SHORT COMMUNICATION

Naturally Occurring Radioactive Materials Measurement of Mineral soil in South-East of Ethiopia

Teklemariam Tessema Teklemariam *, Milkesa Gebeyehu, Abreham Alemu

Department of Physics, College of Natural Sciences, Jimma University, Jimma, Ethiopia

* Corresponding author: teklema@yahoo.ca (TT)

ABSTRACT

This experimental research was designed to determine the concentration of naturally occurring radioactive materials (NORM) in samples of mineral soil being used by people as a free choice mineral for cattle feed in South West Shoa Zone of Oromia Regional State, Ethiopia, particularly around Woliso (Latitude: 8° 24' 59.99" N; Longitude: 38° 14' 60.00" E), and to calculate the parameters connected to radiation hazard due to the mineral. Measurement of the natural radioactivity was performed by gamma spectrometry consisting of 3x3 inch NaI(Tl) detector, multichannel analyzer and associated electronics equipped with software MAESTRO and WINSPAN. The average values of activity concentration in the investigated samples for ^{40}K, ^{226}Ra and ^{232}Th, respectively, were 458.78±3.40, 24.82±0.38 and 67.45±0.12 in Bq Kg^{-1}. This data showed that the risk of radiation from the mineral soil, locally called "Hora, and used as a free choice mineral, is below the limit set in literature with External and internal index level values found below one, (H_{ex} and $H_{in} < 1$). It could be concluded that the current practice associated with the use of 'Hora' minerals will not expose the public to A radiation risk.

Key Words: Activity concentration Mineral Soil, NaI (Tl) gamma spectrometry, NORM

INTRODUCTION

Radioactivity in minerals is caused by the presence of naturally-occurring radioactive materials (NORM) in the mineral's composition. The degree of radioactivity is dependent on the concentration and isotope present in the mineral. For the most part, minerals that contain potassium (K), uranium (U), and thorium (Th) are radioactive; these materials are called naturally occurring radioactive materials (NORM) since they are in existence since the existence of the earth. Many materials that are usually found in the earth's crust contain small but measurable amount of NORM (Krieger, 1981).

Exposure to natural sources of radiation is often influenced or can be influenced by human activities. When minerals or rocks are collected a certain place, compared with that of the undisturbed earth's crust, it may cause excess exposure to external gamma. Some ores/minerals may contain natural radionuclide at levels much higher than usually present in earth's crust (Fathivand et al., 2007); and when these minerals are collected or mined for different purpose in a technological process, they cause a radiation risk to the public or environment.

It is a common practice that people in the rural part of Ethiopia use mineral soils for cattle production, and other purposes (Muluken, 2016; Shewangzaw, 2013). Such a practice leads to accumulation of the mineral soil in a small place near peoples' homes from where radiation might cause substantial exposure associated risk if it contains elevated levels of naturally occurring radionuclide (Todorovic et al., 2015). Accordingly, it is necessary to evaluate the level of gamma exposure on such environment so that the effect of radiation from the soil shall be determined. If the level of the radiation is above the safe level to the environment, appropriate measures necessary to keep the environment healthy has to be taken as per the recommended scientific approaches in radiation-physics. Todorovic et al. (2015), stressed the importance of measuring the concentration of radionuclide in rocks and minerals to assess the radiological risk to human health.

Different measurements and parameters of such mineral soils in Ethiopia were determined (Teklemariam et al., 2015; Tibebu et al., 2017). A nuclear technique has been used to check the concentration of elements in one of such mineral soils called "Ewoa" used in Gurage zone, Ethiopia, in which the potassium concentration was found to be higher compared to other soils measured in different parts of the world (Teklemariam et al., 2015). The physicochemical parameters of "Hora" natural mineral water and soil used by people in Oromia Regional Sate, Ethiopia, was also determined (Tibebu et al., 2017). However, the physicochemical parameters do not give information about the risk on an environment associated to gamma radiation emanating from the soil. The objective of this study was, therefore, to determine the radioactivity of samples of a mineral soil used for the purpose of cattle feeding by people in Debub Mirab Shewa Zone of Oromia Regional State, Ethiopia, and to measure the concentration of naturally occurring radioactive materials and calculate parameters connected to the radiation hazard due to the mineral; and compare to the standard limits to check for any appreciable risks associated to this practice. The mineral soil in the area is known by two interchangeably used names called "Bole" or "Hora", hereafter called Hora across the document.

MATERIALS AND METHOD

Sample Collection: Farmers in Debub Mirab Shewa Zone of the Oromia Regional State, around Woliso (Latitude: 8° 24' 59.99" N; Longitude: 38° 14' 60.00" E), Ethiopia, use Hora as a mineral choice to feed their cattle. Hora soil is gray and fine powdered natural mineral that could be mined directly from the ground. But there is no mining site in the study area; instead people purchase and use the mineral from open markets in the zone which is brought there for commercial activity from other parts of Ethiopia.

During the research, a structured interview was performed for most of the Hora-sellers, in four market areas within the research site, namely "Aleltu", "Seden sodo', "Sumbo" and "Gorki", to get information about the exact place where the mineral soils have been mined. According to the analysis from the interview, all the respondents indicated that the mineral soils were mined from Silte Zone of South Nations Nationality and People Region (SNNP) of Ethiopia, specifically in three places called "Qawaqoto", "Manz" and "Alicho" located in the Central Rift Valley of Ethiopia. Samples of the Hora-soil were collected from the three mining areas separately and transported to Jimma University, Ethiopia, for analysis.

Experiment: The collected Hora samples (mineral soil samples) were brought to the Advanced Physics Laboratory at Jimma University, and exposed to air to dry at ambient temperature for one week. After the samples were checked for drying, each of the three samples were sieved and packed in to cylindrical plastic containers of size that

approximately fill the space left for samples in the lead shielding of the NaI(Tl) gamma spectroscopy. The sample containing containers were labeled with names, “Hora I”, “Hora II”, “Hora III” and “Hora IV”. The sample inside the container “Hora I” was sample collected from Qawaqoto site, “hora II” from Manz, “Hora III from Alicho and “Hora IV” was composite sample contributed from the three sites equally. The sample containers were accurately weighed with digital mass balance before and after they were filled with the Hora-soil samples. This helps to get the dry mass of the soil inside each sample. Accordingly, the samples in “Hora I”, “Hora II”, Hora III and “Hora IV” were containing mineral soils weighing 523.8, 524.2, 524.9 and 524.6 in gram, respectively, and were sealed air tight to protect the escape of radon gas as shown in the Fig. 1 below.

Fig. 1 Sealed Hora-soil samples collected from the study sites

The samples were carefully transported to the laboratory at Ethiopian Radiation Authority, Ministry of Innovation and Technology, Addis Ababa and were kept for four weeks (28 days), in order to allow the re-establishment of secular equilibrium between parents and short-lived daughter products.

The measurements of natural radioactivity level was performed by gamma spectrometry facility consisting of 3x3 inch NaI(Tl) detector housed in a 6 cm thick lead shield and lined with a cadmium and copper sheets connected to multi-channel analyzer (MCA) and associated electronics. Use of such detector is better compared to solid-state detectors for better efficiency (Papastefanou, 2007). The system was equipped with data acquisition and analysis software. With this facility, the room background, standard reference materials and each of the Hora samples have been counted for eight hours after the secular equilibrium was attained and the spectrum was saved for further analysis and calculation. The calibration standard and reference sources used were International Atomic Energy Agency (IAEA) standards, RGU-1, RGTh-1 and RGK-1 for uranium, thorium and potassium, respectively. The reference soil samples were certified to have activity concentrations of 95% confidence interval (C.I) in the interval (13600 – 14400)$BqKg^{-1}$ for RGK-1, (4910 – 4970) $BqKg^{-1}$ for RGU-1, and interval (3160 – 3340) $BqKg^{-1}$ for RGTh-1 (IAEA, 2009).

The reference samples have similar matrices to the analyzed samples and were counted at the same geometry. The analyzed data for the references in this work was with in the 95% C.I values indicating the procedure was correct. The room background was counted using

empty container placed inside the sample position of the detector and its activity concentration was subtracted from the sample activity. The uranium and thorium decay series, ^{238}U and ^{232}Th activities, were determined indirectly via activities of their daughter products. Accordingly, the 2614.5 keV -peak of ^{208}Tl was used to measure the specific activity ^{232}Th, while 1764 keV -peak of ^{214}Bi was used in assessment of the specific activity of ^{238}U and ^{40}K was determined directly by 1460.5 keV photo peak.
The activity concentrations in the samples were obtained using Eq. 1 given below:

$$C = KC_0 \quad \text{..... 1}$$

Where, C is the activity concentration of the radionuclide in the sample in $BqKg^{-1}$, (becquerel pe kilogram), C_0 is the count rate under the corresponding peak in counts per second (Cps); and K is given by $K = 1/(\epsilon P_\gamma M_s)$ such that, "ϵ" is the detector efficiency at the specific gamma ray energy, P_γ is the absolute transition probability of the specific γ-ray and M_s is the mass of the sample (Kg). The activity in cps was obtained by dividing the total peak area to the counting time.

RESULT

During the experiment, the gamma spectrums acquired by the acquisition software were saved in a computer before the analysis. The shape of a spectrum obtained during the activity counting of one of the samples in this work was as shown in Fig. 2.

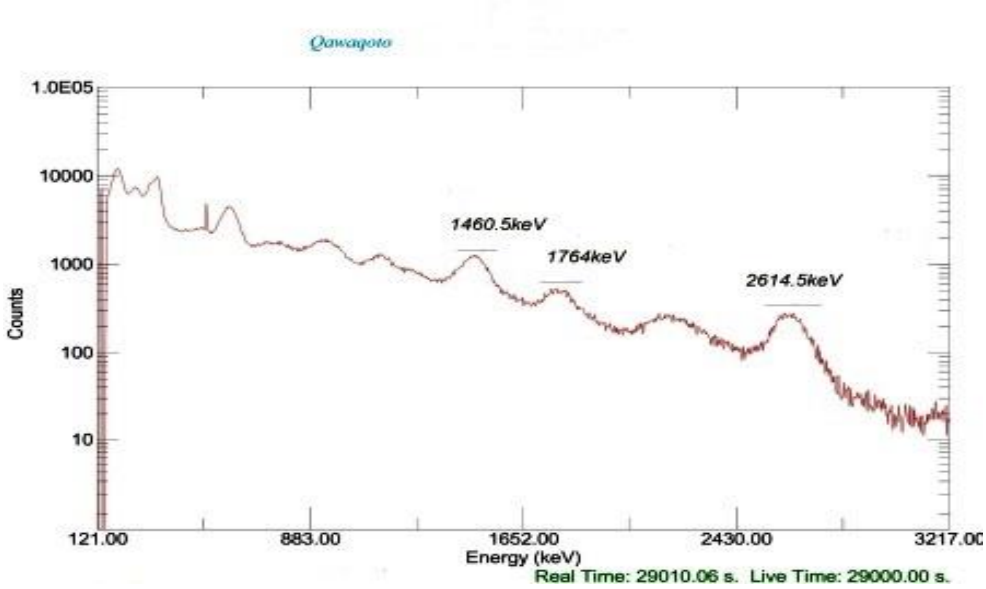

Fig. 2 The spectrum obtained during the activity counting of the mineral sample "Hora I".

Activity concentration: The most important source of natural radiation exposure is caused by gamma rays emitted from members of the uranium and thorium decay chains and radioactive potassium occurring naturally in soils and minerals. In the ^{238}U series, the decay chain branch starting from radium (^{226}Ra) is radiologically more important and therefore reference is often made to ^{226}Ra than ^{238}U (Ravisankar et al., 2012). The specific activities of radium, thorium and potassium measured in the samples, as well as corresponding standard deviations due to counting errors are presented in Table 1.

Table 1: The activity concentration due to ^{40}K, ^{226}Ra and ^{232}Th in the investigated mineral soil samples

Sample Code	Activity concentration of ^{40}K (A_K)		Activity concentration of ^{226}R (A_{Ra})		Activity concentration of ^{232}Th (A_{Th})	
	Cps	Bq Kg^{-1}	Cps	Bq Kg^{-1}	Cps	Bq Kg^{-1}
Qawaqoto	0.296±0.0023	460.34±3.41	0.022±0.0003	25.73±0.53	0.071±0.0002	73.35±0.12
Manz	0.292±0.0018	454.12±3.18	0.021±0.0003	23.92±0.45	0.057±0.0001	61.21±0.11
Alicho	0.297±0.0024	461.89±3.62	0.021±0.0004	24.80±0.17	0.064±0.0002	67.78±0.12
Mixed	0.294±0.0022	457.93±3.80	0.020±0.0004	24.75±0.41	0.064±0.0001	66.98±0.23
Average	0.295±0.0022	458.78±3.40	0.021±0.0003	24.82±0.38	0.064±0.0002	67.45±0.12

Where, Cps = counts per second, ^{40}K=Potassium, ^{226}R= Radium, ^{232}Th= thorium

A_K , A_{Ra} , A_{Th} = activity concentration of ^{40}K, ^{226}Ra , and ^{232}Th in $BqKg^{-1}$, respectively.

The radioactivity concentrations of the three nuclides are relatively similar in the different samples of Hora-soil. As can be seen from the Table 1, no significant difference was seen between the concentrations of the radioactive nuclides in the Hora samples. But the concentration of the radionuclide ^{40}K, ^{226}Ra and ^{232}Th in the same Hora sample were significantly different, with ^{232}Th concentration roughly in the range of three times the concentration of ^{226}Ra in all the samples, and ^{40}K activity was higher.

If we consider the small differences in radionuclide concentration available between the samples, Hora soil sample called "Manz" contained relatively less concentrations compared to the other two. The average values of activity concentrations of the investigated samples for ^{40}K, ^{226}Ra and ^{232}Th, respectively, were 458.78±3.40, 24.82±0.38 and 67.45±0.12 in Bq Kg^{-1}. These values were closer to the experimentally determined concentration with mixed soil samples as shown above (Table 1).

Radium Equivalent Activity (Ra_{eq}): This is an index used to obtain the sum effect of these activities in a sample. It is used in order to compare specific activities of materials containing different concentration of ^{40}K, ^{226}Ra and ^{232}Th. It is given by Eq. 2 based on the estimation that 370 BqKg^{-1} of 226Ra, 259 BqKg^{-1} of ^{232}Th, or 4810 BqKg^{-1} ^{40}K produce equivalent rate of gamma dose (Stranden, 1976; Beretka and Mathew, 1985). UNSCEAR (2000) recommended maximum value of radium equivalent activity to be 370BqKg^{-1} .

$$\mathbf{Ra_{eq}}\ (\mathrm{BqKg^{-1}}) = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots 2$$

Where A_{Ra}, A_{Th} and A_K are activity concentration of ^{226}Ra , ^{232}Th and ^{40}K in BqKg^{-1}.

External and Internal Hazard indices: External Hazard index (H_{ex}) is a criterion to access the radiological suitability of a material, and the internal hazard index (H_{in}) gives the internal exposure to carcinogenic radon and its short-lived progeny. To have less (negligible) hazardous effect, both indexes should have a value less than unity or should not be greater than one. The indexes are given by Eq. 3 and Eq. 4 (Krieger, 1981).

$$H_{ex} = \frac{A_{Ra}}{370BqKg^{-1}} + \frac{A_{Th}}{259BqKg^{-1}} + \frac{A_K}{4810BqKg^{-1}} \quad \ldots\ldots\ldots\ldots\ldots 3$$

$$H_{in} = \frac{A_{Ra}}{185BqKg^{-1}} + \frac{A_{Th}}{259BqKg^{-1}} + \frac{A_K}{4810BqKg^{-1}} \quad \ldots\ldots\ldots\ldots\ldots 4$$

Using the results displayed in Table 1 above and the Eqs. 2, 3 & 4, the calculated values of Ra_{eq}, H_{ex} and H_{in} for each of the samples are as shown below (Table 2).

According to the results shown in Table 2, the levels of natural radioactivity in the samples were within normal ranges because the external and internal hazard indices have values less than one. Furthermore, the values in Table 3 summarize the natural radioactivity levels and radiation hazard indices in soil obtained in literature for some regions in Africa including Egypt (Ahmed and El-Arabi, 2005) and Nigeria (Agbalagba et al., 2011), and beyond Africa such as Yemen (Abd El-Mageed et al., 2011).

Table 2: Radium equivalent activity, external and internal indices values of Hora samples.

Sample	Ra_{eq} ($BqKg^{-1}$)	H_{ex}	H_{in}
Qawaqoto	166.07±0.96	0.451	0.512
Manz	146.42±0.85	0.398	0.464
Alicho	157.05±0.	0.426	0.494

There was no information in literature found during this work which gives data about NORM activity in the soil of a region in Ethiopia. The values of Ra_{eq}, H_{ex}, and H_{in} in this work are within the ranges reported in other regions listed in Table 3.

Table 3: Radiation Hazard indices of soil samples compared with the values reported in literature

Region	Ra_{eq} ($BqKg^{-1}$)	H_{ex}	H_{in}	Reference
Egypt 31	152	___	___	Ahmed and El-Arabi, 2005
Yemen 29	191	0.52	___	Abd El-Mageed et al., 2011
Nigeria 26	50-110	0.29-0.14	0.18-0.37	Agbalagba et al., 2011
Present work	146 - 167	0.398 - 0.451	0.464 - 0.512	This study

CONCLUSION

According to the analyzed result, the activity concentrations for ^{238}U, ^{232}Th, and ^{40}K in soil samples in the present study were within the range of activity values for other regions including Egypt, Nigeria and Yemen, and the levels of natural radioactivity in the samples studied were within normal values of H_{ex} and H_{in} (H_{ex} and $H_{in} < 1$). This shows that the radiation exposure from using the mineral soil Hora, as a free choice mineral, is within acceptable range.

Furthermore, the findings show that the mineral soil Hora has greater internal index value compared to minerals and rocks reported in literature. The calculation in this work was based on the external use of the soil sample. Since cattle eat the soil, it might give higher exposure internally to them so that excessive use of it might cause radiation associated problems, like abortion, lessen resistance, and breathing system related problems to the cattle. In addition, based on experimental procedure and average calculation of concentration of radionuclide in the samples, it was noted that the mixed soil-sample concentration of the radionuclide was approximately equal to the average concentration. The ratio between the radionuclide concentrations of the soil samples was nearer to one indicates that the soil samples have the same composition or belong to the same origin, even though they were mined from different places.

ACKNOWLEDGEMENT

The author acknowledges the financial support of the College of Natural Sciences Research and Post Graduate Office, Jimma University; under the contractual agreement code of CNS –PHYS-005-2017-SP1; the support of facilities by Department of Physics; and the editorial support of Prof. VK Mittal, Professor of Nuclear Physics at Jimma University, Ethiopia.

REFERENCES

Fathivand , A.A., Amidi, J, and Hafezi, S. (2007). Natural radioactivity concentration in raw materials used for manufacturing refractory products, Iran. J. Radiat. Res.; 4 (4): 201- 204

Abd El-Mageed, I., El-Kamel, H., Abbady, A., Harb, S., Youssef, M. and Saleh, I. (2011). Assessment of natural and anthropogenic radioactivity levels in rocks and soils in the environments of Juban town in Yemen. Radiation Physics and Chemistry Journal, 80: 710-715.

Agbalagba, E. and Onoja, R. (2011). Evaluation of natural radioactivity in soil, sediment, and water samples of Niger Delta (Biseni) flood plain lakes, *Nigeria. Environmental Radioactivity Journal*, 102: 667-671.

Ahmed, K. and El-Arabi, M. (2005). Natural radioactivity in farm soil and phosphate fertilizer and its environmental implications in Qena governorate, Upper Egypt, Environmental Radioactivity Journal, 84: 51-64.

Beretka, J. and Mathew, P.I. (1985). Natural radioactivity of Australian building materials, waste and by-products. Health Phys, 48: 87- 95.

Papastefanou, C. (2007). Radioactive Aerosols, 1st Edition; ISBN:9780080555980.

IAEA (2009). Reference products for reference and Trade., IAEA/AL/148. *https://nucleus.iaea.org/rpst/referenceproducts/referencematerials/.../IAEA-RGK-1.htm*

Krieger, R. (1981). Radioactivity of construction materials. Betonwork Fertigteil-Techn, 47: 468 - 473.

Muluken Zeleke, Yisehak Kechero and Mohammed Y. Kurtu, (2016). Practice of Local Mineral Supplementation to Livestock's and Perception of Farmer's in Humbo Woreda, Wolaita Zone, Ethiopia, Global Veterinaria, 17 (2): 114-121

Todorovic N., Bikit I., Krmar M., Mrdja D., Hansman J., Nikolov J., Forkapic S., Veskovic ., Bikit K. and Jakonic I., (2015). Natural radioactivity in raw materials used in building industry in Serbia, Int. J. Environ. Sci. Technol., 12:705–716

Ravisankar, R., Vanasundari, K., Chandrasekaran, A., Rajalakshmi, A., Suganya, M., Vijayagopal, P. and Meenakshisundaram, V. (2012). Measurement of natural radioactivity in building materials of Namakkal, Tamil Nadu, India using gamma-ray spectrometry. Applied Radiat. Isot., 70:699 – 704.

Shewangzaw Addisu, Firew Tegegne and Zeleke Mekuriaw (2013). Effect of Different Levels of Dietary Bole (Lake Soil) Inclusion on Feed Intake, Milk Yield and Composition of Holstein Friesian Cows, Inter J Agri. Biosci, 2(6): 377-382.

Stranden E. (1976). Some aspects on radioactivity of building materials. Phys. Norv., 8: 167 -173

Teklemariam, T., Chaubey, A.K., Birhanu, W., Ahmed Yusuf and Oladipo M. O. (2015). Multi Elemental Analysis of mineral soil "Ewoa" used for cooking, cloth washing, etc. in Gurage Zone, Shamene, Ethiopia. International J. Engineering and Scientific Research (IJSER). 3. 126.

Tibebu Alemu, Eyobel Mulugeta and Miresa Tadese (2017). Determination of physicochemical parameters of “Hora” natural mineral water and soil in Senkele Kebele, Oromia Region, Ethiopia, Cogent Chemistry, 3:1, DOI: 10.1080/23312009.2017.1354800

UNSCEAR (2000). United Nations Scientific Committee on the Effect of Atomic Radiation, Sources and Biological Effects of Ionizing Radiation, United Nations, New York.