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RADON GENERATION AND TRANSPORT IN AND AROUND A GOLD-MINE TAILINGS DAM IN SOUTH AFRICA

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Abstract

Naturally Occurring Radioactive Material (NORM) occurs in most soil and rock, and by mining and mineral processing, some of the radionuclides are significantly enhanced. An in-situ gamma-ray detector called MEDUSA, has been used to produce a map of relative activity concentrations in a gold mine tailings dam on the Witwatersrand in South Africa. A CsI(Na) scintillation detector is used in this system. MEDUSA spectra obtained from the survey were analyzed using the Full-Spectrum Analysis (FSA) procedure to compute the ^{40}K, ^{238}U and ^{232}Th activity concentrations. The activity concentrations are used with global positioning data (GPS) to produce the concentration maps. A hyper-pure germanium gamma-ray detector (HPGe) was used to measure γ-rays from the naturally occurring nuclides for soil samples taken at different points on the site to calibrate the MEDUSA system. Radon soil gas measurements were performed at certain points on the mine tailings with a continuous radon monitor; RAD7, and emanation coefficients were measured with electret technology. These parameters have been combined with the activity concentrations to obtain an average radon exhalation rate of about 0.1 $Bq.m^{-2}.s^{-1}$ (with an uncertainty of about 20%) from the tailings dam. The purpose of the study is to also review and develop a mathematical model for radon activity concentration predictions in gold mine dumps.

Key words: ^{222}Rn flux/ mine dumps/ emanation/ RAD7™/ γ spectroscopy.

1. Introduction

The mine dumps around the gold mines in South Africa are a potentially hazardous source of radon, since the soil in the dumps contains radium activity concentrations of typically 200-300 $Bq.kg^{-1}$. A study of the diffusion of the radon will give us a better understanding of its possible pathways through the soil into the air surrounding the mine dumps [1]. The measurement and modelling of the radon concentrations in disturbed and

undisturbed soil can help to deduce the radon flux to identify the problem areas around gold and uranium mine tailings. Rehabilitation in those cases usually consists of a multilayer cover of solids like crushed rock or clay.

Many techniques for measuring radon releases from soil have been developed. This investigation describes the modelling of a depth profile with respect to the radon activity concentration to understand from what depth radon might be migrating to the surface as well as the effect of different radon diffusion lengths on the depth profile. The study was performed on a gold mine dump in the province of Gauteng in South Africa. Soil samples were taken from the dump and analysed in the laboratory at the University of the Western Cape and at iThemba LABS.

2. Radon emanation

Radon is the direct result when a ^{226}Ra atom decays in the ^{238}U decay series. The radon emanation coefficient is the fraction of the produced radon that enters the pore space. The radium atoms in soils tend to be concentrated near the pore boundaries, and when a radium atom decays, the resulting radon atom could recoil into the pore space [2]. However if the recoil range is small, like in most cases, the recoiling radon atom can be stuck in the host grain as represented in Fig. 1. The pore space in between grains might contain water, and if the radon atom terminates its recoil in the water, the radon might be transported more easily to the air in the pore space where it can be transferred to the air by rapid mass transfer. Nazaroff et al. describe three mechanisms, which influence the emanation coefficient [3]. The possibilities are; direct recoil, indirect recoil and diffusion. Figure 1 represents the different scenarios possible through the emanation process from a soil grain matrix.

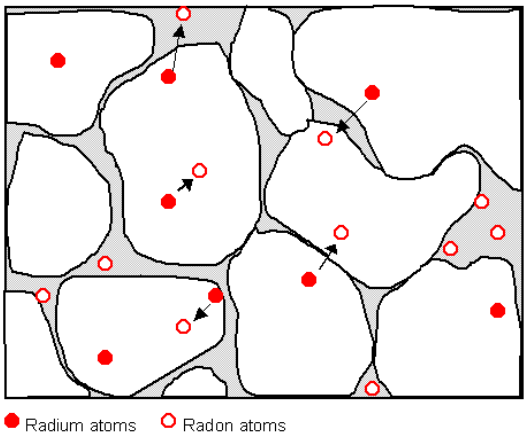

Figure 1. Representation of an enlarged section of pore space with soil grains (larger white areas). It also depicts the radium and radon atoms that might be embedded in the grains. The arrows represent radium atoms that decay to radon. The alpha particle from the decay would be propelled in the opposite direction. The representation of the atoms greatly exaggerates the atomic dimensions (modified after Tanner, 1980 [4]).

3. Radon modelling

Numerical modelling of ^{222}Rn is complex and involves a few important physical processes. Mine dumps are considered to be sources of radon since they largely contain the predecessor of radon; ^{226}Ra. The medium consisting of the processed tailings, in some cases protrude about 60 m high from the ground and covering an area of about 2 km^2; which in essence could be viewed even as a large volume with a radon release problem. However, it is not so straightforward for the radon to escape to the ambient air since it depends on factors like (1) emanation from the soil grains, (2) generation of radon itself, (3) transport of the radon through the mine tailings and (4) the pressure variations in the soil and ambient air.

3.1 One-dimensional diffusion equation

There are two main transport processes that dictate radon release from soil. The two processes are diffusion and advection. Advection will not be considered in this study, since its role depends on small pressure changes that tend to average out. The diffusion equation [5] is given by

$$\frac{\partial C}{\partial t} = D\ \frac{\partial^2 C}{\partial z^2} - \lambda C + S \qquad (1)$$

where C is the activity concentration of radon, λ is the radon decay constant, D is the diffusion coefficient and S is the creation rate of radon. On the right hand side of the equation the first term relates to the change of radon atoms due to diffusion while the second and third terms relate to the decay of radon atoms and the creation of radon respectively. The boundary conditions are: $C \rightarrow C_0$ as $z \rightarrow 0$ at the surface where the z-axis is chosen vertically down and $C \rightarrow C_{Max}$ as $z \rightarrow \infty$. The radon concentration at the surface (C_0) will be close to zero. Assume steady state diffusion as well as no pressure changes. The solution to the above equation is then

$$C(z) = \frac{S}{\lambda} + C_1 e^{\sqrt{\frac{\lambda}{D}}z} + C_2 e^{-\sqrt{\frac{\lambda}{D}}z}. \qquad (2)$$

Since C_1 will be zero due to the 2nd boundary condition, we get $C_2 = C_0 - C_{Max}$ from the first boundary condition where $C_{Max} = S/\lambda$ also follows from the 2nd condition. This leads to

$$C(z) = C_0 e^{-\frac{z}{l}} + C_{Max}(1 - e^{-\frac{z}{l}}) \qquad (3)$$

where $l = \sqrt{\frac{D}{\lambda}}$ is the diffusion length. The effect of different l-values on the radon concentration will be discussed below.

3.2 Radon and soil-gas transport

A numerical modelling tool; RnMod3d was developed by Andersen [6] to model radon and soil-gas entry into houses. A number of soil specific parameters are needed, e.g. water- (ε_w), air-(ε_a), total porosity (ε) of the soil, dry-soil density (ρ_{ds}), volumetric water content and the amount of water per dry mass of soil for RnMod3d to solve the specific transport equation for a given medium. The radon transport equation for a given reference element δV of soil is represented by

$$\frac{\partial \beta c_a}{\partial t} = \varepsilon S - \lambda \beta c_a - \vec{\nabla} \cdot \vec{j}$$

(4)

where S is the creation rate of radon per unit bulk volume, measured in Bq/m^{3}s [7], ε *is the total porosity of the soil, combining the volume of the air and water in* δV*,* β *is the so-called partition-corrected porosity [7] [8].*

$$\beta = \varepsilon_a + L\varepsilon_w + K\rho_{ds}$$

(5)

where L and K is the Ostwald partitioning (measure of radon solubility in water with respect to temperature) and radon surface sorption coefficient respectively, $\vec{j}$ *is the bulk flux density (in units of Bq/m^{2}s) at time t. The first term on the right hand side of eq. (4) includes the radon generation, then the process of radon decay, followed by the diffusion of radon. The concentration of radon is given by* c_a*. The bulk flux density has 2 components; the advective flux and diffusive flux that give*

$$\vec{j} = \vec{j}_a + \vec{j}_d$$

(6)

where

$$\vec{j}_a = c_a \vec{q}$$

(7)

with $\vec{q}$ *representing the bulk flux density of the soil-gas (m^{3}s^{-1} for every m^2). The diffusive flux* $\vec{j}_d$ *is written as*

$$\vec{j}_d = D\vec{\nabla} c_a .$$

(8)

The solution of eq. (4) is the most important factor for this exercise.

4. Experimental methods

4.1 Radon soil-gas measurements

The radon soil-gas in the mine dump was measured using a Durridge RAD7™ continuous radon monitor. Figure 2 shows a schematic of the setup of the RAD7 on the mine dump. The RAD7 pumped the soil-gas for 5 minutes into the cell of the detector, and then waited for 5 minutes, then counted for another 5 minutes. In total, each set of readings included four 5-minute cycles that took 1 half hour.

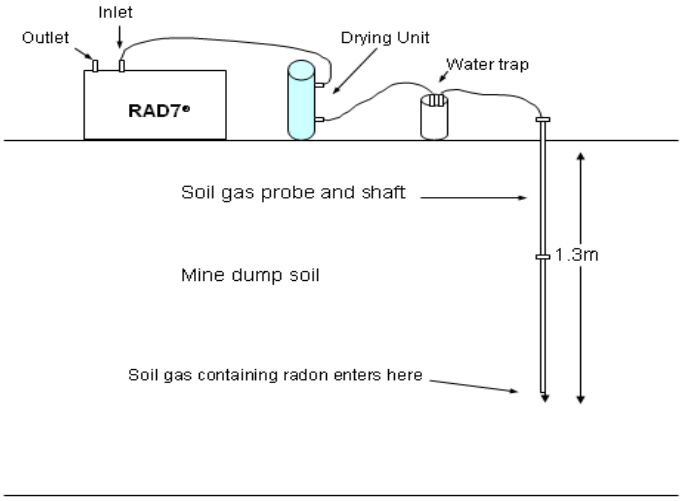

Figure 2. Schematic of the RAD7 soil-gas setup as seen from the side of the mine dump.

The length of the probe only allowed for depths up to 1.25 m. Table 1 shows the results from 2 different sites chosen on the mine dump, designated M1 and M2 located at 26° 24.797' South Latitude, 27° 37.111' East Longitude and 26° 24.802' South Latitude, 27° 37.404' East Longitude respectively.

4.2 Radon emanation and gamma-ray spectrometry with HPGe

The E-PERM (Electret Passive Environmental Radon Monitor) detectors were used to measure the radon emanation (η) of the mine dump soil [9]. The E-PERM system is based on electret ion chamber (EIC) technology that consists of an ion chamber made of an electrically conductive plastic housing an electrically charged Teflon® disk called an electret that is screwed inside the above-mentioned chamber. The EICs were used to measure the Rn concentration that accumulated in a container as a result of the Rn emanating from a thin layer of soil [10] that was placed at the bottom of a sealed accumulator jar. The radon emanation was then calculated using the radium content of the soil samples that was measured using an HPGe detector.

The HPGe detector has a relative efficiency of 45% at 1.33 MeV and a full-width at half maximum of 2.2 keV. It has dimensions 62.5 mm and 59.9 mm for the crystal diameter and length respectively. It is mounted in a lead castle (10 cm thick) with the detector facing upwards holding a sealed Marinelli beaker containing the sample. The soil samples gathered from the mine dump were analysed after a 21-day period after being sealed to establish secular equilibrium and to prevent radon from escaping [11]. The activity concentrations of the natural radionuclides ^{238}U and ^{232}Th are measured indirectly by using the ^{214}Pb and ^{214}Bi γ-rays (found in the ^{238}U decay series) for the ^{238}U and the γ-rays from ^{228}Ac and ^{208}Tl (found in the ^{232}Th decay series) for the ^{232}Th determination.

4.3 MEDUSA detector

The MEDUSA (Multi-Element Detector System for Underwater Sediment Activity) detector as the name suggests was developed to asses the activity concentrations of the natural radionuclides contained in the sediments and hence monitoring the dispersal of dredge spoil over a given surface [11]. The core component of the detector consists of a CsI(Na) crystal with dimensions 15cm in length and 7cm in diameter which is fitted in an aluminum casing together with all the electronics and sensors. Customised software (loaded onto a portable computer preferably) for optimal performance with the MEDUSA detector was also developed and a high precision GPS device for accurate positional data to complete the acquisition system [12].

The detector is fitted (Fig. 3) onto the front end of a 4×4 vehicle (about 60 cm above the ground) that will be traversing (at about 2 $m.s^{-1}$) the surface of the tailings dam together with the GPS receiver to record a spectrum every few seconds (2 s) for later interpretation into maps of activity concentrations [13]. The gamma-ray energies that are recorded are in the range of 0-3 MeV. A 20-minute measurement was made with the MEDUSA detector at a fixed spot for calibration purposes. A soil sample was then collected from exactly underneath the detector and sealed in Marinelli beakers and analysed at the Environmental Radioactivity Laboratory (ERL) at iThemba LABS by means of the HPGe detector.

Figure 3. The MEDUSA detector fitted on a 4×4 vehicle for gamma-ray measurements on the gold-mine tailings dam.

5. Results and discussion

5.1 One-dimensional diffusion equation

The one-dimensional diffusion equation was solved (within certain boundary conditions) in section 3.1 and eq. (3) yields the results plotted in Fig. 4. The radon concentrations are presented as a function of depth for different l (diffusion lengths) values. These predictions assume that only diffusion is considered to have an effect on the radon soil-gas. It is clear from the graph that a very short diffusion length (l) would cause a very

rapid increase in the radon soil-gas with increasing depth. One limitation with this analytical model is that it does not explain the radon concentrations at depths closer to the surface of the tailings. One of the possible reasons is that at these depths, pressure changes on the surface of the mine dump could influence the radon soil-gas close to the surface. The model predicts radon concentrations that tend to the maximum radon concentration assumed in the model round about 500kBq.m^{-3} as indicated in Fig. 4.

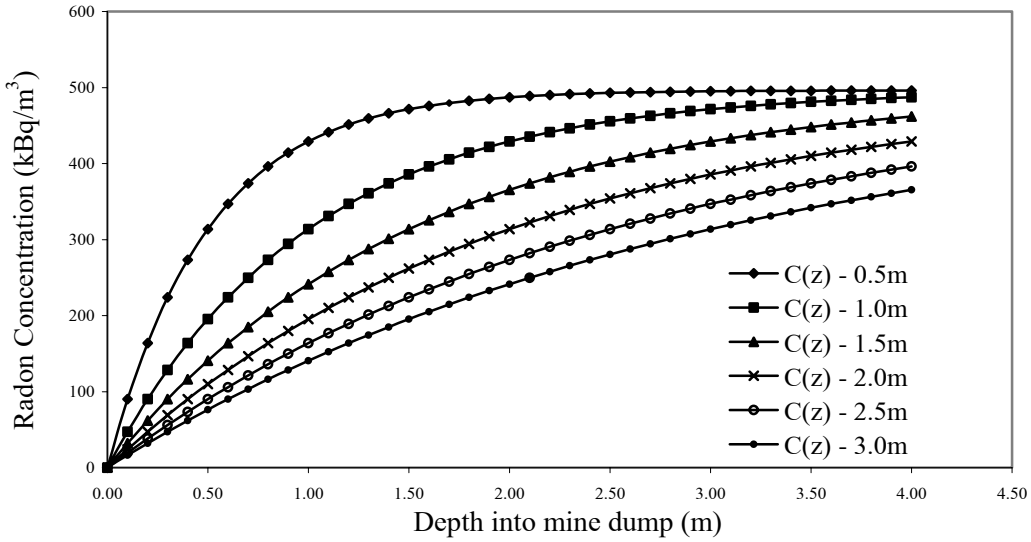

Figure 4. Radon depth profile generated with a one-dimensional analytical model for the prediction of radon activity concentrations in the mine dump. The calculated curves vary considerably with varying diffusion lengths (lengths indicated on plot).

5.2 Radon emanation coefficient

The radon emanation coefficients for the different soil samples are listed in Table 2. Radon emanation is a very important influence on the radon flux from a gold mine tailings dam [13]. In order to determine the radon emanation coefficient, two parameters have to be measured. The first parameter gives a measure of the radon emanating from ^{226}Ra (RnERaC) of the soil. The second parameter includes the approximate determination of the radium activity concentration (A_{Ra}). The RnERaC method [10] incorporates the E-PERM technology whereas A_{Ra} is determined using an HPGe γ-ray detector. The ratio of the RnERaC and A_{Ra} gives the radon emanation coefficient (η),

$$\eta = \frac{RnERaC}{A_{Ra}} .$$

(9)

The soil samples collected at different depths (Table 2) show appreciable differences in emanation coefficient. The increase in the emanation coefficient with depth suggests that more radon is being released deeper in the dump. This is a surprising result, since the mine dump is assumed to be fairly uniform, but this may be due to the possible compaction of the soil as well as possible radium leaching through the dump. The data in the rightmost column show consistent results for soil-gas

measurements at the two sites for the samples collected close to the surface, independent of the preparation method. SST and LST refer to the specific EICs that were used together with the electret type. SST refers to an S-chamber together with a ST-electret (short-term) and LST refers to an L-chamber used with a ST-electret. The S-chamber was used for short-term (≈ 7 days) measurements whereas the L-chamber was used for long-term (≈1month) measurements.

Table 1. Observed radon concentrations at different depths measured with the RAD7™ continuous radon monitor.

M1			M2		
Depth (cm)	Mean Rn Conc.(kBq/m^3)	Std. Dev. (kBq/m^3)	Depth (cm)	Mean Rn Conc.(kBq/m^3)	Std. Dev. (kBq/m^3)
25	8.1	0.8	25	11.6	0.5
50	129.1	4.2	50	49.7	1.3
75	277.6	5.7	75	163.1	2.9
100	350.0	6.3	100	226.5	7.1
120	405.8	13.8	120	271.4	13.9

Table 2. Summary of the radon emanation coefficients measured for mine dump soil samples obtained at 2 different locations and at different depths.

Different Depth Sampling point				M1 and M2 Locations			
Site Location	Preference	SST/LST	Emanation Coefficient ± SD	Site Location	Preference	SST/LST	Emanation Coefficient ± SD
SS5cm	Room-dried	SST	0.17 ± 0.01	M1	Oven-dried	SST	0.23 ± 0.03
SS5cm	20-30% moist	SST	0.17 ± 0.02	M1C	Oven-dried	SST	0.27 ± 0.03
SS5cm	Room-dried	SST	0.13 ± 0.01	M1	Room-dried	SST	0.25 ± 0.02
SS5cm	Room-dried	LST	0.15 ± 0.02	M1C	Room-dried	SST	0.24 ± 0.02
SS5cm	Room-dried	LST	0.16 ± 0.02				
				M2	Oven-dried	SST	0.27 ± 0.03
SS50cm	Room-dried	SST	0.23 ± 0.02	M2C	Oven-dried	SST	0.23 ± 0.03
SS50cm	20-30% moist	SST	0.25 ± 0.02	M2	Room-dried	SST	0.26 ± 0.02
SS50cm	Room-dried	SST	0.19 ± 0.02	M2C	Room-dried	SST	0.21 ± 0.02
SS50cm	Room-dried	LST	0.21 ± 0.02				
SS50cm	Room-dried	LST	0.20 ± 0.02				
SS100cm	Room-dried	SST	0.35 ± 0.02				

SS100cm	Room-dried	SST	0.34 ± 0.03
SS100cm	Room-dried	LST	0.34 ± 0.03
SS100cm	Room-dried	LST	0.39 ± 0.03

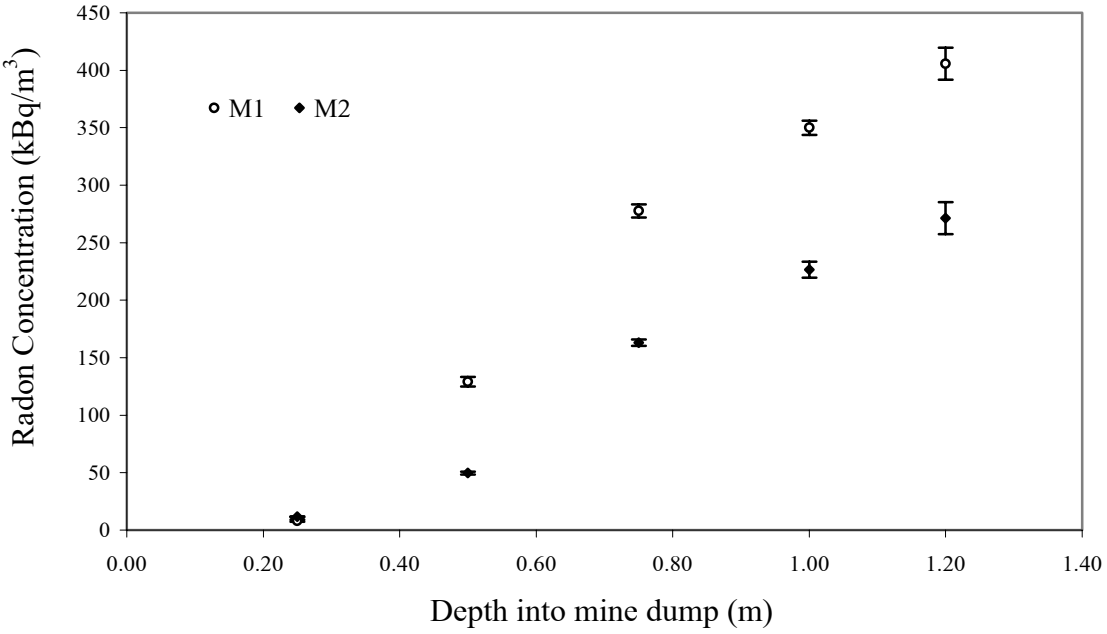

Figure 5. Measured radon depth profile for 2 different locations on the mine dump.

Table 2 and Fig. 5 show the results with standard deviations obtained from the radon soil-gas measurements on the mine dump. The measurement at location M1 shows a much more rapid increase with depth in the radon soil-gas. One of the possible reasons for this could be that the radon diffusion length is different at that location. Another possible explanation could involve the radon diffusion coefficient (D) which would suggest that the radon emanation at M1 is different from that at M2. The actual depth profile in Fig. 5 differs from the predicted depth profile in Fig. 4 in the following sense. The values obtained at the lower depths appear to be too low, as mentioned before. A possible reason could be that the pressure on the surface influences the measurements at these lower depths and it for that reason why radon and soil-gas transport modelling will be investigated for application on typical uncovered mine tailings. The measurements at small depths may also suffer from a leakage of air down the side of the probe when taking measurements.

5.3 MEDUSA data processing and maps

The MEDUSA system records the total count rate from the in-situ measurements on the mine dump. The count rates (mapped in Fig. 6a) are indicative of the activity of the relevant radionuclides being measured. The spectra collected while driving in grid patterns over the surface of the mine dump were stored on a laptop computer for further analysis with the

accompanying software of the system. The method of Full Spectrum Analysis (FSA) was used to analyse the MEDUSA spectra [12] for the calculation of the activity concentrations of ^{40}K, ^{238}U and ^{232}Th. The FSA method takes into account the full energy spectrum. In this case, 3 standard spectra (1 for each radionuclide mentioned) and a background spectrum are fitted to the measured spectrum by means of least-squares minimisation. Separate calibration measurements led to the derivation of the standard spectra. The standard spectra are the response of the detector to a specific activity concentration (1 Bq/kg) for a given nuclide.

The relative activity concentrations of ^{40}K, ^{238}U and ^{232}Th were extracted from the FSA analysis and together with the GPS data, maps could be generated (Fig. 6). These maps could be accessed while capturing data of the top 30 cm of soil on the mine tailings. The absolute activity concentrations could then be calculated by making a correction for the off set of results (ratio) from the MEDUSA calibration and HPGe measurements in the laboratory. The ^{226}Ra activity concentrations are then calculated (from uranium distribution) from the maps for use to calculate the radon flux potential for that site.

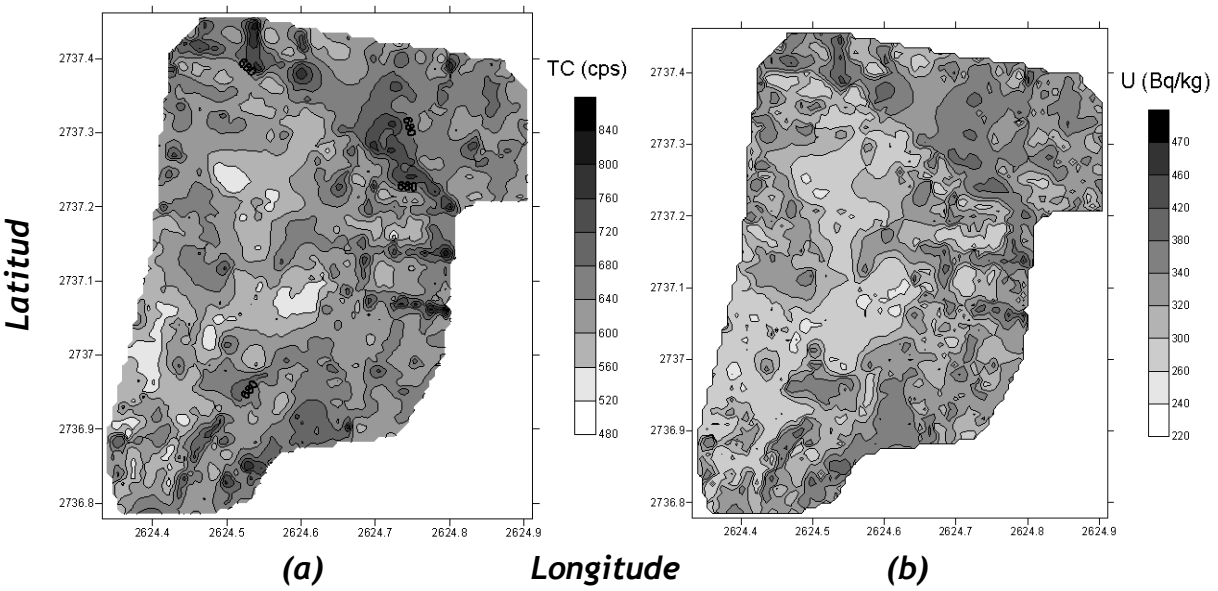

Figure 6. Maps of (a) the registered total counts (TC) in counts per second and (b) the derived ^{238}U absolute activity concentrations (Bq.kg^{-1}) from MEDUSA measurements on the gold mine dump which was interpolated using the Surfer8 software package.

The exhalation rate J of radon was calculated using the following relation [3] [14]

$$J = \sqrt{D\lambda}\rho_b \eta A_{Ra} = \lambda l \rho_b \eta A_{Ra}$$

(10)

where D (~2 $\times$ 10^{-6} m^{2}s^{-1}) [3] is the diffusion coefficient and measurements on the mine tailings soil gave a total soil porosity ε of 0.35, a soil density ρ_b of 1867 kg.m^{-3}, λ is 2.1 $\times$ 10^{-6} s^{-1}, A_{Ra} (radium activity concentration) was

found to be 310 Bq.kg^{-1}, η (radon emanation coefficient) was determined to be 0.12 and l (diffusion length) an assumed value of 1m. The radon flux was then calculated using eq.(10) and a value of 0.1 Bq.m^{-2}.s^{-1} with an uncertainty of about 20% was recorded. This value represented the average radon exhalation rate on this specific mine tailings.

6. Conclusion

Estimating the radon potential of a typical gold mine tailings can be achieved through a number of methods. This investigation suggests the use of a γ-ray detection system (MEDUSA) that yields fast and effective mapping of tailings dam. The relative activity concentrations of either ^{40}K, ^{238}U or ^{232}Th could easily be accessed while capturing data, making assessments and recommendations for sampling points, etc. The absolute activity concentrations could be obtained by making a correction for the slight off-set of results from the MEDUSA and HPGe calibration measurements. The sensitivity of the MEDUSA detector makes it ideal for measurement of NORM since clear differences could be distinguished from the produced maps and hence decreasing sampling and analysis times. The higher radium activity concentrations indicate a high potential of radon gas can be generated and the radon soil-gas measurements (Fig. 5) confirms this. The radon emanation results (Table 2) show that only about 13 - 40% escape the soil grain matrix and hence is made available for transport to the surface for release. However, not much is known about the radon and soil-gas transport of a typical mine dump and hence the interest in using a modelling tool like RnMod3d for understanding radon and soil-gas movement. This study shows that an in-situ gamma-ray mapping technique can be very advantageous in mapping large mine dump areas in a relative short time and it also produces maps of the activity concentrations by using the varying activities of the radionuclides across the dump together with the global positioning data.

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