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Spatial Distribution of Some Heavy Metals in Urban Soil of Western Iraq

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Abstract

Concentrations of major and trace metals (33 elements) in urban soils of Al-Anbar province, Iraq were surveyed. Links between heavy metals pollution and potential sources were investigated. The concentration of 15 elements, including As, Cu, Sb, Cd, Ag, Ni, Cr, V, Cs, Te, Zn, Mn, Ti, Fe and Ca, was found higher than background limits. High detected concentrations indicating enrichment of these metals in urban soil of Al-Anbar province. Besides, the presence of "unexpected" toxic metals such as Rb, Sr and Zr was observed at high levels in the surveyed region. Correlation analysis was employed to evaluate the source similarities of heavy metal pollution in urban soils of the studied area. Four main sources of soil contamination by heavy metals heavy was suggested. Uncontrolled rapid urbanization in the Al-Anbar province is the first contamination source for different trace metals, *e.g.,* Zn, Cd and Cu. Lead, Cr, Mn and Ni are closely related to the socio-economic growth in the studied region. Microelements, *e.g.,* Fe, K, Ca, are appeared to be driven from the natural origin through paedogenesis and from parent materials. Obtained results showed that the military activities could be regarded as a potential source for high levels of unusual trace elements and toxic heavy metals like As, Cd, Cr, Cu, Ni, Pb, Rb, Sr, V and Zr that are usually found only in trace amounts in the natural environment. This study will not only guide soil quality in Al-Anbar province but also can provide a baseline for policy analysis to mitigate soil pollution in urban areas and risk control.

Keywords: Heavy metals; Urban soil; Pollution; spatial distribution; West of Iraq

Introduction

Contamination by heavy metals in urban soils is of increasing worldwide concerns due to the growing demand for metals in industries and rapid urbanization. Heavy metals naturally exist in the earth's crust. Therefore metals concentrations in the soils tend to be at low concentrations (Pan et al., 2016). However, anthropogenic sources of many soil heavy metals, particularly in urban soils, could exceed their natural inputs from pedogenesis (Facchinelli et al., 2001)). Fertilization, metallic or mining and smelting operations, sewage and sludge applications, urban effluent and atmospheric deposition, are deemed the major sources of soil heavy metals pollution (Montagne et al., 2007, Rahi et al., 2014). In addition, military activities and human warfare have also been documented as a potential source for heavy metal pollution and it impacts on the ecosystem structure and function) (Machlis and Hanson, 2011, Lawrence et al., 2015(. Intensive use of some chemical agents, e.g., trace elements (Arsenic, Copper lead and Zinc), explosives or radioactive elements, is well reported during military activities(Clausen et al., 2004). Heavy metals do not tend to be broken down easily by biodegradation processes. Still, they tend to accumulate within the soil over time, which may trigger physical and chemical changes in the soil properties and undesirable legacy on the landscape (Certini et al., 2013) Some of the toxic metals can be potentially accumulated and magnified at high levels through the food chain in contaminated soils, and therefore cause adverse effects to human health when the latter regarded as a final consumer (Nadal et al., 2004, Li et al., 2009) .Most important, heavy metals are identified as co-factors in many diseases (Giaccio et al., 2012, Guagliardi et al., 2012). For example, Lead, one of the most commonly used

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metals as a chemical warfare agent in military activities (e.g., in munitions), has toxic properties that are highly detrimental to many organ systems in mammals including nervous systems (Papanikolaou et al., 2005).

In Iraq, several studies on heavy metals in urban soils at different scales have been conducted (Sulaivany and Al-Mezori, 2007, Salah et al., 2012, Al Obaidy and Al Mashhadi, 2013, Fayad et al., 2013). Al-Anbar is the largest populated province, located in the west part of Iraq, including many urbanized and populated areas. Al-Anbar province, as some of Iraqis territories so-called "hotspots," has witnessed some dramatic military activities after 2003. As a consequence, loads of military wastes contained different heavy metals that were emitted and deposited, which left a legacy on the landscape across the province.

Limited investigations were conducted for mapping heavy metals in the urban soils at Al-Anbar province to provide information about planning management strategies to achieve better urban environmental quality and risk assessment, e.g., (Salah et al., 2013, Al-Anbari et al., 2015). However, two other Possible reasons are associated with the socio-economic growth from one hand and military activities in the province of Al-Anbar from another hand.

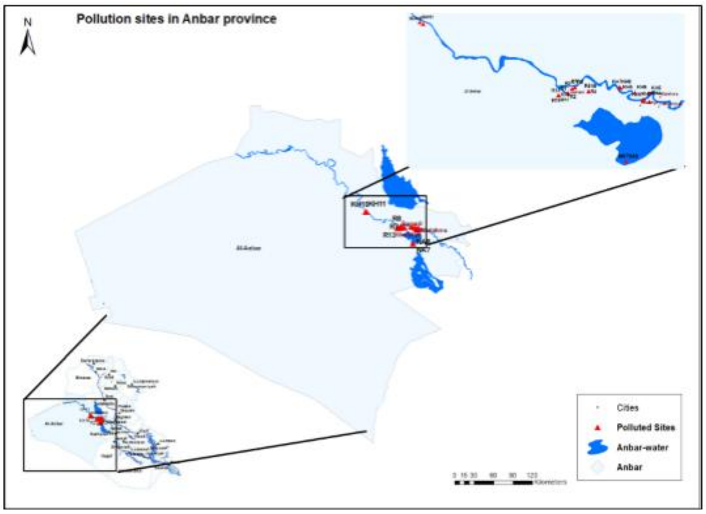
To understand urban soil deterioration in Al-Anbar province, understanding the sources of heavy metals pollution should be considered. Rapid urbanization associated with loss of in agricultural soils during the last five decades, also; gaseous emissions from vehicle exhausts of intensive road traffic in recent years. Last but not least, armed conflict in Al-Anbar province has generated an unexpected potential source for heavy metal pollution.

Few information, if any, about the spatial distribution and availability of these contaminants in urban soil in Al-Anbar province are available. Therefore, the objective of this research is to evaluate the spatial distribution and possible potential sources of several heavy metals in soils of the region of Al-Anbar urban areas.

Materials and Methods

1.1 Study area

Al-Anbar province is located in a plain desert environment of western Iraq, which occupies a surface area of 138,500 km2 (Latitude 31°00' to 36°00' N, Longitude 39°00' to 44°00' E) that represent one-third of the total area of Iraq (Figure 1). The city is some of the vast deserts in the world, namely the Arabian desert located in the central plain of the Arabian Peninsula. The province is densely populated (a total population of approximately 1,600,000 inhabitants (CSO, 2014).



<Figure 1: Soil sampling points the study area>

In recent years, Al-Anbar province acquired ecological importance not only because of its large geographical area but also for the dramatic events of sequence military actions that occurred in this region. After 2003, the survey region is exposed to long-term military events as part of the war against terrorism, where different kinds of military weapons were used.

Soil Sampling and Analysis

The survey region represents urban core area, including Ramadi (R), Al-Khalidya (KH), Fallujah and Hit (NA) districts of the Al-Anbar province (Latitude 33°34' to 33°63' N, Longitude 43°76' to 42°83' E), where different residential and commercial activities along with motorways are located. A total of 96 surface soil samples (0-20 cm) were collected from the survey region using a gridded sampling design. Each study area was divided into

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regular grids of 500x500 m, and samples were collected surrounding each sampling point and then were mixed to form one composite sample. A composite (3 subsamples) soil sample was collected in each grid in residential gardens, school parks, health services sites, roadside fields and agricultural warehouses. At each sampling site, samples were screened to remove gravel-sized materials, residual roots and other unwanted materials and then stored in polyethylene film bags until analysis. The samples were air-dried, ground, passed through a sieve of 2 mm mesh size and homogenized. Each sample weighed about 1 kg. All soil samples were screened for 33 heavy metals by X-ray fluorescence spectrometry (Niton XL3t 950 XRF Analyzer) (Mo, Zr, Sr, U, Rb, Th, Pb, Au, Se, As, Hg, Zn, W, Cu, Ni, Co, Fe, Mn, Cr, V, Ti, Sc, Ca, K, S, Ba, Cs, Te, Sb, Sn, Cd, Ag and Pd). Precision and accuracy of the analysis were performed using duplicate samples, and several referenced soil samples.

Results and Discussion

Descriptive statistics and frequency distribution for multi-elemental concentrations in topsoil samples are reported in Table 1 and Figure 3. For most of the studied metals, total levels were not normally distributed, illustrating slightly skewed data ranged from 0.1 for Ca to 4.67 for Zn. Due to such lack of homogeneity of variances, the median instead of mean values would better describe data distribution precisely. As shown in Table 1 and Figure 2, heavy metal concentrations are varied considerably. To simplify results interpretation, multi-elements examined were grouped into five main categories (A, B, C, D and E). Group A, including nine heavy metals (Mo, U, Th, Au, Se, Hg, W, Co and Pd), are found in the lowest concentrations (approximately zero levels at all studied sites). Concentrations of group B elements (Rb, Pb, As, Cu, Sb, Sn, Cd and Ag) were markedly higher than those of group A. Observed concentrations of group C (Zr, Ni, Cr, V, Sc, Cs and Te) were relatively higher than metal levels from group B but lower than measured concentrations of group D (Sr, Zn, Mn, Ti and Ba). The concentrations of four heavy metals from group E (Fe, Ca, K and S) were found in the highest levels compared to other group's metals.

	Minimum	Maximum	Median	Mean	SEM[a]	Skew[b]	Kurt[c]	LOD[d]	CV[e]
Group A									
Mo	0.0	10.72	0.0	1.34	0.48	1.99	3.40	0.42	2.04
U	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.01	0.0
Th	0.0	6.08	0.0	0.49	0.27	3.01	7.78	0.01	3.18
Au	0.0	10.50	0.0	0.79	0.45	3.10	8.61	0.01	3.20
Se	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.02	0.0
Hg	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.01	0.0
W	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.01	0.0
Co	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.05	5.65
Pd	0.0	29.65	0.0	3.40	1.45	2.20	3.50	0.01	2.41
Group B									
Rb	16.57	53.36	32.63	32.49	1.59	0.19	0.50	0.05	0.27
Pb	0.0	47.23	10.92	10.55	1.72	1.61	5.21	0.02	0.92
As	0.0	19.88	8.33	8.49	0.67	0.48	2.47	0.01	0.45
Cu	0.0	47.67	28.67	22.63	2.93	0.42	1.34	0.27	0.73
Sb	0.0	122	38.62	42.65	3.86	1.21	4.88	0.02	0.51
Sn	0.0	74.3	23.76	25.10	2.62	0.80	3.07	0.02	0.59
Cd	0.0	58.19	17.10	14.62	2.65	0.72	0.34	0.01	1.02
Ag	0.0	27.93	9.30	8.10	1.50	0.46	1.02	0.002	1.05
Group C									
Zr	14.70	175.53	91.75	94.04	4.71	0.01	3.66	0.05	0.28
Ni	67.15	252.42	153.41	155.33	8.89	0.06	0.79	0.01	0.32
Cr	0.0	342.13	115.31	117.4	11.92	1.52	3.90	0.02	0.57
V	0.0	134.72	67.98	68.31	5.13	0.16	0.10	0.05	0.42
Sc	0.0	248.79	31.41	68.88	14.55	0.92	0.65	0.02	1.19
Cs	0.0	217.09	73.29	82.18	6.39	1.36	5.68	0.02	0.43
Te	0.0	354.27	119.8	126.4	11.06	1.40	4.75	0.05	0.49
Group D									
Sr	194.13	1142.9	482.53	490.69	36.68	1.12	1.84	0.05	0.42
Zn	18.34	1142	73.35	126.26	35.0	4.67	23.8	0.01	1.56
Mn	305.16	810.48	577.62	578.24	21.58	0.35	0.04	0.01	0.21
Ti	801.61	3756.1	2179.93	2046.1	139.1	0.12	0.89	0.01	0.38
Ba	173.16	968.94	460.69	485.43	24.93	1.07	3.52	0.05	0.29
Group E									
Fe	7981.2	35057.4	24717.9	24780.8	1110.4	0.56	0.34	0.04	0.25
Ca	31664.2	150462.5	82610.7	87783.3	4662.5	0.10	0.28	0.01	0.30
K	4440.5	15506.9	9091.41	9179.25	549.89	0.13	0.94	0.04	0.33
S	0.0	31579.9	11192.9	12234	1738.1	0.42	1.08	0.01	0.80

<Table 1: Descriptive statistics of multi-elemental concentrations in urban soils of the Al-Anbar province (ppm)>

Median levels of group B metals were 53.36, 10.92, 8.33, 28.67, 38.62, 23.76, 17.10 and 9.30 mg kg-1 for Rb, Pb, As, Cu, Sb, Sn, Cd and Ag, respectively. Background concentrations of these elements in the surface soils are 19.02, 6.75, 22.22, 1.12, 50, 0.38 and 0.71 mg kg-1 for Pb, As, Cu, Sb, Sn, Cd and Ag, respectively (USEPA, 2005). The analysed soils contained As, Cu, Sb, Cd and Ag levels are exceeded background concentrations by 69, 63, 94, 56 and 53 % respectively, however, Pb and Sn levels (91 % and 97 %) are within established values for topsoil's by US EPA (Figure 3). Because of the lack of benchmark concentration for Rb, it was challenging to assess obtained results against standard value. Median soil content of group C heavy metals was 91.75, 153.41, 115.31, 67.98, 31.40, 73.29, and 119.8 mg kg-1 for Zr, Ni, Cr, V, Cs and Te, respectively. The reference values established for soils of public, residential and private areas by US EPA are 130, 18.47, 48.93, 73.36, 3 and 0.001 mg kg-1 for Zr, Ni, Cr, V, Cs and Te, respectively. Total soil content of

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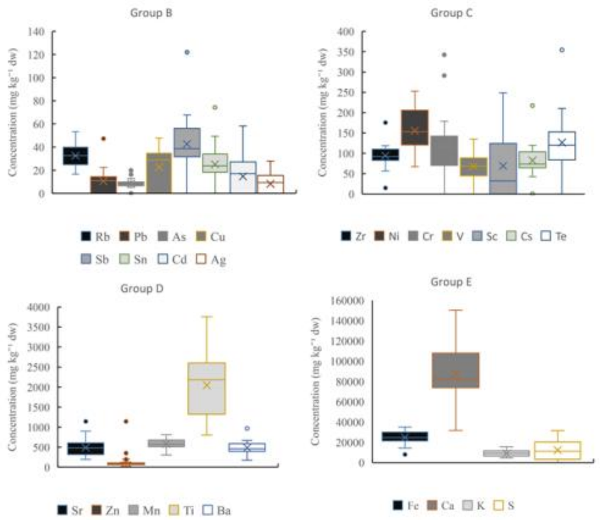
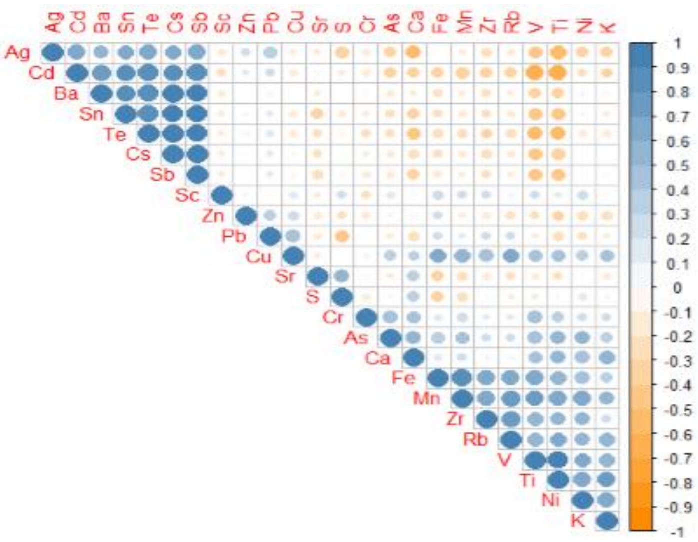
Ni, Cr, V, Cs and Te were above the regularity limits by 100, 91, 44, 97 and 79 %, respectively, while approximately 97% of Zr concentration was observed within the standard limit set. The median levels of Sr, Zn, Mn, Ti and Ba (group D) were 482.53, 73.35, 577.63, 2180 and 460.7 mg kg-1, respectively. The regulatory limits of these metals in surface soils are 200, 58.1, 494.36, 18.47 and 480.8, respectively. A large number of soils were polluted by Sr, Zn, Mn and Ti, with concentrations exceeded background limits by 100, 81.25, 87.5, 100 %, respectively. Nevertheless, approximately 60 % of the analyzed samples shown that Ba concentration was never exceeded the regulatory limit. Median levels of Fe, Ca, K and S (group E) were 24718, 82611, 9091.4 and 11193 mg kg-1, respectively. Benchmark concentrations for Fe and Ca in the soil are 24088 and 14954 mg kg-1, respectively. A percentage of contaminated soil by Fe and Ca was 50 % and 100%, respectively. Due to the lack of regulatory limits established for K and S, it is challenging to compare obtained results with reference values to determine the extent of contamination and percentage of soil pollution.



<Figure 3: Heavy metal concentrations in urban soils from Al-Anbar province. Each box represents the 25th-75th percentiles and the whiskers represent the 10th -90th percentiles. The horizontal line represents the median, and the cross sign represents the mean>

Based on obtained data, the ranking of group's metals follows ascending order as A < B < C < D < E. Specifically, metal concentrations for the five groups were as follows: (Mo, U, Th, Au, Se, Hg, W, Co, Pd) < Ag < As < Pb < Cd < Cu < Sn < Rb < Sb < Sc < V < Cs < Zr < Cr < Te < Ni < Zn < Ba < Sr < Mn < Ti < K < S < Fe < Ca, Respectively (Figure 4).



<Figure 4: The relative contribution of heavy metal concentrations (%) observed in this study>

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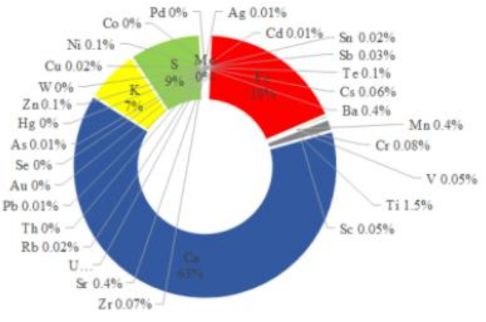
Spearman correlation coefficients were used to investigate the relationships and associations between heavy metal concentrations and to understand the sources of chemical components (Figure 5). Correlation analysis showing that Ag, Cd, Ba, Sn, Te, Cs and Sb were strongly positive related ($P < 0.05$). Ferrous, Mn, Zr, Rb, V, Ti, Ni and K were also significantly positive correlated ($P < 0.05$). Chrome, Arsenic and calcium were closely correlated between each other's from one hand and with V, Ti, Ni and K from another side ($P < 0.05$). Further, Copper showed a relatively significant correlation with Fe, Mn, Zr, Rb, V, Ti, Ni and K, but it is showed weak correlation with As and Ca ($P < 0.05$). Lead is slightly correlated with Ag, Cd, Zn and Cu. Besides, Zinc showed a weak positive correlation with Cu and Fe.
On the other hand, a range of negative relationships between heavy metals concentration was observed ($P < 0.05$). Vanadium, Titanium and, to some extent, calcium was significantly related to Ag, Cd, Ba, Sn, Te, Cs and Sb ($P < 0.05$). However, Cadmium concentrations showed a slight negative correlation with As, Fe, Mn, Zr, Rb, Ni and K. Likewise; Silver concentrations were closely correlated with S, As, Ni and K. Although Zn and Pb are significantly related to S, weak correlations with V, Ti, Ni and K were observed.



<Figure 5: Correlation coefficients of heavy metal concentrations in urban soil of Al-Anbar province>

Based on Spearman correlation analysis, a set of observations could be classified into different groups based on a significant combination of the examined variables. Therefore, significant correlation coefficients between metals could be used to evaluate the source similarities of the screened metals in urban soils of Al-Anbar province. As shown in Figure 5, examined heavy metals were classified into five exclusive groups: (1) Ag, Cd, Ba, Sn, Te, Cs and Sb; (2) Fe, Mn, Zr, Rb, V, Ti, Ni and K; (3) Cr, As and Ca; (4) Cu; (5) V and Ti. These groups are likely to indicate potential sources of metals in urban soils from Al-Anbar province. Accordingly, four main sources of heavy metals pollution can be suggested.
The first source, including trace elements, is mainly related to uncontrolled urbanization dynamics in the Al-Anbar province that has produced a considerable increase for pollution sources of heavy metals via waste disposal, urban effluent and vehicular traffic. For instance, Zn, Cu, and Cd are regarded as trace metal supplements for different urban agronomic practices, while Pb is widely considered as a by-product of vehicle emissions (USEPA, 2005, Sun et al., 2013, Kabata-Pendias and Pendias, 1992) Found that the soil content of Zn in urban gardens and parks in the US was ranged from 20 to 1200 mg kg-1, between 3-140 mg kg-1 for Cu, between 0.02-13.6 mg kg-1 for Cd and 218-10.900 mg kg-1 for Pb. It was also reported that the surface soil of the roadsides and vicinity of highways contained Pb concentrations between 960-7.000 mg kg-1 and between 1-10 mg kg-1 for Cd.
The second source is strictly relative to the socio-economic growth, associated with rapid urbanization, in the surveyed region where multi-industrial and commercial activities, as well as vehicular traffic, are likely to be responsible for the releases and emission of range of heavy metals pollutants to the environment. Several investigations have indicated a close relationship between metal pollution and industrial activity (Schulin et al., 2007, Montagne et al., 2007) .For example, (Schulin et al., 2007)found that the enrichment of Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn in surface soil of Baghdad province was closely associated to industrial activity in the city.
The third source is appeared to be associated with natural occurrence derived from parent materials and inputs from pedogenesis (27). Obtained results showed that the composition of macro-elements and trace elements in the surveyed region varied significantly depending on origin and source of elements. Major elements (e.g., Fe,

K, Ca) appeared to be at the highest levels, while the lowest levels are common for elements usually found at trace levels.

The fourth source for heavy metals pollution is suggested to be belonged to the military activities already occurred in the studied area. ʰeavy metals contamination in areas exposed to military activities can much difference in terms of the type of pollutants compared with natural, domestic, agricultural or industrial territories (Nwaedozie et al., 2013, Vasarevičius and Greičiūte, 2004, Lawrence et al., 2015). As indicated by XRF analysis, there were some "unexpected" trace elements such as V, Rb, As and Sr at high levels. Besides, toxic trace elements associated with man activities such (e.g., Pb, Cd, Cu, Cr, Ni and Zr) observed in the surveyed region suggest point source pollution, probably related to military activities. As evident from the obtained results, trace metal concentrations in the studied locations do revealed significantly higher levels than the EPA standards and averaged values common for national territories (Rahi et al., 2014, Al Obaidy and Al Mashhadi, 2013, Al-Anbari et al., 2015).

Conclusions

To sum up, all studied locations in Al-Anbar province do show significant high contamination levels for most multi-elements screened. Specifically, increased concentrations of trace metals, as well as the presence of "unexpected" elements at high levels alongside with other toxic heavy metals, that are usually found only in trace amounts in natural environment indicates that military activities could be potential anthropogenic source for heavy metals pollution in this region. Considering all suggested pollution sources, this study will provide a baseline for policy analysis to mitigate soil pollution in urban areas and risk control.

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