

Assessment of natural and artificial radioactivity levels and radiation hazards and their relation to heavy metals in the industrial area of Port Said city, Egypt

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Abstract A detailed gamma ray spectrometry survey was carried out to make an action in environmental impact assessment of urbanization and industrialization on Port Said city, Egypt. The concentrations of the measured radioelements U-238, Th-232 in ppm, and K-40 %, in addition to the total counts of three selected randomly dumping sites (A, B, and C) were mapped. The concentration maps represent a base line for the radioactivity in the study area in order to detect any future radioactive contamination. These concentrations are ranging between 0.2 and 21 ppm for U-238 and 0.01 to 13.4 ppm for Th-232 as well as 0.15 to 3.8 % for K-40, whereas the total count values range from 8.7 to 123.6 uR. Moreover, the dose rate was mapped using the same spectrometer and survey parameters in order to assess the radiological effect of these radioelements. The dose rate values range from 0.12 to 1.61 mSv/year. Eighteen soil samples were collected from the sites with high radioelement concentrations and dose rates to determine the activity concentrations of Ra-226, Th-232, and K-40 using HPGe spectrometer. The activity concentrations of Ra-226, Th-232, and K-40 in the measured samples range from 18.03 to 398.66 Bq kg⁻¹, 5.28 to 75.7 Bq kg⁻¹, and 3,237.88 to 583.12 Bq kg⁻¹, respectively. In addition to analyze heavy metal for two high reading samples (a_1 and a_{10}) which give concentrations of Cd and Zn elements (a_1 40 ppm and a_{10} 42 ppm) and (a_1 0.90 ppm and a_{10}

0.97 ppm), respectively, that are in the range of phosphate fertilizer products that suggested a dumped man-made waste in site A. All indicate that the measured values for the soil samples in the two sites of three falls within the world ranges of soil in areas with normal levels of radioactivity, while site A shows a potential radiological risk for human beings, and it is important to carry out dose assessment program with a specifically detailed monitoring program periodically.

Keywords Gamma ray contamination · Dose rate · Industrial area · Environmental assessment · Egypt

Introduction

Human beings are exposed to ionizing radiation every day from natural radionuclides in the ground, building materials, air, food, the universe, and even elements in their own bodies. The assessment of these doses from natural materials is important as external radiation exposures from natural materials contribute about 50 % of the average annual dose to humans from all radiation sources (UNSCEAR 2010).

Gamma radiation emitted from naturally occurring radioisotopes, such as K-40 and the radionuclides from the Th-232 and U-238 series and their decay products (also called terrestrial background radiation), which exist as trace levels in all ground formations, represents the main external source of irradiation to the human body (Harb et al. 2008).

In general, approximately 85 % of the annual total radiation dose of any person comes from natural radionuclides of both terrestrial and cosmogenic origin (Belivermis et al. 2010; NCRP 1994; UNSCEAR 2000). Dose rate is most unlikely to exceed 1 mSv in a year. Very little action needs to be taken in this dose range for evaluating and controlling worker doses. If dose rate ranges between 1 and 6 mSv in a year, a dose assessment program is necessary and can utilize work place monitoring or

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individual monitoring. Wherever dose rate is likely to exceed 6 mSv in a year, individual monitoring of the transport personnel is mandatory (IAEA 2005; Anonymous 2005; ICPR 2009).

Radioisotopes that are present in soil significantly affect terrestrial gamma radiation levels. In the last decade, several studies were carried out to assess the average outdoor terrestrial gamma dose rate in air (e.g., Faheem et al. 2008; Ngachin et al. 2008; Janković et al. 2008; Degerlier et al. 2008; Psichoudaki and Papaefthymiou 2008; Al-Hamameh and Awadallah 2009; Baykara and Dođru 2009; Lee et al. 2009; Taskin et al. 2009; Radenković et al. 2009; Belivermis et al. 2010; El-Taher et al. 2010).

Natural radioactivity in soil, sand, and rock comes from the Ra-226 and Th-232 series and K-40, which in turn depends upon the local geology of each region. As natural radionuclides are not uniformly distributed, the knowledge of their concentration and distribution in materials plays an important role in radiation protection. Such investigations can be useful both for the assessment of public dose rates and keep reference data records, to ascertain possible changes in the environmental radioactivity due to nuclear, industrial, and other human activities (Eissa et al. 2010).

The mining, milling, and industrial uses of naturally occurring radioactive materials (NORM) cover a range of mineral resources and industrial activities. Due to the technological processes evolved in some of these industrial processes, the concentration of natural radioactive elements in the products and in the wastes can be much higher than that in the ore/raw materials. TENORM stands for technologically enhanced naturally occurring radioactive materials. “Technologically enhanced” was added to distinguish clearly between radionuclides which occur naturally and those which are produced by human activity. High levels of NORM and consequently increased exposure to NORM have been identified in several major industries including energy production using fossil fuels, phosphate industry as well as oil and gas production (UNSCEAR 1993). The originated wastes in these activities are released in the environment and, hence, an environmental management of the highest quality is needed to reduce the resultant safety problems for both environment and population. The growing concern among the population about the quality of their environment increases the significance of impact assessment of radioactive wastes that discharged into the environment even if natural radionuclides occur.

In the present research, the radiological risks associated with the presence of natural and artificial radionuclides in Port Said city were assessed. For this purpose, radiation doses of gamma radiation were measured in situ using portable RS-230 BGO gamma ray spectrometer, and then radiation hazards parameters were calculated. Results of this work are important both in assessment of the risk for human health and in the development of environmental assessment tools in Egypt. Also, these results can be used as a reference point for radiological mapping of the area of Port Said city.

Port Said city lies in the northeastern part of Egypt, between latitudes 32° 12' 59" and 32° 19' 15" E and longitudes 31° 12' 4" and 31° 17' 6" N. It has a triangular shape, surrounded by the Suez navigation Canal to the east, the Mediterranean Sea to the north, and the eastern part of Lake Manzala to the west (Fig. 1). Port Said city belongs to the far western part of the Sinai Peninsula coastal plain, with a flat topography alleged that ranges from half a meter to about a meter above sea level as it is part of the sandy coastal strip. The region is characterized by a lack of any manifestation of the topography of the terrain preference and covered parts of the southern region of Lake Manzala. The area dates back to Holocene Era where it is composed of Nile deposits represented by three formations which are the Mit-Ghamer formation; consists of about 115 m of sand, silt sand, and mud with overlaps of limestone sand, especially in the upper part of the underground water-bearing formations, this is due to its geological age Pleistocene, The Bilqas formation; consists of 18 m of soft brown sand structures with overlaps of silt where this configuration represents a natural extension of the Nile River Delta sediments and old age due to geological Pleistocene era and Aeolian sand which covers most of the coastal area of Port Said, in addition to some of the island and barrier sandy sediments; a soft sandy minute graininess good category (SEAM Programme 2005; Environmental Description of Port Said Governorate 2007). Nile River transported black sand from mountain ranges of Sudan and Abyssinia to its delta in the Mediterranean coast of Egypt. This black sand contains anomalies of relatively higher natural radioactive nuclides than the other coastal sands (El-Naggar 1990; El-Khatib et al. 1993; El-Gamal et al. 2004; Hussein 2011). Uranium and thorium isotopes have been used commonly as radiotracers for studies of marine sediments, including their sources, transport, and sedimentation rates in relation to the sedimentary and geochemical processes in the marine environment (Cochran and Krishnaswami 1980; Li et al. 1985; Cole et al. 1986; Suman and Bacon 1989). Three sites (A, B, and C) have been selected to carry out the present study.

Materials and methods

The investigation methods which have been carried out in the studied sites are ground gamma ray spectrometric survey and laboratory measurements for soil samples. A description of these investigation methods is given as follows.

Ground gamma ray spectrometric survey

The three studied sites (A, B, and C) were selected in the study area (Port Said city) according to their spatial relations with probable sources of contamination. Two of them (A and B) are located near the industrial area of Port Said city, whereas site C is located near the solid waste dump and sewage treatment

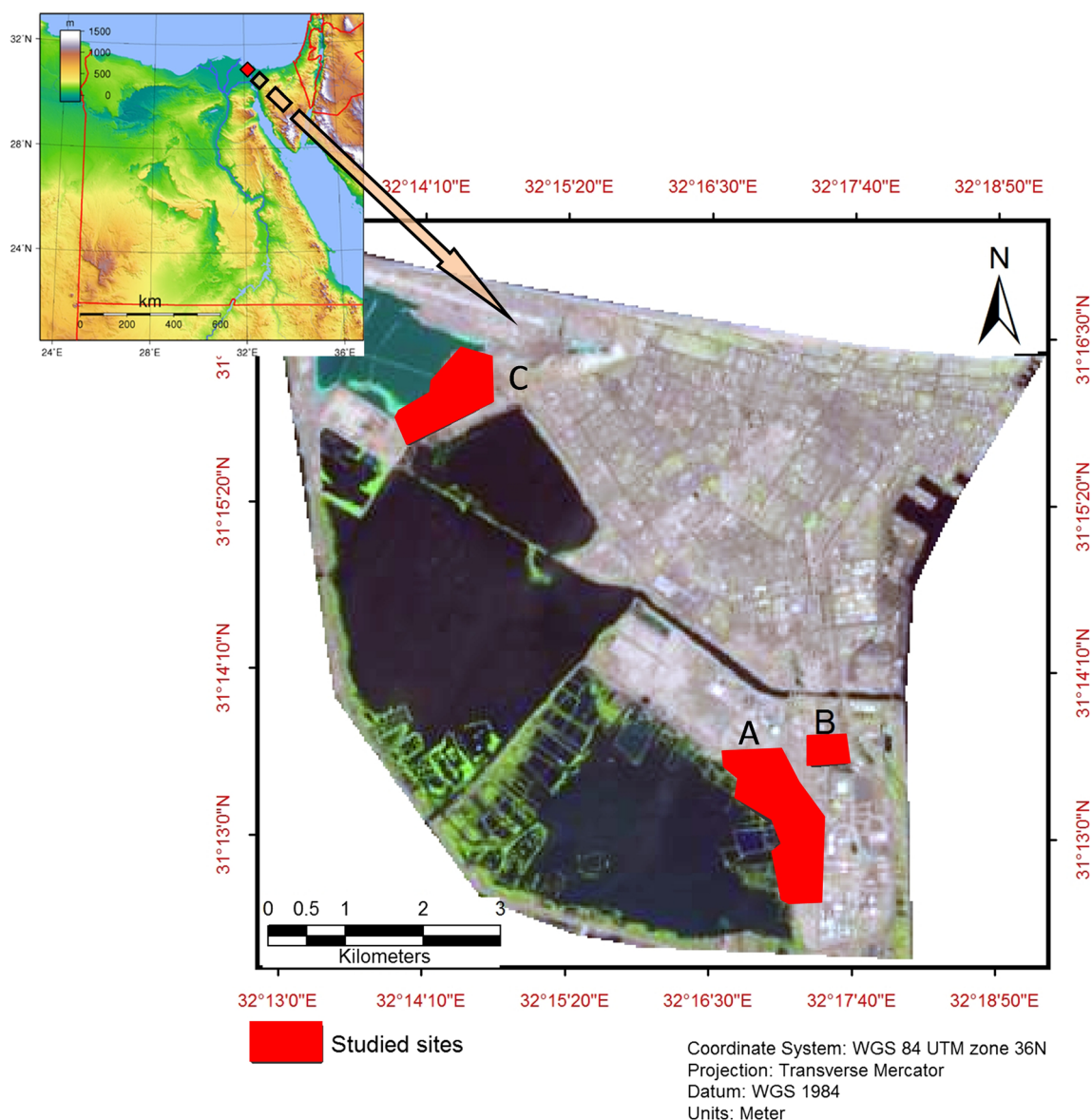


Fig. 1 Location map of the studied sites (a, b, and c) in Port Said city

plant (Fig. 1). Systematic detailed ground gamma ray spectrometric survey was carried out to measure the vertical and lateral spatial distribution of the radioelement concentration and to measure the dose rate using the RS-230 BGO gamma ray spectrometer provided by Radiation Solutions INC.

The spectrometer uses a 6.3 cu in bismuth germinated detector which provides typically $\times 3$ equivalent performance over comparably sized sodium-iodide detectors. The detector is previously calibrated and self-stabilized; it gives direct measurements of total radiation, uranium, and thorium concentrations in ppm, potassium concentration in percent, and the dose rate. Both the spectrometer function and variations in background radioactivity have been checked daily at an established reference station in the survey area. The assay time was set to 2 min to achieve the best accuracy. The assay data were stored

on the internal memory of the spectrometer which is then dumped into the PC using the GeoView software delivered with the spectrometer (Radiation Solution INC. 2009).

The locations of the measuring points were identified using a Garmin GPS model 71 champ and stored on the internal memory of the GPS then dumped into the PC coincide along with the assay measurements of the gamma ray spectrometer. Both data sets were merged into one single sheet for each site, which is then used to produce the radiometric maps for total count, potassium, uranium, thorium, and dose rate.

Soil samples

Eighteen soil samples have been collected from the studied sites (A, B, and C) to measure the activity concentrations of

Ra-226, Th-232, and K-40. These measurements were determined using an n-type coaxial high-purity germanium (HPGe) detector, Canberra model in Gamma Irradiation Unit—Nuclear Research Center—Atomic Energy Authority—Egypt. The detector has an efficiency of about 40 %, energy resolution of 1.9 keV, full width at the half maximum (FWHM) for the 1,332.3 keV gamma line of ^{60}Co and MCA with 8,000 channels. The detector is housed inside a low-background lead shield to reduce the background of the system. The energy calibration and absolute efficiency calibration of the spectrometer were carried out using known amounts of standard thorium, uranium, and potassium oxide samples dispersed in sand. The masses of the collected samples varied between 250 and 350 g. The samples were dried at 105 °C for 48 h to ensure that moisture is completely removed. They were then crushed and grounded to fine grain size of about 1 mm and sieved in order to remove the big sizes. The powdered samples were then packed in a standard plastic container (7.5×5.5 cm), and after property tightening the threatened lid, the containers were sealed with adhesive tape and left for 4 weeks before counting by gamma spectrometry in order to ensure that the daughter products of Ra-226 up to Pb-210 and of Th-232 up to Pb-208 in secular equilibrium with their respective parent radionuclides and then the gamma ray spectrum was accumulated to up to 900 min (Powell et al. 2006).

Heavy metal analysis

The samples that record highest concentrations in radioelements and total radioactivity will be analyzed to heavy metal concentrations (Pb, Zn, Cu, Ni, and Cd). This analysis was measured by atomic absorption spectrophotometer to find if there is a relation between these heavy metal concentrations and the source of this high radioactivity.

The collected soil samples were manually sorted to eliminate pebbles and coarse materials and air-dried under ambient conditions that are inside the laboratory to completely remove moisture. The dried soil samples were pulverized in a disc mill crusher. The resulting powdered samples were screened through a nylon sieve of 2-mm mesh size. Each five grams of soil sample was mixed and classified to quadrant in order to achieve a complete mixing of the sample. A weight 1-g portion of the fine ground sample was digested for complete dissociation with the acid mixture (i.e., H_2SO_4 , HCl , HNO_3 , and HF). The resulting sample digests were filtered into 100-mL volumetric flasks and made up to 100-mL mark with distilled water and closed up to a measuring volume of 100 mL.

The digested sample solutions of soil in 100-mL volumetric flasks were quantified for the heavy metals cadmium Cd, chromium Cr, manganese Mn, nickel Ni, copper Cu, zinc Zn, lead Pb, cobalt Co, Iron Fe, and vanadium V by using flame atomic absorption spectrometry (FAAS) by aspirating the samples into flame of Unicam 969 atomic absorption

spectrometer (at the Nuclear Material Authority of Egypt) fitted with deuterium lamp, over a 2-mm burner using pre-mix fuel (i.e., air/acetylene mixture).

Results and discussion

Results of the ground radioactive measurements of the studied sites (A, B, and C) in the Port Said city are represented in the form of graded colors maps for K %, U ppm, and Th ppm (Figs. 2b–d, 3b–d, and 4b–d). These maps show the spatial distribution features of the apparent and relative surface concentrations of the three radioelements K, U, and Th, in addition to the integral gamma ray response (T.C.) of the study area (Figs. 2a, 3a, and 4a). In the following, an inspection is given for each of these maps starting with some information about the background radiation.

In order to assess for the radioactivity of the study area, the natural radioactivity background should first be determined. This background represents the lowest level of radiation in the area. It was measured over the lake Manzala at 50 m from the shore, where the water depth at the measuring point is about 1.3 m. It was found that the background values are nearly the same as the minimum values of radioactivity in sites B and C. As the purpose of this work is to monitor the environmental radiation and the changes occurred due to human activities, the radioactivity background was chosen to be the minimum values that were measured in site B and C because the soil at this site is a natural soil as shown in Table 1.

Radioelement maps of site A

Total count (T.C.) map

Total count grade map (Fig. 2a) shows a wide range of radioactivity from a very low level reaching 13.4 uR in value to a very high level reaching 123.6 uR. The highest T.C. value (a_1) is located in the northeastern corner of site A where the total count value reaches 123.6 uR. During the field survey in this anomalous zone, it was noticed that the radioactivity value of the soil increased with depth and the values were duplicated at a depth of 30 cm. There are other two anomalies (a_2 and a_3) located to the northwestern part and southeastern part of this site with a maximum value of 27 and 24 uR, respectively.

Potassium (K %) map

Potassium grade map of site A (Fig. 2b) shows values varying from 0.01 to 3.8 %, with two highest anomalies

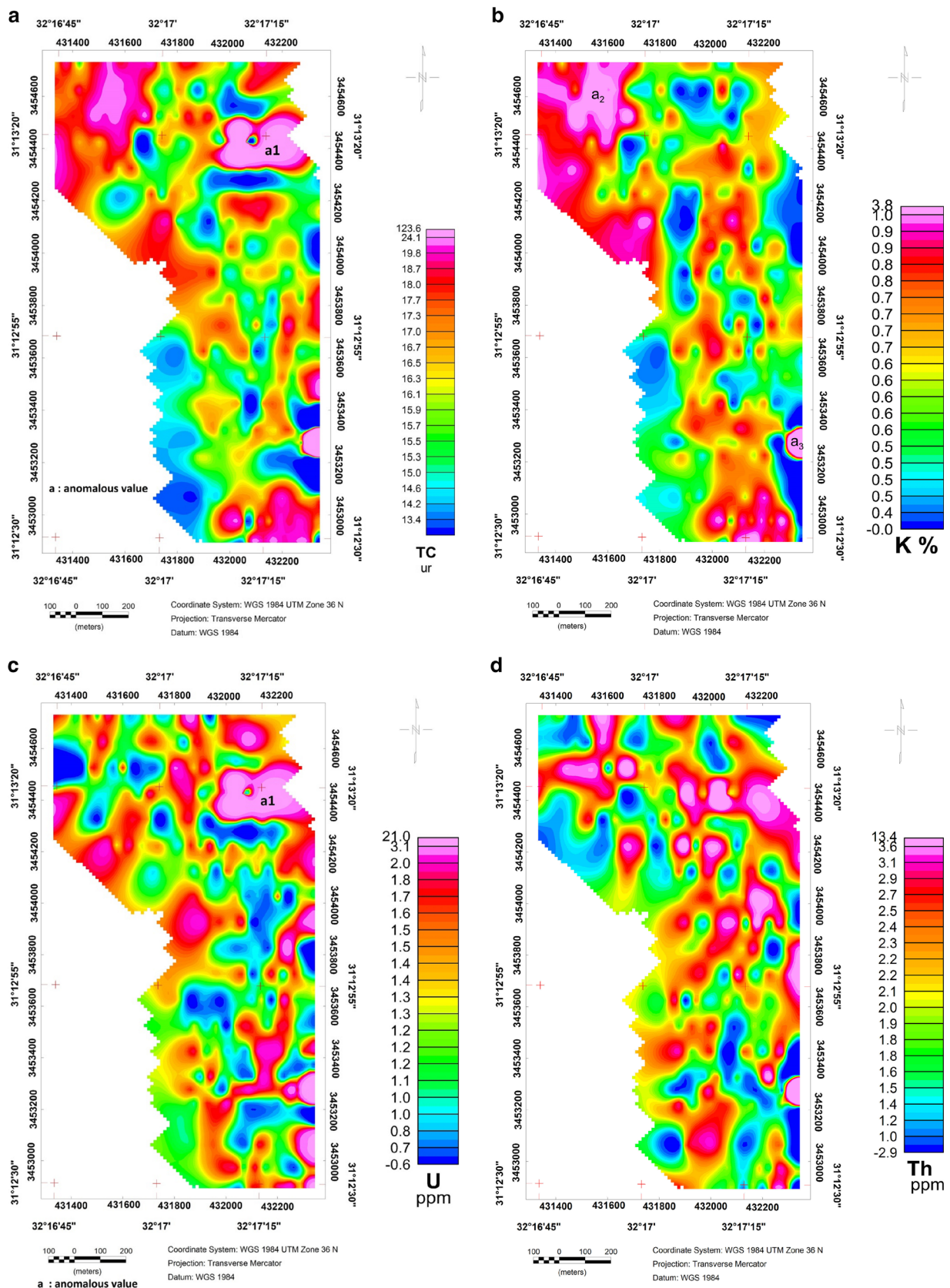


Fig. 2 a, b, c, and d show total count grade map (in uR), potassium grade map (in %), uranium grade map (in ppm), and thorium grade map (in ppm) of site (A) in the south of Port Said city, respectively

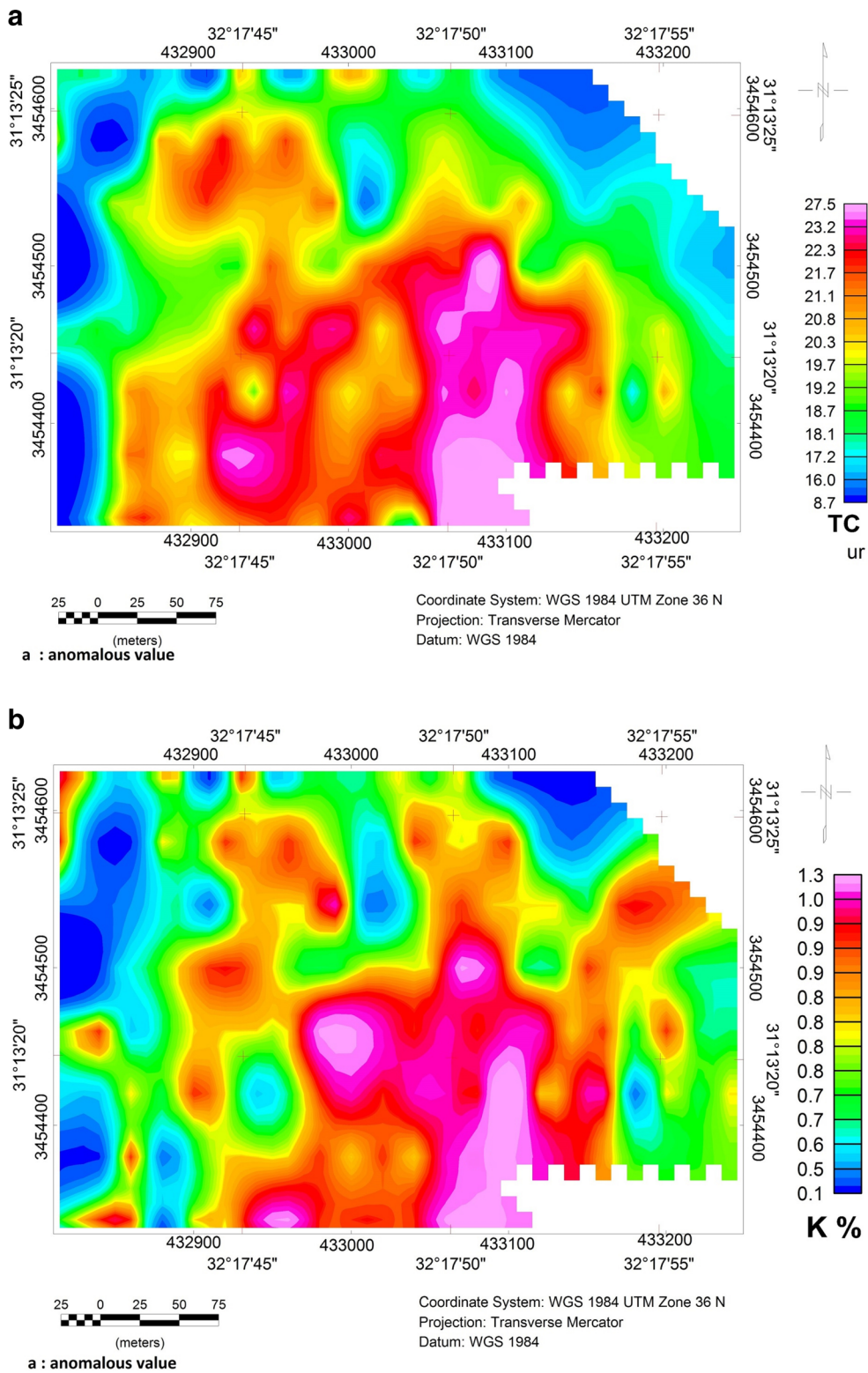


Fig. 3 a, b, c, and d show total count grade map (in uR), potassium grade map (in %), uranium grade map (in ppm), and thorium grade map (in ppm) of site (B) in the south of Port Said city, respectively

(a_2 and a_3) in the northwestern part and southeastern part of site A which coincide with the T.C. anomalies. It

was believed that the main reason of potassium in this site is due to both the dumped wastes of fertilizer and

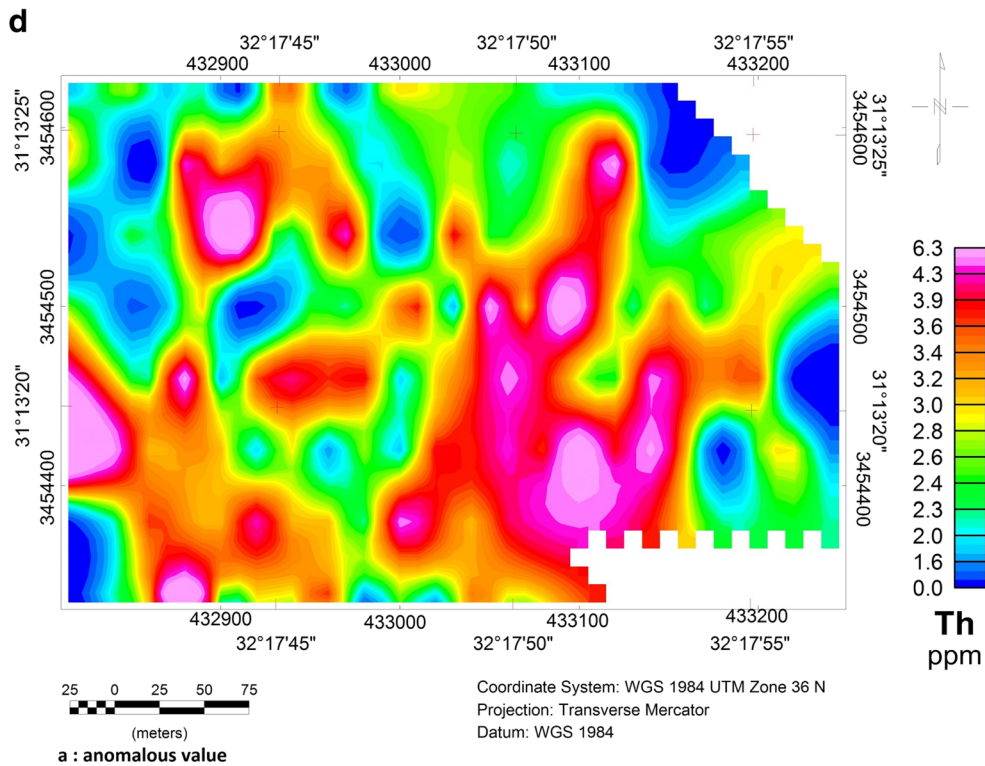
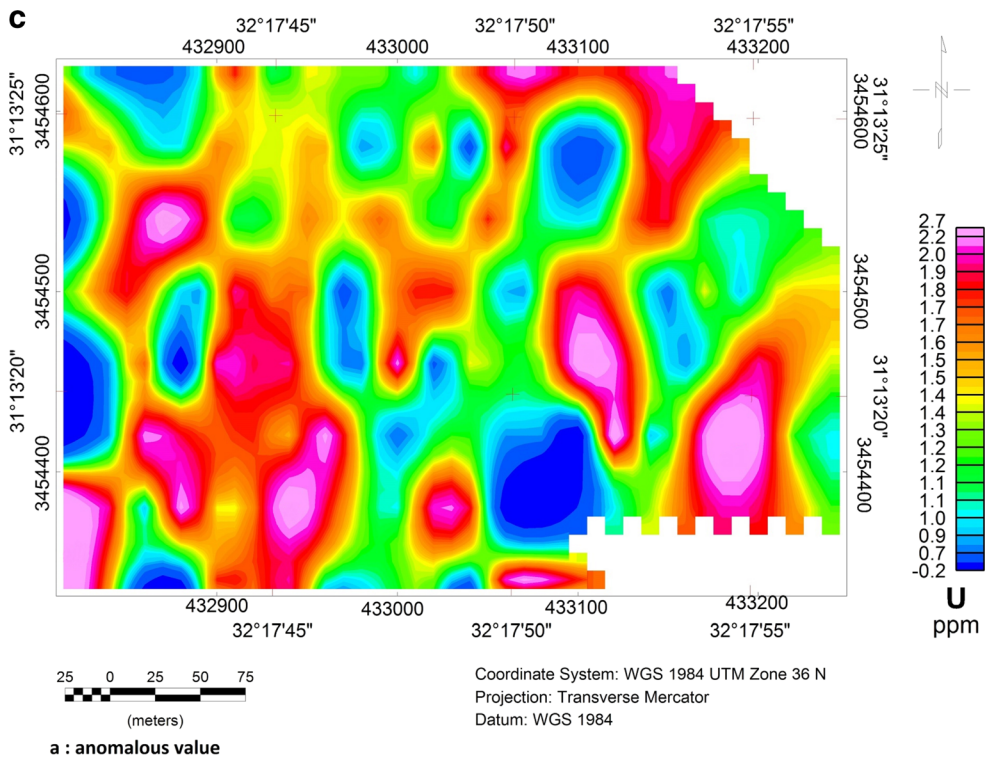


Fig. 3 (continued)

chemical factories and Paints Company nearer to the region of anomaly a_2 in addition to the accumulation

of pieces of granites, coming from a stone refine factory close to the region of anomaly a_3 , all are in site A. It is

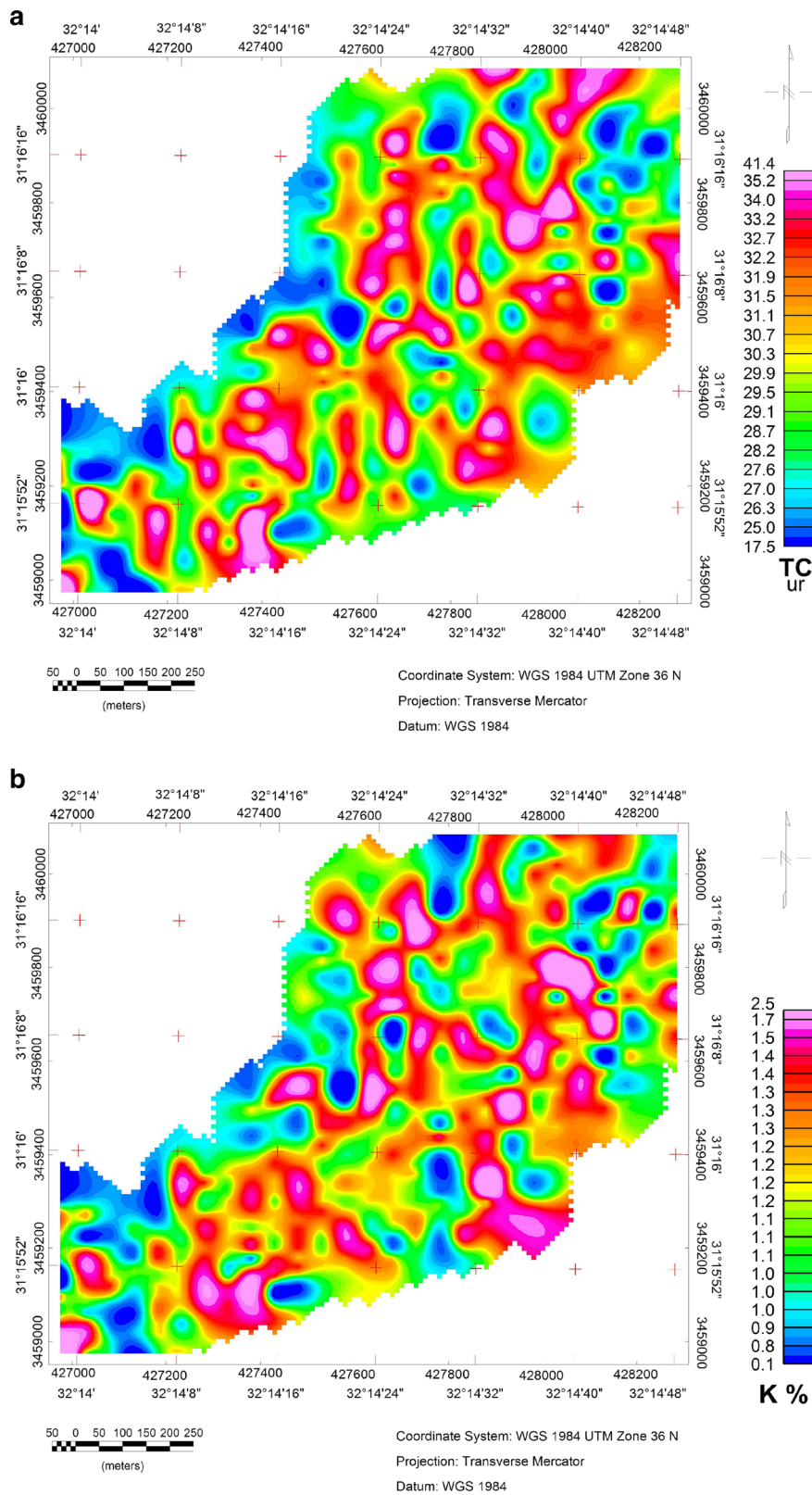


Fig. 4 a, b, c, and d show total count grade map (in uR), potassium grade map (in %), uranium grade map (in ppm), and thorium grade map (in ppm) of site (C) in the south of Port Said city, respectively

also noticed that the other high anomaly appeared in the T.C. map of site A disappeared in the K % map. This

may prove the artificial source containing traces of uranium and thorium.

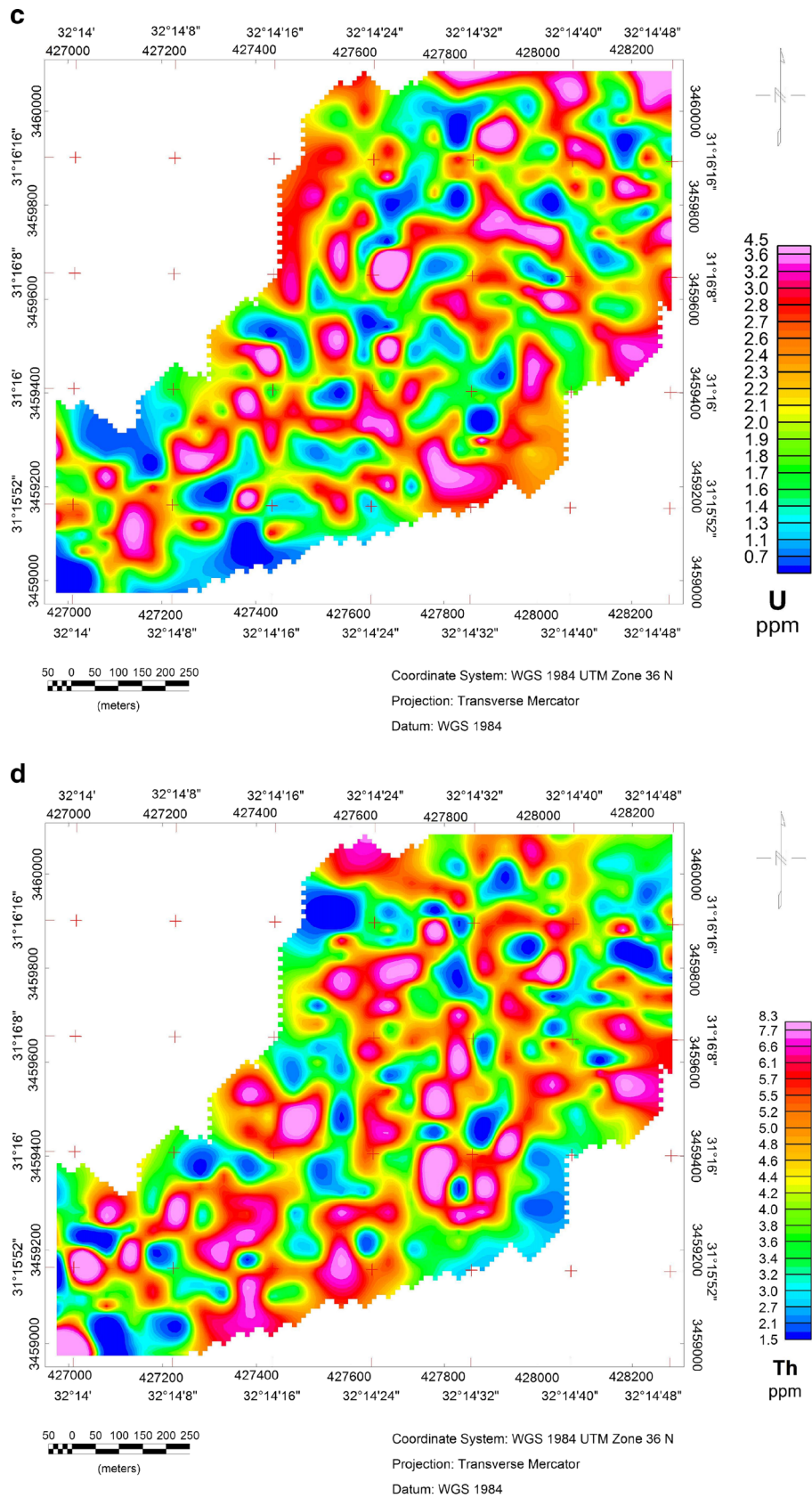


Fig. 4 (continued)

Table 1 Background values of T.C., K, U, and Th for the three studied sites (A, B, and C)

Sites	T.C. in (uR)	K in (%)	U in (ppm)	Th in (ppm)
A	8.7	0.012	0.2	0.01
B	7.4	0.014	0.1	0.02
C	8.2	0.01	0.08	0.01

Uranium (U ppm) map

Uranium grade map of site A (Fig. 2c) shows anomalies coincide with the T.C. anomalies. The uranium values range from 0.6 to 21 ppm, where the highest value (a_1) coincides with the T.C. anomaly that gives the main reason of uranium as the radioelement responsible for this anomaly. This anomalous value may indicate the presence of a buried artificial radioactive source rich in uranium, as site A is located nearby an area with condensed artificial activities containing fertilizer and chemical factories and Paints Company.

Thorium (Th ppm) map

Thorium grade map of site A (Fig. 2d) shows a wide range of values varying from 0.3 to 13.4 ppm. The anomalous values are generally located at the northern part of site A. The highest anomaly value coincides with the “U” and “T.C.” anomalies (a_1) at the northeastern part of this site. Other anomalies are mismatched with uranium anomalies and also are very limited and localized anomalies. This may be due to the difference in mobility behavior between uranium (under oxidation conditions) and thorium (Attia 2009).

Radioelement maps of site B

Total count (T.C.) map

Total count grade map of site B (Fig. 3a) shows a general trend of increment toward the center of this site. The values range from 8.7 to 27.5 uR.

Potassium (K %) map

Potassium grade map of site B (Fig. 3b) shows the same trend of the total count anomaly map with values ranging from 0.1 to 1.3 %. This gives an indication that the source of the high radioactivity in this zone may be due to the high concentration of K-40. This anomaly has nearly the same shape as the total count with the maximum value at the same position and trend. This reflects the presence of natural radioelement (K) due to the contribution of Nile and marine deposits in the study area (El-Gamal et al. 2004; Saleh et al. 2004).

Uranium (U ppm) map

Uranium grade map of site B (Fig. 3c) shows values considerably varying from 0.2 to 2.7 ppm with an average value of 1.5 ppm. Six anomalies, randomly distributed, could be noticed in this site. Also, the central part of this site shows a low uranium concentration which differs completely with the total count and potassium grade maps (Fig. 3a, b). This may indicate that the total count anomaly is not due to the concentration of uranium and may be due to the type of sediments as marine sediments. As many authors concluded, uranium and thorium isotopes have been used commonly as radiotracers for studies of marine sediments, including their sources, transport, and sedimentation rates in relation to the sedimentary and geochemical processes in the marine environment (Cochran and Krishnaswami 1980; Li et al. 1985; Cole et al. 1986; Suman and Bacon 1989).

Thorium (Th ppm) map

Thorium grade map of site B (Fig. 3d) shows values ranging from 0.01 to 6.3 ppm where there are three anomalies spreading in this site. One of them is directed to the north which nearly coincides with the total count grade anomaly. The second anomaly is located in the northwestern corner of site B. It has a spherical shape and a maximum value of 6.3 ppm. The third anomaly occupies the western part of this site with a maximum value of 6.3 ppm.

Radioelement maps of site C

Total count (T.C.) map

The total count grade map of site C (Fig. 4a) shows total radioactivity ranging from 17.5 to 41.4 uR. More than ten small anomalies are distributed in this site. These anomalies have irregular shapes, and all have nearly the same maximum value (i.e., 35 to 41 uR).

Potassium (K %) map

The potassium map of site C (Fig. 4b) shows a range of values from 0.1 to 2.5 % which nearly coincides with the T.C. anomalies. This may give an indication that the T.C. values may be associated with high concentrations of K-40. It is also noticed that the values of K-40 are lower than those in site A and higher than those at site B. Potassium anomalies have random shapes and trends. The shape, value, and size of the anomalies at site C are different from those of site B. This is because the anomalies in site B reflect the natural soil that formed by the effect of marine and Nile deposits, while the anomalies in site (C) look like many limited and localized anomalies spread all over the area, which reflects the artificial

soil (man-made soil) that is made by the random dump of wastes, as this site is considered the solid waste dump and sewage treatment plant of Port Said city.

Uranium (U ppm) maps

The uranium grade map of site C (Fig. 4c) shows a range of values varying from 0.7 to 4.5 ppm. These anomalies coincide with the total count and K-40 maps of site C and have also NW trends. It is also noticed that the maximum values of uranium in site C are lower than those at site A and higher than those at site B as mentioned above in the potassium grade map of site C.

Thorium (Th ppm) map

The thorium grade map of site C (Fig. 4d) shows a range of values varying from 1.5 to 8.3 ppm. The number of the thorium anomalies is larger and more restricted than those of the uranium, which also confirms the difference in mobility of these two radioelements (Attia 2009). The anomalies have a semi spherical shape, and they show a considerable coincidence with the total count anomalies in site C.

Table 2 summarizes the univariate analysis of the total count, potassium, uranium, and thorium values in the three studied sites. It is clear that site A is superior to site C in the all measured parameters, while site B has the lowest values. This is due to the type of soil, as in site B the soil is a natural soil which has a low radioactive background, while the soil in sites A and B are artificial soil that contains wastes from many sources especially the neighboring industrial area which is confirmed by high standard deviation values (i.e., the data points are spread out over a large range of values) of the total count, potassium, uranium, and thorium in these two sites.

Dose rate maps

Dose rate mapping survey was carried out in the three studied sites (A, B, and C) using the RS-230 BGO gamma ray spectrometer. The spectrometer has the capability to give measurements of dose rates in nano-Sievert per hour (nSv/h) (Radiation Solution INC. 2009). A field survey in the three

sites has been taken by the same survey parameters used in radioelement mapping survey. The constructed dose rate maps in milli-Sievert per year (mSv/year) are illustrated below (Fig. 5a–c) and listed as annual effective dose rates for samples collected from the three sites in Table 3.

The dose rate grade map of site A (Fig. 5a) shows a considerable weight variation of values ranging from 0.12 to 1.61 mSv/year. The maximum anomaly is coincident with the anomalous values of the U ppm, Th ppm, K %, and total count anomalies in the northwestern corner of site A (Fig. 2a–d). The value of 1.61 mSv/year is considered to be risky as it exceeds the safe limit (<1 mSv/year) stated by (IAEA 2005; Anonymous 2005; ICPR 2009). Accordingly, a remediation action should be conducted in these locations to get rid of these risky sources. The rest of the dose rate values in site A range between 0 and 1 mSv/year which is within the safe limit of radioactivity.

During phosphate fertilizer manufacture, the phosphate ore is firstly attacked by sulfuric acid to produce the phosphoric acid (green acid) where uranium will be mainly concentrated in the phosphoric acid. Several studies showed also that heavy metals in phosphoric fertilizers can accumulate in soil and become readily available to plants. In terms of fertilizer use, U, As, Cd, Cr, F, Sr, Th, and Zn are the elements that have a potential risk of accumulation in soil (Sauerbeck 1992; Modaihsh et al. 2004).

The dose rate grade map of site B (Fig. 5b) shows a range of values oscillating from 0.12 to 0.46 mSv/year. The highest value is concentrated in the center of the site, and it is coincident with the potassium and total count anomalies (Fig. 3a, b). However, the dose rate values in the rest of this site are considered to be less than 1 mSv/year and have not been affected by any anthropogenic origin and dumped wastes.

The dose rate grade map of site C (Fig. 5c) shows values ranging from 0.17 to 0.67 mSv/year. Some of the relatively high values coincide with the high values of K %, while the others are related to the U ppm and Th ppm anomalies. Also, the dose rate values in site C are in the safe level of radioactivity. But the anomalies of dose rate in site C differs from those in site B. Site C has many limited anomalies, because it is located near the solid waste dump and sewage treatment plant and may be contaminated by random wastes that slightly contain compounds of radioelements.

Table 2 Univariate analysis of T.C., K, U, and Th for the ground spectrometric survey of the three sites (A, B, and C)

Site	Total count (uR)			Potassium (%)			Uranium (ppm)			Thorium (ppm)		
	Min	Max	STD	Min	Max	STD	Min	Max	STD	Min	Max	STD
A	13.4	123.6	1.6	0.01	3.8	0.23	0.6	21	1.7	0.3	13.4	1.13
B	8.7	23.5	0.83	0.1	1.3	0.17	0.2	2.7	0.55	0.01	6.3	1.13
C	17.5	41.4	0.97	0.1	2.5	0.2	0.7	4.5	0.9	1.5	8.3	1.08

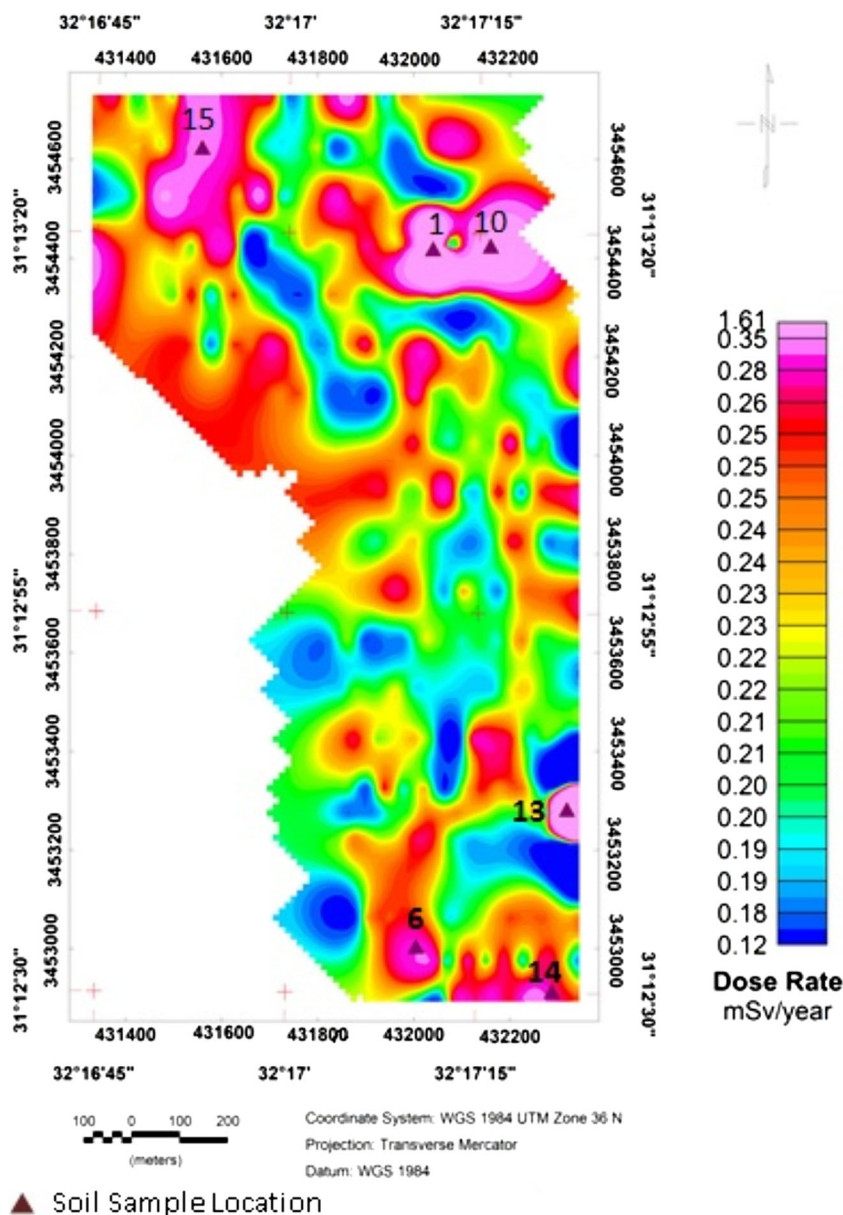


Fig. 5 a, b, and c show dose rate grade maps (mSv/year) of sites (A, B and C) in the south of Port Said city respectively

Activity concentrations

The radioelement concentrations are measured for 18 soil samples which have been collected from the studied sites (A, B, and C). They were converted from part per million (ppm) to activity concentrations in Bq/kg of the Ra-226, Th-232, and K-40 radionuclides using the conversion factors which were produced by IAEA (IAEA 2003). The annual effective dose was measured for all samples using the RS-230 BGO gamma ray spectrometer provided by Radiation Solutions INC. The results are listed in Table 3. The worldwide average concentrations of the radionuclides Ra-226, Th-232, and K-40 which were reported by UNSCEAR (2000) are 35, 30, and 400 Bqkg⁻¹, respectively.

The activity concentration of Ra-226 ranges from 18.03 to 398.66 Bq/Kg. The two highest concentrations of Ra-226 were recorded in samples (*a*₁ and *a*₁₀) which were collected from the risky zone in the northeastern part of site A. The activity concentration of Th-232 ranges from 5.28 to 75.70 Bq/Kg. Also, the highest concentrations of Th-232 were recorded in samples (*a*₁ and *a*₁₀). The activity concentrations of uranium and thorium confirmed what had been estimated from the highest anomalies in T.C., U, and Th grade maps of site A, whereas the activity concentration of K-40 ranges from 237.88 to 538.12 Bq/Kg. The highest activity concentration of K-40 was recorded in sample (*a*₁₃) which has been collected from the southeastern part of site A and confirms the highest

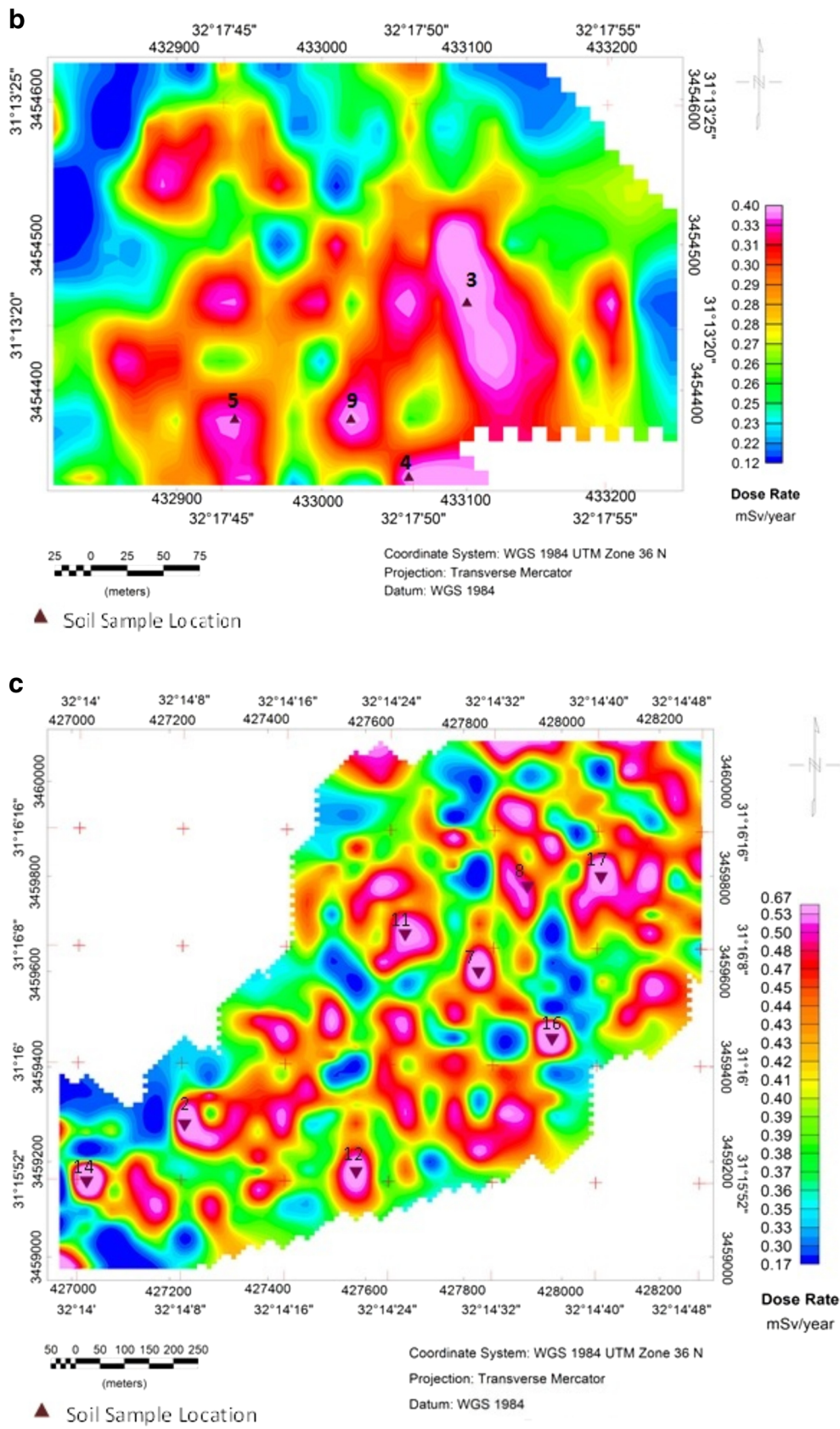


Fig. 5 (continued)

Table 3 Activity concentrations of Ra-226, Th-232, and K-40 Bq/kg and annual effective dose for the soil samples in the sites (A, B, and C)

Site	Sample no.	Ra-226 Bq/kg	Th-232 Bq/kg	K-40 Bq/kg	Annual effective dose rates (mSv/year)
A	1 ^a	348.27	54.00	237.88	1.61
	6	65.46	7.92	403.77	0.31
	10 ^a	398.66	75.70	273.56	1.61
	13 ^a	33.38	6.07	583.12	1.60
	15	75.27	9.10	464.34	0.35
	18	20.74	14.89	388.75	0.31
B	3	35.07	8.93	380.62	0.4
	4	29.02	5.28	507.06	0.37
	5	18.65	10.76	316.13	0.35
	9	18.03	12.95	338.04	0.4
C	2	53.11	10.15	244.14	0.67
	7	32.48	8.24	347.43	0.53
	8	30.13	9.26	494.54	0.49
	11	61.07	11.67	280.76	0.67
	12	40.34	10.27	307.31	0.62
	14	21.45	12.37	363.55	0.58
	16	37.35	9.48	399.54	0.56
	17	34.65	10.65	568.72	0.53
	MAX	398.66	75.70	583.12	1.61
	MIN	18.03	5.28	237.88	0.31

^a Samples collected from anomalous points of site A

anomaly of K-40 due to the dumped remnants of rock cuttings from the close factory.

The measured annual effective dose of the 18 samples in the three sites (A, B, and C), listed in Table 3, shows that the samples named 1, 10, and 13 reach the value of 1.61 mSv/year which is considered to be risky as it exceeds the safe limit (<1 mSv/year) stated by (IAEA (2005); Anonymous (2005); ICPR (2009)). Table 4 gives a clear picture for the great difference between the two sites (A and B) although these two sites are geographically very close together. The only essential difference of these two sites is that the first one is located inside the industrial zone and the second is located outside the industrial zone close to run way of the south entrance to Port Said city. Radioactivity background (T.C., K, U, and Th) in the two sites (A and B) are nearly very

similar, but the average concentrations of these radioelements are greater in site A than in site B, while the maximum activity concentrations (Ra-226, Th-232, and K-40 in Bq/kg) are found very high in some samples in site A than in site B.

Finally, annual effective dose rates are maximum in site A than in site B and exceeds the safe limit (<1 mSv/year) stated by (IAEA (2005); Anonymous (2005); ICPR (2009)).

Heavy metal analysis

Two samples (a_1 and a_{10}) were taken from the risky zone that exceeds safe limits and then analyzed to the concentrations of Pb, Zn, Cu, Ni, and Cd that were measured by atomic absorption spectrophotometer as shown in Table 5 (APHA 1998). The most common heavy metal contaminants are cadmium (Cd), copper (Cu), molybdenum (Mn), lead (Pb), nickel (Ni), and zinc (Zn) (Lasat 2002; USEPA 1997).

In general, the concentrations of the studied heavy metals in the two samples could arrange as $Mn > Zn > Ni > Pb > Cu > Cd$. These elements' concentrations are in the range of phosphate fertilizer products that suggested a dumped man-made wastes in site A. Cd concentration shown in Table 5 which was recorded in the two samples (a_1 0.90 ppm and a_{10} 0.97 ppm) is in the known range of Cd concentration in the phosphate fertilizers. Cd occurs naturally as a minor constituent of phosphate ore and is transferred in variable amounts to phosphorus fertilizers during manufacturing process (Alloway and Steinnes 1999). Rock phosphate contains large amounts of zinc and cadmium as impurities (McMurtry et al. 1995; Kpombekuo and Tabataba 1994). Zn concentration in Table 5 that was recorded in the two samples (a_1 40 ppm and a_{10} 42 ppm) is also in the known range of Zn concentration in phosphate fertilizers. Approximately one-half to two-thirds of the Zn is of anthropogenic origin, whereas for Cd, the value may exceed 90 % (De Carlo and Spencer 1995).

Conclusion

Ground gamma ray spectrometry measurements have been carried out in three sites (A, B, and C) in the

Table 4 Comparison between site A and site B

Close sites	Background values				Average concentrations of T.C., K, U, and Th				Maximum activity concentrations			Maximum annual effective dose rates	
	T.C. (uR)	K (%)	U (ppm)	Th	T.C. (uR)	K (%)	U (ppm)	Th	Ra-226	Th-232 (Bq/kg)	K-40	(mSv/year)	Safety
Site A	8.7	0.012	0.2	0.01	68.5	1.9	10.8	6.85	398.66	75.7	583.12	1.61	Unsafe
Site B	7.4	0.014	0.1	0.02	16.1	0.7	1.45	3.15	35.07	12.95	507.06	0.40	Safe

Table 5 Heavy metal concentrations in (ppm) of samples (a_1 and a_{10}) in the risky site (A)

Site	Sample no.	Heavy metals concentrations in (ppm)					
		Cu	Pb	Cd	Zn	Ni	Mn
A	a_1	12	16	0.90	40	22	70
	a_{10}	11	17	0.97	42	26	67

industrial area of Port Said city. Results indicated that the measured values for the radioelement concentrations in site B have the lowest values as the soil type in this site is an original soil which is characterized by low radioactivity, while the highest radioelement concentrations were recorded in site A as the soil in this site is an artificial soil, composed due to the dumping of wastes from many sources especially the industrial area. This may indicate that these wastes have higher radioactivity than the natural background.

In order to assess for the radiologic effect of these concentrations, the activity concentrations of Ra-226, Th-232, and K-40 in soil samples have been measured and the radiological hazard parameters were calculated. Results indicated that soil samples of sites B and C fall within the world ranges for soil in areas with normal levels of radioactivity, but for site A, a highly radioactive zone (at the northeastern part of this site) is detected which represents a potential contamination and foretells a possible future risk.

The obtained results, in the light of the industrial activities in the area, give an indication about the possible source of these contaminants as the raw materials used in steel production are iron ore, coal, and limestone (HPS 2002). They each contain low levels of naturally occurring radionuclides potassium-40, uranium-238, uranium-235, and thorium-232 decay chains. The steel production process results in releases of materials to the environment with concentrations of NORM that are enhanced with respect to the raw materials used (NRPB 2003). Accordingly, it is expected that the source of these contaminants is the steel industry in the area.

Element concentrations especially Cd and Zn that resulted from heavy metal analysis are in the range of phosphate fertilizer products that suggested a dumped man-made waste in site A.

A remediation action is highly recommended, making in site A, by replacing the contaminated soils by another clean soil and treat the contaminated soil. Also, it is suggested to design a monitoring program that should be carried out periodically in this site to assess for any future contamination by comparing the current radioelement maps with any new consequent maps in this area.

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