RANDOM AND SYSTEMATIC SPATIAL VARIABILITY OF ^{137}Cs INVENTORIES AT REFERENCE SITES IN SOUTH-CENTRAL BRAZIL

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ABSTRACT: The precision of the ^{137}Cs fallout redistribution technique for the evaluation of soil erosion rates is strongly dependent on the quality of an average inventory taken at a representative reference site. The knowledge of the sources and of the degree of variation of the ^{137}Cs fallout spatial distribution plays an important role on its use. Four reference sites were selected in the South-Central region of Brazil which were characterized in terms of soil chemical, physical and mineralogical aspects as well as the spatial variability of ^{137}Cs inventories. Some important differences in the patterns of ^{137}Cs depth distribution in the soil profiles of the different sites were found. They are probably associated to chemical, physical, mineralogical and biological differences of the soils but many questions still remain open for future investigation, mainly those regarding the adsorption and dynamics of the ^{137}Cs ions in soil profiles under tropical conditions. The random spatial variability (inside each reference site) was higher than the systematic spatial variability (between reference sites) but their causes were not clearly identified as possible consequences of chemical, physical, mineralogical variability, and/or precipitation.

Key words: cesium-137, spatial distribution, radioisotope, sediment, radioactive tracer

VARIABILIDADE ESPACIAL RANDÔMICA E SISTEMÁTICA DOS INVENTÁRIOS DE ^{137}Cs DE ÁREAS DE REFERÊNCIA NO SUDESTE DO BRASIL

RESUMO: A precisão da técnica de análise da redistribuição do “fallout” do ^{137}Cs nas avaliações das taxas de erosão são fortemente dependentes da qualidade de um inventário médio tomado em uma área de referência representativa. O conhecimento das fontes e do grau de variação da distribuição do “fallout” do ^{137}Cs desempenham um importante papel na aplicação desta técnica. Quatro áreas de referência foram selecionadas na região Sudeste do Brasil, sendo estas caracterizadas quanto aos aspectos físicos, químicos, mineralógicos do solo, assim como a variabilidade espacial dos inventários de ^{137}Cs. Houve algumas diferenças importantes no padrão da distribuição do ^{137}Cs em profundidade nos perfis de solo dos diferentes locais que, provavelmente, estão associadas às diferenças químicas, físicas, mineralógicas e biológicas dos solos estudados. Mas muitas questões ainda permanecem abertas para futuras investigações, principalmente em relação à adsorção e à dinâmica do ^{137}Cs no perfil dos solos tropicais. A variabilidade espacial randômica (dentro de cada local de referência) foi maior do que a variabilidade espacial sistemática (entre áreas de referência), porém as causas disto não foram claramente identificadas, talvez em conseqüência da variabilidade química, física e/ou mineralógica bem como da precipitação.

Palavras-chave: césio-137, radioisótopo, sedimento, traçador radioativo

INTRODUCTION

Atomic-bomb derived ^{137}Cs fallout has been widely used for the evaluation of soil erosion rates and patterns of soil redistribution within the landscape (Ritchie & McHenry, 1990; Walling, 1995; Basher et al., 1995). A basic premise for the use of the ^{137}Cs technique in soil erosion evaluation is that the ^{137}Cs inventories at the investigated points in the study site can be directly compared with the reference inventory (taken at a reference site) and that the residual values indicate the relative magnitude of erosion or deposition (Walling & Quine, 1990).

The ^{137}Cs inventory at the reference site (local, flat, uneroded, undisturbed, stable site) represents the accumulated atmospheric input per unit surface area, adjusted for radioactive decay. Therefore, the value attributed to the reference inventory plays a crucial role in converting ^{137}Cs measurements to estimates of rates of soil erosion or sediment deposition. The use of inaccurate reference inventories will result in biased and unreliable estimates of soil erosion and sediment deposition (Owens & Walling, 1996; Sutherland, 1996). Despite its crucial importance, there is very little information for the Southern Hemisphere on the spatial variability of ^{137}Cs within areas taken as reference sites.

This paper presents preliminary results from a study that is being carried out in the South-Central region of Brazil with the objective of evaluating the random and systematic spatial variabilities in reference ^{137}Cs inventories. Different potential reference sites, close to areas where the ^{137}Cs technique is being applied for erosion studies, were selected and sampled for a larger investigation. The study is of special interest for Brazilian conditions, where the ^{137}Cs activity in soils is very low. In such condition of very low ^{137}Cs activity, the variability in reference inventories can have even a larger contribution in the uncertainties associated to ^{137}Cs evaluations.

MATERIAL AND METHODS

General characteristics of the selected reference sites

Four reference sites, in Piracicaba, SP, were initially selected for this study. It is intended to extend the number of sites, as well as to expand the number of soil profiles per area, to improve the methodology in future studies. All four sites are under sub-tropical climate conditions, situated in a radius of about 50 km from Piracicaba, with a Cwa climate according to Köppen, which is dry during the winter and rainy during summer. Figure 1 shows a map of the relative positions of the four reference sites.

Reference site R_1 - The first reference site R_1 was taken in Piracicaba, SP (22°42'S, 47°38'W and 560 m of altitude.) The site is a 20,000 m^2 grass field of an old garden. More than 40 years ago the area was embanked in order to establish a very flat grass (*Paspalum notatum* Flugge) field to be used only for recreation. The precipitation is 1,253 mm per year and the soil is an embankment with 180 g kg^{-1} sand, 390 g kg^{-1} clay and 430 g kg^{-1} silt. From this reference site, five soil profiles were taken and analyzed. The sampling points are 25 m distant from each other in a transect that crosses the field in its flat portion.

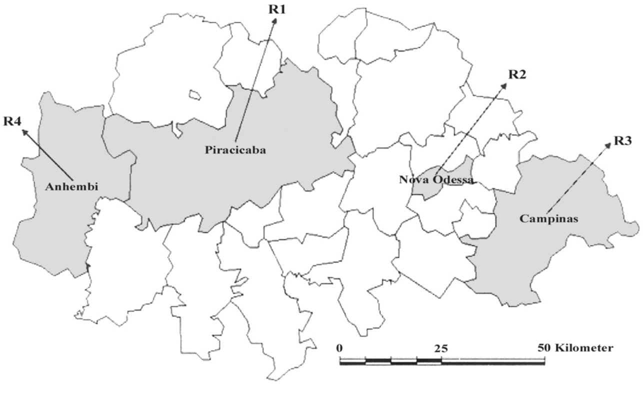

Figure 1 - Relative location of the four reference sites.

Reference site R2 - The second reference site R2 is situated at coordinates 22°47' S, 47°19'W and 600 m of altitude in the municipality of Nova Odessa, SP. The sampling place, with an area of about 30,000 m^2, is a pasture (*Brachiaria decumbens* Stapf) since 1979 after the deforestation and presents an average slope of 0.5%. The total annual precipitation in the region is 1,317 mm. The soil is an Arenic Paleudult with 520 g kg^{-1} sand, 140 g kg^{-1} clay and 340 g kg^{-1} silt. Six soil profiles from this site, taken along a transect, were sampled and analyzed. The distance between sampling points in the transect is 5 m.

Reference site R3 - The third reference site R3 is located at coordinates 22°09' S, 47°01'W and 700 m of altitude in the municipality of Campinas, SP. The total annual precipitation in the region is 1,400 mm. The selected sampling site is in a very flat area of 2,400 m^2 covered with a perennial legume, called "tropical kudzu" [*Pueraria phaseoloides* (Roxb) Benth.]. The area was seeded only once in 1943 and the local soil remained uncultivated since that time. The soil is classified as a Typic Hapludox with 380 g kg^{-1} sand, 380 g kg^{-1} clay and 240 g kg^{-1} silt. Five profiles from this area were sampled and analyzed. The sampling points were taken on a transect with a distance of 5 m between points.

Reference site R4 - The fourth reference site R4 is located in the municipality of Anhembi, SP. It is located at coordinates 22°40'S, 48°10'W and 460 m of altitude. The local annual precipitation is 1,100 mm. The select area of 20,000 m^2 has an average slope of 0.5% and the soil is a Typic Quartzipsamments with 850 g kg^{-1} sand, 60 g kg^{-1} clay and 90 g kg^{-1} silt. The site was deforested more than 40 years ago for the establishment of a pasture (*Brachiaria decumbens* Stapf). Six soil profiles, selected in a randomized way, in a distance of about 10 m from each other, were taken and analyzed.

Soil sampling and ^{137}Cs analysis

The sampling device (Figure 2) used allows the collection of soil samples in 5 cm thickness layers, down to the 50 cm depth in the soil profile, covering a surface area of 672 cm^2, which corresponds to a sampling volume of 3,360 cm^3 for each 5 cm layer.

Soil samples were analyzed for ^{137}Cs activity using a HPGE Coaxial Detector (GEM-20180P, PopTop) with a detection efficiency of 0.7%[1] for the adopted geometry (1 litter Marinelli Beakers) and a minimum de-

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[1] Detector efficiency determined using a IAEA standard soil sample presenting a known activity of 2.97 Bq kg^{-1}.

tectable activity of the order of 0.2 Bq kg^{-1}. Due to the very low ^{137}Cs activity of the soil samples and the very low detection efficiency, the counting time varied from 24 to 56 hours according to soil sample activities in order to assure an acceptable integration error (<10%) during the spectrum analysis.

The soils of the four sites studied were analyzed for chemical and physical properties in depth in order to look for correlations between ^{137}Cs distribution and other soil property, related to the dynamics of the cesium.

RESULT AND DISCUSSION

The average inventories (Table 1) were compared using the statistical t test. At site R1 the inventory was higher ($P < 0.01$) than, the other three sites. No differences were found between average inventories of the three other sites (R2, R3 and R4). In R1 inventories varied from 277 to 367 Bq m^{-2}. The ^{137}Cs activities decreased from 302 Bq m^{-2} in the upper 15 cm of the soil profile to 11 Bq m^{-2} in the 15-25 cm layer. No ^{137}Cs activity was detected below the 20-25 cm layer.

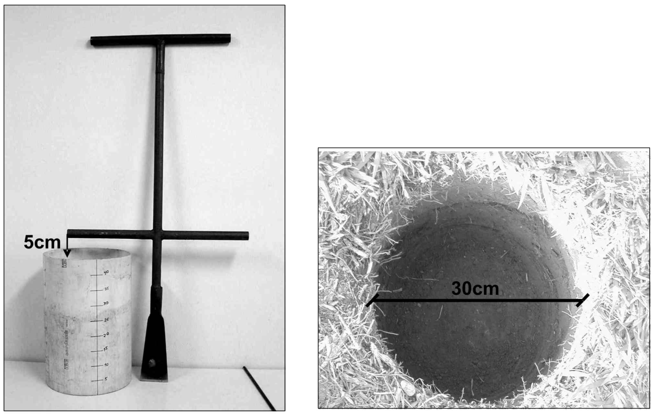

Figure 2 - Equipment used for sampling soil profiles at reference sites.

At site R2 the inventories varied from 163 to 393 Bq m^{-2} and 91% of the ^{137}Cs was found in the first upper 20 cm of the soil profile. At site R3 inventories varied from 177 to 291 Bq m^{-2} with more than 98% of the total ^{137}Cs activity in the upper 20 cm of the soil profile. For R4 the inventories varied from 205 to 290 Bq m^{-2}. The ^{137}Cs activities presented a smooth reduction in depth from the 15-20 cm layer down to 45 cm depth below which no radioisotope was detected.

The conditions found on the site R1, both the ^{137}Cs distribution in the soil profiles and smaller variation between the inventories (very low CV), are closer to the expected and desired conditions of an adequate reference site. In the sandy soil of site R4 ^{137}Cs activity decrease in depth was also observed but the ^{137}Cs distribution in the soil profile was more uniform when compared to the distribution of the other sites.

The average ^{137}Cs activity distribution found in the soil profiles of the four sites is in accordance to reference sites described in the literature (Owens & Walling, 1996; Walling & Quine, 1992; Walling, 1995). In non disturbed soil profiles the higher ^{137}Cs activity is found in the upper 20 to 25 cm of the soil profile (Basher et al., 1995).

The organic matter content of the soil at site R3 is higher ($P < 0.001$) than those of the other sites (Table 2). The same behavior was observed for soil pH. The

Table 1 - Average vertical distribution of ^{137}Cs inventory (Bq m^{-2}) of the reference sites.

Layer	Average ^{137}Cs activity											
	R1			R2			R3			R4		
	p = 5			p = 6			p = 5			p = 5		
	Average	SD	CV	Average	SD	CV	Average	SD	CV	Average	SD	CV
cm	Bq m^{-2}											
0-5	145	30	21	73	24	33	108	32	30	47	12	22
5-10	109	26	24	60	10	17	86	28	33	42	8	23
10-15	48	17	34	54	15	27	41	15	37	45	11	37
15-20	9	12	137	41	21	53	14	10	68	38	11	43
20-25	2	5	224	17	12	72	4	5	141	23	6	34
25-30	0	0	0	4	4	90	0	0	0	16	5	41
30-35	0	0	0	1	2	215	0	0	0	12	5	41
35-40	0	0	0	0	0	0	0	0	0	9	1	9
40-45	0	0	0	0	0	0	0	0	0	9	0	3
45-50	0	0	0	0	0	0	0	0	0	0	0	0
Total	314	34	11	250	77	31	253	46	18	242	37	15

SD = standard deviation, Bq m^{-2}; CV = coefficient variation, %; *p* = number of analyzed soil profiles

Table 2 - Chemical and mineralogical analysis of the soils from the reference sites (average for profiles).

Parameter	Site			
	R1	R2	R3	R4
	p = 5	p = 6	p = 5	p = 6
pH, 0.01 mol L^{-1} ($CaCl_2$)	4.53	4.49	5.55	4.62
OM, g dm^{-3}	14.54	13.79	31.50	12.12
P^+, mg kg^{-1}	45	4	4	6
K^+, $mmol_c$ kg^{-1}	1.40	0.67	1.60	0.27
Ca^{+2}, $mmol_c$ kg^{-1}	42	13	55	10
Mg^{+2}, $mmol_c$ kg^{-1}	11	6	11	5
Al^{+3}, $mmol_c$ kg^{-1}	5	4	0	3
H+Al, $mmol_c$ kg^{-1}	42	27	20	19
SB, $mmol_c$ kg^{-1}	54	19	67	15
CEC, $mmol_c$ kg^{-1}	96	47	87	35
V, %	56	41	74	44
m, %	10	19	0	17
Fe_2O_3, g kg^{-1}	nd	15.4	173.0	5.8
Al_2O_3, g kg^{-1}	nd	41.2	182.0	24.0
SiO_2, g kg^{-1}	nd	82.9	111.5	30.7

p = number of soil profiles analyzed. **nd** = not determined.

amount of potassium of the soil of R3 was similar to the soil of R1 but much higher than that of the soils of R2 and R4. The amounts of oxides in the soil of R3 were also higher in relation to the other soils. Table 3 shows the difference of silt content of the soils ($P < 0.001$).

The amount of organic matter is positively correlated with ^{137}Cs activity for soil sites R1 ($R^2 = 0.94$; $P < 0.01$), R2 ($R^2 = 0.93$; $P < 0.001$) and R4 ($R^2 = 0.86$; $P < 0.001$), while for site R3 the correlation was not good ($R^2 = 0.52$; $P < 0.1$) (Figure 3). Another correlation found was between ^{137}Cs activity and Ca^{+2} concentrations ($P < 0.001$) for site R4. Although some correlations have been identified, it is not sure that there are cause/effect relationships between them. Roque (2003) reports that tropical soils present specific exchange sites for ^{137}Cs, mainly in the silt fraction (Roque et al., 1998), and that the humic organic matter does not present this kind of adsorption sites.

Wässerman et al. (2002) and Sheppard & Thibault (1992) observed that most of the ^{137}Cs of fallout seems to be associated to soil oxides. Unfortunately in the present study the concentration of oxides was determined only for the upper 0-20 cm soil layer and therefore it was not possible to analyze its possible correlation with ^{137}Cs activities.

Table 3 - Physical analysis of the soils from the reference sites (average for profiles).

Site	p	Soil fraction							
		TS	VCS	CS	MS	FS	VFS	SILT	CLAY
		%							
R1	5	18	2	3	4	6	3	43	39
R2	6	52	0	3	15	22	11	34	14
R3	5	38	1	6	16	11	4	24	38
R4	6	85	0	2	28	43	12	9	6

p = number of soil profiles analyzed. TS: total sand (2-0.053 mm); VCS: very coarse sand (2-1 mm); CS: coarse sand (1-0.5 mm); MS: medium sand (0.5-0.25 mm); FS; fine sand (0.25-0.125 mm); VFS: very fine sand (0.125-0.053 mm); SILT(0.053-0.002 mm) and CLAY (<0.002 mm).

The recommendation that reference sites should be taken in places that have not been cultivated or mechanically disturbed since the ^{137}Cs fallout is well known. In such condition, it is expected that the plant nutrients as well as the ^{137}Cs have higher concentrations in the upper first centimeters of the soil profile. It is also expected that the concentrations of such elements decrease exponentially in depth. Such behavior was observed in the soils of the sites R1, R2 and R4 but not in R3. Some facts can explain these results. At areas R1, R2 and R4 the soils were in grass whose root system is composed basically of fine roots that are concentrated in the upper 20 cm of the soil profile. The depth organic matter distribution in these three sites (Figure 3) is very different from that found for site R3, which has been covered for 60 years with a perennial legume (*Pueraria phaseoloides*) that presents a root system composed of a main deep tap-root and many secondary roots. The presence of ^{137}Cs in deeper soil layers in the site R3 could be associated to internal

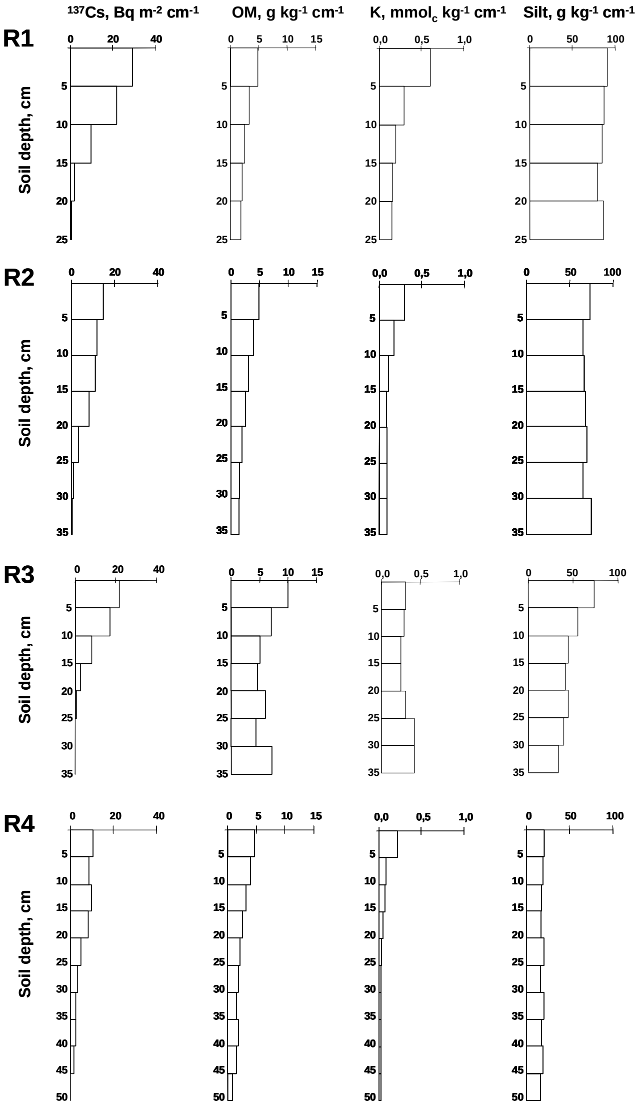

Figure 3 - Depth distribution of ^{137}Cs activities (^{137}Cs, Bq m^{-2} cm^{-1}), organic matter (OM, g kg^{-1}cm^{-1}), potassium (K, mmol$_c$ kg^{-1} cm^{-1}) and silt fraction (Silt, g kg^{-1} cm^{-1}) in the four reference sites.

soil redistribution in the soil profile which could have been occurring in large and continuous pores left after the decomposition of these large deep roots, as well as by the action of large amount of soil organisms present in the area.

The relatively uniform depth distribution of organic matter at site R3 explains the lack of a significant correlation between this variable and the ^{137}Cs activity. The presence of ^{137}Cs in deeper soil layers in the sandy soil profile of site R4 is probably associated with soil texture and structure of the Typic Quartzipsamments. This soil presents high permeability and fast drainage due to its high macro-porosity (Resende et al., 1988) and as a consequence they present higher nutrient leaching susceptibility as compared to clay soils (Raij, 1991).

This study showed some important differences in the pattern of ^{137}Cs depth distribution in soil profiles of reference sites. They are probably associated with the chemical, physical, mineralogical and biological differences of the studied soils but many questions still remain open for future investigation, mainly those regarding the adsorption and dynamics of the ^{137}Cs in soil profiles under tropical conditions.

Taking into account that the sampling and analytical procedures were the same for all soil profiles and that the sample ^{137}Cs activities were of the same order of magnitude, the observed variability can be attributed to:

a) random spatial variability (inside each reference site) – this variability, expressed by the coefficient of variation (CV%) of the average of the inventories of each site, varied from 11%, observed for site R1, to 31%, for site R2. Using the arbitrary categories established by Wilding & Drees (1983), mentioned by Sutherland (1996), these CV values would place the ^{137}Cs random spatial variability in a category of moderate variability, in the same class of other soil characteristics such as the clay and organic matter content (Wilding & Drees, 1983; Dahiya et al., 1984). Therefore the results observed in the present study are in accordance with other authors (Owens & Walling, 1996). These results indicate that the common simple assumption that ^{137}Cs is uniformly distributed in undisturbed landscapes (Ritchie & McHenry, 1990) needs detailed examination.

b) systematic spatial variability (between reference sites) – this variability, expressed by the coefficient of variation (CV%) of the average of the inventories of the four sites, was of 11%. The average inventory of the four sites was 268 ± 30 Bq m^{-2}. The amplitude of variation was from 250 Bq m^{-2} (site R2) to 314 Bq m^{-2} (site R1). Flores et al. (2001) found an average of 491 ± 15 Bq m^{-2} (CV = 3%). The statistically higher average inventory found in the site R1, could be attributed to systematic differences of precipitation, soil and vegetation types, as mentioned by Owens & Walling (1996). It is well known that in tropical regions the precipitation can present a high variability at short distances when they are analyzed for short periods of time and that this variability can be very small when analyzed over the long run (Reichardt et al., 1995). Taking into account that the annual precipitation values for the studied sites are very close, it is difficult to interpret the real effect of the rainfall variability on the observed ^{137}Cs fallout systematic spatial distribution.

The variability of the soil physical and chemical characteristics was not able to explain the random ^{137}Cs spatial variability which was higher than the systematic spatial variability. Since the use of the ^{137}Cs technique for erosion rate evaluation is based on average reference inventories it is clear that all data interpretation will depend strongly on the spatial distribution pattern of the ^{137}Cs fallout in the selected reference site. Based on the observed systematic spatial variability, it is also clear that the reference site must be as close as possible of the study area.

REFERENCES

BASHER, L.R.; MATTHEWS, K.M.; ZHI, L. Surface erosion assessment in the South Canterbury downlands, New Zealand using ^{137}Cs distribution. **Australian Journal of Soil Research**, v.33, p.787-803, 1995.

DAHIYA, I.S.; RICHTER, J.; MALIK, R.S. Soil spatial variability: a review. **International Journal of Tropical Agriculture,** v.11, p.1-102, 1984.

DAVIS, J.J. Cesium and its relationship to potassium in ecology. In: SHULTZ, V.; KLEMENT JR., A.W. (Ed.) **Radioecology**. New York: Reinhold, 1963. p.539-556.

FLORES, O.B.; BENITEZ, J.E.G.; ESTRADA. A.M. Estimación de la tasa de erosión en suelos agrícolas mediante el radioisótopo cesio-137. In: CONGRESSO LATINOAMERICANO, 25; CONGRESSO CUBANO DE LA CIENCIA DEL SUELO, 5., Varadero, 2001. 1 CD-ROM.

LIVENS, F.R.; LOVELAND, R.J. The influence of soil properties on the environmental mobility of caesium in Cumbria. **Soil Use and Management**, v.4, p.69-75, 1988.

OWENS, P.N.; WALLING, D.E. Spatial variability of caesium-137 inventories at reference sites: an example from two contrasting sites in England and Zimbabwe. **Applied Radiation and Isotopes,** v.47, p.699-707, 1996.

RAIJ, B. van. **Fertilidade do solo e adubação**. Piracicaba: CERES; POTAFOS, 1991. 343p.

REICHARDT, K.; ANGELOCCI, L.R.; BACCHI, O.O.S.; PILOTTO, J.E. Daily rainfall variability at a local scale (1,000 ha), in Piracicaba, SP, Brazil, and its implications on soil water recharge. **Scientia Agricola**, v.52, p.43-49, 1995.

RESENDE, M.; CURI, N.; SANTANA, D.P. **Pedologia e fertilidade do solo**: interações e aplicações. Lavras: ESAL; Piracicaba: POTAFOS, 1988. 83p.

RITCHIE, J.C.; MCHENRY, J.R. Application of radioactive fallout cesium-137 for measuring soil erosion and sediment accumulation rates and patterns: A review. **Journal of Environmental Quality**, v.19, p.215-233, 1990.

ROQUE, M.L. Sorção de radiocésio em solos tropicais. Medidas de contenção e de remediação de solos contaminados por ^{137}Cs. Piracicaba:USP/CENA, 2003. 152p. (Tese – Doutorado).

ROQUE, M.L.; TAGLIAFERRO, F.S.; FERNANDES, E.A.D.N. Comportamento de ^{137}Cs e elementos terrígenos em solos e respectivas frações finas. In: CONGRESSO GERAL DE ENERGIA NUCLEAR, 7., Belo Horizonte, 1998. Belo Horizonte: ABEN, 1998.

SHEPPARD, M.I.; THIBAULT, D.H. Desorption and extraction of selected heavy metals from soils. **Soil Science Society of America Journal**, v.56, p.415-423, 1992.

SUTHERLAND, R.A. Caesium-137 soil sampling and inventory variability in reference locations: a literature survey. **Hydrological Processes**, v.10, p.34-54, 1996.

TAMURA, T. Selective sorption reactions of cesium with soil minerals. **Nuclear Safety**, v.5, p.262-268, 1964.

WALLING, D.E. Use of ^{137}Cs and other fallout radionuclides in soil erosion investigations: progress, problems and prospects. In: INTERNATION ATOMIC ENERGY AGENCY. **Use of ^{137}Cs in the study of soil erosion and sedimentations**. Vienna: IAEA, 1995. (TECDOC, 1028).

WALLING, D.E.; QUINE, T.A. Calibration of caesium-137 measurements to provide quantitative erosion rate data. **Land Degradation and Rehabilitation**, v.2, p.161-175, 1990.

WALLING, D.E.; QUINE, T.A. The use of caesium-137 measurements in soil erosion surveys. In: EROSION AND SEDIMENT TRANSPORT MONITORING PROGRAMMERS IN RIVER BASINS, Oslo, 1992. **Proceedings**. Oslo: IAHS, 1992. (Publication, 210).

WÄSSERMAN, M.A.M.; PEREZ, C.V.; BOURG, A.C.M. Behavior of cesium-137 in some Brazilian soils. **Communications in Soil Science and Plant Analysis**, v.33, p.1335-1349, 2002.

WILDING, L.P.; DREES, L.R. Spatial variability and pedology. In: WILDING, L.P.; SMECK, N.E.; HALL, G.F. (Ed.) **Pedogenesis and taxonomy concepts and interactions**. Amsterdam: Elsevier, Amsterdam, 1983. p.83-116.

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