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Hazards Index Analysis of Gamma Emitting Radionuclides in Selected Areas Around the Uranium Mine Sites at Erongo Region, Namibia

ABSTRACT

This study measures the ^{226}Ra, ^{232}Th and ^{40}K activity concentrations using gamma spectrometry to asses first order exposure risks for the persons residing in Walvis Bay and Swakopmund towns in Erongo Region, Namibia. The concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the soil samples vary from 14.94 Bq kg^{-1} to 48.24 Bq kg^{-1}, 17.68 Bq kg-^{1} to 52.51 Bq kg^{-1} and 162.58 Bq kg^{-1} to 259.35 Bq kg^{-1}, respectively, with average values of 30.38 ± 11.28 Bq kg^{-1}, 32.58 ± 10.09 Bq kg^{-1} and 203.62 ± 27.00 Bq kg^{-1} in Walvis Bay town. For Swakopmund town, the concentrations vary from 71.38 Bq kg^{-1} to 155.80 Bq kg^{-1}, 41.63 Bq kg^{-1} to 131.58 Bq kg^{-1} and 360.82 Bq kg^{-1} to 761.76 Bq kg^{-1}, respectively, with average values of 99.59 ± 24.39 Bq kg^{-1} 90.90 ± 31.99 Bq kg^{-1} and 553.07 ± 107.17 Bq kg^{-1}. The radium equivalent activity (Raeq) calculated for the same composite soil samples varies from 62.14 Bq kg^{-1} to 126.69 Bq kg^{-1} with an average value of 92.64 Bq kg^{-1} in Walvis Bay town. In Swakopmund town, it varies from 172.32 Bq kg^{-1} to 332.66 Bq kg^{-1} with an average value of 273.43 Bq kg^{-1}. The average values of absorbed dose and annual effective dose (outdoors) are found to be 42.20 nGy h^{-1} and 123.98 nGy h^{-1}, 0.05 mSv y^{-1} and 0.15 mSv y^{-1} in Walvis Bay and Swakopmund towns, respectively. The average excess lifetime risks of cancer (ELRC) in Walvis Bay and Swakopmund towns were 1.81 x 10^{-4} and 5.33 x 10^{-4}, respectively. This implies that 1 person out of 5555 persons in Walvis Bay town and 1 person out of 1876 persons in Swakopmund town may be affected of cancer related diseases.

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INTRODUCTION

The estimation of external gamma dose due to terrestrial sources is essential not only because it contributes considerably to the collective dose but also because of variations in the individual doses related to this pathway (*Singh et al. 2005*). These doses vary depending upon the concentrations of the radionuclides, ^{238}U, ^{232}Th, their daughter products and ^{40}K, present in the soils and rocks of each region in the world (*Radhakrishna et al. 1993, Quindos et al. 1994*). Natural radionuclides toxicity in soil may pose some health concerns. Some of the major ways through which external radiation get into the human system may be via ingestion of food, soil and water or inhalation of radionuclides as aerosols (*Njinga et al. 2015*). These radionuclides accumulate in various organs once in the system and due to their long half-lives (^{232}Th: 1.4 x 1010 yrs. 238U: 4.47 x 109 yrs. and 40K: 1.28 x 109 yrs.) and chemical behaviour, they may deliver radiation doses which may cause some health related problems. The determination of natural radioactivity of soil samples is usually done from the ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K contents (*Ivanovich and Harmon 1982*). Natural radioactivity measurement due to gamma rays from the dose rate is needed to implement precautionary measures whenever the dose is found to be above or below the recommended limits (*Al-Hamarneh et al. 2009*). There is a growing worldwide interest in natural radiation exposure which has led to extensive surveys in many countries (*Bresson et al. 2011*).

Environmental problems associated with technologically enhanced radionuclides in the uranium mines in Erongo region in Namibia, may result to some health effects. The spread of naturally occurring radionuclide materials (NORMs) in the environment is a means of potential radiation exposure to members of the

public. The uranium mines which are located 40 km and 60 km away from this region produces large volumes of tailings which may be enhanced with some high levels of natural radionuclides. In Namibia, data on radionuclides concentrations in raw materials, residues, fallout from uranium mining and processes and public exposure is still very scanty.

Investigations for the measurement of natural radioactivity in Erongo region, Namibia have been carried out in detail for the first time. The main aim of the present study is to calculate the levels of radioactive exposure through radium, thorium and potassium in Erongo region of Namibia for health risk assessment. This study will provide the baseline data which might be of interest to policy makers, planners and regulators.

MATERIALS AND METHODS

The study areas

The Erongo region is located in the central western part of Namibia and the region covers a land area of 63,549 km^2 and is occupied by the Namib-desert which stretches parallel to the coast of about 120 km to 150 km inland to the study sites (*UNDP 2012*). The two coastal towns, Walvis Bay and Swakopmund are 60 km and 40 km, respectively, away from most of the uranium mine sites (*SEA 2010*) (**Figure 1**).

The landscape is arid and only 10 km^2 of the region is used for cultivation. This includes the area of small-scale farming in the Swakop River bed and the small areas at Omaruru and Okombahe. One of the main activity in the region is mining. The mining industry is the most prominent revenue earner in Swakopmund. The most significant contributors are Rössing and Langer Heinrich Uranium mines. There are also several smaller exploration and mining companies contributing to the uranium rush.

Soil sampling and preparation

A total of twenty composite soil samples were collected from different geographical areas in Walvis Bay and Swakopmund towns (**Figure 2a** and **b**). In Walvis Bay town, soil samples were collected as follows: five collected randomly along the main roads, three collected in the open spaced playground, and two collected in the residential area. In Swakopmund town, the soil samples were collected along the beach (**Table 1**).

Before the collection of the soil samples, the surfaces were carefully cleared of debris and 0.30 m thickness of thesurface soil was removed.

Two kg of soil from each identified point was collected using an auger at a depth of about 0.75 m from the ground so as to get the natural soil. After thorough mixture, 20 composite soil samples of 2 kg each, were transported to the Centre for Applied Radiation and Technology (CARST) laboratory at North-West University, South Africa, for

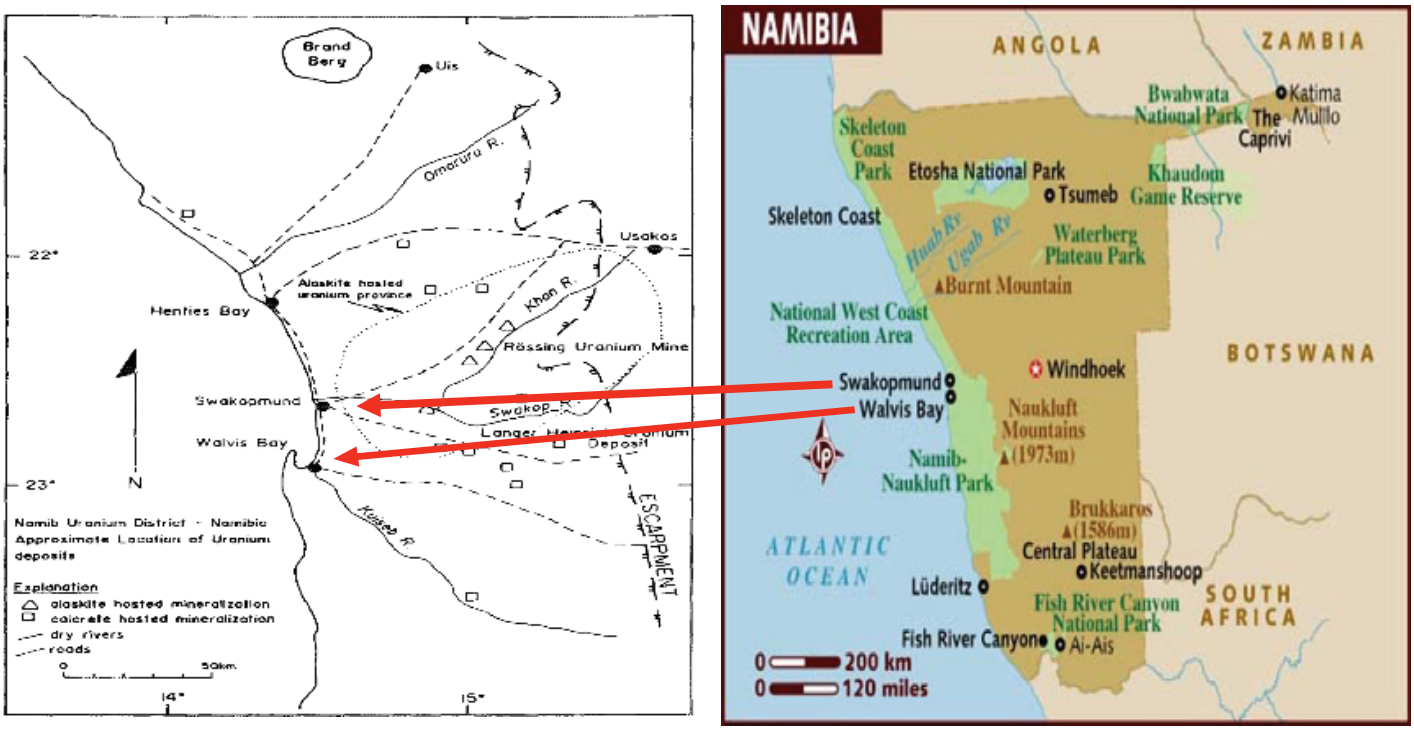

Figure 1. Locations of the study towns in the Western region of Namibia.

Figure 2. Sampling locations within Walvis Bay (a) and Sampling locations along the beach of Swakopmund (b).

Table 1. Description of soil sample collection in Walvis Bay town [a] and Swakopmund town [b].

Sample ID		Description	
Walvis Bay town	**Swakopmund town**	**Walvis Bay town**	**Swakopmund town**
WB8	SK1	WBT^1	$SKAB^1$
WB13	SK4	WBT^2	$SKAB^2$
WB17	SK6	WBT^3	$SKAB^3$
WB10	SK7	WBR^1	$SKAB^4$
WB1	SK8	WBR^2	$SKAB^5$
WB2	SK13	WBR^3	$SKAB^6$
WB7	SK14	WBR^4	$SKAB^7$
WB22	SK15	WBR^5	$SKAB^8$
WB23	SK16	$WBRA^1$	$SKAB^9$
WB20	SK20	$WBRA^2$	$SKAB^{10}$

WBT^1, WBT^2, WBT^3 = At these locations, 10 m^2 area were marked. Four samples collected at the edges and one in the middle of the square. These samples were thoroughly mixed to form a composite sample.

WBR^1, WBR^2,… WBR^5, $WBRA^1$, $WBRA^2$= At these locations, 5 m^2 area were marked. Four samples collected at the edges and one in the middle of the square. These samples were thoroughly mixed to form a composite sample.

$SKAB^1$, $SKAB^2$, $SKAB^3$…$SKAB^{10}$ = At each identified point along the beach, 15 m^2 area was marked. Four samples collected at the edges and one in the middle of the square. These samples were thoroughly mixed to form a composite sample.

analysis. While in CARST, the soil samples were crushed into fine powder using a mortar and pestle. The fine form of each soil sample was obtained using a scientific sieve of 150 micron-mesh size. The samples were dried in an oven at about 383^{o}K for 24 hours before measurement. Each of the sample was packed and sealed in an airtight PVC container and kept for about 28 days to allow radioactive equilibrium among radon (^{222}Rn), thoron (^{220}Rn), and their short lived progenies. On average, 1.25 kg of soil was taken from each sample and put into 1.50 L Marinelli beakers for measurements using the HPGe detector.

Detector calibration

The calibration of the low background counting system was done using a secondary standard which was calibrated with a primary standard obtained from the International Atomic Energy Agency. The activity of samples was counted using a HPGe detector on a high-resolution gamma spectrometry system at the CARST laboratory. The detector was a co-axial n-type high purity germanium detector, which has a resolution of 2.0 keV at 1332 keV of ^{60}Co with a relative efficiency of 20 %. The output of the detector was analyzed using Canberra Genie 2000 software (*Genie™ 2000*).

The detector was lead shielded to reduce the background level of the system (*Xinwei and Xiaolon 2008*). The efficiency calibration for the system was carried out using secondary standard source of uranium ore in geometry available for the sample counting and the values were plotted against energy for particular geometry. The samples were counted for a period of 12 hours and the spectra were analysed for ^{226}Ra, ^{232}Th and ^{40}K.

The concentration of ^{226}Ra was determined using a photon peak of 609 keV (46.1%) from ^{214}Bi. The 186 keV photon peak of ^{226}Ra was not used because of interference with a photo peak of ^{235}U, at an energy of 185.7 keV. Concentration of ^{232}Th was determined using the weighted mean of the gamma-ray transitions associated with the decays of ^{228}Ac, ^{212}Pb and ^{208}Tl. The ^{40}K concentration was determined using the gamma transition of 1461 keV (10.7%). The activity concentrations of radium, thorium, and potassium in Bq kg^{-1} of the radionuclides in the composite soil samples were calculated using the equation (*Olise et al. 2010*):

$$A_{Bq\,kg^{-1}} = \frac{C_{NP}}{B.I \times \varepsilon(E_{\gamma}) \times m} \qquad 1.0$$

where C_{NP}= net peak counts for a given energy line, $B.I$= branching intensity, $\varepsilon(Ey)$= the absolute photo-peak efficiency of the detector and is the mass of the sample in kg.

Radiological risk Analysis

The measured activity concentration of ^{226}Ra, ^{232}Th and ^{40}K were converted into doses by applying the factors 0.461, 0.604 and 0.0417 for radium, thorium and potassium, respectively as:

$$D_R = \sum_k A_k \times F_k \qquad 2.0$$

where D_R is the gamma dose rate in the outdoor air at 1m above the ground, A_K (in unit of nGyh^{-1}/Bq kg^{-1}) is the weighted mean activity of ^{226}Ra, ^{232}Th or ^{40}K, is the corresponding dose conversion factor. The dose conversion factors used in the calculation of ^{226}Ra, ^{232}Th and ^{40}K were 0.461, 0.604, and 0.0417, respectively (*UNSCEAR 1982*). the effective dose received by an adult has to be taken into consideration. This value is 0.7 SvGy^{-1} for environmental exposure to gamma rays of moderate energy published in *UNSCEAR* (*1982; 2000*). The outdoor and indoor occu*pancy factors are 0.2 and 0.8 respectively (*UNSCEAR 1982). The annual effective dose equivalent is given by:

$$AEDE\,(mS/yr) = D_R \times DCF \times F_{IO} \times T \qquad 3.0$$

where F_{10}= the indoor and outdoor occupancy factors (0.8 and 0.2), DCF=dose conversion factor (0.7 SvGy^{-1}) and T= time (8760 hyr^{-1}). The world average annual effective dose equivalent (*AEDE*) from outdoor terrestrial gamma radiation is 0.046 mSv y^{-1} (*Olise et al. 2010*).

The annual effective dose external is given by the equation (*ICRP 1990*):

$$AEDE_{EX}(mSv/yr) = \sum AEDE_{outdoor} + AEDE_{indoor} \qquad 4.0$$

Excess lifetime cancer risk (*ELCR*) was calculated by using equation (4.0):

$$ELCR = AEDE_{outdoor} \times E_{LD} \times C_{RF} \qquad 5.0$$

where E_{LD} = Expected lifetime duration (70 yrs.) and C_{RF}= Fatal cancer risk factor (for stochastic effects, *ICRP 1990* uses a value of 0.05 for the general public).

Radium equivalent activity (Ra_{eq}) is used to assess the hazards associated with materials that contain ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1} (*UNSCEAR 1982*), which is, calculated on the assumption that 370 Bq kg^{-1} of *226*Ra or 259 Bq kg^{-1} of ^{232}Th or 4810 Bq kg^{-1} of ^{40}K produce the same gamma dose rate [7-9]. The Ra_{eq} of the sample in Bq kg^{-1} was achieved using the equation (*ICRP1990*):

$$Ra_{eq} = (A_{Th} \times 1.43) + (A_K \times 0.077) + (A_R) \qquad 6.0$$

where A_{Th}, A_K, A_R =Activity concentrations of ^{232}Th, ^{40}K, and ^{226}R, respectively.

The radium equivalent is the most useful guideline for regulating safety standards on radiation protection for the general public (*UNSCEAR 1982*).

In order to evaluate the external hazard index (H_{ex}), a model proposed by *Beretka and Mathew* (*1985*) was used. This index evaluates the hazard to natural gamma radiation (*Amrani and Tahtat 2001*). However, the prime objective of this index is to limit the radiation dose to the permissible dose equivalent limit of 1mSv y^{-1}. The equation used in evaluating H_{ex} is given as:

$$H_{ex} = (A_R/370) + (A_{Th}/259) + (A_K/4810) \leq 1 \quad 7.0$$

The criterion of this model considers that the external hazard due to gamma-rays corresponds to a maximum radium-equivalent activity of 370 Bq kg^{-1} for the material (*ICRP 1990; Friedrich 2009*).

RESULTS AND DISCUSSION

Activity concentration in Walvis Bay

It can be observed that ^{40}K recorded high values in both towns. In Walvis Bay, a median value of 202.75 Bq kg^{-1} was obtained with minimum and maximum values of 162.58 Bq kg^{-1} and 259.35 Bq kg^{-1} measured in soil sample taken closer to the sand dunes "WB23" (**Table 2**). The mean value of activity concentration of ^{40}K in the ten soil samples from Walvis Bay was 203.62 ± 27.00 Bq kg^{-1}. ^{226}Ra and ^{232}Th activity concentrations in soil samples from Walvis Bay ranged from 14.94 ± 02.24 to 48.24 ± 7.31 Bq kg^{-1} and 17.68 ± 2.39 to 52.51 ± 09.02 Bq kg^{-1}, respectively.

Table 2. Radionuclides concentrations in Bq kg^{-1} for the composite soil samples from [a] Walvis Bay Town and [b] Swakopmund Town, Namibia.

[a] Walvis Bay town

Radionuclides	Mean ± Sd	Median	Min - Max
^{226}Ra	30.38 ± 11.28	28.88	14.94-48.24
^{232}Th	32.58 ± 10.09	31.41	17.68-52.51
^{40}K	203.62 ± 27.00	202.75	162.58 - 259.35

[b] Swakopmund Town

Radionuclides	Mean ± Sd	Median	Min - Max
^{226}Ra	99.60 ± 24.39	91.79	71.38-155.8
^{232}Th	90.90 ± 31.99	96.17	41.63-131.58
^{40}K	553.07 ± 107.17	563.95	360.82 - 761.76

Activity concentrations in Swakopmund

In Swakopmund, the mean values of activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K from the soil samples were 99.60 ± 24.39, 90 ± 31.99 and 553.07 ± 107.17 Bq kg^{-1} respectively (**Table 2**). In comparison with soils from Walvis Bay, these values were high in the magnitude by 3.28, 2.7 and 2.7 times for ^{226}Ra, ^{232}Th and ^{40}K, respectively. This can be attributed to their geographical locations from the uranium mines. Swakopmund and Walvis Bay are located at distances of 40 and 60 km from Rossing Uranium mine, respectively. As a result, Swakopmund town received high levels of dust emissions giving rise to ambient pollution concentrations and deposition levels derived from anthropogenic, natural and biogenic sources (*Neuman et al. 2009*).

Outdoor terrestrial gamma dose rates in Walvis Bay town

The outdoor terrestrial gamma dose rates were determined in composite soil samples (**Table 3**). It was observed that the outdoor terrestrial gamma dose rate values were 56.84 nGy/h for WB1, and 51.34 nGy/h for WB10 and were higher compared to the 51.00 nGy/h limit set by *UNSCEAR* (*1982; 2000*).

Outdoor terrestrial gamma dose rates in Swakopmund Town

The outdoor terrestrial gamma dose rates were higher compared to the limit set by *UNSCEAR* (*1982; 2000*) with highest values of 140.21 nGy/h (SK 15), 143.09 nGy/h (SK 1), 152.02 nGy/h (SK 16) and 143.64 nGy/h (SK 14) found (**Table 3** [b]). It was also observed that the sampling locations, SK 16, SK 15, SK 6, SK 1, SK 14 and SK 20 had about two times higher dose rates compared to the 51.00 nGy/h average value of *UNSCEAR* (*2000*). The other locations were 1.37 to 1.67 times higher. It was observed that most of the locations in Walvis Bay town, were lower compared to the average value obtained for Swakopmund Town and the *UNSCEAR* (*1982*).

Excess lifetime cancer risk in Walvis Bay and Swakopmund towns

The excess lifetime cancer risks were also calculated (**Table 3** [a], [b]). The life expectancy was taken as 70 years (*UNSCEAR 1982*), while the lifetime outdoor gamma radiation was assumed to be 6.0 (**Table 3** [a], [b]). The excess lifetime cancer risks in the two towns were compared to the world average value of 0.29 × 10^{-3} (*UNSCEAR 1982*). All the sampling locations in Swakopmund recorded

higher values with an average value of 1.84 times higher in magnitude.

This study revealed that the average terrestrial gamma dose rate of 123.98 nGy h^{-1} in soils from Swakopmund town were higher compared to the limit according to *UNSCEAR* (*1982; 2000*). This high level of gamma radiation was directly associated with the activity concentrations of the radionuclides in the soil samples. Swakopmund town recorded high activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K which increased the terrestrial gamma dose rates. The locations SK 16, SK 14 and SK 1 in Swakopmund town recorded the highest outdoor gamma dose rate of 152.02, 143.64 and 143.09 nGy h^{-1}, respectively (**Table 3 [b]**). The same town also had higher activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K compared to the other localities. This could be attributed to the mine tailings located a few kms away from the town that produced fugitive dust emissions containing radionuclides such as of ^{226}Ra, ^{232}Th and ^{40}K. The calculated outdoor annual effective dose equivalent in all ten geographical locations of Walvis Bay, varied from 0.04 mSv y^{-1} to 0.07 mSv y^{-1} with an average of 0.05 mSv y^{-1} (**Table 3 [a]**).

Radium equivalent (Ra_{eq}) and external hazard index (H_{ex}) in Walvis Bay town

In this town, the radiation hazard parameters in terms of radium equivalent (Ra_{eq}) and the external hazard index (H_{ex}) were calculated. The maximum value of 126.69 Bq kg^{-1} for WB1 site and a minimum of 62.14 Bq kg^{-1} was recorded for WB8 site (**Table 3 [a]**). All the values were within the permissible limit recommended value of 370 Bq kg^{-1} as recommended by *ICRP* (*1990*). The average external radiation hazard index (H_{ex}) for the two towns were 0.25 and 0.74, respectively. These values were lower than unity, which corresponds to the maximum radium activity of 370 Bq kg^{-1} for all terrestrial material.

Radium equivalent (Ra_{eq}) and external hazard index (H_{ex}) in Swakopmund town

In the town of Swakopmund, the values, were lower than the average world value of 370 Bq kg^{-1} (**Table 3 [b]**). The external hazard index (H_{ex}), ranged from 0.47 Bq kg^{-1} for location SK 8 to 0.90 Bq kg^{-1} for location SK 16.

Table 3. Radiological Hazard index parameters measured in [a] Walvis Bay Town and [b] Swakopmund Town, Namibia.

[a] Walvis Bay town

Sample ID	Ra_{eq} Bq kg^{-1}	H_{ex}	*ELCR*	Dose Rate nGy h^{-1}	$AEDE_{outdoor}$ mSv y^{-1}	$AEDE_{ex}$ mSv y^{-1}
WB 1	126.69	0.34	2.44E-04	56.84 (> Rv)	0.07 (> WAV)	0.35
WB 2	99.23	0.27	1.93E-04	44.89	0.06 (> WAV)	0.28
WB 7	69.93	0.19	1.38E-04	32.15	0.04	0.2
WB 8	62.14	0.17	1.24E-04	28.90	0.04	0.18
WB 10	112.09	0.30	2.21E-04	51.34 (> Rv)	0.06 (> WAV)	0.32
WB 13	111.19	0.30	2.16E-04	50.28	0.06 (> WAV)	0.31
WB 17	90.91	0.25	1.76E-04	40.91	0.05	0.25
WB 20	75.81	0.20	1.50E-04	34.86	0.04	0.21
WB 22	73.96	0.20	1.45E-04	33.73	0.04	0.21
WB 23	104.48	0.28	2.07E-04	48.14	0.06 (> WAV)	0.3

(> Rv) = greater than recommended value of 51 nGy h^{-1} [20], (> WAV) = greater than the World Average
Value of 0.046 mSv y^{-1} (*UNSCEAR 1982*), (> WA) = world's average value of 2.90E-04 for 70 yrs. life expectancy (*UNSCEAR 1982*)

[b] Swakopmund Town

Sample ID	Ra_{eq} Bq kg^{-1}	H_{ex}	*ELCR*	Dose Rate nGy h^{-1}	$AEDE_{outdoor}$ mSv y^{-1}	$AEDE_{ex}$ mSv y^{-1}
SK 1	318.61	0.86	6.15E-04 (> WA)	143.09 (> Rv)	0.18 (> WAV)	0.88
SK 4	202.21	0.55	3.97E-04 (> WA)	92.37 (> Rv)	0.11 (> WAV)	0.57
SK 6	316.68	0.86	6.11E-04 (> WA)	142.24 (> Rv)	0.17 (> WAV)	0.87
SK 7	204.03	0.55	4.06E-04 (> WA)	94.59 (> Rv)	0.12 (> WAV)	0.58
SK 8	172.32	0.47	3.46E-04 (> WA)	80.55 (> Rv)	0.10 (> WAV)	0.49
SK 13	244.34	0.66	4.83E-04 (> WA)	112.43 (> Rv)	0.14 (> WAV)	0.69
SK 14	313.06	0.85	6.17E-04 (> WA)	143.64 (> Rv)	0.18 (> WAV)	0.88
SK 15	311.82	0.84	6.02E-04 (> WA)	140.21 (> Rv)	0.17 (> WAV)	0.86
SK 16	332.66	0.90	6.53E-04 (> WA)	152.02 (> Rv)	0.19 (> WAV)	0.93
SK 20	318.61	0.86	5.96E-04 (> WA)	138.66 (> Rv)	0.17 (> WAV)	0.88

In general, the high radiological indices such as outdoor gamma dose rate, annual effective dose external and excess lifetime cancer risk in Swakopmund town can be attributed to the close proximity of this town to Rossing uranium mine. It can be inferred that Swakopmund town isexposed to high levels of radiation through high wind blow, carrying dust from the mine tailings and disperse it into the air which eventually settle in this nearby town.

CONCLUSION

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil samples from Walvis Bay and Swakopmund towns in Namibia were higher than the world figures reported in *UNSCEAR* (*2000*) in the town of Swakopmund. However, the concentration for ^{40}K is very much comparable and a concentration for ^{226}Ra is lower as compared with world figures. The outdoor terrestrial effective dose due to natural radioactivity of soil samples were averagely low in Walvis Bay town and were high in Swakopmund town when compared to the average national and world recommended value of 1.0 mSv y^{-1}. The calculated values of hazard indices ($\boldsymbol{H_{ex}}$) for the soil samples were lower than unity. Therefore, according to the Radiation Protection 112 report (*European Commission 1999*), soils from these regions are safe. The calculated lifetime risks of cancer were higher in Swakopmund town and lower in Walvis Bay town when compared to the world's average.

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