

Article **Environmental Radiological Impact and Risk Assessment of Natural Radioactivity at the Heap Leach Facility of Tarkwa Goldmine, Ghana: Radiotoxicity and Public Exposure**

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Abstract: In this study, a comprehensive investigation was conducted to determine the radioactivity levels of naturally occurring radioactive materials (NORMs) in heap pads/soil and water samples within and around the operational area of Tarkwa Gold Mine in Ghana. Gamma-ray spectrometry was used to determine the activity concentrations of ²³⁸U, ²³²Th, and ⁴⁰K in heap pads/soil, and ²²⁶Ra, 228 Th, and 40 K in water samples. The average activity concentrations of 226 Ra and 228 Th in all water samples were below the World Health Organisation (WHO) recommended guidelines for drinking water. Similarly, the average activity concentrations of measured radionuclides in heap pads/soil samples taken from depths of 0–20 cm and 20–50 cm were found to be below the worldwide average reported values. The annual effective dose to the public was estimated to be around 0.16 mSv, which is below the recommended limit. The values of the hazard indices are also below the recommended limits, implying that if heap pads/soils are used for building purposes and construction, they will not pose any significant radiation hazard. The results of this study indicate that radiation levels are within the natural background radiation levels reported in the literature and are consistent with findings from similar studies conducted in Ghana.

Keywords: environmental radioactivity; radiation risk assessment; gamma spectrometry; gold mine; public health; effective dose; hazard indices; soil; water

1. Introduction

Natural radioactivity constitutes the largest source of population dose [\[1\]](#page-10-0). The primary sources of external gamma radiation include 238 U and 232 Th, and their decay products, along with $40K$. The internal dose humans receive from naturally occurring radionuclides is mainly due to the short-lived progenies of $226Ra$, including $222Rn$ [\[2\]](#page-10-1). Despite the widespread distribution of these radionuclides in nature, their concentrations depend significantly on local geological conditions and thus vary by location [\[3,](#page-10-2)[4\]](#page-10-3), which is related to the type of rock. For example, low concentrations are found in sedimentary rocks, while higher concentrations are typically associated with igneous rocks, such as granite. However, there are exceptions; some shales and phosphate rocks have a relatively high radionuclide content, and higher concentrations may arise due to human activities such as mining and mineral processing [\[1](#page-10-0)[,5](#page-10-4)[,6\]](#page-10-5).

Mining is one example of potential occupational exposure to naturally occurring radioactive materials (NORMs) [\[1\]](#page-10-0). However, such workplaces are not yet regulated for

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monitoring and assessing direct exposure to NORMs in Ghana. Several ongoing studies in Ghana aim to establish the activity concentrations of radionuclides resulting from industrial activities such as mining and mineral processing. Some of these studies are part of a national project to establish a baseline for environmental radioactivity and to create a national reference database for both new and existing mines. Published studies in this field include references [\[7–](#page-11-0)[11\]](#page-11-1). For instance, the activity ranges of 238 U, 232 Th, and 40 K in soil samples from two mines were reported as approximately 29 and 34, 25 and 21, and 582 and 682 Bq⋅kg⁻¹, respectively, with an average annual effective dose of 0.3 ± 0.1 mSv [\[9\]](#page-11-2).

In this research, efforts were made to conduct a comprehensive radiological survey to assess the radioactivity levels of naturally occurring radioactive materials (NORMs) in heap pads/soils and water samples from the Tarkwa Gold Mine [\[12](#page-11-3)[,13\]](#page-11-4), with the aim of facilitating the decommissioning of the facility. The availability of data from these studies is crucial for all stakeholders involved in environmental protection measures. The specific objectives of the study were as follows:

- To determine the activity concentrations of radionuclides from the U/Th series and 40 K.
- To calculate the doses resulting from these activity concentrations and compare them with internationally recommended dose limits to assess the risk to the public.
- To evaluate the suitability of heap pads/soils for purposes such as building, road construction, block making, etc., by assessing the radioactive hazard indices.

Description of Study Area

The research site is located at the Tarkwa Gold Mine, operated by Gold Fields Ghana Limited, a subsidiary of Gold Fields Limited. This mining concession covers approximately 294.606 km² and is situated in Ghana's Western Region. Positioned approximately 4 km west of Tarkwa town, the Tarkwa Gold Mine benefits from robust infrastructure and easy accessibility. A major roadway connects the mine to Takoradi port, located roughly 140 km southeast along the Atlantic coast. Tarkwa itself is around 300 km from Accra, Ghana's capital, positioned at latitude 5◦15′ N and longitude 2◦00′ W. Serving as the administrative centre for the area, agriculture is the primary occupation of the local population, with mining as the key industrial sector [\[12,](#page-11-3)[14\]](#page-11-5). The region forms part of Ghana's significant gold belt, stretching from Axim in the southwest to Konongo in the northeast [\[15\]](#page-11-6). Tarkwa Township has a population of around 90,477 people, while the district's total population is estimated at 218,664 [\[16\]](#page-11-7).

Geology, Hydrogeology, and Meteorology of the Mining Area

The gold deposits within the study area are located in the Tarkwaian system, which forms an integral part of the Ashanti Belt's stratigraphy in southwestern Ghana. Intrusive igneous rocks make up around 20% of the total thickness of the Tarkwaian system in Tarkwa. The ore body consists of multiple sedimentary banket quartz reef units, similar to those found in South Africa's Witwatersrand. A geological map of the area is shown in Figure 1 (detailed geological information and map of area can be found in pervious published papers [\[17](#page-11-8)[,18\]](#page-11-9)).

The geological structure of the mine area reveals that the gold-bearing ore is situated between non-ore-bearing belts predominantly composed of sedimentary rocks. The Ashanti Belt, where these formations are located, is a northeasterly striking synclinal structure comprising lower Proterozoic sediments and volcanics, underlain by metavolcanics and metasediments of the Birimian system. The interface between the Birimian and Tarkwaian systems is often marked by intense shearing, which hosts several significant shear-hosted deposits such as Prestea, Bogoso, and Obuasi. Dominating the local geology is the Banket Series, which includes well-sorted conglomerates and pebbly quartzites. These quartzites contain clasts typically of Birimian origin and are notable for substantial gold mineralization, forming the Tarkwa ore body.

In the Tarkwaian system, the rock formations comprise the Kawere Group, the Banket Series, Tarkwa Phyllite, and Huni Sandstone. The surface presence of sandstone in most areas results from the weathering of underlying parent quartzites [\[17\]](#page-11-8).

Hydrogeologically, with the exception of Tarkwa Township in the Wassa West District, most major towns and villages rely on groundwater sourced from boreholes and hand-dug wells for their water supply [\[15\]](#page-11-6). This groundwater presence is associated with secondary porosity developed through fissuring and weathering, as the region's rocks generally lack primary porosity due to their consolidated nature. Soils in the Tarkwa–Prestea area are classified into two types: forest oxysol in the southern regions and a mix of forest ochrosol–oxysol in the northern areas [\[15\]](#page-11-6).

2. Materials and Methods

2.1. Sampling and Sample Preparation for Gamma Spectrometry Analysis

A total of 165 samples were collected from selected areas within the mine concession, comprising 120 pads/soil samples and 45 water samples (corresponded sampling locations are listed in Tables [1](#page-2-0) and [2\)](#page-3-0). The soil samples were extracted at two depths: 0–20 cm and 20–50 cm.

σ—standard deviation with coverage factor K = 2.

The soil samples were air-dried for a week and then further dried in an oven at 105 °C until all moisture content was completely removed. Following internal sample preparation guidelines, the samples were pulverized into fine powder and sieved through a 2 mm mesh before being stored in Marinelli beakers. These beakers, sealed tightly, were stored for one month to ensure that short-lived daughters of 238 U and 232 Th decay series attained equilibrium with their long-lived parent radionuclides. For the water samples, following the internal sampling guidelines to maintain radioactive equilibrium between ²²⁶Ra and its short-lived daughters, samples were homogenized and transferred into one-litre Marinelli beakers without filtration. The prepared samples were sealed, weighed, and stored prior to measurement [\[12,](#page-11-3)[19\]](#page-11-10).

Table 2. Activity concentrations, absorbed dose rates, radium equivalent activities, external and internal hazard indices, and annual effective doses of ²³⁸U, ²³²Th, and ⁴⁰K in soil samples at depth of 0–20 cm from Tarkwa Gold Mine.

σ—standard deviation with coverage factor K = 2.

2.2. Instrumentation and Calibration

Direct gamma spectrometry analysis was performed on soil and water samples using a non-destructive method employing a High-Purity Germanium (HPGe) detector system (AMETEK ORTEC, Oak Ridge, TN, USA). The HPGe detector, connected to a multi-channel analyser (MCA) operated via computer, offers 40% relative efficiency with an energy resolution of 2.0 keV at a gamma-ray energy of 1332 keV from ${}^{60}Co$.

For radionuclide identification, characteristic gamma-ray energies were utilized, and quantitative analysis was conducted using the Genie 2000 gamma acquisition and analysis software (Mirion, Atlanta, GA, USA). The detector is shielded by a 100 mm passive shielding consisting of layers of lead, copper, cadmium, and plexiglass (3 mm each) to minimize background radiation from external X-rays.

Calibrations for energy and efficiency were conducted by counting standard radionuclides with known activities and precise energies, ranging from 60 keV to approximately 2000 keV. The efficiency calibration for analysing soil, rock, and water samples utilized standard radionuclides uniformly distributed in a solid water substitute with a volume of 1000 cm³ and a density of 0.98 g/cm³, provided by the Czech Metrology Institute (source number: 9031-OL-146/14).

2.3. Activity Concentration, Dose, and Radiological Hazard Calculations

2.3.1. Calculation of Activity Concentration by Gamma Spectrometry

The concentration of radioactive decay, quantified as activity concentration in environmental samples of soil and water, was evaluated for specific isotopes using the gamma-ray spectrometric method. This method focused on the distinct energy peaks associated with the decay products of these isotopes. For 238 U and 226 Ra, determination was based on the 609.3 keV gamma-ray emission from 214 Bi. The activity concentration of 232 Th in soil and 228 Th in water were determined using the 911.2 keV peak of 228 Ac and 238.6 keV peak of ²¹²Pb, respectively. Additionally, the presence of 40 K was quantified by its 1460.8 keV gamma-ray peak. These measurements enabled the calculation of radioactive content in becquerels per kilogram (Bq⋅kg⁻¹) for soil and becquerels per litre (Bq⋅l⁻¹) for water as outlined in Equation (1), an analytical formula designed to convert the observed gamma-ray data into specific activity concentrations. This rigorous approach leverages gamma-ray spectroscopy to provide a precise assessment of natural radioactivity in these environmental mediums [\[20\]](#page-11-11).

$$
A_{sp} = \frac{N_D}{p.T_c.\eta(E).M}
$$
 (1)

where N_D represents the net counts of the radionuclide in the samples, p denotes the probability of gamma-ray emission (gamma-ray yield), Tc is the counting time of the sample, $\eta(E)$ signifies the counting efficiency of the detector system, and M refers to the mass (kg) or volume (l) of the sample.

2.3.2. External Gamma Dose Rate

Using Equation (2), the external gamma dose rates (D_{γ}) at 1.0 m above ground for the soil samples were estimated [\[5,](#page-10-4)[12,](#page-11-3)[21\]](#page-11-12).

$$
D_{\gamma}(\text{nGy/h}) = DCF_U \times A_U + DCF_{Th} \times A_{Th} + DCF_K \times A_K \tag{2}
$$

where DCF_u, DCF_{Th}, and DCF_K (0.462, 0.604, 0.0417) are the dose conversion for ²³⁸U, ²³²Th, and ⁴⁰K (nGy·h⁻¹/Bq·kg⁻¹), respectively, and A_u, A_{Th}, and A_K are the activity concentrations (Bq·kg⁻¹) of ²³⁸U, ²³²Th, and ⁴⁰K, respectively.

2.3.3. External Effective Dose

The derived average annual external effective dose for 8760 h per year was estimated from the absorbed dose rate calculated using Equation (2), applying an outdoor occupancy

factor of 0.2, and an absorbed dose conversion factor of 0.7 Sv \cdot Gy⁻¹ for environmental exposure to gamma rays as per Equation (3).

$$
E_{\gamma} = D_{r} \times 0.2 \times 8760 \times 0.7
$$
\n(3)

where E_y is the average annual effective dose and D_r is the absorbed dose rate in air.

2.3.4. Committed Effective Dose

The committed effective doses from water samples were calculated using the activity concentrations of each radionuclide and the annual water consumption rate for adults, approximately 730 litres [\[22\]](#page-11-13), applying Equation (4) [\[23,](#page-11-14)[24\]](#page-11-15).

$$
S_{ing}(w) = I_w \sum_{j=1}^{3} DCF_{ing}(Ra, Th, K) A_{sp}(w)
$$
\n(4)

where, $A_{\rm sp}(w)$ is the activity concentration of the radionuclides, (Bq·l⁻¹), I_w is the water consumption rate, and DCF_{ing} is the ingestion dose coefficient per radionuclide (Sv per Bq·l⁻¹) for ²²⁶Ra, ²²⁸Th, and ⁴⁰K.

2.3.5. Radium Equivalent Activity and Hazard Indices

The radiological risk posed by NORMs in heap pads/soils within the study area, potentially used for road construction, housing/building, cement production, block making, etc., was evaluated. This assessment involved calculating the radium equivalent activity (Ra_{eq}) as well as the external and internal hazard indices. Ra_{eq} , a commonly used hazard metric, was determined using Equation (5) [\[12](#page-11-3)[,25\]](#page-11-16). Ra_{eq} serves as a unified index to measure the radioactivity of construction materials, combining the activities of 226 Ra, 232 Th, and 40 K into a single value to assess the potential radiological hazard [\[26](#page-11-17)[,27\]](#page-11-18).

$$
Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_{K}
$$
\n(5)

where C_{Ra} , C_{Th} , and C_K are the activity concentrations of ²²⁶Ra, ²²⁸Th, and ⁴⁰K, respectively. The maximum recommended value of Ra_{eq} in raw building materials and products must be less than 370 Bq⋅kg⁻¹ for safe use. This ensures that the external gamma dose remains below 1.5 mSv per year. The external hazard index (H_{ex}) is another commonly used metric to estimate the level of gamma radiation associated with natural radionuclides in construction materials. It should be less than one, as shown in Equation (6) [\[12,](#page-11-3)[25,](#page-11-16)[28](#page-11-19)[,29\]](#page-11-20).

$$
H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810}
$$
 (6)

Also, the internal hazard index (H_{in}) due to radon and its daughters was calculated from Equation (7).

$$
H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810}
$$
 (7)

The internal hazard index should be less than one for construction materials to be considered safe for utilization. However, the radium equivalent activity (Ra_{eq}) and the external hazard index (H_{ex}) are related and using one index might be equal to other. Ra_{eq} might simplify the overall assessment by combining radionuclide activities, while H_{ex} ensures that this combination does not result in a dose exceeding the safety limit.

2.3.6. Estimation of Total Annual Effective Dose in Soil and Water

The total annual effective dose (E_T) was calculated using Equation (8) based on the ICRP dose calculation method [\[30](#page-12-0)[,31\]](#page-12-1).

$$
E_T = E_{\gamma}(U, Th, K) + E_{ing}(W)
$$
\n(8)

where E_T is the total effective dose (Sv), E_V (U, Th, K) is the external gamma effective dose (the average of the in situ and derived external effective doses, that is, Equations (3) and (4)), and $E_{ing}(W)$ is the effective dose from water.

3. Results and Discussion

Table [1](#page-2-0) summarizes the average activity concentrations of 226 Ra, 228 Th, and 40 K in the water samples. The lowest and highest average values recorded for $226Ra$ were $0.44\,\pm\,0.12$ Bq·l⁻¹ and $0.52\,\pm\,0.17$ Bq·l⁻¹, for ²²⁸Th were $0.60\,\pm\,0.13$ Bq·l⁻¹ and 0.73 ± 0.12 Bq·l $^{-1}$, and for $^{40}\rm{K}$ were 3.24 \pm 0.44 Bq·l $^{-1}$ and 4.86 \pm 0.50 Bq·l $^{-1}$, respectively. These values are comparable to those reported in other studies [\[7](#page-11-0)[–12\]](#page-11-3). The WHO recommends 1.0 Bq \cdot l $^{-1}$ as the guideline level for 226 Ra and 228 Th. As there is no guideline level for ⁴⁰K, the activity concentrations of ²²⁶Ra and ²³²Th in all the water samples were below these recommended guideline levels.

The overall average annual effective dose was calculated to be 0.13 ± 0.04 mSv per year, with a range of 0.13–0.15 mSv per year. The main source of water supply for the Tarkwa Mine is underground, used for both domestic purposes and in the processing plant. The WHO recommended guideline for annual effective dose in drinking water is 0.1 mSv per year [\[19\]](#page-11-10). The average annual effective dose of the water samples in this study is slightly higher than the WHO recommended guideline level. However, it should be noted that some of the water sources in the study area are not intended for drinking or domestic use and therefore are not expected to pose any significant radiation hazard.

Tables [2](#page-3-0) and [3](#page-7-0) present the activity concentrations of 238 U, 232 Th, and 40 K in the heap pads/soil samples collected at depths of 0–20 cm and 20–50 cm, respectively.

For the 0–20 cm depth, as shown in Table [2,](#page-3-0) the average activity concentrations of 238 U ranged from 7 \pm 1 to 13 \pm 1 Bq·kg⁻¹, 232 Th ranged from 16 \pm 1 to 56 \pm 10 Bq·kg⁻¹, and 40 K ranged from 63 \pm 10 to 371 \pm 55 Bq⋅kg⁻¹. Similarly, for the 20–50 cm depth, as observed from Table [3,](#page-7-0) the average activity concentrations of ²³⁸U ranged from 7 ± 1 to 19 \pm 1 Bq⋅kg⁻¹. For ²³²Th, the average activity concentrations ranged from 17 \pm 3 to 48 \pm 11 Bq⋅kg $^{-1}$, and for 40 K, they ranged from 18 \pm to 311 \pm 3 Bq⋅kg $^{-1}$.

The worldwide average activity concentrations of 238 U, 232 Th, and 40 K in soil samples are reported as 33, 45, and 420 Bq·kg $^{-1}$, respectively [\[1\]](#page-10-0). By comparison, the average activity concentrations of ²³⁸U and ⁴⁰K in the heap pads/soil samples from the mine, as shown in the tables, align well with similar studies conducted in Ghana and elsewhere [\[9](#page-11-2)[–12\]](#page-11-3). In terms of depth profile, the results show no significant difference for 238 U. However, for 232 Th, the activity concentration in the 0-20 cm depth was higher than in the 20-50 cm depth. This is expected because 232 Th has low solubility and tends to adsorb to particulate matter, thus not readily leaching unless under acidic conditions. Therefore, the activity concentration of 232 Th is expected to be higher in the topsoil.

In comparison to the International Atomic Energy Agency (IAEA) Exemption Levels, all results are well below the recommended levels of 1000 Bq⋅kg⁻¹ for ²³⁸U and ²³²Th, and 10,000 Bq⋅kg⁻¹ for ⁴⁰K in materials that would require regulatory control [\[22\]](#page-11-13).

As depicted in Figure [1,](#page-8-0) there is a high and consistent correlation between ²³⁸U and ²³²Th at both depths, indicating a stable relationship between these two radionuclides. Conversely, the correlation between 238 U and 40 K is low at both depths, with a noticeable decrease in correlation with increasing depth. This behaviour may be attributed to the solubility and mobility of these radionuclides in soil.

Table 3. Activity concentrations, absorbed dose rates, radium equivalent activities, external and internal hazard indices, and annual effective doses of ²³⁸U, ²³²Th, and ⁴⁰K in soil samples at depth of 20–50 cm from Tarkwa Gold Mine.

σ—standard deviation with coverage factor K = 2.

Figure 1. Correlation matrix between radionuclides at different depths. **Figure 1.** Correlation matrix between radionuclides at different depths.

A moderate correlation is observed between ²³²Th and ⁴⁰K at both depths, suggesting a consistent but modest relationship, likely due to similar interactions of ²³²Th and ⁴⁰K with soil components and environmental factors.

The Student's *t*-test was comparing the means of each radionuclide at both depths and could indicate that there were no statistically significant differences for any of the meters (all p -values > 0.05). parameters (all p -values > 0.05).

Figures 2 and 3 show 3D scatter plots of radionuclide concentration distribution for in the average absorbed dose rate for the 0–20 cm depth was 30.4 ± 5.6 nGyr 1, 5.6 nGyr 1, 5.6 nGyr 1, 5.6 nGyr both depths.

Figure 2. Three-dimensional scatter plot of 238U, 232Th, and 40K concentrations in soil samples taken **Figure 2.** Three-dimensional scatter plot of ²³⁸U, ²³²Th, and ⁴⁰K concentrations in soil samples taken from depth 0–20 cm. from depth 0–20 cm.

Figure 3. Three-dimensional scatter plot of ²³⁸U, ²³²Th, and ⁴⁰K concentrations in soil samples taken from depth 20–50 cm. from depth 20–50 cm.

The average gamma dose rate and annual effective dose from terrestrial gamma rays, calculated from soil activity concentrations, are also presented in Tables [2](#page-3-0) and [3.](#page-7-0) As shown in these tables, the average absorbed dose rate for the 0–20 cm depth was 30.4 \pm 5.6 nGy·h $^{-1}$, with a corresponding average annual effective dose of 0.04 \pm 0.06 mSv. For the 20–50 cm depth, the average absorbed dose rate was 32.5 \pm 5.7 nGy·h⁻¹, with an average annual effective dose of 0.04 ± 0.06 mSv.

According to the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) report, the worldwide average absorbed dose rate measured in air from terrestrial gamma radiation is 59 nGy·h⁻¹. In comparison, the absorbed dose rates measured from the mine, as shown in the tables, are lower than this worldwide average value.

The overall average values of Ra_{eq} from the mine are 66 \pm 13 Bq·kg⁻¹ for the 0–20 cm depth and 71 \pm 14 Bq·kg $^{-1}$ for the 20–50 cm depth, both well below the recommended limit of 370 Bq \cdot kg $^{-1}$. The overall average values of H_{ex} and H_{in} in the heap pads/soil samples are 0.19 \pm 0.02 and 0.22 \pm 0.07, respectively, both below the recommended limit of 1.0.

These results imply that the heap pads/soils from the mine are radiologically safe and can be used for building, road construction, block making, etc., without posing any significant radiation hazard.

The total average annual effective dose due to the external gamma dose rate from soil and the ingestion of drinking water was calculated to be 0.16 mSv \cdot y $^{-1}$, which is less than the ICRP recommended value of 1 mSv⋅y⁻¹ for public [\[30](#page-12-0)[,31\]](#page-12-1).

4. Conclusions

This study aimed to assess radiation doses from activity concentrations of radionuclides in the U/Th series and $40K$, and to evaluate associated hazards. Radiation hazard indices were calculated to determine the suitability of heap pads and soils for building and road construction. Two exposure pathways were considered: direct external gamma-ray exposure from natural radioactivity concentrations in heap pads/soil from 238 U, 232 Th,

and 40 K, and internal exposure due to 226 Ra, 228 Th, and 40 K in water samples from ponds and boreholes. The average activity concentrations of 226 Ra and 228 Th in all water samples were below recommended guideline levels, with the corresponding average annual effective dose slightly exceeding the WHO recommended guideline of 0.1 mSv per year. This suggests that while water sources in the study area, including ponds and boreholes, contain some natural radioactivity, the levels are insignificant and do not pose a radiation hazard. The total annual effective dose from external gamma radiation and ingestion of drinking water was calculated to be 0.16 mSv per year, which is below the 1 mSv per year dose limit recommended by the ICRP for public radiation exposure control. The radium equivalents and hazard index values were below the recommended limits of 370 Bq⋅kg⁻¹ and 1.0, respectively, indicating that heap pads/soils from the mine do not pose a significant radiation hazard.

However, considering the average annual effective dose in water samples from the mine slightly exceeds the WHO recommended guideline of 0.1 mSv/year, it is recommended that ponds and certain boreholes located at the mine be restricted from public use for domestic purposes.

Overall, the results indicate no significant levels of natural radionuclides in the mine. These data can serve as baseline values and reference material for future studies of natural radioactivity in the area, particularly as no existing data were available prior to the commencement of mining activities in the region.

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Conflicts of Interest: The authors declare no conflicts of interest.

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