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PART VIII: Radionuclides

Chapter 18

RADIOACTIVE SOIL CHARACTERIZATION OF THE STATE OF SAO PAULO, BRAZIL

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Abstract: For proper management of soil and groundwater quality, background levels of toxic elements in a given ecosystem must be known. The aim of this study is to determine quality reference values for radioactive content in representative soils from the São Paulo State, Brazil.

Thirty samples were collected and the concentration of U-nat, Th-nat, 228Ra, 226Ra, 210Pb, 137Cs and 40K were evaluated and correlated with soil mineralogical characteristics. The types of soil and sampling places were chosen according to their representativeness and spatial distribution in the State geological formations, taking also into account their nearness to large urban areas. The samples were measured by means of passive gamma spectrometry and sequential chemical extraction, followed by alpha spectrometry. Results showed a wide variation on background levels for natural radionuclides of the uranium and thorium series and very low concentration of 137Cs from radioactive fallout.

Key words: Soil; radioactivity; background levels; characterization contamination.

1. INTRODUCTION

Over the last decades, as a result of past agricultural and industrial activities lacking adequate regulatory control, contaminated areas have become a great environmental issue.

In search for proper solutions to these questions, soil remediation policies have been implemented in several countries (Kolluru et al, 1996). In general, soil remediation is an expensive option. Besides which, typically the elapsed time between the identification of the contaminated area and the decision to undertake corrective action is too great, due to the complexity of the site-specific studies necessary for a proper characterization of all the associated risks. Nevertheless, in face of this, governmental agencies from countries such as the Netherlands (VRON, 1988), the USA (U.S.EPA, 1966) and Germany (Bachmann, 2000), among others, have opted to establish target values for soil quality as a first step in the whole process of risk assessment.

In Brazil, the only state to adopt target values for soil contamination – specific to its characteristics – is the State of São Paulo, through its governmental agency for pollution control - CETESB. Three levels have been established: (1) quality reference value, indicating the quality level of a soil considered as clean soil, generally associated with the natural concentration of the element of interest; (2) preventing value, above which harmful changes in the soil quality may occur; and (3) intervention value, above which potential risks to the human health exist, considering a generic exposure scenario (CETESB, 2005).

The list of the elements for which target values are in force includes organic chemicals, volatile organic compounds and heavy metals, all of them potential carcinogens and, therefore, subject to regulatory control. However, it does not include radionuclides. In addition, Brazilian National Nuclear Energy Commission (CNEN), the federal agency responsible for regulating nuclear activities in the country, has not established target values for radioactive soil contamination either.

The main aim of the present study is to perform a radioactive characterization of representative soils from the São Paulo State, in order to determine background levels for some selected radionuclides, which could be used as a quality reference value.

2. MATERIALS AND METHODS

Surface samples (0-0.2 m depth) of 30 profiles of non-cultivated soils were collected in areas of native vegetation or ancient reforestation in the State of Sao Paulo, Brazil. This state has mainly four kinds of climate (Köppen) according to the slope: tropical wet and dry (Aw), in the shore coastline (20-22°C; 4,154 mm/yr); humid subtropical (Cwa), in the plateau (20°C; 1,300 mm/yr); tropical wet and dry (As), in the northwest region (24°C; 1,000-1,250 mm/yr); and humid subtropical (Cfa), in the south region (18-20°C; 1,500 mm/yr). The remaining natural vegetation is composed by Atlantic Rain forest along the shore and surrounding the plateau. The rest of these areas present fragments of semideciduos forest and a kind of vegetation that includes all the formations of the Cerrado biome, such as clean grass fields, grassland with some shrubs and the cerrado strictu sensu (short savannah), that is arbustive and herbaceous, and the forest formation of “cerradão” (closed savanna approaching a forest). Oxisols, Ultisols, Alfisols, Entisols, Inceptisols, Histosols and Mollisols were found in these regions constituting a group with wide variation on (electro)chemical, physical and mineralogical properties.

Soil types and sampling places were chosen according to their representativeness and spatial distribution in the State geological formations, taking also into account their nearness to large urban areas. Thirty samples were collected in areas presenting low anthropic perturbation - native vegetation or ancient reforestation -, minimizing the chances that selected areas could have sustained anthropic effects in recent past.

Samples were collected using steel stainless instruments, in accordance to the standard procedures recommended by CETESB (CETESB, 2001); they were conditioned in polyethylene bags for transportation and dispersed over benches for natural drying. After that, the soils were sieved to less than 2,0 mm granular size. The granulometric analysis of soil samples followed the densimetry method, after dispersion with $(\text{NaPO}_3)_6 \cdot \text{Na}_2\text{O}$ (0.015 mol L^{-1}) + NaOH (1 mol L^{-1}) (Soares, 2004). Table 1 presents the soil type and the content of clay, silt and sand.

Contents of ^{228}Th , ^{228}Ra , ^{226}Rn , ^{210}Pb , ^{137}Cs and ^{40}K were determined by means of passive gamma spectrometry using a hyperpure germanium detector, in a 100 mL polyethylene flask, for a counting time ranging from 90.000 to 220.000 seconds. The flasks were previously sealed to assure that radioactive equilibrium between ^{226}Ra , as well ^{228}Th , and their short-living daughters would be reached.

Contents of U-nat and Th-nat were determined by means of spectrophotometry, employing Arsenazo III.

Table 1. Types of the soils collected in the São Paulo State and contents of clay, silt and sand.

Soil Type ¹	Geographical Coordinates	Composition (%)		
		Clay	Silte	Sand
1 – Typic Hapludox	22°19'S 47°10'W	18.1	4	77.9
2 – Typic Hapludox	22°15'S 47°49'W	22.1	2	75.9
3 – Typic Hapludox	22°19'S 47°10'W	20.2	6	73.8
4 – Rhodic Eutrudox	22°01'S 47°53'W	20.1	8.1	71.8
5 – Rhodic Hapludox	21°05'S 47°08'W	53	10.2	36.8
6 – Rhodic Eutrudox	22°43'S 47°38'W	68.4	20.7	10.9
7 – Rhodic Acrudox	21°10'S 47°48'W	71.6	14.3	14.1
8 – Xanthic Hapludox	22°15'S 47°49'W	22.2	4	73.8
9 – Xanthic Hapludox	21°57'S 47°59'W	34.2	6	59.7
10 – Anionic Acrudox	20°10'S 48°02'W	47	12.3	40.7
11 – Arenic Hapludult	22°32'S 47°54'W	6	10	84
12 – Arenic Hapludult	22°17'S 49°33'W	10	32	58
13 – Arenic Hapludult	22°38'S 47°11'W	24.7	20.6	54.7
14 – Arenic Hapludalf	22°59'S 47°30'W	20.2	38.4	41.4
15 – Typic Hapludalf	22°59'S 47°30'W	36.6	44.8	18.6
16 – Arenic Hapludult	22°43'S 47°38'W	24.6	12.3	63
17 – Arenic Hapludalf	22°12'S 49°56'W	10	34.1	55.8
18 – Typic Hapludalf	22°06'S 47°07'W	42.7	42.7	14.6
19 – Typic Quartzipsamment	22°32'S 47°54'W	8	4	88
20 – Lithic Udortent	22°12'S 49°39'W	14.2	34.6	51.2
21 – Typic Usthorthent	22°26'S 49°45'W	10.1	30.2	59.7
22 – Typic Hapludult	22°06'S 46°40'W	34.5	18.2	47.3
23 – Kandiuclalfic Eutrudox	21°10'S 47°48'W	65.8	26.7	7.5
24 – Typic Haplaquox	24°43'S 47°52'W	53.2	28.7	18.1
25 – Typic Eutraquox	22°43'S 47°38'W	47.6	38	14.4
26 – Typic Medifibris	24°43'S 47°52'W	27.5	40.1	32.4
27 – Typic Dystrochep	22°22'S 46°56'W	24.3	10.1	65.6
28 – Arenic Albaquilt	22°43'S 47°38'W	20.4	34.7	44.9
29 – Arenic Hapludalf	22°12'S 49°39'W	4	24	72
30 – Typic Argiudoll	22°07'S 47°39'W	54.3	25.1	20.6

¹ Classification according to USDA Soil Taxonomy (Soil Survey Staff, 1999)

3. RESULTS AND DISCUSSION

Results of the radionuclide determination are presented in Table 2. A wide variation in activity concentration could be observed for all the elements analyzed, even for the same type of soil, except for the ¹³⁷Cs. The analysis of the statistical distribution of the given values revealed that that no clear choice between a log-normal or normal pattern can be made. This result was expected for the ¹³⁷Cs, since it is not an isotope naturally present in the soil and fallout precipitation is expected to be relatively uniform over the State area. The activity concentration values for the ¹³⁷Cs obtained are very close to the detection limits for most samples and normal pattern was adopted in this case; for other radionuclides, option was made for a log-normal distribution.

No significant correlation could be observed between the clay, silt and sand content in the samples and the respective activity concentration of the radionuclides.

Table 2. Activity concentration in soil of selected radionuclides (Bq per kg of dry soil)

Soil	Radionuclide							
	U-nat	Th-nat	²²⁸ Th	²²⁸ Ra	²²⁶ Ra	²¹⁰ Pb	¹³⁷ Cs	⁴⁰ K
1	<47	18 ± 3	14.6 ± 0.8	11.9 ± 1.9	8.6 ± 1.2	21 ± 3	1.9 ± 0.4	26.4 ± 8.5
2	<47	32 ± 4	15.7 ± 0.8	16.1 ± 2.0	9.5 ± 1.2	<20	1.2 ± 0.4	34.2 ± 8.6
3	<47	12 ± 2	30.6 ± 0.9	27.2 ± 2.3	10.9 ± 1.2	<20	1.3 ± 0.5	< 11.1
4	<47	12 ± 2	11.9 ± 0.7	9.1 ± 1.6	5.9 ± 0.9	<20	0.8 ± 0.4	49.0 ± 7.3
5	124 ± 14	65 ± 8	94.4 ± 2.1	77.7 ± 4.9	56.2 ± 2.6	56 ± 6	< 0.3	64.8 ± 18.9
6	<47	33 ± 4	32.4 ± 1.8	22.5 ± 4.2	14.6 ± 2.4	60 ± 8	2.9 ± 0.7	15.3 ± 19.1

	Radionuclide							
7	57 ± 8	35 ± 5	32.7 ± 1.7	29.0 ± 4.0	13.3 ± 2.3	60 ± 7	2.0 ± 0.7	50.2 ± 18.3
8	<48	15 ± 2	33.4 ± 0.8	28.1 ± 2.0	15.7 ± 1.1	<20	1.8 ± 0.4	119.8 ± 8.8
9	<47	33 ± 4	41.9 ± 1.5	34.9 ± 3.5	16.0 ± 1.9	26 ± 3	<0.8	15.6 ± 15.0
10	<47	43 ± 6	60.0 ± 1.1	54.6 ± 2.6	34.2 ± 1.4	35 ± 4	0.8 ± 0.5	51.1 ± 9.4
11	<47	<6	8.2 ± 1.1	9.3 ± 2.5	4.5 ± 1.4	<20	1.2 ± 0.5	47.7 ± 12.1
12	<47	18 ± 3	13.5 ± 1.1	12.7 ± 2.7	12.5 ± 1.6	25 ± 6	1.1 ± 0.5	104.8 ± 13.2
13	61 ± 8	76 ± 10	83.8 ± 1.1	70.3 ± 2.7	36.4 ± 1.2	32 ± 8	2.7 ± 0.4	54.1 ± 8.0
14	<48	16 ± 2	19.2 ± 1.4	14.6 ± 3.2	18.7 ± 1.9	33 ± 8	1.3 ± 0.6	96.9 ± 15.5
15	<47	41 ± 5	57.9 ± 1.8	52.8 ± 6.3	21.7 ± 2.3	35 ± 9	1.4 ± 0.7	344.8 ± 19.7
16	56 ± 8	61 ± 8	66.5 ± 0.9	56.8 ± 2.4	40.4 ± 1.2	70 ± 10	2.1 ± 0.4	111.3 ± 7.9
17	<47	23 ± 3	25.9 ± 1.2	24.8 ± 2.8	13.1 ± 1.6	30 ± 7	2.2 ± 0.5	351.3 ± 15.0
18	<48	63 ± 8	76.6 ± 1.8	61.0 ± 4.3	61.8 ± 2.5	64 ± 13	1.9 ± 0.7	38.3 ± 17.6
19	<47	8 ± 2	4.8 ± 0.5	3.3 ± 1.1	1.0 ± 0.7	<20	1.0 ± 0.3	< 13.2
20	<47	18 ± 3	25.5 ± 0.6	21.4 ± 1.4	9.6 ± 0.8	51 ± 10	1.6 ± 0.3	356.2 ± 8.4
21	<47	22 ± 3	20.5 ± 1.3	15.3 ± 3.0	9.2 ± 1.7	37 ± 9	0.9 ± 0.6	301.8 ± 16.1
22	75 ± 9	40 ± 5	62.1 ± 1.9	47.2 ± 4.5	44.9 ± 2.6	80 ± 9	4.1 ± 0.8	< 13.2
23	<47	33 ± 5	30.3 ± 1.5	21.4 ± 3.4	17.7 ± 2.0	33 ± 10	2.5 ± 0.6	33.8 ± 15.9
24	99 ± 12	54 ± 7	72.6 ± 1.2	57.0 ± 2.8	45.8 ± 1.5	94 ± 14	4.0 ± 0.5	516.4 ± 13.4
25	411 ± 45	36 ± 5	71.1 ± 2.4	65.5 ± 5.7	36.5 ± 3.2	121 ± 13	1.7 ± 1.0	68.6 ± 24.6
26	81 ± 10	29 ± 5	50.9 ± 2.0	47.0 ± 4.8	39.6 ± 2.8	(1)	2.2 ± 0.9	233.3 ± 22.4
27	86 ± 10	82 ± 11	119.9 ± 1.9	97.6 ± 4.7	43.0 ± 2.2	75 ± 15	1.1 ± 0.7	67.3 ± 16.2
28	98 ± 12	42 ± 6	60.6 ± 1.7	47.9 ± 3.8	38.7 ± 2.2	50 ± 9	2.1 ± 0.7	93.3 ± 16.5
29	<47	20 ± 3	23.1 ± 1.2	16.5 ± 2.7	10.7 ± 1.5	<20	1.2 ± 0.5	253.8 ± 14.0
30	<47	25 ± 4	22.8 ± 0.8	18.9 ± 1.9	8.3 ± 1.1	37 ± 10	2.7 ± 0.4	153.0 ± 9.1
Range	<47 - 411	8 - 82	4.8 - 120	3.3 - 97.6	1.0 - 61.8	<20 - 121	0.8 - 4.1	15.3 - 516
Mean	93 ⁽²⁾	30 ⁽²⁾	33.2 ⁽²⁾	27.8 ⁽²⁾	17.1 ⁽²⁾	46 ⁽²⁾	1.9	86.7 ⁽²⁾
SD	1.8 ⁽³⁾	1.8 ⁽³⁾	2.2 ⁽³⁾	2.2 ⁽³⁾	2.4 ⁽³⁾	1.6 ⁽³⁾	0.9	2.7 ⁽³⁾

(1) Not determined. (2) Geometric mean. (3) Geometric standard deviation

4. CONCLUSION

Considering the representativeness of the soil samples collected, geometric mean of the activity concentrations obtained in this study could be used by regulatory bodies to establish quality reference values for soils in the Sao Paulo State. The present study covers the main natural radionuclides and those originating from fallout of more radiological concern. Further studies are still in progress, aiming at determining concentration activities of others radionuclides of interest.

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