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Lead in residential soil and dust in a mining and smelting district in northern Armenia: a pilot study^{☆, ☆☆}

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Abstract

This pilot study of sources of lead exposure in residential settings was conducted in a mining and smelting district in northern Armenia. Samples of exterior soil and dust and interior house dust were collected in and around apartment buildings in Alaverdi where the country's largest polymetallic smelter is located, and in nearby mining towns of Aghtala and Shamlugh. The NITON XL-723 Multi-Element XRF analyzer was used for lead testing. Lead levels in samples from Alaverdi were higher than those in Shamlugh and Aghtala. In all three towns, the highest lead levels were found in loose exterior dust samples, and lead concentrations in yard soil were higher than those in garden soil. Many soil samples (34%) and the majority of loose dust samples (77%) in Alaverdi exceeded the US Environmental Protection Agency standard of 400 mg/kg for bare soil in children's play areas. In addition, 36% of floor dust samples from apartments in Alaverdi exceeded the US Environmental Protection Agency standard of 40 µg/ft² for lead loading in residential floor dust. The Armenian Ministry of Health and other interested agencies are being informed about the findings of the study so that they can consider and develop educational and preventive programs including blood lead screening among sensitive populations.

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1. Introduction

Lead poisoning is an environmental health problem worldwide, especially for young children (Silbergeld,

1995). Lead has been used in Armenia since ancient times. It was historically used in construction, water pipes, and ceramic manufacturing. Lead is also mentioned in ancient and medieval manuscripts as an ingredient in medicine and paint (The Armenian Soviet Encyclopedia, 1979).

Armenia, a country with a population of about three million and an area of 29,800 km², is located in the southern Caucasus Mountains (Fig. 1). Armenia was one of the most industrialized republics of the former Soviet Union (USSR). Its diverse economy of more than 2000 industries (including metallurgical and chemical industries, mining and ore reprocessing, machine building, electronics, wood reprocessing, and construction materials and crystal production) comprised 1.2% of total USSR industrial output, even though its land mass

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Fig. 1. Map of Armenia.

covered only 0.13% of the territory of the USSR (TACIS, 1998). Among those industries were several that used lead in production and emitted it into the environment (e.g., metallurgy, machine building, lead-acid battery production, electronics, and crystal manufacturing) (Sarian, 1989).

In Armenia, polymetallic ores have been mined and smelted in the northern (Lori region) and southern regions (near the industrial town of Kapan) (The Armenian Soviet Encyclopedia, 1979). The metals extracted have included copper, molybdenum, gold, zinc, mercury, chromium, arsenic, cadmium, and silver (Kurkjian, 2000). Lead has not been refined in Armenia; it has, however, been emitted into the environment during mining and smelting operations because it is naturally present in high concentrations in ores. During the Soviet era, data on metal production levels were classified as state secrets and thus remain generally unavailable today.

The biggest polymetallic smelter in Armenia is located in Alaverdi in the Lori region. It produces mainly copper; lead is a byproduct of smelting and extracting operations. The smelter was shutdown in the late 1980s, when the shift to a free market economy and the devastating earthquake of 1988 led to an 85% decline in

total industrial production in Armenia (TACIS, 1998). This smelter, which was restarted in the late 1990s, currently operates at about 30% of full capacity and continues to increase its production level (Armenian Bureau of Statistics, 2001). In addition to past contamination, the lack of modern environmental control systems may further increase environmental contamination as the industry continues to recover (Babayan et al., 1998a, 1999). No surveys of the potential public health impact of mining and smelting operations in Armenia have been reported. The purpose of this initial environmental study was to assess the lead content of dust and soil in residential areas near the mining and smelting operations in the mining district of northern Armenia.

2. Materials and methods

2.1. Study areas

The study was conducted in three towns in the Lori region of northern Armenia—Alaverdi, Aghtala, and Shamlugh. These towns are located about 200 km north of Yerevan in the Caucasus mountains (at an average

elevation of about 2000 m above sea level). The town of Alaverdi (population of $\sim 20,000$) has the largest polymetallic smelter in Armenia. The town is situated along a narrow canyon, with residential areas located on both sides of the canyon and separated by the Debet River. The smelter is located at the bottom of the canyon in the middle of the town (Fig. 2). The smelter smokestack is about 20 m high and does not clear the top of the canyon; thus the emissions from the stack dissipate for the most part along the canyon. The predominant wind direction is along the canyon to the East in the morning and to the West in the late afternoon. Aghtala (population of ~ 3500) is located 30 km northeast of Alaverdi. Shamlugh (population ~ 1000) is located about 10 km north of Aghtala. Both towns are located in the hills within a 1 km radius of numerous abandoned and active open polymetallic mines. The latter two towns were built during the Soviet era as residential areas for miners.

Most of the dwellings in all three towns are five-storied apartment buildings with one to four communal entrances. Each entrance serves 10 apartments (2 apartments per story). The area in front of the entrances is typically paved with asphalt or cement. Most of the buildings have yards with bare soil; some also have vegetable gardens.

2.2. Sampling procedures

Samples of soil, exterior dust, and interior dust were collected in and around 30 apartment buildings in

Alaverdi. Buildings were selected from each of the five residential sectors of the town, determined by their locations in relation to the smelter (Fig. 2). Sector 1 was located on the top of the canyon across the river and southwest of the smelter. Sector 2 was located about 1 km west of the smelter on the same side of the canyon. Sector 3 was located about 20 m west of the smelter. Sector 4 was located across the river and about 500 m south of the smelter. Last, Sector 5 was located about 1 km east of the smelter on the same side of the canyon as the smelter. Environmental sampling was conducted in and around randomly selected buildings as follows: Sector 1, 7 buildings; Sector 2, 7 buildings; Sector 3, 3 buildings; Sector 4, 5 buildings; and Sector 5, 8 buildings. The number of buildings sampled in each sector was related to the number of buildings in each residential sector. The exact number of apartment buildings in Alaverdi was not available. We estimated that there were as many as 150 apartment buildings in Alaverdi.

In Aghtala, sampling was conducted in and around 12 (34%) of the total approximately 35 buildings as follows: every other building on both sides of the main street of the town's major residential area ($n = 8$ buildings), and 4 buildings in the Svinetz "lead" neighborhood located on the hillside next to old mine tailings. In Shamlugh, every other apartment building located around a town square was sampled ($n = 8$ buildings of a total of 13 buildings (62%)).

The sample collection was conducted in July of 2001. Exterior samples were collected near each apartment

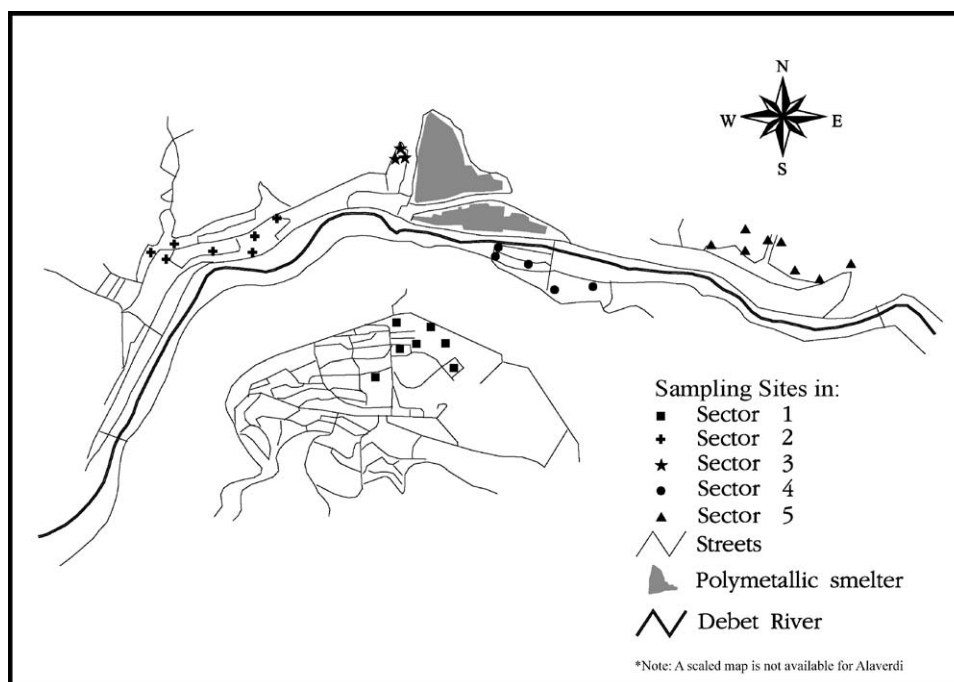


Fig. 2. Sampling sites by sector in Alaverdi in relation to the smelter.

building as follows: (a) one composite soil sample from the yard, one from the garden, and one from the playground (if present); (b) one composite loose dust sample from the front of a randomly selected communal entrance to the building; and (c) one floor wipe dust sample just inside that communal entrance. Also, in each building interior dust wipe samples were collected from two randomly selected apartments on the first floor. First-floor apartments were selected because they were closest to the communal entrance. Dust wipe samples were collected from three locations in each apartment as follows: the interior entryway floor; the center of the living room floor; and the kitchen windowsill. If there was no kitchen windowsill, another window of the same apartment that was frequently opened (as reported by the owner) was sampled. Prior to interior sampling, permission was obtained from an adult occupant of each apartment.

Composite soil samples were collected from the top 2 cm of soil using a reusable metallic scoop. Composite samples were collected from 2 to 10 randomly selected locations depending on the size of the yard or garden (range 20–200 m²). If present, grass and other vegetation were removed from each subsample. The subsamples of soils were combined in an 18 × 20-cm reclosable polypropylene bag. Composite samples of loose dust were collected from exterior asphalt and cement surfaces in front of the communal entrances using a reusable metallic scoop. Subsamples were collected from 1 to 3 random locations, depending on the size of the paved surface, and placed in resealable bags. The metallic scoop was cleaned with a disposable wipe after each use to minimize cross-contamination of samples. Wipe dust samples were collected using Palintest towelettes according to a standard house dust wipe sampling procedure (HUD (U.S. Department of Housing and Urban Development), 2001a). Floor dust wipe samples were collected from a 0.0929-m² (1-ft²) area using reusable templates. The dimensions of each sampled windowsill were measured and recorded. After sampling, each wipe was folded and placed in a 5 × 5-cm plastic bag that was sealed with tape and placed in an 18 × 20-cm reclosable polypropylene bag. A total of 73 soil samples, 47 loose dust samples, and 325 dust wipe samples were collected. Three of the soil samples were collected from kindergarten playgrounds and included in the analysis of the yard soil data.

2.3. Sample preparation and analysis

Soil and loose exterior dust samples were prepared at the hydro-geochemical laboratory of the Institute of Geosciences, the National Academy of Sciences of Armenia, Yerevan. The samples were dried at 30°C, homogenized, crushed with a mortar and pestle, and sieved in order to provide a uniform fine-grained

powder for analysis. Wipe dust samples did not require sample preparation prior to analysis.

Samples were analyzed at the Environmental Conservation and Research Center, American University of Armenia, using a portable NITON XL-723 multi-element x-ray fluorescent (XRF) analyzer. This instrument provided simultaneous analysis of 15 metals when the isotope source was ¹⁰⁹Cd, i.e., Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Pb, Hg, Rb, Sr, Zr, and Mo. The instrument's bulk-sample mode was used for the analysis of soil and loose exterior dust samples, and the results were reported as element-specific concentrations (mg/kg). The instrument's thin-sample mode was used for the analysis of dust wipe samples, and the results were reported as loadings (μg/ft²) (NITON, 1998). Soil and loose dust samples were analyzed in 18 analytical runs. Wipe dust samples were analyzed in 40 analytical runs.

Since the XRF method does not destroy the sample, it was possible to compare the wipe lead loading values based on the NITON XRF instrument with those based on other methods commonly used in the USA. For that purpose, 83 wipe dust samples already analyzed by XRF were leached using a modified US Environmental Protection Agency (USEPA) 3050 method for nitric acid hotplate digestion (Orlova et al., 1999) and analyzed at the Trace Metals Laboratory of the Kennedy Krieger Research Institute in Baltimore, MD (USA) using a Perkin Elmer Optima 2000 inductively coupled plasma optical emission spectrometer (ICP-OES) and US Environmental Protection Agency 6070 method (U.S. Environmental Protection Agency, 1986a, b). This laboratory has a well-established performance record with the US Environmental Lead Proficiency Accreditation Testing (ELPAT) Program (Grunder, 2002) and the US National Lead Laboratories Accreditation Program (Schlecht et al., 1996).

2.4. Quality control/quality assurance

This study included multiple laboratory quality control activities. When measuring soil and loose dust, three standard reference materials (SRM) provided by the NITON Corp. (Billerica, MA, USA) were analyzed once per day as follows (for a total of 4 days): National Institute of Standards (NIST) (USA) SRM 2710 (Montana Soil, 5532 mg/kg lead); NIST SRM 2711 (Montana Soil, 1162 mg/kg lead); and NIST SRM 2709 (18.9 mg/kg lead). After the analysis of each batch of 10 soil or loose dust samples, the instrument was reset for self-calibration and NIST SRM 2711 was analyzed in order to monitor for variation in the instrument's response. The SRM 2711 was chosen because it best reflected the range of measured soil lead concentrations. The reproducibility of the NIST SRM 2711 was ±2% SD of the mean with a mean recovery of 76% [range 71–

80%; 95% confidence interval (CI) for the mean from 72% to 80%]. Based on the mean percentage recovery of NIST SRM 2711, a correction factor of 1.31 was calculated and applied to the instrument-reported lead concentration for each sample.

For wipe dust samples analysis, the wipe standard provided by the manufacturer (range of 450–620 µg of lead) was analyzed after the instrument self-calibration was performed. After the analysis of a group of fifteen wipes, the sample instrument was reset for self-calibration, and the standard was reanalyzed prior to continuing further analysis. The mean XRF reading on the wipe standard of 606 µg lead ($n = 32$, range 581–619 µg of lead per sample; 95% CI of 586–625 µg of lead per sample) was within the range reported by the manufacturer.

The limit of detection for the NITON 700-series for lead in soil is 20 mg/kg and for dust wipe samples is 12 µg of lead per sample (NITON, 1999; D. Schatzlein, NITON Corp., personal communication). Eight of the eleven method blank wipe samples included in analytical runs had measured lead levels below the limit of detection, and three samples had lead levels slightly above the limit of detection (max = 16 µg of lead per sample).

2.5. Data management

Statistical analysis was performed using SAS software (Version 8) (SAS, 1999–2001). The Tukey–Kramer Test within the general linear model procedure (multiple group comparisons) was used to test for significant

differences between groups (e.g., towns or residential sectors in Alaverdi) of unequal sizes (Kirk, 1995).

3. Results

3.1. Soil

Soil lead concentrations by sample type and by study area are presented in Table 1. Alaverdi soil lead concentrations (geometric mean (GM) and maximum) were higher than those measured in Aghtala and Shamlugh. The results from Aghtala and Shamlugh were not statistically significantly different from each other. The soil lead concentrations in Shamlugh but not in Aghtala were statistically significantly different from the Alaverdi results. When combined, however, the results from Shamlugh and Aghtala were statistically significantly different from the Alaverdi results for both garden soil and yard soil lead concentrations.

Statistically significant differences were not detected in soil lead concentrations across the five sectors in Alaverdi. This could be due to very small sample sizes in each sector. However, lead concentrations tended to be higher in sectors close to the smelter. Lead concentrations of 1136 and 1076 mg/kg were found in the yard and garden soils, respectively, of the apartment buildings located within 20 m of the smelter in Sector 3 (Table 1). The sector farthest from the smelter (Sector 1) had the lowest GM lead concentration for yard soil (GM = 127 mg/kg) and garden soil (GM = 105 mg/kg) (Table 1). Three soil samples collected from kindergarten

Table 1
Soil lead concentrations (mg/kg) by study area and by sample type

Study area—sector	Sample type	Number of samples	Mean	SD	Geo ^a mean	Geo SD	Min	Max
Aghtala	Garden soil	12	139	97	113	0.68	48	369
	Yard soil	9	234	98	220	0.35	141	474
Shamlugh	Garden soil	5	77	56	64	0.66	27	173
	Yard soil	9	106	25	103	0.25	68	138
Aghtala and Shamlugh (combined)	Garden soil	17	121	90	95	0.70	27	369
	Yard soil	18	170	96	151	0.49	68	474
Alaverdi—Sector 1	Garden soil	6	139	98	105	0.87	37	274
	Yard soil	5	148	83	127	0.67	45	266
Alaverdi—Sector 2	Garden soil	2	331	316	244	1.16	108	554
	Yard soil	5	600	473	436	0.99	96	1334
Alaverdi—Sector 3	Garden soil	2	640	616	469	1.17	204	1076
	Yard soil	2	748	548	639	0.81	360	1136
Alaverdi—Sector 4	Garden soil	6	293	129	270	0.45	146	502
	Yard soil	1	523	—	523	—	523	523
Alaverdi—Sector 5	Garden soil	4	408	240	319	0.95	80	597
	Yard soil	5	621	300	548	0.60	224	938
Alaverdi—Total	Garden soil	20	308	253	220	0.90	37	1076
	Yard soil	18	493	379	348	0.94	45	1334

^aGeo, geometric.

Table 2
Exterior loose dust lead concentrations (mg/kg) by study area and sector

Study area—sector	Number of samples	Mean	SD	Geo ^a mean	Geo SD	Min	Max
Aghtala	12	318	150	288	0.47	105	700
Shamlugh	5	182	71	172	0.34	127	305
Aghtala and Shamlugh (combined)	17	278	144	248	0.49	105	700
Alaverdi—Sector 1	7	512	466	391	0.75	164	1504
Alaverdi—Sector 2	7	936	769	774	0.60	390	2642
Alaverdi—Sector 3	3	1021	526	916	0.60	470	1518
Alaverdi—Sector 4	5	407	141	388	0.34	243	623
Alaverdi—Sector 5	8	986	629	869	0.50	507	2445
Alaverdi—Total	30	771	591	617	0.66	164	2642

^aGeo, geometric.

Table 3
Wipe dust lead loadings ($\mu\text{g}/\text{ft}^2$) in building communal entrances by study area and sector

Study area—sector	Number of samples	Mean	SD	Geo ^a mean	Geo SD	Min	Max
Aghtala	8	139	79	124	0.50	54	318
Shamlugh	8	40	12	39	0.28	28	63
Alaverdi—Sector 1	7	89	53	75	0.66	30	182
Alaverdi—Sector 2	7	114	19	112	0.18	80	134
Alaverdi—Sector 3	3	164	45	160	0.28	119	208
Alaverdi—Sector 4	5	82	47	74	0.48	45	163
Alaverdi—Sector 5	8	240	158	197	0.70	59	554
Alaverdi—Total	30	142	107	115	0.65	30	554

^aGeo, geometric.

playgrounds in Sectors 4 and 5 of Alaverdi and in Shamlugh had lead concentrations of 523, 938, and 99 mg/kg, respectively.

3.2. Exterior dust

Lead concentrations in exterior loose dust samples are presented in Table 2. Lead loadings in wipe dust samples collected just inside communal entrances to the apartment buildings are presented in Table 3. A statistically significant correlation ($r = 0.60$) was found between the exterior loose dust lead concentrations and communal entrance wipe dust lead loadings for all three towns. The loose dust lead concentrations in Alaverdi ($\text{GM} = 617 \text{ mg/kg}$) were higher than in Aghtala ($\text{GM} = 288 \text{ mg/kg}$) and Shamlugh ($\text{GM} = 172 \text{ mg/kg}$) (Table 2). The GM entrance dust lead loadings in Alaverdi and Aghtala were similar ($\text{GM} = 115$ and $124 \mu\text{g}/\text{ft}^2$, respectively). The lead levels in loose dust and wipe dust samples in Shamlugh ($\text{GM} = 172$ and $39 \mu\text{g}/\text{ft}^2$, respectively) were lower than those found in Alaverdi and Aghtala.

The loose dust results from Aghtala and Shamlugh were not statistically significantly different, but both were statistically significantly different from the Alaverdi results. The statistical significance persisted

when combined loose dust results from Aghtala and Shamlugh were compared to the results from Alaverdi. There was no statistically significant difference between communal entrance wipe dust lead loadings from Aghtala and Alaverdi. The communal entrance wipe dust loadings from Shamlugh were statistically significantly different from the Aghtala and Alaverdi results.

The multiple-group comparisons did not detect statistically significant differences in loose dust lead concentrations across the five residential sectors of Alaverdi. This could be because of very small sample sizes in each sector. However, the pattern of lead concentrations in loose dust in the five residential sectors of Alaverdi was similar to the pattern found for the garden and yard soil lead concentrations. The highest loose dust lead concentration ($\text{GM} = 916 \text{ mg/kg}$) was found in Sector 3, that closest to the smelter, and the lead concentrations in loose dust samples in Sectors 2 and 5 ($\text{GM} = 774$ and 869 mg/kg , respectively) were higher than those in Sectors 1 and 4 (Table 2).

In Alaverdi, the highest communal entrance wipe dust lead loadings were found in Sector 5 ($\text{GM} = 197 \mu\text{g}/\text{ft}^2$). The only statistically significant differences found were between the entrance dust lead loadings in Sector 5 and those in Sectors 1 and 4, where the levels were much lower (Table 3).

3.3. Interior house dust lead loadings

The interior dust lead loadings by surface, room, town, and sector are presented in Table 4. The GM dust lead loadings on all surface types and locations were slightly higher in Alaverdi than in the other two towns (Table 4).

The hallway floor dust lead loadings across three towns were not statistically significantly different; however, the combined hallway floor dust lead loadings in Aghtala and Shamlugh (GM = 33 µg/ft²) were statistically significantly lower than the hallway floor dust lead loadings in Alaverdi (GM = 44 µg/ft²). The living room floor dust lead loadings in Shamlugh were not statistically significantly different from those in Alaverdi and Aghtala. The living room dust lead loadings in Aghtala were statistically significantly different from the living room lead loadings in Alaverdi. The statistical significance persisted when combined living room floor dust lead loadings in Aghtala and Shamlugh (GM = 22 µg/ft²) were compared with the living room floor dust lead loadings in Alaverdi (GM = 34 µg/ft²). The kitchen windowsill dust lead loadings had the same pattern of statistically significant differences as the living room dust lead loadings for all three towns. No

statistically significant differences were found between lead loadings on hallway floors, living room floors, and kitchen windowsills in Alaverdi and Shamlugh. Only in Aghtala, hallway floor lead loadings were statistically significantly higher than living room floor lead loadings. However, when the data from the three towns were pooled, lead loadings on hallway and living room floors and kitchen windowsills were not statistically significantly different from each other.

No statistically significant differences were found between the interior dust lead loadings across the five sectors of Alaverdi. The GM hallway floor dust lead loadings ranged from 31 µg/ft² in Sector 1 to 53 µg/ft² in both Sectors 3 and 5. The GM living room floor dust lead loadings ranged from 24 µg/ft² in Sector 1 to 52 µg/ft² in Sector 3. The GM kitchen windowsill lead loadings ranged from 33 µg/ft² in Sector 4 to 55 µg/ft² in Sector 3.

3.4. XRF versus ICP-OES

The estimates of wipe dust lead loadings obtained by NITON XRF were highly correlated with those obtained by ICP-OES ($r = 0.95, P < 0.0001$). Additionally, the XRF dust lead loading results were statistically

Table 4
Interior dust lead loadings (µg/ft²) by study area, sector, and surface and room type

Study area—Sector	Surface and room type	Number of samples	Mean	SD	Geo ^a mean	Geo SD	Min	Max
Aghtala	Apartment hallway floor	20	41	39	32	0.65	17	152
	Living room floor	20	21	6	21	0.25	15	41
	Kitchen windowsill	19	26	13	24	0.43	13	60
Shamlugh	Apartment hallway floor	15	43	35	35	0.57	17	155
	Living room floor	16	32	30	25	0.71	<LOD ^b	125
	Kitchen windowsill	15	109	302	36	1.05	17	1198
Aghtala and Shamlugh (combined)	Apartment hallway floor	35	42	37	33	0.61	17	155
	Living room floor	36	26	21	22	0.51	<LOD ^b	125
	Kitchen windowsill	34	63	201	28	0.79	13	1198
Alaverdi—Sector 1	Apartment hallway floor	13	33	23	31	0.36	15	67
	Living room floor	14	26	9	24	0.30	18	52
	Kitchen windowsill	12	52	60	37	0.74	17	233
Alaverdi—Sector 2	Apartment hallway floor	14	49	24	44	0.49	21	86
	Living room floor	14	55	42	43	0.71	17	146
	Kitchen windowsill	13	66	58	48	0.81	16	217
Alaverdi—Sector 3	Apartment hallway floor	6	56	19	53	0.32	39	89
	Living room floor	6	80	88	52	0.97	25	242
	Kitchen Windowsill	6	64	40	55	0.64	24	134
Alaverdi—Sector 4	Apartment hallway floor	8	53	39	44	0.61	24	140
	Living room floor	11	46	40	38	0.56	22	160
	Kitchen windowsill	9	37	18	33	0.47	16	72
Alaverdi—Sector 5	Apartment hallway floor	15	73	76	53	0.76	21	299
	Living room floor	16	60	132	30	0.87	<LOD ^b	552
	Kitchen windowsill	15	47	33	38	0.67	16	121
Alaverdi—Total	Apartment hallway floor	56	53	46	44	0.57	15	299
	Living room floor	61	50	77	34	0.72	<LOD ^b	552
	Kitchen windowsill	55	53	46	41	0.69	16	233

^a Geo, geometric.

^b <LOD, below the limit of detection.

Table 5
Metals' Concentrations (mg/kg) in garden and yard soils, and exterior loose dust by study area

Metal	Alaverdi						Aghtala and Shamlugh					
	Garden soil		Yard soil		Loose dust		Garden soil		Yard soil		Loose dust	
	$n(N)^a$	Range (mg/kg)	$n(N)$	Range (mg/kg)	$n(N)$	Range (mg/kg)	$n(N)$	Range (mg/kg)	$n(N)$	Range (mg/kg)	$n(N)$	Range (mg/kg)
As	16 (20)	17–184	17 (18)	17–448	26 (30)	25–490	7 (17)	18–32	11 (18)	20–78	12 (17)	21–136
Cr total	0 (20)	—	1 (18)	352	3 (30)	324–423	0 (17)	—	0 (18)	—	0 (17)	—
Co	3 (20)	292–392	6 (18)	275–828	8 (30)	305–636	1 (17)	277	3 (18)	272–352	6 (17)	270–475
Cu	18 (20)	201–3648	18 (18)	64–5930	30 (30)	147–6989	15 (17)	65–514	17 (18)	64–485	17 (17)	94–406
Fe	20 (20)	9357–48282	18 (18)	22195–82688	30 (30)	25088–100966	17 (17)	23795–34790	18 (18)	24588–46797	17 (17)	24588–51994
Hg	0 (20)	—	0 (18)	—	0 (30)	—	0 (17)	—	0 (18)	—	0 (17)	—
Mn	19 (20)	537–3299	17 (18)	592–1440	24 (30)	590–1779	16 (17)	482–1460	16 (18)	565–1030	12 (17)	534–1190
Mo	15 (20)	14–38	16 (18)	14–183	28 (30)	15–219	12 (17)	13–22	15 (18)	16–63	15 (17)	15–70
Ni	0 (20)	—	2 (18)	157–158	6 (30)	170–337	1 (17)	160	0 (18)	—	0 (17)	—
Rb	8 (20)	11–24	7 (18)	10–27	3 (30)	13–18	16 (17)	10–36	11 (18)	8–34	9 (17)	16–45
Se	3 (20)	10–12	4 (18)	12–31	8 (30)	11–30	0 (17)	—	0 (18)	—	0 (17)	—
Sr	20 (20)	91–579	18 (18)	94–292	30 (30)	125–280	17 (17)	74–247	18 (18)	77–256	17 (17)	23–370
Zn	20 (20)	75–1160	18 (18)	95–1450	30 (30)	221–3709	17 (17)	109–822	18 (18)	192–573	17 (17)	391–1070
Zr	20 (20)	26–77	18 (18)	47–73	30 (30)	34–73	17 (17)	40–89	18 (18)	42–87	17 (17)	46–84

^a n , the number of samples with detectable levels of metals; N , the total number of samples collected.

significantly higher (on average 2.6 times higher) than those measured by ICP-OES (paired t test, $P < 0.0001$).

3.5. Multiple metals in soil and loose dust samples

All soil and loose dust samples were simultaneously analyzed for 14 other metals (i.e., Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Hg, Rb, Sr, Zr, and Mo) (Table 5). The Eikmann–Kloke (E-K) list (Germany) of soil remediation criteria for heavy metals (Visser, 1994) is more comprehensive than the US or Armenian lists and was used as a point of reference for this study. It covers the following metals: As, Cr, Co, Cu, Me, Mo, Ni, Se, and Zn.

Nine of 20 (45%) garden soil samples, 9 of 18 (50%) yard soil samples, and 21 of 30 (70%) loose dust samples in Alaverdi had arsenic levels above 80 mg/kg, the E-K soil remediation criterion. Aghtala and Shamlugh data indicated that only one sample collected in Shamlugh had an arsenic concentration above 80 mg/kg (Table 5). Ten percent of garden soil samples and 22% of yard soil samples in Alaverdi exceeded the 10 mg/kg E-K criterion for selenium. Twenty-five and 35% of garden soil samples in Aghtala and Alaverdi, respectively, and 61% of yard soil samples in Alaverdi were above the 600 mg/kg remediation criterion for zinc in garden soil. The zinc concentration in exterior loose dust samples was above 600 mg/kg in 25% of the samples in Aghtala ($n = 12$), 100% of those in Shamlugh ($n = 5$), and 80% of those in Alaverdi ($n = 30$). Ninety percent of garden soil samples, 78% of yard soil samples, and 93% of loose dust samples in Alaverdi had copper levels above 200 mg/kg, the E-K garden soil remediation criterion for copper. Forty-two percent of garden soil samples ($n = 12$), 78% of yard soil samples ($n = 9$), and 83% of loose dust samples in Aghtala ($n = 12$) had copper concentrations above 200 mg/kg. In Shamlugh, 11% of yard soil samples ($n = 9$) and all loose dust samples ($n = 5$) were above the criterion of 200 mg/kg. Thirteen percent of loose dust samples in Alaverdi had nickel concentrations above 200 mg/kg, the E-K criterion for garden soils. Neither the garden nor the yard soil samples nor the loose dust samples from the three study towns had detectable concentrations of mercury.

4. Discussion

It is important to emphasize that the findings of this study are based on relatively small sample sizes in each town and particularly in each residential sector. Despite the consistency of results within and across towns for various sample types and endpoints, there is a need to confirm these findings with larger sample sizes.

4.1. Lead in soil

Soil lead concentrations were higher in the smelter town of Alaverdi than in Shamlugh and Aghtala (located near the open-pit mines). Our study suggests that smelter operations were associated with more lead contamination than were the mining operations. In Alaverdi, higher lead concentrations were found in the residential sector closest to the smelter (Sector 3) (Fig. 2). The lowest lead concentrations were found in Sector 1, located across the river, on top of the canyon at an elevation above that of the opening of the smelter smoke stack. Elevated lead concentrations and loadings in Sector 5 in comparison with those in Sectors 2 and 4, located about the same distance from the smelter, could be due to the predominantly northeast wind direction that moves the stack emissions toward Sector 5.

The GM lead concentrations were higher in yard soil than in garden soil in each town. This could be due to the tilling of garden topsoil and the common practice of adding to gardens fertile soils imported from other areas.

All soil samples collected in this survey exceeded the Armenian soil lead standard of 20 mg/kg (Babayan et al., 1998a) (Table 1). Armenian environmental standards, including the soil lead standard, were registered at the Management Administration of Standardization, Metrology, and Certification Committee in Moscow and reportedly were based on political motives and without any risk assessment (Babayan et al., 1998a). This may explain why the Armenian environmental standards were often unrealistically strict and hard to achieve. Unfortunately, these standards have not been revisited or revised. The Armenian soil lead standard of 20 mg/kg is an example of a current standard that is an order of magnitude below those accepted internationally. For example, the US Environmental Protection Agency action level for lead in bare soil in children's play areas is 400 mg/kg (US Environmental Protection Agency, 2001). The GM yard soil lead concentrations in Alaverdi were above 400 mg/kg in each residential sector except for Sector 1. Five of 20 (25%) garden soil samples and 8 of 18 (44%) yard soil samples in Alaverdi were above the US Environmental Protection Agency action level for bare residential soil. Soil lead concentrations from kindergarten playgrounds in Sectors 4 and 5 of Alaverdi were 523 and 938 mg/kg, respectively. Only one yard soil sample in Aghtala exceeded this action level. None of the soil samples in Shamlugh and Sector 1 in Alaverdi was above 400 mg/kg. Our findings indicate that soil is a potential source of lead exposure to children in Alaverdi because our soil samples were collected from bare yards that were, in effect, children's playgrounds.

4.2. Lead in exterior dust

Lead concentrations in exterior loose dust samples collected in front of apartment buildings were higher than lead concentrations in soil samples in each town (Tables 1 and 2). This could be related to the fact that the loose dust samples were collected from asphalt or cement surfaces with ~2–3 mm-thick loose dust layers, whereas the soil samples were collected from the top 2 cm of soil—lead concentrations may be lower at increasing depth.

Lead concentrations in exterior loose dust were much higher in Alaverdi than in Shamlugh and Aghtala. In Alaverdi, the highest lead concentrations were found in Sector 3, that closest to the smelter, and in Sectors 2 and 5, those located downwind of the smelter. The similarity in the patterns of lead levels in exterior loose dust and soil in Alaverdi (Tables 1 and 2) suggests that lead in soil and loose dust might be derived from the same source, most likely from the smelter emissions.

There are no standards for lead in exterior loose dust in Armenia, the USA, or internationally. Therefore, we could only relate the lead concentrations in loose dust to existing soil lead standards. The majority (77%) of exterior loose dust samples in Alaverdi (total $n = 30$) exceeded the US action level of 400 mg/kg lead for bare soil. Two of 12 (17%) samples in Aghtala and none of the samples in Shamlugh exceeded the standard. Given that residential exterior dust has been shown to be one of the sources of children's exposure to lead (Lanphear et al., 1998; TerraGraphics, 2000) and that, in general, children spend many hours outside, exterior loose dust represents a potentially important source of lead exposure for children in Alaverdi and Aghtala.

4.3. Lead in interior dust

The interior floor dust lead loadings were higher in Alaverdi than in Aghtala or Shamlugh (Table 4). Only 7 of 35 (20%) hallway floor samples and 4 of 36 (11%) living room floor samples in Aghtala and Shamlugh had lead loadings above the US Environmental Protection Agency standard for lead in residential floor dust of 40 $\mu\text{g}/\text{ft}^2$. Twenty-five of 56 (45%) hallway floor samples and 17 of 61 (28%) living room floor samples in Alaverdi had lead loadings above 40 $\mu\text{g}/\text{ft}^2$. The highest interior floor lead loadings (GM = 53 $\mu\text{g}/\text{ft}^2$ for the hallway and GM = 52 $\mu\text{g}/\text{ft}^2$ for the living room) were found in Sector 3 of Alaverdi, the residential area closest to the smelter (Table 4). None of the floor samples in Sector 1 (farthest from the smelter) exceeded the US Environmental Protection Agency standard for residential floor dust lead loading.

All kitchen windowsill dust lead loadings were below the US Environmental Protection Agency standard for lead in residential dust of 250 $\mu\text{g}/\text{ft}^2$ for windowsills,

except one unusual sample in Shamlugh (1198 $\mu\text{g}/\text{ft}^2$). This unusual sample was collected from a windowsill that had visible dust/soil on it. This finding highlights the potential for a reaccumulation of higher lead levels in the absence of cleaning.

The floor and windowsill dust lead loadings were substantially lower than corresponding levels in high-risk lead-painted residential properties in urban areas in the USA (US Environmental Protection Agency, 1997; HUD, 2001b). One possible explanation could be that all apartment buildings in the study towns were built well after the 1920s, when the former Soviet Union (including Armenia) adopted the White Lead Convention of the International Labor Organization, banning the manufacture and sale of lead-based paint (International Labor Office, 1927). This Soviet ban appears to be one of the most effective national programs for implementing the White Lead Paint Convention (Silbergeld, 1995). Additionally, cultural factors related to housekeeping likely contributed to the low interior lead loadings. It is a common practice among Armenian women to do daily cleaning that includes dusting the windowsills and furniture and sweeping and mopping the floors. The interior floors are typically wooden and often covered by rugs. Heavy cleaning, including washing the windows with detergent, usually takes place every 2–3 months. Rugs and carpets typically get washed with detergent every summer. It is also a common practice for damp cloths to be placed at the entryway of the apartments and for people to remove their shoes upon entering their own apartments.

4.4. XRF versus ICP-OES

Our study found that lead loadings measured by XRF were significantly higher than, and highly correlated with, those measured by ICP-OES methodology following nitric acid digestion. A similar correlation coefficient of 0.93 ($P < 0.0001$) was found by others when wipe dust samples were analyzed by XRF and by flame atomic absorption spectroscopy (FAAS) methods (Sterling et al., 2000). Sterling et al. (2000) also found that lead loadings measured by NITON XRF tended to be higher than those measured by FAAS, but the difference was not statistically significant. They suggested that this pattern might be explained by losses that occurred during the digestion of the wipe samples. Thus, the lead loading estimates reported in this study would likely have been lower if the dust samples had been analyzed using ICP-OES or FAAS methodologies (more commonly used in the USA).

4.5. Other sources of lead exposure in Armenia

This study focused on the surveillance of lead in residential environments close to mining and smelting

operations, where emissions from those operations are considered to be the primary source of environmental lead contamination. We recognize, however, that there are other sources of lead in Armenia that can affect the residential environment. Leaded gasoline was generally used throughout Armenia during the Soviet era despite its ban in major Soviet cities and some capitals of the Soviet republics (Thomas and Orlova, 2001). Ninety percent of lead contamination in urban areas of Armenia is estimated to have been produced by leaded gasoline emissions (Kurkjian et al., 2002). The lead content in gasoline was estimated to be 0.15–0.37 g/L and sometimes up to 0.60 g/L (Kurkjian, 2000; Lovei, 1998). However, gasoline used in Armenia since 1998 has not been leaded (Kurkjian et al., 2002). This might be due to a phase-out of leaded gasoline in the supplier countries and a shift toward purchasing gasoline from countries in which leaded gasoline has already been phased out (Kurkjian et al., 1999; 2002; Thomas and Orlova, 2001). Moreover, effective March 2000, Armenia has prohibited the import of gasoline containing more than 0.15 g/L of lead (Kojima et al., 2000).

Additionally, during the last decade, unregulated small lead-acid battery-recycling shops have been operating in residential back yards in Armenia, creating a new source of lead contamination of the residential environment (Babayan et al., 1996, 1998a, 1998b, 1999; Kurkjian et al., 1999, 2002; Sarian et al., 1995).

5. Conclusion

This pilot environmental survey contributed to the assessment of lead in the residential environments in the mining and smelting district in the Lori region of northern Armenia. We found elevated lead levels in residential soil and dust in the study towns. Lead levels were the highest in the smelter town of Alaverdi, particularly in residential Sectors 3 and 5, which are closest to, and downwind from, the polymetallic smelter. The mining towns of Aghtala and Shamlugh had lower levels of lead in the environment than Alaverdi. This pattern was consistent across all sample types. Moreover, the concentrations of other metals (including arsenic) in the soil and loose exterior dust were also higher in Alaverdi than in the two mining towns. These patterns suggest that the emissions from the polymetallic smelter were primarily responsible for lead contamination in Alaverdi and that mining operations did not result in high lead contamination of the residential environments in Aghtala and Shamlugh.

Children typically spend significant amounts of time outside playing in their yards and could ingest lead-contaminated soil and dust through hand-to-mouth activities. Additionally, the very dry climate, the strong winds, and the presence of bare soil can create

conditions for resuspension of settled lead-contaminated dust into the air, contributing to its dispersion and potential increase of human exposure (Kurkjian et al., 2002). Settled dust lead loadings on interior floors in some cases exceeded the US Environmental Protection Agency residential dust lead loading standards, particularly in Alaverdi. We suggest that efforts be made by the local health authorities to educate the public about the sources of lead in the environment and the importance of preventive actions. Preventive efforts could be especially important if the smelter, which operated at approximately 30% of its capacity at the time of this study, increases future production, adding more lead into an already contaminated residential environment.

The findings of this preliminary environmental survey indicated that lead levels in residential areas of Alaverdi, Shamlugh, and Aghtala were lower than those found in residential soil and exterior dust in other mining and smelting areas, such as the Rudnaya Pristan–Dalnegorsk mining and smelting district in the Russian Far East, where the GM lead concentrations were 1626 mg/kg for garden soil, 1575 mg/kg for yard soil, and 4420 mg/kg for roadside dust (von Braun et al., 2002). These higher lead concentrations could be explained by the fact that the smelter in Rudnaya Pristan is a primary lead smelter that operates at full capacity.

During the study, we found that the participants were very interested and receptive to information about potential lead hazards and preventive measures. This study indicates not only a need for but also interest among the residents of the study towns in education about potential sources of lead in the environment and the importance of minimizing and preventing lead exposure. Preventive actions could include washing children's hands after being outside, in situ soil treatment, soil removal, blood lead testing in children, and installation of appropriate environmental control mechanisms in the smelter to reduce future emissions. The Armenian Ministry of Health has been informed about the study results, and ongoing efforts are being made to inform other interested agencies about the findings of the study so that they can develop educational and preventive programs. These efforts will be especially important as industrial operations in the area increase in the near future.

Because lead concentrations measured in this study exceed action levels in many cases, an ongoing need for monitoring, remediation and education exists in Armenia's northern mining district. Future research efforts might include investigation of the bio-available fraction of lead in soil and dust in the study towns, lead concentration in various particle size fractions of soil, and the lead concentrations in the soil around the rail lines that go through the residential areas and are easily

accessible to children. Other efforts should include investigating blood lead concentrations in children living in the study towns, particularly those in Alaverdi.

An infrastructure to support future work will be needed. Toward this end, it will be important to enroll Armenian laboratories in international proficiency programs (e.g., ELPAT) to assure quality and inter-comparability of environmental and blood lead data. This will help to ensure the collection of reliable data that could serve as the basis for effective environmental health policy in Armenia.

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