Radon escape from mine tailings dams

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Abstract.
Gold mine tailings dumps contain considerable amounts of ^{226}Ra and have therefore been identified as large sources of radon. Gamma ray measurements at the field (Kloof mine dump) were done using MEDUSA while HPGe was used for laboratory based measurements. Field activity concentrations were analyzed using the Full Spectrum Analysis. Thirty four samples were collected from the field, analyzed and were found to contain an average of 308 ±12 Bq/Kg, 255±12 Bq/Kg and 18 ±1 Bq/Kg for ^{238}U, ^{40}K and ^{232}Th respectively. The link between MEDUSA and HPGE was established through normalisation factors and after normalising the activity concentrations and correcting for moisture content the normalised radon flux was computed to be 0.105±0.015 Bq m^{-2} s^{-1}.

1. Introduction

In Gauteng province, South Africa where mining activities take place, gold is extracted from rocks which are crushed and processed. After extraction, the waste is disposed in tailing dumps. The tailings usually contain naturally radioactive elements mainly uranium (^{238}U), potassium (^{40}K) and thorium (^{232}Th). ^{238}U decays into several products (see Figure 1) and among them is a radioactive gas known as radon (^{222}Rn).

Radon further decays to form short lived radioactive particles which enter the body through inhalation and can adhere to the lining of the lung. The deposited particles decay emitting alpha radiation which has a potential to damage cells in the lung [1]. Therefore radon exhalation from these tailing dumps poses a health concern.

The discussion that follows highlights a novel technique [7] for mapping radon exhalation from Kloof mine dump near Carletonville by using MEDUSA (**M**ulti **E**lement **D**etector for **U**nderwater **S**ediment **A**ctivity) γ-ray detection system [4] at the field in conjunction with a laboratory based hyper-pure germanium (HPGe) detector. The dump is approximately 2 square kilometres and it has been inactive for over seven years.

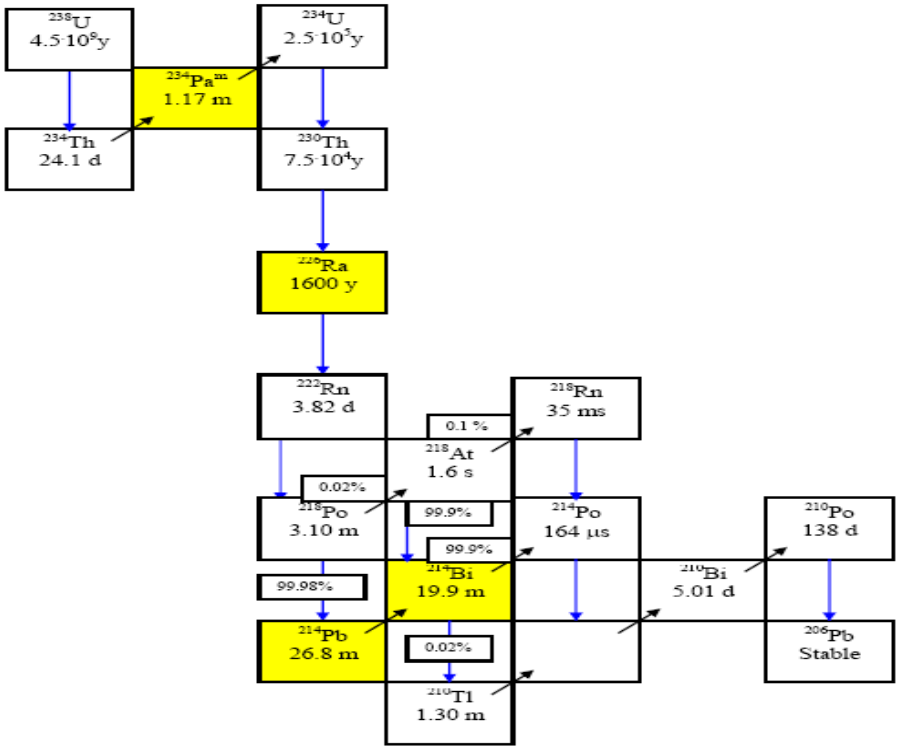

Figure 1. A schematic illustration of the uranium decay series. The half-life of each radionuclide in the series is indicated in years (y), days (d), minutes (m) and seconds (s). The yellow box represents the γ-ray emitters. The vertical arrow (blue) indicate α-decay while upward, slanted arrows (black) indicate β-decay.

2. Radon exhalation

Radon generated from the decay of radium in mineral grains migrates and is released to the atmosphere. Radon exhalation studies have been conducted and among the methods that have been used include: flow through method, the can technique and the accumulator [6]:

Unlike the mentioned techniques above MEDUSA technology which is based on gamma ray spectrometry will be used to map radon exhalation from Kloof mine dump. This technology was developed in the Netherlands by the Nuclear Geophysics Division of Groningen.

MEDUSA system incorporates a gamma-ray detector (CsI (Na)), ALADIN box, MPA (MEDUSA Post Analysis) software tool and global positioning system (GPS). The CsI (Na) crystal with 70 *mm* diameter and 150 *mm* length was integrated into the system mainly because of its stability in light output at variable temperatures. The ALADIN box contains data acquisition system and MPA software tool for gamma-ray spectra analysis. The gamma-ray intensities are interfaced on the surveyed area from the GPS.

Previously, the system has been used to map sand/mud ratios in rivers and dispersion of dredge spoil dumped on the sea floor [3; 9]. Furthermore, the system has been used in doing airborne surveys at iThemba LABS [5] and also at the mine dam [7].

The detector system was mounted 0.55 *m* off the ground on the front of a 4× 4 vehicle and accessible parts of the dam were transversed at 2 *m/s* (~7.2 *km/h*) with the spectra recorded every 2 *s* as also the GPS location logged-in for the given data.

A map with radiometric data was used to select locations where 30-60 *minutes* MEDUSA stationary measurements were done and thereafter five samples were collected that is one sample just

beneath the detector and four more samples roughly 80 *cm* from the detector at North, South, East and West to be analyzed using HPGe detector.
The acquired MEDUSA spectra were analyzed using full spectrum analysis [4] to extract radiometric information such as total counts (Figure 2a) and then activity concentrations of ^{238}U, ^{232}Th and ^{40}K (Figure 3a). These maps indicate accessible parts of the dam which are in colour while the inaccessible parts are in grey and beside them are their corresponding interpolated maps using Golden Software Surfer 8.

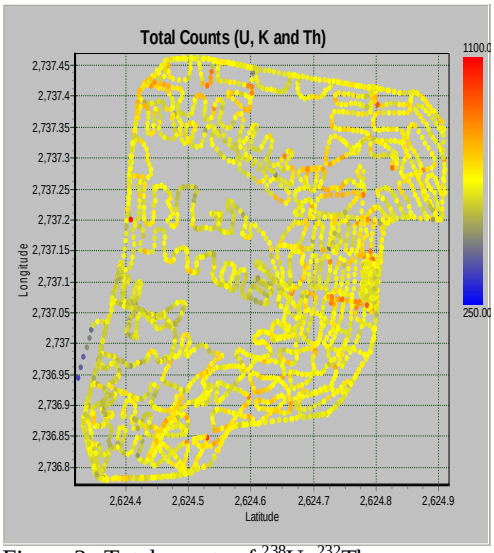

Figure 2a Total counts of ^{238}U, ^{232}Th and ^{40}K. Inaccessible parts on the map indicated by grey

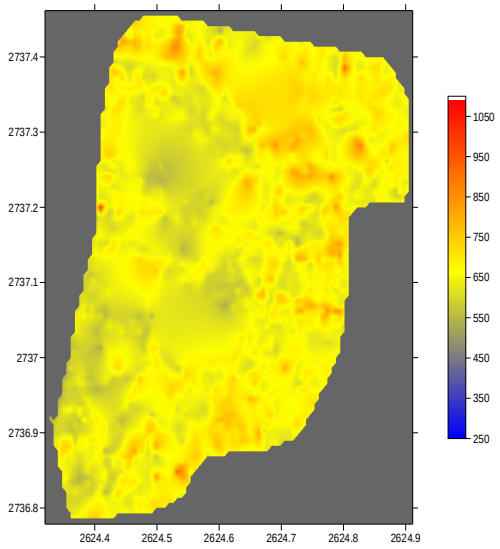

Figure 2b. Interpolated total counts of ^{238}U, ^{232}Th and ^{40}K using Golden Software Surfer 8

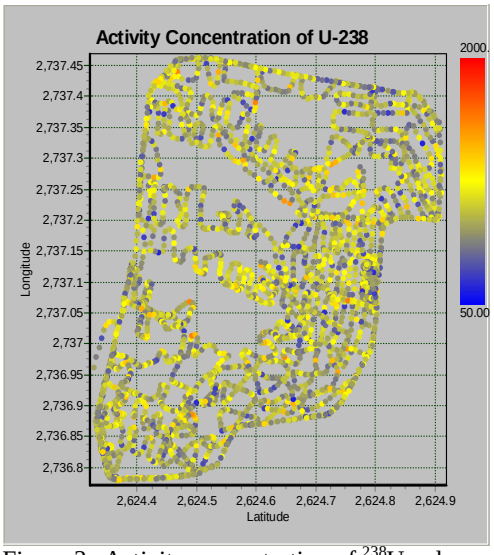

Figure 3a Activity concentration of ^{238}U only after Full Spectrum Analysis

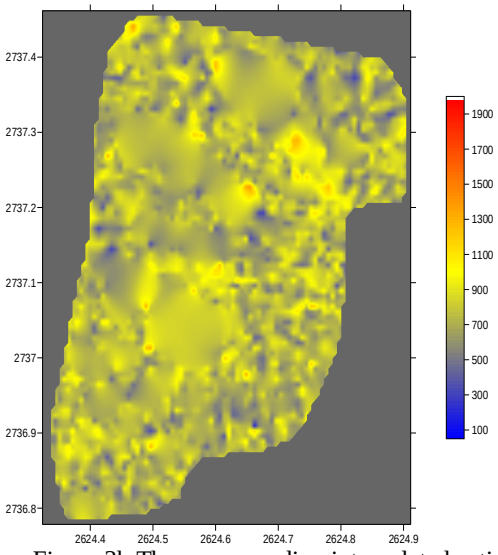

Figure 3b The corresponding interpolated activity concentration of ^{238}U only using Golden Software Surfer 8

Thirty four samples collected from the mine dump were further analyzed using laboratory based HPGe detector. This detector is a Canberra GC4520, p-type detector with 45 % relative efficiency and 2 keV FWHM resolution at 1.33 MeV [8]. The detector is encased in a 10 cm thick lead castle fitted with a 2.0 mm thick copper inner lining in order to reduce the background in the sample spectra. Standard nuclear electronics are used to process the detector signals. The pulses from the amplifier are collected and sorted by the ATOMKI Palmtop software multi-channel analyzer installed on a desktop PC. The HPGe detector system is energy and efficiency calibrated regularly with certified ^{232}Th, ^{238}U, ^{40}K ore (International Atomic Energy Agency/RGTh-1, RGU-1, RGK-1, respectively) and liquid cocktail reference source (^{60}Co, ^{137}Cs and ^{152}Eu) from National Metrology Institute of South Africa.

The soil samples were dried, sieved, sealed in Marinelli beakers and then stored for three weeks before counting. Radionuclides present in soil samples were identified and activity concentration (*A*) calculated using the following expression;

$$A(Bq/kg) = \frac{counts}{Br \cdot \varepsilon \cdot t \cdot m} \quad , \tag{1}$$

where *counts* represent the full-energy peak content corrected for continuum contribution, *Br* is the branching ratio (that is the percentage of the decay of the nuclide that will proceed via the emission of a particular gamma-ray), ε the detection efficiency, *t* the live time (in s) for spectrum acquisition and *m* (in *kg*) the sample mass. The counts for a given full-energy peak from the spectrum is determined by manually setting a region of interest (ROI) around it. The ATOMKI Palmtop MCA software uses an algorithm to calculate the net area (counts) associated with the ROI.

The average activity concentrations for the thirty four samples were determined to be: 308 ±12 Bq/Kg, 255±12 Bq/Kg and 18 ±1 Bq/Kg for ^{238}U, ^{40}K and ^{232}Th respectively.

In order to work out radon exhalation from the tailing dump a link between field based MEDUSA and laboratory based HPGe was established. The normalisation factor between MEDUSA data and HPGe was calculated using this expression:

$$Normalisation\_Factor = \frac{HPGe\_Concentration}{Medusa\_Concentration} \tag{2}$$

MEDUSA concentration in equation 2 refers to the concentration extracted from measurements done while the detector was stationary for a period of 30-60 *minutes*, whereas the HPGe concentration refers to the average concentration of the five samples picked beneath and around MEDUSA detector after the measurement was completed. These stationary measurements were done at four different spots spanning the mine dam and the average normalisation factors were 0.54±0.05, 0.36±0.03 and 0.21±0.01 for ^{238}U, ^{40}K and ^{232}Th respectively.

Further, normalised activity concentrations of ^{238}U, ^{40}K and ^{232}Th were obtained before they were corrected for moisture content. Moisture correction factors were obtained using the following expressions [2]:

$$C_{wet} = (1 - w_a)C_{dry} \tag{3}$$

$$C_{wet} = (1 - (1 + \alpha)w_a)C_{dry} \tag{4}$$

where C_{wet} is the wet activity concentration, C_{dry} is the dry activity concentration; w_a is the water content and α is a radionuclide specific constant that can be determined from the slope.

Since ^{40}K and ^{232}Th activity concentrations are not or hardly affected by radon exhalation, then it means that their concentrations will be independent of the radon loss and therefore the ratio of ^{238}U and ^{40}K and /or ^{232}Th is a measure of the radon exhalation at a particular location which is determined using the following expressions [7].

$$F = 1 - \left\{ (U_{dis} / K) / U_{eq} / K \right\} \quad (5)$$

$$F = 1 - \left\{ (U_{dis} / Th) / (U_{eq} / Th) \right\} \quad (6)$$

where F is the fraction of radon that escapes, U_{dis} is the ^{238}U activity concentration from the field (MEDUSA), U_{eq} is the activity concentration of ^{238}U from the laboratory (HPGe), K is potassium and Th is thorium.

Finally the flux obtained by equation 5 and 6 is normalised and the average flux obtained for Kloof mine dump was 0.105±0.015 Bq m^{-2} s^{-1}. Figure 4 shows a normalised flux which was computed and for parts which were inaccessible interpolation was done using Golden Software Surfer 8. The computed range of flux was 0.02-0.26 Bq m^{-2} s^{-1}.

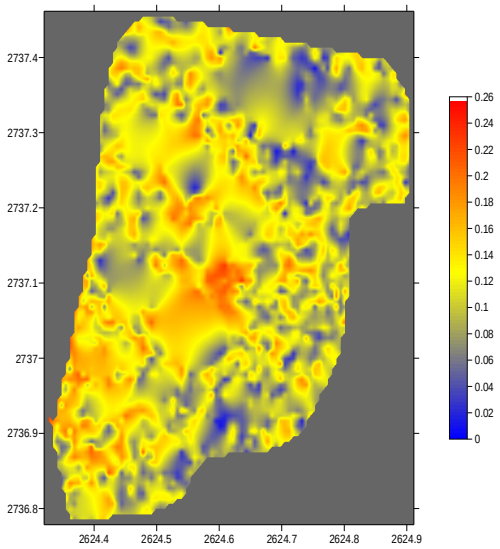

Figure 4 Map of interpolated flux for Kloof mine dump

3. Conclusion

A gamma ray technique namely MEDUSA technology has been used to determine the radon exhalation from Kloof mine dump. The analysis demonstrates that other than knowing the radium content in a particular point another parameter namely moisture content is also important in determining radon flux from an area. This is method practically provides a quick and accurate way to determine radon exhalation from an area at a relatively short period of time.

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