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LOCAL ENHANCEMENT OF THE NATURAL RADIONUCLIDES IN BEACH SAND IN AL-ARISH, EGYPT

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Radionuclide concentrations in Al-Arish city area, north Sinai, Egypt, were measured. The activities of the ^{232}Th and ^{238}U series in soil samples from two locations in harbor area and a location ~3 km east the harbor were higher than those from other sites, with activities of 81.6±0.7, 50.6±0.5, and 24.8±0.4 Bq/kg for ^{222}Rn and 98.9±1.3, 67.2±1.1, and 52.2±0.8 Bq/kg for ^{228}Ac. The average contents for the other sites were 8.0±7.2 and 7.5±6.7 Bq/kg for ^{222}Rn and ^{228}Ac, respectively. No significant difference in the ^{40}K activity was found between these two groups. The measured depth profiles in the site with the highest activity are studied. Samples with the smallest grain size (<0.2 mm) were found to contain the highest level of activities, due to its high content of black sand.

Keywords: natural radionuclides, profile distribution, coastal environment, Sinai, Egypt.

INTRODUCTION

Studies of natural environmental radioactivity are of great important and interest in health physics for practical and fundamental scientific reasons. The natural radiation to which humans are exposed consists of two distinct components: the internal exposure originated from radionuclides in diet and from inhalation of radon gas, and the external one generated by cosmic rays and γ-rays emitted by radioactive elements in soils, rocks, and building materials. The terrestrial radiation is originated from the so-called primordial radioactive nuclides that were made in the early stage of the formation of the solar system. ^{238}U series (^{214}Bi, ^{214}Pb, and ^{226}Ra), ^{232}Th series (^{208}Tl, ^{212}Bi, ^{212}Pb, and ^{228}Ac) and potassium are, however, the main elements contributing to the natural terrestrial γ-ray radioactivity.

Beach sands are weathering-resistance remainders of geological formations, which may have come to their place by rivers and are deposited on the beaches by actions of waves and currents. In the beach environment, metal-rich sands are generally found in the upper zone of the beach, or back shore, as waves concentrate the heavy minerals (HMs) that are not easily removed back to the sea by the low-energy backwash flow [1]. Beach sand minerals are the term generally used for definition of beach deposits of these accessory minerals. HMs are accessory constituents having high specific gravity. They are hard and stable minerals such as garnet, zircon ($ZrSiO_4$), monazite (Ce, La, Nd, $Th(PO_4)$), rutile (TiO_2), and ilmenite ($FeTiO_3$) [2,3]. They are economically important coastal placers [2]. While HMs are characterized by

high concentrations of the ^{232}Th and ^{238}U in their crystalline structure, the light minerals (e.g., quartz and feldspar) have relatively low concentrations of these radioisotopes.

MATERIALS AND METHODS

1. Study area

The whole area lies in Al-Arish district, governorate of Northern Sinai. Al-Arish is the biggest city in north Sinai. It has one third of the total Sinai population. Figure (1) shows the area under study in the surroundings of Al-Arish city (31.13° N and 33.80° E). A total of 11 locations were included in this study. The western limit of the studied area was about 15 km west Al-Arish city in the eastern limit of the Zaranik protected area (location 1). The locations 10 and 11 were both about 6 km east and south Al-Arish City, respectively. The sampling locations 4-8 are within Al-Arish city area. Locations 6 and 7 are directly in the western and eastern neighborhood of the local harbor, respectively. Surface samples collected from the locations marked 1–9 are beach sands while locations 10 and 11 are dune sands.

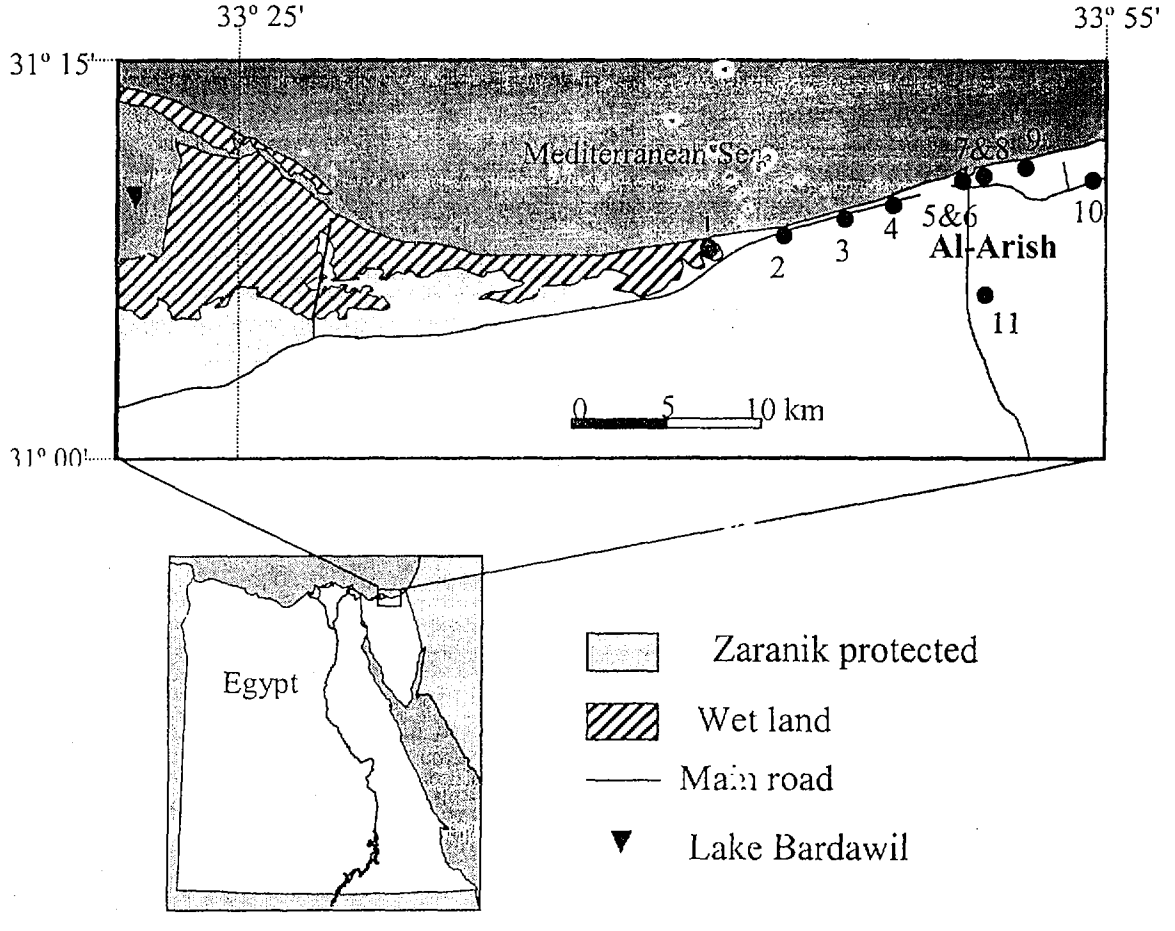

Fig. 1. The map of the study area in the surrounding of Al-Arish city showing the 11 sampling locations.

2. Sampling

Sampling sites were selected to be open, reasonably level. No samples were taken close to a field boundary, trees, buildings, or other obstructions. The sand at the sites must not be covered with grass or other vegetations. Sites were not chosen based on their

radioactivities. In each location, a sample was obtained from five positions, one central and four close to the corners of a square of ~10 m length. Samples were taken from the upper 10 cm of the surface layer. Based on the analysis of these samples, second sampling was carried out at the two locations with the highest (#8) and lowest (#5) activities. In addition, core samples were taken twice from location 8. The first core sample (May 2001) was down to a depth of 30 cm (3 layers of 10 cm each), whereas the second one (January 2002) extended to 1m (10 layers of 10 cm each).

3. Sample preparation

The five collected samples at each location were thoroughly mixed together at the laboratory, air-dried for several days under ambient temperature and hand-cleaned to remove stones, leaves, and other foreign particles. The samples were then oven dried at 100°C until constant mass, homogenized, screened with a laboratory test sieve 1.0 mm. Each sample (~650 cm^3) was placed in a PVC cylindrical container of diameter ~10.3 cm and height ~8 cm. The containers were sealed tightly with a thick vinyl tape around their necks to limit as far as possible any gas escape from them, and stored for a minimum period of four weeks to allow equilibrium between ^{222}Rn and its daughter products. The pH and total organic matter content (TOM) was determined for each sample. For grain size study, some samples were directly sieved after drying into five classes of particle size, namely, less than 200, 200–315, 315–400, 400–500, and 500–640 μm.

4. Activity measurements

The activity concentrations in the samples were measured using a 204 cm^3 p-type HPGe detector (Tennelec) with a relative efficiency of 50%. It has an energy resolution (FWHM) of 1.92 keV for the 1332 keV ^{60}Co γ-line. The detector was shielded using a 10 cm thick low-background lead shield (Canberra 747E). The amplified signals of the detector were acquired with an 8k ADC MCA (Oxford PCA3). The γ-ray background spectra were measured frequently to check the stability of the background and to correct the net count rate of the selected γ-ray photopeaks. The minimum measuring time was 18,000 s. The statistical errors (1σ) in the net peak area were 3–5%. The efficiency calibration method was carried out using standard radioactive point sources and KCl [4,5]. Both the detector efficiency calibration curve and the gas-tightness of the containers were examined using the certified reference materials IAEA soil-6, 326, and 327. The activity of the various isotopes present in these certified materials, obtained by the use of the measured detector efficiency, was in agreement with the values provided by the IAEA with ~3% deviation in the worst case. This includes ^{226}Ra activity, which supports the good tightness of the sample containers.

RESULTS AND DISCUSSION

1. Radionuclides concentrations

The pH of the sand samples ranged from 7.8 to 8.4, which is considered weak base. The TOM was found to be less than 1% for all samples. An example of γ-ray spectrum of a sand sample with relatively high ^{238}U and ^{232}Th activities is shown in Figure (2). The activities of some samples were repeatedly measured after sealing up to 60 days. The variations of the ^{40}K, ^{222}R,n and ^{228}Ac activities with time are shown in Figure (3) for three samples. Naturally, the activity of ^{40}K and ^{228}Ac in the same sample did not vary with time. On the other hand, the measurements of ^{222}Rn show that radon emanation is negligible, which suggests the possibility of excluding the sample sealing and storing from the sample

preparation procedure. This agrees with other reported measurements of radon emanation from sand samples (e.g., [6,7]). Based on these multi-measurements, the coefficient of variation (CV) in the measured ^{40}K, ^{222}Rn, and ^{228}Ac activities were 5-10, 5-10, and 2-7%, respectively.

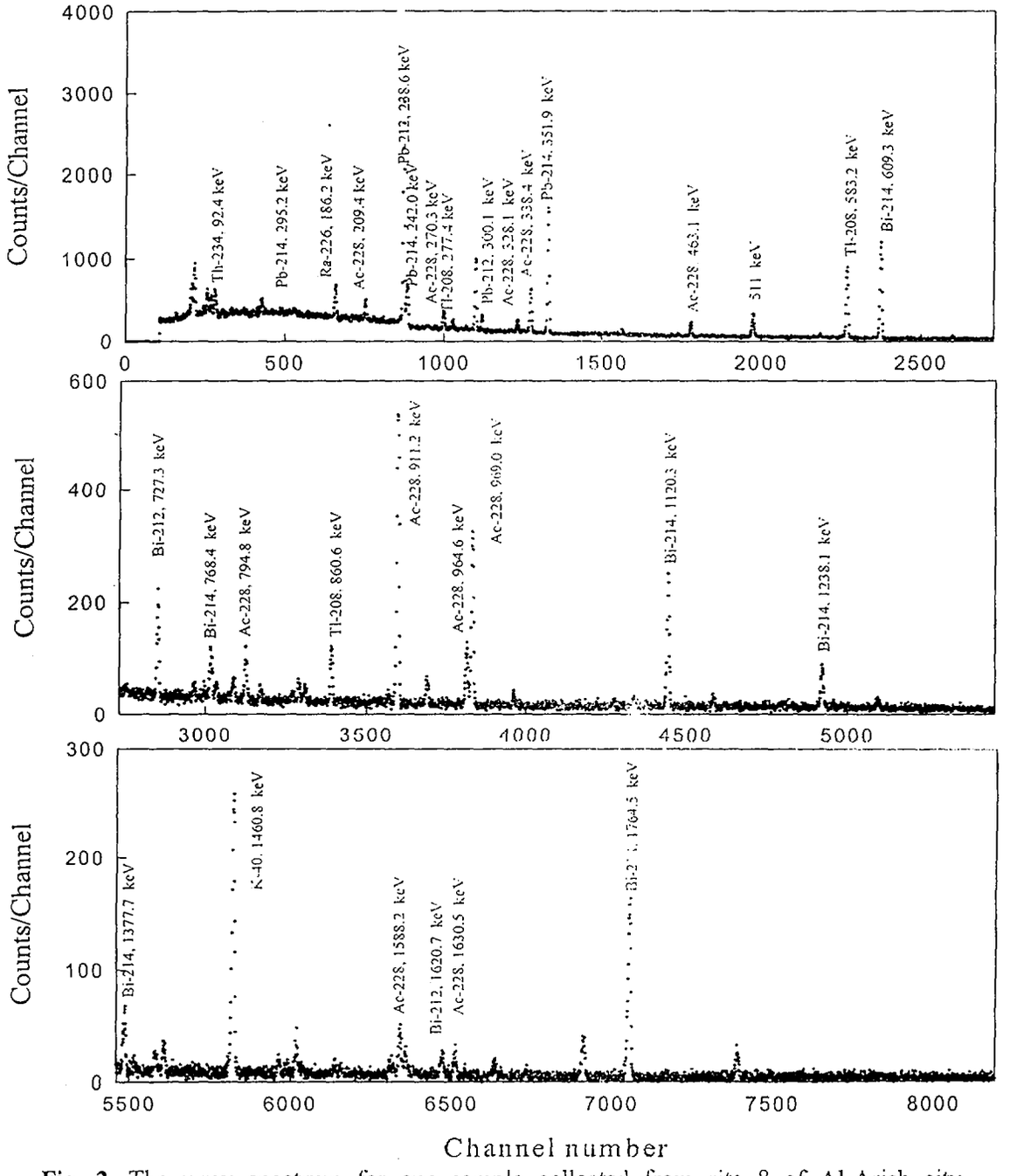

Fig. 2. The γ-ray spectrum for one sample collected from site 8 of Al-Arish city represents isotope identification and γ-ray energies of the well-defined peaks.

Figure (4) shows the measured activity concentrations of ^{222}Rn, ^{228}Ac, and ^{40}K in Bq/kg dry weight (dw) for each location. The average concentrations of ^{226}Ra (^{222}Rn) and ^{232}Th (^{228}Ac) for the three locations 7, 8, and 9 were 52.3±28.5 and 63.8±37.0 Bq/kg dw, respectively. These values are significantly larger than the average concentration of the rest of the locations; 8.0±7.2 Bq/kg dw for ^{226}Ra and 7.5±6.7 Bq/kg dw for ^{232}Th. On the other hand,

the average activities of ^{40}K were generally comparable for the three locations 7, 8, and 9 (101.4±10.3 Bq/kg) and the rest of the locations (122.0±55.8 Bq/kg). Only the two locations 10 and 11 showed a relatively elevated level of ^{40}K (~72% higher than the average value). Table 1 gives the statistics summary for ^{214}Bi and ^{214}Pb (^{222}Rn daughters), ^{228}Ac and ^{212}Bi (^{232}Th daughters) and ^{40}K activities in sand samples. The activities of Egyptian sand used as building materials are relatively lower than the present values (activities were 7.8, 9.0, and 155 Bq/kg for ^{222}Rn, ^{228}Ac, and ^{40}K, respectively) [8].

Location 8 had the highest activity (98.9 Bq/kg for ^{228}Ac and 81.6 Bq/kg for ^{222}Rn) and location 5 had the lowest activity (2.7 Bq/kg for ^{228}Ac and 2.6 Bq/kg for ^{222}Rn); see Fig. 4. Sampling was carried out ~8 month later from these two locations. The ^{40}K activity in the two samples collected from both locations did not show any significant variation. On the other hand, the measured activities in the second sampling were 4.5 Bq/kg for ^{228}Ac and 4.1 Bq/kg for ^{222}Rn for location 8 and 4.0 Bq/kg for ^{228}Ac and 3.4 Bq/kg for ^{222}Rn for location 5. This indicates the measurements may vary from place to place and from time to time due to the dynamic of sand system; radionuclides migration and beach environment, e.g. sea waves, wind, etc.

Table 1. Summary statistics for ^{214}Bi, ^{214}Pb, ^{228}Ac, ^{212}Bi and ^{40}K activities in sand samples.

	Bi-214	Pb-214	Ac-228	Bi-212	K-40
Mean±SD	20.8±25.9	19.5±24.4	22.6±31.5	23.4±33.3	118±50.1
Min	84.9	78.9	98.8	100.6	221.3
Max	3.0	2.3	2.3	2.7	85.2
Kurtosis	3.24	2.93	2.93	2.09	2.05
Skewness	1.88	1.83	1.91	1.69	1.86

The radiation hazard index (radium equivalent; Ra_{eq}) was used to evaluate the γ-ray radiation hazards due to the naturally occurring radioactive materials (NORM) as given by (Beretca and Mathew, 1985) [9]

$$Ra_{eq} = C_{Ra} + 1.43 \times C_{Th} + 0.077 \times C_K. \quad (1)$$

The dose rates in air 1 m above the ground surface for the studied sites were calculated using the following formula (UNSCEAR, 1988) [10]

$$D = 4.27 \times 10^{-2} C_U + 6.62 \times 10^{-2} C_{Th} + 4.32 \times 10^{-3} C_K \ (10^{-8} \text{ Gy/h}) \quad (2)$$

where C_U, C_{Ra}, C_{Th} and C_K are the activities of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K, respectively. It is assumed that ^{238}U and ^{226}Ra are in radioactive equilibrium. The effective dose equivalent was calculated by assuming a conversion coefficient of 0.72 Sv/Gy for the absorbed dose in air to the effective dose in the human body and an in-door and out-door occupancy factors of 0.8 and 0.2, respectively [10]. The calculated radium equivalent, absorbed dose rate in air due to the presence of the natural radionuclides in the sand and effective dose equivalent are given in Table 2. Even for the three high activity sites, the radium equivalent and the effective dose equivalent values are much lower than the world allowed of 370 Bq/kg and 410 μSv/y, respectively [10].

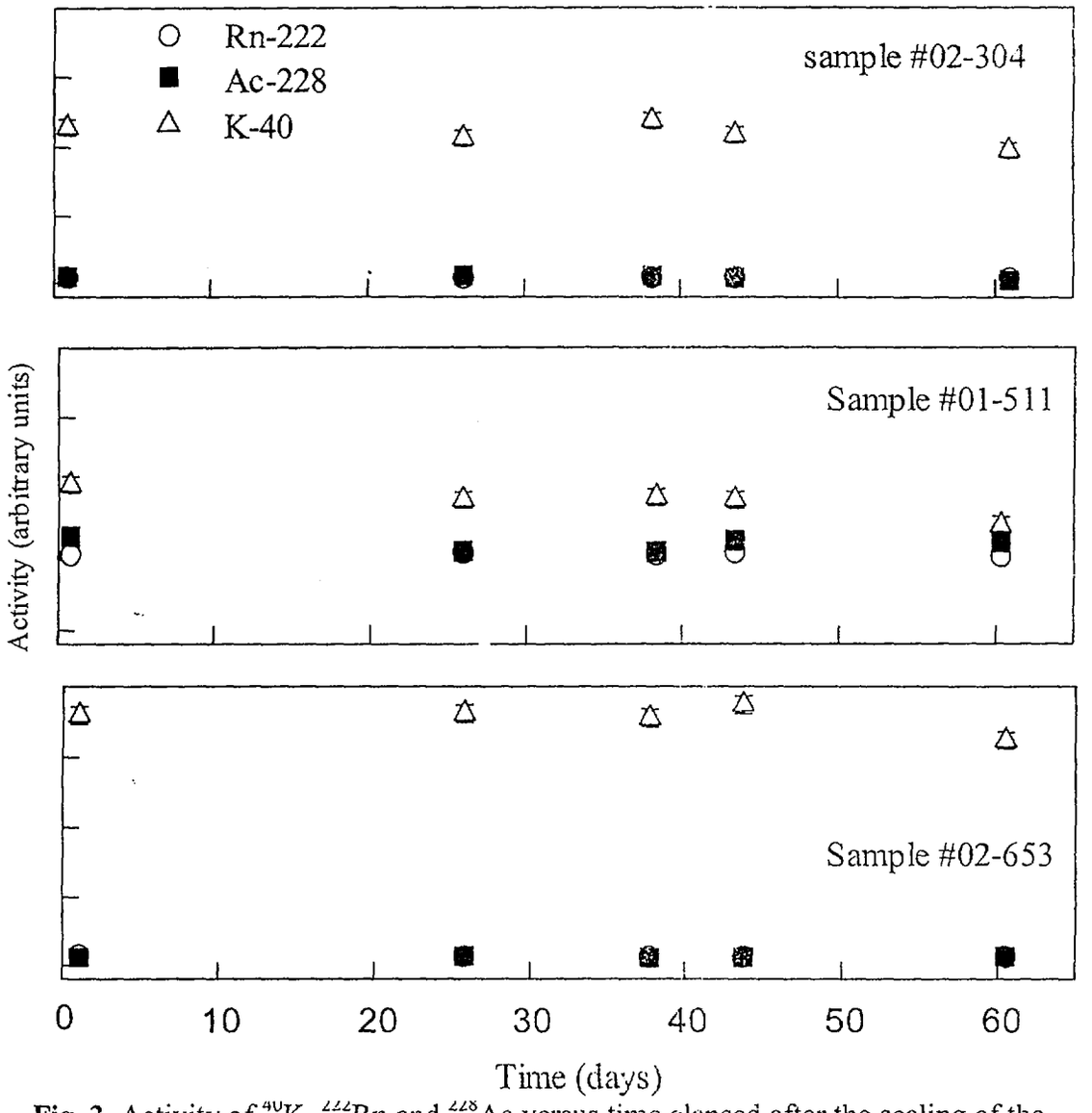

Fig. 3. Activity of ^{40}K, ^{222}Rn and ^{228}Ac versus time elapsed after the sealing of the sample container for three samples.

2 Activity Dependence on the Grain Size

Sand sample collected from location 7 was also subjected to grain size analysis. It was separated into five classes without crashing, namely <200, 200–315, 315–400, 400–500 and 500–640 μm. The activities in each grain size were determined as indicated before (see sec.2.4). Fig. 5 shows the mean contribution of activity concentrations for the individual grain-size fractions. It is clear that the activity of sand with grain size <200 μm is much higher than that in other grain size fractions due to the possible presence of black sand [11]. The tendency of ^{238}U and ^{232}Th series to increase in concentration with decreasing particle size was also found in other studies of beach sand [12]. On the other hand, ^{40}K activities did not show any strong dependence on the grain size in the smallest three grain size ranges and drops with increasing grain size in the two large ranges. The fraction of sand with grain size larger than (500 μm) is very poor in NORM radioactivity.

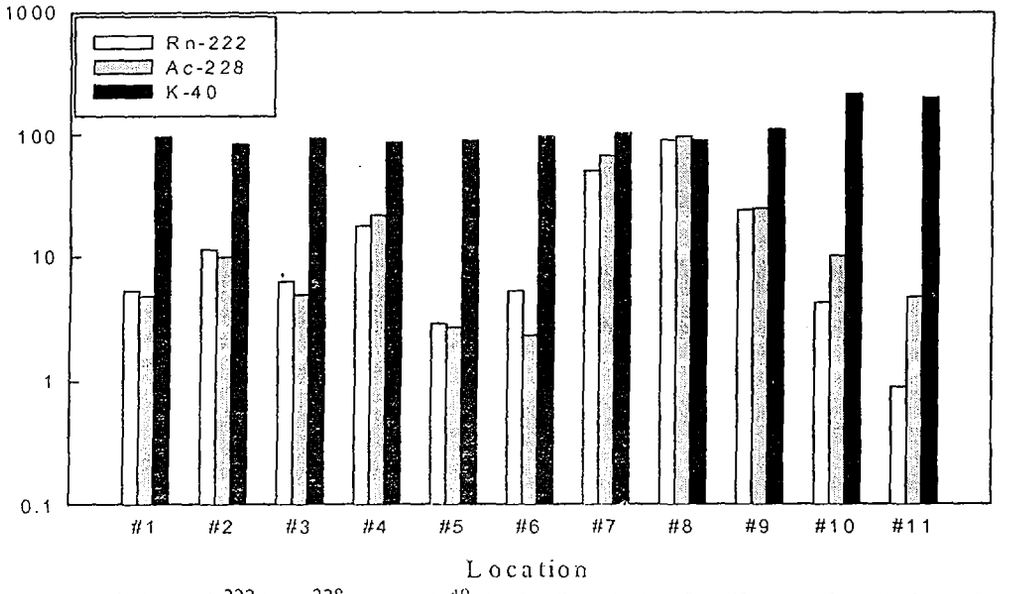

Fig. 4. Activity of ^{222}Rn, ^{228}Ac and ^{40}K (Bq/kg dw) of soil samples in locations included in the present study.

Table 2. The calculated radium equivalent, absorbed dose rate, and effective dose equivalent for each location of the studied area.

Location	Ra_{eq} (Bq kg^{-1})	Dose rate (nGy h^{-1})	Eff. D. eq. (μSv y^{-1})
1	17.0	8.7	11.0
2	32.0	15.1	19.0
3	19.2	10.0	12.7
4	62.8	26.2	33.1
5	13.5	7.0	8.8
6	14.1	8.0	10.1
7	154.4	70.7	89.3
8	229.9	108.5	136.9
9	69.3	32.0	40.5
10	39.0	18.0	22.8
11	26.9	12.2	15.4
recommend limit	370		410

3 Radionuclides Migration

The downward migration of radioisotopes is important from the radiation protection point of view because this process results in natural reduction of the external exposure rate from deposited γ-ray emitting isotopes. Therefore, the depth profile of radioisotope concentrations in the area of Al-Arish harbor (location 8) was measured twice. The sampling times were May 2001 and January 2002 with a time gap of ~8 months. In the first depth profile investigation, three core samples from depth 0–30 cm were analyzed (each from a layer of 10 cm thickness). While in the second, the measurements were extended up to a depth of one meter with ten core samples. The measured distributions of natural radionuclides are presented in Fig. 6. For ^{222}Rn and ^{228}Ac, a peak at 15 cm depth was observed in the first

profile, while it migrates downward to 25 cm in the second one. No obvious trend can be observed from the ^{40}K activity. In addition, no change of the TOM with the depth was found. The vertical distribution of ^{222}Rn and ^{228}Ac are strongly correlated (r=0.99). The ratio $^{222}Ra/^{228}Ac$ is consistent with unity (0.9), which indicates the same origin of both series. The vertical distribution of ^{222}Ra and ^{228}Ac in sand from three location on the coast (2,3 and 4) and two location far from the coast (10 and 11) were also measured up to 30 cm. All these locations show relatively low variation with depth.

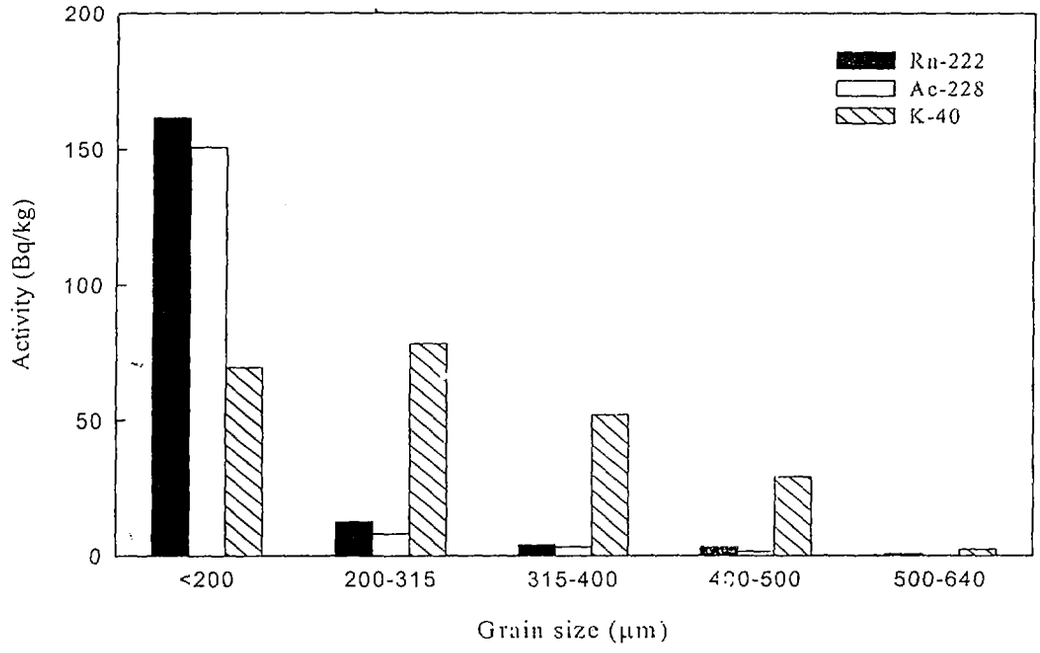

Fig. 5. Dependence of the NORM activities on grain-size for sand sample collected from location #7.

CONCLUSION

The results of this study indicate that the activities of the ^{238}U and ^{232}Th series in samples obtained from the harbor area of Al-Arish city and about 3 km east are higher than those from other sites. The highest site in the harbor area had activities of 81.6±0.7 Bq/kg for ^{222}Rn and 98.9±1.3 for ^{228}Ac. The ^{40}K contents was higher in dune sand with an average value of 211.7±13.6 Bq/kg compared with 97.1±9.8 Bq/kg for beach sand. Although, it is remarkable that the present activity concentrations of ^{222}Rn, ^{228}Ac and ^{40}K of the samples are close to the world average of 25 (10-50), 25 (7-50) and 370 (100-700) Bq/kg, respectively [10] except for some locations that possibly contained black sand. The present results show that the beach sand in these locations are not radiation hazardous. However, it is not advised to use sand from this area in building construction. Otherwise, the inhabitants will receive a higher external dose.

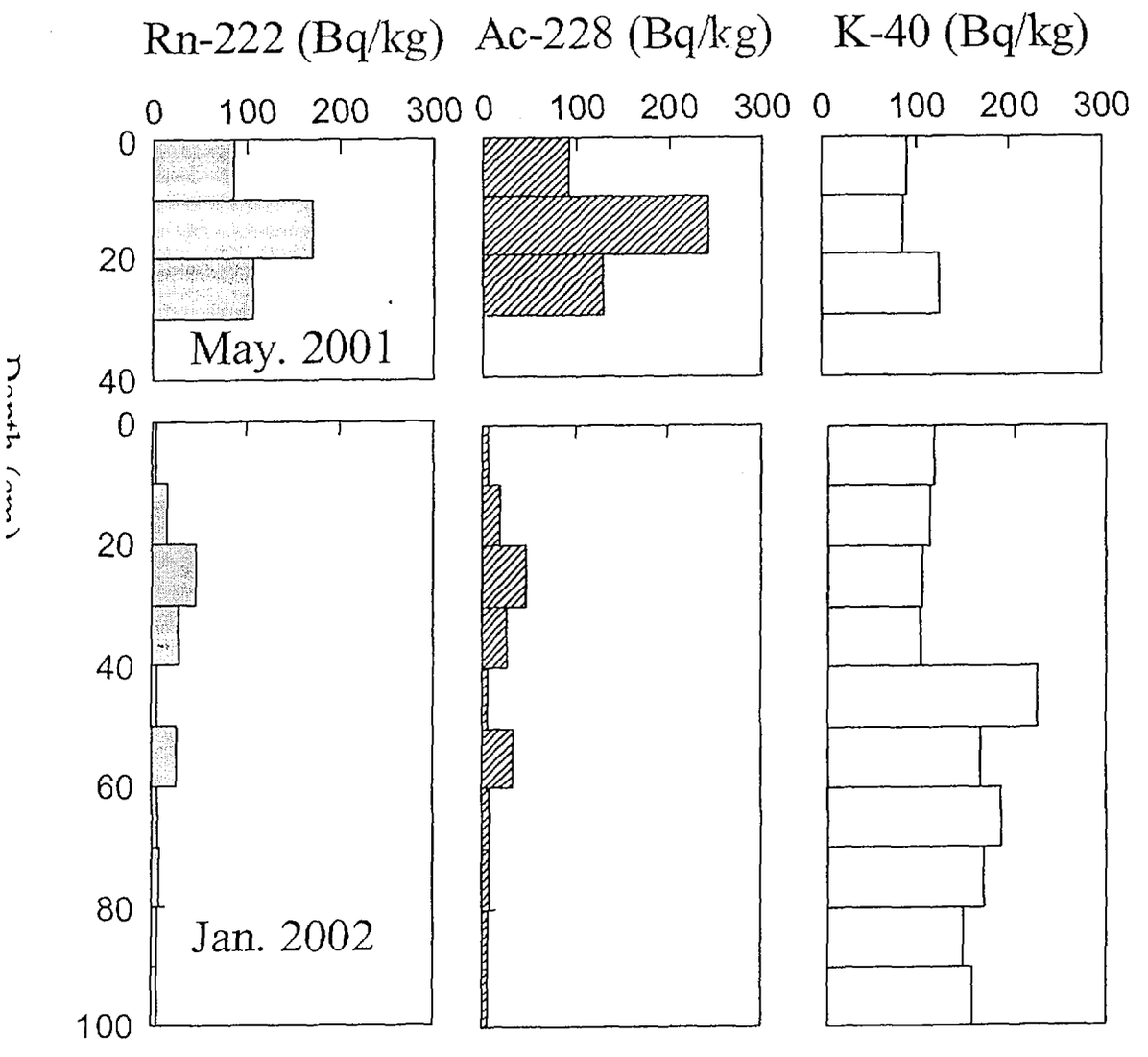

Fig. 6. Depth profile for core samples taken from site #8 for ^{222}Rn, ^{228}Ac and ^{40}K isotopes collected in May 2001 and Jan 2002.

Repeated sampling from two locations with the highest and lowest activities showed that ^{222}Rn and ^{228}Ac activities vary with time in one case. Samples with grain size <200 μm were found to have the highest level of ^{222}Rn and ^{228}Ac activities. The measured depth profile showed evidence for correlation between ^{222}Rn and ^{228}Ac activities and their migration in sand with time.

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