SOIL RADON IN THE NIGERIAN YOUNGER GRANITES

B. B.Muhammad Dewu
Centre for Energy Research and Training, Ahmadu Bello University, Zaria, Nigeria
Bbmdewu@yahoo.com

(Submitted: 21 October,2006; Accepted: 15 October, 2007)

Abstract

Soil radon measurements were carried out to determine the applicability of soil radon surveys in the investigation of uranium in the Nigerian Younger Granites. The study forms part of a series of studies in the investigation of uranium in the Nigerian Younger Granites. Radon activity was measured with a radon detecting system consisting of a cartridge of radon/alpha cards and a card reader. The cards were exposed in holes where soil samples were collected. Results of the measurement indicate a wide variation in soil radon activity over the Nigerian Younger Granite; both inter and intra-soil-types. The results show that soil radon activity mainly controlled by the uranium content of the underlying granites. The results thus suggest that in the Younger Granites, soil radon measurements are better indicators of uranium at depth than the soil uranium content. Findings further suggest that uranium in the soils is young and has not had enough time to attain equilibrium with its daughters. In general, the results suggest that with proper control, soil radon measurements over the Younger Granite can be used for uranium exploration in the region.

***Keywords**: Radon, younger granite, soil uranium, half-life and thorium*

Introduction

Radon (Rn) is a chemically inert radioactive gas produced by the decay of radium (Ra) in the U and Th decay series. In the natural environment it occurs in three isotopes, radon (^{222}Rn), thoron (^{220}Rn) and actinium (^{219}Rn) due to ^{238}U, ^{235}U and ^{232}Th, respectively. In each series it undergoes radioactive decay into polonium (Po) with the release of alpha-particles. These transmutations together with the half-life of the Rn isotopes are shown below:

$$^{226}Ra \ ? \ ^{222}Rn + ^{4}He$$
$$^{222}Rn \ ? \ ^{218}Po + ^{4}He \qquad 3.825 \text{ days}$$

$$^{223}Ra \ ? \ ^{219}Rn + ^{4}He$$
$$^{219}Rn \ ? \ ^{215}Po + ^{4}He \qquad 3.92 \text{ sec.}$$

$$^{224}Ra \ ? \ ^{220}Rn + ^{4}He$$
$$^{220}Rn \ ? \ ^{216}Po + ^{4}He \qquad 55.6 \text{ sec.}$$

Of the three isotopes, the geochemically most important is ^{222}Rn, both because of its abundance and the relatively long half-life compared to the other two.

In soils, Rn is mostly supported in the soil water and the pore spaces. Most of the Rn in soil is also dependent on sources within the soil except where the soils lie on faults or fractures in the underlying rock formations. In such cases soil Rn may reflect sources from the underlying rocks.

Being a daughter in the main U decay series, the presence of Rn in an environment can be used to infer the presence of U. In principle thus, measurement of Rn in soils and surface waters can be used in U exploration programmes, especially because of its relative short half-life, its presence in an environment is considered to be associated with U-bearing minerals in the immediate environment. Successes in using Rn measurements as a U exploration method have been documented, for example, Ridland (1945) and Dyck (1969). Despite these successes however, various environmental factors put restrictions on the use of Rn surveys in U exploration. These factors were found to have profound control on the Rn content in an environment, for example, Kovach (1946), Kraner *et. al.* (1964) and Caneer and Saum (1974) have documented environmental effects on soil Rn content. Therefore, although the presence of U mineralization may manifest itself by an increase in ^{222}Rn, other factors not related to mineralization may also produce the same effect. Because of the complexity of the factors controlling ^{222}Rn release, transport and accumulation in the natural environment, Rn surveys are often simply used to delineate areas of anomalous Rn concentration. However, with proper control on the meteorological, geological, hydrogeological and geochemical setting, Rn surveys can be made to yield valuable information regarding U mineralization.

With the understanding as presented above, and instrumental development thus far, Rn studies has found application in the exploration for U deposits especially in areas where thick overburden present problems for gamma-ray detection. As part of a series of investigations to study U occurrence in the Nigerian Younger Granite, a soil Rn survey was carried out to determine the applicability of soil n studies as a means for U exploration in the region. The survey was thus carried out to investigate the distribution of Rn in the soils of the Nigerian Younger Granite and discover any relationship between Rn activity and the U and Th content of both the soil and its associated rocks.

Geology and Radioactivity of the Younger Granites

The Nigerian Younger Granites form a series of ring complexes in the northern part of the country. These granites form a series with similar occurrences in Niger and Cameroon, and in most part of North Africa. Detailed geology of the Younger Granites has been discussed elsewhere. Bowden (1985), for example, gave a detailed account of the geology of the Younger Granites; Butt and Cole (1985) discussed details of the geology of the Nigerian Younger Granites, while Macleod (1954) detailed the geology of the Jos-Bukuru ring complex. The origin of the Younger Granites has, in general, been a subject of debate. For the Nigerian Younger Granites, however, it is generally believed that they were emplaced through three major eruptive cycles, each producing distinct type of mineralisation.

The Younger Granitic province contains a host of granites of many textural variations including fine, medium and coarse-grained biotite granites, fayalite - hornblende granite, microgranite, aegerine granite and riebeckite granites. Both late-stage and post magmatic alterations have, to a large extent, affected these granites, modifying their mineralisation. Albitization, in particular, was pervasive and has greatly affected a large portion of the area covered by these granites. The process is believed to have enriched the granites with the radioelements U and Th. The elements are mostly concentrated in the mineral pyrochlore in these granites. Other alteration processes that were recorded in the region include wall rock alterations, kaolinisation, argillization, fluorization, sericitization, and greisenisation and silification (Kinnaird, 1985). These late-stage alteration processes are associated with the wide spread mineralisation in the granite province of the Younger Granites.

The Younger Granites are specialised with high radioactivity and enhanced levels of U and Th, and other Rare Earth Elements (REE). The U, Th and 40K content of the granites average 12 ppm, 26ppm and 4.2%, respectively (Dewu, 1989). Higher U values occur in some regions, more especially in the albite riebeckite granite where U is believed to be held in the mineral pyrochlore, which may record U concentration of up to 5% U3 08. Generally, the granites are anomalously enriched in the elements Sn, Nb, W, and also enriched in the Large Ion Lithophile (LIL) elements (Dewu, 1989).

The Younger Granite hosts a number of important radioactive accessory minerals in addition to some other minerals of economic importance. The radioactive minerals include pyrochlore, zircon, monazite, Xenotime, thorite and uraninite (Dewu, 1994). Some of these minerals are associated with specialised rock-types e.g. pyrochlore is usually concentrated in the albitized phases of the granites, thorite, the U-rich type, has been found concentrated in association with cassiterite bearing granites in the Jos-Bukuru complex. Thorite in particular has been recovered in large quantities as by-product in tin mining operations (Butt and Cole, 1985; Kinnaird, 1985).

The occurrence of these minerals in the granites and their high abundance combine to give the Younger Granites characteristically high radioactivity with the result that they are classified within the High Heat Production granites (Kinnaird *et al.*, 1985).

Radon in U Exploration

Being a daughter in the main U decay series, the presence of Rn in an environment can be used to infer the presence of U. Indeed, over the years, Rn studies have found application in the exploration for U ore deposits especially in areas where thick overburden presents problems for gamma-ray detection. Because of its short half-life (3.82days), Rn has always been considered to be associated with U-bearing minerals in its immediate environment, but this is not always the case.

Rn survey techniques in U exploration have recorded a number of successes. Investigations which show Rn measurements, either alone or in conjunction with other techniques to have been responsible for detecting U occurrence have been reported. In the first application of the method (Ridland, 1945), detected a vein of pitchblende about 18m below the surface. Dyck (1972) found close correlation between Rn anomalies and gamma ray activity in the determination of soil Rn over a known U deposit in Bancroft. Development of instrumental technique largely pioneered by the Geological Survey of Canada (Dyck, 1969) has led to wide application of the method for U exploration (Butt and Cole, 1985; Gregory, 1987). In areas that lack soil cover, such as in parts of Canada, Rn measurements on surface and ground waters yield good results. For example a regional surface water survey over the Bancroft and Elliot Lake areas by Dyck and Smith (1968) outlined zones of U enrichment. Similarly, Dyck and Cameron (1975) and Reimer et al.,(1979) reported detecting Rn anomalies related to U mineralisation based on measurements on groundwater from boreholes in areas of known U mineralisation.

Although successes have been reported both for soil and surface and/or ground water surveys, the present situation is that soil surveys have proved to be the more successful in indicating U mineralisation. For example, in an investigation of a uranium vein reported by Gregory (1987), both soil and stream ^{222}Rn determinations were carried out. The soil survey detected the vein quite clearly, but water ^{222}Rn values were barely above background even directly over the vein.

Applications of Rn measurements in U exploration are not without limitations.

Various environmental factors, which control Rn content place restrictions on the use of Rn surveys for U exploration. The soil Rn content is subject to a number of varying environmental factors such as, soil moisture content, and atmospheric conditions. These not withstanding however, with proper control on these factors, soil Rn surveys can be used to yield such important information as U mineralisation in the environment.

Methodology

In the investigation of uranium occurrence in the Nigerian Younger Granite, a number of methods were employed to study the Jos-Bukuru Ring complex. These studies include aero-radiometric and ground four-channel gamma-ray spectrometric survey, alpha-particle autoradiography including microprobe analysis, laboratory gamma-ray spectrometric analysis, and alpha-particle spectroscopy and whole rock and soil elemental analysis using X-ray fluorescence techniques. These techniques, particularly the gamma ray spectrometric ones, indicate the Younger Granites to be enriched in the radioelements U, Th and 40K. As part of these series of studies, a soil radon survey was carried out to determine the applicability of soil Rn studies as a means of U exploration in the region.

The Rn survey involved measuring soil Rn activity in holes where soil samples were collected. The soil samples were collected in close association with the various rock-types from which the soils were derived. This made it possible to relate the soil Rn activity to the U and Th content in both the soils and their associated rocks. For the measurements, an alpha/radon detecting system was used. The system consists of a cartridge of alpha-cards (thin aluminised Mylar discs) as the Rn detectors and a card reader system designed to read the Rn activity on the alpha cards.

As Rn survey is sensitive to environmental conditions, such factors that could affect the results were taken into consideration in the treatment of the data obtained. Cards found to be coated with moisture were re-exposed in order to remove the effect of moisture on the results. A base station was also established to monitor any environmental effects on the Rn activity in the region during the period of measurement.

Soil Rn activity was measured in-situ using an alpha card system. The system made by alphaNUCLEAR consists of a cartridge of alpha/Rn cards and a card reader. The cards are made of thin aluminised Mylar discs supported by a rectangular plastic frame. To take a measurement using this system, holes (with diameters between 15 cm and 20cm) where soil samples were collected were used to expose the cards to Rn from the soil. To expose the cards, the cards were simply suspended on cross-sticks wedged between the sides of the hole. The holes were then covered with hard boards, usually cardboard sheets and then polythene sheets spread over the boards. The assembly of board and polythene sheets is then covered with a layer of soil. The arrangement is shown in Figure 1. The card number, the time of burial and the station number were then recorded for each station. The Cards were exposed for a period of between 18 and 24 hrs. A base station was set up to monitor any environmental effect on the readings on the cards in the region. Using this arrangement, soil Rn activity in the area was measured. Details on card reading, isotope discrimination from the card readings are reported Dewu (1989). Sites of data collection are shown in Figure 2.

Analyses, Results and Discussion

In addition to the soil Rn activity measurements, soil samples from these sites and their associated rock samples were analysed for their U and Th content. Elemental composition of both the soil and rock samples was determined using X-ray Fluorescence analysis with a fully automated Philips, PW1400, XRF system. The reliability and precision of the instrument had been fully determined and found to be good, especially for the elements U and Th (Heath, 1982). The results (Figure 2) indicate a wide variation in soil Rn activity in the soils of the Younger Granite province. The Rn activity range from as low as 4cpm to high counts of up to 71cpm. High counts

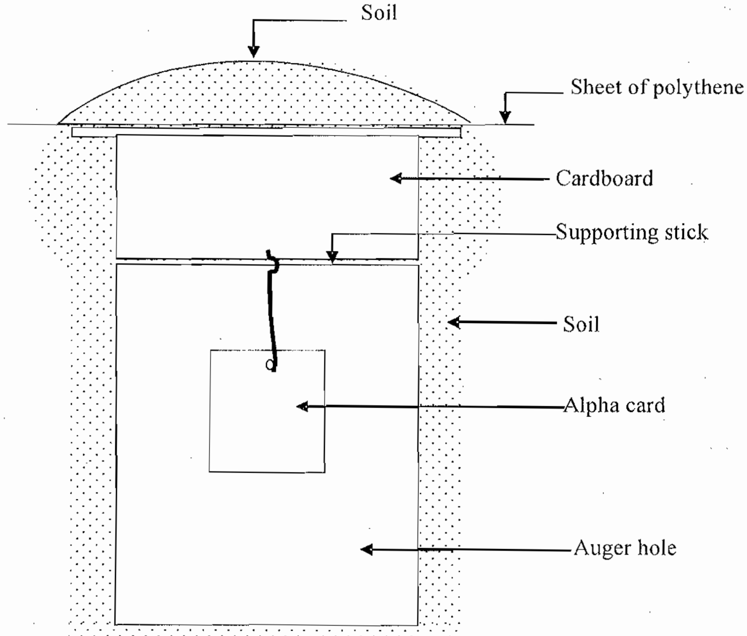


Fig.1: Arrangement for soil radon measurement

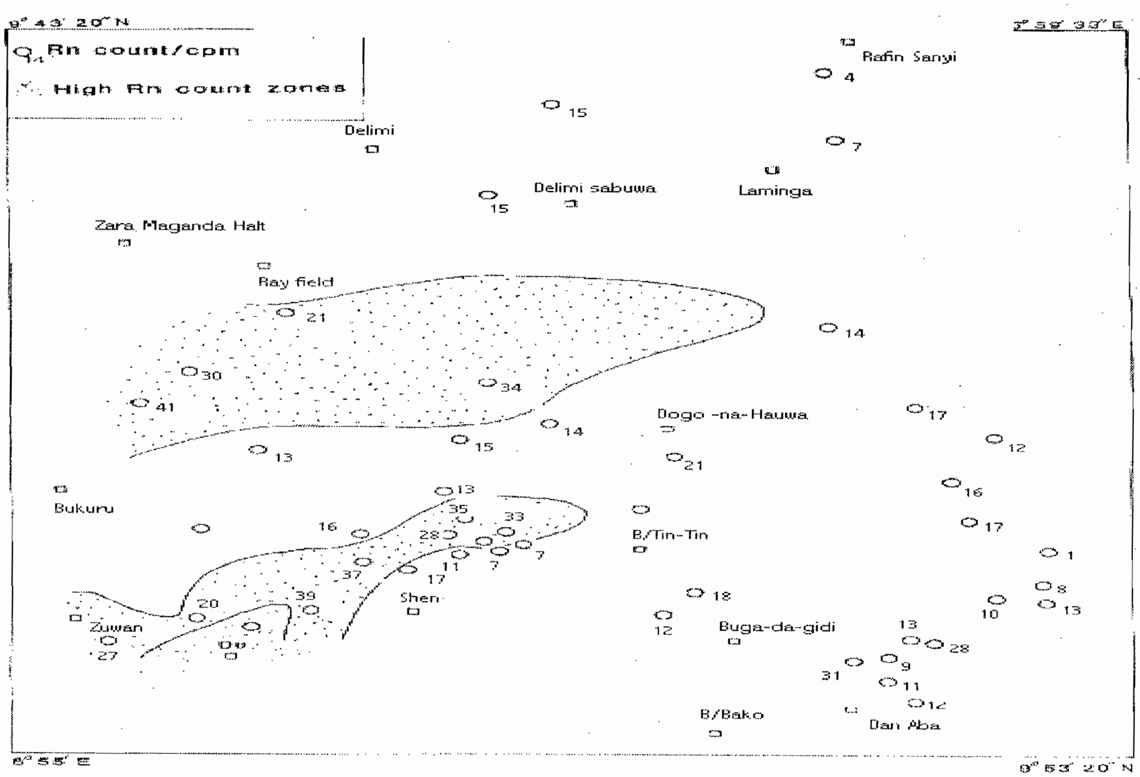


Fig. 2: Rn distribution in Jos plateau soils

are normally associated with soils of high porosity, which suggest the importance of diffusion in controlling Rn transport in the region.

.In addition to total Rn activity, the activities of the individual Rn isotopes ^{222}Rn and ^{220}Rn were also calculated. The results are given in Table 1, in which is displayed the total Rn activity, the activities of the isotopes, ^{222}Rn and ^{220}Rn; and the soil and rock U and Th concentration for the sites where these measurements were taken.

The survey was carried out towards the end of the rainy season in the region, and, therefore, the soil was moist with a high rate of radon diffusion expected. The heavy rainfall characteristic of the region has been found to wash down Rn to lower soil horizons (Tanner, 1954). These factors thus need to be considered in interpreting the results.

To investigate the relationship between Rn activity and the U and Th content of the soil on the one hand and the U and Th content in the rocks associated with these soils on the other, a number of correlations were considered. In these, correlation of soil Rn activity with sum total of U and Th in both the soil and its associated rocks, respectively, were considered. These correlations show that it is only the latter that gave any significant correlation. The generally low correlation in both the soil and the associated rocks suggests the importance of factors other than the U and Th content of the soil on the Rn activity. Because of the low correlation also, it is difficult to determine the major contributor to the soil Rn from these plots. In Figure 3 are shown plots of soil Rn activity against the U and Th content in the rocks. In Figures 4 and 5 are shown the plots of Rn activity against soil and rock U content, respectively; and soil and rock Th content, respectively. In both plots, soil Rn is more associated with the rock radioelement contents than with the soil radioelement contents. Clearly this suggests the dominance of rock U and Th in contributing to the soil Rn content. The closer association of the soil Rn with the rock U and Th than with the soil U and Th; plus the closer association of ^{220}Rn

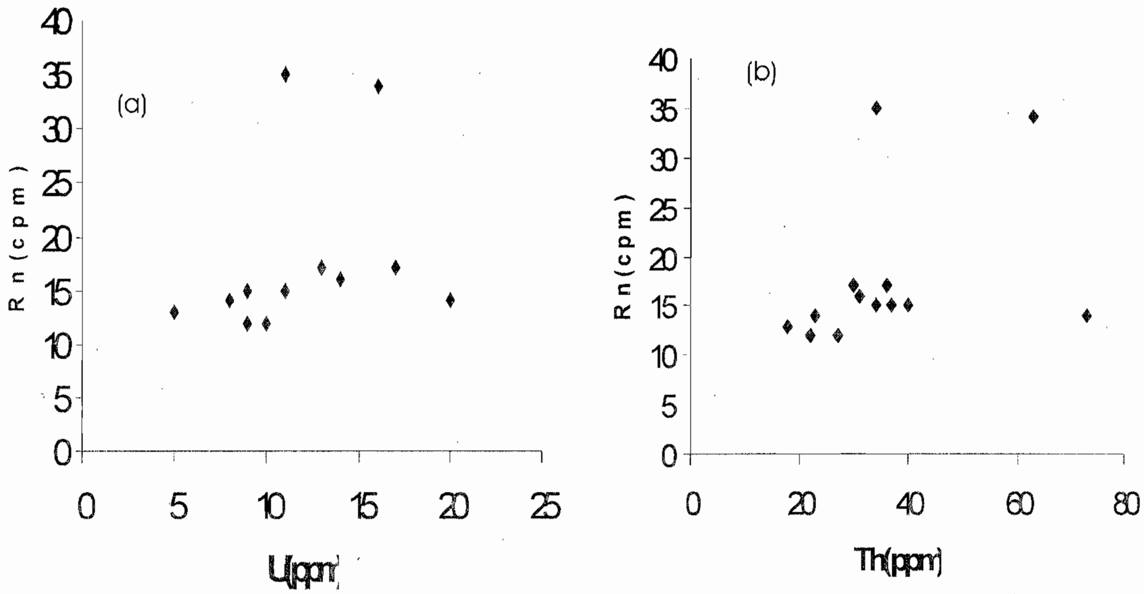
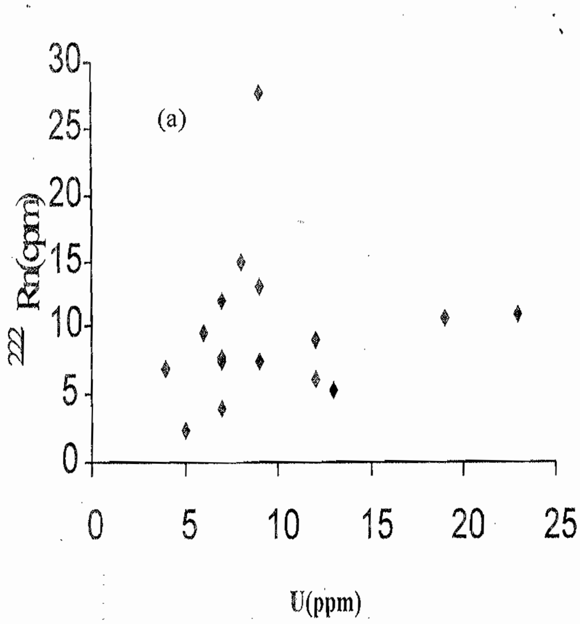


Fig. 3: Plot of soil Rn versus rock U(a) and rock Th(b)



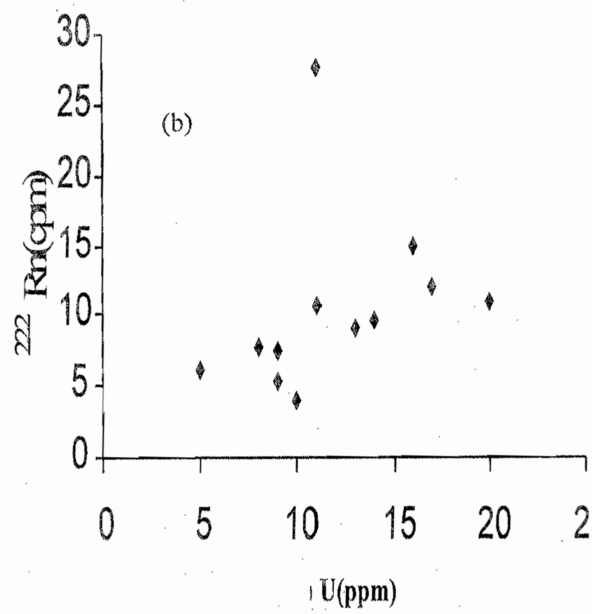
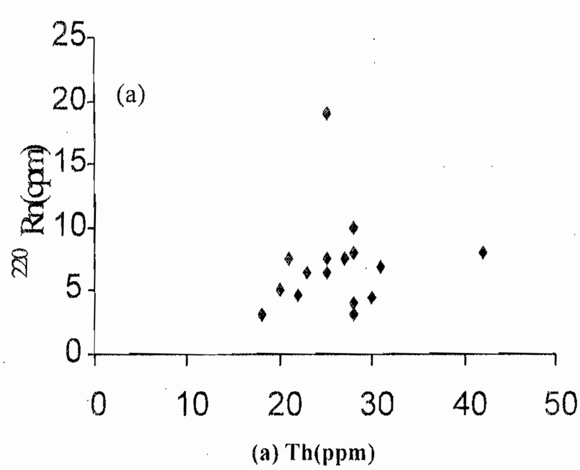


Fig. 4: Plot of soil ^{222}Rn versus soil U(a) and rock U(b)

with Th than ^{222}Rn with U suggest that U in the soil is young, ^{238}U in the soil has not had enough time to reach secular equilibrium with its daughters.

In general the results suggest that ^{222}Rn is associated more with U in the underlying rocks than with U in the soils in the Younger Granite province of the Jos-Bukuru Complex. Soil Rn surveys can thus be used to investigate U occurrence in the underlying rocks of the region. Th in the soil on the other hand, possibly occurring in resistate minerals is able to support the soil ^{220}Rn content. This is because the ^{232}Th decay series contains daughters with relatively short half-lives, it therefore takes relatively very short time to reach secular equilibrium with its daughters.

That soil Rn is associated more with the radioelement content of the underlying rocks implies that some active upward transport is taking place. These may either be Rn transported in its gaseous phase or ^{226}Ra in solution. The former means the presence of atmospheric pumping where the gas is conveyed through fractures while the latter may



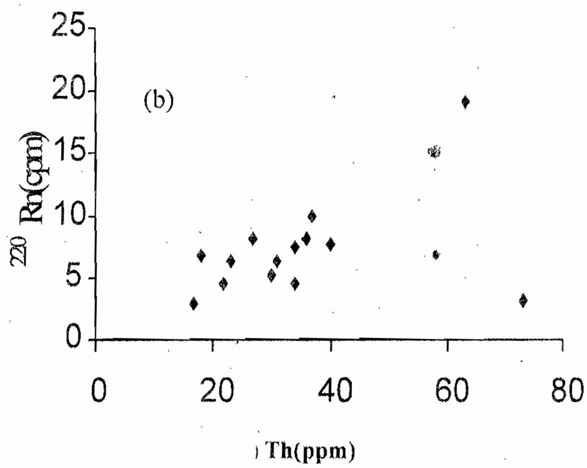


Fig. 5: Plot of soil ^{220}Rn versus soil Th(a) and rock Th(b)

Table 1: Soil Rn activity with associated soil and rock U and Th concentrations

Site	Radon activity in cpm			U and Th concentration in ppm			
	Rn(Total)	^{222}Rn	^{220}Rn	U_s	Th_s	U_r	Th_r
JP08	12	7.4	4.6	7	22	9	22
JP09	10	7.0	3.0	4	18	-	17
JP14	13	6.2	6.8	12	31	5	18
JP15	15	5.2	9.8	13	28	9	37
JP16	34	14.9	19.1	8	25	16	63
JP17	14	10.8	3.2	23	28	20	73
JP28	35	27.5	7.5	9	25	11	34
JP41	17	9.0	8.0	12	42	13	36
JP42	17	11.9	5.1	7	20	17	30
JP43	16	9.6	6.4	6	23	14	31
JP44	12	4.0	8.0	7	28	10	27
JP46	14	7.6	6.4	7	25	8	23
JP48	15	7.4	7.6	9	27	9	40
JP49	15	0.5	4.5	19	30	11	34
JP61	17	12.9	4.1	9	28	-	-
JP62	04	4.0	-	7	20	-	-
JP65	10	2.4	7.6	5	21	-	-

Note: 1:U_s and Th_s refer to soil U and Th, respectively
2:U_r and Th_r refer to rock U and Th, respectively

mean upward movement of groundwater (supported either by a fracture head or a thermal head). All these are likely in the Younger Granite regions.

Conclusion

The investigation reported above shows that rocks underlying the associated soils mainly support soil Rn in the Nigerian Younger Granite province. In view of the strong correlation between ^{222}Rn in the soil and the U content of the underlying rocks, it is believed that soil Rn activity is a better indicator of U at depth than the U content of the soil itself. The investigation also provide evidence which show that U in the soil is young, having not had enough time to reach equilibrium with its daughters.

The result indicates that where the need arises, and with proper environmental control, soil Rn survey can be applied to delineate U occurrence in the region. However, any such surveys should take into cognizance the contribution of ^{220}Rn to the total Rn.

References

Bowden. P. (1985): The Geochemistry and Mineralisation of Alkaline Ring Complexes in Africa - A review. Journ. Afr. Earth Sci. 3 (1/2), 17-39.

Buchanan, M. S. (1977): The Geology of the Jos Plateau. Bull. Geo. Surv. Nig., vol. 2&3

Butt, C. R. M., and Gole, M. J. (1985): Helium in Soil and Overburden as an Exploration Pathfinder - An Assessment. Journ. Geochem. Explo. 24, 141-173.

Caneer, W. T. and Saum, N. M., (1974): Radon emanotry in uranium exploration. Ming. Eng. 26, (5), 26-29.

Dewu, B. B. M. (1989): Distribution of Uranium in Granites and Uranium Mobility During Low Temperature Alteration Processes, Ph. D. Thesis, Univ. of Exeter.

Dewu, B. B. M. (1994): Radioactive Accessory Minerals of the Nigerian Younger Granites. (Abstract) NMGS Book of Abstract, 1994.

Dyck, W. (1969): Development of Uranium Exploration Methods Using Radon. Pap. Geol. Surv. Can., 69-96.

Dyck, W. (1972): Radon Methods of Prospecting In Canada. In: S. H. U., Bowie, M. Davis and D., Ostle (Eds), Uranium Exploration Handbook, Inst. Ming. Metall., London.

Dyck, W. and Cameron, E. M. (1975): Surface Lake Water Uranium Radon Survey of Lineament Lake Area District of Mackenzie. Geol. surv. Can. papers, McGraw-Hill, New York.

Dyck, W. and Smith, A. Y. (1968): Use Of Radon-222 in Surface Waters for Uranium Geochemical Prospecting. Can. ming. Journal, 100-103.

Gregory, R. G. (1987): Soil Gas Emanometry and Hydrothermal Mineralisation in Southwest England. Unpubl. Ph. D. Thesis, Univ. of Exeter.

Heath, M. J. (1982): Uranium in the Dartmoor Granite: Geochemical and Radiological Investigations in Relation to the south-west England Geothermal Anomaly. Unpubl. Ph. D. Thesis, Unive. Of Exeter.

Kinnaird, J. A. (1985): Hydrothermal Alteration and Mineralisation Of The Alkaline Anorogic Ring Complexes Of Nigeria. J. Afr. Earth Sci. 3, (1\2), 229-251.

Kinnaird, J. A., Batchelor, R. a., Whitley, J. E., and Mackenzie, A. B. (1985): Geochemistry, Mineralisation and Hydrothermal Alteration Of The Nigerian High Heat Production Granites. In: High Heat Production (HHP) Granites Hydrothermal Circulation and Ore Genesis, Inst. Ming. Metall., London.

Kovach, E. M., (1946), Diurnal variations in radon content of soil gas. Terr. Magnetism and Atmosph. Electricity 51, 45-46.

Kraner, H. W., Schoeder, G. L. and Evans, R. D., (1964), Measurements of the effects of atmospheric variables on radon-222 flux and soil gas concentrations. In: Adams, J. A. S. and Lowder, W. M. (eds.), The Natuural Radiation Environment, Chicago Univ. Press, 191-215.

Macleod, W. N. (1954): The Geology of the Jos-Bukuru Younger Granite Complex with Particular Reference to the Distribution Of Columbite. Mem. Geol. surv. Nig., vol. 33.

Reimer, G. M., Derton, E. H., Friedman, I. and Otton, J. K. (1979): Recent Developments in Uranium Exploration Using The U. S. Geological Survey's Mobile Helium Detector. Journ. Geochem. Explo., 11, 1-12.

Ridland, G. C. (1945): Use of Geiger_Muller counter in the Search for Pitchblende-Bearing Veins at Great Bear lake, Canada. Trans. Am. Inst. Min. Metall. And Eng. 164, 117-124.

Tanner, A. B. (1954): Radon Migration In The Ground: A Review. In: Adams, J. A. S. And Lowder, W. M. (Eds), Natural Radiation Environment. Chicago Univ. Press, 161-190.