Measurement of Natural Radioactivity and Radiation Hazard Indices for Dust Storm Samples from Qassim Region, Saudi Arabia

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Abstract: This work presents a study of natural radioactivity levels and radiation hazard indices for dust storms samples from Qassim region, Saudi Arabia. Distributions of naturally occurring radioactive materials (NORM) of 226 Ra, 232 Th and 40 K were determined using NaI (Tl) γ -ray spectrometry. The Radiological effects from 23 samples were estimated. Activity concentrations were determined for 226 Ra (from 1.8 to 102 with average value of 10 Bq kg-1), 40K(from 1.5 to 57.6 with average value of 8.1 Bq kg⁻¹) and 232 Th (range from 88.9 to 433.6 with average value of 306.6Bq kg⁻¹). To assess the radiological hazard of the natural radioactivity in the samples, the radium equivalent activity (Raeq), Gamma Index (I γ) and the absorbed dose rate were calculated and found to be within those values recommended by the International Commission on Radiological Protection (ICRP1990) as the maximum annual dose to members of the public. This study provides a baseline map of radioactivity background levels in the Saudi environment and will be used as reference information to assess any changes in the radioactive background level due to geological processes.

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1. Introduction

A dust (sand) storm is a meteorological phenomenon common in arid and semi-arid regions. Dust storms arise when a gust front or other strong wind blows loose sand and dust from a dry surface. Particles are transported by saltation and suspension, causing soil erosion from one place and deposition in another. The Sahara and dry lands around the Arabian peninsula are the main terrestrial sources of airborne dust, with some contributions from Iran, Pakistan and India into the Arabian Sea. It has been argued that recently, poor management of the Earth's dry lands, such as neglecting the fallow system, are increasing dust storms from desert margins and changing both the local and global climate, and also impacting local economies. Dust storms cause soil loss from the dry lands, and worse, they preferentially remove organic matter and the nutrient-rich lightest particles, thereby reducing agricultural productivity. Also the abrasive effect of the storm damages young crop plants. Other effects that may impact the economy are: reduced visibility affecting aircraft and road transportation; reduced sunlight reaching the surface; increased cloud formation increasing the heat blanket effect; high level dust sometimes obscures the sun over cities; effects on human health of breathing dust. Movement of mineral dust by the air atmosphere has been underway for ages, but efforts to assess its magnitude and significance have come largely in the last century.

Attempts to identify and understand effects of the dust on soils are even more recent, mostly in the last 30 years. Airborne movements have several forms. Dust is transported by large storms that can be spectacular. Dust is also moved by smaller pulses and in a rather continuous flux. Most movement is within continents or from continents to nearby oceans, but fine dust (fine silt and clay) is carried from one continent to another. Prime sources of the dust are the deserts and semi-arid regions of the world. For example, large quantities of dust are known to be carried out of the Sahara every year. Quantities of dust carried by individual pulses, from the small ones to the massive storms, extend from a few thousand to 150 million metric tons.

Simonson (1995) reported that the Quantities of dust carried by individual pulses, from the small ones to the massive storms, extend from a few thousand to 150 million metric tons.. That the dust could add significant quantities of nutrient elements as, for example, south of the Sahara, came into the picture even later. The beneficial effects of airborne dust to many soils raise an interesting question. Is the dust more, or less, useful in its new location than it was in its source area? An adequate answer to that question would provide an improved basis for assessing the effects of wind erosion. Chen et al., (2009) explained that The dust concentrations over the saline land of the western Songnen Plain (SSL), Northeastern China, are circumstantially higher than those from the

northwestern Chinese deserts. These concentrations are sensitive to the surface soil conditions and wind velocity on the ground. The dust samples collected during dust storm events on the SSL contain abundant Na, Mg, Al, K, Ca, Fe and Ti, as well as toxic elements such as Cu, V, Zn and Ba. Individual particle analysis reveals that fine saline particles (< 10 μ m in diameter) on the saline land, consisting largely of carbonate, halite and sulfate together with lithogenic minerals such as SiO2 and aluminosilicate, are eventually uplifted during the interval from spring to autumn. Guo et al., (2004) determined The chemical characteristics of selected elements and solventextractable organic compounds (SEOC) to identify the origin of these materials. The elements Fe, Mg, Ca, Al, Ti, Mn and V in the dust samples were found to be of crustal origin and were transported to Oingdao from outside the area, while Pb and Zn were attributed to local (regional) emissions. The results suggest that the dust episodic aerosols possess strong local characteristics superimposed with a heavy crustal and possibly pollution influx transported with the dust particles during the storm. Because the sources of the aerosols were different, high pollutant concentrations were observed at Oingdao in two waves: the locally emitted aerosols such as vehicular exhaust rose first before the long-distance transported materials arrived and the locally emitted pollutants tended to stay in the atmosphere for a longer time. Tiantao et al., (2005) examined the Mass concentrations and concentrations of the chemical elements associated with aerosols he found that the average mass concentration of TSP during the field period were 0.545 mg m-3 at Sanggen Dalai tall-tower station, 0.366 mg m-3 at Sanggen Dalai ground-based station, 0.360 mg m-3 at Beijing, and 2.778 mg m-3 at Sunite Zuogi. The average concentrations of PM10 accounted for more than 61% of TSP loadings at all stations. The average concentrations of special crustal elements (Al, Ca, Fe, Mg, Mn) in PM10 accounted for 65-86% of each of their loadings in TSP, and the average concentrations of important pollutant elements (Pb, Zn, Cu) in PM10 accounted for 63-79% of each of their loadings in TSP. During the dusty days with wind speeds rising, the aerosol mass loading severely increased, and the trend of chemical element increasing in dust particles was associated with the aerosol loading ascending. Crustal elements in aerosols were dominated by dust particles.

Natural radioactivity is wide spread in the earth's environment. It exists in soil, plant, water, air, coal and phosphate. The natural radioactivity in soil comes mainly from ²³⁸U series, ²³²Th series and ⁴⁰K. Naturally occurring radioactive materials (NORMs) in soil are one of the components of external gamma-ray exposure to which persons are exposed to regularly.

Natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on geological and geographical conditions and exists at different levels in the soils of each region in the world. The specific levels of terrestrial environmental radiation are related to the geological composition for each lithologically separated area, and to the content of uranium, thorium and potassium in the rock from which the soil originated in each area (Chowdhury *et al.*, 2006; Gbadebo, and Amos 2010)

Measurement of natural radioactivity in soil is very important to determine the amount of change of the natural background activity with time as a result of any radioactive release, where monitoring of any release of radioactivity to the environment is important for environmental protection and is useful to set the standards and national guidelines in the light of international recommendations. The aim of the present work is: 1) measuring the natural radioactivity levels of ²²⁶Ra, ²³²Th and ⁴⁰K in dust storm samples from Qassim and the radiation hazard indexes due to these radio-nuclides. 2) Calculate the radiation hazard parameters due to these natural radionuclides in dust storm soil. 3) Determining the amount of pollutants in dust storms which include heavy metals. This database will be used as reference to gauge any inputsand transboundary radioactive release in future. Study area:

The study samples were collected from Qassim region, which located in the center of Saudi Arabia and its area is about 65,000 km2, and classified as arid climate and mean monthly air temperature is 34 °C and 15 °C, in summer and winter seasons, respectively However, Dust storms can occur anytime of the year but they are mostly frequent during March, April and May. According to data dust/sand storms are continuously present 29.78% throughout the year in Qassim area

2. Materials and Methods Samples collection and preparation

23 dust samples were collected from different locations in Qassim area (see table 1) between September 2012 to July 2013, by used dry stainless steel dust fall collectors($1.2m \times 1.2$ square meters) at 1.5 m altitude above ground level. The location of these samples were stated in the following Table 1. The collectors were situated in some cities in Qassim province and to get enough materials for analysis the samples collected at the end of study period (approximately one year, from September 2012to July 2013). Samples were sieved using 0.125 mm sieve to avoid ash. The location of these samples were stated in the following Table 1.The Weighted samples were ground, homogenized and sieved to about 100 meshes by a crushing machine. The samples were then placed for drying at 110°C to ensure that moisture is completely removed. Weighted samples were placed in polyethylene bottles, of 350 cm³ volume, each. The bottles were completely sealed for more than one month to allow radioactive equilibrium to be reached. This step was necessary to ensure that radon gas is confined within the volume and that the daughters will also remain in the sample (IAEA, 1989; El-Taher et al., 2003; El-Taher et al., 2007 and El-Taher, 2010 ae).

Samples counting

The naturally occurring radionuclides considered in the present analysis of the measured γ -ray spectra are : ²¹²Pb (with a main gamma energy at ~239 keV and a gamma yield of ~ 43.1%), 214 Pb (~ 352 keV, ~ 37.1%),²¹⁴Bi (~ 609, 1120 and 1765 keV, ~ 46.1, 15 and 15.9% respectively), 228Ac (~911 keV, ~ 29%), 208Tl (~2615 keV, ~ 35.9%) and 40 K (~1461 keV, ~10.7%). Under the assumption that secular equilibrium was reached between²³²Th and 238 U and their decay products, the concentration of ²³²Th was determined from the average concentrations of ²¹²Pb, ²⁰⁸Tl and ²²⁸Ac in the samples, and that of ²³⁸U was determined from the average concentrations of the ²¹⁴Pb and ²¹⁴Bi decay products (Hamby and Tynybekov, 2000; El-Taher& Madkour., 2011; El-Taher, 2011; El-Taher, 2012). Gamma-spectrometric measurements were performed with HPGe detector. The detector had a photopeak efficiency of about30% and an energy resolution of 1.85 keV for the1.33MeV γ -transition of $^{60}\text{Co.}$ The $\gamma\text{-ray}$ spectrometer energy was calibrated using ⁶⁰Co and ¹³⁷Cs point sources. The measuring time for gamma-ray spectra ranged was 12 h. In order to determine the background distribution due to naturally occurring radionuclides in the environment around the detector, an empty polystyrene container was counted in the same manner as the samples. After measurement and subtraction of the background, the activity concentration was calculated (El-Taher& Makhluf., 2010; 2011).

2. Results and Discussion

Activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K

The variation of the activity concentration (Bq/kg) of ²²⁶Ra, ²³²Th and ⁴⁰K radionuclides in the dust storms soil collected from Qassim area were represented in Tables 2. From all samples (23 samples), the ²²⁶Ra activity (or ²³⁸U activities for samples assumed to be in radioactive equilibrium) ranges from 1.8 Bq/kg to 102.2 Bq/kg with an average of 10 (Bq/kg). The activity concentration of ²³²Th ranges from 1.5 Bq/kg to 57.6 Bq/kg with an average of 8.1 (Bq/kg). Finally, the activity concentration of ⁴⁰K ranges from 88.9 (Bq/kg) to 433.6 Bq/kg with an average of 306.6 (Bq/kg). These results are in

agreement with those reported in UNSCEAR (2000) for Egyptian soil. The average activities (range) of 226 Ra (238 U) series, 232 Th series and 40 K in Egyptian soil are 17 (5–64), 18 (2–96) and 320 (29–650) Bq/kg dry weight (UNSCEAR, 2000). Table 3 presents a comparison of the average value of the specific activity of the radionuclides in dust storm soil with literatures, worldwide.

Estimation of dose rate

Conversion factors to transform specific activities $A_{\rm K}$, $A_{\rm Ra}$ and $A_{\rm Th}$ of K, Ra and Th, respectively, in absorbed dose rate at 1m above the ground (in nGy h⁻¹ by Bq kg⁻¹) are calculated by Monte Carlo method and the values are:

 $D(nGy h^{-1}) = 0.0417A_{K} + 0.462A_{Ra} + 0.604 A_{Th}$ (1)

The annual effective dose rate outdoors in units of $\mu Sv/y$, is calculated by the following formula (Satio *et al.*, 1990):

Annual Effective Dose Rate $=D \times T \times F$ (2)

Where D is the calculated dose rate (in nGy h⁻¹), T is the outdoor occupancy time (0.2x24 h x365.25 $d\approx 1753 \text{ h y}^{-1}$), and F is the conversion factor (0.7x10⁻⁶ Sv Gy⁻¹).

Radiation hazard indexes

The natural radioactivity of building materials is usually determined from ²²⁶Ra, ²³²Th and ⁴⁰K contents. As 98.5 % of the radiological effects of the U series are produced by Ra and its daughter products, the contribution from the ²³⁸U has been replaced with the decay product ²²⁶Ra. Since sand beach minerals, rejected light sands and sea beach soils can be used in industries and building constructions, the γ -ray radiation hazards due to the specified radionuclides were assessed by three different indices Radium equivalent activity is an index that has been introduced to represent the specific activities of ²²⁶Ra, ²³²Th and ⁴⁰K by a single quantity, which takes into account the radiation hazards associated with them. This first index can be calculated according to (Beretka and Mathew, 1985) *as*

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K,$$
(3)

where A_{Ra} , A_{Th} and A_{K} are the specific activities of ²²⁶Ra, 2³²Th and ⁴⁰K in Bqkg⁻¹, respectively. The $Ra_{e\alpha}$ is related to the external γ -dose and internal dose due to radon and its daughters. The maximum value of Ra_{eq} in building materials must be less than 370 Bqkg⁻ ¹ for safe use. Beretka and Mathew 1985 dened two other indices that represent the external and internal radiation hazards). The radium equivalent concentration Raeq for all samples was calculated using radionuclides concentration and given in Table 4. The calculated Ra-equivalent activities of the samples under study are less than the recommended maximum value of 370 Bq.kg⁻¹.

No S	Site(aitx)	location	location			
NO. 5	Sile(city)	Ν	Е	elevation		
1	Shmaceah	26/35/126	44/17/307	582 m		
2	Bandareh	26/32/673	44/17/445	585m		
3	Robeeah	26/24/460	44/11/867	616 m		
4	Shmaceah East	26/20/969	44/11/662	619 m		
5	Bureydah Southeast	26/19/377	44/05/222	616 m		
6	Bureydah South	26/12/325	44/01/455	632 m		
7	Muleeda South	26/18/153	43/46/898	647 m		
8	Muleeda North	26/17/627	43/47/513	644 m		
9	Bureydah Center	26/21/555	43/57/164	607 m		
10	Unezah Southeast	26/09/651	43/57/980	651 m		
11	Unezah East	26/06/958	43/54/866	727 m		
12	Unezah North	26/07/981	43/52/180	727 m		
13	Bdeya North	25/56/373	43/30/136	681 m		
14	Bdeya Northeast	26/02/876	43/41/125	675 m		
15	Aras North	25/48/576	43/19/112	702 m		
16	Aras Southeast	25/15/895	43/22/463	684 m		
17	Aras East	25/55/686	42/26/991	685 m		
18	Bureydah west	26/18/504	43/53/325	661 m		
19	Bureydah East	26/02/032	43/59/498	598 m		
20	Bureydah North	26/24/409	43/55/590	633 m		
21	Bureydah Northeast	26/27/039	43/58/138	608 m		
22	Bureydah Northeast(2)	26/26/670	43/58/278	604 m		
23	Bureydah North(2)	26/21/848	43/57/427	618 m		

Table 1. Docations of this bothe Dust concered Sample.	Table 1: Location	ns of Airborne l	Dust collected	Samples.
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 Table 2: Activity concentrations (226Ra, ²³²Th and ⁴⁰K) in dust storms samples from Qassim province, Saudi Arabia

Sample	Activity Bq/Kg			
No.	Ra-226	Th-232	K-40	
1	4.5	3.1	253.7	
2	6.4	4.6	406.6	
3	4.7	2.9	287.1	
4	18.8	23.9	176.0	
5	1.8	1.5	88.9	
6	5.0	7.1	401.2	
7	7.6	8.2	437.6	
8	3.2	3.3	184.1	
9	12.6	11.7	373.2	
10	4.6	4.5	393.4	
11	2.8	1.8	273.1	
12	9.3	6.8	410.3	
13	6.1	6.3	385.9	
14	4.1	3.8	355.7	
15	5.5	18.5	360.2	
16	102.2	57.6	318.4	
17	2.6	2.2	197.0	
18	4.4	3.7	323.9	
19	4.0	1.9	270.5	
20	7.1	5.0	402.8	
21	1.6	0.8	95.2	
22	8.4	5.4	433.6	
23	2.9	1.6	224.1	

 Table 3
 Comparison of specific activities of radionuclides in dust storm soil samples from Qassim province with soil in other countries as given in [UNSCEIR., 2000].

Samplas	Specific activity of radionuclides) Bq/kg(
Samples	²²⁶ Ra	²³² Th	⁴⁰ K	
Egypt	17	18	320	
United States	40	35	370	
Argentina	-	-	650	
Bangladesh	34	-	350	
China	32	41	440	
Hong Kong SAR	59	95	530	
India	29	64	400	
Japan	33	28	310	
Korea	-	-	670	
Iran	28	22	640	
Denmark	17	19	460	
Belgium	26	27	380	
Luxemburg	35	50	620	
Switzerland	40	25	370	
Bulgaria	45	30	400	
Poland	26	21	410	
Romania	32	38	490	
Greece	25	21	360	
Portugal	44	51	840	
Spain	32	33	470	
Worldwide mean	33	36	474	
Thailand	48	40	400	
OAP data	172	211	511	
Present study	10	8	306	

 Table 4: Radiation hazard parameters in dust storms samples from Qassim province, Saudi Arabia.

Sample No.	Raeq	gamma-index (I)	Ann. dose equ. (μSv/y)
1	28.5	0.1	18.3
2	44.4	0.2	28.7
3	31.0	0.1	20.0
4	66.5	0.2	38.6
5	10.8	0.0	6.9
6	46.0	0.2	29.6
7	53.1	0.2	33.9
8	22.1	0.1	14.1
9	58.0	0.2	35.8
10	41.3	0.2	26.9
11	26.5	0.1	17.4
12	50.6	0.2	32.1
13	59.7	0.2	37.0
14	44.8	0.2	28.7
15	209.1	0.2	117.2
16	36.9	0.7	24.0
17	20.9	0.1	13.6
18	34.6	0.1	22.5
19	27.6	0.1	18.0
20	45.2	0.2	29.1
21	10.1	0.0	6.5
22	49.5	0.2	31.8
23	22.3	0.1	14.6

 Table 5: Gamma-index values suggested by the European Commission (EC,1999).

Dose criterion	0.3mSv y ⁻¹	1 mSv y ⁻¹
Materials used in bulk amounts, e.g. bricks Superficial and other materials with restricted use: tiles, boards, etc	$\begin{array}{c} I_{\gamma} \leq 0.5 \\ I_{\gamma} \leq 2 \end{array}$	$\begin{array}{l} I_{\gamma} \!\! \leq 1 \\ I_{\gamma} \!\! \leq 6 \end{array}$

Gamma index

A number of indexes dealing with the assessment of the excess gamma radiation originating from building materials (frequently called "gammaindexes" or "external-indexes") have been proposed. In this study, the gamma-index was calculated as proposed by the European Commission (EC,1999; Moharram. et al., 2012). The Commission suggests that the building materials should be exempted from all restrictions concerning their radioactivity if the excess gamma radiation originating from the increases the annual effective dose of a membe of the public by 0.3 mSv at the most. On the contrary, doses higher than 1mSv should be accepted only in some very exceptional cases where materials are used locally. The gamma-index($I\gamma$) proposed by the European Commission is calculated using the following

 $I\gamma = CU/300 + CTh / 200 + CK / 3000$ (4) where C_U , C_{Th} , C_K are the ²³⁸U^{. 232}Th and ⁴⁰K activity concentrations in Bqkg⁻¹, respectively, in the granite samples. The index shall not exceed the values given in Table 5, depending on the dose criterion and the way and amount the material is used in a building. Gamma-indexes of the samples are shown in Table 4. Gamma- index (I γ) ranged from 0. 1 to 0.7 in dust storms soil used. No exceedance of the recommended upper limit was noted.

Conclusions

It is important to determine background radiation level in order to evaluate the health hazards. Therefore, radiometric survey is recommended for dust storm soil from Oassim area to assess natural radioactivity levels and heavy metals. From this study, the mean activity concentrations for $^{226} Ra^{,}_{}^{232} Th$ and $^{40} K$ are 10, 8.1 and 306.6 Bq.kg^-1, respectively. The values of Raea ranging from 10.1 to 209.1 with mean value 45.2 Bq kg⁻¹. From the results it can be seen that the values Raeq, Gamma index and gamma absorbed dose rates of dust storm samples varies appreciably from sample to another due to the variation of radium, thorium and potassium. The average Raeq values for dust storm samples in the studied area are below the internationally accepted values 370 Bq kg^{-1} . The values of annual effective dose rates in air of dust storm samples from Qassim area, Saudi are within the world range (6.9-117.2 (μ Sv/y). The data obtained here are reference value to be used a data baseline for drawing a radiological map of the region. The data obtained in this study will serve as baseline data for the proper assessment of radiation exposure of dust storm samples.

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