




















































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










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2 Al-Baha, southwest Saudi Arabia
3 Fahd Al-Shehri, Abdelbaset S. El-Sorogy*, Yasser Al-Zahrani
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8
9 Abstract
10 This study documents the spatial distribution and assesses the environmental risk of heavy metals
11 (HMs) in Al-Baha soils in western Saudi Arabia.^[34] For analysis, 30 surface soil samples were
12 collected and the hazard assessment of the HMs were determined using various pollution
13 measurements, sediment quality guidelines (SQGs), and statistical methods. The average
14 concentrations of HMs ($\mu\text{g/g}$) are listed in the following decreasing order: Fe (5487.73) Mn
15 (323.54) Cr (37.52) Cu (30.25) Zn (24.55) Ni (17.48) Co (9.51) Pb (7.50) Cd (0.81).
16 The Al-Baha soil was very severe enriched and moderate risk with Cd, moderate severe enriched
17 with Cu and Pb, and moderate enriched with Co, Cr, Ni and Zn. Cd, Zn, Pb, and Co reported values
18 lower than the ISQG-Low values and few samples reported levels of Ni, Cr, and Cu between the
19 ISQG-Low and ISQG-High values, indicating a low risk of exposure to these HMs with some
20 anthropogenic effects. Multivariate statistical methods indicated natural sources for Cr, Mn, Fe,
21 Co, and Ni; and anthropogenic sources for Cu, Zn, Cd, and Pb.
22 Keywords: Agricultural soil; Heavy metals; Pollution indices; Risk assessment.
23

Introduction

Land, water and air pollution are from environmental problems in Saudi Arabia caused due to the urbanization and high standards of living whereas consumption of natural resources and agriculture leads to deforestation and desertification (Elimam 2022). Heavy metals are important environmental pollutants and their presence with higher concentrations in plants, atmosphere, soil and water can cause serious problems to all organisms (Ghosh et al. 2013; Al-Hammad and Abd El-Salam 2016). Heavy metals are not easily biodegradable and can consequently be transferred into the human food chain via many pathways and accumulated in important human organs causing significant health problems, particularly for children (Farooq et al. 2008; Antoniadis et al. 2017, 2019; Rinklebe et al. 2019).

Al-Baha region is located in the southwest of Saudi Arabia between the Hijaz Mountains and the Tihama Plain, with an area of 10,362 Km². Rainfall is the main source of water in the region, whether for domestic consumption or for drinking water. Al-Janabin Valley is one of the most important sites in Baljurashi governorate, western Saudi Arabia where the floodwaters are collected and which are stored by Al-Janabin Dam, which is one of the strategic dams in the region and secure a drinking source for the population of the Al-Balshahem village (more than 20,000 residents). Herein, we estimate the extent of HM contamination in agricultural soils in the Al-Baha soils, around Al-Janabeen Dam and determine the significantly enriched HMs contaminating the studied soils. Moreover, we examine potential sources of HMs in the study area using various contamination metrics and multivariate analytical methods.

Materials and methods

Study area

47 Al-Baha area is located in the southwestern part of the Arabian Peninsula and is bordered
48 to the north and west by the Makkah region and to the south and east by the Asir region. The study
49 area was geologically studied (Greenwood 1975a; Anderson 1977; Greenwood et al. 1980; Prinz
50 1983; Greenwood et al. 1986; Johnson and Woldehaimanot 2003; Al-Shanti 2009). It composed
51 of two major Precambrian assemblages: 1) Baish and Baha groups of metamorphosed basalt,
52 graywacke, and chert, and 2) Jiddah and Ablah groups of meta-andesitic and coarse clastic
53 assemblage. The Baish, Baha, and Jiddah Groups are folded, metamorphosed to greenschist facies,
54 and intruded by gabbroic to quartz dioritic plutons about 960 My ago during the Aqiq orogeny.
55 Rocks of the Ablah Group were deposited unconformably on older dioritic and layered rocks.

56

57 Sampling and analytical procedures

58 Thirty soil samples were collected the study area, between 19.9023171-19.911044N and
59 41.7103489 - 41.7119024 E (Fig. 1).^{[19]►} The samples were collected at a depth of 30 cm from the
60 soil surface using a plastic hand trowel, placed in plastic sample bags, and stored in an ice box.^{[0]►} Cr,
61 Mn, Fe, Co, Ni, Cu, Zn, Cd, and Pb were analyzed using inductively coupled plasma-mass
62 spectrometry (ICP-MS) in laboratories of the College of Science, King Saud University, according
63 to USEPA 3050B (USEPA, 1996). Soil samples were air-dried and sieved before chemical
64 analysis.^{[15]►} Approximately 200 mg of samples were accurately weighed into a dry and clean Teflon
65 beaker, and then 2 ml of HNO₃, 6 ml of HCl, and 2 ml of HF were added (Trabzuni et al., 2014;
66 Suleman et al., 2013). Samples were digested on a hot plate with sand at gentle heat 60–120°C for
67 approximately 40 min. The resulting digest was filtered and transferred to 25 ml plastic tubes.^{[60]►} A
68 blank digest was conducted in the same way. ICP-MS calibration was conducted via external
69 calibration.

70 Supplementary Table 1 summarizes the coordinates of sampling sites, and results of HM analysis.^[0]

71 Herein, the enrichment factor (EF), contamination factor (CF), and pollution load index
72 (PLI) were calculated to estimate the HM contamination in soil (Kowalska et al. 2018).^[3] The PLI
73 was used to determine the integrated pollution status of the associated hazardous groups at the
74 sampling sites and the potential ecological risk index (RI) was employed to measure the level of
75 risk of metal accumulation in the soil to community health (Hakanson 1980; Bhuiyana et al. 2010).
76 Furthermore, the sediment quality guideline (SQG) procedure was used to estimate the detrimental
77 effects of polluted soils on microorganisms (US EPA 1992; Long et al. 1995; Crane and
78 MacDonald 2003). The aforementioned indices are classified in Supplementary Table 2.

79 The last mentioned pollution indices were calculated based on the following equations:

80
$$EF = (M/X)_{\text{sample}} / (M/X)_{\text{background}},$$

81
$$I_{\text{geo}} = \text{Log}_2 (C_{\text{m sample}} / 1.5 \times C_{\text{m background}}),$$

82
$$CF = C_o / C_b,$$

83
$$PLI = (CF_1 \times CF_2 \times CF_3 \times CF_4 \dots \times CF_n)^{1/n},$$

84
$$Eri = Tr_i \times Cf^i,$$

85
$$RI = \sum (Tr^i \times Cf^i),$$

86 where M represents the analyzed metal and X denotes the level of the normalizing element.^[31] Fe was
87 selected as the normalizing element in this study, $C_{\text{m sample}}$ represents the analyzed metal within
88 the sample and $C_{\text{m background}}$ denotes the level of the normalizing element, C_o represents the HM
89 concentration in the sediment and C_b represents the normal background value of the HM, CF_n
90 denotes the CF for metal n, Eri denotes the potential ecological RI for each individual HM; its
91 biological toxic response factor and CF are represented by Tr_i and Cf^i , respectively.^[69]

Multivariate statistical techniques—namely hierarchical cluster analysis (HCA), Pearson's correlation coefficient, and principal component analysis (PCA)—were used to determine the likely sources of HMs in the soil.^[3] However, Hg and Cd were not included in the Pearson's correlation analysis and PCA because of their low amounts in the samples.^[3]

96

97 Results and discussion

98 Spatial distribution of HMs

99 The coordination of the sampling sites, concentrations of the analyzed HMs ($\mu\text{g/g}$), and the
100 results of PLI and RI are listed in Supplementary Table 1. The average HM levels are listed in
101 decreasing order: Fe (5487.73) Mn (323.54) Cr (37.52) Cu (30.25) Zn (24.55) Ni (17.48)
102 Co (9.51) Pb (7.50) Cd (0.81). Sample 3 reported the highest concentrations of Cu, Cd, and
103 Pb (41.6, 1.1, and 9.5 $\mu\text{g/g}$, respectively). Samples 5, 18, and 20 reported the highest concentrations
104 of Zn, Mn, and Ni (32, 4733.6, and 45.2 $\mu\text{g/g}$, respectively). Sample 22 reported the highest
105 concentrations of Cr, Fe, and Co (155.2, 24400, and 44.1 $\mu\text{g/g}$, respectively). In the other hand
106 Samples 7, 10, 13, 19, 21, and 23 reported the lowest HM concentrations. Results of the Q-mode
107 HCA supported for a great extent the last mentioned distribution of HMs in the studied soil. It
108 clustered the soil samples into two clusters (Fig. 5). The first cluster comprised samples 18, 20,
109 21, and 22, which reported mostly the highest values the investigated HMs, while the second
110 cluster accounted the remaining samples.^[11]

111 The average values for the HMs in the soil samples are listed in Table 1, along with their
112 comparison to diverse soils in Saudi Arabia, worldwide background references, and SQGs.^[16] The
113 average Ni, Zn, and Pb in the study area were less than those reported in soils worldwide (Kabata-
114 Pendias 2011), earth's crust (Turekian and Wedepohl 1961; Yaroshevsky 2006), and continental

crust (Taylor 1964; Rudnick and Gao 2003), as well as the SOGs and other Saudi soils (Al-Boghdady and Hassanein 2019; Alharbi and El-Sorogy 2021).^[0] Alternatively, average values of Cu, Cd, Cr, and Co were greater than those reported in Al Uyaynah soil, Saudi Arabia (Alharbi and El-Sorogy 2021), moreover, average Cd value exceeded those reported from reported in soils worldwide and continental crust which may be attributed to extensive fertilizer usage and other agricultural activities in the study area.^[70]

121

Assessment and possible sources of HMs

Pollution indices can be used for a comprehensive geochemical assessment of the condition of the soil environment (Caeiro et al. 2005; Kowalska et al. 2016; Weissmannová and Pavlovský 2017; Alharbi and El-Sorogy 2021). Results of the EF (Table 3) indicated that the Al-Ahsa soils are significantly very severe enriched with Cd (average EF = 35.58), moderately severe enriched with Cu and Pb (average EF = 8.64 and 5.10, respectively), moderately enriched with Co, Cr, Ni and Zn (average EF = 3.89, 3.75, 3.33, and 3.29, respectively), and minor enriched Mn (average EF = 2.65). Cadmium is one of the most dangerous of soil pollutants and easily transfers from soil to plants through root absorption (Oliver, 1997). Chronic exposure to cadmium can affect the nervous system, liver, cardiovascular system and may lead to renal failure and death in mammals and humans (Semerjian, 2010). Some individual samples exhibited high enrichment, such as sample 10 with very high Cd and Cu enrichment (EF = 54.39 and 13.36, respectively), and sample 18 with significant Mn enrichment (EF = 14.64). The high EF values indicated anthropogenic activities in the case of these HMs, mostly from the extensive use of fertilizers and insecticides (Kitagishi and Yamane 1981; Kabata-Pendias 2011; Al-Kahtany et al. 2018). However, Mn yielded average EF = 2 (Table 3), indicating a geogenic source for this HM.

Results of the I_{geo} indicated that the investigated soil is moderately polluted with Cd (average $I_{geo} = 1.26$) and unpolluted with the remaining HMs (Average $I_{geo} = 0$). Moreover, CF results indicated a moderate contamination of Cd (average $CF = 2.70$), and low contamination of the other HMs (average $CF = 1$). However, samples 18 and 22 yielded EF values of 4.73 and 3.39 for Mn and Co, respectively, suggesting considerable contamination of the two HMs. The PLI was used to assess HM contamination at a particular site (Rabee et al. 2011; Hossain et al. 2021). The average PLI values indicated that the study area was unpolluted with HMs ($PLI = 1$).^[21]

The potential ecological RIs can be used to determine the effects of HM pollution on the ecology of a particular site (Hossain et al. 2021). The average values of the ecological RIs suggested a moderate risk of Cd ($Eri = 80.94$) and no to low risk of the other HMs ($Eri = 40$). The potential ecological RI ranged from 68.08 (sample 29) and 121.70 (sample 3), with an average of 92.99 (Supplementary Table 1), indicating a considerable risk. Cd, Zn, Pb, and Co exhibited values less than the ISQG-Low values (Simpson et al. 2013), indicating a low risk of these HMs in the soil. However, 5 samples of Ni, 4 samples of Cr, and 3 samples of Cu were reported values between the ISQG-Low and ISQG-High values, indicating a low risk of exposure to these three HMs with some anthropogenic effects.^[20]

The Pearson's correlation coefficient was used to determine the relation between HMs to identify their potential sources (Liu et al. 2016; El-Sorogy et al. 2016). Table 4 lists positive correlations between the members of the two elemental groups: “Cr, Mn, Fe, Co, Ni” and “Cu, Zn, Cd, Pb” indicated similar sources for each group.^[24] Presence of Mn and Fe in the first group indicated natural sources owing to the weathering of clay minerals in the soil (Al-Kahtany et al. 2015; El-Sorogy et al. 2020). Alternatively, members of the second groups implied anthropogenic sources, mainly from agricultural activities (Kahal et al. 2020).

PCA enables researchers to summarize large datasets with many variables into fewer principal components that can be easily visualized and analyzed. This method contributes to our understanding of the main processes involved in soil contamination and its possible sources (Jolliffe and Cadima 2016). Herein, PCA supported the Pearson's correlation coefficient and revealed two principal components that cumulatively explained 81.95% of the total data variance (Table 5). The first component explained 59.39% of the total variance and showed a strong association of Cr, Mn, Fe, Co, and Ni, indicating mixed natural and human sources (Alharbi et al. 2017). The presence of Fe and Mn indicated a natural process. The average EF values of Cr, Co, and Ni were slightly greater than 2 (Table 3), suggesting minor anthropogenic effects (Javed et al. 2018; Kahal et al. 2020). The second component represented 22.55% of the total variance and was highly associated with Zn, Cd, and Cu, which showed higher EF values, indicating an anthropogenic process may be associated with the use of various agricultural chemicals and P fertilizers (Weissmannová and Pavlovský 2017, Alharbi and El-Sorogy 2019).

Conclusions

The present work highlighted the hazardous HMs in the agriculture soil in Al-Baha area using several pollution indices and SQGs. Results of assessment indicated that the Al-Baha soils are significantly very severe enriched and moderate risk with Cd, moderate severe enriched with Cu and Pb, and moderate enriched with Co, Cr, Ni and Zn. Cd, Zn, Pb, and Co reported values lower than the ISQG-Low values and few samples reported levels of Ni, Cr, and Cu between the ISQG-Low and ISQG-High values, indicating a low risk of exposure to these HMs with some anthropogenic effects. Multivariate statistical methods indicated natural sources for Cr, Mn, Fe, Co, and Ni, primarily originating from the weathering of earth materials and atmospheric

deposition; and anthropogenic sources for Cu, Zn, Cd, and Pb, which might be originated from sewage and agricultural activities.^[21] The concentrations of HMs in the soil of Al-Baha must be monitored periodically to control and prevent increased HM levels, particularly those of Cu, Zn, Cd, and Pb. Moreover, farmers should use biofertilizers and manure and reduce their dependency on chemical fertilizers and pesticides.

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