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ORIGINAL PAPER

Cadmium Health Risk Assessment and Anthropogenic Sources of Pollution in Mount-Lebanon Springs

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Abstract

Cadmium (Cd) is a highly toxic heavy metal with low permissible limits. This study aims at conducting a health risk assessment for Cd and examining potential sources of pollution in springs of the Mount-Lebanon Governorate, a semi-urbanized area in Lebanon. A total of 31 spring samples were collected during the wet and dry seasons and tested for water quality. Graphite furnace atomic absorption spectrometry was used to quantify Cd levels in water. Risk assessment was performed by means of a survey on water consumption and usage, and followed the USEPA Hazard Quotient (HQ) model. Correlations with different human activities were examined using different linear regression models and spatial analysis tools. Results highlight springs within El Metn and Keserwan Districts as posing a non-carcinogenic health risk in (1) Ain Saadeh both in adults ($HQ_{wet} = 1.94$) and iron-deficient children ($HQ_{wet} = 1.29$), and (2) Faraya both in healthy ($HQ_{wet} = 1.33$; $HQ_{dry} = 1.19$) and iron-deficient children ($HQ_{wet} = 2.67$; $HQ_{dry} = 2.37$), as well as in iron-deficient adults ($HQ_{wet} = 1.57$; $HQ_{dry} = 1.39$). In addition, Cd levels increased significantly with proximity to upstream open dumps (Pearson's $r = -0.511$; $p = 0.003$), agricultural land surface area, increased upstream industrial activity, mainly jewelry and fine stones industries in both bivariate ($r = 0.33$; $p = 0.001$) and multivariate analyses ($r = 0.41$; $p = 0.002$), plastic industries (Pearson's $r = 0.36$; $p = 0.05$), and furniture manufacturing (Pearson's $r = 0.38$; $p = 0.038$). Our findings constitute the first report assessing Cd health risk and identifying sources of pollution in Mount-Lebanon, and make a strong basis for a risk management strategy.

Keywords Cadmium · Health risk assessment · Water pollution · Heavy metals · Lebanon

Introduction

Cadmium (Cd) is a chemical of major public health concern on a global scale (Zang et al. 2018). It is utilized in various types of products and industries including batteries, pigments, photography, printing, dyed products, the metallurgic industry, and as a stabilizer for plastics (Stephen 2015). Cd pollution can be detected in water, air, and food. According to the USEPA, major sources of Cd contamination in water include corrosion of galvanized pipes, erosion of natural deposits, discharge from metal refineries, and run-off from waste batteries and paint products (EPA 2018b).

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Other important sources consist of anthropogenic activities that use phosphate fertilizers, fossil fuel combustion, and improper waste disposal and incineration (Morrow 2010). In addition, tobacco smoking is considered another source of Cd exposure in the general population (Mezynska and Brzoska 2018). Upon ingestion, up to 10% of Cd is absorbed by the intestine in healthy individuals while it can reach up to 20% in iron-deficient individuals (Silver et al. 2013; WHO 2011). Cd can accumulate in various organs and tissues, with more than half of the body burden concentrating in the kidney cortex and a half-life of 11–23 years (Ishizaki et al. 2015). Chronic Cd exposure leads to proximal tubular damage, renal anemia, low bone mineral density, osteoporosis, and increased risk of fractures (Cabral et al. 2015). In addition, experimental evidence and epidemiological studies show that Cd exposure is associated with an increased risk of renal, prostate, and lung cancer (Nawrot et al. 2015; Song et al. 2015), although no cancer slope factor (CSF) has been determined for Cd by ingestion so far (Ju-Kun et al. 2016; Song et al. 2015). Currently, The International Agency for Research on Cancer (IARC) classifies Cd in Group 1 as "Carcinogenic to Humans" (IARC 2018).

Health risk assessment studies for water sources have been conducted for a wide panel of agents, including nitrate, chromium, fluoride, and other hazardous substances, in different regions of the world (He and Wu 2018; Khemis et al. 2017; Li et al. 2014b, 2018); however, studies that examined Cd are still limited. Among those, one study assessed the health risk in 18 communities exposed to Cd from borehole drinking water near gold mining sites in Tarkwa, Ghana (Bortey-Sam et al. 2015). Another study, conducted in 5 different areas in Northwestern China, assessed the health risk for Cd in local children, from both tap and surface water (Wu et al. 2017). Similarly, a study in Southern China investigated the health risk for heavy metals, including Cd, from groundwater sources used for drinking (Peng et al. 2016). Results from these studies showed significant geographical variations in assessed levels of risk.

In Lebanon, an Eastern Mediterranean middle-income country, pollution of surface and groundwater have been associated with inappropriate practices in waste disposal, abuse of fertilizers and pesticides usage, industrial effluents and agricultural runoff, as well as excessive burning of fossil fuel (World Bank 2011). In addition to the country's persistent economic and political instability, the continuous influx of refugees from neighboring countries with armed conflict has increased pressure on the country's infrastructure (Cherri et al. 2016). This has aggravated misuse and mismanagement of Lebanon's natural resources, and thus amplified the country's environmental problems. In parallel, the recent solid waste crisis, which acquired wide media coverage in July 2015, has led to the emergence of over 900 dumps, where 77% of Lebanon's waste is either openly dumped or burned (Hilal et al. 2015). Previous studies have reported mobile forms of heavy metals including cadmium, chromium, zinc, and nickel, as contaminants of groundwater (Kassir et al. 2012). Yet, no studies have been carried out to assess the Cd health risk associated with drinking water in the country. Of particular interest are the springs of Mount-Lebanon Governorate, a major source of drinking and domestic water for more than 40% of the total Lebanese population (CAS 2009). Our Preliminary data had revealed water pollution in springs and water networks in that area. Therefore, in continuation of our earlier investigation, launched by the Associate Research Unit on Potable Water Quality and Management (ARU-PWQM) at the National Council for Scientific Research in Lebanon (LCNRS), the current study aims to (1) conduct a health risk assessment for Cd in Mount-Lebanon springs, and (2) determine the anthropogenic factors influencing Cd levels in water.

Materials and Methods

Study Area

The Mount-Lebanon-targeted study area is one of 8 Governorates in Lebanon, and is divided into 6 districts: Aley, Baabda, Chouf, Jbeil, Keserwan, and El Metn. It is a semi-urbanized region with a total surface area of 1984 Km^2 (OCHA 2014), and an estimated population of over 1.5 M spanning along the Mediterranean coast (MOPH 2014). The majority of trade and industrial activities take place in that area, which also includes 10% of the country's agricultural lands (OCHA 2014). In general, Mount-Lebanon has a predominant Mediterranean temperate climate with an average yearly precipitation of 800–1000 mm at 500 m altitude, and reaching up to 1400 mm at the higher altitudes (UNDP and MOEW 2016). The highest peak is located in the North at an elevation of more than 3000 m. The Mount-Lebanon region has an abrupt and rugged topography characterized by valleys and deep clefts, with steep slopes that form drainage basins for waterways and springs. Geologically, it mainly consists of Cretaceous formations (C2, C3, C4 and C5), and some Jurassic formations (J4) which have relatively high infiltration and groundwater flow rates (UNDP et al. 2014). Precipitation and snow melting are the two main sources of groundwater replenishment. Water in aquifers tends to flow along well-developed karstic underground channels at relatively high velocity, thus making water storage difficult (UNDP et al. 2014).

Water Sampling

A total of 409 springs in Lebanon were pre-screened by the ARU-PWQM earlier in 2009 in order to identify those used as a source of drinking water. Identified springs were then sampled and preliminarily examined first in 2011. In the present study, a second sampling campaign was conducted in 2015. Water samples were collected in triplicates from 31 identified water spring locations in Mount-Lebanon, with an altitude range of 47–1541 m (Fig. 1). Obtained replicates from each source were then combined to construct a 1L representative sample in High-Density Polyethylene (HDPE) plastic bottles, stored on site at 4 °C using a portable cooler, and maintained until analysis. HDPE bottles were used based on their inert chemical properties, thus preserving the quality of the samples (EPA 2009). GPS coordinates of each sample location were recorded. In order to account for seasonal variations, samples were collected both at the end of the wet and dry seasons (April and September 2015, respectively).

Water Quality and Cadmium Analysis

Laboratory analysis was conducted at the ARU-PWQM premises, located at the Faculty of Health Sciences of the American University of Beirut. Parameters tested during laboratory analysis were selected based on the WHO's standard protocol guidelines for drinking water quality analysis (WHO 2008). Laboratory testing included physical and chemical parameters, such as color, turbidity, electrical conductivity, total dissolved solids (TDS), pH, alkalinity, total hardness, chlorides, ammonia, nitrites, nitrates, sulfates, and sodium. Heavy metal analysis was also conducted for Cd, iron, manganese, nickel, copper, zinc, and lead (Pb). In addition, water microbial quality was examined using total and fecal coliform. Graphite furnace atomic absorption spectrometry (AAS Atomic Absorption Spectrometer, G95 graphite furnace, SOLAAR Thermofisher) was used to measure Cd and Pb concentrations in collected water samples. For Cd, the ash and atomization temperatures set during analysis were 600 °C and 1000 °C, respectively, and the emission wavelength was set to 326.1 nm. For Pb, the ash and atomization temperatures were 800 °C and 1200 °C, respectively, and the emission wavelength was 405.8 nm. The standard curve for Cd was constructed based on solutions of 0.6, 4.8, and 6 µg/L using a certified stock solution (TraceCERT®, 51994-250ML, Sigma-Aldrich Co. LLC; Darmstadt, Germany), and 20 µg of magnesium nitrate as a matrix modifier. The standard curve for Pb was based on solutions of 40, 80, 120, 160, and 200 µg/L also using a certified stock solution (TraceCERT®, 16595-250ML) in addition to 50 µg of magnesium nitrate as a matrix modifier.

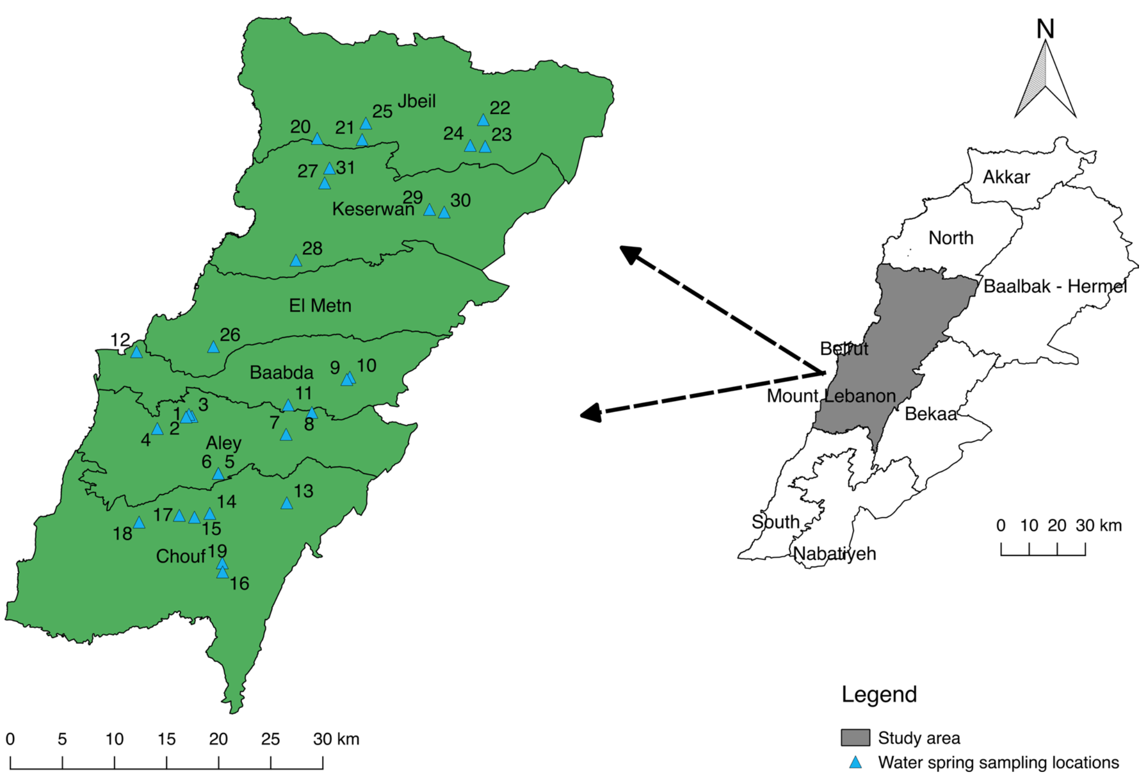

Fig. 1 Geographical distribution of 31 sampled water springs in the Mount-Lebanon Governorate

Quality Assurance and Control

All parameters were analyzed under standard conditions and using standard methods of water analysis following the American Public Health Association (APHA 2012; EPA 2018a; WHO 2008). The auto sampler was washed and purged prior to running blanks in order to avoid cross contamination. A blank was run for every batch of samples. All plastics used for analysis were rinsed twice with distilled water prior to usage. Measurements were conducted twice per sample for quality control, and the average concentration was reported.

Water Consumption Survey

After analyzing the water samples, areas with springs showing water Cd levels significantly exceeding international standards were surveyed in order to collect information on water usage and consumption rates. A systematic sampling method was adopted, where a total of 300 households—50 households per identified area—were consecutively selected from the White Pages and surveyed by phone. Surveying took place between March and November 2017 in native Arabic, using a structured questionnaire, in order to collect information on the following: (1) number of household occupants (including children under the age of 6 and adults) (2) patterns of usage and consumption rates of spring water (3) methods of spring water collection, and (4) adoption of domestic water treatment. An Institutional Review Board approval was obtained for the project by the ARU-PWQM.

Cd Health Risk Assessment

The health risk assessment was conducted based on the Agency for Toxic Substances and Disease Registry's Public Health Assessment Guideline Manual (ATSDR 2005) and as described (Li et al. 2014b); only exposure by ingestion was considered, as being the predominant one. After collecting consumption data for each area, the Chronic Daily Intake (CDI) was determined in µg/Kg/Day for both children and adults according to Eq. (1) (ATSDR 2005):

$$\mathrm{CDI} = \frac{(C_{\mathrm{water}} \times \mathrm{IR} \times \mathrm{CF} \times \mathrm{FI} \times \mathrm{ABS} \times \mathrm{EF} \times \mathrm{ED})}{\mathrm{BW} \times \mathrm{AT}}, \qquad (1)$$

where C_{water} is the average Cd concentration determined in drinking water (µg/L); IR is the average ingestion rate (L/day) estimated from the household survey; FI is the estimated fraction ingested from the contaminated source (ranging from 0.5 to 1); CF is the conversion factor (since all are expressed in the same unit, CF is equal to 1); ABS is the chemical-specific absorption from the GI tract (%) considered to be 10% for healthy individuals (ATSDR 2012) and 20% for iron-deficient individuals based on WHO guidelines (WHO 2011); EF is the exposure frequency (days/year) assumed to be 365 days/year; ED is the exposure duration assumed to be 70 years for adults and 6 years for children based on the ATSDR's Expo Fact Book (ATSDR 2005); BW is the body weight (in Kg) considered to be 70 for adults and 16 for children (ATSDR 2005); and AT is the averaging time (in days).

The CDI was determined for both children and adults in the following exposure scenarios: (a) subjects who drink only spring water, (b) subjects who have complementary sources of potable water, (c) subjects with proper nourishment both for (a) or (b), and (d) iron-deficient subjects both for (a) or (b) scenarios. The USEPA Hazard Quotient (HQ) was then computed to assess a non-carcinogenic health risk (Li et al. 2016; Wu and Sun 2016; Zhang et al. 2018), according to Eq. (2)

$$HQ = \frac{CDI}{RfD}, \qquad (2)$$

where CDI is the chronic daily intake, and RfD is the EPA Cd reference dose (0.5 µg/Kg/day) (IRIS 2008). An HQ above 1 indicates the existence of a health risk. In addition, the determined CDI was also compared to the Cd WHO Provisional Tolerable Monthly Intake (PTMI) of 25 µg/kg-BW/month (0.8 µg/Kg/day), and the Cd ATSDR Minimal Risk Level (MRL) of 0.1 µg/Kg/day (ATSDR 2012; WHO 2013).

Data Collection on Human Activities and Spatial Analysis

A list of all open dumps and waste sites, including data on open burning, year of establishment of dump, surface area and volume, and GPS coordinates in Mount-Lebanon, were obtained from the United Nations Development Program (UNDP), based on the 2017 report on "Updating of the Master Plan for The Closure and Rehabilitation of Uncontrolled Dumpsite throughout Lebanon" (UNDP and MOE 2017). In addition, a list of all relevant industries in Mount Lebanon was obtained from the Ministry of Industry, specifying types of industries and their GPS coordinates. The different industries were grouped by type of manufactured products into 9 categories: wood products, plastics, furniture, machinery and electrical appliances, jewelry and fine stone, metal products, tile and cement, paint and paper products, and hunting cartridges. At the same time, the latest available data of all agricultural activities in Mount Lebanon were obtained from the Lebanese Centre National des Recherches Scientifiques (LCNRS) based on a 2010 survey; agricultural areas were grouped by type of crops into 15 categories: banana trees, deciduous fruit trees, vineyards, citrus trees, olive trees, greenhouses, open fields, cultivation on large surfaces, cultivation on small surfaces, urban sprawl on (i) intensive croplands (ii) orchids (iii) open fields, poultry breeding, agricultural equipment, and abandoned agricultural lands. On the other hand, aquifer flow directions, in the form of GIS interface soft data, were obtained from the Earth Link & Advanced Resources Development Group (ELARD) based on a consent from UNDP (UNDP et al. 2014). Using QGIS (version 2.14 for windows), the different layers of gathered data described above were overlaid with the measured Cd levels for the different springs (Fig. 2). In order to identify potential sources of contamination, two different approaches were used. In the first, we made a justified assumption that an aquifer lying underneath a spring is its main feeding source, guided by aquifer water flow data and expert opinion. Data were then clustered based on the aquifer source. In this approach, all human activities, with the exception of agricultural lands, were labeled as “upstream” or “downstream” from a spring based on the aquifer flow direction and using QGIS; only upstream activities were included in the analysis, while all downstream activities were excluded. In rare cases where an aquifer did not have a clear direction, all human activities located on that aquifer were considered in the analysis. In the second approach, we determined all human activities within 2 km and 5 km radiuses from a given

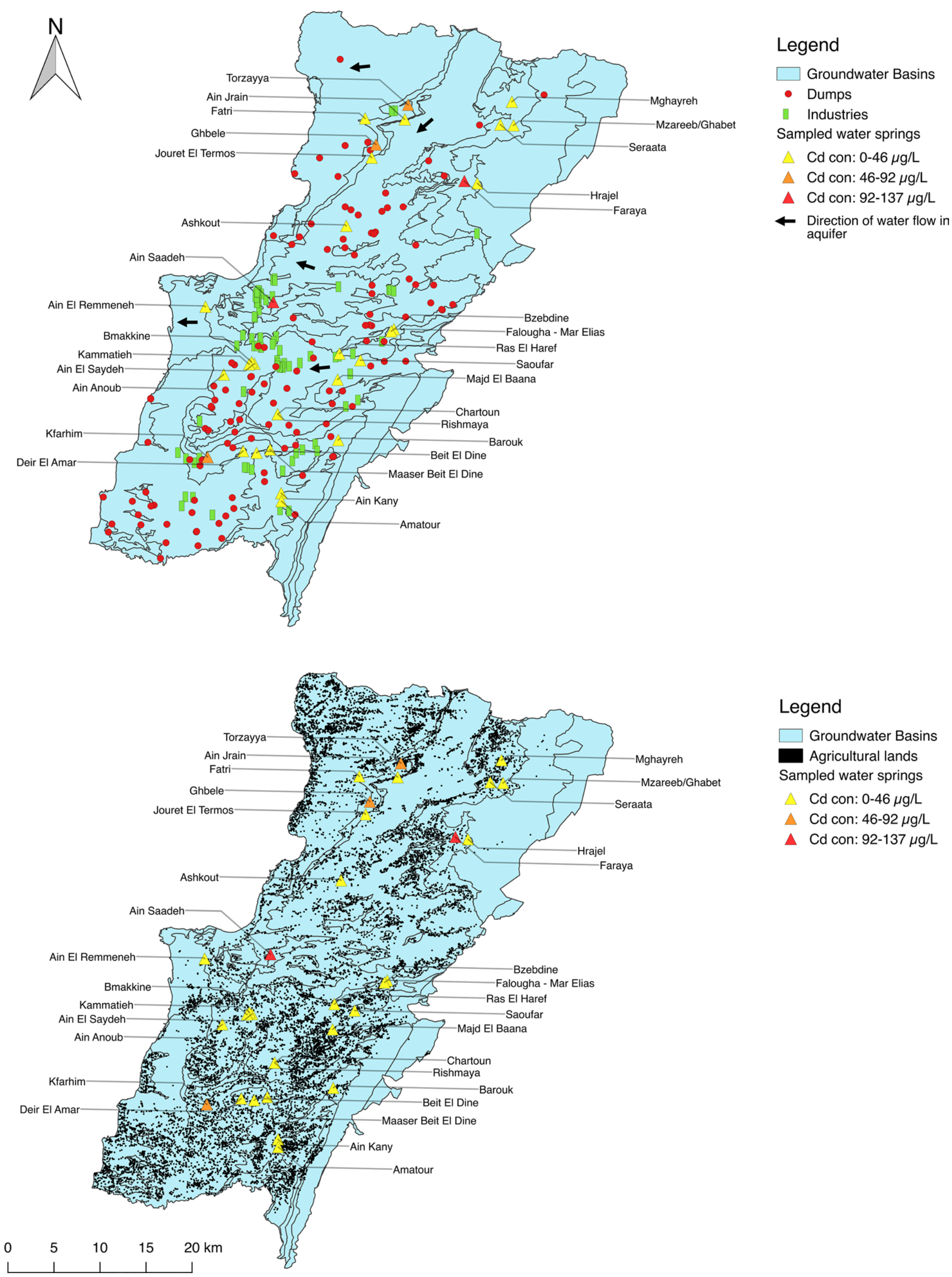

Fig. 2 Geographical distribution of human activities overlaid with measured water Cd concentrations in Mount-Lebanon Springs

spring, irrespective of the aquifer feeding source. In both approaches, data on human activity were merged in a comprehensive database in order to conduct statistical analyses. In addition, all waste sites having a year of establishment that does not precede the period of our water sampling campaign were excluded from the analysis.

Statistical Analysis

Statistical analysis was conducted using Stata/SE (windows version 11.2). For statistical analysis purposes, Cd levels below the limit of detection (LOD) were considered to be equal to LOD. Non-normality of the average Cd levels was treated by taking its natural logarithm; to avoid negative values, any Cd concentration below 1, and/or below detection limit, was considered to be equal to 1. In the first approach on potential sources of contamination described above, a bivariate linear regression with cluster analysis by aquifer was conducted for each of the upstream human activities. All variables that showed significance at the bivariate level were then entered in a multivariate linear regression model. In the second approach, correlations between Cd concentrations and upstream human activities, within 2 and 5 km radiuses, were examined using Pearson's r. Correlations with $p < 0.05$ were considered statistically significant.

Results

Water Physicochemical and Microbial Quality

Microbial and physicochemical parameters of collected water samples are summarized in Tables 1 and 2. Total coliform was present in water samples of 4 out of the 6 districts during the wet season and in all 6 of them during the dry season, whereas fecal coliform was present in 3 and 4 of the sampled districts during the wet and dry seasons, respectively. Chlorides, nitrates, nitrites, sulfates, iron, and zinc did not exceed the USEPA Maximum Contaminant Level (MCL) set for drinking water, whereas manganese's MCL was exceeded in 5 districts during the wet season and 4 during the dry season. In addition, all samples collected during wet and dry seasons exceeded the Pb MCL set by the USEPA at 15 µg/L. For Cd, the measured water concentrations ranged from 0.029 to 145.3 µg/L, and the limit of detection (LOD) was 0.029 µg/L (Table 3). During the wet season, more than 60% of the samples exceeded the MCL set at 5 µg/L, while 48% of the dry season samples exceeded that limit. All wet season samples obtained from El Metn and Keserwan springs exceeded MCL (100%), followed by Jbeil (83%), Baabda (67%), Chouf (43%), and Aley (28%). Dry season samples from El Metn were also all in excess of MCL, followed by Keserwan (60%), Baabda and Jbeil (50%), and Aley (38%). The highest Cd levels were found in Faraya springs (District of Keserwan) and persisted across both seasons. When examining the correlation between Pb and Cd levels in water, results were not statistically significant ($p = 0.401$).

Water Usage Survey

Fifty-five percent of surveyed households in Mount-Lebanon relied on springs as a source of drinking water. The average number of household occupants in the surveyed areas ranged between 3.9 and 5.0. The average spring water consumption rate ranged between 1.73 and 3.85 L/day for adults, and 0.36–1.21 L/day for children. Bzebdine water consumption rate was none due to an issued municipality warning following a microbial quality test. The frequency of households that used spring water was higher in Keserwan and Chouf (84%), followed by Jbeil (56%), Aley (54%), Baabda (34%), and lowest in El Metn (20%). In addition, households that used complementary sources of drinking water, other than the spring water, were 14% of the total surveyed sample. The main method of water spring transportation to households was the use of plastic gallons in all areas, except for the Chouf district, where a pipe network was reported for spring water (Table 4).

Cd Human Health Risk Assessment

When comparing computed CDI values to Cd international standards, the following can be highlighted. First, when compared to the PTMI (0.8 µg/Kg/day), children who may be iron deficient and drinking from Faraya springs as the only source of water, in both dry and wet season, had an excessive Cd exposure. Second, when compared to the MRL (0.1 µg/Kg/day), adults drinking from springs of Amatour, Mghayreh, Ain Saadeh, Faraya, and Gheble, during the wet season, had an excessive Cd exposure, whereas, during the dry season, adults drinking from springs in all the aforementioned towns (except Mghayreh), in addition to Kfarhim, Richmaya and Ain El Remmeneh, all had an excessive exposure, particularly for subjects that are iron-deficient and those that use the spring as the only source of potable water. Further, all iron-deficient children who drink from any of the springs in Richmaya, Amatour, Mghayreh, Ain Saadeh, Faraya, and Ghbele, during the wet season exceeded the Cd MRL, while children drinking from springs in Richmaya, Amatour, Ain Saadeh, Faraya, Ain Remmeneh and Kfarhim, during the dry season also exceeded the Cd MRL.

On the other hand, the computed HQ shows a health risk for both children and adults drinking from springs in Ain Saadeh and Faraya during both dry and wet seasons (Tables 5 and 6). The average Cd health risk distribution for the examined springs is illustrated in Fig. 3. The HQ

Table 1 Physicochemical and microbial quality of Mount-Lebanon springs during both wet and dry seasons in 2015

District & Season (# of Springs)	Total coliform/100 mL	Fecal coliform/100 mL	Chlorides mg/L as Cl^-	Ammonia mg/L as NH_3N	Nitrates mg/L as NO_3N	Nitrites mg/L as NO_2N	Sulfates mg/L as SO_4^{-2}	Sodium mg/L as Na^+
	Mean (SD)	Mean (SD)	Mean (SD)	Mean (SD)	Mean (SD)	Mean (SD)	Mean (SD)	Mean (SD)
Aley								
Wet season ($N=7$)	12.00 (18.97)	0.75 (2.12)	24.29 (9.8)	0.16 (0.02)	0.81 (0.47)	0.004 (0.001)	37.14 (34.6)	0.011 (0.0029)
Dry season ($N=8$)	16.33 (26.58)	11.86 (17.91)	13.13 (3.72)	0.14 (0.06)	1.34 (0.86)	0.004 (0.001)	30.38 (32.2)	0.011 (0.0037)
Baabda								
Wet season ($N=3$)	1.33 (2.31)	0 (0)	30 (5)	0.11 (0.04)	1.17 (1.42)	0.003 (0.001)	60.67 (57.14)	0.013 (0.0041)
Dry season ($N=4$)	13.25 (26.50)	6.75 (13.5)	12.5 (8.66)	0.07 (0.03)	1.1 (1.29)	5.25 (10.5)	35.78 (25.32)	0.012 (0.0057)
Chouf								
Wet season ($N=7$)	23.50 (47.36)	15.71 (25.42)	45.71 (19.24)	0.17 (0.02)	0.79 (0.60)	0.006 (0.002)	11.14 (13.18)	0.009 (0.0047)
Dry season ($N=7$)	11.29 (10.55)	2 (3)	18.57 (15.73)	0.13 (0.01)	0.76 (0.69)	0.001 (0.001)	11.43 (13.67)	0.008 (0.0049)
Jbeil								
Wet season ($N=6$)	35.00 (40.10)	16.83 (31.94)	21.67 (8.17)	0.02 (0.02)	0.98 (0.81)	0.004 (0.001)	15.33 (18.95)	0.008 (0.0054)
Dry season ($N=6$)	1.40 (2.07)	0 (0)	16.67 (9.31)	0.16 (0.01)	0.98 (1.03)	0.01 (0.001)	16.67 (18.95)	0.008 (0.0066)
El Metn								
Wet season ($N=1$)	0 (0)	0 (0)	25 (0)	0.01 (0)	2.4 (0)	0.006 (0)	52 (0)	0.015 (0)
Dry season ($N=1$)	6.00 (0)	0 (0)	32.5 (0)	0.13 (0)	2.7 (0)	ND	53 (0)	0.018 (0)
Keserwan								
Wet season ($N=4$)	0 (0)	0 (0)	13.75 (2.5)	0.15 (0.01)	1.25 (0.91)	0.005 (0.001)	8.75 (9.39)	0.008 (0.0043)
Dry season ($N=5$)	1.00 (2.00)	0.5 (1)	17 (8.37)	0.16 (0.06)	1.04 (0.84)	0.001 (0.001)	5.6 (5.68)	0.006 (0.0053)
Standard limit								
MCL (USEPA)			250		10	1	250	
GDWQ (WHO)			250	1.5	50	3	250	200

*Values figuring as "ND" indicate levels below LOD

positively correlated with water Cd levels ($r^2=0.751$) (Fig. 4); the two areas with springs that had the highest Cd levels (Faraya and Ain Saadeh) also exhibited the highest HQs in both adults and children. In addition, variation in spring water consumption had a significant impact on the identified risks. For instance, areas such as Amatour and Kfarhim, exhibited a higher HQ mainly due to the relatively higher average consumption of spring water, even though the water Cd levels were relatively lower. By the same token, in Ghbele, low water consumption conferred a lower risk even though water Cd levels were relatively higher than other sampled areas.

Potential Sources of Cd Pollution

Although QGIS did not reveal a visual pattern of association between Cd levels and human activities (Fig. 2), a number of indicators of polluting sources were identified. In the first approach using bivariate linear regression clustered by aquifer, Cd levels significantly increased with increased number of upstream jewelry and fine stones industries, increased surface area of neighboring agricultural lands, decreased median distance between spring and upstream waste dumps, and decreased median distance between spring and upstream industries (Table 7). Statistical significance was maintained

Table 2 Levels of heavy metals in Mount-Lebanon springs during both wet and dry seasons in 2015

District & Season (# of Springs)	Iron µg/L as Fe Mean (SD)	Manganese µg/L as Mn Mean (SD)	Lead µg/L as Pb Mean (SD)	Nickel µg/L as Ni Mean (SD)	Copper µg/L as Cu Mean (SD)	Zinc µg/L as Zn Mean (SD)
Aley						
Wet season ($N=7$)	40 (20)	60 (50)	70 (88)	ND	ND	35 (29)
Dry season ($N=8$)	20 (20)	60 (30)	20 (0.4)	ND	ND	13 (9)
Baabda						
Wet season ($N=3$)	30 (6)	90 (80)	20 (0.4)	ND	ND	58 (43)
Dry season ($N=4$)	10 (10)	40 (10)	21 (0.8)	ND	ND	37 (19)
Chouf						
Wet season ($N=7$)	43 (42)	37 (3)	20 (0.5)	ND	ND	70 (116)
Dry season ($N=7$)	9 (23)	73 (15)	20 (4)	ND	ND	10 (9)
Jbeil						
Wet season ($N=6$)	23 (20)	140 (75)	20 (0.9)	ND	ND	116 (240)
Dry season ($N=6$)	3 (10)	30 (10)	21 (0.5)	ND	ND	13 (9)
El Metn						
Wet season ($N=1$)	10 (0)	205 (0)	21 (0)	ND	ND	84 (0)
Dry season ($N=1$)	30 (0)	57 (0)	20 (0)	ND	ND	9 (0)
Keserwan						
Wet season ($N=4$)	28 (13)	60 (11)	20 (1)	ND	ND	40 (49)
Dry season ($N=5$)	64 (79)	51 (11)	18 (2.4)	ND	ND	1.5 (9)
Standard limit						
MCL (EPA)	300	50	15		1300	5000
GDWQ (WHO)	300	10–50	10	700	2000	3000

*Values figuring as “ND” indicate levels below LOD

at the multivariate analysis level for the number of upstream jewelry and fine stones industries ($p=0.002$) and the median distance between spring and upstream industries ($p=0.001$); In the second analysis approach, within a 2 km radius, Cd levels significantly increased with increased number of plastic industries and furniture manufacturers, and with decreased median distance between spring and agricultural lands. Within a 5 km radius, the median distance between spring and waste dumps was also significantly associated with Cd levels ($p=0.003$) (Table 7).

Discussion

Contamination of water bodies is one of the most alarming contemporary challenges (WWAP 2017). Groundwater is considered susceptible to human activities, particularly in environmentally degrading societies (Khatri and Tyagi 2015; Li et al. 2017; Morris et al. 2003). Out of 5050 springs across Lebanon, only 409 are considered to be major sources of water for drinking and domestic use, based on their flow discharge (UNDP et al. 2014). In this study, we examined water quality in 31 springs distributed in 6 districts across Mount-Lebanon. Our results highlight, on one hand, fecal coliform in a number of springs, indicating potential human sewage contamination, and on the other hand, an alarming widespread of heavy metals contamination, particularly for Cd and Pb. Heavy metals are considered contaminants of potential concern in groundwater, primarily from human activities, as well as from natural processes (Ikem et al. 2003; Li et al. 2014a). Previous studies that examined heavy metals reported much lower levels for Cd in water bodies in various other countries. For instance, in Pakistan and Iran, the average Cd concentrations in groundwater were found to be 6.25 and 45 µg/L, respectively (Arain et al. 2015; Ebrahimi et al. 2010). Another study conducted in Ghana found Cd concentrations to range from 16 to 92 µg/L in groundwater (Affum et al. 2015). Further, Cd levels were found to be within WHO limits in Irbid, Jordan, a neighboring country in the Eastern Mediterranean region (Alomary 2013). Hence, the levels of Cd found in Mount-Lebanon springs (up to 145.3 µg/L) suggest an alarming globally high water Cd pollution. The observed seasonal variations in water quality may be explained by runoff water in the wet season (Wilson 2018). This is supported by the fact that the fractures and karst formation of aquifers in Lebanon facilitate the percolation of contaminants carried by runoff into groundwater, making aquifers prone and vulnerable to pollution (DAR-IAURIF 2005). The observed patterns

Table 3 Cd levels mapping for sampled Mount-Lebanon springs during both wet and dry seasons in 2015

District	Spring (corresponding # in Fig. 1)	Cd Concentrations (μg/L)	
		Wet season	Dry season
Aley	Bmakkine (#1)	–	6.66
	Kammatieh (#2)	41.95	0.029
	Ain El Saydeh (#3)	0.029	19.86
	Ain Anoub (#4)	0.029	4.52
	Rishmaya (#5)	4.53	17.63
	Chartoun (#6)	0.029	4.01
	Majd el Baana (#7)	81.32	1.22
	Saoufar (#8)	0.029	1.21
	District mean (SD)	18.27 (31.77)	6.89 (7.64)
Baabda	Falougha – Mar Elias (#9)	33.75	0.43
	Bzebdine (#10)	9.73	25.40
	Ras El Haref (#11)	0.029	0.74
	Ain El Remmeneh (#12)	–	22.34
	District mean (SD)	14.50 (17.36)	12.23 (13.50)
Chouf	Barouk (#13)	1.77	14.94
	Maaser Beit El Dine (#14)	18.41	0.029
	Beit El Dine (#15)	18.13	2.05
	Amatour (#16)	15.45	15.02
	Deir El Amar (#17)	0.75	3.53
	Kfarhim (#18)	1.69	112.29
	Ain Kany (#19)	1.70	0.029
	District mean (SD)	8.27 (8.53)	21.13 (40.72)
Jbeil	Fatri (#20)	8.36	9.95
	Ain Jrain (#21)	0.029	0.89
	Mghayreh (#22)	41.23	9.09
	Mzareeb/Ghabet (#23)	23.60	0.029
	Seraata (#24)	10.07	4.98
	Torzayya (#25)	137.96	5.71
	District mean (SD)	36.86 (51.58)	5.11 (4.08)
El Metn	Ain Saadeh (#26)	129.36	77.55
	District mean (SD)	129.36 (0)	77.55 (0)
Keserwan	Jouret El Termos (#27)	–	4.77
	Ashkout (#28)	29.36	4.60
	Faraya (#29)	145.30	129.22
	Hrajel (#30)	64.19	8.84
	Ghbele (#31)	59.28	64.42
	District mean (SD)	74.53 (49.63)	42.37 (54.76)
Standard Limit			
Cd MCL (USEPA)		5 μg/L	
Cd GDWQ (WHO)		3 μg/L	

showing increased Cd levels during the wet season may be attributed to site-specific activities, source of wastes, and velocity of water flow (Islam et al. 2015). At the same time, polluted sediment particles may be suspended in the bottom layer during the dry season, which may cause lower heavy metal concentrations (Islam et al. 2015). The water pH can also impact Cd solubility in water and may contribute to the observed seasonal variations.

Natural population growth, influx of refugees escaping the Syrian conflict, progressive urbanization, and the wide skepticism about the quality of tap water are all factors putting pressure on groundwater reservoirs in Lebanon as the primary drinking water resource. In this study, HQ values for Cd were consistently above 1 across seasons for various users of Ain Saadeh and Faraya water springs, in the districts of El Metn and Keserwan, respectively, hence

Table 4 Water consumption patterns in surveyed households in the districts of Mount Lebanon in 2017, ($N=300$ households)

District	Average number of occupants per household	Average consumption of spring water for an adult per household per day (in Liters)	Average consumption of spring water for a child per household per day (in Liters)	Households that utilize spring water for drinking purposes	Households that utilize additional sources of water	Households that use spring water for washing of food	Methods of transporting Spring Water		Households that domestically treat spring water
							Gallons & Buckets	Network	
Aley ($n=50$)	4.11	1.92	1.21	27 (54%)	8 (16%)	23 (46%)	20 (40%)	7 (14%)	4 (8%)
Baabda ($n=50$)	3.94	2.08	0.40	17 (34%)	7 (14%)	17 (34%)	9 (18%)	8 (16%)	8 (16%)
Chouf ($n=50$)	4.52	3.85	0.99	42 (84%)	3 (6%)	39 (78%)	7 (14%)	35 (70%)	8 (16%)
Jbeil ($n=50$)	4.76	1.93	0.36	28 (56%)	7 (14%)	28 (56%)	15 (30%)	13 (26%)	2 (4%)
El Metn ($n=50$)	4.3	2.62	0.40	10 (20%)	1 (2%)	8 (16%)	7 (14%)	3 (6%)	2 (4%)
Keserwan ($n=50$)	5.04	1.73	0.45	42 (84%)	15 (30%)	42 (84%)	27 (54%)	15 (30%)	4 (8%)
Total ($N=300$)	4.52	2.10	0.66	166 (55%)	41 (14%)	157 (52%)	85 (28%)	81 (27%)	28 (9%)

indicating a persistent health risk to residents and water consumers in that area. The computed HQ values attributed to Cd in water for both adults and children residing in most of the areas selected in this study were higher than values reported in the literature in different parts of the world. For instance, a study conducted in Xi'an and Xining in China, reported the absence of a health risk ($HQ=3\times10^{-4}$) in residents exposed to Cd in drinking water (Wu et al. 2017). Another study conducted in Southern China also reported no health risk in Cd-exposed residents ($HQ=0.570$) (Peng et al. 2016). Similar results were reported in an exposed African population in Tarkwa, Ghana, with an HQ range of $(0.2–18)\times10^{-3}$ (Bortey-Sam et al. 2015). Therefore, our findings call for an intervention to mitigate the identified risk. One study estimates that a population with an average daily Cd intake of 30 µg/day (i.e., 0.43 µg/kg/day) will suffer from an increased renal damage of 1%, and up to 5% in vulnerable groups (Jarup et al. 1998). Given that the estimated Cd exposure from water consumption exceeds 0.43 µg/kg/day in many Mount-Lebanon towns, and knowing that the USEPA Cd RfD is 0.5 µg/kg/day, together, these imply a higher risk of renal damage in exposed residents, particularly in Ain Saadeh and Faraya. The risk may be higher in women due to higher absorption rates (Nishijo et al. 2017). Even though a number of surveyed residents reported that they treat the water domestically, however, the employed methods—boiling, exposure to sunlight, and usage of crude filters—cannot remove dissolved heavy metals, and hence have no impact on the estimated risk. The same applies to spring water transported through networks, as in the case of Chouf and Aley districts. In Aley specifically, although spring water consumption is much more common, predominantly among residing groups that seek "natural water" and boycott tap water based on religious and cultural beliefs, we did not detect a Cd health risk in that district; this is mainly because Cd water levels were relatively lower compared to levels in other districts in El Metn and Keserwan. However, these "natural water seekers" in particular may be at a higher risk in the future if Cd levels continue to rise in springs.

On the other hand, when examining sources of Cd pollution, a number of points need to be emphasized. First, heavy metals pollution did not correlate with fecal coliform, hence suggesting that improper discharge of wastewater cannot explain the observed surge in springs' heavy metals (Chakarvorty et al. 2015). In addition, the fact that Pb contamination levels do not correlate with Cd levels suggests different sources of contamination that needs further investigation. Second, the inverse correlation between Cd levels and spring-to-dump median distance—identified using two different analytical models—is the first indicator to show a significant relationship between the Cd surge in Mount-Lebanon springs and the solid waste crisis. Mount-Lebanon has witnessed a fast and chaotic rise of dumpsites since

Table 5 Cd health risk assessment for adults in springs during the wet and dry seasons in Mount-Lebanon

District	Spring	HQ							
		Wet season				Dry season			
		Spring water only		With complementary water sources		Spring water only		With complementary water sources	
		Healthy individual	Iron-deficient individual	Healthy individual	Iron-deficient individual	Healthy individual	Iron-deficient individual	Healthy individual	Iron-deficient individual
Aley	Ain Saydeh	0.0001	0.0003	0.0001	0.0001	0.09	0.18	0.05	0.09
	Rishmaya	0.03	0.06	0.01	0.03	0.11	0.23	0.06	0.11
Baabda	Bzebdine	0	0	0	0	0	0	0	0
	Ain El Remmeneh	–	–	–	–	0.13	0.27	0.07	0.13
Chouf	Barouk	0.01	0.01	0.003	0.01	0.06	0.11	0.03	0.06
	Amatour	0.42	0.84	0.21	0.42	0.41	0.82	0.2	0.41
	Kfarhim	0	0.01	0.002	0.003	0.23	0.46	0.11	0.23
Jbeil	Fatri	0.05	0.1	0.03	0.05	0.06	0.12	0.03	0.06
	Mghayreh	0.2	0.4	0.1	0.2	0.04	0.09	0.02	0.04
El Metn	Ain Saadeh	0.97	1.94*	0.48	0.97	0.58	1.16*	0.29	0.58
Keserwan	Faraya	0.78	1.57*	0.39	0.78	0.7	1.39*	0.35	0.7
	Ghbele	0.27	0.53	0.13	0.27	0.29	0.58	0.14	0.29

*Indicating a health risk

Table 6 Cd health risk assessment for children in springs during the wet and dry seasons in Mount-Lebanon

District	Spring	HQ							
		Wet season				Dry season			
		Spring water only		With complementary water sources		Spring water only		With complementary water sources	
		Healthy individual	Iron-deficient individual	Healthy individual	Iron-deficient individual	Healthy individual	Iron-deficient individual	Healthy individual	Iron-deficient individual
Aley	Ain Saydeh	0.0002	0.0003	0.0001	0.0002	0.11	0.23	0.06	0.11
	Rishmaya	0.11	0.22	0.06	0.11	0.43	0.86	0.22	0.43
Baabda	Bzebdine	0	0	0	0	0	0	0	0
	Ain El Remmeneh	–	–	–	–	0.11	0.22	0.06	0.11
Chouf	Barouk	0.01	0.02	0.01	0.01	0.09	0.19	0.05	0.09
	Amatour	0.38	0.76	0.19	0.38	0.37	0.73	0.18	0.37
	Kfarhim	0.01	0.02	0.01	0.01	0.7	1.4	0.35	0.7
Jbeil	Fatri	0.03	0.06	0.02	0.03	0.04	0.07	0.02	0.04
	Mghayreh	0.22	0.44	0.11	0.22	0.05	0.1	0.02	0.05
El Metn	Ain Saadeh	0.65	1.29*	0.32	0.65	0.39	0.78	0.19	0.39
Keserwan	Faraya	1.33*	2.67*	0.67	1.33*	1.19*	2.37*	0.59	1.19*
	Ghbele	0.12	0.24	0.06	0.12	0.13	0.26	0.06	0.13

*Indicating a health risk

2011, which may explain our results. This is also supported by similar findings in Ghana, where groundwater Cd levels were associated with leachates from open dumpsites and septic tanks (Affum et al. 2015). Similar findings were also reported in North-East India, in which a significant association was identified between Cd water levels and proximity

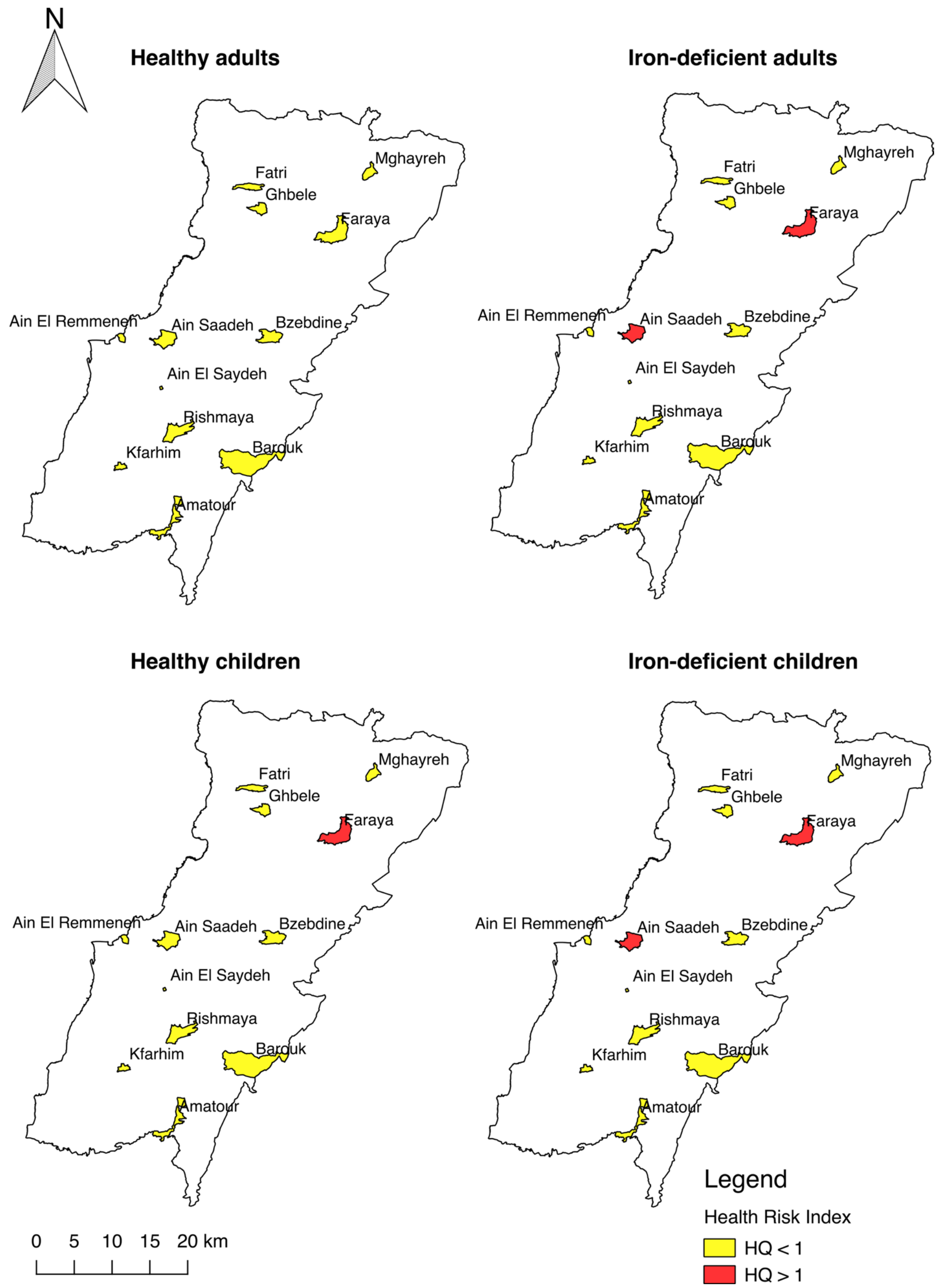

Fig. 3 Geographical distribution of Cd health risk associated with water springs when used as the sole source of drinking water

to dumpsites (Choudhury and Gupta 2017). Third, the study also showed that increased upstream industrial activity, particularly jewelry and fine stones, plastic industries, as well as furniture manufacturing, significantly impact spring Cd levels. Previous studies have reported that Cd concentrations were higher in samples taken at a close proximity to similar industries (Bhagure and Mirgane 2011; Khlifi et al. 2014; Yu et al. 2010). In addition, agricultural activity seems be an important contributor to Cd water pollution. Studies have shown that the use of phosphate fertilizers is an important contributor to Cd soil and water pollution (Yan et al. 2015); however, in our context, this remains highly speculative since we were unable to find any data on the use of such fertilizers in the country.

This study may be limited by a number of factors. First, exposure may be underestimated due to other sources of human Cd exposure such as contaminated food, polluted air near industrial sites, and through smoking. This is important considering that 50–60% of the Lebanese population is estimated to smoke either cigarettes or water-pipe tobacco (Salti et al. 2015). Second, this study did not gather medical information from surveyed household

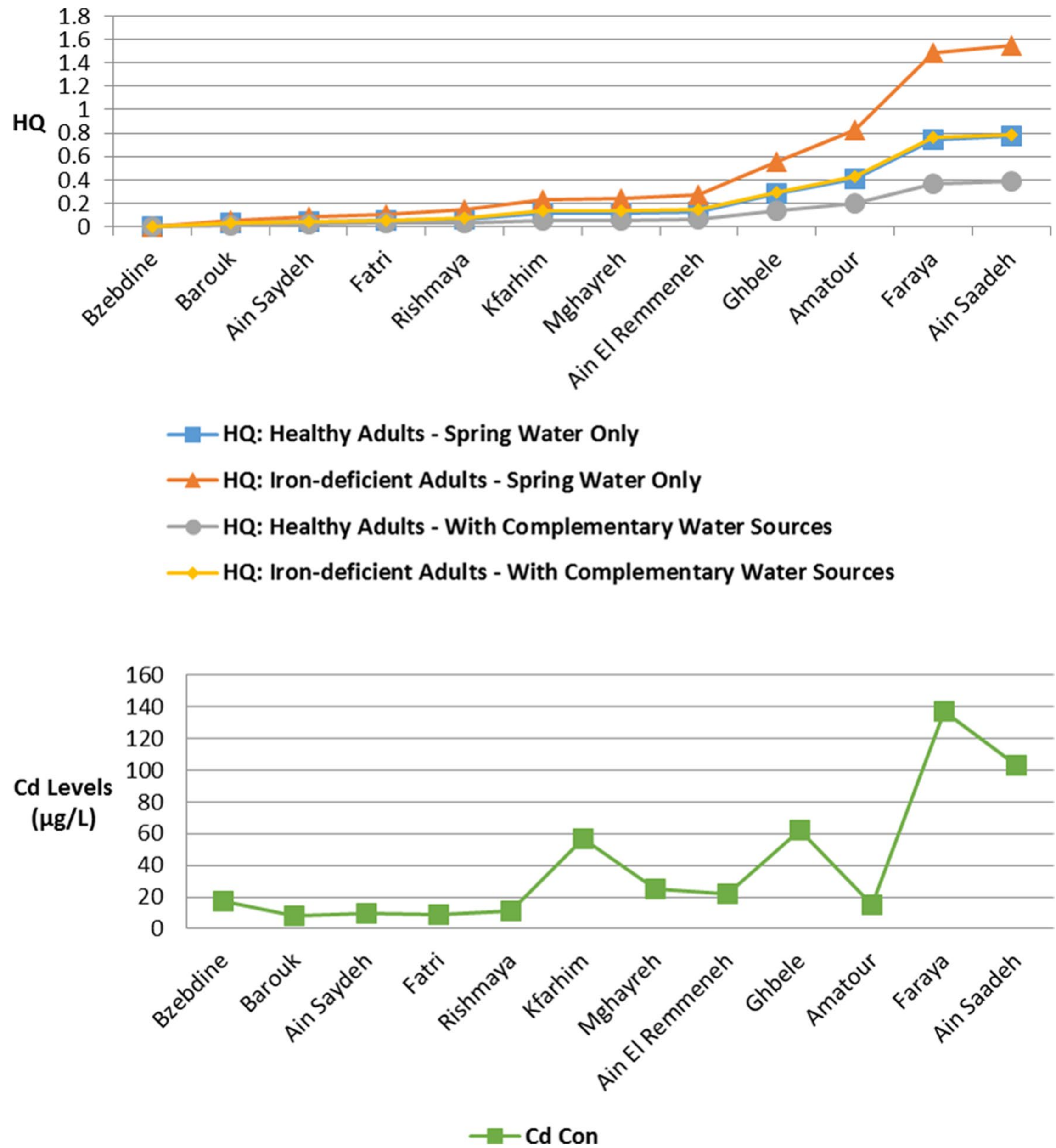

Fig. 4 Correlations between the Hazard Quotient (HQ) indicating health risk and Cd water levels in Mount-Lebanon Springs

residents in order to examine a correlation between spring water Cd exposure levels and putative adverse effects. Further research in this area is needed in the future. Third, used analysis models, particularly those employed in identifying sources of contamination, may be biased by data outliers and model overfitting. We tried to overcome these challenges by pre-cleaning the data and normalizing the outcome variable. Another major challenge that we overcame in this study was identifying and collecting geocoded data on various human activities, as well as connecting with communities and local authorities in order to identify patterns of water usage and quantify consumption in the different districts and towns of Mount-Lebanon.

Conclusion

Monitoring heavy metal pollution in reservoirs of Mount-Lebanon is of great importance in preserving a reliable drinking water source for residents. Our findings indicate widespread Cd and Pb pollution, as well as fecal contamination, with great spatial and seasonal variations among

Table 7 Significant indicators of sources of Cd pollution in Mount-Lebanon springs

Cluster Analysis	Indicators	Regression coefficient	*P* value	CI (95%)
Bivariate analysis	Median area of agricultural land	0.0004065	0.033	0.0000551, 0.0007579
	Median distance between spring and dumps	−0.0000557	0.007	−0.0000865, −0.0000249
	Median distance between spring and industries	−0.0000609	0.001	−0.000081, −0.0000408
	Number jewelry and fine stones industries	0.3328866	0.001	0.2174987, 0.4482744
Multivariate analysis	Number jewelry and fine stones industries	0.4079081	0.002	0.2506252, 0.5651909
	Median distance between spring and industries	−0.0000551	0.001	−0.0000711, −0.0000391

Analysis by Radius	Pearson's r	*P*-value
Within 2 km radius		
Number of plastic industries	0.355	0.05
Number of industries that manufacture furniture	0.375	0.038
Median distance between spring and agricultural land	−0.613	0.001
Within 5 km radius		
Median distance between spring and dumps	−0.511	0.003

Mount-Lebanon springs. Analysis of anthropogenic sources suggests a contribution of multiple sources to Cd pollution, particularly open dumps and industrial activity. Our results establish valuable baseline data that will help in prioritizing areas for groundwater sustainability and water safety. In addition, an unusually globally elevated Cd health risk was found for residents of El Metn and Keserwan Districts, particularly in Ain Saadeh and Faraya where the hazard quotient exceeded 1 for both children and adults, mostly influenced by the high Cd levels in the water. Globally, several studies have reported a positive association between human Cd ingestion of similar magnitude to those we found and higher renal damage risk, which calls for further epidemiological investigation. Our alarming findings on prevalent Cd health risks should trigger a response by local authorities and concerned ministries, leading into an effective risk management plan, which may include restrictions on land use and access, a national drinking water advisory, and an elaborate spring water monitoring plan for Mount-Lebanon.

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References

Affum AO, Osae SD, Nyarko BJ, Afful S, Fianko JR, Akiti TT, Adomako D, Acquaah SO, Dorleku M, Antoh E, Barnes F, Affum EA (2015) Total coliforms, arsenic and cadmium exposure through drinking water in the Western Region of Ghana: application of multivariate statistical technique to groundwater quality. Environ Monit Assess 187:1. https://doi.org/10.1007/s10661-014-4167-x

Alomary A (2013) Determination of trace metals in drinking water in Irbid City-Northern Jordan. Environ Monit Assess 185:1969–1975. https://doi.org/10.1007/s10661-012-2680-3

APHA (American Public Health Association), AWWA (American Water Works Association), WEF (Water Environment Federation) (2012) Standard methods for the examination of water and wastewater, 22nd edn. American Public Health Association, Washington, DC

Arain SS, Kazi TG, Afridi HI, Brahman KD, Naeemullah Khan S, Panhwar AH, Kamboh MA, Memon JR (2015) Erratum to: preconcentration and determination of lead and cadmium levels in blood samples of adolescent workers consuming smokeless tobacco products in Pakistan. Environ Monit Assess 187:538. https://doi.org/10.1007/s10661-015-4724-y

ATSDR (2005) Public health assessment guidance manual. Atlanta, Georgia

ATSDR (2012) Toxicological profile for Cadmium. https://www.atsdr.cdc.gov/toxprofiles/tp5.pdf. Accessed 28 May 2018

Bhagure GR, Mirgane SR (2011) Heavy metal concentrations in groundwaters and soils of Thane Region of Maharashtra, India. Environ Monit Assess 173:643–652. https://doi.org/10.1007/s10661-010-1412-9

Bortey-Sam N, Nakayama SMM, Ikenaka Y, Akoto O, Baidoo E, Mizukawa H, Ishizuka M (2015) Health risk assessment of heavy metals and metalloid in drinking water from communities near gold mines in Tarkwa, Ghana. Environ Monit Assess 187:397. https://doi.org/10.1007/s10661-015-4630-3

Cabral M, Toure A, Garcon G, Diop C, Bouhsina S, Dewaele D, Cazier F, Courcot D, Tall-Dia A, Shirali P, Diouf A, Fall M, Verdin A (2015) Effects of environmental cadmium and lead exposure on adults neighboring a discharge: evidences of adverse health effects. Environ Pollut 206:247–255. https://doi.org/10.1016/j.envpol.2015.06.032

CAS (2009) Population and Housing in Lebanon. Central Administration of Statistics. http://www.cas.gov.lb/index.php/demographic-and-social-en/population-en. Accessed 19 June 2018

Chakarvorty M, Dwivedi AK, Shukla AD, Kumar S, Niyogi A, Usmani M, Pati JK (2015) Geochemistry and magnetic measurements of suspended sediment in urban sewage water vis-a-vis quantification of heavy metal pollution in Ganga and Yamuna Rivers, India.

Environ Monit Assess 187:604. https://doi.org/10.1007/s10661-015-4794-x

Cherri Z, Arcos Gonzalez P, Castro Delgado R (2016) The Lebanese-Syrian crisis: impact of influx of Syrian refugees to an already weak state. Risk Manag Healthc Policy 9:165–172. https://doi.org/10.2147/RMHP.S106068

Choudhury D, Gupta S (2017) Impact of waste dump on surface water quality and aquatic insect diversity of Deepor Beel (Ramsar site), Assam, North-east India. Environ Monit Assess 189:540. https://doi.org/10.1007/s10661-017-6233-7

DAR-IAURIF (Dar-al-Handasah and the Institut d'Amenagement et d'Urbanisme de la Region Ile-de-France) (2005) National Physical Master Plan of The Lebanese Territory. http://www.cdr.gov.lb/study/sdatl/English/NPMPLT.PDF. Accessed 15 June 2018

Ebrahimi A, Amin MM, Hashemi H, Foladifard R, Vahiddastjerdi M (2010) A survey of groundwater chemical quality in Sajad Zarinshahr. Health Syst Res 6:918–926

He S, Wu J (2018) Hydrogeochemical characteristics, groundwater quality, and health risks from hexavalent chromium and nitrate in groundwater of huanhe formation in Wuqi County. Expo Health, Northwest China. https://doi.org/10.1007/s12403-018-0289-7

Hilal N, Fadlallah R, Jamal D, El-Jardali F (2015) K2P Evidence Summary: Approaching the Waste Crisis in Lebanon: Consequences and Insights into Solutions Knowledge to Policy (K2P) Center. https://www.aub.edu.lb/k2p/Documents/K2P%20Evidence%20Summary%20Waste%20Management_Final_%20Dec%2014%202015.pdf. Accessed 01 Dec 2018

IARC (2018) Monographs on the Evaluation of the Carcinogenic Risk of Chemicals to Humans. International Agency for Research on Cancer. https://monographs.iarc.fr/wp-content/uploads/2018/07/List_of_Classifications.pdf. Accessed 29 July 2018

Ikem A, Egiebor NO, Nyavor K (2003) Trace elements in water, fish and sediment from Tuskegee Lake, Southeastern USA. Water Air Soil Pollut 149:51–75

IRIS (Integrated Risk Information System) (2008) Cadmium Compounds A. https://www.epa.gov/sites/production/files/2016-09/documents/cadmium-compounds.pdf. Accessed 20 July 2018

Ishizaki M, Suwazono Y, Kido T, Nishijo M, Honda R, Kobayashi E, Nogawa K, Nakagawa H (2015) Estimation of biological half-life of urinary cadmium in inhabitants after cessation of environmental cadmium pollution using a mixed linear model. Food Addit Contam A 32:1273–1276. https://doi.org/10.1080/19440049.2015.1052573

Islam SM, Uddin KM, Tareq, Shammi M, Kamal KA, Sugano T, Kurasaki M, Saito T, Tanaka S, Kuramitz H (2015) Alteration of Water Pollution Level with the Seasonal Changes in Mean Daily Discharge in Three Main Rivers around Dhaka City, Bangladesh. Environments. https://doi.org/10.3390/environments2030280

Jarup L, Berglund M, Elinder CG, Nordberg G, Vahter M (1998) Health effects of cadmium exposure–a review of the literature and a risk estimate. Scand J Work Environ Health 24(Suppl 1):1–51

Ju-Kun S, Yuan DB, Rao HF, Chen TF, Luan BS, Xu XM, Jiang FN, Zhong WD, Zhu JG (2016) Association between Cd exposure and risk of prostate cancer: a PRISMA-compliant systematic review and meta-analysis. Med (Baltimore) 95:e2708. https://doi.org/10.1097/MD.0000000000002708

Kassir LN, Lartiges B, Ouaini N (2012) Effects of fertilizer industry emissions on local soil contamination: a case study of a phosphate plant on the east Mediterranean coast. Environ Technol 33:873–885. https://doi.org/10.1080/09593330.2011.601765

Khatri N, Tyagi S (2015) Influences of natural and anthropogenic factors on surface and groundwater quality in rural and urban areas. Front Life Sci 8:23–39. https://doi.org/10.1080/21553769.2014.933716

Khemis IB, Besbes Aridh N, Hamza N, M'Hetli M, Sadok S (2017) Heavy metals and minerals contents in pikeperch (Sander lucioperca), carp (Cyprinus carpio) and flathead grey mullet (Mugil cephalus) from Sidi Salem Reservoir (Tunisia): health risk assessment related to fish consumption. Environ Sci Pollut Res Int 24:19494–19507. https://doi.org/10.1007/s11356-017-9586-0

Khlifi R, Olmedo P, Gil F, Feki-Tounsi M, Hammami B, Rebai A, Hamza-Chaffai A (2014) Biomonitoring of cadmium, chromium, nickel and arsenic in general population living near mining and active industrial areas in Southern Tunisia. Environ Monit Assess 186:761–779. https://doi.org/10.1007/s10661-013-3415-9

Li P, Qian H, Howard K, Wu J (2014a) Heavy metal contamination of Yellow River alluvial sediments, northwest China. Environ Earth Sci. https://doi.org/10.1007/s12665-014-3628-4

Li P, Wu J, Qian H, Lyu X, Liu H (2014b) Origin and assessment of groundwater pollution and associated health risk: a case study in an industrial park, northwest China. Environ Geochem Health 36:693–712. https://doi.org/10.1007/s10653-013-9590-3

Li P, Li X, Meng X, Li M, Zhang Y (2016) Appraising groundwater quality and health risks from contamination in a semiarid region of northwest China. Expo Health 8:361–379. https://doi.org/10.1007/s12403-016-0205-y

Li P, Tian R, Xue C, Wu J (2017) Progress, opportunities, and key fields for groundwater quality research under the impacts of human activities in China with a special focus on western China. Environ Sci Pollut Res. https://doi.org/10.1007/s11356-017-8753-7

Li P, He X, Li Y, Xiang G (2018) Occurrence and health implication of fluoride in groundwater of loess aquifer in the Chinese Loess Plateau: a case study of Tongchuan. Expo Health. https://doi.org/10.1007/s12403-018-0278-x

Mezynska M, Brzoska MM (2018) Environmental exposure to cadmium-a risk for health of the general population in industrialized countries and preventive strategies. Environ Sci Pollut Res Int 25:3211–3232. https://doi.org/10.1007/s11356-017-0827-z

MOPH (Lebanese Ministry of Public Health) (2014) Statistical bulletin 2014. https://www.moph.gov.lb/en/DynamicPages/page_download_file/1611. Accessed 20 June 2018

Morris BL, Lawrence ARL, Chilton PJC, Adams B, Calow RC, Klinck BA (2003) Groundwater and its susceptibility to degradation: a global assessment of the problem and options for management. Early warning and assessment report series, RS. 03-3. UNEP (United Nations Environment Programme)

Morrow H (2010) Cadmium and Cadmium Alloys. In: Suslick KS (ed) Kirk-Othmer encyclopedia of chemical technology. Wiley, New York

Nawrot TS, Martens DS, Hara A, Plusquin M, Vangronsveld J, Roels HA, Staessen JA (2015) Association of total cancer and lung cancer with environmental exposure to cadmium: the meta-analytical evidence. Cancer Causes Control 26:1281–1288. https://doi.org/10.1007/s10552-015-0621-5

Nishijo M, Nakagawa H, Suwazono Y, Nogawa K, Kido T (2017) Causes of death in patients with Itai-itai disease suffering from severe chronic cadmium poisoning: a nested case-control analysis of a follow-up study in Japan. BMJ Open 7:e015694. https://doi.org/10.1136/bmjopen-2016-015694

OCHA (Office for the Coordination of Humanitarian Affairs) (2014) Beirut and Mount-Lebanon Governorate Profile. https://reliefweb.int/sites/reliefweb.int/files/resources/20140811-Beirut%20MountLebanonGovernorateProfile.pdf. Accessed 19 June 2018

Peng C, Cai Y, Wang T, Xiao R, Chen W (2016) Regional probabilistic risk assessment of heavy metals in different environmental media and land uses: an urbanization-affected drinking water supply area. Sci Rep 6:37084. https://doi.org/10.1038/srep37084

Salti N, Chaaban J, Nakkash R, Alaouie H (2015) The effect of taxation on tobacco consumption and public revenues in Lebanon. Tobacco Control 24:77–81. https://doi.org/10.1136/tobaccocontrol-2012-050703

Silver MK, Lozoff B, Meeker JD (2013) Blood cadmium is elevated in iron deficient US children: a cross-sectional study. Environ Health-Glob. https://doi.org/10.1186/1476-069x-12-117
Song J, Luo H, Yin X, Huang G, Luo S, du Lin R, Yuan DB, Zhang W, Zhu J (2015) Association between cadmium exposure and renal cancer risk: a meta-analysis of observational studies. Sci Rep 5:17976. https://doi.org/10.1038/srep17976
Stephen FD (2015) Cadmium. In: Hamilton RD (ed) Hamilton & Hardy's industrial toxicology. Wiley, Hoboken
UNDP, MOE (Lebanese Ministry of Environment) (2017) Updated master plan for the closure and rehabilitation of uncontrolled dumpsites throughout the country of Lebanon, Vol A. http://www.lb.undp.org/content/lebanon/en/home/library/environment_energy/MASTER-PLAN-FOR-THE-CLOSURE-AND-REHABILITATION.html. Accessed 20 June 2018
UNDP, MOEW (Lebanase Ministry of Energy and Water), ELARD (Earth Link & Advanced Resources Development Group) (2014) Assessment of groundwater resources of Lebanon. http://www.lb.undp.org/content/dam/lebanon/docs/Energy%20and%20Environment/Publications/Assessment%20of%20Groundwater%20Resources%20of%20Lebanon.pdf. Accessed 19 May 2018
UNDP, MOEW (Lebanese Ministry of Energy and Water) (2016) National guideline for rainwater harvesting systems. http://www.lb.undp.org/content/dam/lebanon/docs/Energy%20and%20Environment/Publications/RWHG.pdf. Accessed 23 Nov 2018
USEPA (2009) Industrial waste resource guidelines: sampling and analysis of waters, wastewaters, soils and wastes. https://www.epa.vic.gov.au/~/media/Publications/IWRG701.pdf. Accessed 23 Nov 2018
USEPA (2018a) Clean water act analytical methods. https://www.epa.gov/cwa-methods. Accessed 23 Nov 2018
USEPA (2018b) National Primary Drinking Water Regulations. https://www.epa.gov/ground-water-and-drinking-water/national-primary-drinking-water-regulations. Accessed 15 June 2018
WHO (2008) Guidelines for drinking-water quality. http://www.who.int/water_sanitation_health/dwq/fulltext.pdf. Accessed 19 July 2018
WHO (2011) Cadmium in drinking-water. http://www.who.int/water_sanitation_health/dwq/chemicals/cadmium.pdf. Accessed 27 May 2018
WHO (2013) Cadmium. http://apps.who.int/food-additives-contaminants-jecfa-database/document.aspx?docID=9002. Accessed 21 July 2018
Wilson DC (2018) Potential urban runoff impacts and contaminant distributions in shoreline and reservoir environments of Lake Havasu, southwestern United States. Sci Total Environ 621:95–107. https://doi.org/10.1016/j.scitotenv.2017.11.223
World Bank (2011) Republic Lebanon Country Environmental Analysis. http://documents.worldbank.org/curated/en/360011468265768992/pdf/622660ESW0Gray0c060601100BOX361480B.pdf. Accessed 20 June 2018
Wu J, Sun Z (2016) Evaluation of shallow groundwater contamination and associated human health risk in an alluvial plain impacted by agricultural and industrial activities, mid-west China. Expo Health 8:311–329. https://doi.org/10.1007/s12403-015-0170-x
Wu T, Li X, Yang T, Sun X, Mielke HW, Cai Y, Ai Y, Zhao Y, Liu D, Zhang X, Li X, Wang L, Yu H (2017) Multi-elements in source water (drinking and surface water) within five cities from the semi-arid and arid region, NW China: occurrence, spatial distribution and risk assessment. Int J Environ Res Public Health. https://doi.org/10.3390/ijerph14101168
WWAP (World Water Assessment Programme) (2017) Water pollution is on the rise globally. http://www.unesco.org/new/en/natural-sciences/environment/water/wwap/facts-and-figures/all-facts-wwdr3/fact-15-water-pollution/. Accessed 17 Aug 2018
Yan Y, Zhou YQ, Liang CH (2015) Evaluation of phosphate fertilizers for the immobilization of Cd in contaminated soils. PLoS ONE 10:e0124022. https://doi.org/10.1371/journal.pone.0124022
Yu C, Ling Q, Yan S, Li J, Chen Z, Peng Z (2010) Cadmium contamination in various environmental materials in an industrial area, Hangzhou, China. Chem Speciat Bioavailab 22:35–42. https://doi.org/10.3184/095422910X12631439471494
Zang Y, Devleesschauwer B, Bolger PM, Goodman E, Gibb HJ (2018) Global burden of late-stage chronic kidney disease resulting from dietary exposure to cadmium, 2015. Environ Res 169:72–78. https://doi.org/10.1016/j.envres.2018.10.005
Zhang Y, Wu J, Bin X (2018) Human health risk assessment of groundwater nitrogen pollution in Jinghui canal irrigation area of the loess region, northwest China. Environ Earth Sci 77:273. https://doi.org/10.1007/s12665-018-7456-9