

Environmental radioactivity near the central coast of Venezuela and its radiological impact

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The concentrations of ^{40}K , ^{226}Ra , ^{232}Th and ^{137}Cs were determined in the upper layers of soils in the central coastal region of Venezuela. The activities of ^{137}Cs are higher in the areas where the forest is well developed, oriented towards the wind and at higher elevations. The origin of the ^{137}Cs deposition is from water input from the clouds directly in the cloudforest and rainfall from the northeast trade winds. Even though the values of ^{137}Cs are much higher in these areas, there is little or no significant increase in the health risk. The natural radioactivity is correlated with the geology in the region except in the area of Urama. The values for the natural radiation background are as follows: for potassium between 1–3%, for radium between 1–3 ppm and for thorium the range was 6–39 ppm. The corresponding amounts of absorbed dose rates in air, the exposure rates and the annual effective dose equivalents are in the following ranges respectively: 11–39 pGy/s, 4–16 $\mu\text{R}/\text{h}$ and 0.25–0.86 mSv/y. The annual effective dose equivalents include the contribution of the global average (2.57 mSv/y) of the rest of the natural sources of radiation. Finally, the largest natural radioactivity background, was found near Chichiriviche as a result of the massive granite deposits in this area, but again there is no significant health risk.

Introduction

The exposure to radiation emitted from radionuclides in the upper layers of soil is a major component of the annual effective dose equivalents that affects the human population. This exposure can be externally or internally from the ingestion of vegetable material or other parts of the food cycle, and depends upon the specific activity of the radionuclides in the soil. Thus, the measurements of both natural and artificial radionuclides in soils generate important information on the location of risk areas from the view point of radiological impact.

The most important sources of internal radiation from the natural radiation background are ^{40}K and ^{222}Rn , while ^{137}Cs and ^{90}Sr are considered the most dangerous artificial radionuclides produced in nuclear explosions and accidents. The interaction of ^{137}Cs with the environment increase the risk factor of exposure to its radiations, because of the ease to incorporate into the food chain, similarly as potassium. The distribution of ^{137}Cs in soils in general is related to latitude; more at the middle latitudes and less at the poles. About, twice as much in the northern hemisphere than the southern, where most of the nuclear weapon tests and accidents have occurred. It has also been correlated with rainfall and topography,¹ which help to explain the distribution of the higher levels in some regions of Venezuela. Recent studies have reported anomalous high ^{137}Cs concentrations on the island of Margarita, just off the coast of Venezuela in respect to those measured for the

southern side of El Avila, located on the Venezuelan coastal mountain chain.^{2,3} In this work, we have concentrated on the northern side of this coastal mountain chain, since it is directly affected by the northeastern trade winds, which transported the ^{137}Cs from its man-made sources.

The major sources of external exposure is from ^{40}K and the radionuclides of the uranium and thorium series. These radionuclides are mostly natural and related to the geology of the area with higher concentrations found in feldspars and granites. In the central coastal region of Venezuela there are massive granite deposits with the presence of micas and potassium-feldspars. In this area, it is planned for developing tourism, which could modify the environment and redistribute the concentrations of the radionuclides. Thus, another reason to study presently the natural radiation background in this region.

In Venezuela, a program of monitoring the environmental radioactivity background, both natural and artificial, with the purpose to control and protect the environment has resulted from a grant from the Venezuelan National Research Foundation (CONICIT RP VII 7600026) and an earlier one (PC-075). These grants have resulted in various publications on environmental radioactivity and its impact.^{2–9} In this work, we have measured the major radionuclides in soils to study their distribution and their impact (the absorbed dose rates in air and the annual effective dose equivalents) on the human population.

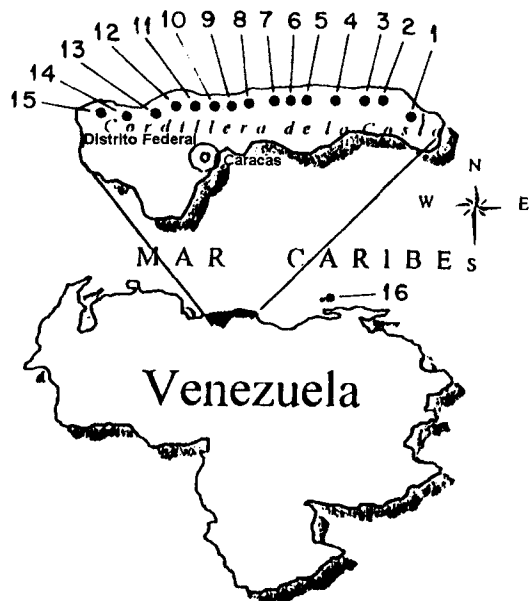


Fig. 1. Map of Venezuela showing the locations of the sampling sites:
 1 – Aricagua, 2 – Chuspa, 3 – Caruao, 4 – La Sabana, 5 – Urama,
 6 – Todasana, 7 – Oritapo, 8 – Osma, 9 – Naiguatá, 10 – Macuto,
 11 – Maiquetía, 12 – Catia la Mar, 13 – Oricoa, 14 – Chichiriviche,
 15 – Puerto Cruz, 16 – Isla de Margarita

Experimental

Description of the region studied

The coastal central region is located in the middle of the northern coast of Venezuela between $66^{\circ}05'$ and $67^{\circ}30'$ longitude west and between $10^{\circ}20'$ and $10^{\circ}39'$ latitude north (see Fig. 1). In general the area consists of metamorphic rocks that rise from the coast to the mountain range abruptly. The soils are generally sandy near the coast and their origins are predominately sedimentary and alluvial. There are two distinct climates: a tropical semiarid zone that extends along the coast and areas of low elevations; the other is a tropical mountaneous climate, which includes cloud forests at the higher elevations, which is located on the mountain chain.¹⁰

Sampling and sample preparation

The soils were collected in both virgin and disturbed areas from the top layer (0–20 cm). Then, the rocks, organic and other foreign materials were separated manually before passing a 2 mm particle size sieve.

Finally, the samples were air dried for three weeks in the laboratory.

Gamma-ray measurements

The dried soil samples were transferred to one liter Marinelli beakers for analysis. The gamma-ray spectra of the samples and standards (Soil-6 and IAEA-326) were obtained similarly employing a hyperpure Germanium detector with an energy resolution of about 2 keV for the 1.33 MeV peak of ^{60}Co and an efficiency of more than 20%. The samples and standards were measured for 60,000 seconds real time and the dead time was less than 5%. The gamma-ray data were collected with a compatible IBM computer with a multichannel/interface card (PCA-P) from Oxford Instruments Inc. (Oakridge, TN). The concentrations of the radionuclides were determined as follows: ^{137}Cs was measured employing the 661 keV gamma-rays from $^{137\text{m}}\text{Ba}$, ^{40}K from its 1461 gamma-rays, ^{226}Ra from the gamma-rays of ^{214}Pb (352 keV) and ^{214}Bi (609 keV), and ^{232}Th was calculated from the following gamma-rays: ^{228}Ac (911 keV), ^{212}Pb (239 keV) ^{212}Bi (727 keV) and ^{208}Tl (583 keV). The higher energy gamma-rays for ^{214}Bi (1.76 MeV) and ^{208}Tl (2.62 MeV) were not employed since this data collection system only had 1024 channels rather than 4096 channels. The identification and activity calculations were performed using the DIMEN program.¹¹

Results and discussion

The results from the measurements of ^{137}Cs and the other parameters of the soil samples are presented in Table 1. A multiple regression analysis of the different parameters of the the soil samples independently versus the concentration of ^{137}Cs resulted with low correlation coefficients (<60%). But, a cluster analysis showed three clear groups with the following mean ^{137}Cs activities: 1.8, 2.6 and 6.3 Bq/kg. The group with the highest deposition was closely related to soil samples taken at higher elevations in the cloud forest where the climate is very tropical and foggy as well as, the vegetation being well developed. This is logical since much of the ^{137}Cs deposition in Venezuela from the nuclear weapons tests took place in the dry season and the water input by direct condensation from the clouds in this time is similar to the amount from rainfall in the wet season.^{12,13} The relatively high content of clays and organic matter in the surface soil in these areas also help the retention of ^{137}Cs and reduce the lost from lixivation. Finally, the very developed forest also can help to retain the ^{137}Cs from resuspension and erosion too.

Table 1. Mean concentration of ^{137}Cs (in Bq/kg) and its standard deviation ($X \pm \text{SD}$) in soils at the different locations and other relevant soil parameters

Sample location ^a	^{137}Cs , ^b Bq/kg	General description ¹	Elevation ²	Geographic orientation ³	Distance from coast ⁴	Soil alteration ⁵	Surface inclination ⁶	Vegetation type ⁷	Soil type ⁸
1	8.6 ± 0.7	B	C,D	A,C	T	Y	Y	B	B,D
1-2	7.3 ± 0.2	B	C,D	A,C	S,T	Z	Y	A,B	C,D
1-2	6.4 ± 0.6	B	C,D	A,C	S,T	Z	Y	A,B	C,D
2	4.9 ± 0.4	B	C,D	A,C	R,S	Y		B,D	B,D
3	7.3 ± 0.3	B	C,D	A	R,S	Y	Y	A,B	C,D
3	10.1 ± 0.4	B	C,D	A	S	Z	X	B	C,D
3	6.9 ± 0.1	B	C,D	A	S	Z	X	B	C,D
4	4.7 ± 0.7	A,B	B	E	R	Z	X	B	C,D
4	3.8 ± 0.4	A,B	B	E	R	Z	X	B	C,D
4-5	7.5 ± 0.6	B	C,D	A,C	R	X	Y	A	C,D
4-5	4.3 ± 0.3	B	C,D	B,D	S	Y	Y,Z	A	A,C
4-5	6.4 ± 0.6	B	C,D	A	S	Y	X	A,B	C,D
5	2.6 ± 0.1	B	C,D	A	S	Y	X	A,B	C,D
5	3.5 ± 0.1	A,B	C	A	P	Y	Y	A,B	C,D
6	3.2 ± 0.2	A,B	C	A	P	Y	Y	A,B	C,D
7	≤	A	C	A	P	Y	Y	B,D	B,D
8	2.4 ± 0.2	A	B	A	Q	Y	X	B	C,D
8	5.8 ± 0.1	C	B	E	S	Z	X	B,C	B
8	5.2 ± 0.1	C	B	B	S	Z	X	B	C,D
8	9.3 ± 0.1	B	C	B	S	Y	Y	A,B	C,D
8	9.6 ± 0.7	B	D	A	S	X	Y	A,B	C,D
8	4.4 ± 0.2	B	B	E	S	X	X	A,B	B,C
8	0.3 ± 0.1	C	B	E,F	S	Z	X	B,C	B
8	4.0 ± 0.3	C	B	E	S	Y	X	C	B
8	≤	A	B	E	S	Y	X	A	E
8	2.9 ± 0.3	A	B	E	S	X	X	A,B	B
8	3.8 ± 0.6	B	D	B,C	T	Z	Y	B	C,D
8	2.9 ± 0.1	B	D	B	T	Z	Y	B	C,D
8	7.8 ± 0.1	B	D	A	T	Z	Y	B,C	C,D
8	3.5 ± 0.4	C	C	EF	T	X	Y	A	C,D
8	6.3 ± 0.5	B	D	B,A	S	Y	Y	A	C,E
8	8.3 ± 0.1	B	D	A	S	X	Y	A	C,D
8	3.5 ± 0.1	C	C	E	R,S	Z	X	B,C	B
8	1.4 ± 0.1	C	B,C	E,F	R,S	Z	X	C	B,E
8	≤	C	A,B	E	R,S	Y	X	B	E
8	2.3 ± 0.1	C	A,B	E	R,S	Y	Y	D	B
8	6.0 ± 0.2	B	B,C	B,C	R,S	Y	Y	B	C,D
8	5.8 ± 0.2	C	A,B	E	R,S	Y	X	A,B	C,D
8	2.4 ± 0.6	C	A,B	E	R,S	Y	X	C	A,B
8	5.2 ± 0.5	B	B,C	B,C	R,S	Y	Y	B	C,D
8	3.1 ± 0.3	A,B	C,D	B,C	R,S	Z	Y	D	A,B
8	3.8 ± 0.1	B	D	B,C	R,S	Y	Y	B,C	B,D
8	0.8 ± 0.2	C	B	E	R,S	Y	X	B,C	E
8	1.1 ± 0.1	A	A	E	P	Z	X	B,C	A,D
8	≤	C	B	B	R,S	Y	Z	D	A,D
8	≤	A	A	E	P	Y	X	B	A,D
8	2.4 ± 0.4	A,C	B	E	P,Q	Y	X	B	B
8	3.4 ± 0.3	A,C	B	E	P,Q	Y	X	B	B
8-9	≤	A,B	C,D	A,B	P,Q	Y	Y	D,E	A
9	0.6 ± 0.1	A	C	A	Q	Z	Y	D,E	A,D
14	≤	A	B	A	Q	Z	Y	B	A
14	0.6 ± 0.1	A	B	A	Q	X	Y	B,D	B,D

Codes for soil parameters:

¹ General description: A – semi-arid tropical; B – tropical forest; C – valley;² Elevation (m. a. s. l.): A – close to sea level; B – 10–150; C – 50–100; D – 100–500; E – >500;³ Geographical orientation: A – facing north; B – facing south; C – facing east; D – facing west; E – flat; F – south side (wind blocked by mountains);⁴ Distance from coast (meters): P – <100; Q – 100–500; R – 500–1000; S – 1000–2000; T – 2000–5000⁵ Alteration of sample zone: X – undistributed; Y – slightly distributed; Z – cultivated;⁶ Inclination: X – flat; Y – slight inclined; Z – very inclined;⁷ Type of vegetation: A – large trees (>3 m); B – small trees (<3 m); C – grass; D – weeds; E – cactus;⁸ Type of surface: A – rocky; B – sandy; C – with organic residues; D – clay; E – river sand.^a The number indicates the sample site on the map in Fig. 1.^b The ≤ symbol is for levels below our detector limit of 0.3 Bq/kg.

Table 2. The mean concentration of ^{40}K , ^{226}Ra and ^{232}Th and their respective standard deviations ($X \pm \text{SD}$) at the different sample locations

Sample location* (x)	Number of samples	^{40}K , Bq/kg ($X \pm \text{SD}$)	^{226}Ra , Bq/kg ($X \pm \text{SD}$)	^{232}Th , Bq/kg ($X \pm \text{SD}$)
Aricagua (1)	8	616 \pm 45	27.5 \pm 2.5	56.4 \pm 5.2
Aricagua-Chuspa (1-2)	7	519 \pm 34	30.8 \pm 3.1	43.8 \pm 4.0
Chuspa (2)	10	822 \pm 70	26.2 \pm 1.7	41.4 \pm 3.2
Caruao (3)	8	414 \pm 36	36.5 \pm 2.7	35.4 \pm 3.3
La Sabana (4)	9	558 \pm 39	22.8 \pm 2.2	46.2 \pm 3.9
El Salto (4-5)	8	630 \pm 51	13.6 \pm 1.4	35.3 \pm 3.1
Aguas Calientes (4-5)	7	721 \pm 62	17.1 \pm 1.8	36.5 \pm 3.7
Campamento Aventura (4-5)	7	327 \pm 28	22.7 \pm 2.1	26.9 \pm 2.4
Urama (5)	7	637 \pm 47	36.5 \pm 3.3	59.4 \pm 4.8
Todasana (6)	9	325 \pm 32	20.6 \pm 1.6	27.1 \pm 2.3
Oritapo (7)	9	403 \pm 21	15 \pm 1.6	24 \pm 2.3
Osma (8)	33	378 \pm 21	20.6 \pm 2.1	29.2 \pm 2.2
Osma-Los Caracas (8-9)	8	384 \pm 22	23.7 \pm 2.1	32.2 \pm 2.9
Chichiriviche (14)	10	811 \pm 62	26.9 \pm 2.2	141 \pm 12

* The number in parentheses indicates the location on the map in Fig. 1.

Table 3. The mean concentration of potassium, radium and thorium with their respective standard deviations ($X \pm \text{SD}$), as well as the calculated mean absorbed dose rates, exposure rates and annual effective dose equivalents for the different sample locations

Sample location* (x)	Number of samples	Potassium, ppm ($X \pm \text{SD}$)	Radium, ppm ($X \pm \text{SD}$)	Thorium, ppm ($X \pm \text{SD}$)	Absorbed dose rate, pGy/s	Exposure rate, $\mu\text{R/h}$	Annual effective dose equivalents, mSv/a
Aricagua (1)	8	1.98 \pm 0.14	2.27 \pm 0.206	14.07 \pm 1.30	21.16	8.77	0.48
Aricagua-Chuspa (1-2)	7	1.67 \pm 0.11	2.54 \pm 0.26	10.93 \pm 1.01	18.19	7.54	0.40
Chuspa (2)	10	2.64 \pm 0.22	2.16 \pm 0.14	10.33 \pm 0.80	20.79	8.61	0.47
Caruao (3)	8	1.33 \pm 0.12	3.01 \pm 0.22	8.83 \pm 0.82	16.19	6.71	0.36
La Sabana (4)	9	1.79 \pm 0.12	1.88 \pm 0.18	11.53 \pm 0.97	18.02	7.46	0.40
El Salto (4-5)	8	2.02 \pm 0.16	1.12 \pm 0.12	8.81 \pm 0.73	15.70	6.50	0.35
Aguas Calientes (4-5)	7	2.32 \pm 0.20	1.41 \pm 0.15	9.11 \pm 0.92	17.48	7.24	0.39
Campamento Aventura (4-5)	7	1.05 \pm 0.09	1.87 \pm 0.17	6.71 \pm 0.60	11.78	4.88	0.26
Urama (5)	7	2.05 \pm 0.15	3.01 \pm 0.27	14.82 \pm 1.20	23.16	9.59	0.53
Todasana (6)	9	1.05 \pm 0.10	1.7 \pm 0.13	6.76 \pm 0.57	11.52	4.77	0.26
Oritapo (7)	9	1.29 \pm 0.07	1.24 \pm 0.13	5.99 \pm 0.57	11.15	4.62	0.25
Osma (8)	33	1.21 \pm 0.07	1.7 \pm 0.17	7.29 \pm 0.55	12.52	5.19	0.28
Osma-Los Caracas (8-9)	8	1.23 \pm 0.07	1.96 \pm 0.17	8.03 \pm 0.72	13.55	5.61	0.30
Chichiriviche (14)	10	2.60 \pm 0.20	2.22 \pm 0.18	35.28 \pm 3.00	38.58	15.98	0.87

* The number in parentheses indicates the location on the map in Fig. 1.

The estimated annual addition dose from the ^{137}Cs component from ingesting cultivated products from the highest sampled area, about 10 Bq/kg was calculated to be only about 0.016 mSv/y, using the equations and information in Reference 14. This estimation is negligible as compared to the estimated global annual dose of 1.6 mSv/y for the internal exposure from the primordial radionuclides including the inhalation of ^{222}Rn gas.

A cluster analysis employing the activities of the different radionuclides used to estimate the natural radiation background, immediately showed the sample site at Chichiriviche to be anomalous, about twice as high as at any other site. After excluding this site, the cluster analysis resulted with two clearly defined groups. The mean activities of the different radionuclides and their

respective standard deviations (1σ) are shown in Table 2. The difference in the groups can be explained by the geology of the sites, except Urama.

The mean concentrations of potassium, radium and thorium and their respective calculated absorbed dose rates, exposure rates and annual effective dose equivalents are expressed for each sampling location in Table 3. Here, the high values can readily be seen for the location of Chichiriviche, which can also be explained by the geology of the area. But, again the high values in respect to the group in which it fell for the Urama area can not be explained by its geology. Finally, the average estimated global value for external exposure from the primordial radionuclides is about 0.41 mSv/y, which in general is similar to most of the sites we studied.

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