dl PCBs. The copper smelting plant was shut down in 2018; however, pollution most probably lingers in the soil and dust.

Comparison of PCDD/Fs and dl PCBs in eggs with other studies

The levels of 5-40.1 pg WHO-TEQ g^{-1} in the eggs from the vicinity of the copper smelting plant in Alaverdi are comparable to those measured in the neighbourhoods of other metallurgical plants, e.g. in Beihai, China (24-37 pg BEQ g^{-1} fat) [18] or near a copper smelting plant in Balkhash, Kazakhstan (12.7-30.1 pg WHO-TEQ g^{-1} fat) [19].

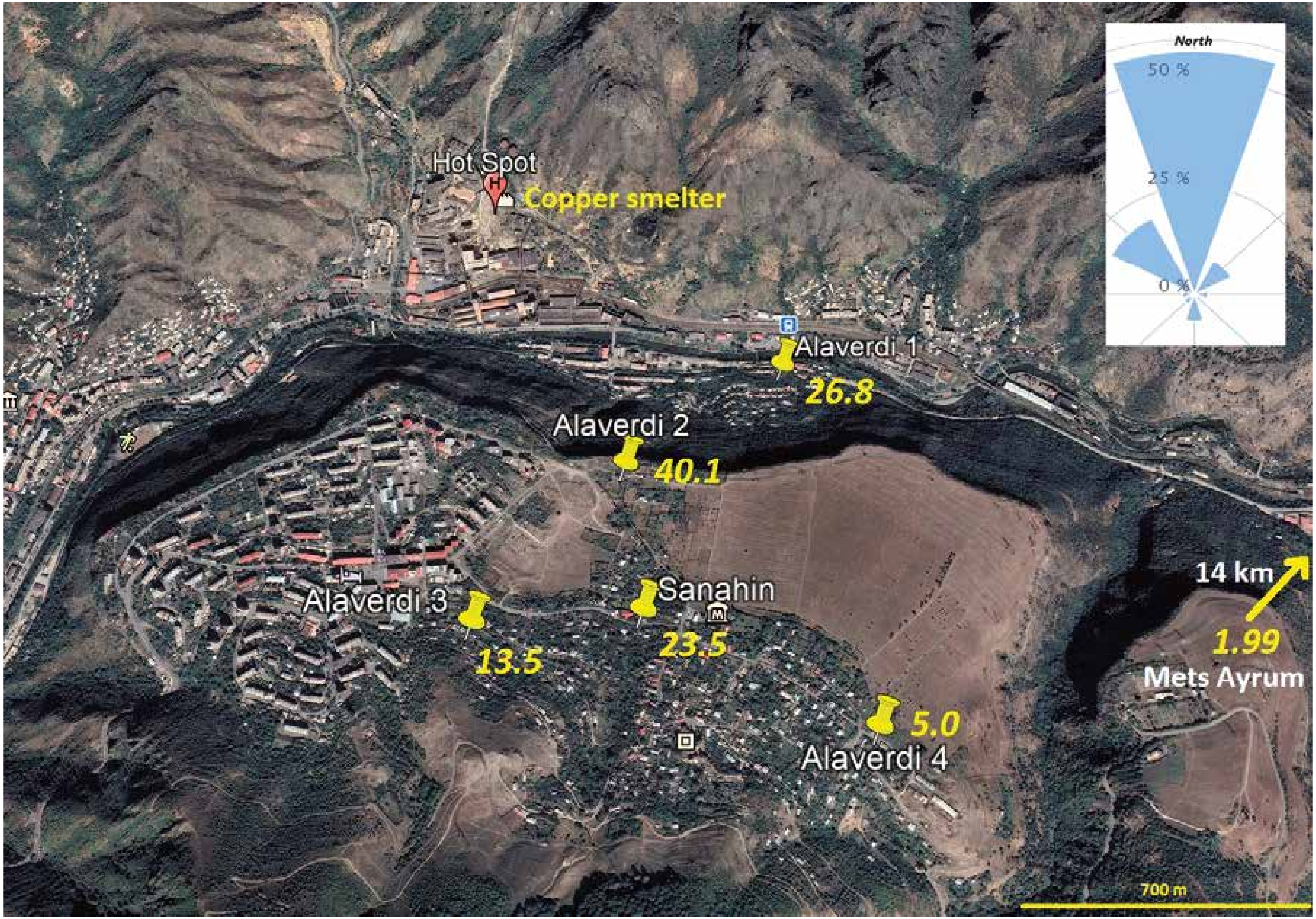

Figure 12 *Map of Alaverdi with marked sampling sites and measured levels of PCDD/Fs/dl PCBs. The directions of the prevailing winds are shown in the diagram in the upper right corner of the map. Mets Ayrum is 14 km to the north-east.*

Health risks posed by consumption of eggs

Average consumption of eggs (1/3 of an egg per day) would lead to the exceeding of the TDI for PCDD/Fs/dl PCBs by 260% and 520% for a 70-kg adult man and a 35-kg ten-year-old child respectively. This calculation was made from the levels in all five pooled egg samples from Alaverdi. For the egg sample with the highest level, the exceedance would be much higher, at levels of 457% and 915% respectively. Free-range eggs from Mets Ayrum and a supermarket in Yerevan reached 23% and 3% of the TDI for an adult. The percentage is doubled for a ten-year-old child.

5.2.2.2 Dioxins and dioxin-like PCBs in fish

PCDD/Fs and dl PCBs levels of 0.1 and 1.25 pg WHO-TEQ g^{-1} in chub from the Debed River (Table 11), sampled in the surroundings of the town of Alaverdi, were below the EU ML for PCDD/Fs and PCDD/Fs/dl PCBs of 3.5 and 6.5 pg WHO TEQ g^{-1} ww respectively set for fish meat [16]. They are also lower than the levels of PCDD/Fs and dl PCBs of 0.48-0.88 and 3.9-4.4 pg WHO-TEQ g^{-1} ww respectively measured in chub samples from the Elbe (Labe) River in 2005 [20]. Levels of PCDD/Fs and dl PCBs of 0.04-0.09 and 0.53-1.4 pg WHO-TEQ g^{-1} ww, respectively, which were also found in chub samples caught in the upper part of the Sava River, Croatia in 2015 [21] are almost the same as those observed in the pooled sample of chub from the Debed River. Chub is also considered to be a nomadic species [20], which means that the levels found in chub from Alaverdi do not necessarily show just localized contamination.

5.2.2.3 Heavy metals in fish and cheese

HM in fish

The highest concentration of the metals of interest was determined for Zn (13.9 mg/kg), but this value is not considered to be harmful to human health (Table 12). The fish samples were assessed as safe for consumption by the local population. Our result showed a lower heavy metal presence in comparison to the HM presence in the Kura scraper fish, sampled from the Debed River [10]. In addition, methylmercury was tested in the fish samples, with a maximum MeHg value of 0.023 mg/kg.

Table 12 *HM concentrations in fish samples from the Debed River (mg/kg) <LOQ – below the limit of quantification*

	As	Cd	Cu	Pb	Zn	Hg	MeHg
				mg/kg			
min	0.04	0.013	0.67	<LOQ	11.1	0.016	0.012
max	0.11	0.026	0.83	<LOQ	15.8	0.027	0.023
mean	0.08	0.020	0.73	<LOQ	13.9	0.019	0.014

HM in cheese

Homemade cheese made from cow's milk is a common food in this region. Analyses of the determined metals did not reveal high levels of the pollutants of interest. In most cases, the cheeses sampled were made from the milk of cows that graze in another region during the summer season and are not exposed to possible pollution all year round.

In all samples, minor amounts of Cu, which is a natural component of milk (a biological trace element), were detected [22]. In two samples of cow's-milk cheese from Mghart, there were detectable amounts of both Mo and Ni compared to the samples from other locations. Most of the metals were below the limit of quantification (see Annex – Table 20).

6 Biological samples

6.1.1 Heavy metals in hair

Cu, Pb, and Hg were detected in almost all the samples (95-98%), whereas As was detected in only 27% of the samples. An overview of the results is shown in Table 13. In all the samples that were tested the amount of Mo was below the limit of quantification (<LOQ).

It is not possible to compare the quantities of pollutants in samples in different age categories for females and males because of the narrow age range in the case of male samples (age range two-17 years). When the samples from children below 18 years of age were compared, the presence of Pb, Ni, and Cd was higher in the girls. With regard to the rest of the metals, their content was either the same or slightly different (Table 13).

Table 13 *Heavy metal presence in hair samples from the Tumanyan and Stepanavan Regions (mg/kg), (* - location with one sample, ** - one outlying sample was excluded, *** - mean concentration of HM in female samples aged <18)*

	As	Cd	Cu**	Mo	Ni	Pb	Hg	Age range/mean
				mg/kg				(yrs)
>LOQ (№/%)	11 (27%)	20 (74%)	39 (95%)	0 (0%)	13(48%)	39 (95%)	40 (98%)	
min	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ	
max	0.480	2.24	53.40	<LOQ	45.10	23.20	0.650	
mean	0.190	0.151	13.27	<LOQ	13.96	1.73	0.086	2-85/28.4
Alaverdi	0.134	0.028	13.92	<LOQ	14.65	1.37	0.072	
Akori	0.140	0.040	8.46	<LOQ	35.70	0.98	0.196	
Sanahin	<LOQ	0.020	9.60	<LOQ	2.30	0.57	0.107	
Teghut	<LOQ	<LOQ	11.33	<LOQ	<LOQ	0.45	0.059	

	As	Cd	Cu**	Mo	Ni	Pb	Hg	Age range/mean
Shnogh	<LOQ	0.055	12.50	<LOQ	2.10	1.16	0.035	
Mghart*	<LOQ	0.090	14.00	<LOQ	1.3	0.46	0.097	
Haghpat	0.440	1.130	19.68	<LOQ	5.20	5.30	0.099	
Mets Ayrum	<LOQ	0.023	11.00	<LOQ	6.57	0.50	0.032	
Shamlugh	<LOQ	0.065	10.47	<LOQ	1.80	0.77	0.084	
mean (F)	**0.19**	**0.22**	**13.9**	**<LOQ**	**14.1**	**1.95**	**0.09**	**4-85/ 32.6**
mean (M)	**0.10**	**0.10**	**10.5**	**<LOQ**	**17.4**	**0.8**	**0.1**	**2-17/ 9.4**
Mean (F) ***	0.20	0.50	10.6	<LOQ	20.3	1.7	0.1	4-11/ 7.75

When the total datasets of females and males are compared, higher concentrations of metals can be seen in the female samples. This may be due to a different metabolic rate and capacity for accumulation, with higher accumulation in females and a greater age range in the group of women's samples. The average age of a woman was determined to be almost 33 years, which is higher than the average age of the male population (nine years) that took part in the testing. It is therefore necessary to take into account the longer exposure of the women compared to the men in this study.

The highest Cu concentration in hair was observed in the Haghpat, Alaverdi, and Mghart samples. Moreover, in one sample of female hair (37 yrs) Cu was detected at a level of 1426 mg/kg and Pb at 23 mg/kg. This leads to concern regarding the health risk for women of reproductive age. A significantly higher amount of Ni was found in the hair samples taken in Alaverdi and Akori.

The findings of our research did not find that the HM of interest were significantly deposited in the hair. Compared to the scientific studies, our results were mostly below or at the edge of the results from other reports [23, 24]. Therefore, the occurrence of HM and their effect on human health in the local population should be investigated further, especially the concentration in blood or other body tissues.

6.1.2 Heavy metals in urine

When the average values of samples from the potentially contaminated Tumanyan and Stepanavan regions and from the control region of Tavush Province were compared, higher values were recorded for the affected area for all metals. In most cases, the difference was more than twofold. The distribution of heavy metals in the urine samples from local residents is shown in Table 14.

Table 14 *Average HM concentrations in communities of interest in the Tumanyan and Stepanavan regions (µg/g creatinine)*

	As	Cd	Cu	Ni	Pb
	µg/g creatinine				
>LOQ (№/%)	134 (97%)	71 (51%)	130 (94%)	8(6%)	78 (57%)
min	<LOQ	<LOQ	<LOQ	<LOQ	<LOQ
max	63.0	16.0	50.0	15.0	10.0
mean	19.8	0.9	13.0	8.6	2.3
mean ref	12.8	0.3	7.2	<LOQ	1.5
Alaverdi	22.5	3.5	15.5	<LOQ	4.0
Haghpat	20.9	0.4	12.5	5.0	1.5
Akori	28.1	0.7	13.4	7.5	2.2
Alaverdi-Kayaran	21.0	0.8	28.7	<LOQ	3.5
Teghut	14.3	0.4	12.6	10.7	1.3
Shnogh	10.3	0.6	8.8	8.0	2.3
Akhtala	22.7	0.6	24.0	<LOQ	2.8
Shamlugh	26.0	0.4	13.5	<LOQ	3.3
Mets Ayrum	18.6	0.3	11.3	<LOQ	1.9
Pokr Ayrum	25.8	0.4	8.5	<LOQ	1.8
Chochkan	16.3	0.3	9.9	<LOQ	2.3
Armanis	10.5	<LOQ	10.0	<LOQ	1.0
Mghart	15.0	0.3	19.0	<LOQ	1.0

The graphs in Figure 13 and Figure 14 show the difference between the results for As and Cu, metals that were found in almost all of the urine samples. The male and female population samples were divided into two age groups (under 18 years and over 18 years).

The higher content of As and Cu belongs to the children's age group. The mean As level in the child population was higher in the group of boys (25.7 μg/g creatinine) compared to the girls (21.6 μg/g creatinine). Cu levels, on the other hand, were higher in the child population in the girls (18.1 μg/g creatinine) compared to those (15.7 μg/g creatinine) found in the boys. In the adult age group, Cu and As levels were higher in the women's group, with 11.9 and 7.5 μg/g creatinine for Cu and 20.4 and 17.1 μg/g creatinine for As respectively.

Comparison of HM content in urine with legislation

In the case of three samples collected from Alaverdi, Pokr Ayrum, and Shamlugh belonging to boys aged 4, 10, and 13 years, the professional limit according to Decree 432/2003 Coll. for the protection of workers' health in the Czech Republic was exceeded (0.05 mg/g creatinine or 0.075 μmol/mmol creatinine for the samples taken at the end of the working week). The presence of arsenic in these three children's samples was 55, 58, and 68 μg/g creatinine. This raises concerns about the high risk and health burden, especially for children's health.

Comparison of HM content in urine with other studies

Our findings were compared with results obtained in other studies. The average As content was higher than in urine samples studied in the USA and Germany [25, 26], but still lower than in a historically polluted area in the Czech Republic, Kaňk [27]. The average Cu and Cd concentrations in this study were higher than those shown in data from other mining regions in the USA, Spain, and Germany [25, 26, 28].

6.1.3 Heavy metals in nails

The basic statistics are shown in Table 15. The highest values were recorded in the village of Chochkan. Molybdenum was not present in any of the nail samples analysed. Comparing the results of nails from the reference area, the samples of interest contained on average higher amounts of five out of eight metals, namely As, Cd, Cu, Pb, and Zn.

Nail samples collected from locals from the Chochkan community showed higher levels of As, Cu, Fe, Ni, and Zn when compared to the Mets and Pokr Ayrum communities. The average pollutant concentrations from this site exceeded the HM sampled at the reference site. For As, Cu, and Pb, the content was more than twice as high.

For samples collected and analysed separately, there was a difference in the distribution of HM between fingernails and toenails. Cu was present in higher amounts in the fingernails than in the toenails in all samples. In four of the five samples, higher concentrations of Fe and Zn were also present in the fingernails.

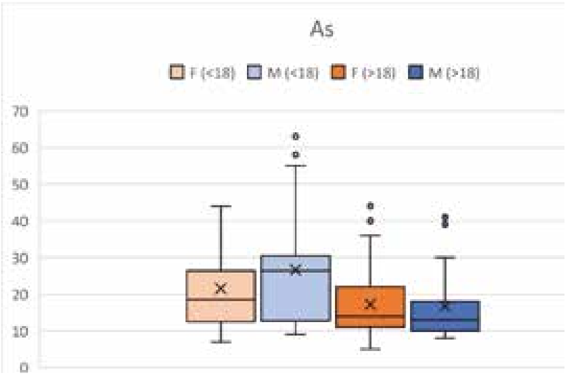

Figure 13 *Distribution of arsenic content (μg/g creatinine) in urine (light blue – M <18, blue >18, light orange F<18, orange – F >18)*

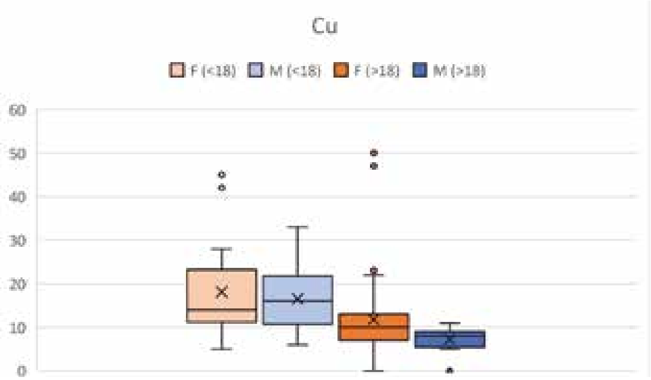

Figure 14 *of copper content (μg/g creatinine) in urine (light blue – M <18, blue >18, light orange F<18, orange – F >18)*

Table 15 *Average HM concentrations in nail samples of locals from communities of interest in the Tumanyan region (mg/kg) (* - location only with one sample)*

	As	Cd	Cu	Fe	Mo	Ni	Pb	Zn
				mg/kg				
>LOQ (№/%)	18 (78%)	22 (96%)	23 (100%)	23 (100%)	0 (0%)	10 (44%)	23 (100%)	23 (100%)
min	0.10	0.01	0.8	45.0	<LOQ	1.3	0.1	11.0
max	0.74	0.16	166	485	<LOQ	31.7	3.9	234
mean	0.31	0.04	13.5	198	<LOQ	9.3	1.4	119
mean ref	0.18	0.02	9.03	232	<LOQ	13.2	0.74	98.0
Mets Ayrum	0.35	0.04	5.3	164	<LOQ	8.5	1.2	113
Pokr Ayrum*	<LOQ	0.04	4.8	4.8	<LOQ	4.0	0.6	113
Chochkan	0.28	0.05	21.8	240	<LOQ	11.7	1.7	124

7 Conclusion

This study investigated the distribution of heavy metals and persistent organic pollutants in the vicinity of potential sources of pollution in the Tumanyan and Stepanavan regions, located in northern Armenia.

On the basis of our results, state that there is a direct impact of the activities of the mining and manufacturing industry and its infrastructure on the environment of the region, where about 40 thousand people live.

The results obtained in this work show a certain impact of industry on the environment. Our data in many cases shows a decrease in the quantities of pollutants in the direction from the source of pollution, thereby identifying the main sources of pollution. It can be assumed that the industrial facilities located in Alaverdi, Akhtala, Teghut, Armanis, and Mghart are sources of environmental pollution and a burden on the health of local residents.

This statement is confirmed by our results, which can be summarized in the following points:

» on average, at all the points that were studied, the indicators for heavy metals in the soil and river sediments were higher than in the control area;

» in soil samples, higher values were detected in the vicinity of polluting entities; moreover, in the case of Alaverdi, the spread of the pollution corresponded to the direction of the prevailing winds. The highest concentrations were found for Cu, Zn, and Pb;

» the results for sediments showed a difference in the content of heavy metals in the points located upstream from the polluted places, as compared to the points located in the affected zone. This trend was well mapped on the Debed and Akhtala Rivers, where the difference was especially visible for such elements as Cu, Mo, Zn, and Cr.

The increased presence of pollution in the environment as a result burdens the food chain:

» the free-range chicken eggs sampled in Alaverdi showed high levels of dioxins and dioxin-like compounds, with concentrations decreasing with distance from the copper smelting plant in Alaverdi. The content of dioxins in domestic eggs exceeded by 2-73 times the dioxin content in the control eggs bought in Yerevan; for dioxin-like compounds, the excess was in the range of 11-182 compared to the control eggs that were bought;

» the presence of heavy metals and POPs in the environment and food affects the general health of local residents who have been under the direct influence of pollutants for a long time.

This impact on human health can be characterized in the following points:

» the risk assessment in the case of arsenic and cadmium showed increased health risks to local residents, both in the child population (67% of samples) and in the adult population (30%) in the case of arsenic. In three samples tested for arsenic and in one sample tested for cadmium their presence was unacceptable;

- the free-range chicken eggs sampled in Alaverdi showed high levels of dioxins and dioxin-like substances, exceeding the TDI by 520% for a ten-year-old child and 260% for an adult. In the case of the Alaverdi egg with the highest dioxin content (40.1 pg WHO TEQ g-1 fat), this limit was exceeded by 415% for an adult and 915% for a child;
- the results of the study found higher levels of heavy metals in the child urine samples than in the adult subsample. The presence of arsenic in high concentrations in the child population is of high concern. Samples taken from boys aged 4, 10 and 13 exceeded the Czech occupational limit for workers. This is of particular concern for the safety of the child population;
- The values for all the recoverable metals (As, Cd, Cu, Ni, and Pb) in the urine of the residents of the Tumanyan region were higher than in the control group, which lives in a non-industrialized region in Armenia.

Since heavy metals and persistent organic pollutants degrade slowly, these substances remain in the environment for a long time. Therefore, local residents are susceptible to the long-term effects of pollution on their health. The most vulnerable group is the child population, because of their lower body weight and more frequent hand-to-mouth contact. These results confirm and draw attention to the need for regular control and monitoring of pollution.
These results confirm and draw attention to the need for regular control and monitoring of pollution in the Tumanyan and Stepanavan regions.

8 Annex

8.1 Overview of heavy metals and their health impact

8.1.1 Arsenic

Arsenic (As) occurs naturally in many forms, either as inorganic compounds (mainly sulphides) or as organic compounds, and accompanies some ore deposits such as gold, silver, lead, copper, nickel, cobalt, antimony, iron, etc [29]. A significant anthropogenic source of arsenic is the mining and metallurgical industry [30], as well as the burning of coal (especially lignite) [31].

Acute (short-term) inhalation exposure to arsenic dust or vapours at high levels leads to gastrointestinal effects (nausea, diarrhoea, abdominal pain); disorders of the central and peripheral nervous system can occur in workers acutely exposed to inorganic arsenic [32, 33]. Chronic (long-term) inhalation exposure to inorganic arsenic in humans is associated with irritation of the skin and mucous membranes and effects on the brain and nervous system [34-36].

Arsenic itself and arsenic trioxide were allocated by the International Agency for Research on Cancer (IARC) [37] into the group 1 as human carcinogens and their link to the occurrence of lung and bladder cancer is considered sufficient. The IARC considers the evidence of the influence of arsenic on prostate, liver, and kidney cancer only as partially sufficient. Non-carcinogenic health risks of arsenic exposure are connected with the deceleration of foetal development, influencing the neuropsychic development of children, influence on the central and peripheral nervous system, and with heart and vessel diseases [38]. The toxicity of arsenic is dependent on its speciation; arsenic (III) compounds are more toxic than AS(V) [39].

8.1.2 Cadmium

Cadmium (Cd) is a heavy metal chemically related to zinc, which is most commonly naturally present with copper, zinc, or lead in ore as sulphides of these metals [40]. Cadmium is used as a stabilizer of selected plastics and in metallurgy and other industrial branches. In the past, cadmium was abundantly used in many electric and electronic devices as well; consequently, another significant source of cadmium is the processing of electronic waste [41, 42].

Cadmium has the ability to accumulate within living organisms, including the human body (bioaccumulation). About half of the total amount of cadmium in the body is found in the kidneys, where it binds to metallothionein, a cysteine-rich protein, and itcause damage to these organs [43]. In addition, complementary cadmium displaces and replaces calcium absorbed by the intestines. Calcium deficiency in the body is reflected in demineralization, lower bone density, and impaired vitamin D3 metabolism [44]. Cadmium also has a negative effect on the hormonal system, especially sex hormones [45].

According to the IARC, cadmium is considered as a confirmed human carcinogen (group 1) which can disrupt the genetic information in cells (genotoxicity) and cause damage to human foetuses during their prenatal development (teratogenicity). In the case of exposure to cadmium, the IARC confirms the existence of sufficient evidence for the development of lung cancer, and additionally, the influence of cadmium has also been observed in kidney, mammary, and prostate cancer [46].

8.1.3 Lead

Lead (Pb) is present in the soil as a result of human activity or natural processes such as pedogenesis and rock weathering. In soil, lead forms sulphides, sulphates, or carbonates. The presence of lead in the environment is mainly associated with the mining and processing of ores, the disposal of tailings, transport, and the operation of smelting plants, gasworks, and incinerators [47-49]. Lead contamination can penetrate the soil to a depth of up to twenty centimetres, thus adversely affecting the quality of biological systems in the long term [50].

Lead has been proved to have a wide spectrum of toxic effects; it is considered one of the persistent HM and is one of the global environmental pollutants. Its presence in the blood is unfavourable in all concentrations as it is a xenobiotic for all life forms. Lead can cause effects on the blood, as well as the nervous, immune, renal, and cardiovascular systems. Exposure to high levels of lead can cause gastrointestinal symptoms and severe damage to the brain and kidneys and may have effects on reproduction [51]. Experts consider the so-called developmental neurotoxicity, i.e. damage to brain development in childhood (slow cognitive development, low IQ), to be a possible critically negative effect of lead that can severely affect children [52]. Once taken into the body, lead becomes distributed throughout the body in the blood and is accumulated in the bones [53].

The IARC has placed the inorganic compounds of lead into the group of "likely" carcinogens (group 2A), with the explanation that there have been studies conducted on animals, with sufficient but limited results [54].

8.1.4 Chromium

In nature, chromium (Cr) is a component of many minerals. It is used in a wide range of manufacturing, for example in the metallurgical and textile industries and in papermaking, and is also a component of dyes, catalysts, and fertilizers. It enters the environment through release from landfill leaching, contamination during ore extraction, or the combustion of petroleum and coal [55, 56].

Chromium (VI) is known to cause various health effects, such as skin rashes, respiratory problems, a weakened immune system, kidney and liver damage, involve oxidation stress, alterates and damages DNA and proteins [57, 58]. The inhalation of chromium (VI) compounds can result in ulceration and perforation of the mucous membranes of the nasal septum, irritation of the pharynx and larynx, asthmatic bronchitis, bronchospasms, and oedema. Respiratory symptoms may include coughing and wheezing, shortness of breath, and nasal itching. Chromium (VI) is classified by the IARC in group 1.

Significantly, chromium (III) is an essential nutrient for humans, and occurs naturally in many vegetables, fruits, meats, yeasts, and grain [59, 60].

8.1.5 Copper

Copper (Cu) is a metal naturally present mainly in the forms of sulphides, oxides, and carbides and to a lesser extent can also exist in pure metal form. Copper has been used by humans for thousands of years, but the mining and processing of its ores can be a significant source of environmental contamination [61, 62].

On one hand, copper, from a biological point of view, is one of the so-called essential elements; it is involved in the function of many enzymes and in the catalysis of significant enzymatic processes such as cellular respiration or the formation of neurotransmitters [63]. On the other hand, higher exposure to copper may pose a health risk. Acute copper toxicity can result in a variety of pathological conditions and, in extreme cases, even death. Chronic toxicity can lead to liver and kidney damage [64] and severe neurological damage [65, 66]. It has also been suggested that excess copper could also play a role in Alzheimer's disease [67].

8.1.6 Molybdenum

Molybdenum (Mo) often accompanies copper in ores and it has a strong connection to the copper system in living organisms too. Molybdenum is an element that is present in various enzymes [68]. Its toxicity is mainly associated with impaired copper metabolism and induction of copper deficiency in the body [69]. An imbalance in one's copper-molybdenum-sulphur intake can lead to anaemia, gastrointestinal disturbances, bone disorders, and growth retardation [70, 71]. Molybdenum causes a more pronounced excretion of copper from the body [69].

Both acute and prolonged exposure to excessive molybdenum may give rise to morphological changes in the liver, kidneys, and spleen. Proteinuria and functional disturbances of the liver have been reported. Other symptoms after prolonged exposure are anaemia, diarrhoea, and deformities of the joints and long bones, as well as the mandibular exostoses [72, 73].

8.1.7 Nickel

Nickel (Ni) is present in the environment especially in the form of sulphides and silicates. The contamination of the environment occurs primarily through ore mining and the metallurgical industry [31].

Prolonged oral exposure to higher doses of nickel causes changes in blood count composition, reduced iodine content in the thyroid gland, and skin irritation, and displaces essential divalent metals such as copper, zinc, calcium, magnesium, or iron from enzymes [74, 75]. Some studies on animals have proved a variety of damage done to hereditary information – DNA [37]. Moreover, considering its health impacts, nickel also acts as a very strong contact allergen, causing dermatitis [76].

The IARC has classified some compounds of nickel as confirmed human carcinogens (group 1) and nickel itself into group 2B, which is the classification of possible carcinogens.

8.1.8 Mercury

Mercury (Hg) is found naturally in many chemical and physical forms, e.g. in a solid or gaseous state. Each form of mercury has a unique toxicological profile, and differs in the mechanisms of transport and disposition in the body and metabolic fate. Mercury is spread via rock dust particles as a result of the erosion and weathering processes, deposited on the earth's surface, or delivered to water surfaces. The primary route of Hg exposure for humans is the inhalation of mercury vapour. The anthropogenic origin of mercury is associated with combustion processes, the burning of coal or municipal waste, and mining [77].

Elemental Hg is toxic to the central and peripheral nervous system, causing neurological and behavioural disorders. Symptoms include tremors, insomnia, memory loss, neuromuscular effects, headaches, and cognitive and motor dysfunction. The inhalation of mercury vapour can have harmful effects on the immune, nervous, and digestive systems, lungs, and kidneys and can be fatal [78, 79].

In aquatic environments inorganic mercury can be converted into an extremely toxic organic form – methylmercury (MeHg). MeHg accumulates in fish and shellfish and has the highest bioavailability compared to the other forms [80]. Its ability to penetrate cell membranes causes toxicity of the whole organism, specifically, for example, damage to the nervous system, the development of cardiovascular problems, or problems with the liver and kidneys. The hormonal system is affected even at low concentrations of this metal as a result of the inactivation of receptor sites and suppression of hormone synthesis. Methylmercury is able to cross the placenta and cause brain and nerve disorders in a developing foetus. The presence of mercury in plant tissues inhibits plant growth and affects the intensity of adsorption and accumulation of essential elements [81].

According to the IARC, methylmercury compounds are possibly carcinogenic to humans (Group 2B).

8.1.9 Zinc

Zinc (Zn) is an essential trace element. It is considered to be relatively non-toxic, particularly if taken orally [82]. Rather than zinc toxicity, zinc deficiency is observed [82]. However, manifestations of toxicity symptoms (nausea, vomiting, epigastric pain, lethargy, and fatigue) will occur with extremely high zinc intakes. Excessive zinc concentrations may lead to the deterioration of copper or iron metabolism [83].

8.2 Dioxins (PCDD/Fs) and other unintentionally produced POPs

Annex C of the Stockholm Convention lists seven unintentionally produced POPs: HCB, hexachlorobutadiene (HCBD), pentachlorobenzene (PeCB), PCBs, polychlorinated dibenzo-p-dioxins (PCDD), polychlorinated dibenzofurans (PCDF), and polychlorinated naphthalenes.

8.2.1 PCDD/Fs and dl-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 that can be divided into two groups according to their toxicological properties: 12 congeners exhibit toxicological properties similar to dioxins and 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) [16]. Technical mixtures of PCBs are characterized by six, sometimes seven indicator PCB congeners (i-PCBs). 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 [3]. These WHO TEFs were used to evaluate dioxin-like toxicity in the pooled samples of chicken eggs, soils, ash and other samples from Indonesia in this study.

Chlorinated dioxins (PCDD/Fs) are known to be extremely toxic. Numerous epidemiologic studies have revealed a variety of human health effects linked to chlorinated dioxin exposure including cardiovascular disease, diabetes, cancer, porphyria, endometriosis, early menopause, alteration of testosterone and thyroid hormones, and altered immune system response among others [84,85]. Laboratory animals given dioxins suffered a variety of effects, including an increase in birth defects and stillbirths. Fish exposed to these substances died shortly after the exposure ended. Food (particularly from animals) is the major source of exposure for humans [86].

Chlorinated dioxins became known to the public in the 1970s as a result of their contamination of Agent Orange, a defoliant pesticide mixture sprayed by the U.S. during the Vietnam War.[3] The production of 2,4,5 T pesticide as a basic ingredient for Agent Orange left one of the most seriously contaminated sites in Europe [87-89] and sick workers with many symptoms of exposure to the most toxic of dioxin congeners 2,3,7,8-TCDD [90,91].

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[3] According to estimates provided by the Government of Vietnam, 400,000 people were killed or maimed by the pesticide; 500,000 children were born with birth defects ranging from retardation to spina bifida; and an additional 2 million people have suffered cancers or other illnesses, which also can be related to dioxins as impurities in the Agent Orange mixture. It is estimated that in total, the equivalent of at least 366 kilograms of pure dioxin were dropped. York, G. and H. Mick. (2008, April 27, 2018). "Last ghost‘ of the Vietnam War." Retrieved 19-11-2018, 2018, from https://www.theglobeandmail.com/incoming/last-ghost-of-the-vietnam-war/article1057457/?page=all.

8.3 Legal standards

Table 16 *Legal standards for heavy metals in soils (mg/kg FW)*

Soil standards	As	Cd	Cu	Mo	Ni	Pb	Cr
Armenia[4]	2	-	3	-	4	32	6
France	37	20	190	-	-	400	-
Dutch	34	1.6	40	254	38	140	100
Czech Republic[5]	40	20	300	-	200	400	-
Levels of pollution limits – industrial areas (US EPA)[6]	2.4	800	41,000	5100	20,000	800	-
Levels of pollution limits – other areas (US EPA)[7]	0.61	70	31,000	390	1500	400	-

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[4] Order No. 01-N of 25 January 2010 of the Minister of Health of the Republic of Armenia "On Approving Sanitary Rules and Norms N 2.1.7.003-10 for Sanitary Requirements for Land Quality"

[5] Czech Decree No. 153/2016 issued by the Ministry of Agriculture.

[6] US EPA. Regional Screening Levels. [online] [cit. 2021-11-25]. Available at: http://www.epa.gov/reg3hwmd/risk/human/rb-concentration_table/Generic_Tables/docs/params_sl_table_run_JAN2015.pdf

[7] US EPA. Regional Screening Levels. [online] [cit. 2021-11-25]. Available at: http://www.epa.gov/reg3hwmd/risk/human/rb-concentration_table/Generic_Tables/docs/params_sl_table_run_JAN2015.pdf

8.4 Results of some samples

Table 17 *HM concentration in sediment samples (mg/kg dry weight, * - community with one sample)*

	As	Cd	Cu	Mo	Ni	Pb	Zn	Fe
				mg/kg DW				
Min	3.6	0.0	4.0	46	0.9	2.8	0.8	34
Max	108.0	13.9	72.8	59,148	719.0	60.0	327.0	4322
Mean	28.2	1.7	26.8	3633	112.2	15.3	40.4	681
Mean-ref	6.8	0.1	10.8	26	0.7	9.5	9.2	55
Alaverdi	32.2	0.7	29.7	12,688	206.4	11.5	26.5	430
khtala	42.9	4.0	25.6	3004	97.7	14.4	90.4	1628
eghut	7.4	0.2	20.7	928	117.0	12.6	5.6	135
Armanis	19.8	0.9	57.8	47	3.2	47.5	22.0	333
Chochkan	47.6	1.1	17.4	951	89.7	5.1	39.7	436
Mets Ayrum*	15.8	0.5	36.9	255	3.5	30.8	28.4	166

Table 18 *Concentration of HM in dust samples (µg/sample, * - community with one sample)*

	As	Cd	Cu	Mo	Ni	Pb	Zn	Fe
	µg/sample							
Min	1	0	10	0	4	5	25	1090
Max	42	4	652	8	67	269	1230	17,300
Mean	8	1	102	2	16	55	280	5285
Yagran (control)*	10	1	57	3	29	75	691	11,500
Alaverdi	5	1	57	3	10	38	284	4098
Akori	11	1	104	2	20	43	506	5375
Haghpat*	8	1	38	1	8	20	274	6330
Akhtala*	42	3	224	4	67	193	676	13,000
Shamlugh	8	1	55	3	11	81	907	5275
Teghut	5	0	184	4	14	29	224	6907
Shnogh	6	0	59	3	8	63	117	5205
Mets Ayrum	6	1	124	1	16	46	202	3855
Pokr Ayrum*	1	0	10	0	4	5	25	1360
Chochkan	8	1	105	2	20	79	129	7032
Mghart	14	2	101	5	23	97	522	7445
Armanis*	6	1	30	1	16	37	211	8250

Table 19 *Summary of HM concentrations in foodstuff samples (<LOQ - below limit of quantification)*

	Hg	As	Cd	Cu	Mo	Ni	Pb
	mg/kg fresh weight						
Min	<LOQ	<LOQ	<LOQ	0.15	<LOQ	<LOQ	<LOQ
Max	0.001	0.07	0.03	6.81	8.85	0.61	0.22
Mean	0.001	0.02	<LOQ	1.22	1.06	0.1	0.01

Table 20 *HM concentrations in homemade cow's-milk cheese samples (mg/kg) (<LOQ - below limit of quantification)*

	Sample ID	As	Cd	Cu	Mo	Ni	Pb	Hg
		mg/kg						
1	**MGHART-CHEESE-2/21**	<LOQ	<LOQ	0.2	0.1	0.7	<LOQ	<LOQ
2	**AKR-CHEESE-2/21**	<LOQ	<LOQ	0.5	<LOQ	0.4	<LOQ	<LOQ
3	**AKR-CHEESE-2021-1**	<LOQ	<LOQ	0.2	<LOQ	<LOQ	0.03	<LOQ
4	**MGH-CHEESE-2021-1**	<LOQ	<LOQ	0.2	0.1	0.3	<LOQ	<LOQ
5	**META-CHEESE-1/21**	<LOQ	<LOQ	0.2	<LOQ	<LOQ	<LOQ	<LOQ

9 References

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10 Photos

Alaverdi, 2010

1 Arsenic graveyard in Alaverdi
2 Interior of Alaverdi copper smelting plant
3 Arsenic graveyard in Alaverdi
4 Alaverdi copper smelting plant


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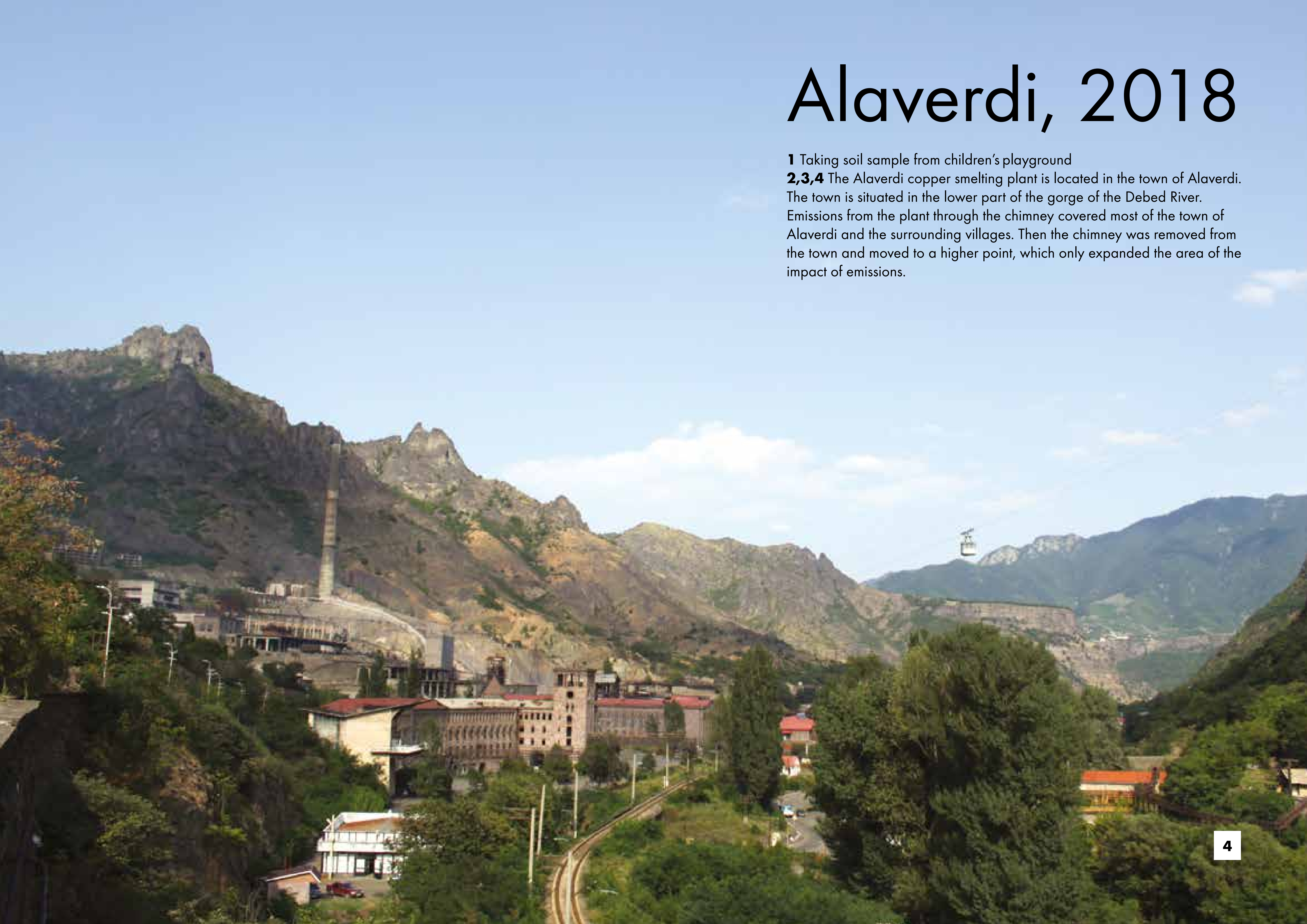


Alaverdi, 2018

1 Taking soil sample from children's playground
2,3,4 The Alaverdi copper smelting plant is located in the town of Alaverdi. The town is situated in the lower part of the gorge of the Debed River. Emissions from the plant through the chimney covered most of the town of Alaverdi and the surrounding villages. Then the chimney was removed from the town and moved to a higher point, which only expanded the area of the impact of emissions.

Sanahin

1 View of the Sanahin Monastery (behind the smoke from Alaverdi copper smelting plant)
2 Sanahin Monastery is founded in the 10th century in the Lori Province of Armenia, recognized as a UNESCO World Heritage Site.
3 Sampling in Sanahin village
4 Sanahin Monastery

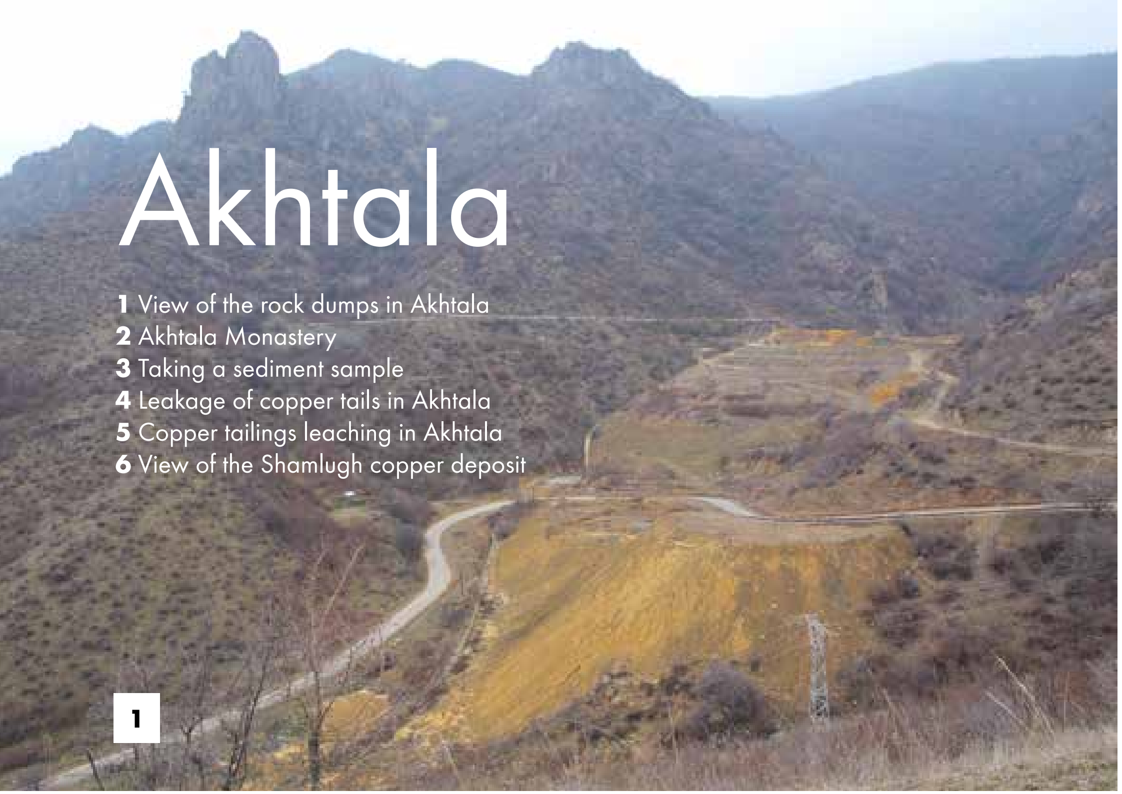
2

3

4

Akhtala

1 View of the rock dumps in Akhtala
2 Akhtala Monastery
3 Taking a sediment sample
4 Leakage of copper tails in Akhtala
5 Copper tailings leaching in Akhtala
6 View of the Shamlugh copper deposit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Mets Ayrum

1 Taking sediment samples from Naatak tailings pond
2 CCMS, EcoLur and Arnika team
3 Mining tailings
4 Naatak tailings pond in Mets Ayrum


2


3


4

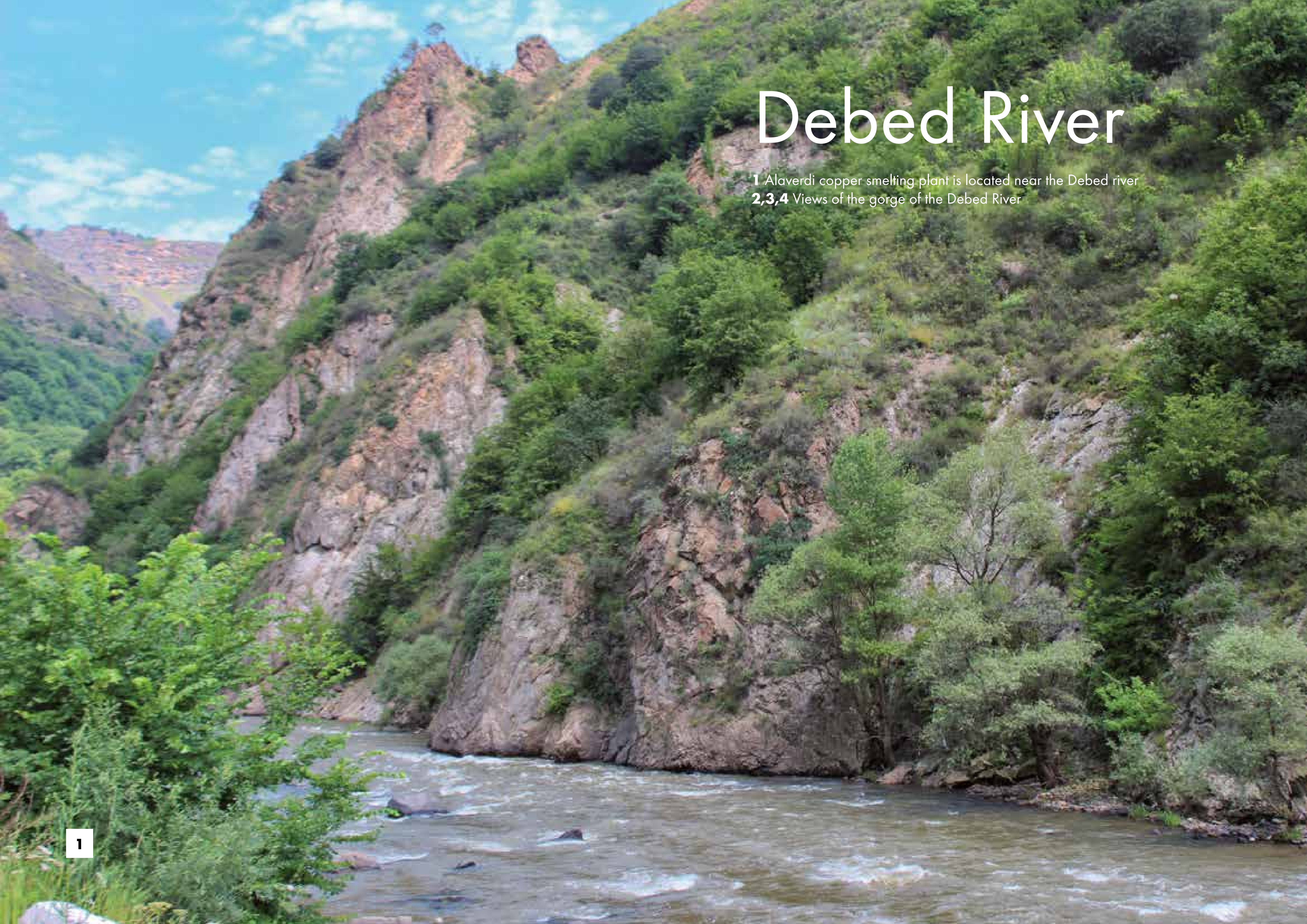
Teghut

1 Teghut tailings pond
2 River flowing from Teghut tailings pond

Akhtala River

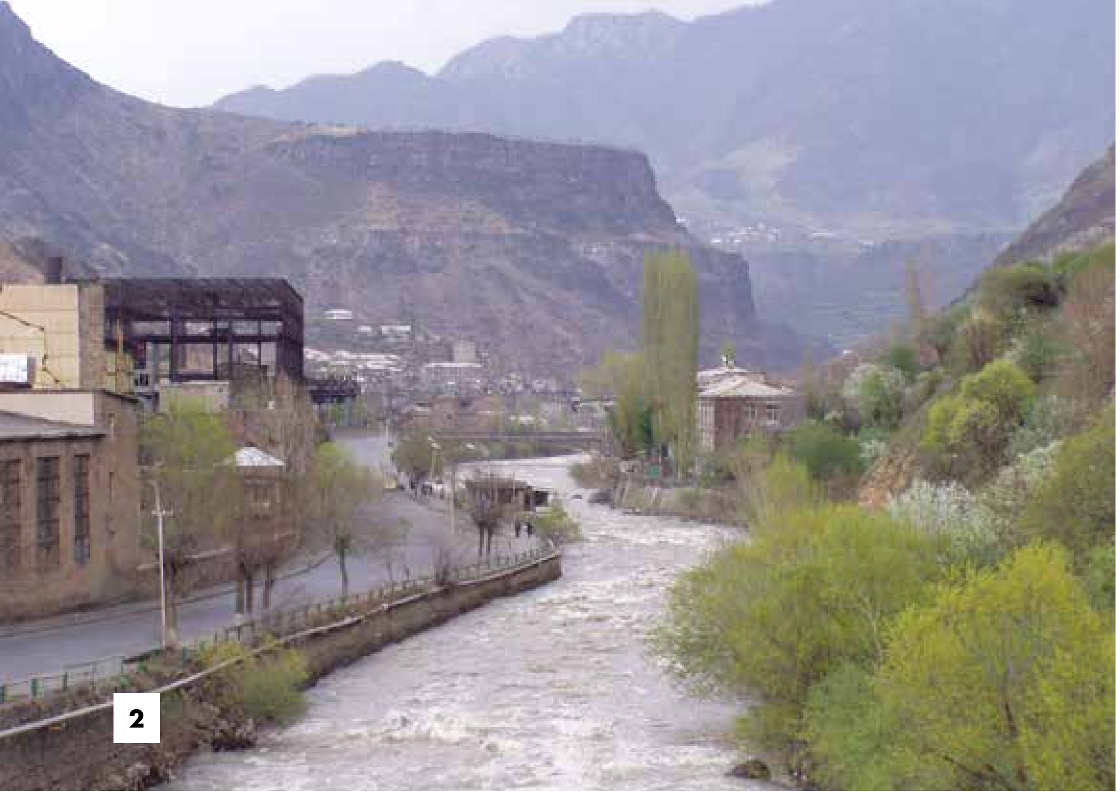
1 The Akhtala River flowing next to reclaimed tailings dam and ore waste piles in Akhtala village

Debed River



1

1 Alaverdi copper smelting plant is located near the Debed river
2,3,4 Views of the gorge of the Debed River



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Chqnagh River

1 The Chqnagh River flows near the ore mining residue dump
2 The ore mining residue dump

This report was prepared and published as a part of the project “Raising awareness on pollution in the Tumanyan region”, with the financial assistance of the Ministry of Foreign Affairs of the Czech Republic under the Transformation Cooperation Programme. The production of this publication was also made possible thanks to the Global Greengrants Fund.

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