



Contamination levels and human health risk assessment of mercury in dust and soils of the urban environment, Vanadzor, Armenia

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ABSTRACT

Mercury has unique chemical features which predetermine its migration and accumulation behaviors in the environment. In this regard, the studies devoted to the identification of Hg contents and pollution issues should consider both transit and deponent medium such as air dust and soils, respectively. This paper aimed at exploring spatial distribution characteristics, pollution levels and human health risk of Hg in dust and soils of Vanadzor urban area. A total of 42 street dust, 13 leaf deposited dust and 84 soil samples were collected from an area covering the entire territory of Vanadzor, encompassing residential and industrial sites. The potential hazard associated with Hg was assessed with the use of geo-accumulation index, enrichment factor and potential ecological risk index. The spatial distribution of concentrations of Hg suggested that high values were detected near the Chemical Plant and on high traffic streets, indicate the possible anthropogenic origin of Hg. According to geo-accumulation index, most of the samples in street dust were characterized as moderately polluted, in leaf deposited dust – from unpolluted to moderately polluted levels and all soil samples were characterized by unpolluted levels. Enrichment values are significantly higher in dust samples compared with soils. The potential ecological risk assessment showed that high, considerable, and low levels of ecological risk dominated in the street dust, leaf deposited dust and soils in the City area, respectively. The results of health risk assessment showed that detected contents of Hg are not risky for the health of the population.

1. Introduction

Mercury (Hg) is one of the most toxic elements entering the environment from natural and anthropogenic sources (Bernhoft, 2012a,b; Kabata-Pendias and Mukherjee, 2007). Natural sources of Hg include volcanic action, weathering of rocks, forest fires etc. (Sheu and Lin, 2011). The majority of Hg in urban areas comes from the anthropogenic sources and uses of Hg (Bradl, 2005): gaseous, liquid and solid wastes, pharmaceutical industry, scientific instruments, Hg vapor lamps, paint and paper industry, coal-fired power, ceramic and cement production, dentistry (Bradl, 2005), biomass/fossil fuels burning in vehicles (Howard et al., 2017) etc. Hg pollution is of international concern because of its considerable potential hazard to the environment and public health (Bernhoft, 2012a,b; Kabata-Pendias and Mukherjee, 2007). Hg accumulating by airborne particles and soils (LIANG et al., 2009), can enter human bodies through inhalation, ingestion and dermal contact and cause adverse acute and chronic effects on human health (Zheng et al., 2015). Children are the most vulnerable (Tucaliuc

et al., 2014) to its effects as their central nervous system is still in the development process. Hg may cause central nervous and renal dysfunction, memory deficits, and a decrease in psychomotor skill. Hg may also have a deleterious effect on fetus, when passed from a pregnant mother to fetus (RAIS, 2018).

Investigations around the world have been conducted by studying two medium simultaneously: soils and street dust, as reservoirs for toxic elements (Cai et al., 2013; Christoforidis and Stamatis, 2009; Dehghani et al., 2017; LIANG et al., 2009; Nicholas Hewitt and Candy, 1990; Ordóñez et al., 2003; Xinmin et al., 2006). The investigation of these medium is of particular importance for two main reasons: 1) the soil gives the picture of long-term pollution (Perelman and Kasimov, 2000), street dust is characterized by a shorter residence time, as it is influenced by different factors such as wind, road runoff, and cleanup (Cai et al., 2013; Zhang et al., 2013), 2) following the deposited airborne particles, the adjacent soil particles are the second main source of street dust (Hu et al., 2011) and vice versa: pollutants in street dust can be released into soil. Mercury gets enriched in particles (Bradl, 2005)

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which can be transported and deposited not only on urban surfaces (Lu et al., 2014), but also on biological substances such as plant leaves (Wang et al., 2015). Therefore, tree leaf deposited dust was also studied.

In this study, the investigation of Hg was conducted in the city of Vanadzor, which was one of Armenia's leading industrial cities. The main industrial branches were machinery, chemicals, light and food industries. The most powerful chemical plant was “Vanadzor Qimprom” CJSC, which included three enterprises: chemical fibers, a chemical factory and the “Polimer-glue” scientific-production association (Mesropyan, 1984). In 1978, after the catastrophic accident in the Chemical Plant, the explosion of vacuum pumps resulted in the release of an essential quantity of Hg into the city environment. Investigation conducted in 1987 has shown that even after 9 years, high contents of Hg was again detected in the City environment (Jugaryan, 2000). Although the City lost its industrial power after the collapse of the Soviet Union, the Chemical Plant operated with small working capacity until 2017. Additionally, some factories and enterprises are still functioning nowadays and may be a potential source of Hg in the City area, as well.

The objectives of the present study were 1) to determine the concentration of Hg in street dust, leaf deposited dust and soil, 2) to evaluate contamination levels and spatial distribution of Hg in street dust, leaf dust and soil, and 3) to assess the potential ecological and health risk of Hg for children and adults via dust and soil exposure.

2. Material and methods

2.1. Study area

The urban area of Vanadzor (Armenia, Lori region) covers about 26 km², with a population of 82.2 thousand. Vanadzor is the third largest city of Armenia and is located in the northern part of the country (40° 48' 46" N, 44° 29' 18" E) at the heights of 1350 m (RA National Statistical Service, 2016). The City is located in the mountain-steppe landscape (State Committee of the Real Estate Cadastre. Center of Geodesy and Cartography, 2007). Its geological structure mainly represents loam, loamy sand and rubbles/shingles. Andesite, granite, basalt, obsidian, perlite and limestone rocks are also presented in the geological base of the City (State Committee of the Real Estate Cadastre. Center of Geodesy and Cartography, 2007).

Three rivers (Pambak, Tandzut and Vanadzor) flow through the City. The climate is characterized by warm, mild summers and moderately cold winters. The annual average temperature is 7.4 °C (Avagyan, 1997). The average air temperature in January is −4.2 °C (min. −32 °C), and in July −19.7 °C (max. 34.0 °C). Snow cover persists 90 days, and its depth ranges from 34 cm to 100 cm on average. The weather is relatively wet during the year. The annual average relative humidity is 70% (Baghdasaryan et al., 1971). In the months of January and February, southern winds dominate; in the month of January southwest winds dominate, too. The annual atmospheric precipitation is 600 mm (Babayan, 1989). Most rainfall days occur in the months of May, June, and July (Avagyan, 1997).

2.2. Sampling

The sampling of street dust, leaf deposited dust and soil was carried out in July–August 2016 (Fig. 1).

2.2.1. Street dust sampling

A total of 42 street dust samples (Fig. 1) were collected through the City, including the main and secondary streets exposed to different traffic density. The sampling was carried out based on guidance from international literature (Alecios Demetriades; Manfred Birke, 2015; Lu et al., 2010; Zhang et al., 2013) and our experience. At each sampling site, from 9 to 21 subsamples were taken depending on the length of the

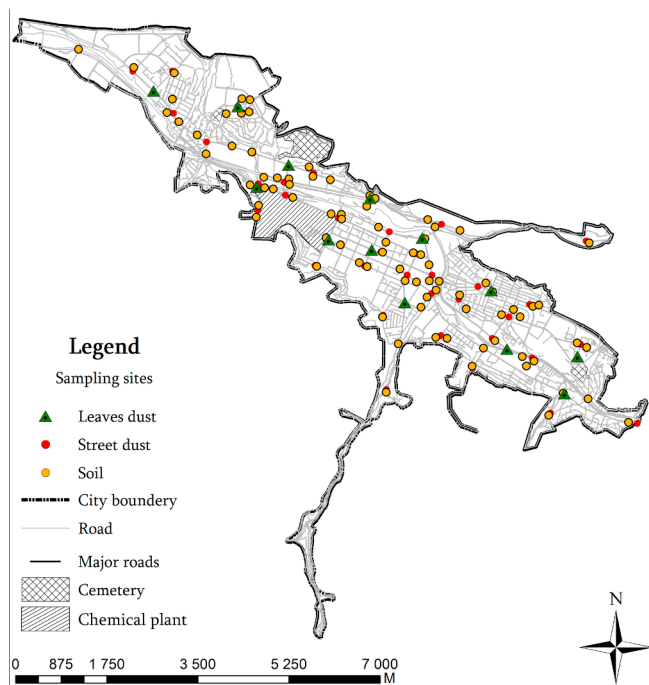


Fig. 1. Distribution map of sampling sites.

street and the presence of the intersection. The subsamples were mixed thoroughly to obtain a bulk sample. Each dust sample weighed 200–500 g and was collected from both sides along the street (including approximately 20 m of distance in each side) with a clean plastic dustpan and a brush, and then stored in plastic bags and transported to the laboratory. All the dust samples were air-dried in the laboratory in room conditions (under the < 25 °C) in order to avoid the volatilization of Hg. Dried samples were homogenized using an agate mortar and then sieved via a nylon sieve with a diameter of 1 and 0.063 mm. The samples were then stored in sealed bags until the analyses stage commenced.

2.2.2. Leaf deposited dust sampling

42 tree-leaf samples (Fig. 1) were collected from the entire area of the City, generally from the same sampling sites as street dust samples. Tree species such as *Ulmus parvifolia* L., *Juglans regia* L., *Fraxinus excelsior*, *Acer platanoides* L., *Populus alba* L. and *Populus nigra* L. were sampled considering the presence and the amount of the same species in the sampling site. Tree leaves sampling was performed based on international literature guidance (Ilkun, 1978; Saet Yu et al., 1982; Tomašević and Aničić, 2010) and our experience (Maghakyan et al., 2017). Three to four subsamples were taken with the purpose of generating one bulk sample. Tree leaves (to avoid loss of deposited dust amount) were carefully sampled at the height of 1.5–2 m above the ground and then placed in paper bags for transfer. In the laboratory, the leaf samples were air dried in room temperature, and then washed with de-ionized water (MilliQ). The generated liquid was filtered using a weighed ash free filter (retention limit set at 0.45 µm). In some cases, because of insufficient amount of dust in the filters for the Hg content analysis, two to six filters (from the samples spatially located close to each other) were combined. Finally, 13 leaf dust samples were gained. Before the use of the filters, they were first placed in a drying chamber (from 30 °C to 40 °C) for at least 8 h and then in a desiccator until stable weight is maintained. Afterwards, filters were weighed by analytical balance (VLR-200) with the 4 digit (0.0001 g) weighing precision. Two filters have been prepared as a blank sample. For the blank sample, the same amount (appropriate for washing the leaves) of de-ionized water

(MilliQ) was retrieved from the clean weighted filter. All filters (blank filters and filters with leaf deposited dust) were dried, weighed again and stored for the analysis.

2.2.3. Soil sampling

The geochemical soil survey of Vanadzor was implemented using the scale of 1:25000. A total of 355 soil samples were taken. In this study, only soil samples located close to street dust samples were considered (total of 84 samples, combined into 42) (Fig. 1). Also, 20 soil samples were collected from a pristine area outside the City having similar geological features as the City territory, in order to establish the local soil background value of Hg.

Soil samples (topsoil, 0–5 cm) were collected with a stainless-steel shovel. At each sampling site, three to five subsamples were taken as bulk samples. Samples (weighing 800–1500 g) were stored in polyethylene bags for transportation and storage. They were air-dried in the laboratory and then sieved consistently through 2 and 1 mm mesh according to ISO-11464 (US EPA, 2007). Approximately 50 g of homogenized soil were ground in the anagate mortar and stored in sealed bags until the analyses stage.

2.3. Analytic methods and QA/QC

The total concentration of Hg in the street dust and soil was determined by applying the X-ray fluorescence spectrometry (Olympus Innov-X-5000 (USA)), according to the EPA standard method 6200 (US EPA, 2007) in the Center for Ecological-Noosphere Studies (CENS). The QA/QC of the analysis has shown that according to the Standard Reference Materials (NIST 2711 and NIST 2710, USA) the accuracy of the digestion of street dust samples was 12.3% and for soils - 14.4%, which did not exceed the acceptable threshold of 20%. The precision was 13.6% for street dust and 4% for soils. For interlaboratory control, 12.5% (5 sample) of street dust samples were analyzed in the CENS Central Analytical Laboratory (accredited by ISO-IEC 17025).

To determine Hg in leaf deposited dust, the dried filters were placed in the Erlenmeyer flask and 10 mL of aqua regia (HNO₃ -HCl 1:3v/v) was added. Then the solution stood for 12 h after which it was carefully heated on a hot plate (60 °C) until the production of red NO₂ fumes had ceased. The flask was cooled and a small amount (1–2 mL) of H₂O₂ was added. The solution was heated again and left to evaporate to a smaller volume of liquid. The solution was then transferred to a 100-mL volumetric flask and diluted to a volume with distilled water. Mercury contents were determined by cold vapor atomic absorption spectroscopy (AAnalyst 800 AAS PE, USA). The relative standard deviation (RSD) of triplicate measurements was within 5%. The Standard reference material (CRM024-50G) was used to assess the accuracy of the analyses. The accuracy error (9.87%) was within the allowable limit of 15% (Albanese and Cicchella, 2018).

2.4. Statistical analysis and mapping

Statistical analyses was conducted using the SPSS 20 Statistical software. The normality of data distribution was tested by Shapiro Wilke test. Pearson and Spearman correlation analysis (CA) was performed to reveal the relationship of Hg concentrations in the studied medium. ArcGIS software and IDW method were used for data mapping and interpolation.

Statistical treatment of the data collected at the background site was performed. Soil local geochemical background was established using boxplot and interactive 2σ methods (Reimann et al., 2018; G. O. Tepanosyan et al., 2017).

2.5. Pollution assessment

Common indices acknowledged and used in professional academic literature were used to assess the pollution levels of Hg in street dust,

leaf dust and soils. These were the concentration coefficient (K_c) (Golovin et al., 2000; Perelman and Kasimov, 2000), the Geo-accumulation index (I_{geo}) (Ghrefat et al., 2011; Muller, 1969; Tang et al., 2017) and the Enrichment factor (EF) (Cai et al., 2013; Ghrefat et al., 2011; Gope et al., 2017).

The Concentration coefficient was calculated using the following equation (Golovin et al., 2000; Perelman and Kasimov, 2000)

$$K_c = \frac{C_i}{C_f} \quad (1)$$

where C_i - is the concentration of Hg in each media: street dust, leaf dust, soil, C_f - is the geochemical background value of Hg for Vanadzor soil ($Hg = 0.05 \text{ mg/kg}$). The K_c is classified as acceptable ($K_c < 4$), moderately dangerous ($4 < K_c < 8$), dangerous ($8 < K_c < 16$), highly dangerous ($16 < K_c < 32$) and extremely dangerous ($K_c > 32$) (Golovin et al., 2000).

The Geo-accumulation index was calculated using the following equation (Ghrefat et al., 2011; Muller, 1969; Tang et al., 2017).

$$I_{geo} = \log_2 \frac{C_n}{1.5B_n} \quad (2)$$

where C_n was the concentration of Hg in each media: street dust, leaf deposited dust, and soil, B_n was the geochemical background value of Hg for Vanadzor soil ($Hg = 0.05 \text{ mg/kg}$). For the I_{geo} , the following classifications are defined: uncontaminated ($I_{geo} \leq 0$); uncontaminated to moderately contaminated ($0 \leq I_{geo} \leq 1$); moderately contaminated ($1 \leq I_{geo} \leq 2$); moderately to strongly contaminated ($2 \leq I_{geo} \leq 3$); strongly contaminated ($3 \leq I_{geo} \leq 4$); strongly to extremely contaminated ($4 \leq I_{geo} \leq 5$); and extremely contaminated ($I_{geo} \geq 5$) (Ghrefat et al., 2011; Muller, 1969; Tang et al., 2017).

The Enrichment factor (EF) is an important parameter to evaluate the degree of anthropogenic influence. EF was calculated using the following equation (Cai et al., 2013; Ghrefat et al., 2011; Gope et al., 2017).

$$EF = \frac{C_{n_{\text{sample}}}/C_{\text{conservative element}}}{C_{n_{\text{background}}}/C_{\text{conservative background}}} \quad (3)$$

where $C_{n_{\text{sample}}}$ is the concentration of Hg in the samples, $C_{\text{conservative element}}$ is the concentration of conservative element (Ti) in the sample, $C_{n_{\text{background}}}$ is the background concentration of Hg, and $C_{\text{conservative background}}$ is the background concentration of the conservative element. The Enrichment factor was categorized as follows: $EF < 2$ - deficiency to minimal enrichment; $EF = 2-5$ - moderate enrichment; $EF = 5-20$ - severe enrichment; $EF = 20-40$ - very high enrichment; and $EF > 40$ - extremely high enrichment (Cai et al., 2013; Ghrefat et al., 2011; Gope et al., 2017).

2.6. Potential ecological risk assessment

The potential ecological risk was calculated using the following equation (Liang et al., 2015; Saeedi et al., 2014; Yuan et al., 2014; Zhang et al., 2013):

$$E_r = T_r \times C_f \quad (4)$$

$$C_f = \frac{C_n}{C_f} \quad (5)$$

where E_i is the potential ecological risk factor of Hg, T_i is the toxic-response factor of Hg - and is 40, C_f is the background value of Vanadzor soils, and C_n is the concentration of Hg in a sample. The ecological risk degree was classified as low ($E_r < 40$), moderate ($40 < E_r < 80$), considerable ($80 < E_r < 160$), high ecological risk ($160 < E_r < 320$) and a very high ecological risk ($E_r > 320$) (Saeedi et al., 2014; Yuan et al., 2014; Zhang et al., 2013).

2.7. Non-carcinogenic health risk assessment

Health risk assessment of the exposure of both adults and children to Hg in street dust, leaf deposited dust and soil was performed based on the model developed by the US Environmental Protection Agency (RAIS, 2018; US EPA, 2002, 1989). Three pathways of exposure to Hg were considered: ingestion of particles, inhalation of particles and vapor, and dermal absorption by the skin.

The chronic daily intake (CDI mg/kg-day) of Hg for each pathway was calculated using the following Eqs. (6)–(8) (RAIS, 2018; US EPA, 2002, 1989).

$$CDI_{ing} = \frac{C \times IRS \times EF \times ED \times RBA \times 10^{-6}}{AT \times BW} \quad (6)$$

$$CDI_{inh} = \frac{C \times InhR \times EF \times ED}{AT \times BW \times (PEF + VF)} \quad (7)$$

$$CDI_{dermal} = \frac{C \times EF \times ED \times SA \times AF \times ABSd \times 10^{-6}}{AT \times BW} \quad (8)$$

The Hazard quotient for each pathway and the Hazard index were calculated using the following formulas (9), (10) and (11) (RAIS, 2018; US EPA, 2002, 1989).

$$HQ_{ing} = \frac{CDI_{ing}}{RfD_{ing}} \quad (9)$$

$$HQ_{inh} = \frac{CDI_{inh}}{RfD_{inh}} \quad (10)$$

$$HQ_{dermal} = \frac{CDI_{dermal}}{RfD_{dermal}} \quad (11)$$

$$HI = \sum HQ \quad (12)$$

The description and values of factors used in formulas (6)–(12) are outlined in Table 1.

3. Results and discussion

3.1. Hg concentrations and distribution in dust and soils in Vanadzor

The descriptive statistics of Hg concentrations in the street dust, leaf deposited dust and urban soils of Vanadzor, as well as background values of Vanadzor soils, are summarized in Table 2.

The concentration of Hg in leaf deposited dust varies from 0.03 to 3.29 mg/kg, with the mean of 0.57 mg/kg. High concentration of Hg was detected near Vanadzor Chemical Plant. The mean values were

Table 1
Description and values of factors used in the risk assessment formulas.

Factors (measurement unit)	Description	Value		Source
		Adults	Children	
IRS (mg/day)	Ingestion rate	100	200	US EPA (2002)
EF (day/yr)	Exposure frequency	350*	350*	—*
ED (yr)	Exposure duration	26	6	US EPA (2002)
BW (kg)	Average body weight	70	15	US EPA (2002)
InhR (m ³ /day)	Inhalation rate	20	7.6	Van den Berg (1994)
SA (cm ²)	Skin area	5700	2800	US EPA (2002)
AF (mg/cm ²)	Skin adherence factor	0.07	0.2	US EPA (2002)
PEF (m ³ /kg)	Particle emission factor	1.36 × 10 ⁹	1.36 × 10 ⁹	US EPA (2002)
VF (m ³ /kg)	Volatilization factor (Hg)	3.47 × 10 ⁴	3.47 × 10 ⁴	US EPA (2002)
RBA	Relative bioavailability	1		RAIS (2018)
AT	Average time	AT = 365×ED		US EPA (2002)
ABS _d	Dermal absorption	ABS = 0.03		RAIS (2018)
RfD _{ing} (mg/kg -day)	Chronic reference dose, Chronic reference concentration	1.6E-04		RAIS (2018)
RfC _{inh} (mg/m ⁻³)		3E-04		RAIS (2018)

Note: «*» - values used for this particular research.

Table 2

Descriptive statistics of mercury concentrations (mg/kg) in street dust, leaf dust and soils of Vanadzor.

Descriptive	Street dust	Leaf dust	Soil
Mean	0.266	0.570	0.041
Median	0.259	0.162	0.038
Std. Deviation	0.123	0.910	0.012
CV %	46.03	159.49	29.79
Skewness	−0.248	2.572	0.700
Minimum	0.040	0.027	0.013
Maximum	0.547	3.295	0.070
Background	0.05		

Table 3

Mean, minimum and maximum concentrations of Hg (mg/kg) in street dust and soils in some cities.

City	Hg in Street/road/roadway dust (mg/kg)			
	Mean	Min	Max	Reference
Baoji, China	1.1	0.5	2.3	Lu et al. (2010)
Nanjing, China	0.12	0.05	0.34	Hu et al. (2011)
Shanghai, China	0.3	—	—	Hu et al. (2011)
Huainan, China	0.11	0.02	0.56	Zheng et al. (2015)
Huludao, China	1.222	0.119	5.212	Xu et al. (2013)
Xiamen, China	0.28	0.034	1.4	Liang et al. (2009)
Ahvaz, Iran	2.53	0.02	8.75	Nazarpour et al. (2017)
Aviles, Spain	2.56	1.2	10.8	Ordóñez et al. (2015)
Kavala, Greece	0.1	—	—	Christoforidis and Stamatis (2009)
Bhilai, India	2.1	0.3	3.0	Ambade and Litrupa (2012)
Shiraz, Iran	1.05	0.004	4.504	Keshavarzi et al. (2015)
Hg in Soil				
Lithgow, Australia	0.053	0.019	0.0149	Rouillon et al. (2013)
Athenas, Greece	0.166	0.01	1.08	Kelepertzis and Argyraki (2015)
Aviles, Spain	0.57	0.17	9.83	Nazarpour et al. (2017)
Kavala, Greece	0.1	—	—	Christoforidis and Stamatis (2009)
Xiamen, China	0.46	0.071	1.2	LIANG et al. (2009)
Beijing, China	0.3	0.022	9.4	Chen et al. (2010)
Wuhu, China	0.207	0.024	2.844	Fang et al. (2011)
Berlin, Germany	0.42	—	71.2	Birke and Rauch (2000)
Yerevan, Armenia	0.115	0.005	0.270	(G. Tepanosyan et al., 2017a)
Gyumri, Armenia	0.07	0.01	0.38	(G. Tepanosyan et al., 2017b)

compared with the results received in the city of Yerevan, where similar studies using tree leaves were implemented. The mean value of Hg in the leaf deposited dust from Yerevan areas was 0.57 mg/kg; this was the same value as in the case of Vanadzor. The maximum value of Hg was

1.4 times higher in Vanadzor, compared to the maximum value detected in Yerevan (2.37 mg/kg), which may also be explained by the presence of the Chemical Plant - a known source of Hg in the city of Vanadzor.

The concentration of Hg in street dust varies from 0.04 to 0.55 mg/kg with the mean of 0.27 mg/kg. Table 3 shows the reported concentrations of Hg in dust and soils from different cities. The mean value of Hg in street dust is much lower than the concentrations reported for Ahvaz (Iran) (Nazarpour et al., 2017), Aviles (Spain) (Ordóñez et al., 2015), Huludao (Xu et al., 2013), Baoji (China) (Lu et al., 2010), Shiraz (Iran) (Keshavarzi et al., 2015) and Bhilai (India) (Ambade and Litrupa, 2012) cities. These cities are important industrial centers for the countries where such probations have been conducted and which are characterized with high level of traffic and presence of intensive industrial activities and plants. The concentrations of Hg in the above-mentioned cities are reported to have an anthropogenic origin. Particularly, in the case of Aviles (with the mean of 2.56 mg/kg) the observed high concentration of Hg was mainly associated with steel industry (Ordóñez et al., 2015), meanwhile for Ahvaz (with the mean of 2.53 mg/kg) the main sources of Hg were traffic emissions, oil-drilling, and steel smelting-related industries (Nazarpour et al., 2017) and for Shiraz - cement and coal power plants (Keshavarzi et al., 2015). However, the mean value of Hg in Vanadzor's street dust was higher compared with Nanjing (China) (Hu et al., 2011), Shanghai (China) (Hu et al., 2011), Huainan (China) (Zheng et al., 2015), Xiamen (China) (LIANG et al., 2009) and Kavala (Greece) (Christoforidis and Stamatis, 2009). However, it needs to be mentioned that the direct comparison of the results from different urban areas has some scientific limitation and while doing these we should remember that the differences in the mean contents of Hg in the dust of urban areas may be related not only to the differences observed in pollution sources, climate and weather conditions, but may also depend on the sampling methods, density and the applied analytical technique.

The concentration of Hg in soils varies from 0.01 to 0.07 mg/kg, with the mean of 0.04 mg/kg. In comparison with other cities in Armenia, the mean of Hg concentration in soils used in this study was lower than the mean concentrations detected in Gyumri (0.07 mg/kg) (G. Tepanosyan et al., 2017b) and in Yerevan (0.115 mg/kg) (G. Tepanosyan et al., 2017a). The mentioned cities are the main industrial cities of the country where Hg contents have been reported to be originating from mixed sources (G. Tepanosyan et al., 2017a, 2017b). The mean concentration of Hg in Vanadzor soils was also lower than the indicator reported for other cities around the world, such as Lithgow (Rouillon et al., 2013), Athens (Kelepertzis and Argyraki, 2015), Aviles (Nazarpour et al., 2017), Kavala (Christoforidis and Stamatis, 2009), Xiamen (China) (LIANG et al., 2009), Beijing (China) (Chen et al., 2010), Wuhu (China) (Fang et al., 2011), and Berlin (Germany) (Birke and Rauch, 2000). Therefore, the Hg concentration in soils of Vanadzor, with limited exceptions, may be considered as low - close to the local background.

In all medium (street dust, leaf deposited dust, soil), the mean values of Hg exceeded the median, which is the anticipated difference of the current distribution from a normal distribution. The asymmetry of Hg was proven by skewness (> 1 values) and its error values. The Hg concentrations in street dust and soil were between 2σ , meanwhile in the case of leaf deposited dust, they were out of 2σ , which suggested that the concentration of Hg in soils and street dust had an approximately normal distribution, and show non-normal distribution in leaf deposited dust. The presented boxplots (Fig. 2) and histograms also indicated the normal distribution of Hg in soils and street dust, and non-normal distribution in leaf deposited dust, which was verified by Shapiro-Wilk normality test. The non-normal distribution of Hg observed in the leaf deposited dust indicated its anthropogenic origin.

The results of Pearson correlation have shown a significant positive correlation (0.438 at the level $P < 0.01$) between concentrations of Hg in street dust and soils. This may propose the existence of the same

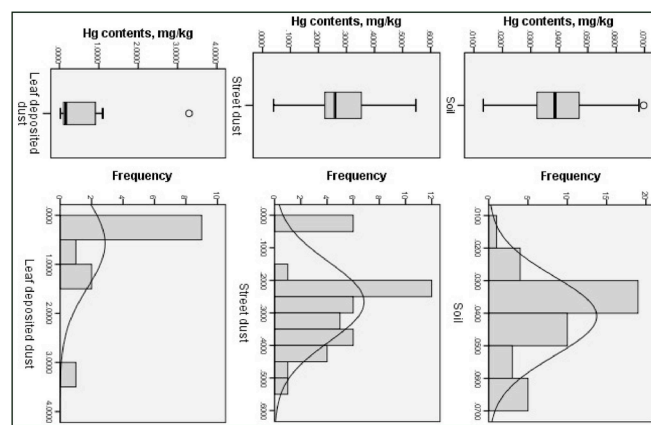


Fig. 2. Boxplots and histograms of Hg concentrations in street dust, leaf deposited dust and soils.

potential sources near the sampling sites of the both medium. Meanwhile, the Spearman correlation coefficient was not significant between the Hg concentrations in leaf deposited dust and street dust (correlation coefficient -0.121), and between leaf deposited dust and soils (correlation coefficient -0.06). These results can be explained by the unusual high volatility of Hg and its long atmospheric lifetime (Beckers and Rinklebe, 2017). We suppose that the low concentrations of Hg in soils compared with dust samples may be connected with the unstable behavior of Hg in soils. Particularly, as mentioned by Bradl (2005) organic and inorganic Hg compounds decompose to elemental Hg, and due to the conversation may be volatilized into the atmosphere and thus be easily accumulated by air dust particles. Moreover, the results of the similar study conducted by Xinimin Zh. et al. in the Beijing's area also detected a comparably higher concentration of Hg in dust samples vs soils, reporting the tendency of the enrichment of mercury in dust (Xinmin et al., 2006).

The distribution map (Fig. 3) of Hg concentrations in street dust has shown that Hg high values ($> 95\%$) were detected in 2 samples: one collected in the western part of the city (in the intersection of a residential area) and the other, in the southern part of the city (in an industrial area, near the Vanadzor Chemical Plant) occupying an area of 0.3 km^2 (0.07% of the territory). Concentrations between 95 and 75% were observed in 8 samples: those collected from the southern, south-western and western parts of the city and also from a small section in the central part of the City and those samples occupy an area of 2.5 km^2 (10.55% of the territory). The main area of the City is (an area of 10.98 km^2 - 47.82% of the territory) characterized with concentrations between 75 and 50%. These concentrations are observed in the north-west and the south-east, as well as the central parts of the City including both residential and industrial sites. The concentrations between 25-50% and 25-5% occupy an area of 4.84 km^2 (10 samples take up the

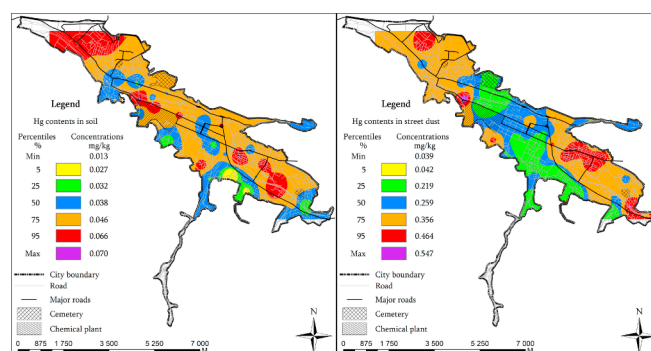


Fig. 3. Spatial distribution of Hg concentrations in Vanadzor street dust and soils.

Table 4
Geoaccumulation index and ecological risk values of Hg in dusts and soil.

Media	I _{geo} values			Er values		
	min	max	mean	min	max	mean
Street dust	−0.92	2.87	1.56	31.7	437.8	212.93
Leaf deposited dust	−1.47	5.46	1.55	21.6	2636.0	456.22
Soil	−2.50	−0.11	−0.94	10.6	55.7	32.71

20.5% of the territory) and an area of 4.96 km² (9 samples take up the 21.04% of the territory) including mainly the central part of the City. Concentration under 5% is observed in only one sample.

The distribution map of Hg (Fig. 3) concentrations in soils have shown that high values (> 95%) of Hg were detected in one sample (occupying an area of 0.01 km², the 0.05% of the territory) within the industrial area, near the Vanadzor Chemical Plant. Concentrations from 75 to 95% were detected in 9 samples and occupy an area of 3.62 km² (the 15.34% of the territory) covering both residential and industrial areas. Concentrations between 50 and 75% were detected in 11 samples and occupy an area of 13.04 km² – the 55.26% of the territory. Concentrations between 25 and 50% were detected in 11 samples and occupy 5.55 km² (the 23.53% of the territory). Concentrations between 5 and 25% were detected in 9 samples and occupy 1.18 km² (5.01% of the territory). Concentrations < 5 were detected in one sample.

Overall, the spatial locations of high Hg concentrations in street, leaf dust and soils have proven that the main sources of Hg in the City are prescribed to Vanadzor Chemical Plant, car technical services and vehicular emissions. High concentrations of Hg exceeding the local background value were detected in a different part of the City and are supposed to be associated with local point sources, the behaviors of particulate Hg and the possibility of its diffusion in a wide area (Xinmin et al., 2006). Additionally, the prevailing wind direction may also have its influence on the distribution of Hg in the City territory. During the year the prevailing wind directions in the area are southern and southwestern (the wind direction, based on the perennial data for the city of Vanadzor). The hotspots of street and leaf deposited dust Hg concentrations located in the eastern and northwestern parts of the City may also be influenced by the distribution of Hg contents from potential sources through prevailing wind directions.

The local background value of Hg in Vanadzor soils is estimated to be 0.05 which is in line with the geological base. The latter is mainly represented by granite, basalt and limestone rocks and, as mentioned in the literature the average natural content of Hg in granite is 0.03 mg/kg, in basalt it is 0.09 mg/kg and 0.04 mg/kg in limestone (Bradl, 2005).

Excess values of Hg concentrations vs background were detected in all investigated medium. The Hg concentrations exceed the background concentration in 85% of the samples (11 samples) of the leaf deposited

dust samples: from 1.61 to 65.9 times. The mean concentration exceeds the background by 11.4 times. In street dust samples, Hg concentration exceeds the background concentration in 85.7% of the samples (36 samples): from 3.53 to 10.9 times. The mean concentration exceeds the background by 5.3 times. In case of soils, Hg concentration exceeds the background concentration in 11.9% of the samples (5 sample). Soil Hg mean concentrations were below the background concentration. Moreover, in 89.1% of the soil samples, the concentration of Hg is lower or close to the background value, which indicates the natural origin of Hg in soil samples, as the geological base of the City is characterized with rocks, which have a similar content of Hg (content of Hg in granite is 0.03 mg/kg, in basalt it is 0.09 mg/kg, in limestone it is 0.04 mg/kg (Bradl, 2005)). In all medium, the maximum concentration of Hg was detected near the Vanadzor Chemical Plant, indicating the possibility of Hg emissions from the Plant area, which again prove, that the Chemical Plant is still considered to be the main potential source of Hg and has a direct influence on Vanadzor environment.

3.2. The geo-accumulation index (I_{geo}) and the enrichment factor (EF) of Hg

The calculated values of Geo-accumulation indices (I_{geo}) for Hg in dust and soils are presented in Tables 4 and 5.

I_{geo} values in leaf deposited dust range from −1.47 to 5.46, with the mean of 1.54. Five levels of contamination were detected: unpolluted (in 2 samples – the 15.38% of the total samples), unpolluted to moderately polluted (in 4 samples – the 30.77% of the total samples), moderately polluted (in 3 samples constituting the 23.07% of the total samples), strongly polluted (in 3 samples taking up the 23.07% of the total samples) and extremely polluted (in 1 sample constituting the 7.69% of the total samples) (Table 5, Fig. 4.).

I_{geo} values in street dust range from −0.92 to 1.56, with the mean of 2.87. The following three levels of pollution were detected: unpolluted (in 6 samples constituting the 14.29% of the total samples and occupying an area of 0.85 km² (4% of the territory)), moderately polluted (in 19 samples taking up the 45.24% of all samples and occupying an area of 14.87 km² (63% of the territory)), moderately to strongly polluted (in 17 samples constituting the 40.48% of all samples, and occupying an area of 5.92 km² (25% of the territory)). Meanwhile, in soils, only an unpolluted level of contamination was detected. I_{geo} values in soils range from −2.5 to −0.11, with the mean of −0.94. This suggests that the volatile feature of Hg and the Hg accumulative potential of city soils may play a significant role on the way of Hg transportation from the dynamic medium to soils. Moreover, as mentioned by Xinmin Zh. et al. (Xinmin et al., 2006) soils represent the local pollution with point source, meanwhile dust pollution with Hg is very complex and is presented as a non-point source of pollution.

I_{geo} values (Fig. 4) have shown that pollution levels of Hg in street and leaf deposited dust have different spatial distribution which may be the result of differences of migration of Hg incorporated with air dust and street dust particles. However, near the Vanadzor Chemical Plant,

Table 5
Geoaccumulation index of Hg in street dust, leaf deposited dust and soils.

Value	Pollution levels	Hg in street dust	Hg in leaf deposited dust	Hg in soils
Amount of samples (%)				
I _{geo} < 0	Unpolluted	14.28	15.38	100
0 < I _{geo} < 1	Unpolluted to moderately polluted	0	30.77	0
1 < I _{geo} < 2	Moderately polluted	45.24	23.07	0
2 < I _{geo} < 3	Moderately to strongly polluted	40.48	0	0
3 < I _{geo} < 4	Strongly polluted	0	23.07	0
4 < I _{geo} < 5	Strongly to extremely polluted	0	0	0
5 < I _{geo}	Extremely polluted	0	7.69	0

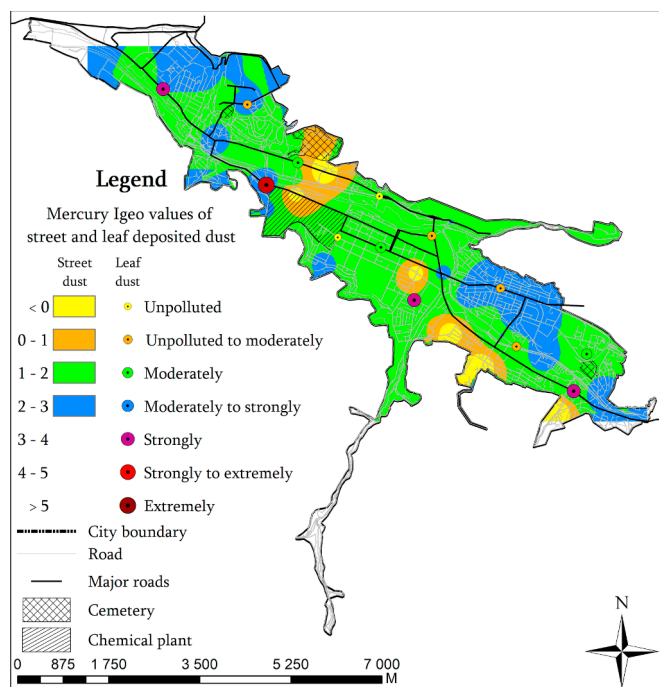


Fig. 4. Spatial distribution of I_{geo} in street and leaf deposited dust.

which is known to be the main source of Hg in the City's territory, the extremely polluted level in leaf deposited dust and the moderately to strongly polluted level in street dust were observed.

The results of Enrichment factor estimation showed that EF values in all soil samples were less than 2 indicating the deficiency to minimal enrichment. Meanwhile, in 85.7% (36 samples) of street dust samples, EF values were between 2 and 10, which show a moderate enrichment in these samples. Moreover, EF values above 3 reflect the anthropogenic pollution with Hg in the area (Shi and Wang, 2013). These values again support the statement that higher concentration of Hg in the City is mainly associated with industrial activities and heavy traffic.

3.3. Potential ecological risk assessment

The potential ecological risk (E_r) values of Hg in street dust, leaf deposited dust and soils are outlined in Table 4. According to the results, the City area is characterized with 5 levels of ecological risks in street dust and leaf deposited dust, and only with two levels in the case of soils.

In street dust, the E_r value ranges from 31.7 (indicating a low level of ecological risk) to 437.76 (indicating a very high level of ecological risk) with a mean of 212.9 (indicating a high level of ecological risk). In 14.28% of all samples (6 samples) E_r values were < 40 , indicating a low level of ecological risk and occupying an area of 0.06 km² (the 0.25% of the territory). A considerable level of ecological risk is observed only in one sampling site. In 69.05% of all samples (29 samples), the ecological risk was classified as a high level risk occupying an area of 20.29 km² (the 58.98% of the territory). A very high level of ecological risk was detected in 14.28% of all samples (6 samples) and occupies an area of 0.55 km² (the 2.32% of the territory).

In leaf deposited dust, the mean value of potential ecological risk is 21.6, indicating a low level of ecological risk. The maximum value is 456.2, indicating a very high level of risk. In 15.4% samples (2 samples), a low level of ecological risk was observed, in 7.69% of samples (1 sample) – a moderate level, in 38.46% (5 samples) the risk was classified as significant, in 7.69% (1 sample) it was high and in 30.77% of all samples (4 samples) a very high level was detected. In the samples collected from the north-western to the north-eastern part of the City, a

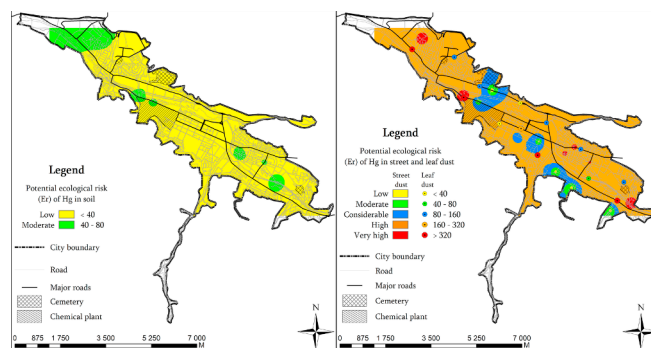


Fig. 5. Spatial distribution of Ecological risk index levels in studied medium.

significant level of ecological risk was dominant, while samples representing the area spanning from south-east to south-west part demonstrated a high level of ecological risk.

In comparison with street dust and leaf deposited dust, only two levels of risk were observed in soils. Their E_r values ranged from 10.6 to 55.7, with the mean of 31.9. In 80.95% of samples (34 samples), a low level of ecological risk was observed, while in 19.05% of samples (8 samples), a moderate level was detected.

In Fig. 5, the spatial distribution of the ecological risk index is illustrated indicating that the highest values of E_r in soil, street and leaf deposited dust are identified at the sampling points located near the streets with heavier traffic and in industrial zones including the Chemical Plant surrounding area.

3.4. Human health risk assessment

Non-carcinogenic health risk assessment results both for children and adults caused by Hg exposure in street dust, leaf deposited dust and soils are listed in Table 6. The HQ values for Hg from each exposure pathway (ingestion, inhalation and dermal) in all medium are below the allowable level 1. These results indicated the absence of adverse effects on human health both for children and adults.

HI values from the exposure to Hg in street dust range from 3.18E-03 to 4.38E-02, with the mean of 2.13E-02 for children, and from 3.41E-04 to 4.70E-03, with the mean of 2.29E-03 for adults.

HI from leaf deposited dust ranges from 2.4E-03 to 1.9E-01, with the mean of 5.0E-02 for children and from 2.6E-04 to 2.0E-02, with the mean of 5.4E-03 for adults and in soils from 1.06E-03 to 5.58E-03, with the mean of 3.28E-03 for children, from 1.14E-04 to 5.99E-04, with the mean of 3.52E-04 for adults. The HI values for the different exposure pathways from different medium occurred in the following order: ingestion > dermal contact > inhalation. Our results are consistent with those of other studies, which have shown that the main pathway of the exposure of Hg in dust and soils was ingestion (Huang et al., 2014; Wang et al., 2014). All HI values were below 1, which indicated that there was no risk from multiway exposure, too.

4. Conclusion

The study of Hg in interlinked environmental compartments of the urban area of city of Vanadzor reveals the differences of Hg contents and associated pollution in air dust, street dust and soil medium. Particularly, Hg contents in dust and soils of Vanadzor showed, that the mean concentration of Hg in leaf deposited dust, street dust and soil were 0.57, 0.26, 0.04, respectively. Significant correlation between the concentrations of Hg in street dust and soil samples observed, whereas correlation between leaf deposited dust with street dust and soil was not significant.

The distribution of the concentration of Hg in dust and soil indicated that the highest concentrations of Hg were found in industrial

Table 6

Non carcinogenic health risk of Hg in dust and soils.

Indexes	Values	Street dust		Leaf deposited dust		Soil	
		Child	Adult	Child	Adult	Child	Adult
HQ_{ing}	min	3.17E-03	3.39E-04	2.4E-03	2.6E-04	1.06E-03	1.13E-04
	max	4.37E-02	4.68E-03	1.9E-01	2.0E-02	5.57E-03	5.96E-04
	mean	2.13E-02	2.28E-03	5.0E-02	5.3E-03	3.27E-03	3.50E-04
HQ_{derm}	min	8.87E-06	1.35E-06	6.7E-06	1.0E-06	2.96E-06	4.53E-07
	max	1.22E-04	1.87E-05	5.3E-04	8.1E-05	1.56E-05	2.38E-06
	mean	5.96E-05	9.09E-06	1.4E-04	2.1E-05	9.15E-06	1.40E-06
HQ_{inh}	min	9.31E-08	9.31E-08	7.1E-08	7.1E-08	3.11E-08	3.11E-08
	max	1.29E-06	1.29E-06	5.6E-06	5.6E-06	1.64E-07	1.64E-07
	mean	6.26E-07	6.26E-07	1.5E-06	1.5E-06	9.61E-08	9.61E-08
HI	min	3.18E-03	3.41E-04	2.4E-03	2.6E-04	1.06E-03	1.14E-04
	max	4.38E-02	4.70E-03	1.9E-01	2.0E-02	5.58E-03	5.99E-04
	mean	2.13E-02	2.29E-03	5.0E-02	5.4E-03	3.28E-03	3.52E-04

zones and residential areas with heavy traffic density. The detected high values are significantly higher than those observed in Vanadzor soil background in the street and leaf deposited dust. A significant contamination of leaf deposited dust and street dust was observed near the Vanadzor Chemical Plant, which was the main known source of Hg in the area. The results of the Geo-accumulation index evaluation and the ecological risk assessment also have shown that the highest values of I_{geo} , E_r and EF were detected in the industrial and high traffic density areas. The health risk assessment having detected the concentration of Hg in all medium does not represent a risk for the City's population. This Paper helps to draw three following conclusion: depending on the behavior of Hg, the investigation of different medium is of particular importance. To understand the process and dynamics in more detail, it is recommended to conduct further investigations on identifying pollution sources and carry out a comprehensive investigation of the known sources including the territory of the Chemical Plant.

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