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

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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 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 U238, Th232 and K40 (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 Cs137 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 CaCO3 and NaSo4 as well as heavy metals (HMs) (Mandal and Voutchkov, 2011). Such mixture of hazardous HMs as cobalt (Co), iron (Fe), lead (Pb), cadmium (Cd), chromium (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;
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 arthropods from field studies (Van Straalen et al., 2001; Blanas’á et al., 2002). Insects were usually used as bio-indicators of HMs in soils, due to their mobility and abundance (Numblrën 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 (Chemari 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 $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$ (Dizman et al., 2016). These radionuclides occur naturally in the soil and transported to the environment through plants and water (Usiku 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 (Ayyaz 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 ($^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$) 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 CaCl$_2$ 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$_2$SO$_4$, 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 HNO₃ in the beaker and slightly heated up at 80 °C to dehydration in the oven. After cooling, added 5-mL of concentrated sulphuric acid (H₂SO₄), and the mixture was heated for 1 h then allowed to cool down. Added 2-mL of 30% hydrogen peroxidase (H₂O₂) 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 ²²²Ra 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 ²³⁸U, ²³²Th and ⁴⁰K. It was calculated using the relation:

\[
Raeq = CRa + 1.43 \times CTh + 0.077 \times Ck
\]

Where CRa, CTh and Ck are the activity concentration of ²³⁸U, ²³²Th and ⁴⁰K 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 Sv/Gy⁻¹ that was used to convert the absorbed rate to human effective dose. A summarized effective dose rate was calculated by the following formula:

\[
E = T \times Q \times D \times 10^{-6}
\]

(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.

\[
Hex = \frac{AU}{370} + \frac{ATh}{259} + \frac{Ak}{4810} \quad \text{(Hussain and Hussain, 2011)}
\]

Where AU, ATh, and Ak are activity concentration of ²³⁸U, ²³²Th and ⁴⁰K, 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 (Schulmacher et al., 2009), and most of them originate from raw materials (Achterbosch 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.
Based on the one way of ANOVA, there were significant differences between the concentrations of MHs in \textit{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 \textit{Z. decumbent} collected from cement site, did not agree to those obtained by Raajasubramanian et al. (2011).

The recorded concentrations of HMs in \textit{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 \textit{Enchytraeus crypticus} of highly contaminated habitats.
Table IV. Correlation coefficients matrix of HMs concentrations (µg/g) in soil, Z. decumbent and A. duodecimguttata collected from cement site.

<table>
<thead>
<tr>
<th>Pearson correlation coefficient</th>
<th>Mg</th>
<th>Cu</th>
<th>Zn</th>
<th>Fe</th>
<th>Pb</th>
<th>Cd</th>
<th>Ni</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1</td>
<td>0.966178*</td>
<td>0.556459*</td>
<td>0.968562*</td>
<td>0.989381*</td>
<td>0.989597*</td>
<td>0.922833*</td>
</tr>
<tr>
<td></td>
<td>1</td>
<td>0.7519*</td>
<td>0.745663*</td>
<td>0.671313*</td>
<td>0.994435*</td>
<td>0.431132</td>
<td>0.193465*</td>
</tr>
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<td>0.745663*</td>
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</tr>
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<td>0.994435*</td>
<td>0.431132</td>
<td>0.193465*</td>
</tr>
</tbody>
</table>

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 (r²= 0.431) could be attributed to the insectability 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.

<table>
<thead>
<tr>
<th>Activity concentration</th>
<th>Reference site</th>
<th>Cement site</th>
<th>One way ANOVA</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{238}$U</td>
<td>0.0087±0.000</td>
<td>1.138±0.10</td>
<td>114.9348*</td>
</tr>
<tr>
<td>$^{232}$Th</td>
<td>141.0±3.21</td>
<td>171.67±2.19</td>
<td>62.23529*</td>
</tr>
<tr>
<td>$^{40}$K</td>
<td>2.48±0.45</td>
<td>3.78±0.22</td>
<td>6.776803*</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>9.10±2.49</td>
<td>6.78±0.48</td>
<td>1.710129</td>
</tr>
</tbody>
</table>

*, 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.

<table>
<thead>
<tr>
<th></th>
<th>Reference site</th>
<th>Cement dust site</th>
<th>One way ANOVA</th>
</tr>
</thead>
<tbody>
<tr>
<td>Absorbed gamma dose rate in year</td>
<td>8.66±0.83</td>
<td>12.11±0.44</td>
<td>5.148936*</td>
</tr>
<tr>
<td>Radium equivalent activity</td>
<td>18.13±1.47</td>
<td>24.41±0.95</td>
<td>13.92632*</td>
</tr>
<tr>
<td>Annual effective dose rate</td>
<td>0.05±0.005</td>
<td>0.07±0.002</td>
<td>12.88356*</td>
</tr>
<tr>
<td>External hazard index</td>
<td>0.05±0.006</td>
<td>0.065±0.003</td>
<td>13.50885*</td>
</tr>
</tbody>
</table>

*, Statistically significant at p ≤ 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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