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Indoor Radon Monitoring in Residential Areas in the Vicinity of Coal Mining Operations in the Mpumalanga Province, South Africa

Paballo M. Moshupya [1,2,*], Seeke C. Mohuba [1], Tamiru A. Abiye [1], Ian Korir [2] and Sifiso Nhleko [2]

[1] School of Geosciences, University of the Witwatersrand, Johannesburg 2050, South Africa; 839981@students.wits.ac.za (S.C.M.); tamiru.abiye@wits.ac.za (T.A.A.)
[2] Centre for Nuclear Safety and Security, National Nuclear Regulator, Centurion 0046, South Africa; ikorir@nnr.co.za (I.K.); snhleko@nnr.co.za (S.N.)
* Correspondence: 729843@students.wits.ac.za

Abstract: Coal mining and combustion have the potential to increase exposure to radon, a form of radioactive gas recognized as one of the major contributors to lung cancer incidents. In South Africa, coal is used as the primary energy source for producing electricity and for heating, predominantly in informal settlements and township communities. Most of the existing coal-fired power plants are found in the Mpumalanga province. This paper presents long-term radon (^{222}Rn) measurements in dwellings surrounding coal mining centres in the Mpumalanga province and evaluates their contributions to indoor radon exposures. The indoor radon measurements were conducted using solid-state nuclear track detectors and were performed during warm and cold seasons. It was found that the overall indoor radon activity concentrations ranged between 21 Bq/m^3 and 145 Bq/m^3, with a mean value of 40 Bq/m^3. In all the measured dwellings, the levels were below the WHO reference level of 100 Bq/m^3 and 300 Bq/m^3 reference level recommended by the IAEA and ICRP, with the exception of one dwelling that was poorly ventilated. The results reveal that individuals residing in the surveyed homes are not exposed to radon levels higher than the WHO, ICRP, and IAEA reference levels. The main source influencing indoor radon activity concentrations was found to be primarily the concentration of uranium found in the geological formations in the area, with ventilation being an additional contributing factor of radon levels in dwellings. To maintain good air quality in homes, it is recommended that household occupants should keep their dwellings well ventilated to keep indoor radon levels as low as possible.

Keywords: coal mining; Karoo Supergroup; Mpumalanga province; radon gas; South Africa

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1. Introduction

Coal is predominantly composed of organic matter and contains trace elements such as uranium (U), thorium (Th), and their associated decay progeny; radium (Ra) and radon (Rn), which are naturally radioactive [1]. Coal, in South Africa, is found in nineteen coalfields and mainly mined in areas of the Mpumalanga province [2]. It is used as a primary resource for electric power generation and about 77 percent of South Africa's energy is produced from its combustion in coal-fired power plants [3,4]. During coal combustion uranium, thorium, and the associated decay daughters contained in the coal matrix are partitioned between the gas and solid phase combustion residuals. Radon gas is transferred to the gas phase and released into the atmosphere [1]. On the other hand, the elements that are less

volatile such as uranium, thorium, and the majority of their less volatile decay products remain in the solid wastes [1]. Thus, the combustion of coal has the potential to enhance the background radioactivity concentrations in the surroundings of the coal-fired power plants and also produce fly ash, a by-product that is enriched with radioactive elements [5–8]. The other cause of concern in the Mpumalanga province arises from the coal mining activities and their associated waste piles that are found in the area. The coal reserves in this region are mined using open-pit, underground, and surface mining techniques. During these excavation processes coal seams are exposed to the surface, and, therefore, may provide favourable conditions for radon gas to easily escape into the atmosphere.

The studies that assessed the concentrations of specific trace elements and radionuclides in the coal found in South Africa [9–12], recorded uranium concentrations that exceeded the world average concentration of 1.9 ppm (mg/kg) in hard coal [13]. Radium-226 (^{226}Ra) measured in the coal used in some of the power plants in South Africa was found to range between 21.69 Bq/kg and 52.63 Bq/kg [12]. Uranium (^{238}U) and radium (^{226}Ra) are the primary and immediate parent radionuclides of radon gas (^{222}Rn), respectively. ^{222}Rn is the most significant contributor of ionizing radiation received by the human population and is classified as one of the largest causes of lung cancer [14]. It is, therefore, important to characterize radon levels in areas around the coal operations and their associated residual products to assess if there are elevated exposures that may present a health risk to the human population in close proximity.

The selected areas of focus for this study were the residential areas in the vicinity of coal operations in the Mpumalanga province of South Africa (Figure 1). As far as the authors are aware, to date, only two studies have been conducted in some parts of the Mpumalanga Province to assess the influence of coal-fired power stations on indoor radon activity concentrations [15,16]. The first study was conducted in the Secunda area and recorded an average indoor radon activity concentration of 76.4 Bq/m^3 [15]. The second study was performed in eMalahleni and reported an average concentration of 122.1 Bq/m^3 [16], which is above the reference level of 100 Bq/m^3 recommended by the World Health Organization (WHO 2009) for indoor radon exposure [14]. However, this value is well below the reference level of 300 Bq/m^3 recommended by the ICRP and IAEA [17,18]. These studies were performed over a short period, for about 3 to 4 days. As such, to obtain long-term exposure levels, the current study investigated radon exposure levels over months and during various seasons to account for temporal variations. The study was also performed on a larger scale in coal industrial areas of the Mpumalanga province, which is situated in the north-eastern part of South Africa (Figure 1). The areas covered in this study included the Middleburg, eMalahleni, Phola, Kriel, Secunda, and Delmas areas (Figure 1). These areas are dominated by coal operations such as mines, coal-fired power stations, coal liquefaction plants, and coal residual products that are in the vicinity of residential areas, which include both formal and informal settlements (Figure 1).

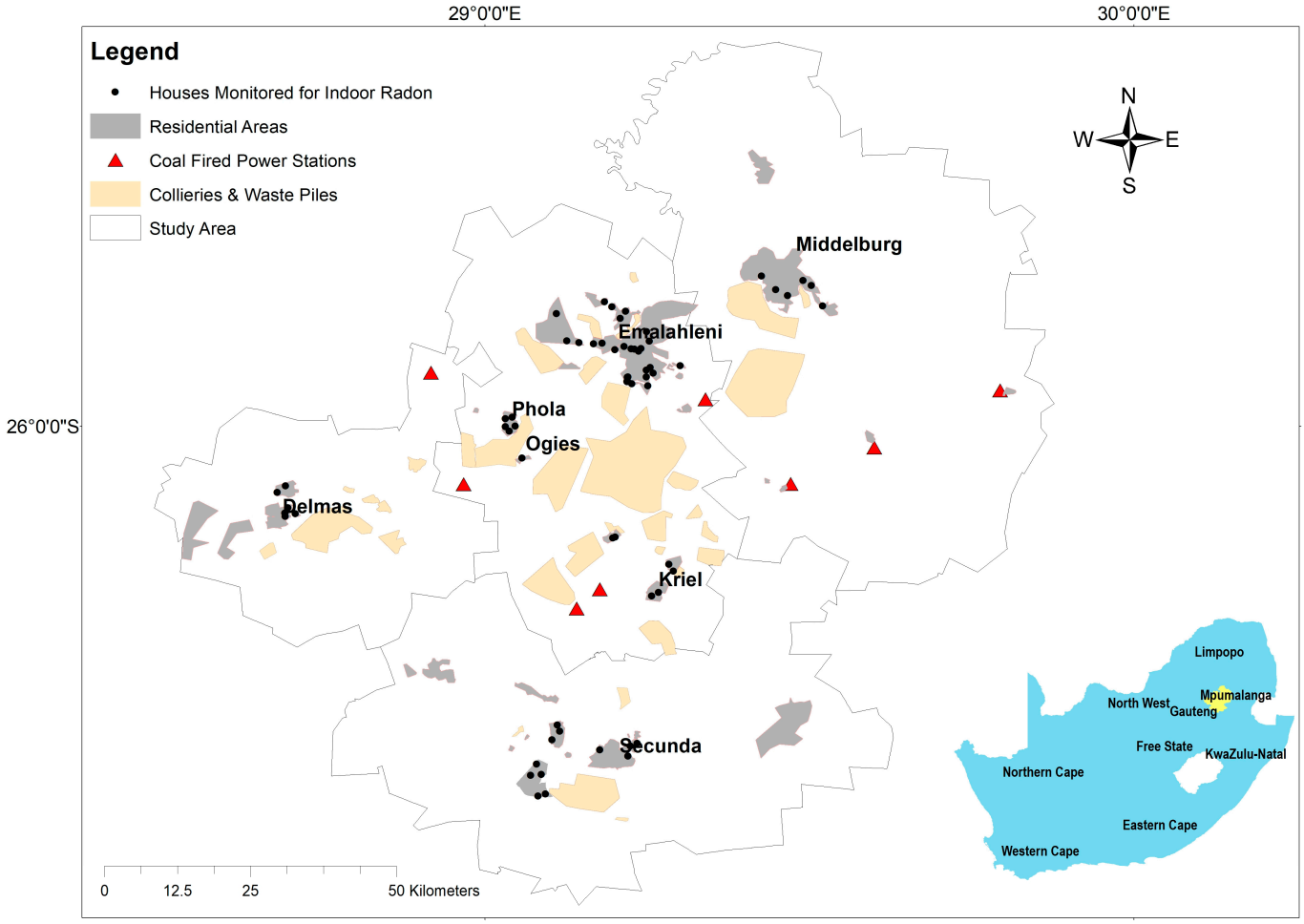

Figure 1. Location of the study area depicting the coal industrial operations and residential areas surveyed as well as the location of the study area in the country (shown in yellow on the blue map).

Geological Overview of the Area

The study area is underlain by rocks of the Transvaal Supergroup, Bushveld Igneous Complex, Waterberg Group, and Karoo Supergroup (Figure 2). However, a large portion of the area is covered by the rocks of the Karoo Supergroup. These include the diamictites of the Dwyka Group appearing towards the western edges of the area and the sandstones, shales, and coal seams of the Ecca Group, which are the dominant lithologies with widespread dolerite dykes and sills. The other main lithologies found in the area are the sandstones, conglomerates, and shales of the Waterberg Group, which extend in the north-western part of the area. The rocks of the Bushveld Igneous Complex are disseminated across the study area and only cover small portions of the study extent. The rhyolites of the Rooiberg Group, which belong to the Transvaal Supergroup, also occupy a larger portion in the north-eastern part of the study area with the Chuniespoort and Pretoria Group rocks covering small portions in the south-western part around the Delmas area.

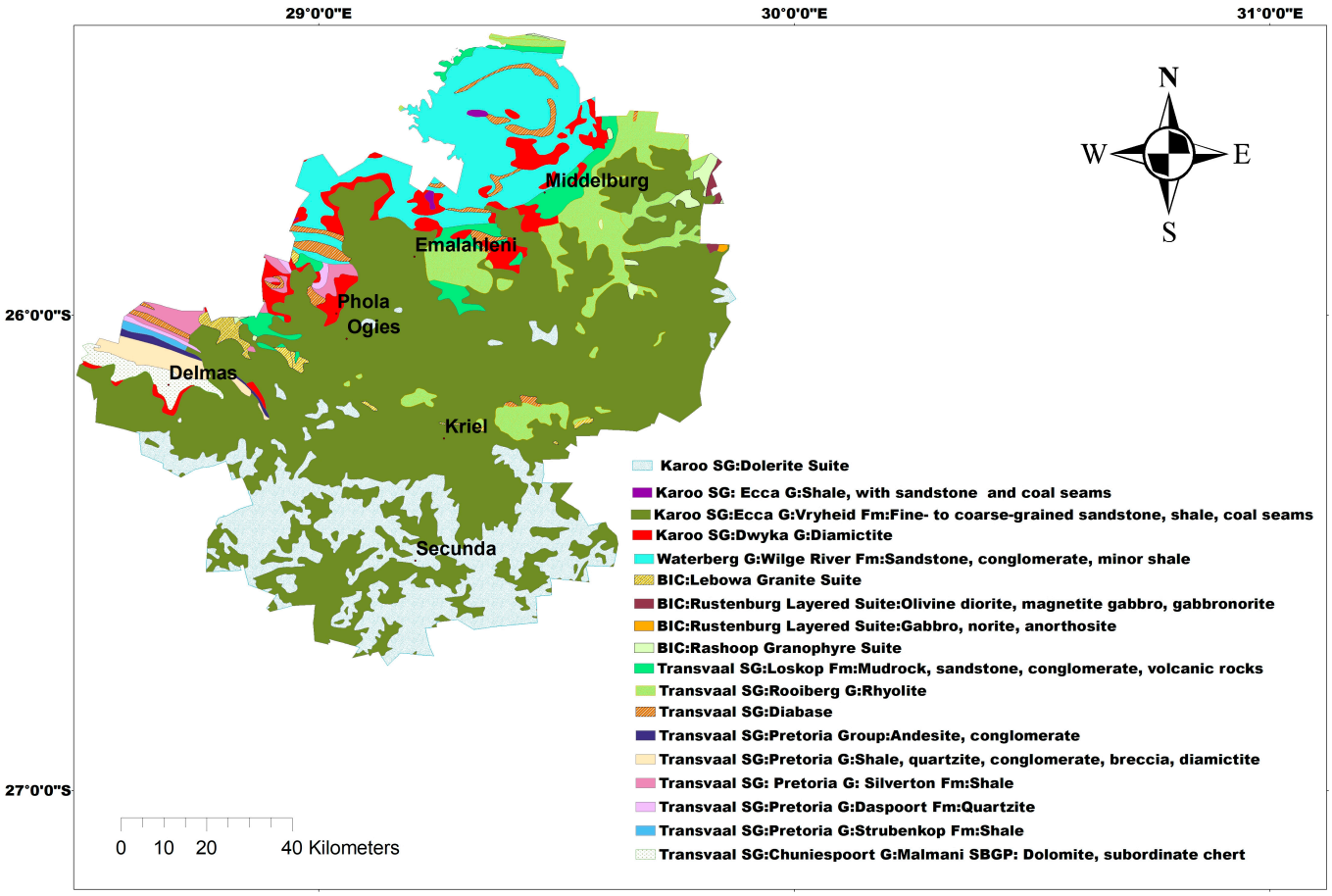

Figure 2. The geology of the area compiled using geological data from Council for Geoscience of South Africa.

2. Materials and Methods

In the Mpumalanga province, indoor radon measurements were conducted in the vicinity of areas dominated by coal operations such as mines, coal-fired power stations, coal liquefaction plants, and coal residual products in order to characterize radon exposure levels and to assess these levels against reference levels recommended by different advisory bodies. The study used mainly a door-to-door approach and referrals to randomly select houses within the targeted area. The measurements were performed predominantly during the cold and warm seasons. During the cold season, the detectors were deployed in early July and retrieved in early October 2022 when the ambient temperature typically ranges between 5 °C and 27 °C [19], whereas, for the warm season, the measurements were conducted from late October 2022 to early March 2023 when the temperatures fall between 14 °C and 28 °C [19]. In every household two detectors were deployed except in one-room dwellings where only one detector was placed. During the cold season, a total of 156 detectors were placed in 83 dwellings, out of which 118 detectors were successfully retrieved and analyzed. During data analysis, 4 of the 118 analyzed detectors were excluded due to incorrect exposure as they were covered during the measuring period, therefore, resulting in the total sample of 114 detectors representing 64 dwellings (Table 1). During the summer months, a total of 74 radon detectors were deployed in 38 dwellings, out of which 59 detectors from 33 dwellings were successfully recovered and analyzed. However, 2 detectors from 1 dwelling were excluded during data analysis due to incorrect exposure, therefore, resulting in the total sample of 57 detectors representing 32 dwellings (Table 1). To observe seasonal variations under the same conditions, 26 dwellings were assessed

during warm and cold seasons. This was to ensure that the seasonal effect is assessed under the same geographical conditions, house characteristics, and living habits. As can be observed, full recovery of the deployed detectors was not achieved because some of the radon detectors were lost or damaged by homeowners. The recovery rates of 76% and 80% were achieved during winter and summer measuring periods, respectively.

Table 1. Statistical summary of the indoor radon measurements performed in the Mpumalanga province during winter and summer months.

Winter Indoor Radon Measurements							
Province	**Area**	**Statistical Summary of Radon Concentrations (Bq/m^3)**					
		No. of measurements (No. of dwellings)	**Min**	**Median**	**Mean**	**Max**	**SD**
Mpumalanga	Middleburg	12 (6)	25	46	51	98	25
	eMalahleni	50 (29)	11	44	42	92	16
	Phola	11 (6)	35	40	42	60	10
	Delmas	8 (5)	27	39	42	56	12
	Kriel	10 (6)	30	37	43	73	17
	Secunda	23 (12)	23	29	39	154	36
	All Areas	114 (64)	11	39	42	154	21
Summer Indoor Radon Measurements							
	Area	**Statistical Summary of Radon Concentrations (Bq/m^3)**					
		No. of measurements (No. of dwellings)	**Min**	**Median**	**Mean**	**Max**	**SD**
Mpumalanga	All areas	57 (32)	16	31	35	135	21

This study used the solid-state nuclear track detectors (SSNTDs) supplied by Radosys, Ltd. (Budapest, Hungary) to measure the radon levels in dwellings. They comprise a CR-39 chip, which is sensitive to alpha particles and operates in such a way that, when alpha particles from radon are generated, they leave microscopic tracks on the chip. Each detector is sealed into a radon-proof bag. The detectors have individual serial numbers for identification. The lowest level of detection (LLD) for the detectors used is about 4.5 kBqh/m^3 (or 0.3 track/mm^2). The measurements were conducted in single-storey houses made of bricks and a few constructed with corrugated iron sheets (shacks). The detectors were placed in highly occupied rooms in the household, such as bedrooms, living rooms, and kitchens. The detectors were positioned at a typical breathing height of 1 m to 1.5 m above the floor, approximately 1 m away from windows, doors, and other potential openings to avoid interferences from outdoor air and about 0.5 m away from the wall. The procedure followed in this study for the deployment of radon detectors aligns with the guidelines outlined by the IAEA Safety Report on Design and Conduct of Indoor Radon Surveys [20], United State Environmental Protection Agency [21], and Health Canada [22]. During the deployment of radon detectors, information related to the characteristics of the dwelling such as the building materials used for construction, ventilation, floor type, and the age of the building structure were collected. The period of exposure for the detectors was 4 months, to obtain average concentration levels and account for temporal changes. After the period of exposure lapses, the radon detectors were collected from the dwellings and submitted to the Radosys Ltd. laboratory in Hungary for analysis. The uncertainty associated with the activity concentration was calculated for each measurement. For all the

measurements reported in this work, the uncertainty values are in a range of 5% and 27%, averaging 11%.

Data Analysis

In dwellings where two measurements were taken, an average value was computed to obtain a representative mean radon concentration of the dwellings. In houses where the measurements were taken in winter and summer months, the values were averaged to obtain representative concentration levels throughout various seasons. The average values were then used for statistical analysis and correlations with various parameters to better understand the distribution and factors influencing indoor radon levels. To compare radon exposure levels in the coalfields and goldfields areas of South Africa, radon measurements that were conducted in gold mining sites of the Gauteng Province during the warm months (October 2021 to January 2022) and cold months (mid-June to early October 2022) were used [23]. This study also used the solid-state nuclear track detectors (SSNTDs) from Radosys Ltd. (Budapest, Hungary) with the lowest level of detection (LLD) of 4.5 kBqh/m^3 (or 0.3 track/mm^2). Similarly to the coalfields, the measurements that were taken in the winter and summer months were averaged to obtain the representative average radon concentration for a dwelling. The averaged values obtained were then used in this study.

3. Results

3.1. Indoor Radon Activity Concentration

The statistical summary of indoor radon measurements performed in the Mpumalanga province during winter and summer months in areas prominent in coal industries and associated coal residual material is presented in Table 1. A total of 171 measurements were conducted in 96 dwellings during both the winter and summer seasons. The results show variabilities between the indoor radon activity concentrations recorded in the winter and summer months. In winter, the mean value of the indoor radon activity concentrations was found to be 42 Bq/m^3, with a maximum value of 154 Bq/m^3, whereas in summer the average and maximum values were 35 Bq/m^3 and 135 Bq/m^3, respectively. To further assess the variations in indoor radon activity concentrations with seasons, the activity concentrations of the 26 dwellings that were measured in both winter and summer months were compared (Figure 3). In general, it was found that the dwellings measured have higher indoor radon activity concentrations during winter months and lower levels in summer (Figure 3). The average ratio of winter to summer indoor radon levels was found to be 1.2. This shows the effect of seasonal differences on indoor radon activity concentrations.

To further understand the variations in indoor radon levels, the concentrations measured in different rooms of the same dwelling for the same time frame were compared (Figure 4). Minor variations were found with an average ratio of 0.99. The slight variation in the measurements within the same dwelling are due to differences in the ingress routes of radon and ventilation rate [20]. This, therefore, indicates that radon levels vary spatially between different rooms in a house, thus, emphasizing the significance of conducting multiple radon measurements in various rooms of the house when assessing indoor radon exposures.

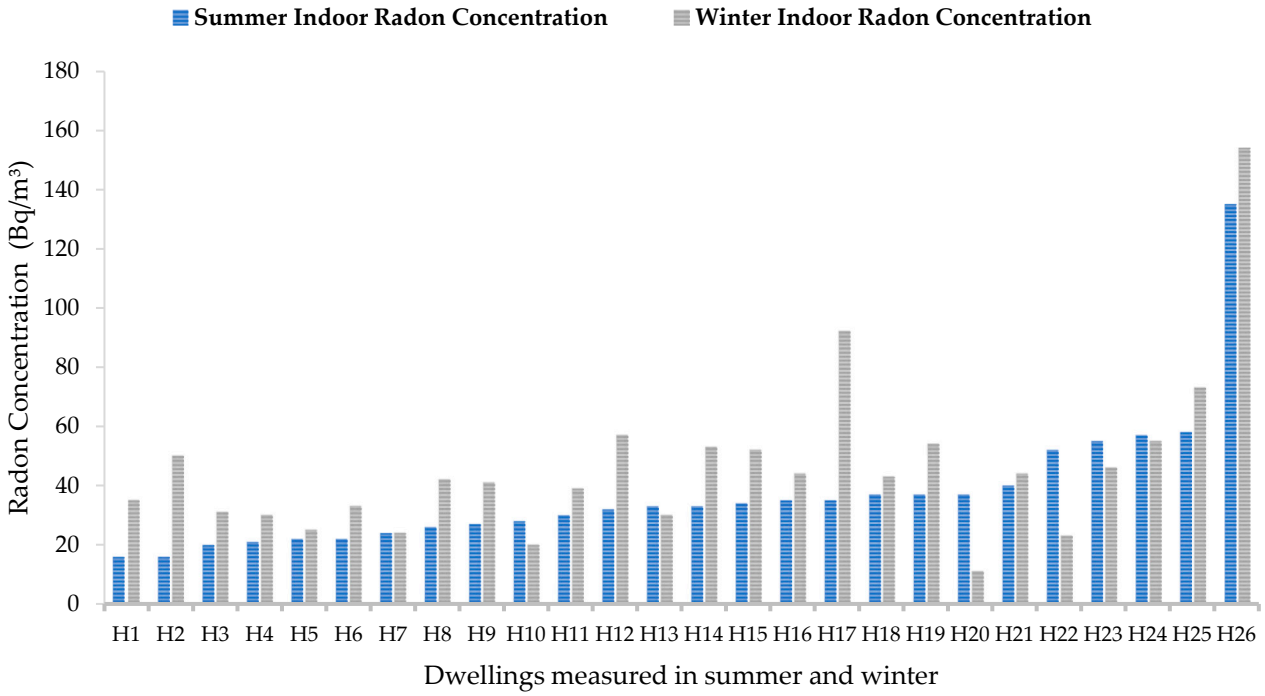

Figure 3. Comparison of summer and winter radon measurements performed in the same dwelling.

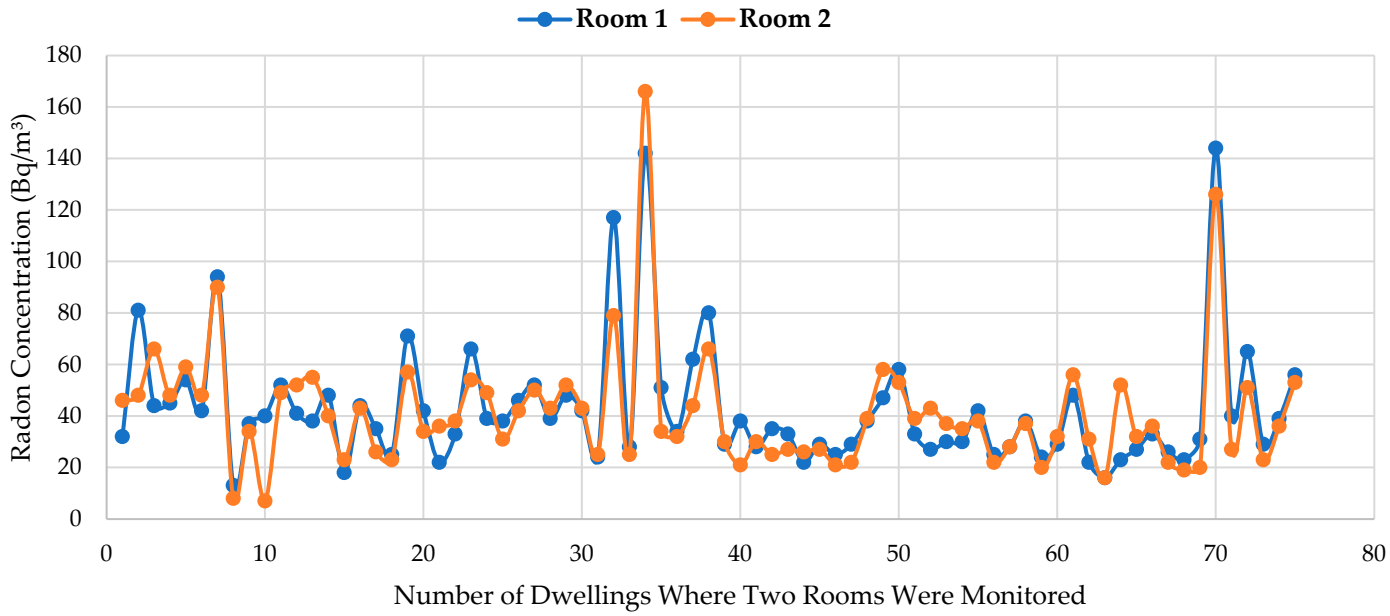

Figure 4. Comparison of radon concentrations found in different rooms in houses with multiple rooms.

To obtain the representative indoor ^{222}Rn concentrations for all the dwellings surveyed in the Mpumalanga Province, the measurements conducted in the same locations during both warm and cold periods were averaged. However, in households where measurements were only conducted during one season, the obtained values were then used as representative concentrations. The average radon concentrations ranged from 21 Bq/m^3 to 145 Bq/m^3, with a mean value of 40 Bq/m^3. Most of the measured dwellings (70%) recorded concentrations that fall in a range between 26 and 50 Bq/m^3 (Figure 5). In comparison with the reference level of 300 Bq/m^3 recommended by the International Commission on Radiological Protection (ICRP) [17] and the International Atomic Energy Agency (IAEA) [18] for

indoor radon exposure, the measured levels are well below the reference level. When the obtained concentrations are compared with the reference level of 100 Bq/m^3 recommended by the World Health Organization (WHO) [14], only one dwelling was found to exceed this value. This dwelling is located in Secunda. Based on the house characteristics information acquired during the survey, it was observed that the house has sufficient ventilation openings, but it is kept closed most of the time as it is not occupied on a regular basis. As such, the extremely low ventilation rate of the house could be the cause of higher indoor radon activity concentrations due to low indoor–outdoor air exchange rate. This demonstrates the effect of ventilation rate on indoor radon activity concentrations.

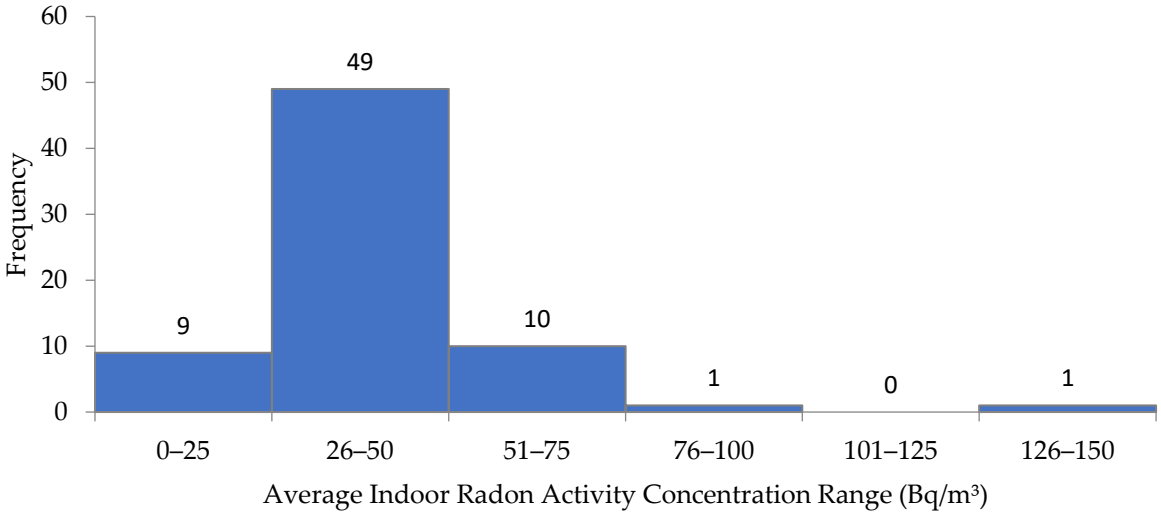

Figure 5. Representative indoor ^{222}Rn concentrations for all the dwellings surveyed in Mpumalanga.

3.2. Comparison with Other Studies

3.2.1. Comparison with Previous Studies Conducted in the Mpumalanga Province

There are two published studies conducted in some parts of the Mpumalanga Province to assess the indoor radon activity concentrations. These studies were performed in Secunda and eMalahleni, and measured indoor radon levels over a short period, for about 3 to 4 days [15,16]. In the Secunda area, an average indoor radon activity concentration of 76.4 Bq/m^3 was recorded [15], whereas in eMalahleni, an average concentration of 122.1 Bq/m^3 was reported [16]. In this study, the measurements conducted in Secunda and eMalahleni during the cold season when higher levels are expected averaged 39 Bq/m^3 and 42 Bq/m$^{3,\text{ respectively}}$ (Table 1). The average concentrations obtained in the current study were found to be lower when compared with those of previous studies. The differences observed between the average values obtained in this study and that of previous studies may be due to various factors such as temporal variations associated with the differences in the measurement periods as well as differences in the location, building materials and occupancy patterns of the houses selected for measurements.

3.2.2. Radon Levels in Coalfields vs. Goldfields

The mining and processing of economic minerals such as gold and coal combustion are typically known to contribute to radon levels in the environment [5,24]. In the coal mining areas studied in this work, it was found that the overall activity concentrations ranged from 21 Bq/m^3 to 145 Bq/m^3, with a mean value of 40 Bq/m^3 (Table 2). In the gold mining areas, the average concentrations for all homes ranged from below detection limit to 96 Bq/m^3, with a mean value of 33 Bq/m^3 (Table 2). It can be observed from Figure 6 that there is no significant variation in the distribution trend of indoor radon activity concentrations in both

the coal and gold mining areas. In both areas, most of the households have indoor radon activity concentrations in a range of 26 to 50 Bq/m^3. However, in the gold mining areas a slight peak in the number of dwellings (38%) that falls in the lower concentration range of 0–25 Bq/m^3 is observed as compared to coal industrial areas (12%). This could be because in the gold mining areas most of the measurements were taken during summer months when the ventilation rate of the dwellings is high whereas in coal mining areas most of the measurements were taken predominantly in winter months. In general, the distribution pattern follows the same trend (Figure 6), and the average concentrations recorded in both sites in almost all homes do not exceed the proposed level of 100 Bq/m^3 recommended by WHO for indoor radon exposures [14] and are all well below the ICRP and IAEA reference level of 300 Bq/m^3, Table 2. The average values of 40 Bq/m^3 and 33 Bq/m^3 obtained in the coal and gold mining sites, respectively, are comparable with the worldwide average indoor radon activity concentration of 39 Bq/m^3 [14].

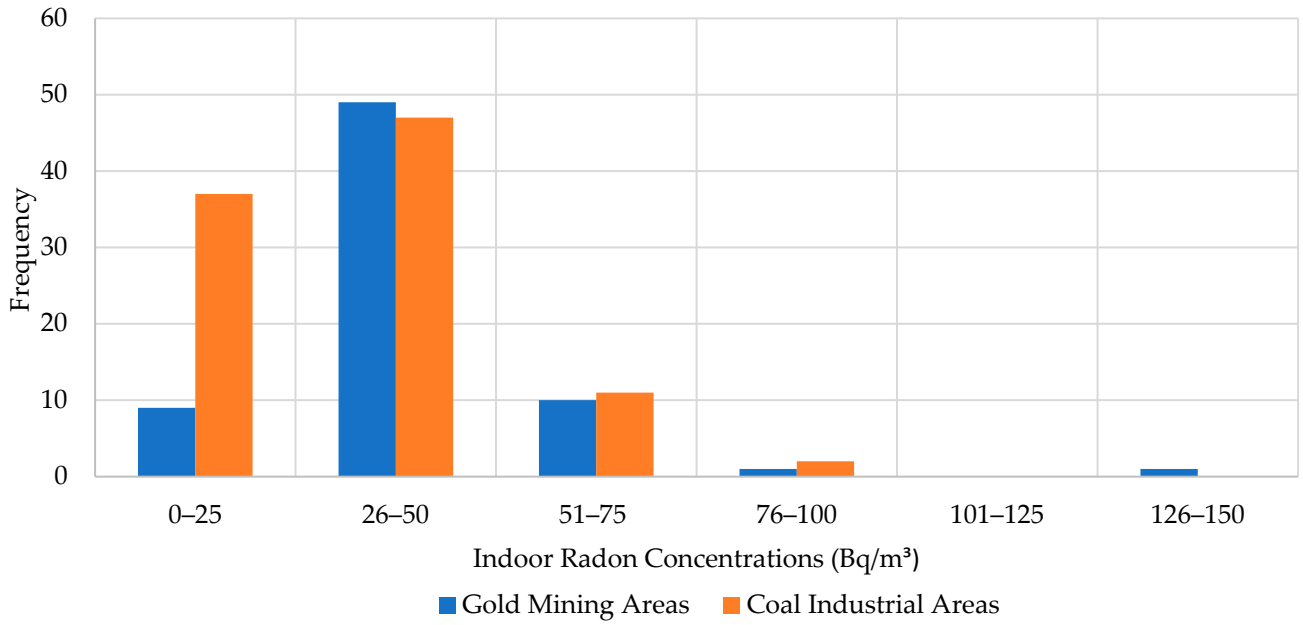

Figure 6. Comparison of indoor radon activity concentrations found in dwellings in the vicinity of the coalfields of the Mpumalanga province and goldfields of the Gauteng province.

Table 2. Statistical summary of indoor radon results obtained in the coalfields and goldfields areas. LLD stands for Low Level of Detection.

Class Interval	Frequency (Coal Industrial Areas)	Frequency (Gold Mining Areas)
0–25	9	37
26–50	49	47
51–75	10	11
76–100	1	2
101–125	0	0
126–150	1	0

Table 2. *Cont.*

Class Interval	Frequency (Coal Industrial Areas)	Frequency (Gold Mining Areas)
Statistical Summary (Bq/m^3)		
No of measurements	70	97
Min	21	<LLD
Max	145	96
Mean	40	33
Median	35	31
SD	19	17

3.3. Evaluation of Main Indoor Radon Sources and Controls

To evaluate the impact of coal industrial operations and residues on indoor radon activity concentration, the associated concentrations found in houses located at various distances from coal impacted areas were assessed. The findings show that there is no definite trend observed on indoor radon levels with increasing or decreasing distance from the coal operations with a very weak correlation of R^2 = 0.0005 (Figure 7). There is no significant variance in the radon concentration range for houses that are located in the immediate vicinity of coal industries and those that are further away. Moreover, the dwelling that recorded the highest activity concentration of 145 Bq/m^3 is found to be located further away from coal industrial areas and residues (>5 km). Based on the findings presented in Figure 7, it is clear that the numerous coal mines, coal-fired power stations and residual material found in the Mpumalanga province do not contribute significantly to the indoor radon activity concentrations in the six main residential areas located in the vicinity of these operations. The findings of the long-term measurements presented in this paper corroborate with earlier studies performed using short-term measurement techniques in eMalahleni and Secunda, which found that the fallout from the coal industrial operations surrounding these towns does not enhance the activity concentration of radon [15,16].

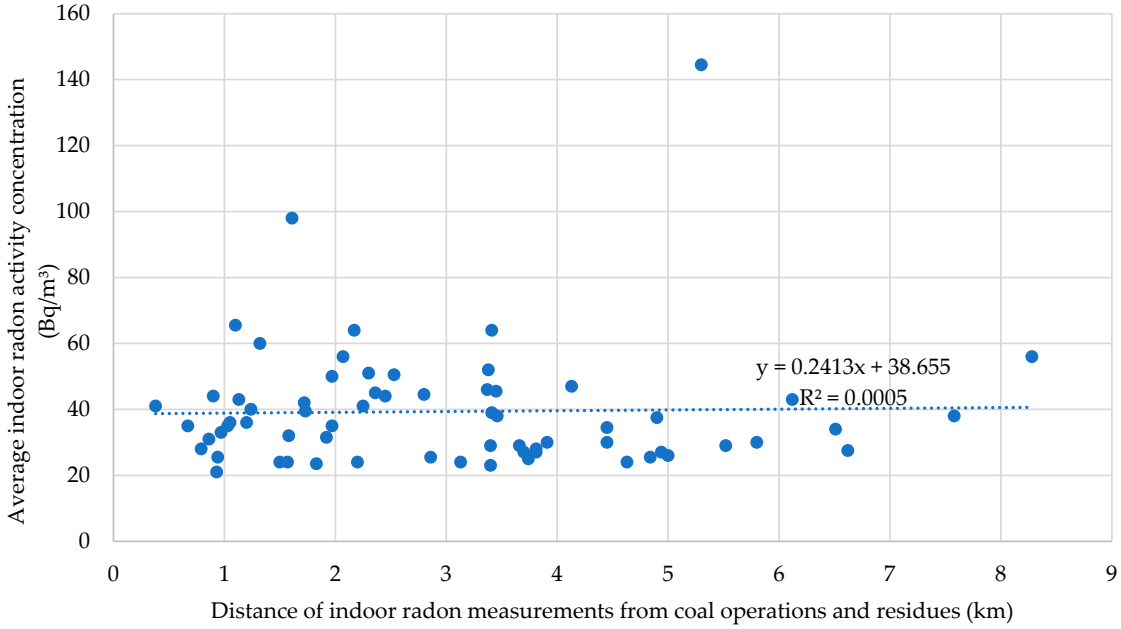

Figure 7. Correlation of indoor radon results with distance from coal operations.

3.4. Influence of Geology on Indoor Radon

To assess the influence of geology on indoor radon levels, the concentrations associated with various geological formations were analyzed. The analysis was performed using Geographic Information Systems (GIS) by overlaying the measured indoor radon data with the geological map presented in Figure 2 to identify the geological units found beneath the dwellings that were monitored. Thereafter, dwellings that fall within the same geological formations were grouped, and statistical analysis was performed to observe trends in activity concentrations associated with various rock types. Overall, the higher radon average values are associated with areas underlain by mudrocks, shale, rhyolite, and diamictites. These are rock types commonly known to contain higher levels of uranium [25]. The higher indoor radon levels associated with the Dwyka diamictite may be due to the rich phyllosilicate minerals found in its matrix [26], which have a high affinity to absorb uranium isotopes [27]. On the other hand, the rock types with radon concentrations in the lower range are sandstones, dolomite, and dolerite (Figure 8). It can be observed in Figure 9 that the rock types associated with lower indoor radon activity concentrations contain low levels of uranium and those with higher radon activity concentrations contain higher uranium levels. There is a general increasing trend in indoor radon activity concentrations with increasing uranium concentrations in rocks (Figure 9). This is expected as the higher the uranium concentration in an area, the greater the probability of high radon accumulation in indoor air [25]. These findings reveal that the uranium content found in rocks has a major impact on radon levels found in dwellings across various areas.

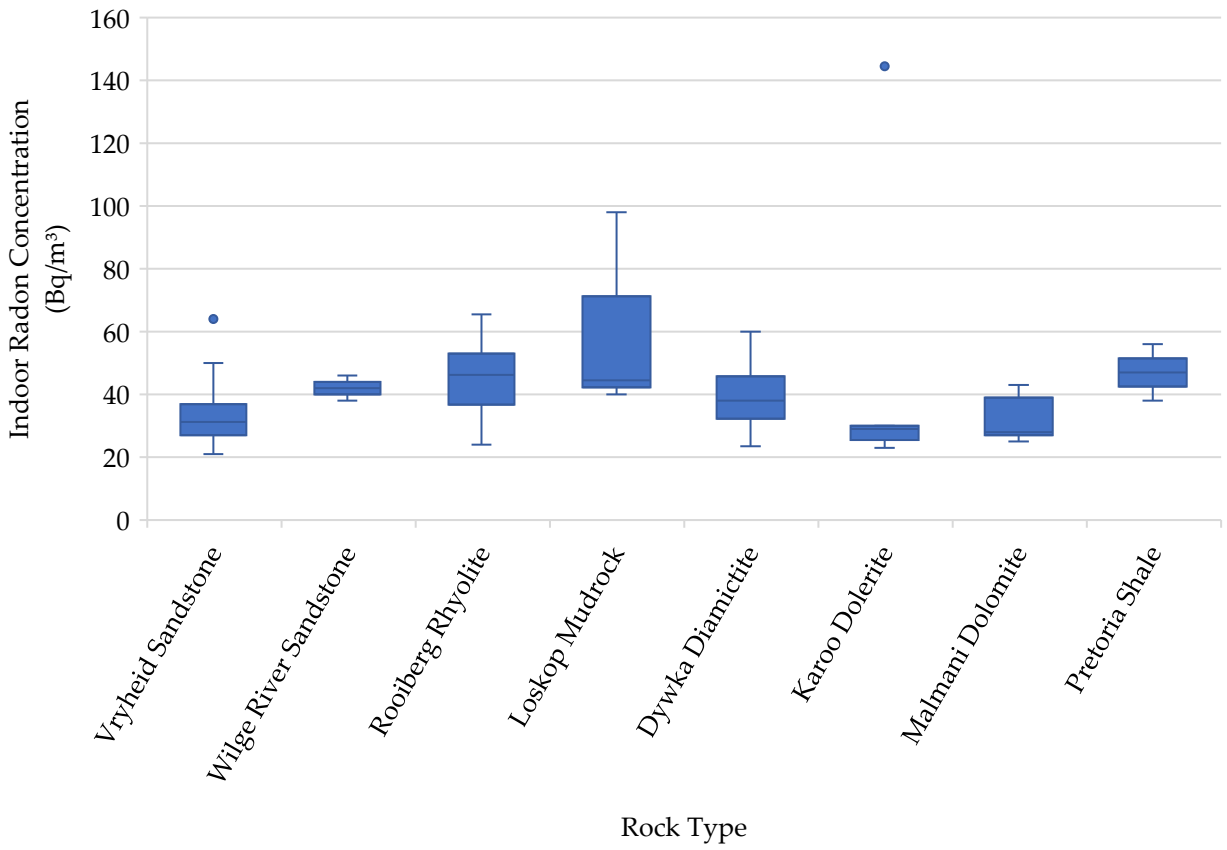

Figure 8. Correlation of indoor radon with the underlying geological formation found in the study area.

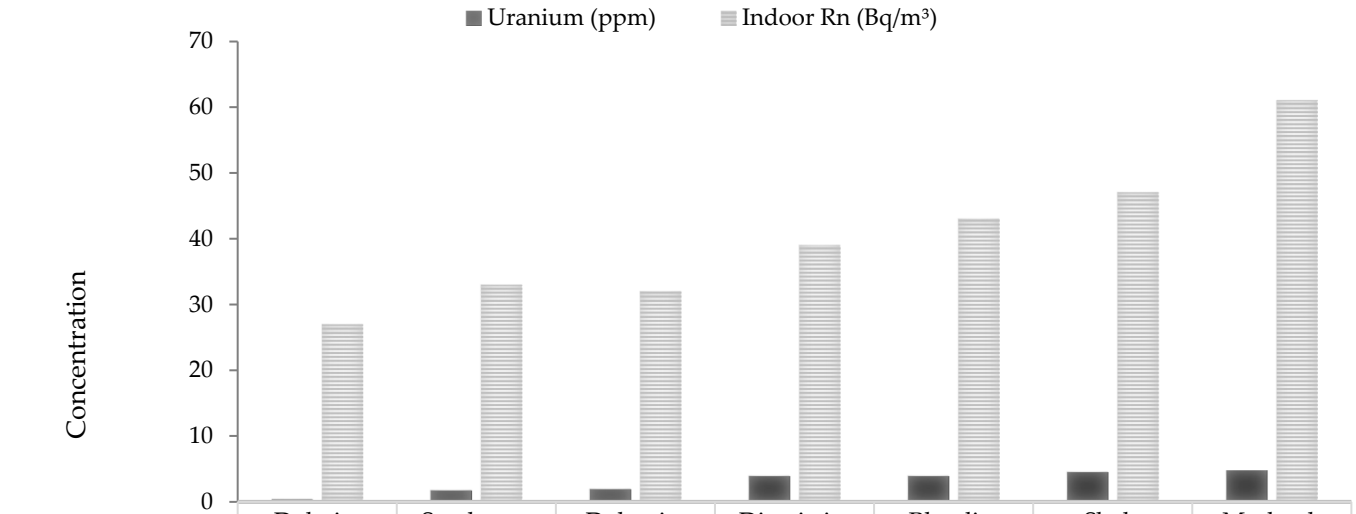

Figure 9. Correlation of indoor radon activity concentrations found in the coal industrial areas surveyed in this study with the uranium concentrations in rocks reported by Bezuidenhout (2021) [28].

4. Discussion

Radon activity concentrations in buildings result from a combination of different sources and depend on multiple factors. This study evaluated radon exposure levels in residential areas in the vicinity of coal operations in the Mpumalanga province of South Africa. The findings revealed that the indoor radon activity concentrations found in dwellings located in the vicinity of coal industrial operations do not indicate elevated concentrations. There is no correlation observed between indoor radon levels and the proximity of dwellings to coal industrial operations. This, therefore, implies that the emissions from coal operations found in the study area do not result in significant indoor radon levels in the homes that were surveyed. It could be that the materials used for house construction and the underlying geology are the major contributing factors to indoor radon levels.

Based on the findings of this study and the recent study that was conducted in gold mining sites [23], it can be observed that the radon emissions released from these anthropogenic sources do not contribute significantly as major sources of indoor radon. The only condition where these anthropogenic sources had an impact on indoor radon activity concentrations was in a case where the dwelling was found to be situated on soils mixed with tailings materials [23]. The impact may also be observed in instances where mining residues such as tailings are used as construction materials; however, such instances were not encountered in these two studies.

In both studies, there is no correlation in indoor radon activity concentrations with the distance of dwellings from the mining operation sites. In the gold mining sites, the radon parent radionuclide, ^{238}U, ranges between 209.95 and 2578.68 Bq/kg in tailings materials in the vicinity of the surveyed homes [29], and in the coalfields the average uranium concentration of 2.24 mg/kg (27.66 Bq/kg) and radium-226 (^{226}Ra) of 21.69 Bq/kg were found in the coal used in some of the coal-fired power plants in the Mpumalanga province [12]. Despite the differences in the activity concentrations of the radon parent

radionuclides found in the NORMs in these two sites, the radon levels found in homes were comparable (Table 2 and Figure 6). Thus, indicating that there is no clear relationship between indoor radon activity concentrations and these mining operations.

The general trend that is deduced from this work and the study conducted in gold mining sites [23] is that the rocks and soils found in the area serve as a primary source of indoor radon. Based on these findings, it is deduced that sources that contribute significantly to indoor radon activity concentrations are those that are directly in contact with or beneath the house to permit direct ingress of radon gas into the dwelling. On the contrary, there is no significant impact on indoor radon from sources that are not in contact with dwellings. This indicates that once radon is released into the atmosphere from mining operations and residues, its dispersion is primarily controlled by the effects of atmospheric winds [30]. In addition, it may be that the confined nature of building structures provides a barrier and constrains the ease of entry of radon from outdoor air. As such, to thoroughly assess the radon exposures associated with the NORM related industrial activities, it is important to also conduct outdoor measurements to quantify if there are any potential health hazards due to outdoor radon exposures and to better understand the extent of radon dispersion from these sources. It is also noted that the variations in indoor radon activity concentrations are dependent on the ventilation conditions of the dwelling, which is dependent on seasonal temperature changes and household occupant's behaviour. This observation was made following that higher radon concentrations were found in winter (Figure 3) when the ventilation rate is low and in a house that remains closed for the most part of the year as it is not occupied on a regular basis.

5. Conclusions

This study conducted a regional indoor radon survey in residential areas in the vicinity of coal operations in the Mpumalanga province to assess radon exposure levels against reference levels recommended by different advisory bodies. The measured indoor radon activity concentrations are well below the proposed international reference levels, with only one dwelling exceeding the WHO recommended value of 100 Bq/m^3. It was found that the coal operations and their associated residues do not result in significant radon levels in the dwellings located in their vicinity. The rocks and soils found in an area besides materials used for house construction serve the primary sources of indoor radon, and the ventilation rate of a building controls the levels that accumulate. It is recommended that future indoor radon studies should also assess contributions from building materials to better identify the primary significant source of indoor radon levels.

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Abbreviations

The following abbreviations are used in this manuscript:

NORM	Naturally Occurring Radioactive Materials
SSNTDs	Solid State Nuclear Track Detectors
LLD	Low Level of Detection

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