

## CORRELATION OF NATURAL AND ARTIFICIAL RADIONUCLIDES IN SOILS WITH PEDOLOGICAL, CLIMATOLOGICAL AND GEOGRAPHIC PARAMETERS

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Various types of soil samples were collected in the southern part of Brazil, with depth intervals of 5 cm, down to 50 cm, using a specially designed sampler. Pedological analysis of these soils were performed. Nuclear activities of <sup>137</sup>Cs (expressed in Bq m<sup>-3</sup>) and radioactive natural element (<sup>226</sup>Ra, <sup>228</sup>Ra and <sup>40</sup>K) concentrations were determined by low background gamma-ray spectrometry. <sup>137</sup>Cs concentrations were correlated with radioactive natural element concentrations and pedological, climatological and geographic parameters related to the soil samples collected.

The concentrations of natural radioactive elements such as <sup>226</sup>Ra, <sup>228</sup>Ra and <sup>40</sup>K and man-made <sup>137</sup>Cs were determined in soil samples and the results are expressed as Bq m<sup>-2</sup>. <sup>137</sup>Cs is a man-made radionuclide and was deposited in soil as fallout produced by atmospheric nuclear tests and accidents.<sup>1</sup> Gamma-ray spectrometry was used to determine nuclear activities.<sup>2</sup> Samples of soils, provided by the International Atomic Energy Agency (IAEA), were used to calibrate the detector.<sup>3</sup>

SCHUCH et al.<sup>1</sup> have studied <sup>137</sup>Cs behavior in soil samples collected in 1991 in Paraná State, Brazil. These samples were compared with others collected in the same locations in March 1977, and November-December 1983. In all samples for which pedological analysis were performed, concentrations of <sup>137</sup>Cs, <sup>226</sup>Ra, <sup>228</sup>Ra and <sup>40</sup>K were measured. This study has shown the latitudinal dependence of <sup>137</sup>Cs fallout, as well as the impossibility to measure <sup>137</sup>Cs contribution from the Chernobyl accident among <sup>137</sup>Cs concentrations measured, due to the intense leaching within these soils from high rainfall regions. It was also shown that, on the contrary of what

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happens with  $^{137}\text{Cs}$ , natural radioactive elements were practically not affected by the effects of leaching, confirming the fact that they are strongly fixed to soil matrix or included in insoluble mineral of the bedrock.

The main purpose of this work was to determine possible correlations<sup>4-6</sup> between  $^{137}\text{Cs}$  concentrations, natural radionuclide concentrations ( $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$  and  $^{40}\text{K}$ ), and pedological (Ph, clay and organic carbon, exchangeable bases  $\text{Ca}^{++}$ ,  $\text{Mg}^{++}$ ,  $\text{K}^+$  and  $\text{Na}^+$ , cation exchange capacity and percentage of base saturation), climatological (mean annual rainfall and temperature) and geographic (altitude, latitude and longitude) parameters.  $^{137}\text{Cs}$  distributions within the soil profiles were also analyzed.

### Experimental

In June 1991, in Rio Grande do Sul State, 7 profiles of soils were collected down to 50 cm, with depth intervals of 5 cm, using a carbon steel sampler (area of 20 cm x 10 cm and depth of 5 cm) specially designed for this purpose, as shown in Table 1.

Samples were collected, prepared, and the concentrations of radioactive elements measured, following the methodology described in SCHUCH et al.<sup>1</sup> Pedological analysis was performed by the Soil Department of the Rural Sciences Center of the Federal University of Santa Maria. Table 2 presents the mean values of 10 samples obtained for each soil profile for the concentrations of radionuclides and for the pedological parameters of all soil from Rio Grande do Sul State. Climatological and geographic parameters of all soil samples are given in Table 1.

### Results and Discussion

The coefficients of linear correlations for  $^{137}\text{Cs}$  concentrations per unit and the rest of the parameters shown in Tables 1 and 2 are presented in Table 3.

From Table 3, we can conclude that there is a certain linear correlation between the concentrations per unit area of  $^{137}\text{Cs}$  and the concentrations of K, C,  $\text{Ca}^{++}$ ,  $\text{Mg}^{++}$  and CEC (cation exchange capacity). This result seems to show that the fixation of  $\text{Ca}^{++}$  and  $\text{Mg}^{++}$  by clays prevents a larger absorption of  $^{137}\text{Cs}$  by the clays, even though the  $^{137}\text{Cs}$  has a greater affinity for the clays.<sup>5,6</sup> The result obtained for the CEC is explained in the same way. The larger the CEC, the easier the way  $^{137}\text{Cs}$ , eventually adsorbed by the clays, will be substituted by extractable bases.

The soils taken for that work were chosen in locations of similar mean annual rainfall, to eliminate this possible influence in the correlations made with pedological parameters. However,

TABLE 1  
Soil Samples Collected in Rio Grande do Sul State, Brazil

Samples	Classification	Latitude (S)	Longitude (W)	Altitude (m)	Mean Annual Rainfall (mm)	Mean Annual Temperature (°C)
50-59	Reddish Brunizem	29°09'44"	51°31'50"	640	1704	17.2
60-69	Dusky Red Latosol	28°30'00"	54°05'00"	390	1729	18.2
80-89	Dark Red Podzolic	29°30'18"	53°40'40"	470	1575	18.0
120-129	Structured Dusky Red Earth	28°39'44"	56°00'15"	90	1523	20.1
130-139	Vertisol	28°40'00"	56°00'00"	180	1523	20.1
140-149	Structured Dusky Red Earth	28°15'40"	52°24'45"	709	1664	17.5
150-159	Brown Latosol	28°30'09"	50°56'12"	955	1412	15.2

TABLE 2  
Activities per Unit Area of  $^{137}\text{Cs}$  and Mean Values Obtained, for Each Soil Profile With 10 Samples,  
for Concentrations of Natural Radionuclides and for Pedological Parameters

SAMPLES	$^{137}\text{Cs}$ ( $\text{Bq m}^{-2}$ )	eq Th ( $^{228}\text{Ra}$ ) (ppm)	eq U ( $^{235}\text{Ra}$ ) (ppm)	K (%)	pH $\text{H}_2\text{O}$ 1:25	CLAY <0.0002 mm (%)	ORG. C (%)	EXTRACTABLE BASES meq/100 g					EXTR. ACIDITY $\text{Al}^{+++} + \text{H}^+$	CATION EXCH. CAP. CEC	BASE SAT. V (%)
								Ca $^{++}$	Mg $^{++}$	K $^+$	Na $^+$	SUM S			
50-59	1566	15.6	4.3	0.47	4.89	21	1.38	6.89	2.57	0.15	0.13	9.74	3.07	12.81	76
60-69	1388	11.8	4.0	0.13	5.18	66	2.01	2.77	2.23	0.23	0.18	5.41	3.58	8.99	60
80-89	1239	14.7	5.3	1.31	4.78	35	1.79	7.61	3.26	0.13	0.11	11.11	8.17	19.28	58
120-129	1228	19.8	5.3	1.64	5.11	56	1.58	3.93	3.88	0.08	0.10	7.99	3.57	11.56	69
130-139	501	6.6	1.4	3.59	5.88	48	3.11	39.71	15.60	0.14	0.34	55.79	3.60	59.39	94
140-149	601	20.9	5.7	1.45	4.49	53	1.61	7.28	4.18	0.17	0.10	11.73	6.31	18.04	62
150-159	913	13.8	3.5	1.38	4.52	70	2.74	1.47	1.11	0.12	0.19	2.89	7.19	10.32	27

TABLE 3  
Linear Correlation Coefficients Calculated

$^{137}\text{Cs}$	$^{228}\text{Ra}$	$^{228}\text{Ra}$	K	pH	Clay	C	Ca $^{++}$	Mg $^{++}$	K $^+$	Na $^+$	SUM S	$\text{Al}^{+++} + \text{H}^+$	CEC	V	rainf.	temp.	alitt.
	0.19	0.38	-0.79	-0.36	-0.62	-0.61	-0.62	-0.51	-0.62	-0.25	-0.67	-0.15	0.43	-0.13	-0.03		

according to SCHUCH et al.,<sup>1</sup> total  $^{137}\text{Cs}$  per unit area in soils collected from several locations of humid regions in the central part of Bahia State, Brazil (yearly rainfall: 1700 to 2000 mm), was about 40% greater than others collected in dry regions (yearly rainfall: 600-1000 mm).

Table 4 shows the percentage of  $^{137}\text{Cs}$  total concentration per unit area within each depth interval. We can observe that the larger concentrations of  $^{137}\text{Cs}$  are found in samples collected between 0 and 15 cm of depth. More than 60% of the total concentration was confined to the top 25 cm of all sites. In this work, we observe the distribution of  $^{137}\text{Cs}$  to be deeper under the ground as compared to that obtained by ARNALDS et al.,<sup>5</sup> where 90% of the  $^{137}\text{Cs}$  was confined to the

TABLE 4  
Percentage of  $^{137}\text{Cs}$  Total Activity per Unit Area  
Within Each Depth Interval

Depth Interval (in cm)	Percentage of total $^{137}\text{Cs}$ activity per unit area (0%)						
	Samples						
	50-59	60-69	80-89	120-129	130-139	140-149	150-159
0-5	20	17	31	28	18	24	33
5-10	11	32	19	34	14	12	27
10-15	46	15	9	9	16	13	8
15-20	17	14	10	7	6	12	15
20-25	6	10	6	9	6	nd	6
0-25	100	88	75	87	60	61	89
25-50	nd	12	25	13	40	39	11

nd = Value not determined. Deviations were greater than 95 percent.

top 20 cm at the 10 analyzed sites. This larger depth profile is due to the high rainfall which is characteristic of Rio Grande do Sul State.

There is a high positive linear correlation among the  $^{137}\text{Cs}$  concentrations per unit area in the superficial soil samples of each profile and the content of organic carbon. However, if we take into account the entire profiles (collected down to 50 cm depth) we find the linear correlation coefficient to be only  $r = -0.62$  (Table 3).

The linear correlation between the  $^{228}\text{Ra}$  and the  $^{226}\text{Ra}$  shows  $r = 0.91$  and, as was expected from pedological soil studies, there is a high correlation between the various pedological parameters such as: Ph and  $\text{Ca}^{++}$  ( $r = 0.76$ ), and  $\text{Mg}^{++}$  ( $r = 0.81$ ), and S ( $r = 0.78$ ); and between CEC and  $\text{Ca}^{++}$  ( $r = 0.90$ ), and  $\text{Mg}^{++}$  ( $r = 0.98$ ) and S ( $r = 0.99$ ).

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