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Occupational Natural Radiation Exposure at the Uranium Deposit of Kitongo, Cameroon

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Soil samples were collected around the uranium deposit of Kitongo to determine activity concentrations of 238U, 232Th and 40K using a NaI(Tl) detector. Ambient dose equivalent rates were measured using a survey meter. 222Rn-220Rn discriminative measurements were performed using passive type monitors. The activity concentrations of 238U, 232Th and 40K ranged from 20±5 to 337±78 Bq kg⁻¹, 14±4 to 53±13 Bq kg⁻¹ and from 21±4 to 897±190 Bq kg⁻¹ respectively, with the average values of 99±24, 27±4 and 592±125 Bq kg⁻¹, respectively. Ambient dose equivalent rates ranged from 0.05 µSv h⁻¹ to 6.20 µSv h⁻¹. Higher values were recorded inside the two galleries of the uranium deposit. The maximum dose to prospective miners was calculated to be 15.2 mSv y⁻¹. In case of the miner staying permanently inside the galleries, the external dose received would vary from 32 to 54 mSv y⁻¹ with an average of 43 mSv y⁻¹. Radon concentrations ranged from 416±13 to 523±14 Bq m⁻³ with the average value of 486±14 Bq m⁻³. Occupational inhalation dose ranged from 2.8 to 3.5 mSv y⁻¹ with an average value of 3.2 mSv y⁻¹.

Key Words: uranium deposit, gallery, radon, ambient dose equivalent rate

1. Introduction

There are three natural radioactive decay series occurring in the environment, 238U, 235U and 232Th. To these three series, 40K can be added as natural radionuclides contributing to the primary radiological exposure of the public. These radionuclides are evenly distributed in soil or rocks. In the case of uranium, its average concentration in soils is in the order of 3 g per ton. In seawater uranium has an average concentration of 3 mg m⁻³. Its most abundant natural ore is pitchblende, which can appear in the form of veins. Uranium ore is considered as a mineable ore at a level of 1 to 2 kg of uranium per ton of
 ore, several hundred times than its natural concentration in soil.

Uranium ore is a slightly radioactive material because of the very long half-life of uranium in the order of several billion years and does not pose considerable danger to the environment if it remains in its natural state. However, after drilling, mining, or dismantling a mine, radioisotopes elements are more exposed. Wind diffuses them in all directions, streaming water transports them, and they infiltrate into the water table, gaseous radioactive elements radon \((^{222}\text{Rn})\) and thoron \((^{220}\text{Rn})\) are freely diffused.

The Kitongo uranium deposit belongs to the uranium-bearing region of Poli in Northern Cameroon. Since 1950 several investigations have been conducted for ore assessment. It was concluded that the Kitongo deposit is of metasomatic type and could contain an ore reserve of 10,000 t \(\text{U}_3\text{O}_8\) at a grade of 0.1%. Additional ongoing prospection could lead to an inventory significantly greater than 13,000 t \(\text{U}_3\text{O}_8\). The International Atomic Energy Agency through the World Distribution of Uranium Deposits (UDEPO) software estimates the resource range as 10,000–25,000 t \(\text{U}_3\text{O}_8\) at the grade of 0.05–0.1%. These investigations have certainly exposed buried radioisotopes. Several cores drilled from granite were abandoned at the site. Two galleries with the length of 37 and 74 m from their respective entrances were opened in the rock of the Kitongo Mountain. Residues of rocks and dust were scattered in the area. Near this summit, there are some inhabited villages. No environmental radioactivity monitoring has been done around Kitongo uranium deposit after the ore assessment. Saidou et al.\(^3\), \(^4\) reported natural radioactivity measurements and corresponding dose assessment in the uranium-bearing region of Poli except for the surroundings of the deposit.

This paper presents an evaluation of radiological exposure, with a highlight on \(^{238}\text{U}\), \(^{232}\text{Th}\) and \(^{40}\text{K}\) activity concentrations in the collected soils, on ambient dose equivalent rates registered at the site, and on radon concentrations inside galleries of the deposit.

This work is a preliminary part of an incoming large environmental radioactivity monitoring campaign at the Kitongo uranium deposit area.

2. Materials and methods

2.1 Study area

The uranium deposit of Kitongo is located in the uranium-bearing region of Poli in the North region of Cameroon. It is a metasomatic uranium deposit type. Fig. 1 shows the location of the uranium deposit and the populated areas around the deposit: Poli, Gouna, Gompou, Fignole, Hoy and Gormaya. The most populated town is Poli.

2.2 Gamma-ray spectrometry

Ten soil samples were collected around the uranium deposit of Kitongo. All measurements were performed using a NaI(Tl) detector (Model 802) with a crystal size of 7.6 cm\(\times\)7.6 cm, a multiple channel analyzer of 1024 channels and a resolution of 7.5% at 662 keV. The detector is mounted inside a cylindrical lead shielding. Spectrum acquisition and analysis were carried out using the GENIE 2000 software (Canberra). The spectrometer was calibrated using a 500 mL Marinelli Beaker-resin containing \(^{155}\text{Eu}\), \(^{57}\text{Co}\), \(^{113}\text{Sn}\), \(^{137}\text{Cs}\), \(^{54}\text{Mn}\) and \(^{65}\text{Zn}\) traceable to international standards and emitting gamma-rays in the 60.0–1115.5 keV energy range. The same geometry was used with the counting time of 100000 seconds to measure radioactivity in soil samples. The full energy peaks of interest for \(^{214}\text{Bi}\) (1120.0 and 1764.5 keV) were considered to determine \(^{238}\text{U}\) activity concentration after reaching secular equilibrium between \(^{226}\text{Ra}\) and daughter products. The full energy peaks of interest for \(^{228}\text{Ac}\) (338.8, 409.5 and 911.6 or 969.1 keV depends on the sample) were considered to determine \(^{232}\text{Th}\) activity concentration. The specific full energy peak 1460.8 keV of \(^{40}\text{K}\) was
used to determine its activity concentration.

2.3 Ambient dose equivalent rate measurements and outdoor external dose assessment

A pocket survey meter (RadEye PRD-ER, Thermo Scientific) was used for the ambient dose equivalent rate measurements. It incorporates a high sensitivity NaI(Tl) scintillation detector with a miniature photo-multiplier allowing the detection of very low radiation levels with an emphasis on gamma emissions below 400 keV. Measurements were conducted at 1 m above the ground surface. The weather conditions were sunny throughout the entire measurement period.

External dose around the deposit and inside the two galleries was assessed applying the formula expressed in equation (1).

\[
E = \hat{D} \times t
\]  

Where \( \hat{D} \) is the arithmetic mean of ambient dose equivalent rate measured by the above pocket survey meter, \( t \) is the occupancy time taken to be 1848 h per year for workers, assuming 7 h per day, 6 days per week and 11 months per year.

2.4 Indoor \(^{222}\)Rn concentrations and inhalation dose assessment

Indoor radon studies for health risk assessment are widely conducted in many countries using different types of detectors. It has been known that thoron \((^{220}\text{Rn})\) concentration is sometimes much higher than that of radon. Radon, thoron and their progenies all contribute to the lung dose, the health risk assessment due to indoor radon extends to the measurements of thoron and their progeny. In this
study, a passive type radon–thoron discriminative monitor called a RADUET (Radosys Ltd, Hungary, Fig. 2) using a solid-state track detector (CR-39; Radosys Ltd) was used to evaluate the average radon concentration.\textsuperscript{5)} CR-39 plates were fixed at the bottom of the chambers with sticky clay. Radon gas diffuses into the chamber through an invisible air gap between the lid and bottom of the chamber. Since this air gap functions as a high diffusion barrier, little thoron enters the chamber due to its very short half-life (55.6 s), compared to that of radon (3.82 d). To detect thoron more effectively, six holes of 6 mm in diameter are opened at the side of the other chamber and are covered with an electroconductive sponge.

There are two galleries excavated in the Kitongo Mountain with 37 m and 74 m of length from their respective entrances. They were drilled during prospection phase of the Kitongo deposit. RADUETs were placed at the height of 1.5 m from the ground inside the galleries for two months. After two months of exposure, the detectors were collected and sent back to Hirosaki University. The CR-39 plates were taken out of the chambers, chemically etched with a 6 M NaOH solution at 60°C for 24 h, and photos of the alpha tracks were taken by a digital camera using a microscope. The number alpha tracks were then counted using Image-J which is a public domain, Java-based image processing program developed at the National Institute of Health. To determine conversion factors of radon and thoron concentrations, these detectors were placed into the radon and thoron calibration chambers at Hirosaki University, respectively.\textsuperscript{6)} The average radon and thoron concentrations ($C_{\text{Rn}}$ and $C_{\text{Tn}}$) were calculated using equations (2) and (3) which were reported by ISO.\textsuperscript{7)}

$$C_{\text{Rn}} = \frac{(d_L - b_L) f_{\text{Tn}2} - (d_{\text{H1}} - b_{\text{H1}}) f_{\text{Tn}1}}{t (f_{\text{Rn}1} f_{\text{Tn}2} - f_{\text{Rn}2} f_{\text{Tn}1})}$$ \hspace{1cm} (2)

$$C_{\text{Tn}} = \frac{(d_{\text{H1}} - b_{\text{H1}}) f_{\text{Rn1}} - (d_L - b_L) f_{\text{Rn}2}}{t (f_{\text{Rn}1} f_{\text{Tn}2} - f_{\text{Rn}2} f_{\text{Tn}1})}$$ \hspace{1cm} (3)

where, $d_L$ and $d_{\text{H1}}$ were alpha track densities (track
cm$^{-2})$ for the low and high air-exchange-rate chambers, respectively. $f_{Rn1}$ and $f_{Tn1}$ were the respective conversion factor from alpha track densities to radon and thoron activity concentration for the low exchange-rate air chamber [(tracks cm$^{-2}$ h$^{-1}$) (Bq m$^{-3}$)$^{-1}$]. $f_{Rn2}$ and $f_{Tn2}$ were the respective conversion factors from alpha track densities to radon and thoron activity concentration for the high exchange-rate air chamber [(tracks cm$^{-2}$ h$^{-1}$) (Bq m$^{-3}$)$^{-1}$].

t was the exposure time (h) and $\bar{b}$ were the backgrounds of the alpha track density (track cm$^{-2}$) on the CR-39 detector.

For inhalation dose assessment equation (4) was used.\(^8\)

$$E_{inh} = (A_{inh} \times e_{inh} \times F_{eq})_{Rn} \times F_{occ} \times t \quad (4)$$

$A_{inh}$ is the average radon concentration, $e_{inh}$ is the inhalation dose conversion factor of 9 nSv (Bq h m$^{-3}$)$^{-1}$, $t$ is the occupancy same as above, and $F_{eq}$ is the equilibrium factor 0.4.

3. Results and discussion

3.1 Activity concentrations of $^{238}$U, $^{232}$Th, and $^{40}$K

Activity concentrations of $^{238}$U, $^{232}$Th and $^{40}$K for the ten soils sampled are summarized in Table 1. The lowest $^{238}$U concentration of 20 ± 5 Bq kg$^{-1}$ was measured in soil sampled at 2 km from the galleries’ location, and the highest concentration of 337 Bq kg$^{-1}$ was found in soil sampled near the galleries. Fig. 3 shows the distribution of the measured radionuclide activity concentrations in the ten soils sampled.

Regional studies investigating uranium and another radionuclide occurrence were performed in the region. Using gamma spectrometry in the laboratory, the activity concentrations ranged from 13 to 57 Bq kg$^{-1}$ with an average of 24 Bq kg$^{-1}$ for $^{238}$U, from 15 to 58 Bq kg$^{-1}$ with an average of 28 Bq kg$^{-1}$ for $^{232}$Th and from 112 to 1124 Bq kg$^{-1}$ with an average of 506 Bq kg$^{-1}$ for $^{40}$K.\(^3\)

By in-situ gamma spectrometry, the activity concentrations ranged from 12.5 to 57 Bq kg$^{-1}$ with an average of 31.5 Bq kg$^{-1}$ for $^{238}$U, from 10 to 67 Bq kg$^{-1}$ with an average of 31 Bq kg$^{-1}$ for $^{232}$Th and from 242 to 777 Bq kg$^{-1}$ with an average of 510 Bq kg$^{-1}$ for $^{40}$K. Therefore, results agree well with the world average values for soil; 33 Bq kg$^{-1}$ for $^{238}$U, 45 Bq kg$^{-1}$ for $^{232}$Th and 420 Bq kg$^{-1}$ for $^{40}$K.\(^8\)

Furthermore, the large difference between $^{238}$U and $^{232}$Th activity concentrations found at the Kitongo deposit shows clearly that the area contains more uranium than thorium.

Radioactivity of $^{238}$U, $^{232}$Th and $^{40}$K in loess soils collected in Shanxi and Shaanxi provinces in China ranged from 30 to 37 Bq kg$^{-1}$, 41 to 47 Bq kg$^{-1}$ and 578 to 670 Bq kg$^{-1}$ with arithmetic mean of 34, 44, and 614 Bq kg$^{-1}$ respectively.\(^9\) $^{238}$U average activity concentration found in the present study is higher than those found in the studies mentioned above. However, it is comparable to the activity concentration found around former uranium mine
of Gabrovnica. Where values ranged from 45 to 442 Bq kg\(^{-1}\) for \(^{238}\)U, from 33 to 170 Bq kg\(^{-1}\) for \(^{232}\)Th and from 520 to 1510 Bq kg\(^{-1}\) for \(^{40}\)K. Measurements taken at the former uranium mines in Portugal gave an average activity of 200 Bq kg\(^{-1}\) for \(^{238}\)U, 200 Bq kg\(^{-1}\) for \(^{226}\)Ra and 91 Bq kg\(^{-1}\) for \(^{232}\)Th. In Sabugal region in Portugal, radionuclides measurements showed enhanced radioactivity levels in surface water streams related to water discharges from old uranium mines. Same authors analyzed radionuclides for soil, irrigation water, and vegetable products from areas receiving drainage from old uranium mines. Results showed enhancement of radionuclide concentrations, especially \(^{228}\)Ra, in vegetables from kitchen gardens. Further studies evidenced the importance of environmental monitoring of uranium deposits, mines or former mines areas. The airborne survey taken at the uranium mines of former Eastern Germany registered activity concentrations of \(^{226}\)Ra and \(^{232}\)Th, respectively, equal to 370 Bq kg\(^{-1}\) and 45 Bq kg\(^{-1}\) at the Ronneburg site; 1200 Bq kg\(^{-1}\) and 40 Bq kg\(^{-1}\) at the Crossen site and 470 Bq kg\(^{-1}\) and 57 Bq kg\(^{-1}\) at the Seelingstädt site. Data contained in Table 2 summarize the previous comparisons with other data.

3.2 Ambient dose equivalent rate and dose assessment

Ambient dose equivalent rate was measured at 20 points in the study area, 18 around the deposit and 2 inside the galleries. Fig. 4 shows its evolution. The first measurement point (13°16’602“E, 8°25’393“N) is located at approximately 2 km from the top of Kitongo Mountain. The eighth measurement point (13°16’702“E, 8°24’897“N) was carried out where the drill cores collected for the prospection were abandoned. An average of 0.09 μSv h\(^{-1}\) was registered at this point, which is above the regional background level. The two higher values of 3.7 and 6.20 μSv h\(^{-1}\) were recorded respectively at the entrance and inside of the gallery with 37 m of length. The high ambient dose equivalent rate found inside the gallery reveals that there is more uranium ore in the rock. The maximum value of air-kerma rate found on the sand surface of a beach in high background radiation area of Kerala was 2.1 μGy h\(^{-1}\). By applying the ratio
between ambient dose equivalent rate to air-kerma rate which corresponds to 1.25 $\mu$Sv $\mu$Gy$^{-1}$, the calculated maximum ambient dose equivalent rate of the area corresponds to 2.6 $\mu$Sv h$^{-1}$. Which is less than that found in this survey. Tokonami et al.9) performed short-term measurements of gamma-ray dose rate with the 1"×2" NaI(Tl) scintillation spectrometer indoors (cave residents) and outdoors in Shanxi and Shaanxi provinces in China at 32 sites. The scintillation spectrometer reading ranged from 0.12 to 0.18 $\mu$Sv h$^{-1}$ indoors and from 0.1 to 0.14 $\mu$Sv h$^{-1}$ outdoors, which is comparable to radiation dose found in some places in the present study.

Annual external radiation dose obtained from ambient dose equivalent rates recorded outside the galleries ranges from 0.4 to 2.9 mSv y$^{-1}$ with the average value of 1.2 mSv y$^{-1}$. It is considered as the annual external radiation dose received by the members of the public living in the surrounding area of the deposit. This average value is above the public reference level of 1 mSv y$^{-1}$. The regional value obtained by car-borne survey ranged from 0.20 to 0.83 mSv y$^{-1}$ which is mostly below of the value obtained in the present study.17) Furthermore, a dose received by a prospective miner working inside the galleries would range from 6.8 to 11.5 mSv y$^{-1}$. In case of a miner staying permanently inside the galleries, an annual effective dose that he would receive varies from 32 to 54 mSv y$^{-1}$ with the average value of 43 mSv y$^{-1}$, largely above the ICRP reference level of 20 mSv y$^{-1}$ for occupational exposure.18) For comparison, the maximum annual effective dose in Karunagappally Taluk was estimated to be 13 mSv y$^{-1}$.15)

### Table 2 Comparison of $^{238}$U, $^{232}$Th and $^{40}$K activity concentrations with corresponding data in other countries

| Study area                        | $^{238}$U concentration (Bq·kg$^{-1}$) | $^{232}$Th concentration (Bq·kg$^{-1}$) | $^{40}$K concentration (Bq·kg$^{-1}$) |
|-----------------------------------|---------------------------------------|----------------------------------------|--------------------------------------|
| Present study                     | Range 20 – 337 | Average 99 | Range 14 – 53 | Average 27 | Range 21 – 887 | Average 592 |
| Uranium bearing region of Poli, Lab Cameroon | Range 13 – 57 | Average 24 | Range 15 – 58 | Average 28 | Range 112 – 1124 | Average 506 |
| Shanxi and Shaanxi (China)        | Range 12.5 – 57 | Average 31.5 | Range 10 – 67 | Average 31 | Range 242 – 777 | Average 510 |
| Former uranium Mine of Gabronicia | Range 30 – 37 | Average 34 | Range 41 – 47 | Average 44 | Range 578 – 670 | Average 614 |
| Former Uranium mine (Portugal)    | Range 45 – 442 | Average - | Range 33 – 170 | Average - | Range 520 – 1510 | Average - |
| World average                     | Range - | Average 33 | Range 45 | Average 420 |
lery to measure indoor radon concentrations. In the gallery having 37 m of length, \(523 \pm 14 \text{ Bq m}^{-3}\) was measured by the first detector and \(501 \pm 14 \text{ Bq m}^{-3}\) by the second. In the gallery having 74 m of length, \(505 \pm 14 \text{ Bq m}^{-3}\) was measured by the first detector and \(416 \pm 13 \text{ Bq m}^{-3}\) by the second. Thoron concentrations provided by these detectors were very low and sometimes below the detection limit. The inhomogeneous distribution of thoron indoors disallows the assessment of its contribution to the lung dose. Its large variability indoors was proven in previously reported studies.\(^{19}\) Although, thoron progeny monitor can be used for assessment of thoron contribution to the lung dose.

The calculated occupational inhalation dose ranges from 2.8 to 3.5 mSv y\(^{-1}\) with an average value of 3.2 mSv y\(^{-1}\). Radon and thoron were measured in 100 houses of some villages of the region. Authors found concentrations ranging from 46 to 143 Bq m\(^{-3}\) and from 18 to 238 Bq m\(^{-3}\) with arithmetic mean values of 82 and 94 Bq m\(^{-3}\) for radon and thoron respectively. The inhalation dose due to radon ranged from 0.9 to 3 mSv y\(^{-1}\) with the average value of 1.6 mSv y\(^{-1}\).\(^{17}\) In the high background radiation areas of Southern-Cameroon, radon and thoron were measured using the same type of detectors, concentrations found there followed lognormal distributions and ranged respectively from 27±26 to 937±5 Bq m\(^{-3}\) and from 48±40 to 700±128 Bq m\(^{-3}\) respectively for radon and thoron. With arithmetic mean concentrations of 92±3 Bq m\(^{-3}\) for radon and 260±13 Bq m\(^{-3}\) for thoron.\(^{20}\) Many surveyed dwellings in the observed areas had higher thoron concentrations than those of radon, which indicates that the region could contain more thorium than uranium in average, in contrast to the present study where thoron concentrations are very low.

In the study of radon and thoron exposures for cave residents in Shanxi and Shaanxi provinces in China, measured with the same type of detectors, among 193 dwellings indoor radon concentrations ranged from 19 to 195 Bq m\(^{-3}\) with a geometric mean of 57 Bq m\(^{-3}\), indoor thoron concentrations ranged from 10 to 856 Bq m\(^{-3}\) with a geometric mean of 153 Bq m\(^{-3}\) which revealed that presence of thoron is not negligible for accurate radon measurements.\(^{9}\)

For houses surrounding former uranium mine of Gabrovnic, radon concentration measured ranged from 33 to 903 Bq m\(^{-3}\), and the study showed that good ventilation greatly affected \(^{222}\text{Rn}\) activity concentrations.\(^{10}\) The ventilation of galleries could greatly reduce radon concentration.

Viega et al.\(^{21}\) studied a radon exposure in Brazilian underground coal mine, authors found that average radon concentrations were above the reference action level for workplaces. Contrarily, the present study reveals radon concentrations below the reference level for workplaces. A different study conducted for evaluating the radiation dose of workers originating from radon in show Cave of Tapolca in Hungary showed that inhalation dose received could exceed the dose limit of 20 mSv y\(^{-1}\) for occupational exposure.\(^{22}\) However, radon measurements conducted around closed uranium mines in Japan reported an effective dose of 1 mSv y\(^{-1}\) at the fence boundary of waste rock site, which is less than the value found in the present study.\(^{23}\)

4. Conclusion

Radioactivity measurements and corresponding dose assessment at the Kitongo uranium deposit have been conducted for environmental monitoring and radiation protection of prospective miners as well as members of the public living in the area surrounding the deposit. The external radiation dose received by a prospective miner inside the deposit galleries ranges from 6.8 to 11.5 mSv y\(^{-1}\). In case of miners staying permanently inside galleries, the annual dose will range from 32 to 54 mSv y\(^{-1}\) with an average of 43 mSv y\(^{-1}\), largely above of
the ICRP reference level for occupational exposure established at 20 mSv y\(^{-1}\). These results indicate the need to complete and extend this study to the whole surrounding area of the deposit, particularly in the populated areas at the immediate vicinity of the deposit, to carry out indoor radon-thoron measurements, air-kerma rates measurements by car-borne survey and activity concentration measurements using in-situ gamma spectrometry. There is a need for natural radiation risk mapping covering the whole uranium-bearing region.

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