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Original Article

Human Risk Due to Radon and Heavy Metals in Soil

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Abstract

Background: We investigated the human risk due to radon and heavy metals (HM) in soil.

Methods: Samples were collected in 2017 from 10 representative geographical locations at Jazan region of southwestern Saudi Arabia and analyzed for elements (Al, Ca, Cu, Ni, Sr Fe, Mg, B, Co, Cr, V, Zn, Mn, Ba, Cd, and Pb). Elements were measured using inductively coupled plasma optical emission spectrometer (ICP-OES). Radon (Rn) was measured using solid-state nuclear detector (SSNTDs).

Results: The maximum human risk was due to Al, which had the highest concentration, where the lowest human risk was due to Cd. The maximum radon concentration was obtained at El-Mazab area with value of 381.05 Bq/m^{3}which leads to 6.55 mSv/y and 78.94 Bq/m^{2}d annual effective doses and radon exhalant rate respectively. Average equivalent and effective dose to different organs due to radon concentration was estimated. Hazard Index due to both carcinogenic and non-cancer hazards were calculated it exceeds permissible level for child due to Nickel and Chromium hence there is a significant risk on children in the study area.

Conclusion: HM concentrations were over limits in some places according to the human activities, municipal waste disposal, fertilizers and pesticides in agriculture. In addition, soil is porous permit dispersion of radon to the atmosphere.

Keywords: Radon; Heavy metal; Risk; Inductively coupled plasma

Introduction

Naturally Occurring Radionuclides (NORM) and heavy metal are distributed through soil in significant amounts depending upon the geological formations of a given area (1-2). Radon and its decay products are the largest source of natural radioactivity we are exposed to it through inhalation of dust particles (3). Radon isotopes are (Rn-222) in the uranium series, thoron (Rn-220) in the thorium series and (Rn-219) in the actinium series (4-6). The annual effective dose equivalent per capita due to inhalation of radon and its progeny reaches to 1.23 mSv, which is about 52% of both natural and artificial radiation doses received by individuals (7). Thus, radon is recognized as the second most important cause of lung cancer after smoking (8-9).

Metals having densities higher than 5g/cm^3 called heavy metals (HM) as As, Cd, Pb, Hg Cr, Co, Cu, Ni, U and Zn, which leads to a problematic pollution due to their persistence and not degradability in the environment. They exist in soils naturally due to volcanoes emissions transport of

continental dusts, and weathering of metal-enriched rocks or due to anthropogenic activities, which increase its concentration to harmful level to human (10-12). The main sources of human activities: exploitation of mines, smelters, application of metal-based pesticides, metal-enriched sewage sludge in agriculture, and combustion of fossil fuel, etc. (13-15). Trace elements have significant important roles in many biological functions but it can be come toxic in higher concentrations.

We investigated the soil concentrations of radon, heavy metals and trace elements then compare the findings with international standards and calculate the human risk due to these contaminants.

Materials and Methods

Sample Collection

Overall, 40 samples were collected from 10 different locations with different biological and geographical conditions at Jazan region of south-western Saudi Arabia as shown in Fig. 1. Samples were collected from depth of 10 cm (surface layer). About 200g were put in a chamber designed to measure radon, its daughters. The specimen is placed at the bottom of the chamber, covered by a filter paper, and the radon detector is located at a distance of 9 cm from the filter paper.

Sample Analysis

Radon Analysis

The CR-39 (solid-state nuclear track detector) is a polymer used for detecting alpha particles which interact with the polymer results in the formation of latent tracks which can be made by chemical etching of the polymer. The three-dimensional shapes of these etch-pits were measured in much detail using a laser scanning con-focal microscope. The efficiency of etching CR-39 using alcohol/water solutions of sodium hydroxide was examined. CR-39 sheets of 25 cm x 30 cm surface area and 1mm thickness were cut into small detectors of area 1.2 cm x 1.5 cm each. The exposure time was 30 days (to reach secular equilibrium) for ^{222}Rn determination. After exposure the CR-39 detectors were etched in 6.25 Normal NaOH at 70 °C for 6 h and the track density ϱ was counted using the optical microscope.

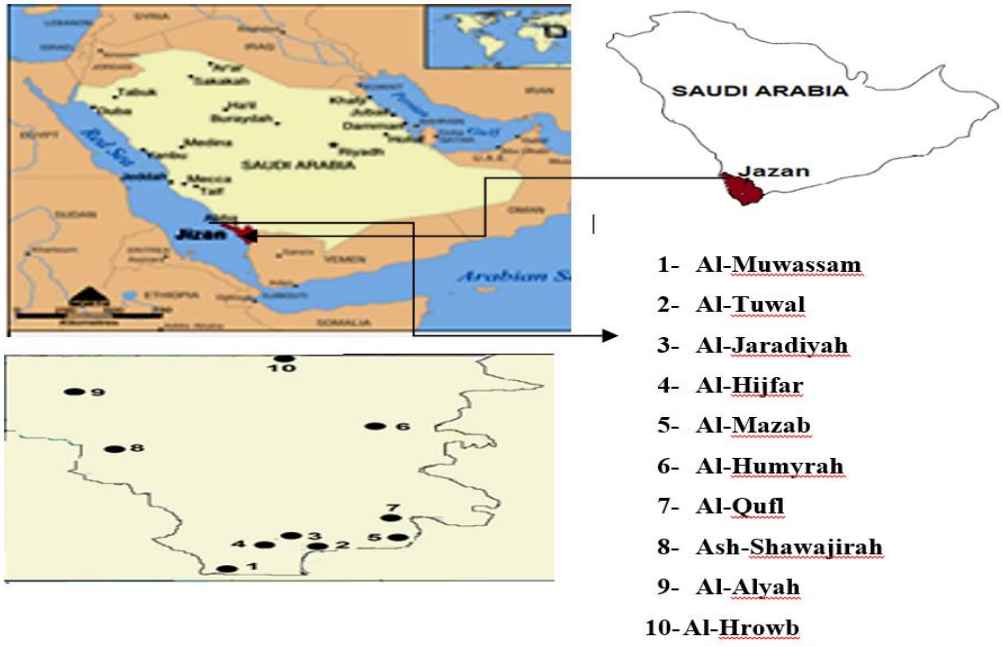

Fig. 1: Map of Jazan region with location of sampling sites 1- 10

The concentration of radon R (Bq/m^3) determined from the track density by using the following relation:

$$R = \frac{\rho}{Kt} \quad [1]$$

where t is the exposure time in hours and the calibration factor K of the SSNTDs can either be determined using standard Rn-chamber K = 0.17 tracks $cm^{-2}d^{-1}/Bqm^{-3}$ of radon .The annual effective dose (D) was calculated from the following relation (16).

$$D = R \times 0.0172\ mSv/yr \quad [2]$$

The surface exhalation rate of the sample for the release of radon can be calculated by the formula (17-18).

$$\text{Exhalation Rate (Ex)} = \frac{RV\lambda}{A\left[T+\frac{1}{\lambda}\left(e^{-\lambda T}-1\right)\right]} \quad [3]$$

Where A is the area of cup (m^2) , V is the effective volume of the cup in m^3, λ the decay constant for radon in h^{-1}, and T the exposure time in hours, respectively.

External Organ Doses Assessment

DFEXT code was used to calculate organ doses to public who live in study area due to external irradiation. In this code, dose coefficients were computed during the preparation of Federal Guidance Report No. 12 (19). Effective doses to different organs were calculated and the tissue-weighting factor (W_T) was taking into considerations. From these coefficients, equivalent dose (H_T) to any organ at height 1 meter from homogenously distributed soil can be calculated as follow:

$$H_T = R * T * 3600\ (sec/hr) * h_t * 10^3\ (mSv/y) \quad [4]$$

Where: R activity concentration of soil (Bq/m^3), T: exposure time (365.25 d/year * 24hr/d), h_t: equivalent dose in tissue t per unit integrated exposure ($Sv\ m^3/sec\ Bq$), the effective dose (E) can be calculated as follow:

$$E = R * T * 3600 * e * 10^3\ (mSv/y) \quad [5]$$

Where: e ($Sv\ m^3/sec\ Bq$) is the effective dose per unit-integrated exposure computed as: $\sum W_T h_t$. For the remainder tissues, the committed dose equivalent per unit-integrated exposure (h_{rem}) can be calculated as: $h_{rem} = 1/5 \sum h_t$ and the tissue weighting factor for the remainder (W_{rem}) is equal 0.2 (20). The dose limit for public must not exceeds 1 mSv/y (21)

Heavy Metal Analysis

Soil samples were oven-dried at 60 °C to constant weight then ground in a mill and sieved to ensure a maximum particle size of 100 µm to minimize the variability due to grain size composition. Soil samples were digested in an Ethos Pro closed system microwave (aquaregia digestion AD) digestion lab (Milestone Inc., Italy) which was chosen as such that the minimum amount of acids could be used with the microwave apparatus.All solutions were prepared with ultrapure water. Soil samples were washed with 10% v/v nitric acid before rinsingwith copious amounts of ultrapure water and drying in air before use. Calibration standard solutions were prepared using serial dilutions for all metals. The standard reference soil materials (GBW07408 and GBW08302 for soil) were used to verify accuracy and repeatability of elemental analytical process. The digestion procedure was as following: concentrated, high-purity HNO_3 (14 ml) was added to the sample until the soil was fully submerged. The mixture was heated until completely dry; then 50 ml of 5% (v/v) high-purity HNO_3 was added following the initial acid digestion. Heavy metals concentrations were determined using an inductively coupled plasma optical emission spectrometer (ICP-OES) (ICAP 6500 Duo. Thermo Scientific, UK). The analysis was carried out at the Desert Research Center (DRC) in Egypt.

Health Risk Assessment of Heavy Metals

Human exposure to soil metals can be accumulated through different pathways as the ingestion (from surface and sub-surface soil), inhalation (from fugitive dust, indoor and outdoor vapors) and dermal contact (from surface and shallow sub-surface soil) of dust particles (22). Model assessment of health risk used in this study was performed by the US Environmental Protection

Agency (23-24). The doses are calculated as follows:

$$D_{ing} = \frac{CxIngRxEFxED}{BWxAT} x10^{-6} \quad [6]$$

where D_{ing} is the average daily intake through ingestion in mg/kg/d, C is the concentration of metal in the soil (mg/kg), IngR is the ingestion rate of soil (mg/day), EF is the exposure frequency (day/year), and ED is the exposure duration (year), BW is the average body weight (kg), and AT is the average time (day).

$$D_{inh} = \frac{CxInhRxEFxED}{PEFxBWxAT} \quad [7]$$

Where D_{inh} is the average daily intake through inhalation in mg/kg.day, InhR is the inhalation rate of soil (m^3/day), PEF is the particle emission factor in m^3/kg.

$$D_{dermal} = \frac{CxSAxSLxABSxEFxED}{BWxAT} x10^{-6} \quad [8]$$

Where D_{dermal} is the average daily intake through dermal absorption in mg/kg/d, SA is the exposed skin area (cm^2), SL is the skin adherence factor, ABS is the dermal absorption factor. A hazard quotient (HQ) (or non-cancer risk) can be calculated as follow:

$$HQ\ (\text{non} - \text{cancer risk}) = \frac{Calculateddose}{\text{RfD}} \quad [9]$$

Where, RfD is the reference dose (mg/kg/d) for the specified element, whereas for carcinogens, a level of cancer risk can be calculated as:

$$cancerrisk = calculateddosexSF \quad [10]$$

Where, SF is the slope factor $(mg/kg/d)^{-1}$. The hazard index (HI) is then the sum of HQ (25).

$$HI = \sum HQ \quad [11]$$

There is no significant risk of non-carcinogenic effects at HI < 1, but the magnitude of risk will be increases with the increasing of HI (25). Carcinogenic risk is used to estimate the probability of an individual developing any type of cancer from the lifetime exposure to carcinogenic hazards. The acceptable risk for regulatory purposes is in the range of 10^{-6}–10^{-4} (21). In this study, hazard index methods and cancer risk methods were used to assess health risks of metal exposure to children and adults in the study areas (22-26).

Results

Radon Measurement

The mean average activity concentration of radon in soil was 189.66 Bq/m^3. Al-Hijfar region showed the minimum value of 88.39 Bq/m^3, while Al-Mazab had the highest value of 381.05 Bq/m^3 and the maximum value of radon exhalant rate and effective dose as shown in Table 1.

Table 1: Radon activity, annual effective dose and exhalation rate in different locations

Location	*Rn concentration (Bq/m³)*	*Dose (mSv/y)*	*Exhalation rate (Bq/m²d)*
Al-Muwassam	229.04	3.94	47.45
Al-Tuwal	256.94	4.42	53.23
Al-Jaradiyah	247.40	4.26	51.25
Al-Hijfar	88.39	1.52	18.31
Al-Mazab	381.05	6.55	78.94
Al-Humyrah	143.73	2.47	29.78
Al-Qufl	273.61	4.71	56.68
Ash-Shawajirah	216.52	3.72	44.86
Al-Alyah	162.82	2.80	33.73
Al-Hrowb	135.17	2.32	28.00
Minimum	88.39	1.52	18.31
Maximum	381.05	6.55	78.94

External Organ Doses Assessment

Table 2 shows the minimum and maximum average equivalent and effective dose to different organs due to exposure to Rn-219, Rn-220, and Rn-222 in different locations. Body surface received the maximum equivalent dose with value of 1.2E-5 mSv/y.

Table 2: Average annual equivalent and effective dose due to exposure to radon, mSv/y

Organ	*Maximum (Al-Mazab)*	*Minimum (Al-Hijfar)*	*Organ*	*Maximum (Al-Mazab)*	*Minimum (Al-Hijfar)*
R Marrow	6.1E-06	1.4E-06	Lungs	6.3E-06	1.5E-06
Adrenals	5.3E-06	1.2E-06	Ovaries	5.1E-06	1.2E-06
B Surface	1.2E-05	2.9E-06	Pancreas	5.0E-06	1.2E-06
Brain	6.1E-06	1.4E-06	Skin	7.7E-06	1.8E-06
Breast	7.3E-06	1.7E-06	Spleen	5.8E-06	1.3E-06
G Bladder	5.1E-06	1.2E-06	Testes	7.0E-06	1.6E-06
Esophagus	5.0E-06	1.2E-06	Thymus	6.0E-06	1.4E-06
ST Wall	5.6E-06	1.3E-06	Thyroid	5.9E-06	1.4E-06
SI Wall	5.2E-06	1.2E-06	U Bladder	5.6E-06	1.3E-06
ULI Wall	5.3E-06	1.2E-06	Uterus	5.1E-06	1.2E-06
LLI Wall	5.4E-06	1.2E-06	Muscle	6.4E-06	1.5E-06
Heart	5.6E-06	1.3E-06	H_{rem}	6.3E-06	1.5E-06
Kidneys	5.8E-06	1.4E-06	E	6.2E-06	1.4E-06
Liver	5.8E-06	1.3E-06			

Concentration of Nutrients Soil Elements (Al, B, Ca, Mg)

Fig.2 shows soil concentrations of Mg, Ca, Al and Fe in different locations. Al-Qufl area had the highest concentrations of Al with values of 28615.2 mg/kg and 2268.42 mg/kg of Ca. B concentrations were the highest (≥ 8 mg/kg) in 50% of locations as shown in Fig. 3.

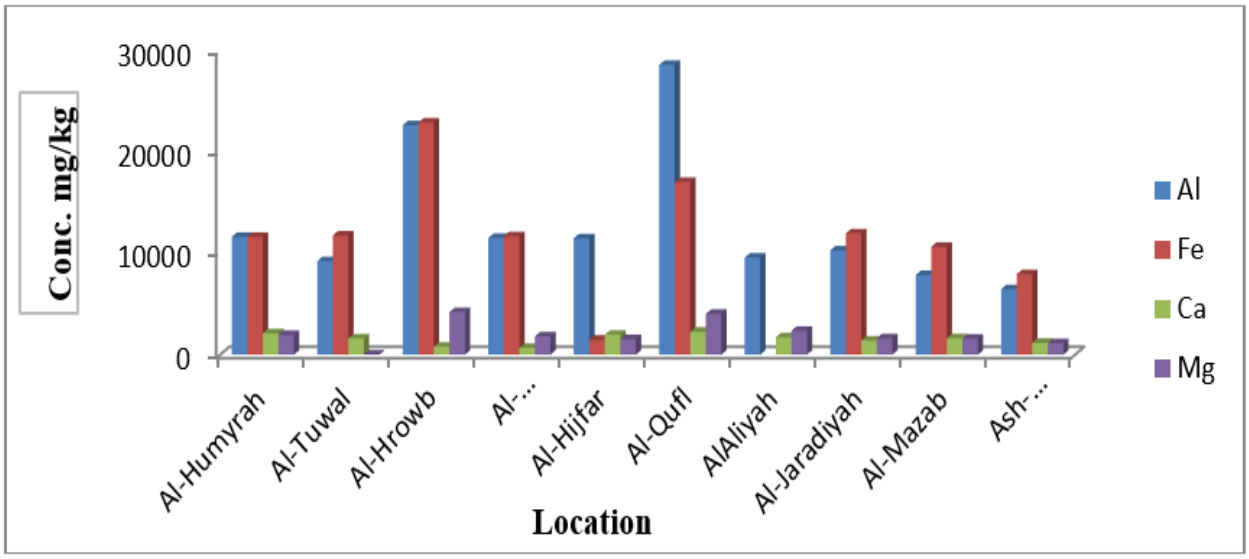

Fig. 2: Al, Fe, Ca and Mg concentrations in different locations

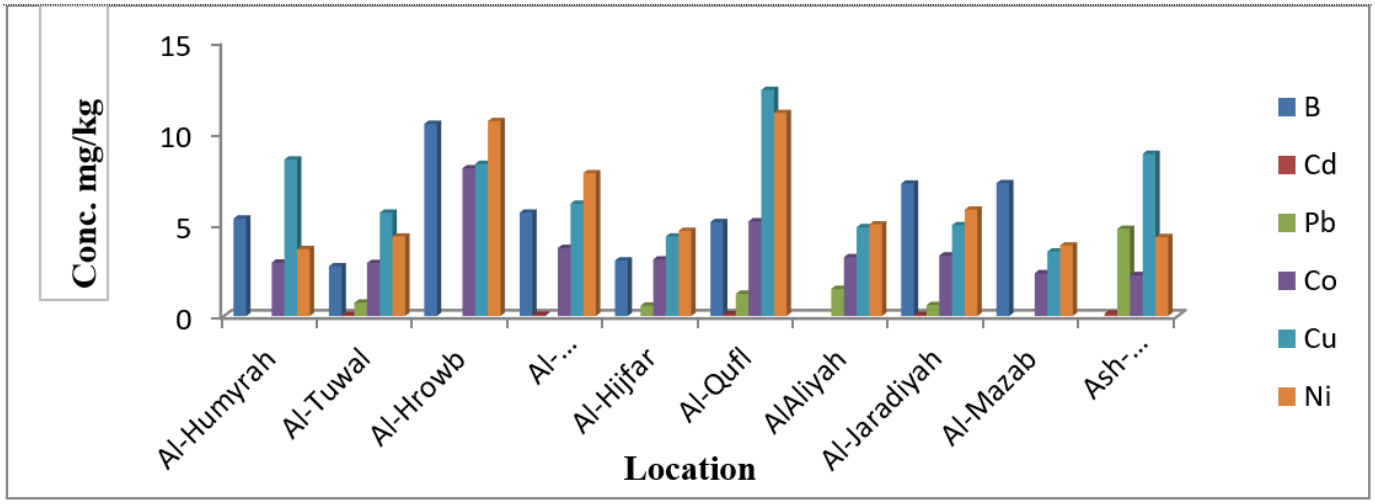

Fig. 3: B, Cd, Pb, Co, Cu and Ni concentrations at different locations (mg/kg)

Concentrations of Heavy Metal

Al-Hrowb area showed the highest concentrations of Fe, 22917.5 mg/kg as shown in Fig. 2. As shown in Fig. 4, Mn and Ba had average soil concentrations of 146.28 mg/kg and 57.27 mg/kg respectively. They showed concentrations less than 200 mg/kg at the most locations except Al-Muwassam.

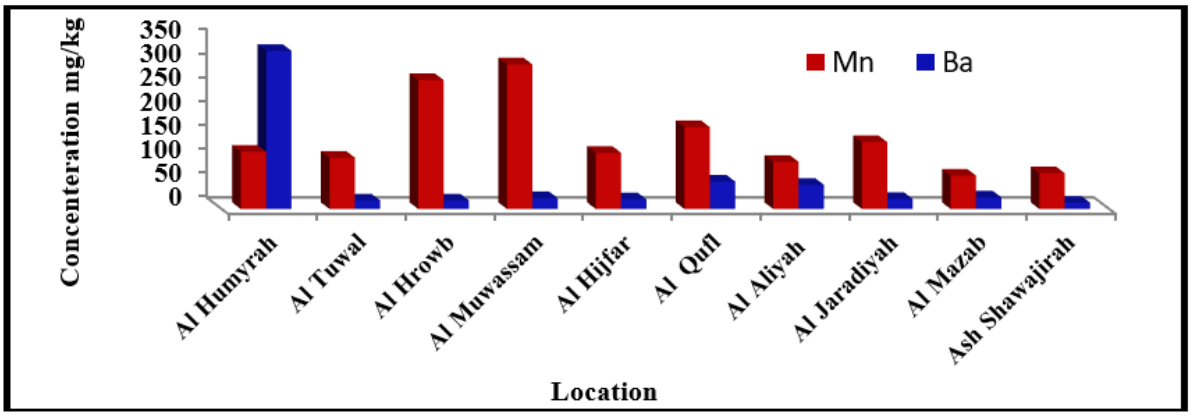

Fig. 4: Manganese and barium concentrations at different locations (mg/kg)

Concentration of Elements (Cu, Co, Ni, Cr, Zn, Sr, V, Cd, and Pb)

As shown in Fig. 3, concentration of copper was varying from 3.53 to 12.36 mg/kg with average of 6.76 mg/kg. Ni concentration was varying from 3.66 to 11.1 mg/kg. Cr, Sr, V and Zn concentrations are shown in Fig. 5. The highest concentrations values of Sr were 24.49 and 21.14 mg/kg in Al-Qufl and Al-Hijfar areas respectively. Zn concentrations were less than 35 mg/kg in all locations with mean value 18.56 mg/kg. Very little concentrations of Cd were measured. The highest concentration of Pb was detected in Ash-Shawajirah area with 5 mg/kg. A comparison between element concentrations of this study and that from other countries for the same soil category is shown in Table 3.

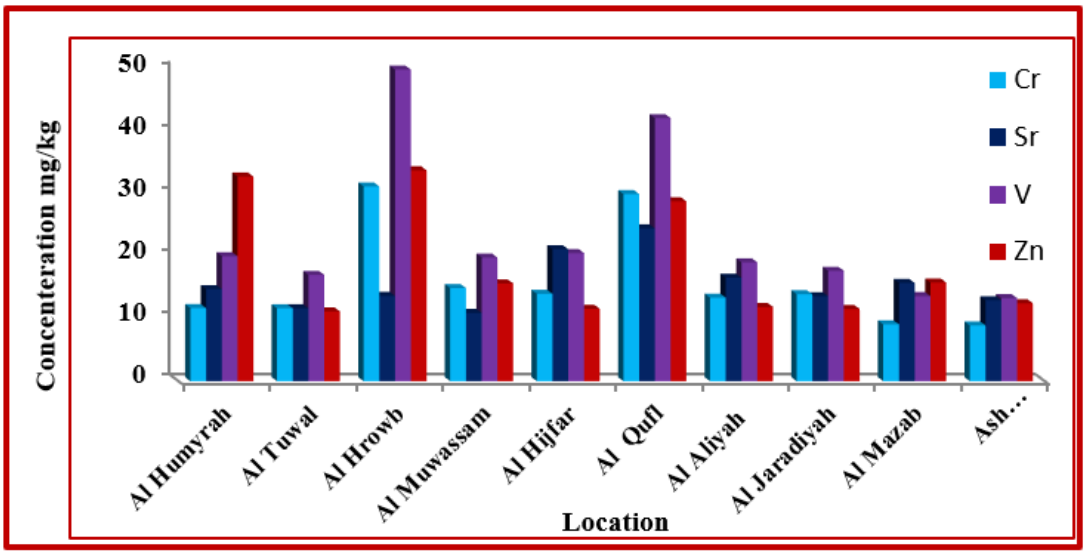

Fig. 5: Chromium, strontium, vanadium and zinc concentrations at different locations (mg/kg)

Table 3: Comparison between element concentrations of this study and that from other countries for the same soil category

Elements	*Current study*	*Spain (27)*	*Hungary (28)*	*Japan (29)*	*China (30)*
AL	12937.36	-	-	-	-
Ca	1534.92	6729	-	-	-
Fe	11621	43725	2070	-	-
Mg	2192.92	482.43	-	-	-
B	4.69	-	-	-	-
Co	3.7	-	-	12.5	-
Cu	6.76	39.24	11.8	448	36.4
Ni	6.13	25.86	5	153	23.87
Cr	15.89	34.29	2.8	71	53.11
Sr	15.55	-	-	-	-
V	23.32	-	5.1	67	-
Zn	18.56	1454.7	52	227	167.03
Mn	146.28	1817.60	-	325	-
Ba	57.27	-	-	-	-
Cd	0.04	4.36	0.9	0.8	1.05
Pb	0.94	-	19.4	114	36.71

Table 4 shows human risk due to different elements. Table 5 shows a non-cancer hazard quotient (HQ) for both child and adult. Its range was from 4.57E-8 for adult due to lead to 3 for child due to chromium. Table 6 shows carcinogenic hazard quotient and the total hazard Index due to both carcinogenic and non-cancer hazards.

Table 4: Human Risk due to different elements

Element		D_{ing}		D_{inh}		D_{dermal}	
	C (mg/kg)	Child	adult	Child	adult	Child	Adult
AL	1.3E+04	1.7E-01	1.8E-02	2.8E-05	1.8E-04	1.4E-01	5.4E-05
Ca	1.5E+03	2.0E-02	2.1E-03	3.3E-06	2.2E-05	1.7E-02	6.4E-06
Fe	1.2E+04	1.5E-01	1.6E-02	2.5E-05	1.6E-04	1.3E-01	4.9E-05
Mg	2.2E+03	2.8E-02	3.0E-03	4.7E-06	3.1E-05	2.4E-02	9.2E-06
B	4.7E+00	6.0E-05	6.4E-06	1.0E-08	6.6E-08	5.2E-05	2.0E-08
Co	3.7E+00	4.7E-05	5.1E-06	7.9E-09	5.2E-08	4.1E-05	1.6E-08
Cu	6.8E+00	8.6E-05	9.3E-06	1.5E-08	9.5E-08	7.4E-06	2.8E-09
Ni	6.1E+00	7.8E-05	8.4E-06	1.3E-08	8.6E-08	6.7E-05	2.6E-08
Cr	1.6E+01	2.0E-04	2.2E-05	3.4E-08	2.2E-07	1.8E-04	6.7E-08
Sr	1.6E+01	2.0E-04	2.1E-05	3.3E-08	2.2E-07	1.7E-04	6.5E-08
V	2.3E+01	3.0E-04	3.2E-05	5.0E-08	3.3E-07	2.6E-04	9.8E-08
Zn	1.9E+01	2.4E-04	2.5E-05	4.0E-08	2.6E-07	4.1E-06	1.6E-09
Mn	1.5E+02	1.9E-03	2.0E-04	3.1E-07	2.1E-06	1.6E-03	6.1E-07
Ba	5.7E+01	7.3E-04	7.9E-05	1.2E-07	8.1E-07	6.3E-04	2.4E-07
Cd	4.0E-02	5.1E-07	5.5E-08	8.6E-11	5.6E-10	6.2E-08	2.4E-11
Pb	9.4E-01	1.2E-05	1.3E-06	2.0E-09	1.3E-08	6.2E-08	2.4E-11
Min.	4.0E-02	5.1E-07	5.5E-08	8.6E-11	5.6E-10	6.2E-08	2.4E-11
Max.	1.3E+04	1.7E-01	1.8E-02	2.8E-05	1.8E-04	1.4E-01	5.4E-05

Table 5: A non-cancer hazard Quotient (HQ) due to toxic elements

Element	HQ ing		HQ		HQ	
	Child	adult	Child	adult	Child	Adult
Cu	2.15E-3	2.33E-4	3.75E-7	2.38E-6	6.17E-4	2.33E-7
Ni	3.90E-3	4.20E-4	6.31E-7	4.17E-6	1.24E+0	4.81E-4
Cr	6.67E-2	7.33E-3	1.19E-3	7.69E-3	3.0E+0	1.12E-3
Zn	8.00E-4	8.33E-5	1.33E-7	8.67E-7	6.83E-1	2.67E-4
Cd	5.10E-4	5.50E-5	--	--	6.2E-3	2.40E-6
Pb	3.43E-3	3.71E-4	6.15E-7	4.00E-6	1.18E-4	4.57E-8

Table 6: Carcinogenic hazard and non-cancer hazards

Element	SF inh	Cancer risk		Hazard Index = ΣHQ	
		Child	adult	Child	adult
Cu	--	--	--	2.77E-03	2.35E-04
Ni	8.40E-01	1.09E-08	7.22E-08	1.24E+00	9.06E-04
Cr	4.20E-01	1.43E-08	9.24E-08	3.07E+00	1.61E-02
Zn	--	--	--	6.84E-01	3.51E-04
Cd	6.30E+00	5.42E-10	3.53E-09	6.71E-03	5.74E-05
Pb	--	--	--	3.55E-03	3.75E-04

Discussion

All samples represent upper limit of external exposure due to radon compared with the recommended safe limit of 1 mSv/y for public (21), while, organ and effective doses due to Rn-219, Rn-220, and Rn-222 still below recommended level. Over doses of aluminum in food reducing

the metal skeleton (osteopenia) and can be observed in infants.

Al-Hrowb site show the highest concentrations of Mg (4206.32 mg/kg), which is higher than other countries while Ca concentrations are lower as shown in Table 4 (27-30). Fe lead to tissue damage, it is higher than those reported in other countries (31). Study area includes activities associated with the petroleum sector, intensive traffic and agricultural and the addition of fertilizer to the soil has led to the deposition of heavy metals (32). The high concentration of Ba in Al-Muwassam region, mainly due to painting automobiles (31).

Cu concentrations were less than the permissible limits of 50-140 mg/kg, and those reported in other countries (Table 3). Sources of copper include metal factories, copper-based pesticides, and fungicides used in agriculture. The concentrations of Ni were less than the permissible limits of 30-75 mg/kg (33), and that found in other countries expect Hungary (28). The main sources of Ni are the steel industry, oil burning, smelters, and coal burning. Strontium was detected in current study rather than other countries. All studied locations show chromium concentrations less than the recommended level of 500 mg/kg (33) and other countries expect Hungary. Chromium sources include the chemical sector and oil refining. Similarly to Ni, the sources of V include steel works, oil burning, smelters, refineries, and coal burning.

Zinc concentration was less than the permissible limit of 150- 300 mg/kg (33) and less than other countries. The sources of Zn are the chemical industry and brake pads.The concentration of Cr, Sr, V, and Zn were lower than the respective permissible limits except Al-Qufl and Al-Hrowb locations. The concentration of Pb was less than permissible value of 50-300 mg/kg. The very little Pb concentrations or absence of detectable concentrations in different locations indicates that little or no relevant anthropomorphic activities take place at these localities. Hazard index for child due to Nickel and chromium exceeds unity (1.24 and 3.07 respectively).

Conclusion

ICP-OES is a very rapid and accurate method for analyzing different elements in soil. The study area have high soil concentrations of Mg, Ca, Fe, and Al due to human activities and the use of agricultural fertilizers. The concentrations of Mn and Ba are attributed to emissions from automobile paint shops located near Al-Hrowb, Al-Muwassam and Al-Humyrah locations. Cd and Pb are particularly toxic, but their concentrations were low at all locations. The maximum human risk was due to Al while Cd has the lowest risk.The total hazard Index due to both carcinogenic and non-cancer hazards due to Nickel and Chromium lead to a significant risk on children in the study area.

Ethical considerations

Ethical issues (Including plagiarism, informed consent, misconduct, data fabrication and/or falsification, double publication and/or submission, redundancy, etc.) have been completely observed by the authors.

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Not applicable

Conflict of interest

The authors declare that there is no conflict of interest.

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