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TECHNICAL ARTICLE

Exposure Risks Assessment Due to Gamma Emitting Radionuclides in Soils and Consumable Waters Around Princess Gold Mine Dump in Roodepoort, South Africa

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Abstract This study evaluated the activity concentration of ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K around the Princess Gold Mine dump in Roodepoort, South Africa using gamma spectrometry and ICP-MS. The soil contained a high concentration of ^{238}U at a depth of 1 m and at the surface. Doses due to ingestion of the untreated water were estimated as being far above the limit of 0.10 mSv/a set by the World Health Organization. The South African National Nuclear Regulator's reference value of 0.25 mSv/a was also exceeded by almost all of the analyzed water samples. Thus, the dose the public could receive from drinking this water poses a potential radiological health risk.

Keywords Bio-remediation · Radio-toxic · Environmental risks · Potential radiological health risk

Introduction

Mining in South Africa's gold-rich Witwatersrand Area has brought naturally occurring radioactive material (NORM) to the surface, negatively affecting water bodies, the people near the mine tailings, and ecological systems (Durand 2012; Winde 2010a; Winde and Jacobus van der Walt 2004). ^{238}U, ^{232}Th, and ^{40}K release radiation to the environment, posing a health hazard to humans (Borylo et al. 2012; Boukhenfouf and Boucenna 2011; Charro et al. 2013). Although natural radioactivity exists in the soils, varying from one type of soil to another (Boukhenfouf and Boucenna 2011), it has been significantly enhanced by mining. The ^{238}U and ^{232}Th progeny, both of which are radiotoxic, are the major contributors to the technologically enhanced radioactivity hazard (Arogunjo et al. 2009). The daughters from ^{238}U and ^{232}Th (^{226}Ra, ^{212}Pb, and ^{214}Pb) and the daughter of ^{226}Ra (^{222}Rn) are serious pollutants in confined facilities with poor ventilation. ^{212}Pb decays to its stable isotope, ^{208}Pb and ^{214}Pb decays to its stable isotope ^{206}Pb. These stable isotopes of lead are, depending on their concentration, potentially toxic (Bolhar et al. 2007; Borylo et al. 2012; Clulow et al. 1998; de Caritat et al. 2001).

Residents near the Princess Gold Mine Dump in Roodepoort, South Africa have complained to the South African government that the presence of the tailings poses a health risk. The Council for Geoscience was commissioned by the government to investigate and tendered the work to the Center for Applied Radiation Science and Technology (CARST). Before this, Ngigi (2009) had studied this gold mine dump, but they focused mainly on the water quality and acid mine drainage (AMD). A risk-based methodology was used to assess the health exposure based on a simple conceptual process that defines the contaminant linkage to the potential environmental inhabitant. Three essential factors were evaluated: (1) the source (soil/water) of the contaminant(s) that could potentially impact human health or the environment, (2) the way in which the contaminant is released and migrates to the environmental media, i.e. soil to plant, and (3) the inhabitants that could be adversely affected.

A risk only exists when the linkage is complete and the inhabitants are exposed to the contaminants. The risk in this study was assessed in quantitative terms and expressed in terms of probability. To accurately predict contaminant transport, it was essential that the geochemical processes

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affecting contaminant transport be identified and accurately measured. Major geochemical processes affecting transport include dissolution/precipitation, adsorption/desorption, chemical gradient diffusion, and diffusion into micro-pores. The distribution coefficient (K_d) measures the partitioning of a contaminant between the solid and aqueous phase (Kaplan et al. 1995).

This paper investigates the transfer of radionuclide contaminants from the Princess Dump to the Victory Park community in the Roodepoort area of South Africa. The exposure risks to the community associated with the exposure pathways due to ^{238}U (and daughters), ^{232}Th (and daughters), and ^{40}K were estimated. The geochemical composition of the tailings soil was also investigated for its concentration of potentially toxic elements.

Materials and Methods

Study Area

Roodepoort is one of the towns that has sprung up on the western side of Johannesburg. On the upper Klip River catchment (Gelb 1991; Ngigi 2009), there is a sizable community called Davidsonville less than a kilometer from the abandoned Princess Gold mine dump. Victory Park, a smaller community, lies just a few meters from the dump, in the corner created by the L-shape of the dump.

The tailings from this dump pose a radiological risk to the Victory Park community, especially those who pan gold from the tailings or who plant their vegetable gardens in the tailings soil. In addition, dust from the dump blows into the community and the tailings are a source of acidic, saline water, leached metals, and NORMs, which affects the soil, groundwater, and the entire watershed (Chevral et al. 2008; Mariette 2009). The geology of the area was described in detail by Njinga et al. (2015).

A total of 26 1 kg soil samples were collected at a depth of 1 m in plastic bags and sealed. Sediment soil cores were collected using an auger of 10 cm diameter and 15 cm length. The sampling points were marked using GIS and labeled A2 to Z2. Samples collected 15 cm from the surface were labelled A1 to Z1, but some of these are not presented here. Generally the dump soil was brown silt of fine grain size and soil samples were collected all around the mine tailings, about 100 m apart. On the sides of the tailings, samples were collected by drilling 1 m horizontally into the tailings to avoid contamination due to rain wash-down. These same soils are used by the as building materials and by some of the Victory Park dwellers for their gardens. A total of 14 vegetation samples were collected near (within 30 cm) to each soil sampling point to determine the transfer factors for ^{238}U and ^{232}Th. Three water samples were collected from the bottom of the tailings dump, where AMD starts to flow, and three more at the entry of the drainage pipe into the Klip River, where it is discharged. Core sample localities are shown in Fig. 1 (Njinga et al. 2015). All the soil samples were subdivided; one portion was analyzed by inductively coupled plasma mass spectrometry (ICP-MS; Perkin Elmer, NeXION 300Q) and the other with a high-purity germanium (HPGe) detector (Canberra Detector-35 190r).

At least six water samples were collected in 500 mL bottles and acidified with concentrated HNO_3 (10 mL/L) and kept refrigerated. Leaf samples collected about 1 m above the ground from each tree and closest to the soil GPS point

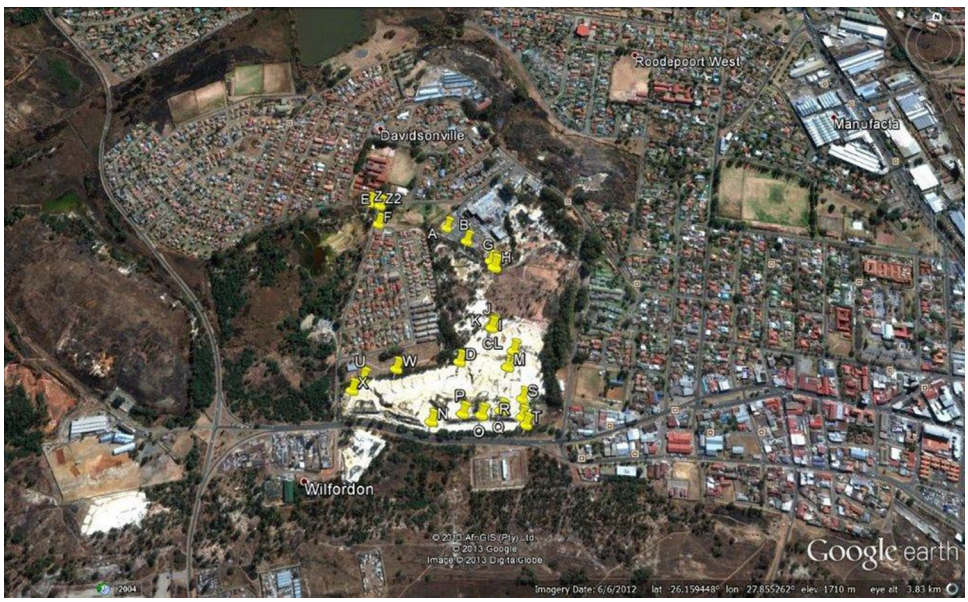

Fig. 1 Sampling locations near the L-shaped Princess dump

were sealed in perforated plastic bags and transferred to the laboratory for preparation and analysis. Table 1 gives a description of each sample ID and the type.

Soil Sample Preparation

Soil samples were digested using a Multiwave 3000, Anton Paar microwave oven. About 1 g of each sample was weighed into a microwave digester vial and then digested with *aqua regia* acid (3 mL of 55% HNO_3, 9 mL of 32% HCl, and 2 mL of 2% H_2O_2), following standard procedures. Then, the sample vials were transferred to a microwave reactor system for 20–45 min. An aliquot (10 mL) of each sample was transferred into a volumetric flask and topped up with ultrapure distilled water up to 100 mL. They were stored at 4 °C for another 24 h. The reagents used were of Suprapur analytical grade (Merck Laboratory Supplies Pvt., Ltd).

Water Sample Digestion Protocol

Each 5 mL of the water samples were mixed with 5 mL of HNO_3 and 1 mL of HCl and then left to digest in the microwave oven (Multiwave 3000, Anton Paar) for 24 h. A 10 mL aliquot of each digested sample was then transferred into volumetric flasks and topped up with Millipore Milli-Q system (resistivity 18 MΩ cm^{-1}) distilled water up to 100 mL and left overnight for ICP-MS analysis.

Table 1 Sample ID and description

Sample ID	Sample type	Comment	Sample ID	Sample type	Comment
A	Water	Water seeping from the tailings	O	Soil	Soil from tailings
B	Water	Water seeping from the tailings	P	Soil and vegetation	Soil samples from tailings, vegetation sample at 1 m from GPS point
C	Water	Water seeping from the tailings	Q	Soil	Soil samples from tailings
D	Water	Water seeping from the tailings	R	Soil	Soil from tailings
E	Water	Water seeping from the PIPE discharge	S	Soil	Soil from tailings
F	Water	Water seeping from the PIPE discharge	T	Soil and vegetation	Soil samples from tailings, vegetation sample at 1 m from GPS point
G	Soil and vegetation	Soil samples from tailings	U	Soil samples from tailings	Soil
H	Soil	Soil samples from tailings	V	Soil and vegetation	Soil samples from tailings, vegetation sample at 1 m from GPS point
I	Soil and vegetation	Soil samples from tailings	W	Soil samples from tailings	Soil samples from tailings, vegetation sample at 1 m from GPS point
J	Soil	Soil samples from tailings	X	Soil and vegetation	Soil samples from tailings, vegetation sample at 1 m from GPS point
K	Soil	Soil samples from tailings	V	Soil and vegetation	Soil samples from tailings, vegetation sample at 1 m from GPS point
L	Soil and vegetation	Soil samples from tailings, vegetation sample at 1 m from GPS point	W	Soil samples from tailings	Soil samples from tailings, vegetation sample at 1 m from GPS point
M	Soil and vegetation	Soil samples from tailings, vegetation sample at 1 m from GPS point	X	Soil samples from tailings	Soil samples from tailings, vegetation sample 1 m from GPS point
N	Soil and vegetation	Soil samples from tailings, vegetation sample at 1 m from GPS point	Z1	Stream water mixed with tailings discharge water	Stream water mixed with tailings discharge water
Zm	Stream water/ tailings discharge water	Stream water/ tailings discharge water	Z2	Stream water mixed with tailing discharge water	Stream water mixed with tailings discharge water

Digestion of Vegetation Samples

Prior to the processing of samples, crucibles were cleaned i.e. they were soaked overnight in analytical grade 12 M HCl and then rinsed with Millipore Milli-Q system (resistivity 18 MΩ cm^{-1}) distilled water. The porcelain crucibles (Sigma-Aldric) were then dried at 600 °C in an oven overnight and then placed in a desiccator for cooling for 6 h. 1 g of the each vegetable sample was weighed and the weight was recorded. The samples were dried in an oven furnace model (G132-002) for 16 h at 800 °C. They were then placed in a desiccator for 6 h. The dry weight was recorded after ashing and the samples were transferred into the rotor vessels. 1 mL of HNO_3 was added to the samples followed by 10 mL of HCl and then placed into the microwave reactor for 20–45 min. A 10 mL aliquot of the digested samples were transferred into a volumetric flask and topped up with water to 100 mL and left to sediment overnight. The supernatant was then transferred into centrifuge tubes for ICP-MS analysis. All digested sampled were preserved at 4 °C before analysis (Kamunda et al. 2016).

Gamma Analysis: Soil Samples

The soil samples were dried by evaporation in the laboratory in their original bags before about 250 g of soil was sealed in a plastic bottles. All of the sealed soils samples were stored for at least 25 days to allow the parent ^{226}Ra radionuclides to attain secular equilibrium with her daughters (Ahtar et al. 2005; Arogunjo et al. 2009; Church et al. 2008; Lawrie et al. 2000), before being analyzed with the HPGe Coaxial Detector (Baltic Scientific Instruments GCD-35190). The detector was placed inside a 10 cm thick lead shield and the samples were placed over it with a counting time set for 24 h. Spectrum acquisition and analysis was done using the Win SPEC and IDENTIFY software, respectively. The energy and efficiency calibration of the detector was done using calibration files supplied with the detector by the manufacturer. Energy and efficiency calibrations were checked daily to maintain the quality of the measurements.

^{238}U, ^{226}Ra, ^{232}Th, their progeny (^{212}Pb, ^{212}Bi, ^{214}Pb, ^{214}Bi), and ^{40}K were analyzed using IDENTIFY, which calculates the activity and error in Bq. For ^{226}Ra, the activity concentration was calculated as the weighted average of activity determined using the gamma line of 295.1 (19.2%) keV and 351.9 (37.1%) keV from ^{214}Pb, 609.3 (46.1%), keV, 1120.3 (15%) keV and 1764.5 (15.4%) 94 keV gamma line from ^{214}Bi; and 186.2 (3.59%) keV the specific gamma line of ^{226}Ra (possible interference from ^{235}U was checked and corrections made where applicable). The mean specific activity concentrations of ^{238}U, ^{232}Th, and ^{40}K (Bq kg^{-1}) in the soil samples were used to calculate the total air absorbed dose rate at 1 m above the ground (Abbady et al. 2005; Nursama et al. 2013; Turhan and Gundiz 2008). The annual effective dose equivalent can be estimated (Chang et al. 2008; IAEA 2003; Turhan and Gundiz 2008).

ICP-MS Analysis: Soil and Leaf Samples

The ICP-MS was used for all sample analysis in this work. The *aqua regia* (Ming and Lena 2001) digestion method described above was used for complete digestion of both the soil and ashed vegetation samples (Lawrie et al. 2000; Ghanthimathi et al. 2012). Run time with the ICP-MS was set to 100 s for each sample. The digested soil and leaf samples were then analyzed for metals, in the surface water samples. The Perkin Elmer Pure Plus NexION dual detector calibration solution was used as the atomic spectrometric standard.

Results and Discussions

Activity Concentrations

The activity concentration of ^{212}Pb in the soil was less than that of ^{214}Pb (Fig. 2). This might be due to the long half-lives of the ^{214}Pb parent. However, the ^{212}Pb is more toxic in water and vegetation due to its ease of uptake or dissolution and formation of compounds with other reagents. On the other hand, ^{214}Pb poses a toxicological

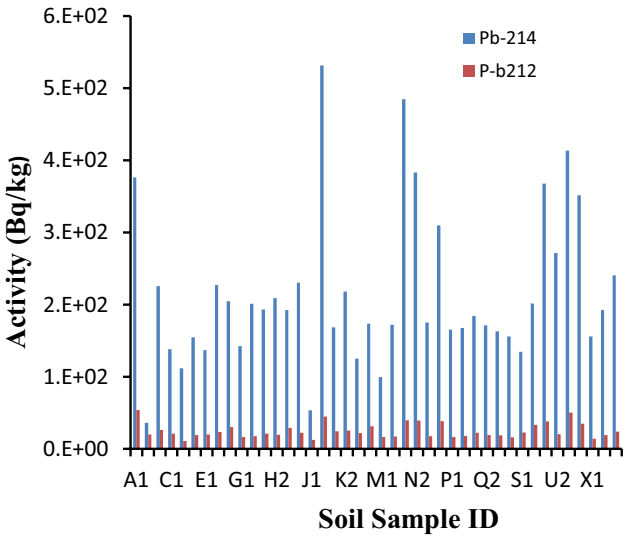

Fig. 2 ^{214}Pb and ^{212}Pb specific activity comparison. A1, A2, etc. denotes sample from 15 and 100 cm deep respectively

hazard. Its concentrations on the surface are sustained by the decay of ^{226}Ra. The ratio of ^{238}U/^{232}Th in the soil was larger than the ratio of ^{214}Pb/^{212}Pb (Fig. 3), which was expected since the decay of the NORM parents have several branches. One branch that causes a ratio imbalance is the loss of ^{222}Rn and ^{220}Rn in the decay of both parents (UNSCEAR 2000).

The mass concentration ratio of U and Th in soil and vegetation at the Princess dump is presented in Fig. 4. The U was easily taken up by plants, especially the blue gum trees (*E. globulus*-types), the leaves of which show a high U concentration. This suggests that this specie of plant could be used for bio-remediation of U in mine dumps for rehabilitation. Gum trees are heavy water absorbers and a good source of firewood and furniture. A uranium mine dump planted with gum trees, which grow fast, might be habitable in a few decades. The authors observed that most of the mine tailings in the Witwatersrand catchment area of the Gauteng Province are surrounded by gum trees to intercept the seepage and to visually obscure the tailings dams.

The activity distribution of ^{238}U, ^{226}Ra, ^{228}Ra, and ^{40}K within the dump at different depths, referred to as top, middle, and bottom, are presented in Fig. 5. Our results confirm the observation that radium crystalline compounds are simple ionic salts (nitrates, chlorides, and bromides), which are soluble in water (Kirby and Salutsky 1964). Thus, these radium salts will wash down the mine dump easily. ^{40}K and ^{238}U increase slightly with the level of the mine dump. It can be concluded that ^{238}U and ^{40}K are not easily washed away. Hence due to their long half-life and in the presence of AMD (Durand 2012), they will continue to release radio-toxic daughters (e.g. ^{226}Ra) and calcium salts to the environment for a long time.

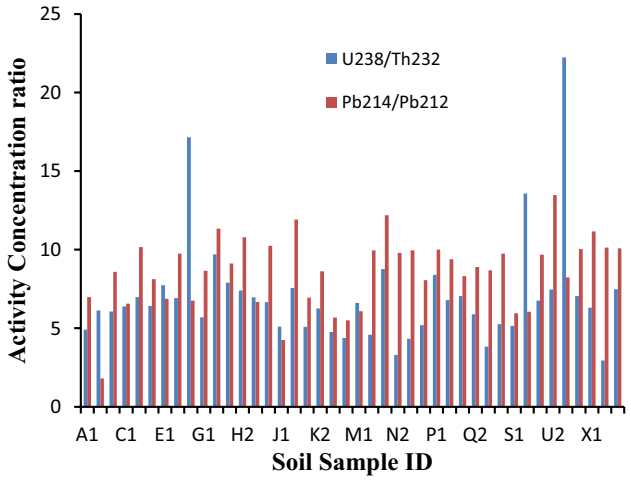

Fig. 3 Comparison of ^{238}U/^{232}Th and ^{214}Pb/^{212}Pb ratios

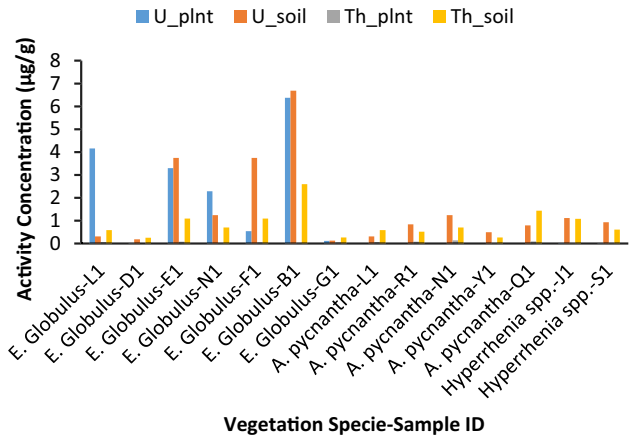

Fig. 4 Total mass concentration of U and Th per vegetation specie at Princess dump

Effective Dose Estimation

Dose coefficients were used to determine the effective dose, which is based on radiation exposure and assessment of human health risks. It is defined as the effective dose equivalent per unit water activity concentration (Sv Bq^{-1}) of the radionuclide. The annual effective dose is calculated using the following equation:

$$AED\left(mSv\,y^{-1}\right) = A\,C\left(Bq\,L^{-1}\right) \times DC\left(Sv\,Bq^{-1}\right) \times AWC\left(L\,y^{-1}\right) \times 1000 \quad (1)$$

where *AED* is the annual effective dose, *AC* the activity concentration (in Bq L^{-1}) of the water, *DC* the dose coefficient (4.5×10^{-8}; ICRP 1995; WHO 2003), and *AWC* is the annual water consumption (731 L year^{-1}). In this study, Eq. (1) was used to determine the annual effective dose of the surfaces water samples for ^{238}U (Fig. 6).

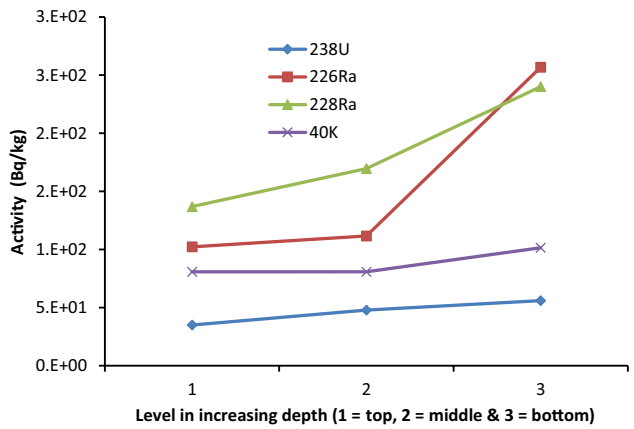

Fig. 5 Specific activity at different levels of the 25 m high tailing

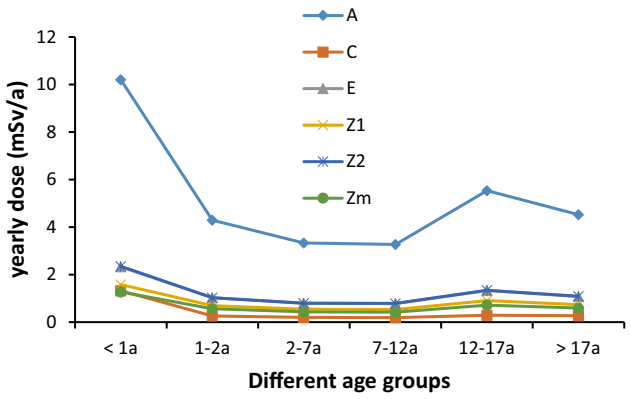

Fig. 6 A plot of the yearly dose for the different age groups compared at all sampling points

From the plot, it can be seen that the annual dose ranges from 0 to 2 mSv/a for all age groups, except at sampling point A, where the dose exceeded 10 mSv/a for infants ≤1 a. This is likely because sampling point A is right at the bottom (30 m) of the dump, while the other sampling points are further away (up to 1 km) from the dump.

The results of this study show and confirm that pollution of the environment via AMD and leaching of U and Th and their daughter products is still significant, though 10 years have passed since the last study was done. Most of the recommendations proposed by previous researchers (Mariette 2006; Ngigi 2009; Winde 2010b; Winde and Jacobus van der Walt 2004) have not been implemented by the South African Government. This study also showed that the communities are at risk of radiation exposure through ingestion of drinking water, with children being particularly vulnerable. It was confirmed that background NORM levels are high in the Witwatersrand Basin because the six billion tons of pyrite tailings containing low-grade uranium (Mariette 2006). Today there are more than 120 tailings dams containing 100,000 tons of U. Percolation contributes 24 tons U (1000 to 1 million times background U concentrations). These tailing dams are now a source of technologically enhanced NORM (TENORM). The dust also releases radon and radionuclide particulates into the atmosphere. Notably, the U and Ra levels exceed the stipulated regulatory limits.

Agricultural Water Use: Irrigation

To decide whether the water from the study area can be used by the residents for irrigation, the concentrations of selected toxic elements were compared against the Food and Agriculture Organization (FAO) standards given for irrigation quality by South Africa's Dept. of Water Affairs and Forestry (DWAF 2002; Table 2). The aluminium, chromium, copper, manganese, nickel, and zinc all exceeded the FAO limits at all of the sampling points. Cadmium was within the limits at all sampling points except for sampling point A. This indicates that the water from these sampled points should not be used for irrigation.

Agricultural Water Use: Livestock Watering

Table 3 compares established contaminant limits for water considered fit to be consumed by livestock (Chap. 5, DWAF 1996) with the concentrations measured at the different sampling points. V and Cd exceeded the DWAF limit only at sampling points C and A, respectively. The rest of the considered elements all exceeded the DWAF limits. Therefore, the water is not fit to be consumed by livestock. Contamination of agricultural crops (pasture, vegetables) is also possible by deposition of dust particles, which we observed to be significant during windy months.

Conclusions and Recommendations

The higher ranges of ^{238}U at the surface than at a depth of 1 m show the potential hazard posed by this radionuclide. Doses due to ingestion of the untreated water were estimated in this study using conversion factors provided in the literature and were found to be above the 0.10 mSv/a

Table 2 Selected water quality criteria for irrigation water (mg/L)

Element	FAO	A	C	E	Z1	Z2	Zm
Aluminum	5.0	480.25	ND	163.32	208.22	284.48	150.11
Arsenic	0.1	0.09	0.29	0.42	0.10	0.57	0.09
Cadmium	0.01	0.05	0.01	0.009	BL	0.001	BL
Chromium	0.1	1.32	8.20	0.73	0.84	4.65	0.70
Copper	0.2	3.00	1.81	1.17	0.88	1.40	0.68
Manganese	0.2	39.47	37.53	7.58	6.40	11.00	4.82
Nickel	0.2	15.04	2.01	5.33	4.15	6.93	3.32
Zinc	2.0	21.10	4.87	21.02	6.61	12.17	5.38

ND not detected, *BL* below detection limit

Table 3 The comparison of limit set for water to be consumed by livestock against concentrations at each sampling point (mg/L)

Element	DWAF limit (mg/L)	A	C	E	Z1	Z2	Zm
Al	(0–5)*, (5–10)***	480.25	ND	163.32	208.22	284.48	150.11
As	(0–1)*, (1–1.5)**	0.093	0.29	0.42	0.097	0.57	0.095
B	(0–5)*, (5–50)**	0.19	1.79	0.29	0.20	0.70	2.19
Cd	(0–0.01)*, (0.01–0.02)**	0.049	0.009	0.008	0	0.002	0
Ca	(0–1000)*, (1000–2000)**	1920.63	4440.69	1250.35	986.40	1407.83	770.93
Cl	(0–1500)*	151.01	984.08	0	533.23	140.88	271.73
Cr	(1–2)**	1.31	8.20	0.73	0.84	4.65	0.70
Co	(0–1)*, (1–2)**	7.52	0.84	3.21	2.42	4.19	1.81
Cu	(0–0.5)*, (1–2)**	3.00	1.81	1.17	0.88	1.40	0.69
Fe	(0–10)*, (10–50)**	419.97	1901.84	979.91	634.71	1172.27	466.49
Pb	(0–0.01)*, (0.1–0.2)**	0.05	0.57	0.27	0.065	0.10	0.065
Mg	(0–500)*, (500–1000)**	90.38	223.32	75.56	63.80	91.48	42.36
Mn	(0–10)*, (10–50)**	39.47	37.53	7.58	6.40	11.00	4.82
Hg	(1–6)**	0.0027	0.0025	0.0025	0.0043	0.0031	0.0035
Mo	(0–0.01)*, (0.01–0.02)**	0.029	0.030	0.028	0.16	0.056	0.078
Ni	(2–5)**	15.04	2.01	5.33	4.15	6.93	3.32
Se	(0–50)*, (50–75)**	0.077	0.063	0.033	0.034	0.034	0.032
Na	(0–2000)*, (2000–2500)**	100.04	77.60	71.18	114.72	91.17	95.37
V	(0–1)*, (1–2)**	0.020	3.36	0.085	0.040	0.089	0.023
Zn	(0–20)*, (20–40)**	21.10	4.87	21.02	6.61	12.17	5.38

*No adverse effects, **adverse chronic effects, ***neurotoxicity

limit set by the World Health Organization (WHO 2008). It was thus concluded that the dose that the public can receive from drinking the untreated water is a potential radiological health risk. The reference value of 0.25 mSv/a, which has been set as the dose limit by the National Nuclear Regulator licensing guide LG-1032 was exceeded by almost all of the water samples analyzed. Ingestion of drinking water was found to be the main exposure pathway, especially to children.

Many recommendations regarding the negative impacts of AMD pollutions have been suggested to the South African Government (Coetzee et al. 2006; DWAF 1996; Winde 2005). The residents of Victory Park have asked the government to remove the tailings, but there are two problems with this. First; remove it to where? The government does not have a repository site for non-functional mine tailings. Second; the cost of moving tons of sand is enormous. The government could potentially move the residents but this would also be expensive. Since the main exposure risk is through drinking the untreated water or using it for irrigation or livestock support, the government could potentially circumvent the challenge of relocating the residents by providing sufficient, clean drinking, washing, and irrigation (garden) water to the community.

This research also suggests that the Government of South Africa should consider planting gum trees around the mine tailings. There is a need to set up a water quality management programme for this site and to initiate environmental remediation of both the surface. This programme could then be extended to similar tailings in the Witwatersrand Basin, according to the DWAF water Quality Management Plan (DWAF 1999).

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