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Bioassessment of Cement Dust Pollution and its Radiation in Al-Ahsa Oasis in the Eastern Province of Kingdom Saudi Arabia

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ZAE analyzed the data, wrote the manuscript and supervised project administration. ZAE, WS and THA conducted the field survey. ZAE, HA, WS and THA helped in lab work. ZAE and WS identification specimens.

Key words
Ecotoxicology, Ground beetles, Heavy metals

ABSTRACT

The objective of this study was to quantify the concentration of heavy metals (Mg, Cu, Pb, Fe, Zn, Cd, and Ni), in the soil, plant (*Zygophyllum decumben*) and ground beetles (*Anthia duodecimguttata)* at two sampling sites. The selected sites located in the Eastern region, Saudi Arabia. The first site was around to the cement factory, in Al-Ahsa (cement site). While the second one was at 15 Km away (reference site). The obtained results showed that there were no differences in the heavy metals (MHs) concentration in soil collected from the two sites ($p>0.05$), except Fe that its concentration was significantly higher in the cement site. In addition, six mean differences of MHs out of seven were proven highly significant in plants and insects collected from the cement site. Mg level was the same in plants and insects collected from the two inspected sites. The concentrations of different MHs were higher in beetles than soil and plant. Only, the concentration of Zn in plant (163.59 µg/g) was higher than that recorded in soil (1.67 µg/g) and insect (80.99 µg/g). The concentration of Mg, Cu, Fe, Pb, Cd and Ni increased and accumulated by transferring from soil to plant to beetle whereas the accumulation of Zn decreased by transferring from plant to insect. The concentration of U^{238}, Th^{232} and K^{40} (Bq/kg) in the soil collected from the cement site is significantly higher than that recorded in the soil collected from the reference site. Except Cs^{137} that showed insignificant difference between the two studied sites. The concentrations of all recorded HMs and radioactivity level in the soil collected from the cement site were less than the acceptable limit.

INTRODUCTION

The cement industry plays a vital role in the imbalances of the environment and produces air pollution hazards (Lamare and Singh, 2020; Olatunbosun *et al*., 2020). Cement dust is considered as a major source of toxic air emission. It contains many toxic substances such as $CaCo_3$ and $NaSo_4$ as well as heavy metals (HMs) (Mandal and Voutchkov, 2011). Such mixture of hazardous HMs as cobalt (Co), iron (Fe), lead (Pb), cadmium (Cd), chomium (Cr), nickel (Ni), manganese (Mn), and arsenic (As) at different proportions, have been considered to be toxic to the living organisms (Gbadebe and Bankole, 2007; Gupta and Singh, 2011). Accumulated large quantities of these elements are potentially harmful to plants, animals and human health (Gupta and Sharma, 2013; Darweesh and El-Sayed, 2014; Azam *et al*., 2015; Khan *et al*., 2015; Rebechi *et al*., 2016; Al-faifi and El-Shabasy, 2021).

It was found that the absorption and translocation characteristics of HMs in plants were associated with plant species, soil properties, environmental conditions, etc. (Leguizamo *et al*., 2017; Schuck and Greger, 2020). The presence of high concentration of cement dust pollutants causes a decline in the plant physiological process;

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such as growth, protein synthesis and photosynthesis (Raajasubramanian *et al*., 2011), a reduction in productivity and concentration of chlorophyll (Sato *et al*., 1993) as well as blocking the stomata and a reduction in number of annual crops (Farmer, 1993).

HMs in the soil accumulate into invertebrates and into higher trophic levels though the food web from soil via detrivores or plants. Although the assumption of biological accumulation of some xenobiotics from the phytophages to carnivores was confirmed (Price *et al*., 1974). Another study carried out by Butovsky (2011) proves that there are no differences in HMs concentrations between omnivorous and carnivorous species.

There are some data on HMs accumulation among various arthopods from field studies (Van Straalen *et al*., 2001; Blanus˘a *et al*., 2002). Insects were usually used as bio-indicators of HMs in soils, due to their mobility and abundance (Nummelin *et al*., 2007). They can tolerate the accumulation of HMs in their tissues and organs, and they can survive in polluted soil because of their genetic adaptation (Ghemari *et al*., 2018; Dar *et al*., 2019). Coleoptera, including the Carabidae family (ground beetles) are accumulators of HMs, particularly the remarkably toxic ones stored in gut (Nasr *et al*., 2020). Consequently, they are excellent indicators of habitat quality and are widely used in biological surveys (Kheirallah *et al*., 2016). Moreover, there were correlations between the HMs concentrations in ground beetles and soil or leaf litter (Jelaska *et al*., 2007; El-Moaty *et al*., 2016).

One of the main determinants of the radiation in soil is radionuclide concentration, such as ^{238}U, ^{232}Th and ^{40}K (Dizman *et al*., 2016). These radionuclides occur naturally in the soil and transported to the environment though plants and water (Usikalu *et al*., 2015). Radiation could influence on the living organism; e.g., maize yield decreased by an average of 16.2% when exposed to radiation (Anda, 1986). Laboratory studies revealed that radiation has adverse effects on the percentage egg hatch, mortality and the time from oviposition to larval development or adult emergence of flour moth *Ephestia kuehniella* (Ayvaz *et al*., 2007).

There is no study that has focused on the HMs accumulation in the Eastern region in Saudi Arabia, especially nearby the cement factory and their transferring on the living organisms of this region. Therefore, in this study, the HMs concentration in the soil, plant and beetle collected from this region were assessed and compared to those from reference site. Moreover, the radionuclides (^{238}U, ^{232}Th and ^{40}K) concentration were evaluated in the soil collected from this region.

MATERIALS AND METHODS

The study area

Two studied areas were selected; the first one was the polluted (cement) site and the second one was the unpolluted (reference) site. The cement site surveyed at 0.5 km downwind from a cement factory in Al-Ahsa, about 30 Km to the south of Al-Hofuf city, in the Eastern region of Saudi Arabia. This area showed signs of cement dust deposition on the vegetation as well as soil surface. The reference site was away from the first one by about 15 Km.

Sampling procedure

Preliminary collections of plants and coleopterous insects inhabiting the selected sites showed that *Zygophyllum decumben* is the dominant plant and *Anthia duodecimguttata* Bonelli, 1813 is one of the dominant carnivore beetle species.

Soil, plants and beetles' samples were collected from the two studied sites. From each site, ten soil samples, each weighed about 0.5 kg, were collected from the top 10 cm by using a hand metallic soil scoop. Also, ten samples of the whole plant were collected. Plants and soil samples were transported to the laboratory in porous nylon bags. The collected beetles were separated, sorted by gender and kept in containers in native soil and plants for 24 h to void gut contents.

Chemical analyses

Soil samples were air-dried at room temperature and ground to pass through a 2-mm sieve. Macro- and microelements were extracted from duplicate samples with DTPA (DTPA: 0.005 M DTPA, 0.1 M triethanolamine (TEA) and 0.01 M CaCl2 at pH 7.3.) with a 1: 2 (v: v) volume ratio (Lindsay and Novell, 1978).

Plant samples were dried at 70°C and has been digested in concentrated sulfuric acid (H_2SO_4, 95-97%) on hotplate at approximately 270 °C. Then, deionized water was added to the final volume of 50 ml in a volumetric flask. 0.5 g of powdered samples of the plant were carefully weighed in a 50 ml standard flask to which 5 ml of concentrated nitric acid was added. The mixture was left for 24 h and then the samples were digested on a hot plate and the temperature was gradually increased to 100 °C. When all nitric oxide fumes were expelled, the temperature was raised and stabilized at 150 °C until the mixture was clear. Distilled water was used to dilute the solution.

Beetle samples (n=15) from each site were anesthetized in ethanol (95%). After cutting the head, elytra, wings, and legs, the beetles dissected in Petri dishes containing drops of 1% Ringer's saline solution and soft

tissues removed from the body, it was grounded using a porcelain mortar and pestle. The prepared whole-body tissue samples subjected to digestion by adding 5-mL of supra-pure grade concentrated HNO_3 in the beaker and slightly heated up at 80 °C to dehydration in the oven. After cooling, added 5-mL of concentrated sulphuric acid (H_2SO_4), and the mixture was heated for 1 h then allowed to cool down. Added 2-mL of 30% hydrogen peroxidase (H_2O_2) solution to the beaker contents and reheated. The final treatment reiterated until a clear solution was acquired to obviate decolorization (Talarico *et al*., 2014).

The determination of HMs; Mg, Cu, Zn, Fe, Pb, Cd and Ni concentration of all collected samples were performed by using Atomic Absorption and Emission Spectrometry model Shimadzu-AA7000. Concentrations of heavy metals were determined according to Allen *et al*. (1986). All data are presented as concentration per unit wet weight of the sample (as µg/g). All analyses were carried out on triplicate samples.

Measurement of radioactivity

To measure the radioactivity in soil; samples were collected from the studied sites, crushed into fine powder by using Mortar and pestle and were dried in an oven at a temperature of 110 °C for 24 h. According to AERB (2003), 200 g of each sample was packed and sealed in an airtight PVC container and kept for about 4 weeks. This period is necessary to allow radioactive equilibrium among the daughter products of radon ^{222}Ra before gamma counting (Veiga *et al*., 2006). HPGe detector based on high resolution gamma spectrometry was used to determine the soil radioactivity. The details of the technique are the same as reported by Meha *et al*. (2007).

Radium equivalent activity was calculated by considering the hazards that are connected with the use of soil containing ^{238}U, ^{232}Th and ^{40}K. It was calculated using the relation:

Raeq = CRa + 1.43 CTh + 0.077 Ck (Yu *et al*., 1992)

Where CRa, CTh and Ck are the activity concentration of ^{238}U, ^{232}Th and ^{40}K in (Bq/kg), respectively (Dhawal *et al*., 2013; Alaamer, 2008). The annual estimated average effective dose equivalent by using a conversion factor of 0.7 $SvGy^{-1}$ that was used to convert the absorbed rate to human effective dose. A summarized effective dose rate was calculated by the following formula:

E = T x Q x D x 10-6 (Dhawal *et al*., 2013; Alaamer, 2008)

Where D is the absorbed dose rate in air, Q is the conversion factor of 0.7 Sv/Gy, and T is the time during one year (8760 h). The characteristics of irradiation risk for the population was considered by the so-called external hazard index that was calculated by the equation.

Hex = AU /370 + ATh /259 + Ak /4810 (Hussain and Hussain, 2011)

Where AU, ATh, and Ak are activity concentration of ^{238}U, ^{232}Th and ^{40}K, respectively.

Statistical analysis

Statistical analysis was performed using the program SPSS. Data were analyzed statistically by One-way analysis of variance (ANOVA) to determine the difference in the HMs concentrations between the studied sites and the Pearson Correlation test to evaluate the relations between HMs concentrations in soil, plant and beetles.

RESULTS AND DISCUSSION

Bioaccumulation of heavy metals

Cement dust contains HMs that deposited into soil at various distances (Schuhmacher *et al*., 2009), and most of them originate from raw materials (Achternbosch *et al*., 2003). They are toxic to insects and plants even at low concentrations (Kabata–Pendias and Mukherjee, 2007) and could accumulated to a different extent in different organ/tissues. The concentrations of HMs in soil samples collected from inspected sites were represented in Table I. The concentration of HMs in soil collected from the two inspected sites were in order of Fe > Mg > Zn > Cu > Ni > Pb > Cd. Statistical analysis showed that the concentrations of Mg, Cu, Zn, Pb, Cd and Ni in soil collected from cement site were obviously insignificantly differed from the those recorded in the soil collected from the reference site. Only, the concentration of Fe was obviously significantly higher ($p < 0.05$) in soil collected from the cement site than that of the reference one (Table I). These findings were dissimilar with those obtained by Eckert *et al*. (1999), Poon and Chen (1999) and Silva *et al*. (2021) who demonstrated that cement dust and its products incorporated a high quantity of lead. However, the current results have the same trend with those obtained by Aitta *et al*. (2019) that the assessed Cd, Co, Pb, and Mn concentration enduring in studied soil were lower than the permissible limits; Cd (0.2 lg/g), Co (0.12 lg/g), Pb (0.3 lg/g) and Mn (0.4 lg/g).

The transfer of MHs from soil to plant is dependent on many factors, such as soil properties, plant species, and metals bioavailability for uptake in the soil-plant system (Adriano, 1986). HMs in the cement dust have hazardous effects on plants richness and diversity (Fakhy, 1994), and the plant growth (Zafar and Shafiq, 1998). In addition, alkaline nature of cement dust reduces the absorption of mineral substances form the soil, this leads to changes in the plant physiology and morphology. Comparing with the HMs' concentrations in soil, *Z. decumben* accumulated high concentrations of HMs in order Fe > Zn > Cu > Mg

> Pb > Ni > Cd. Based on the one way of ANOVA, there were significant differences between the concentrations of MHs in *Z. decumbent* collected from the reference site and those collected from the cement site, except Mg concentration, it was the same concentration in the plants collected from the two studied sites (Table II). These data confirm those obtained by some previous studies (Fakhy, 1994; Butovsky, 2011; Al-faifi and El-Shabasy, 2021). The present recorded concentration of Fe was higher in plant collected from the cement site, confirmed results obtained by Raajasubramanian *et al*. (2011). On the other hand, the higher concentration of Zn and Cu and lower concentration of Mg in *Z. decumbent* collected from cement site, did not agree to those obtained by Raajasubramanian *et al*. (2011).

Table I. Heavy metals concentrations (µg/g) in the soil collected from the two studied sites.

Heavy metals	Concentrations of heavy metals (Mean± SE)		One way ANOVA	
	Reference site	Cement site	F	P
Mg	2.20±0.23	1.77±0.29	1.330	0.31291
Cu	0.987±0.15	1.23±0.06	1.997	0.23045
Zn	1.493±0.29	1.67±0.03	0.364	0.57837
Fe	6.30±0.53	8.58±0.46	10.506*	0.03163
Pb	0.015±0.0001	0.014±0.0003	4	0.11611
Cd	0.006±0.0001	0.006±0.000	0.0000	1.0000
Ni	0.040±0.001	0.034±0.001	1.085	0.35628

*, Statistically significant at $p \leq 0.05$

Table II. Heavy metals concentrations (µg/g) in *Z. decumbent* collected from the two studied sites.

Heavy metals	Concentrations of heavy metals (Mean± SE)		One way ANOVA	
	Reference site	Cement site	F	P
Mg	7.53±0.69	9.30±0.55	3.961	0.11738
Cu	10.47±0.53	25.07±1.66	70.073*	0.00111
Zn	57.04±3.67	163.59±5.18	282.333*	0.00010
Fe	236.34±24.34	110.28±8.02	24.233*	0.00791
Pb	0.019±0.001	0.067±0.0097	23.671*	0.00824
Cd	0.015±0.0008	0.033±0.004	17.11392*	0.01441
Ni	0.0183±0.001	0.045±0.006	13.80752*	0.02054

*, Statistically significant at $p \leq 0.05$

The recorded concentrations of HMs in *A. duodecimguttata* collected from the two studied sites were represented in Table III and it was obvious that beetle accumulated MHs in the same order recorded for the studied plant (Fe > Zn > Cu > Mg > Pb > Ni > Cd). The statistical analysis showed significant differences between the concentration of all HMs in beetle collected from cement site and those from the reference site, except Mg concentration, it was the same concentration in beetles collected from the two studied sites (Table III). These results concord with He *et al*. (2005), Walker *et al*. (2012), Naccarato *et al*. (2020) and Nasr *et al*. (2021). Our findings showed that the mean HMs levels in beetles decreased in the subsequent order Fe > Zn > Cu > Mg > Pb > Ni > Cd. The same trend of HMs diminishing was recorded for carabid beetles studied by Butovsky (2011) and Ghannem *et al*. (2018). Additionally, the present results cleared that beetle accumulated Cd and Pb less than Cu and Zn. These results are similar to those recorded by Jelaska *et al*. (2007) and Nasr *et al*. (2020). In contrast to our findings, Zhang *et al*. (2017) noticed a high Pb level in *Enchytraeus crypticus* of highly contaminated habitats.

Table III. Heavy metals concentrations (µg/g) in *A. duodecimguttata* collected from the two studied sites.

Heavy metals	Concentrations of heavy metals (Mean± SE)		One way ANOVA	
	Reference site	Cement site	F	P
Mg	14.87±0.29	15.03±0.70	0.044253	0.84364
Cu	28.56±0.62	30.83±0.42	8.989293*	0.04001
Zn	40.70±1.67	80.99±1.05	422.8665*	0.0000
Fe	45.23±6.17	136.3±4.43	13.18531*	0.02213
Pb	0.013±0.001	0.090±0.004	302.382*	0.0000
Cd	0.013±0.000	0.067±0.006	74.98286*	0.00097
Ni	0.013±0.000	0.085±0.007	98.51255*	0.00057

*, Statistically significant at $p \leq 0.05$

The percentage of HMs concentrations obviously differed among soil, plant and beetle. The concentrations of different HMs were higher in beetles than soil and plant. Only, the concentration of Zn in plant is higher than that recorded in soil and beetles (Fig. 1). It has been reported that metal associated with heat-stable, low molecular weight metal binding proteins, is highly available to predators. Therefore, some complex mechanisms might exist in the higher tropic level of the food chain to tolerate the contaminated environment (Zhang *et al*., 2017). Statistical analysis for the mean differences of each metal revealed that the mean differences were highly significant between soil, plant and beetles collected from the cement site (F= 149.90, 250.96, 704.59, 20.84, 41.41, 50.00, 21.51 for Mg, Cu, Zn, Fe, Pb, Cd, Ni, respectively all values $p < 0.002$).

Table IV. Correlation coefficients matrix of HMs concentrations (µg/g) in soil, *Z. decumbent* and *A. duodecimguttata* collected from cement site.

Pearson correlation coefficient						
Mg	**Cu**	**Zn**	**Fe**	**Pb**	**Cd**	**Ni**
1						
0.966178*	1					
0.556459*	0.7519*	1				
0.968562*	0.999956*	0.745663*	1			
0.989381*	0.993399*	0.671313*	0.994435*	1		
0.989597*	0.919027*	0.431132	0.922695*	0.958178*	1	1
0.922833*	0.792288*	0.193465*	0.797994*	0.857047*	0.968651*	1

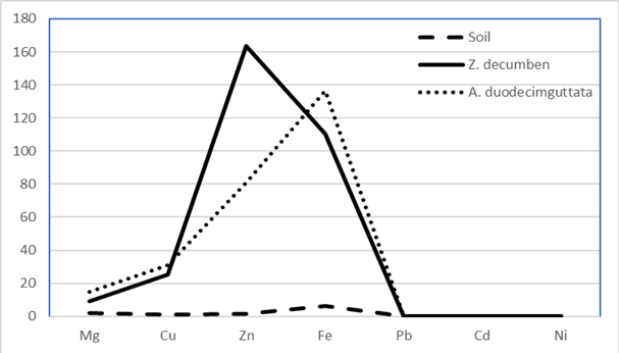

Fig. 1. Percentage of heavy metal concentrations in soil, *Z. decumbent* and *A. duodecimguttata* collected from cement site.

Pearson correlation test showed significant correlation of the concentration of Mg, Cu, Fe, Pb, Cd and Ni in soil, plant and beetles (Table IV). This indicated that these HMs increased and accumulated by transferring from soil to plant to beetle. In the present study, the concentrations of HMs were increased during transfer from soil to plant to insects. This biomagnification of metals has been reported at previous studies (Leita *et al*., 1991; Goodyear and McNeill, 1999; Jelaska *et al*., 2007). This increase of Pb and Cd concentration transferring from soil to plant to insect was observed in respect that Cd and Pb are non-essential elements. These results were in accordance with Van Gestel *et al*. (1993) and Heikens *et al*. (2001). While the accumulation of Zn decreased by transferring from plant to insect (r^2= 0431) could be attributed to the insectabitlity to accumulate higher concentrations of Cu than plant ($p < 0.05$), which may result from the fact that Cu are essential microelement involved in several key physiological processes in insects (Roeder, 1953).

Concentrations of radionuclides

Result of gamma spectrometry analysis for soil samples activity concentration of ^{238}U, ^{232}Th and ^{40}K (Bq/kg) was determined and their contents in (k/kg) was calculated. The results were provided in Table V, showed that the concentration of ^{238}U, ^{232}Th and ^{40}K (Bq/kg) in the soil collected from the cement site is significantly higher than that recorded in the soil collected from the reference site. Except ^{137}Cs that showed insignificant difference between the two studied sites.

Table V. Activity concentration of ^{238}U, ^{232}Th, ^{40}K, and ^{137}Cs (Bq/Kg) in soil samples collected from the two studied sites.

	Activity concentration		**One way ANOVA**	
	Reference site	**Cement site**	**F**	**P**
^{238}U	0.0087±0.000	1.138±0.10	114.9348*	0.000429
^{232}Th	141.0±3.21	171.67±2.19	62.23529*	0.001396
^{40}K	2.48±0.45	3.78±0.22	6.776803*	0.059844
^{137}Cs	9.10±2.49	6.78±0.48	1.710129	0.261063

*, Statistically significant at $p \leq 0.05$

From Table VI, the statistical analysis showed that the mean values of absorbed gamma dose rate in year (nGy/h), radium equivalent activity (Bq/kg), annual effective dose rate (mSv/y) and external hazard index, in the cement site, were significantly higher than that recorded from the reference site. According to our results the radium equivalent activity (Bq/kg), equals to 24.41Bq/kg, which is less than recommended maximum value (UNSCEAR, 2000). According to the results given in Table VI, the mean annual effective dose rate was 0.07mSv/y and the mean value of external hazard index was 0.065. The distribution of natural radionuclides was related to the type of parent, mean value of absorbed dose rate calculated according to natural radioactivity nuclide concentration in the soil. In our studies, it was about 12.11nGy/h. This result was less than the world mean value

Table VI. Absorbed dose rate (nGy/h), radium equivalent activity (Bq/kg), annual effective dose rate (mSv/y) and external hazard index in soil collected from cement site.

	Reference site	Cement dust site	One way ANOVA	
			F	P
Absorbed gamma dose rate in year	8.66±0.83	12.11±0.44	5.148936*	0.08581
Radium equivalent activity	18.13±1.47	24.41±0.95	13.92632*	0.022975
Annual effective dose rate	0.05±0.005	0.07±0.002	12.88356*	0.020265
External hazard index	0.05±0.006	0.065±0.003	13.50885*	0.021289

*, Statistically significant at $p \leq 0.05$

(UNSCEAR, 2000; Dhawal *et al*., 2013). In addition, the mean value of annual effective dose rate and radium equivalent activity were lower than the world mean value (UNSCEAR, 2000; Hussain and Hussain, 2011). The external radiation index mean values were less than one; this indicated that the populated localities on the studied sites is not exposed to radiation hazard that exceeds the limit. Similar results were obtained by Ibraheem *et al.* (2018) who showed that the natural radioactivity levels in the soil samples were well below the acceptable limits (370 Bq/kg) in the soil samples from Abha, Saudi Arabia.

CONCLUSION

The heavy metals in the cement dust are present at different levels in the soil, plants and insects in the area around the industrial cement factory, Eastern region of Saudi Arabia. The data recorded in this study for HMs concentrations accumulated by transferring from soil to plant to insect and both HMs concentrations and radioactivity levels in the soil samples collected from the cement site were below the acceptable limits.

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Statement of conflict of interest

The authors have declared no conflict of interest.

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