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In situ gamma ray measurements of radionuclides at a disused phosphate mine on the West Coast of South Africa

Jacques Bezuidenhout

Faculty of Military Science, Stellenbosch University, South Africa

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ABSTRACT

High levels of uranium and its radioactive progeny like radium is normally associated with phosphate mining. *In Situ* gamma ray spectroscopy as a survey tool has been successfully applied to assess radionuclide concentrations in various geographical environments. A transportable and robust gamma ray detection system (GISPI) was therefore employed to determine the concentrations of naturally occurring radionuclides at a disused phosphate mine on the West Coast of South Africa. The concentrations of radium, thorium and potassium were measured and plotted. The measurements showed fairly high concentrations with medians of 320 Bq/kg for ^{226}Ra, 64 Bq/kg for ^{232}Th and 390 Bq/kg for ^{40}K. The highest concentrations were however confined to specific areas of the mine. The effective dose due to gamma irradiation for the various areas of the mine was also estimated and the highest estimated level was 0.45 mSv/y. The article finally draws conclusions as to the origins and impact of the radiation.

1. Introduction

The most significance naturally occurring radionuclides are uranium (^{238}U), thorium (^{232}Th) and potassium (^{40}K). Distinctive concentrations of these nuclides relate to geological and morphological features of specific locations. Igneous rock and regolith originating from such rock typically has higher concentrations of naturally occurring radionuclides than sedimentary rock. Sedimentary phosphate and some shale are however exceptions to the rule. Sedimentary phosphate rocks that originated from marine environments are usually characterized by activity concentrations of uranium much higher than those of volcanic and biological rocks (Sahu et al., 2014). Sedimentary phosphate usually contains high amount of uranium with typically concentrations of 1500 Bq/kg (Harb et al., 2008). The mining and refinement of phosphate can therefore potentially distribute and enhance uranium concentrations in the environment. High levels of radioactive uranium consequently have an important impact on humans as well as the environment (UNSCEAR, 2000; Vandenhove et al., 2015).

Natural uranium comprises of ^{238}U (99.28%) and ^{235}U (0.71%), which are both radioactive. These nuclides have long decay chains consisting of radioactive daughters that radiate numerous particles and gamma rays. The gamma emitting progeny of ^{238}U are therefore also present in natural material and the decay chain would not necessary be in secular equilibrium. This is especially true if the soil were disturbed by anthropogenic activities, like mining. The most prominent daughter in the decay chain of ^{238}U is the long lived radium (^{226}Ra) nuclide. Gamma rays from the bismuth (^{214}Bi) daughter of ^{238}U are generally utilised to estimate uranium concentrations, under condition that secular equilibrium exist in the decay chain. ^{226}Ra is however before ^{214}Bi in the uranium decay chain and the measurement of the ^{214}Bi gamma rays therefore provides a better estimation of ^{226}Ra concentration than that of ^{238}U, especially when secular equilibrium is uncertain. This study will therefore refer to ^{226}Ra concentrations rather than of ^{238}U concentrations, but still keeping in mind the underlying relationship between these nuclides.

The West Coast Fossil Park on the West Coast of South Africa was founded on the premises of the disused phosphate mine. The fossil and phosphate deposits were formed as a result of the area being a river delta dating back to the late Miocene and early Pliocene epochs (Sciscio et al., 2013). The mine uncovered numerous fossils during its operation but subsequently closed to the end of the previous century. The closing gave the opportunity to establish a fossil park on the premises. Elevated levels of uranium are therefore expected in the area especially due to the marine origin of the phosphate deposits. A radionuclide survey was consequently

E-mail address: jab@ma2.sun.ac.za.

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undertaken to estimate the prevalence of naturally occurring nuclides and in specific that of radium. This following paragraphs deal with the methods and results of this survey on the fossil farm and possible reasons for variations in the concentrations of naturally occurring nuclides.

2. Experimental

2.1. Measuring system

The Gamma ray *In Situ* Portable Instrument (GISPI) was used in the radiometric survey the West Coast Fossil Farm (Bezuidenhout, 2015). The GISPI consists of a NaI(Tl) scintillation detector, a digital Multi Channel Analyser (MCA), a rugged tablet PC with an on-board GPS and real time analysis software that also controls the whole system (see Fig. 1). The NaI(Tl) detector (7.62 × 7.62 cm) is coupled to the MCA and sealed in a padded case to protect the instruments from mechanical shock and dust. The scintiSPEC® MCA (http://gs.flir.com/) that is produced by FLIR® has a USB connection that acts as the power source for operation and allows data transfer. A Trimble® Yuma rugged tablet PC (http://www.trimble.com/) with on-board GPS manages the systems and stores the data.

The system settings and spectrum acquisition was controlled by the winTMCA32® software (with 1024 channels), that is also produced by FLIR®. Program code was developed in the winTMCA32 software to analyse spectra and obtain geographical locations, as well as export this data in GIS compatible format. The winTMCA32 code directly acquired the geographical position coordinated from the onboard GPS via a virtual communications port. The code also corrects for detector dead time and the corrected counts from the various energy windows are then combined with the positions data of each measured point. The code finally stored all hardware settings, *in situ* spectra and the results in files.

The fossil park stretches over a large area and the detector system was therefore mounted on a quad motorcycle. The detector was fitted to the front metal structure of the motorcycle, 20 cm from the ground. The Yuma rugged table PC was mounted on the front carrier to make it easily accessible to the operator (see Fig. 1).

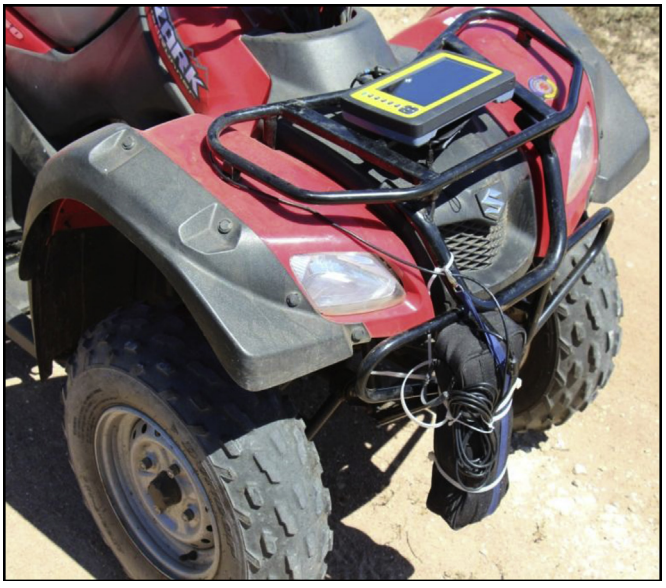

Fig. 1. Photographs of the measuring system that was utilised during the *in situ* measurements. The photographs show the casing with the detector mounted to the frame and the tablet with an inbuilt GPS on the carrier.

2.2. Calibrations

Energy calibrations were performed before and after each period of measurements. The calibrations were done in the range from 0.2 to 2.7 MeV by using anthropogenic nuclides and natural environmental spectra. The following nuclides and associated gamma ray emissions were used for the energy calibration: ^{214}Pb (351.3 keV), ^{137}Cs (661.7 keV), ^{60}Co (1173.2, 1332.5 keV), ^{40}K (1460.8 keV), ^{214}Bi (1764.5 keV) and ^{208}Tl (2614.5 keV).

The efficiency calibrations measurements of the system were done at radiation reference pads of the Nuclear Energy Corporation of South African (NECSA) according the method describe by Chiozzi et al. (2000) and Corner et al. (1979). The facility comprises of 11 surface standard sources, each consisting of a solid circular pad with a 2 m diameter. Calibration spectra were acquired by positioning the detector 20 cm above the standard and then recording for a time period of 10 min. The detector was exposed to three standards that contained 4400 ppm uranium oxide (U_3O_8), 14,200 ppm thorium oxide (ThO_2) and 12.2% potassium oxide K_2O, respectively. These calibration spectra were used to extract efficiency parameters for each of the nuclides according to the method described in Paragraph 2.3. The nuclide concentrations were consequently expressed in Becquerel per kilogramme.

The symmetry assumptions and corrections described by McCay et al. (2014) were adopted for the calibrations and surveys. Ground surface unevenness was neglected and it was hence assumed that the detector was always at constant height and perpendicular to the ground, during field measurements. The radiometric influence of the quad motorcycle was also neglected.

2.3. Measurements and analyses

The GISPI acquired, analysed and stored *in situ* spectra at intervals of 20 s during the field measurements. A typical *in situ* spectrum that was acquired in the refinement area of the mine is illustrated in Fig. 2. The different Regions of Interest (ROIs) that are associated with ^{40}K, ^{226}Ra and ^{232}Th are also marked.

The ^{40}K nuclide emits a single gamma ray with an energy of 1460.8 keV and solely this decay is utilized to obtain the relative concentrations of natural potassium in an area. The nuclide ^{40}K has an abundance of 0.0117% of naturally occurring potassium (http://www.ciaaw.org/isotopic-abundances.htm) but the ^{40}K emission is commonly very strong in natural spectra, due to the abundance of naturally occurring potassium in nature. An active stabilisation function in the winTMCA32 software was used to correct for energy drift, which results from temperature changes while surveys were conducted on the site. The 1460.8 keV emission of ^{40}K was chosen as centroid for stabilisation and the fine gain were automatically adjusted by the winTMCA32 software to correct for any drift from the centroid.

The following procedure was followed for efficiency calibration. Three counting ROIs that were adapted were 1460.8 keV, 1764.5 keV and 2614.5 keV and are denoted by i equal to 1, 2, and 3, respectively. The calibration standards of potassium, uranium and thorium are indicated by j equal to 1, 2 and 3, respectively. The net count rate $R_{i,j}$ in the ith ROI of a calibration standard j is then proportional to the activity $A_{n,j}$ of each investigated nuclide n. If $n = 1, 2$ and 3 denoted the ^{40}K, ^{226}Ra and ^{232}Th nuclides, the net count rate can be expressed as

$$R_{i,j} = \sum_{n=1}^{3} e_{i,n} A_{n,j} \quad (1)$$

where $e_{i,n}$ give the counting efficiency in the ith ROI for the nuclide