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Determination of Natural Radioactivity in the North East Beach Sands of Madagascar

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Abstract: Exploration and exploitation of radioactive ores (ilmenite, zircon and monazite) are considered as the main source of exposure to ionizing radiation of the population living in the coast of Analanjorofo Region (Fénérive-Est Districts, Rural municipality Ampasimbe Manantsatrana). Radioactivity measurements have been performed in this region. The distribution of natural radionuclide gamma-emitters (^{238}U, ^{232}Th and ^{40}K) and their respective annual effective dose rates have been determined for sand(s), water and air. The samples have been collected along and around the coast of the region. The radiation emitted from the natural radionuclide containing in the environmental samples has been determined by gamma spectrometry system and Radon meter (SARAD). Exposure dose rates at 1 m above the ground have been measured along the coast and in the villages around the exploitation sites. Dose rate measurements have been performed by Dosimeter Graetz X5 DE, equipped of Geiger Müller Counter. Results have been compared with the reference values provided by IAEA (BSS 115) and UNSCEAR 2000.

Keywords: Coastal Sands, ^{238}U, ^{232}Th and ^{40}K Concentrations, Annual Effective Dose Rate, Gamma Spectrometry, Geiger Müller Counter

1. Introduction

Natural radioactivity represents the main source of human exposure to ionizing radiation. Public and workers are exposed to natural radionuclides from the environment via external and internal pathways. The terrestrial gamma rays are essentially derived from ^{40}K and radionuclides belonging to ^{238}U and ^{232}Th series that are present in the earth's crust [6].

The study of the distribution of primordial radionuclides allows the understanding of the radiological contribution of these elements due to the γ -ray exposure of the body and irradiation of lung tissues from inhalation of radon and its daughters. It represents a significant asset to the assessment of the radiation hazards associated with the exploitation of radioactive ores. Hence, the assessment of gamma radiation dose from radioactive ores presents a particular importance to study the contribution of external and internal doses for public around the sites and workers in the sites. The dose rates vary depending upon the concentration of the natural

radionuclides such as ^{238}U and ^{232}Th series, and ^{40}K present in water, sand and air [5].

The objective of this study is to evaluate the natural levels of radioactivity from radioactive ores in the study areas and surrounding villages; to quantify the impact of activities related to the exploitation of radioactive ores; to ensure the radiological protection of workers in the site, handling radioactive substances, and publics living around the sites.

Laboratory measurements are performed by gamma spectrometry system using NaI(Tl) detector in order to determine the natural radionuclide activities in soil, sand and water samples. The annual effective dose rates and external hazard indexes have been evaluated and compared to the limits proposed by United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2000) and the measurements of dose rate have been carried out using GRAETZ X5 DE Ratemeter equipped with Geiger Muller detector.

2. Location and Delimitation of Study Site

The investigated area is located in the north-east of Madagascar, Region of Analanjirofo, District of Fenoarivo Atsinanana, Rural Commune of Ampasimbe (Figure 1).

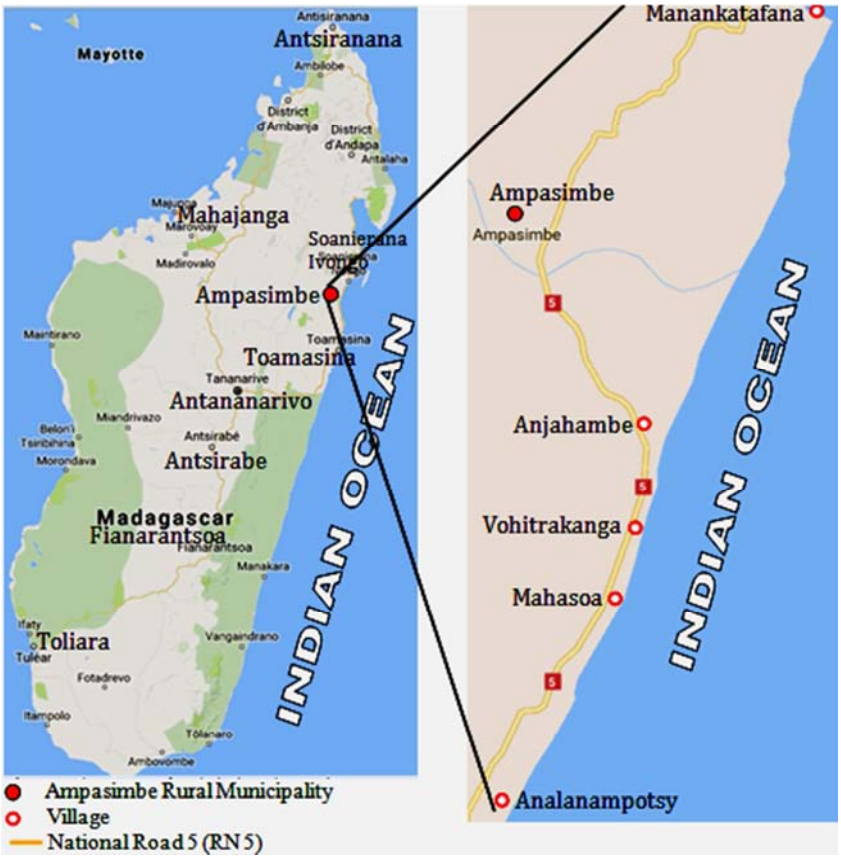


Figure 1. *Location of study site.*

Several villages have been studied such as Analanampotsy, Mahasoa, Anjahambe, Manankatafana and Vohitrakanga. In addition, the study has been perfomed in all village surrondings in order to rake an approximate area.

3. Materials and Methods

3.1. Dosimetry Methodology

Measurements have been carried out using GRAETZ X5 DE ratemeter equipped with Geiger Muller detector type whose the measuring range for gamma radiation varies from 0.06 µSv. h^{-1} to 20 mSv. h^{-1}. The ratemeter detection limit is 0.06 µSv. h^{-1}, values below this limit are not detected. Measurements have been performed at 1 m above the ground. In each point of measurement, the geographical coordinates were recorded using a Global Positioning System (GPS) device. In situ, dosimetric measurements were carried out at a variable mesh depending on the expanse of the area of the map. The obtained longitude and latitude were converted in plan coordinates, using Laborde projection.

3.2. Sampling Strategy

3.2.1. Soil Samples

Soil samples were taken randomly and in the places where measured dose rates were relatively high compared with the mean dose rate value. The samples were put into plastic bags and the quantities varied from 0.5 kg to 1.0 kg. Each point is recorded by GPS device. The samples are analyzed in terms of radioactivity.

3.2.2. Water Samples

Water samples have been collected in streams, in rivers and underground water. These waters are used by the population as household and drinking water. The samples have been put into plastic bottles of 1.5 L. The geographical coordinates were identified using GPS device.

3.2.3. Radon in Air

Measurements of radon were performed in-situ by radon-meter RTM-2100 SARAD. This equipment is able to measure the radioactive gas concentration, especially radon-222. Measurements were carried out inside and outside of sites, in high occupation factor premises or in the populations who live nearby. The affected villages are located within the sites or its surroundings. Measurement times vary from 12 hours to 24 hours depending of the radon concentration level. Similarly, the geographical coordinates of sampling are identified using GPS device. The obtained results were compared with the reference limits, which are 400 Bq. m^{-3} for the public and 1,000 Bq. m^{-3} for the workers.

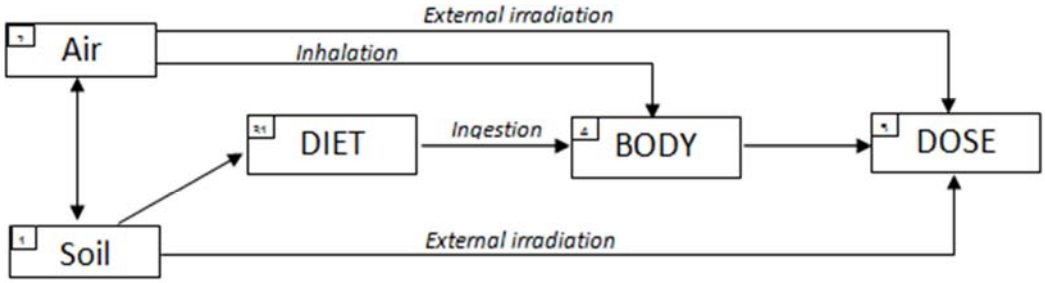
3.3. Laboratory Works

All samples were analysed at the "Institut National des Sciences et Techniques Nucléaires" (INSTN – Madagascar). During sample preparation, soil samples were oven-dried at 80°C, grinded, sieved, homogenized and placed into 100 cm^3 polyethylene cylindrical containers. These containers were hermetically sealed during one month to reach equilibrium of ^{226}Ra and its daugthers before analysis. While water samples were directly put in 500 cm^3 Marinelli beakers. Analyzes were performed by gamma spectrometry system, with NaI (Tl) detector. The samples have been counted between 12 to 24 hours. The equipment is able to measure qualitatively and quantitatively of emitted gamma-rays. The obtained results were compared to the global averages of the same sample types, which are given in reference documents such as UNSCEAR 2000. These values have been used to estimate the minimum and maximum dose rates related to the external exposure, the inhalation and the ingestion.

3.4. Exposure Dose Assessment

3.4.1. Concept

The basic standard model (UNSCEAR 2000) is used to determine the transfer pathway and to evaluate the dose rates related to the environment natural radioactivity (Figure 2).



Source: UNSCEAR, 2000

Figure 2. *Terrestrial pathways of radionuclide dose transfer.*

The main human dose rates are produced by the internal (inhalation and ingestion) and the external exposures. These dose rates proceed mainly from the soil, the air and the food.

3.4.2. Estimation Method

The estimation of radiation exposure consists to calculate the doses caused by the concentration of radionuclides in various environmental patterns. The calculation is based on the activity determination. For people living and working in the exposed area, the annual dose rates were determined such as soil and rock radioactivities, internal exposure of drinking water and internal exposure of radon inhalation. Ingestion and inhalation are the most important pathways for the migration of radionuclides inside the human body.

Estimation dose rates are calculated according to the three following hypotheses:

1. For external exposure, the annual working duration is 8 hours per day, 5 days per week and 52 weeks. The outdoor absorbed dose rate ($\dot{D}$), at 1 m above the ground level, is calculated using equation 1 [1, 2, 3].

$$\dot{D}\ (\mathrm{nGy.h^{-1}}) = (0.462 \times C_U) + (0.604 \times C_{Th}) + (0.0417 \times C_K) \quad (1)$$

where 0.462, 0.604 and 0.0417 are the dose coefficients (nGy. h^{-1} per Bq. kg^{-1}) for ^{238}U, ^{232}Th and ^{40}K respectively. C_U, C_{Th} and C_K are the activity (Bq. kg^{-1}) of ^{238}U, ^{232}Th and ^{40}K respectively.

The annual effective dose rate per year is calculated by the following relation (2):

$$\dot{D}(\mathrm{mSv.y^{-1}}) = \dot{D}\ (\mathrm{nGy.h^{-1}}) \times (8 \times 5 \times 52) \times 0{,}7 \times 10^{-6} \quad (2)$$

UNSCEAR 1993 reported, the conversion coefficient for adult is 0.7 Sv. Gy-1 [4].

2. For ingestion, the annual average consumption is about 500 liters. The estimation of dose rate is calculated from

226Ra activity because of its dissolution in water. The annual effective dose rate is given by the following formula (3):

$$\dot{D}\,(\mathrm{mSv.y^{-1}}) = (0.28 \times C_{Ra}) \times 500 \times 10^{-3} \quad (3)$$

where 0.28 is the conversion factor (µSv. Bq^{-1}) of ^{226}Ra and C_{Ra} is the activity (Bq. L^{-1}).

3. For inhalation, the annual duration in confined place is 7000 hours. The annual effective dose rate is calculated by the following formula (4):

$$\dot{D}\,(\mathrm{mSv.y^{-1}}) = C_{Rn} \times 0.4 \times 7000 \times 9 \times 10^{-6} \quad (4)$$

where C_{Rn} is the ^{222}Rn concentration (Bq. m^{-3}). The dose conversion factor is 9 nSv/Bqh. m^{-3} [U3, U4] which it is still considered as appropriate calculation of average effective dose. According to UNSCEAR 2000, the indoor equilibrium factor is 0.4.

The exposure doses are reported in minimum and maximum values.

The calculated values of the total absorbed dose rate of the all samples (soil, water and air) measured are represented in *Table 7*.

4. Results and Discussions

4.1. Dosimetric Results

In total, 272 ambiante dose rate points have been measured of which 250 points spreaded inside of mining area and 22 points in Analanampotsy storage zone of ilmenite and zircon ores (*Table 2*). In addition, five villages arround the mining zone have been also performed such as Rural Commune of Ampasimbe Manantsatrana (*Table 1*). The minimum and maximum values have been gathered. The ratemeter detection limit is up to 0.06 µSv. h^{-1}, in which values below this limit cannot be measured. However, these low values do not present hazards either to the public or to workers. The measured exposure dose rates vary from 0.06 µSv. h^{-1} to 16.20 µSv. h^{-1}. The values show that environmental radioactivity levels in the studied areas vary according to the geological formation of the environment. In comparison with regulatory limits, measured dose rates are below the limit values which are 2.5 µSv. h^{-1} for public and 7.5 µSv. h^{-1} for worker [7, 8, 9].

At Analanampotsy village where the zircon and ilmenite ores have been stored, the dose rates exceeded the limit values. The maximum dose rates of 2.80 µSv. h^{-1} and 16.20 µSv. h^{-1} were found in ilmenite and zircon storages respectively. The last value is higher than the controlled area limit value of 7.5 µSv. h^{-1} [7, 8, 9].

Table 1. *Exposure dose rates measured in mining sites and in villages at Rural Commune of Ampasimbe Manantsatrana.*

Location	Number of measurements (Village/Site)	Minimum value (µSv. h^{-1})	Maximum value (µSv. h^{-1})
Analanampotsy	88 (23/65)	0.07	0.48
Anjahambe	72 (42/30)	< 0.06	0.32
Mahasoa	28 (28/00)	0.07	0.24
Vohitrakanga	21 (21/00)	0.06	0.13
Manankatafana	41 (25/16)	< 0.06	0.27
Total	250 (139/111)	-	-

Table 2. *Limit values of exposure dose rates measured in temporary storage of mining sites.*

Location	Number of measurements	Storage area of ilmenite ore		Storage area of zircon ore	
		Minimum value (mSv. h^{-1})	Maximum value (mSv. h^{-1})	Minimum value (mSv.h^{-1})	Maximum value (mSv.h^{-1})
Anjahambe	-	-	-	-	-
Analanampotsy	22	0.20	2.80	11.80	16.20
Mahasoa	-	-	-	-	-
Vohitrakanga	-	-	-	-	-
Manankatafana	-	-	-	-	-
Total	22	0.20	2.80	11.80	16.20

Tables 3. *Minimun and maximum values of specific radioactivity of soil samples.*

1. Site of Analanampotsy

Number of samples	Radionuclide	Activity (Bq. kg^{-1}) (content)	
		Minimum	Maximum
07	Potassium	327 ± 16 (1.01 ± 0.05) %	1 045 ± 117 (3.23 ± 0.36) %
	Thorium	910 ±64 (224 ±16) ppm	87 160 ± 28 872 (21 484 ± 7 117) ppm
	Uranium	437 ± 99 (35 ± 8) ppm	17 295 ± 6 779 (1 400 ± 549) ppm

2. Site of Anjahambe

Number of samples	Radionuclides	Activity (Bq. kg^{-1}) (content)	
		Minimum	Maximum
03	Potassium	222 ± 18 (0.7 ± 0.07) %	616 ± 31 (1.9 ± 0.1) %
	Thorium	3505 ± 255 (864 ± 63) ppm	11 004 ± 930 (2 712 ± 229) ppm
	Uranium	1123 ± 253 (91 ± 20) ppm	5 034 ± 1 166 (408 ± 94) ppm

3. Site of Manankatafana

Number of samples	Radionuclides	Activity (Bq. kg^{-1}) (content)	
		Minimum	Maximum
02	Potassium	161 ± 7 (0.5 ± 0.02) %	343 ± 27 (1.1 ± 0.08) %
	Thorium	1 574 ± 112 (388 ± 28) ppm	15 391 ± 1 328 (3 794 ± 327) ppm
	Uranium	444 ± 99 (36 ± 8) ppm	3 199 ± 738 (259 ± 60) ppm

Table 4. *Reference values in the specific radioactivity in soils.*

Radionuclide	Average value (Minimum – Maximum)	
Potassium	400 (140 – 850) Bq. kg^{-1}	1.2 (0.4 – 2.6) %
Thorium	30 (11 – 64) Bq. kg^{-1}	7 (3 – 16) ppm
Uranium	35 (16 – 110) Bq. kg^{-1}	3 (1.3 – 8.9) ppm
Th/U ratio	Roches primaires	3.5 (3.5 – 6.3)
	Soils and sands	5 (1.5 – 21)

Source: UNSCEAR 2000 and J. J. W. ROGERS, J. A. S. ADAMS – Handbookof Geochemistry

4.2. Radioactivity of Water Samples

Table 5. *Specific Radioactivity in water samples.*

Number of samples	Radionuclides	Activity (Bq. L^{-1})	
		Minimum	Maximum
08	Potassium	6.9 ± 0.2	36.2 ± 1.4
	Thorium	3.4 ± 0.2	17.7 ± 1.2
	Uranium	0.6 ± 0.1	19.5 ± 4.4

The results show the normal values of specific radioactivity for the water samples. It can be deduced that the radionuclides contained in material radioactive ores in the investigated area not contaminate the surrounding waters. However, special precautions must be taken into consideration.

4.3. Specific Radioactivity of Radon from Air

Summary values of the emanation of radon in air are showed in Table 6.

The measurement values are given in average concentrations of which all are below of reference value. Similarly for acceptable values of public of 400 Bq. m^{-3} and worker of 1000 Bq. m^{-3}, the measured values are relatively low. On the one hand, this can be explained that the existence of constantly sea wind blows along the coastline and contributes to scatter of radioactive gases in suspension; on the other hand by the natural ventilation of local dwellings. Modern architecture of buildings in the camp (floors, sealed walls) also contributes to attenuate the emanation phenomenon of radon gas from the soil surface.

Table 6. *Specific radioactivity of radon gas in air.*

Code	Counting time (hour)	Concentration (Bq. m^{-3})		
		Average	Minimum	Maximum
R01 / Anjahambe	12	< 1.2	-	< 1.2
R02 / Anjahambe	12	8.1 ± 3.1	7.7 ± 1.2	15 ± 2
R03 / Anjahambe	24	2.6 ± 1.4	2.3 ± 1	16 ± 2
R04 / Anjahambe	12	7.1 ± 3.6	3.2 ± 1.6	10 ± 1
Reference value	-	46	-	480

4.4. Minimum and Maximum Exposure Doses

The estimated Internal and External exposure doses for humans staying and working on sites or its surroundings are calculated from the environmental samples. The summary results are given in *Table 7*.

Table 7. *Minimum and maximum of calculated exposure dose rates.*

Origin	Exposure pathway	Calculated annual dose rate (mSv.y^{-1})	
		Minimum	Maximum
Soil	External	1.1 ± 0.0	87.88 ± 12.93
Water	Internal (ingestion)	0.08 ± 0.0	2.73 ± 0.62
Air	Internal (inhalation)	0.03 ± 0.0	0.40 ± 0.05
Total		1.21 ± 0.0	91.01 ± 12.91
Reference value	1 - 20		

In assumption of a person constantly working near of high radioactive level, the estimated maximum value of annual exposure dose rates is (91.01 ± 12.91) mSv. y^{-1}. This rate is calculated from the zircon samples, the temporary storage and the current operation sites. The obtained value is widely higher than the annual limit value of 20 mSv. y^{-1} for workers [7, 8, 9].

5. Conclusion

The exposure dose rate values vary from 0.06 µSv. h^{-1} to 16.20 µSv. h^{-1}. The measured exposure doses inside and around the villages are below the acceptable limit value for the public. In the mining sites and outside of storage areas, values do not exceed the controlled area limit value for workers. In storage areas, some values exceed the controlled area limit value. The maximum value was obtained in the temporary storage area of zircon mining site of Analanampotsy.

For soil sample measurements, the specific activities values are higher than the world reference given by the *Table 4*.

For water sample, the specific activities values are relatively in the order of world reference magnitude.

In-situ, radon gas measurements show that the average concentrations in the air are below the reference value provided by the *Table 6*. Taking the assumption that a person is staying constantly in the vicinity of high radioactivity area, the maximum exposure rate is (91.01 ± 12.91) mSv. y^{-1}. Such value is higher than the regulatory limit for workers (20 mSv. y^{-1}) [7, 8, 9].

In comparison of available data, the obtained dose rate levels in the all investigated sites are lower than the beach sand from Guarapari (Brazil) 90 µSv. h^{-1} (UNSCEAR 2000).

Appendix

Table A1. *Radioactivity of soil samples.*

Code	Activity (Bq. kg-1)			Content (ppm)		
	^{40}K	^{232}Th series	^{238}U series	^{40}K	^{232}Th	^{238}U
S01	1045 ± 117	7535 ± 823	3206 ± 783	32306 ±3630	1857 ± 203	260 ± 63
S02	< 854	57825 ± 13239	14780 ± 4822	< 26392	14253 ± 3263	1197 ± 390
S03	663 ± 33	2140 ± 158	437 ± 99	20505 ± 1023	528 ± 39	35 ± 8
S04	327 ± 16	1861 ± 137	681 ± 153	10109 ± 506	459 ± 34	55 ± 12
S05	619 ± 27	910 ± 64	461 ± 103	19142 ± 821	224 ± 16	37 ± 8
S06	< 878	63398 ± 17735	17295 ± 6779	< 27133	15627 ± 4371	1400 ± 549
S07	< 900	87160 ± 28872	16606 ± 7255	< 27831	21484 ± 7117	1345 ± 587
S08	< 273	3505 ± 255	1419 ± 319	< 8455	864 ± 63	115 ± 26
S09	616 ± 31	3732 ± 274	1123 ± 253	19051 ± 951	920 ± 67	91 ± 20
S10	222 ± 18	11004 ± 930	5034 ± 1166	6866 ± 561	2712 ± 229	408 ± 94
S11	161 ± 7	1574 ± 112	444 ± 99	4965 ± 231	388 ± 28	36 ± 8
S12	343 ± 27	15391 ± 1328	3199 ± 738	10611 ± 843	3794 ± 327	259 ± 60

Table A2. *Geographical coordinates of soil samples.*

Code	Location	Latitude (S)	Longitude (E)
S01	Analanampotsy	17° 09' 42,5"	49° 29' 12,7"
S02		17° 09' 43,1"	49° 29' 12,5"
S03		17° 09' 44,5"	49° 29' 10,3"
S04		17°09' 51,9"	49° 29' 05,7"
S05		17° 09' 48,1"	49° 29' 09,8"
S06		17° 10' 11,8"	49° 28' 56,4"
S07		17° 10' 12,1"	49° 28' 56,5"
S08	Anjahambe	17° 07' 12,8"	49° 30' 16,4"
S09		17° 06' 42,8"	49° 30' 29,3"
S10		17° 07' 43,8"	49° 30' 08,4"
S11	Manankatafana	17° 04' 24,8"	49° 31' 25,3"
S12		17° 04' 21,6"	49° 31' 26,2"

Table A3. *Radioactivity of water samples.*

Code	Activité (Bq. L-1)		
	^{40}K series	^{232}Th series	^{238}U series
W01	32,0 ± 1,2	10,6 ± 0,7	11,8 ± 2,6
W02	17,1 ± 0,6	< 12	19,5 ± 4,4
W03	< 112	17,7 ± 1,2	10,1 ± 2,3
W04	7,3 ± 0,3	16,5 ± 1,1	3,6 ± 0,8
W05	13,4 ± 0,5	3,4 ± 0,2	8,7 ± 1,9
W06	6,9 ± 0,2	16,9 ± 1,2	8,7 ± 1,2
W07	36,2 ± 1,4	8,1 ± 0,6	10,5 ± 2,3
W08	12,3 ± 0,4	17,7 ± 1,2	0,6 ± 0,1

Table A4. *Geographical coordinates of water samples.*

Code	Location	Latitude (S)	Longitude (E)
W01	Analanampotsy	17° 07' 24,5"	49° 30' 08,8"
W02		17° 09' 42,9"	49° 29' 12,4"
W03		17° 09' 43,0"	49° 29' 12,0"
W04		17° 09' 42,4"	49° 29' 12,1"
W05	Anjahambe	17° 06' 24,9"	49° 30' 36,5"
W06	Manakatafana	17° 03' 34,4"	49° 31' 26,8"
W07		17° 06' 00,6"	49° 30' 32,7"
W08		17° 05' 51,1"	49° 29' 55,3"

Table A5. *Specific radioactivity of radon gas in air.*

Code	Time of measurement (hours)	Concentration of radon (Bq. m^{-3})		
		Average	Minimum	Maximum
R01/Anjahambe	12 h	< 1,2	-	< 1,2
R02/ Anjahambe	12 h	8,1 ± 3,1	7,7 ± 1,2	15 ± 2
R03/Anjahambe	24 h	2,6 ± 1,4	2,3 ± 1	16 ± 2
R04/Anjahambe	12 h	7,1 ± 3,6	3,2 ± 1,6	10 ± 1

Table A6. *Geographical coordinates (radon in air).*

Code	Location	Latitude	Longitude
R01	Anjahambe	17° 07' 25,0"	49° 30' 09,3"
R02		17° 07' 25,1"	49° 30' 09,4"
R03		17° 07' 24,4"	49° 30' 05,8"
R04		17° 07' 24,4"	49° 30' 05,8"

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