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Research Article

Modelling Of Radiological Health Hazards in Tailing Soils from Uranium Mines in Erongo Region, Namibia

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Abstract

RESRAD-OFFSITE model 4.0 was performed with ICRP 107 based radionuclide transformations transfer factors and ICRP 60 external, inhalation and ingestion dose conversion factors. The cancer morbidity risks modelled for NORM in both mines' samples had shown that ²²⁶Ra was the highest contributor. The RESRAD model included water, plants, soil and atmosphere exposure pathways by external gamma, inhalations and ingestions and had shown risk factors in descending order as ²²⁶Ra > ²³²Th > ⁴⁰K > ²³⁸U. In RESRAD-OFFSITE model code, the total cancer morbidity risks in descending order were recorded as 3 persons per 1 000 populations (3 x 10⁻³) by tailings (stockpiles) and surrounding soil samples from mine 1 < 7 – 8 persons per 1 000 populations (7 x 10⁻³ – 8 x 10⁻³) by tailings (stockpiles) soil samples in mine 2 < 7 – 9 persons per 1 000 populations (7 x 10⁻³ – 9 x 10⁻³) were at risks of developing cancer. This could explicitly prove that the modelled cancer risks in the region was higher than the recommended level of 1 x 10⁻⁵ factor for a population and 1x10⁻³ for a subpopulation documented by the World Health Organization (2011) as well as the world average (0.29 x 10⁻³) documented by the United Nations Scientific Committee on the Effects of Atomic Radiation.

INTRODUCTION

The continuous human exposure to radiation from the environment are a serious concern worldwide. This is due to ubiquitously and unevenly distributed natural radionuclides, such as potassium, uranium and thorium and their decay products, like radon and radium, in the earth's crust. These are chief sources of terrestrial radiations [1,2]. The word 'radiation' is as scary as the word 'cancer'. However, it is scientifically known for its power that keeps warmth on the planet and influences volcanic eruptions, tsunamis, earthquakes [3]. Assessments of radioactive elements in the natural environments has become one of the crucial activities that need to be carried out, mostly in areas accessible to a human population [4,5]. Natural radioactivity in the environment is found in soils, sand, rocks, plants, water and air. Exposures of living organisms, including humans, to natural radioactivity at different levels depends on natural radionuclides present in each area [6,7]. Terrestrial and cosmic radiations are chief sources of a continuous external exposures to human beings.

Uranium is one of the trace elements in the crust of the earth

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- Cancer morbidity risks

with a concentration of 1 - 10 ppm in granitic rocks and also sediments of granitic origin, while thorium ranges between 3 and 30 ppm of concentration for crustal minerals origin [8]. In addition to that, potassium is found ranging from 0.1% to 5%, or even more, with an average of 2.5% in crustal rocks. All these radionuclides are regarded as terrestrial elements that are labelled as naturally occurring radioactive materials (NORM). The radioactivity concentrations of these nuclides obtained from NORM can be used to perform modelling of possible radiological hazards over the years. Radium is a progeny that is most likely present in tailings for uranium mines' stockpiles. Tailing soils are one of the carriers of NORM. There are contaminants or hazardous substances that could be released from uranium mining and milling activities in the form of either chronic or acute [9]. Chronic contaminants are low concentrated substances released over a long period of time whilst acute contaminants are high concentrated substances which are released over a shorter time.

Uranium mining contributes highly on human exposures to NORM in the world. In Namibia, uranium mining was first commissioned in the Rossing Mountains of the Namib Desert,

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Erongo region [10]. This region was nicknamed as the "Uranium Province of Namibia", reportedly with high levels of background radiations in some areas [11]. The estimated average background radiation in this region is high, presumably associated with NORM containing uranium, thorium and potassium radionuclides [12]. Groundwater in the region is therefore likely to have uranium contaminations from primarily deposits in bedrock, the saline aquatic environments of paleo-channels, calcrete in carnotite precipitates, sodium bicarbonates or acid treated uranium and leaching of other radionuclides from tailings.

Cosmological contribution to the background radiation in Namibia is about 0.3 mSv.y⁻¹ at the coastal areas, whilst about 0.7 mSv.y⁻¹ was observed in the central highlands of the country [12]. Majority of inhabitants in the region are living on the coastal towns of Swakopmund, Henties Bay and Walvis Bay, whilst a reasonably but still moderate number live in Arandis, Gobabeb, Karibib, Omaruru, Uis and Usakos towns. The population-weighted average of region due to cosmic radiation is reportedly similar to the world's population-weighted average of 0.38 mSv.y⁻¹ [13]. Terrestrial sources contribute to a maximum of about 7.3 mSv.y⁻¹ in the region, with an average of 0.7 mSv.y⁻¹ doubling the world average of 0.33 mSv.y⁻¹, assessed by radiometric surveys [14]. The contribution to populationweighted average of terrestrial radiations in the region is lower than the average level of the region (0.7 mSv.y⁻¹) and natural terrestrial gamma radiation exposures, both indoor and outdoor, are comparable to the world average of 0.48 mSv.y⁻¹, with typical values ranging from 0.3 - 1 mSv.y⁻¹ [15]. UNSCEAR report has indicated low, intermediate, high and very high background areas receiving doses approximated at annual doses of 5 mSv.y⁻¹ or doses doubling the world average (2.4 mSv.y⁻¹), 5 – 20 mSv.y⁻¹, $20 - 50 \text{ mSv.y}^{-1} \text{ and } > 50 \text{ mSv.y}^{-1}, \text{ respectively } [2].$

Some studies investigated excess lifetime cancer risk (ELCR), the probability of radiation-induced cancer in populations, as a result of lifetime exposure to primordial radionuclides [16,17]. The world average value of ELCR was estimated at 0.29×10^{-3} as documented by international standard organizations [18].

The aim of this study was to model radiological health hazards arising from radioactivity concentrations measured in samples collected from stockpiles of uranium mining tailings using RESidual RADioactivity (RESRAD). Radioactivity concentrations in mining environments may lead to long-term exposures of radon through inhalation which has several human health effects such as acute leucopoenia, chronic lung diseases, anemia, and, to some extent, the necrosis of the mouth [19]. In addition, there are concerns on long-term of radium exposure which causes developments of bone, cranial, and nasal tumors in a human body.

METHODOLOGY

RESRAD-OFFSITE Code, version 4.0 was applied on ICRP 107 based radionuclide transformations transfer factors and ICRP 60 external, inhalation and ingestion dose conversion factors. A web diagram of environmental and exposure pathways was considered, as shown in Figure 1, for all possible transfers of all radionuclides of ²²⁶Ra, ²³⁸U, ²³²Th and ⁴⁰K in NORM as measured in soil. These radionuclides could be interchanged between soil, water and atmosphere and are bioaccumulated in crops or vegetation and may lead to humans through ingestions, inhalations and external gamma radiations. Their activity concentrations measured by a well-calibrated high-purity germanium (HPGe) detector, were converted into Bq.g⁻¹, shown in Table 1. The assumption was made that environmental exposure pathways are the most likely conditions with high probability of transfers contributing to deterministic effects of cancer to members of the public in the region. The extent of possible health risks to the population within the study area has not been documented before. Hence the modelling of the hypothetical risks scenario using RESRAD Code (developed by Argonne, Lemont, IL, USA).

Table 2 shows the site and default values of specific characteristics and parameters entered as inputs on RESRAD-OFFSITE model. The exposure media considered were water, plants, soil and atmosphere. The site-specific mean values of estimated transfer factors for ²³⁸U, ²³²Th and ⁴⁰K radionuclides were obtained from a study conducted on some edible but not the only plants grown in the region such as beetroots, spinach and eggplants [16].

RESULTS AND DISCUSSIONS

Estimation of summed absorbed dose

The absorbed dose summed for all radionuclides concentrations for the two mines considered in modelling as estimated in RESRAD-OFFSITE are graphically shown in Figures 1 and 2.

The modelled effective doses for NORM in tailings samples from uranium mine 1 (section A), had initial value of effective doses of 8.06×10^{-1} mSv per year. RESRAD estimated ²³²Th with an increase from 8.95×10^{-4} mSv.y⁻¹ within 12 months (at year 0) to 8.06×10^{-2} mSv.y⁻¹ after 30 years, and that contributed significantly to the increase of total annual effective dose from 8.06×10^{-1} to 8.76×10^{-1} mSv.y⁻¹ in the same range of years. ²³⁸U was estimated with insignificant contribution to the total effective dose as it remained with almost constant values (at 1.29 x 10^{-4} at

Table 1: The average activity concentrations determined in samples (top of table) actual values in Bq.kg⁻¹, and (bottom of table) as inputs on RESRAD-OFFSITE in Bq.g⁻¹.

| RESidual RADioactivity (RESRAD) modeling the activity concentration values | | | | | | | | | |
|--|------------------|---|------------------|-------------------|-----------------|--|--|--|--|
| Study area | Samples | Actual average activity concentrations in Bq.kg ¹ | | | | | | | |
| | | ²²⁶ Ra | ²³⁸ U | ²³² Th | ⁴⁰ K | | | | |
| А | Tailings soil | 1589.35 | 557.36 | 215.59 | 1079.12 | | | | |
| В | Tailings soil | 4414.15 | 842.59 | 436.74 | 2225.00 | | | | |
| | | Input activity concentrations in Bq.g ⁻¹ on RESRAD – OFFSITE vs 4.0 | | | | | | | |
| А | Tailings soil | 1.59 | 0.56 | 0.21 | 1.08 | | | | |
| В | Tailings soil | 1.41 | 0.84 | 0.44 | 2.23 | | | | |

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Table 2: The site-specific data of radionuclides considered in RESRAD-OFFSITE vs 4.0 modelling of doses and risks in sections A and B, with Arandis as the nearest town.

| Parameters | Interest | Site specific | | Default | Reference | |
|---|-------------------|---------------|-------|---------|---------------------------|--|
| Radionuclide: | | u | ata | uata | | |
| | ²²⁶ Ra | | | 70 | Yu <i>et al.,</i> 2001 | |
| Distribution | ²³⁸ U | | | 50 | Yu <i>et al.,</i> 2001 | |
| (cm ³ .g ⁻¹) | ²³² Th | | | 60000 | Yu <i>et al.,</i> 2001 | |
| | ⁴⁰ K | | | 5.5 | Yu <i>et al.,</i> 2001 | |
| | ²²⁶ Ra | | | 0.04 | Yu <i>et al.,</i> 2001 | |
| Transfer factors (nonleafy | ²³⁸ U | 0.0038 | 0.014 | | Amakali, 2021 | |
| and leafy vegetables) | ²³² Th | 0.0065 | 0.020 | | Amakali, 2021 | |
| | ⁴⁰ K | 1.37 | 2.77 | | Amakali, 2021 | |
| Fynosure | Arandis | 70 | | | Current study | |
| duration (years) | Section A, B | | | 30 | Yu <i>et al.,</i> 2001 | |
| Possible contamination zone: | | | | | | |
| Area (m ²) | Arandis | 11000 | | | Current study | |
| | Section A, B | 750 000 | | | RUL, 2019 | |
| Precipitation (mm) | | 50 | | | MME, 2010 | |
| Erosion rate (m.y ⁻¹) | | | | 2 | Yu <i>et al.,</i> 2001 | |
| Density | | | | 1.55 | Yu <i>et al.,</i> 2001 | |
| Thickness (m) | Arandis | | | 2 | Yu <i>et al.,</i> 2001 | |
| | Section A, B | | | 100 | RUL, 2019 | |
| Radon | ²²² Rn | | | 0.25 | Yu <i>et al.,</i> 2015 | |
| coefficient | ²²⁰ Rn | | | 0.15 | Yu <i>et al.,</i> 2015 | |
| Inhalation rate (m ³ .y ⁻¹) | | | | 8400 | Yu <i>et al.,</i> 2015 | |
| Modelled duration (years) | Two sections | 200 | | | Current study | |

0 year to 1.30×10^{-4} at 200 years). ⁴⁰K was modelled with 8.13 x 10^{-4} mSv.y⁻¹ at 0 year to 8.00 x 10^{-4} mSv.y⁻¹ at 200 years. The total effective doses recorded 8.21 x 10^{-1} mSv.y⁻¹ at 200 years were still greater than the initial value (at 0 year), and again greater than the average level of 0.48 mSv.y⁻¹ from terrestrial sources documented by WHO [15].

The annual effective doses modelled for activity concentrations of NORM in the samples from mine 2 were recorded with a total of 2.16 mSv.y⁻¹ at 0 year with ²²⁶Ra dominating among all other

radionuclides, in Figure 3. An increase in total effective dose was recorded to a maximum of 2.26 mSv.y⁻¹ due to a buildup of ²³²Th doses between 0 and 20 years. RESRAD model estimated a total effective doses above 1.85 mSv.y⁻¹ over 200 years. The estimated doses were greater than the levels from terrestrial sources at 0.48 mSv.y⁻¹ (0.3 – 1 mSv.y⁻¹) of world average by WHO (2011) and average level at 0.7 mSv.y⁻¹ of the region by Wackerle [14], whilst lesser than the maximum of 7.3 mSv.y⁻¹ measured in the region by radiometric surveys [14]. According to UNSCEAR report of 2017, the intermediate to high background in the region is attributed to the emission of radionuclides from the mines with annual doses approximated at 2 mSv.y⁻¹.

Estimation of summed cancer risks

The cancer morbidity risk results modelled on RESRAD-OFFSITE are shown in Figure 4 and 5 for the two mines. The World Health Organization (WHO) report of 2011 recommended acceptable effective lifetime radiation attributable risks which may result in one additional cancer case per 100 000 of a population exposed, that is 1×10^{-5} and one cancer case per 1000 of a subpopulation [20]. USNCEAR has determined the world average cancer risks at 0.29×10^{-3} .

Soils from the stockpiles of mine 1, in Figure 4, shows that the modelled morbidity cancer risks for 40 K, 226 Ra, 232 Th and 238 U, at 0 year were: 1.96×10^{-6} , 2.90×10^{-3} , 7.03×10^{-5} and 2.85×10^{-7} ; at 50 years it is: 1.95×10^{-6} , 2.84×10^{-3} , 1.09×10^{-4} and 2.86×10^{-7} with a total of 2.95×10^{-3} ; and at 200 years, it is: 2.04×10^{-6} , 2.66×10^{-3} , 1.09×10^{-4} and 2.86×10^{-7} with a total of 2.77×10^{-3} . The total cancer risks determined were greater than the world limit of 1.45×10^{-3} documented by ICRP (2012).

The initial morbidity cancer risks determined for ⁴⁰K, ²²⁶Ra, ²³²Th and ²³⁸U in samples of mine 2, in Figure 5, reported at 0, 50 and 200 years were: 3.64×10^{-6} , 7.72×10^{-3} , 1.42×10^{-4} , 4.27×10^{-7} with a total of 7.86×10^{-3} ; 2.57×10^{-6} , 7.34×10^{-3} , 2.20×10^{-4} and 4.12×10^{-7} with a total of 7.56×10^{-3} ; and 1.17×10^{-6} , 6.32×10^{-3} , 2.20×10^{-4} and 3.67×10^{-7} with a total of 6.54×10^{-3} , respectively. Samples from the stockpile have, a high probability of cancer cases in humans. A maximum of 8 persons per 1000 human population had a chance of developing cancer in the region and this chance of cancer case may degrease to 6 person after 200 years of exposure to these stockpiles. The cancer morbidity risks for individual radionuclides of ⁴⁰K, ²²⁶Ra, ²³²Th and ²³⁸U measured in both mines were estimated greater than the world average cancer risks of 0.29 x 10^{-3} documented by UNSCEAR (2008) and WHO (2011).

Ingrowth progeny

The modelled cancer risks had high contribution from daughter products of NORM concentrations considered in the study. Of all modelled radioactivity concentrations in samples, ²²⁶Ra was recorded with the highest cancer risk factors. A practical example was the cancer morbidity risks posed by individual radionuclides in tailings' soil samples of a uranium mine 1 (section A). Figure 6 shows the excess cancer risks from individual radionuclides

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Figure 5 The modelled morbidity cancer risks, individual and summed, for 40K, 226Ra, 232Th and 238U in soil samples from section B (mine 2) stockpiles.



progenies, except for 40K (bottom right).

with their respective ingrowth progenies. Cancer risks by ²³⁸U radioactivity concentration over 200 years arises mostly from daughters of ²³⁴U and ²¹⁰Po, with negligible contributions from ²³⁰Th, ²²⁶Ra, ²¹⁰Pb as shown in Figure 6 (top left). The cancer risks from ²²⁶Ra radioactivity concentration arose from ²¹⁰Po daughter, modelled and resulted in almost the same risks as the parent, with a zero significant contribution from ²¹⁰Pb as shown in Figure 6 (top right). The morbidity risks modelled for both ²³⁸U and ²²⁶Ra show decreasing linear slopes over time. ²³²Th ingrowth progenies that contributed to the cancer morbidity risks were ²²⁸Ra and ²²⁸Th over 200 years, shown in Figure 6 (bottom left). ⁴⁰K was the only radionuclide of NORM under this study with stable daughters and its cancer morbidity risks significantly increasing exponentially between 150 and 200 years as shown in Figure 6 (bottom right).

The cancer morbidity risks for individual radionuclide with their progeny (radon inclusive), considering water, plants, soil and atmosphere exposure pathways by external gamma, inhalations and ingestions were recorded in descending order as 226 Ra > 232 Th > ⁴⁰K > ²³⁸U. In RESRAD-OFFSITE code, the total cancer morbidity risks in descending order were recorded as 3 persons per 1 000 populations (3 x 10⁻³) by tailings (stockpiles) samples from section A (mine 1) < 7 – 8 persons per 1 000 populations (7 x 10^{-3} – 8 x 10^{-3} ³) by tailings (stockpiles) soil samples in section B (mine 2) < 7 - 9 persons per 1 000 populations (7 x $10^{-3} - 9 x 10^{-3}$) were at risks of developing cancer. This could prove that the modelled cancer risks in the region was higher than the recommended level for a population (1×10^{-5}) and a subpopulation (1×10^{-3}) documented by the WHO (2011) as well as the world average (0.29×10^{-3}) documented by UNSCEAR (2008). According to these findings on RESRAD-OFFSITE's long term evaluation, the residents of the region who live closer to mining activity areas would be at high risks of developing cancer on prolonged exposures.

CONCLUSIONS

The cancer morbidity risks modelled for NORM in mine 1 and mine 2 samples has shown that ²²⁶Ra dominantly contributed to exposures. Modelling code show that a high risks factor of ²²⁶Ra is attributed to the build-up of its daughter product, lead-210 (²¹⁰Pb) over 30 years of exposure. ²¹⁰Pb is a hazardous daughter with a half-life of 22 years and it decays by emitting a beta particle (β) to form a bismuth-210 (²¹⁰Bi). It was also crucial to note that ²²⁶Ra existence in tailings soils decays by emission of alpha to form radioactive gas 222 Rn (t_{1/2} = 3.8 days) which escapes from the piles, inhaled and could causes lung cancer in humans [21]. All other daughters in a uranium-238 decay chain have very low half-lives, within 12 months, and therefore their activity concentrations are regarded of less contribution to cancer risks into the human body. The other contributing radionuclide to cancer risks is ²³²Th with build-ups of ²²⁸Ra ($t_{1/2}$ = 5.8 years) which decays by β particle to form a short-lived actinium-228 (^{228}Ac) (t_{1/2} = 6.1 hours), and ^{228}Th (t_{1/2} = 1.9 years) which decays by alpha (α) into a short-lived product radium-224 (²²⁴Ra) with a half-life of 3.6 days. Although activity concentrations for ⁴⁰K were recorded to be high in the region, its contribution factor was recorded less but slightly higher than that of ²³⁸U in all samples.

The study has showed that the modelled cancer risks in the Erongo region is higher than the recommended level of 1×10^{-5} for a population and 1×10^{-3} for a subpopulation documented by the World Health Organization (2011), as well as the world average value of 0.29 x 10^{-3} documented by the United Nations Scientific Committee on the Effects of Atomic Radiation (2008). According to these findings from the RESRAD-OFFSITE's long term evaluation, the residents of the region who live closer to mining activity areas would be at high risks of developing cancer on prolonged exposures.

ETHICAL APPROVAL

The article has not been submitted to a Committee on Publication Ethics (COPE) and therefore no ethics approval received so far.

COMPLIANCE WITH ETHICAL STANDARDS

The authors of this article declare that they have no known financial interests or relationship that could appear to be of influence to the research work discussed and reported in this paper. None of the authors is involved or part of the editorial board which may influence the publication of this article in any form. The research involves only human participants.

Consent to participate and publish

The authors have received explicit consent to submit and that they obtained consent from the responsible authorities at the institute/organization where the work has been carried out, **before** the work is submitted. This article is an extract of a PhD thesis submitted and accepted by North-West University in 2022.

CONSENT TO PUBLISH

All authors have consensus agreement to publish this research work.

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COMPETING INTERESTS

Authors Vaino Indongo and Manny Mathuthu declare they have no competing financial interests. The authors have no relevant financial or non-financial interests to disclose.

AUTHOR CONTRIBUTIONS

All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by both Vaino Indongo and Manny Mathuthu. The first draft of the manuscript was written by Vaino Indongo. Supervision was carried by Manny Mathuthu and all authors commented and approved on previous versions of the manuscript.

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AVAILABILITY OF DATA AND MATERIALS

All data used in this article will be provided upon request by the journal and/or reviewers of this article.

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