UPTAKE OF RADIONUCLIDES BY PLANTS GROWING ON BRAZILIAN SOIL: THE EFFECT OF SOIL AGEING

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Abstract. The behaviour of radionuclides in soil is governed by several mechanisms that can vary significantly according to the specific reactivity of each element and soil properties. The ^{137}Cs is one of radionuclides that generally reduces with time its mobility and phytoavailability due to irreversible fixation in high activity clay mineral such as illite, vermiculite and montmorilonite. A long-term experimental essay using Brazilian soils was done in order to determine the effect of ageing of contamination on ^{137}Cs mobility in soils and transfer to plants. To perform this study, 4 different soils with different properties were contaminated with ^{137}Cs at different period: The older contamination refers to an urban soil contaminated at the Goiânia accident (1987). A similar type of Goiânia's soil (Ferralsol rich in Gibbsite) was artificially contaminated with ^{137}Cs in 1993. A subtropical class of soil (Nitisol) was contaminated in 1996 and two other tropical soils were contaminated in 2000 (Acrisol and Ferralsol rich in Goethite). The time's effect was studied by characterizing the evolution of soil properties and the changes in the distribution of radionuclides between phases till 2006. In addition, the phytoavailability was evaluated by carrying out experiments in lysimeters where radish was sowed at different periods:1996, 2000 and 2004. These results showed that the phytoavailability changed with time only in 2 situations: after changes in some soil properties such as pH or due to Cs fixation in high activity clay mineral when it was present in the soils even as trace mineral. The ^{137}Cs distribution in soil showed that Fe oxides are the main sink for this element in all type of soil and 14 years after contamination, the ^{137}Cs was still available for plants in the Ferralsol Gbbiste rich. In the Nitisol, 5 years after contamination, the ^{137}Cs was not detected as in the slightly acidic phase of sequential extraction neither detectable in radish roots or leaves, indicating strong sorption mechanism, only explained by the presence of traces of vermiculite in these soils. Older contamination or younger contamination than contamination in Nitisol did not reduced plant uptake in that extreme way. These results suggest that soils originated from highly weathered areas, identified by the absence of high activity clay minerals and low fertility, the ^{137}Cs uptake by plants can remains high for a long period.

KEYWORDS: radioecology; soil contamination; Cs-137;

1. Introduction

Environmental studies after the accident of Chernobyl contributed to improve the knowledge on the behavior of artificial radionuclide in temperate areas, supplying consistent information to the management of contaminated agricultural areas. In Brazil, the Goiânia accident with ^{137}Cs in 1987, pulled to reinforce the knowledge related to the management of urban contaminated areas and remedial actions, whereas for tropical agricultural areas very few information was available at that time.

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The behavior of radionuclides in soil is governed by several mechanisms that can vary significantly according to the specific reactivity of each element and soil properties. The ^{137}Cs is one of the radionuclides that generally reduces with time its mobility and phytoavailability due to irreversible fixation in high activity clay mineral such as illite, vermiculite and montmorilonite.

Highly weathered soils as Ferralsol and Acrisol are predominant in the Brazilian territory, occupying extensive cultivated areas. They represent important natural reservoirs, with a great potential role for solving problems associated with to the food production and environmental protection.

Since 1993 at the Environmental Impact Assessment Service of the Institute of Radioprotection and Dosimetry, experimental studies were carried out on Brazilian soils artificially contaminated with ^{137}Cs, ^{60}Co and ^{90}Sr and an urban soil contaminated with ^{137}Cs from the Goiânia accident [1, 2, 3, 4, 5, 6, 7, 8, 9]. These studies integrated results of soil to plant transfer factor for several crop groups growing in lysimeters or pots, with results of experiments conducted in laboratory, to understand the main components or properties of soils involved on the mechanisms of sorption for a given radionuclide.

A long-term experimental essay using Brazilian soils was done in order to determine the effect of ageing of contamination on ^{137}Cs mobility in soils and transfer to plants. To perform this study, 3 different soils with different properties were contaminated with ^{137}Cs at different periods. The effect of time was studied by characterizing the evolution of soil properties and the changes in the distribution of radionuclides between phases till 2006.

2. Materials and methods

Lysimeters measuring 1 m^2 of area and with a depth of 1 m were installed in a restricted area of the Institute for Radioprotection and Dosimetry (CNEN/Brazil). The lysimeters were filled, from bottom to top, with: 15 cm of coarse material (sand and gravel), 30 cm of uncontaminated soil and 40 cm of contaminated soil. Each lysimeter received a liter of solution containing approximately 60 $\mu Ci\ L^{-1}$ of ^{137}Cs. The solution was sprayed in every 2 cm of soil, layer by layer, up to 40 cm.

Since 1993, plants have been cultivated in Ferralsol (gibbsite rich Ferralsol- Al), Nitisol (collected in the South of Brazilian territory under subtropical climate rich in goethite with traces of vermiculite), and one urban soil (gibbsite rich) contaminated in 1987 with ^{137}Cs in the accident of Goiânia (refereed to as Goiânia soil). The values of soil activity and year of contamination for ^{137}Cs are shown in Table 1.

Table 1. ^{137}Cs mean activity in soil and year of contamination.

	Goiânia soil	Ferralsol-Al	Nitisol subtropical
^{137}Cs ($Bq.kg^{-1}$ dry weight)	1.8E+3 ± 7.7E+2	7.5E+3 ± 7.8E+2	10.1E+3 ± 8.1E+2
Year of contamination	(1987)	(1992)	(1996)

The Soils Office of the Brazilian Agricultural Research Enterprise (EMBRAPA-Solos) performed the soil analyses, according to their standards manual [10]. The soil to plant transfer factors (TF) were determined as the ratio of the activity in the edible parts of the plant (given in Bq kg^{-1} dry weight) and the activity in the first 20 cm of the soil (given in Bq.kg^{-1} dry weight), following the International Union of Radioecologists recommendations for the contamination and crop treatments [11].

Samples of soil were collected in 1996 (Ferralsol and Goiânia soil), 2000 (Ferralsol, Nitisol and Goiânia soil) and 2004 (Ferralsol, Nitisol and Goiânia soil) to perform sequential extraction analyses. Sequential extractions were performed following Tessier *et al.* protocol [12] for samples collected in 1996 (method 1) and following a modified version for tropical soil (Method 2, [7]), for samples collected in 2000 and 2004 (Table 2). For the sequential chemical extraction, 3g of soil samples were used in duplicates.

Table 2. Geochemical phases and geochemical meanings of considered protocols.

Geochemical phases Considered	Geochemical meanings and phases of different protocols
Slightly Acid	Method 1: bound to carbonates + exchangeable phase Method 2: potentially phytoavailable
Reducible	Method 1: bound to Fe and Mn oxides Method 2: bound to Mn oxides and bound to resistant organic matter or iron compounds (alkaline phase)
Oxidizable	Method 1: bound to organic matter Method 2: bound to organic matter
Residual	Method 1: incorporated to crystal structure Method 2: not potentially available for transfer processes.

Plants and soils were dried, ground and sieved through a 2 mm screen before direct measurements of the of ^{137}Cs activity by gamma ray spectrometry using a Ge detector Ortec. Efficiency was known as to have an accuracy of about 10% for the used geometry (pot of 250 g). Counting errors were less than 3%.

3. Results

The Ferralsol together with Acrisol, represent more than sixty percent of agricultural Brazilian soils. Their main restrictions for some crops are generally the low nutrient contents and acidity, as showed in Table 3.

One of the most important characteristics of highly weathered soils is the predominance of 1:1 clay minerals type and Fe, Al and occasionally, Mn oxides in the clay fraction, while the 2:1 clay mineral is generally absent. Among the studied soils, the Nitisol, was the only soil with traces of vermiculite (Table 3). This 2:1 phyllosilicate is recognized to be responsible for specific adsorption of ^{137}Cs and for its strong fixation, reducing the soil to plants transfer with time [13]. As so, it is important to note that the Goiânia soil did not show reduction of ^{137}Cs TF for radish with time, even 17 years after soil contamination (Fig. 1), and indeed, a small increase in TF was associate to natural decrease of pH and Exchangeable K content occurred in this soil since the contamination event, 8.3, to 7.7 in 2004, as presented in table 3.

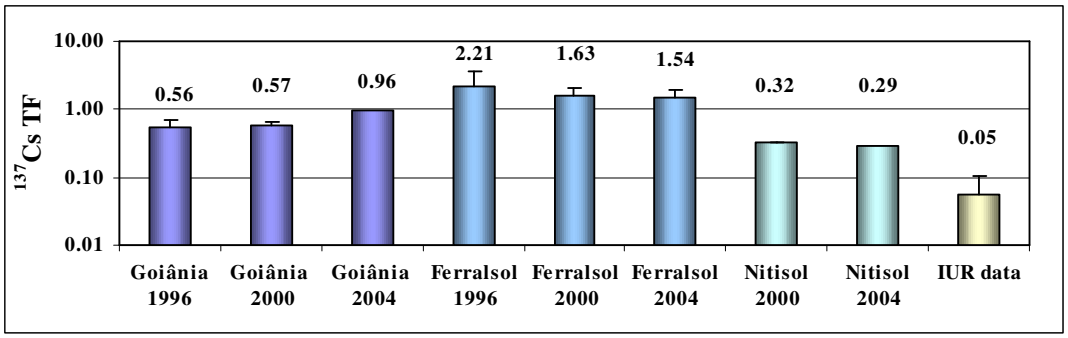
Table 3. Pedological analyses.

	Goiânia soil	*Ferralsol*	*Nitisol subtropical*
Exchangeable K *(cmol kg^{-1})*	**0.25-0.11***	0.12	0.18
Exchangeable Ca *(cmol kg^{-1})*	4.0	3.9	4.0
CEC (cmol kg^{-1})	4.8	4.8	6.1
pH in KCl	**8.3-7.7***	**4.3-7.4***	4.2
OM (%)	1.9	2.1	2.1
Fe_2O_3 (%)	5	3	13
Clay (g kg^{-1})	160	180	490
Sand (g kg^{-1})	680	710	100
Main clay mineral type	gibbsite, kaolinite	gibbsite, kaolinite	hematite, goethite, vermiculite

*natural or artificial changes in properties.

On the other hand, the decrease in the ^{137}Cs TF, observed at Ferralsol, from1996 to 2000, was associated with change in soil pH due to liming: from a pH = 4.8 before 1996 up to 7.4 at subsequent years (Fig. 1).

Figure 1. ^{137}Cs Soil to plant transfer Factor for radish in different years.

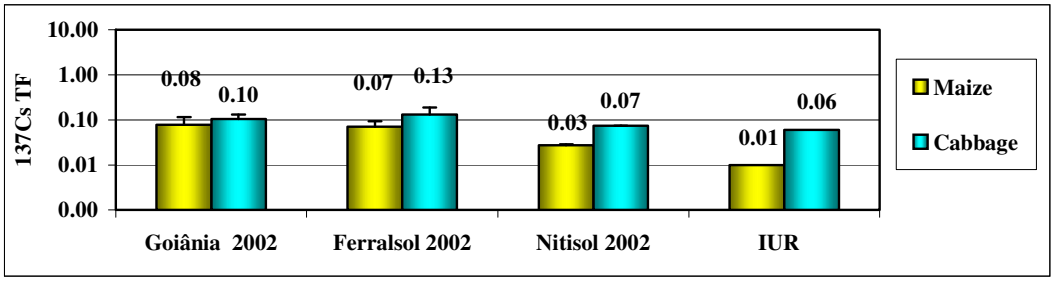


More than 10 years after the contamination, the Goiânia soil and Ferralsol still present ^{137}Cs TF for radish more than one order of magnitude higher than TF recommended by IUR to be used for dose assessment in the absence of regional values [11]. Higher TF was also observed for maize and cabbage in these soils (Figure 2).

On the other hand, the lower ^{137}Cs TF that occurred in Nitisol (subtropical soil), independent of time elapsed from the contamination or crop type (Fig. 1 and 2), indicated a different mechanism in the control of the radionuclide behavior compared with the other tropical soils. This is probably due to the presence of vermiculite, although as a trace compound.

Despite of this natural high vulnerability to ^{137}Cs contamination, Ferralsol respond quite well to agricultural practices like liming and organic amendments as demonstrate by WASSERMAN et al. [8], reducing its soil to plant transfer factors.

Figure 2. ^{137}Cs Soil to Plant Transfer Factor for Maize and Cabbage.



The Figures 3 and 4 presents, respectively, the results of sequential chemical extraction in different years after contamination for Goiânia and Ferralsol.

Figure 3. Geochemical partitioning of ^{137}Cs in Goiânia soil.

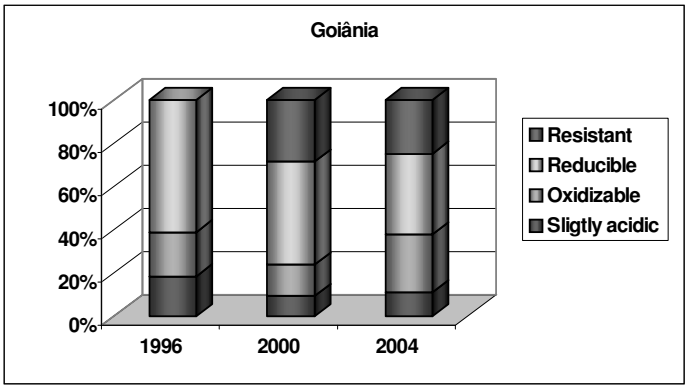
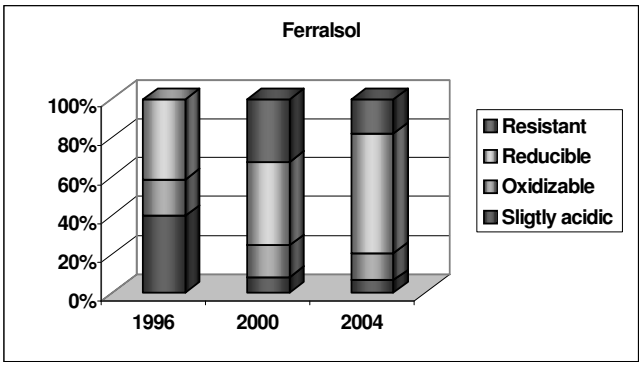


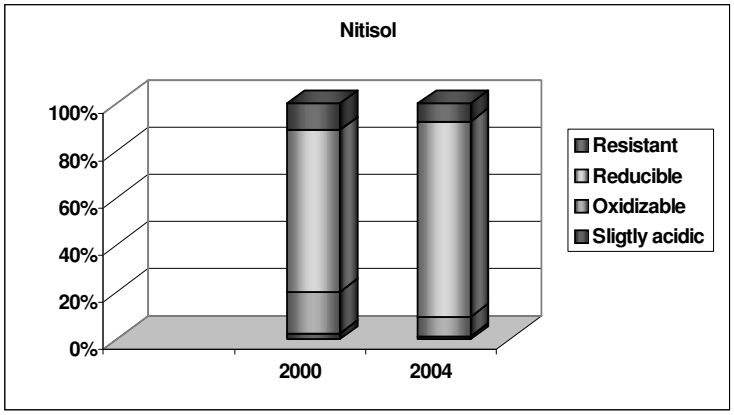
Figure 4. Geochemical partitioning of ^{137}Cs in Ferralsol soil.



The geochemical partitioning of ^{137}Cs, revealed similar partition between Ferralsol and Goiânia in 2000, consistent with similar soil properties at these data and similar TF values for maize and cabbage. The geochemical partitioning of ^{137}Cs in tropical and subtropical soils observed the important affinity of this radionuclide for Fe oxy-hydroxides (reducible phase) constituting an important sink.

In the case of the Nitisol, the alkaline phase can retain 67% of added ^{137}Cs. According to these results, around 10% of ^{137}Cs in soil was still easily available for root uptake 10 years after contamination in the Ferralsol, while very low percentage of this radionuclide (3%) was available for root uptake in the Nitisol (Fig. 5 slightly acid phase) in the 4^{th} years after contamination, what was also corroborated by the low TF observed in this soil for all crops (Fig. 1 and 2). This result indicates strong sorption mechanism in Nitisol, only explained by the presence of traces of vermiculite in these soils, as observed soils from temperate climate countries where an important decrease in transfer factors for ^{137}Cs is observed with time, reflecting a the strong fixation of the contamination in the soil [13].

Figure 5. Geochemical partitioning of ^{137}Cs in Nitisol soil.



Older contamination in Nitisol did not reduce plant uptake in that extreme way. These results suggest that soils originated from highly weathered areas, identified by the absence of high activity clay minerals and low fertility, the 137Cs uptake by plants can remains high for a long period.

4. Conclusions

These results demonstrate that well weathered soils, depleted of high activity clay minerals, such as vermiculite, were the most vulnerable soil for radioactive contamination, but for these areas, good agricultural practices guaranty better radiological protection, since it improves crop production and minimizes transfer process on soil system.

The environmental studies also stressed the relevance of the use of site specific data for environmental impact assessments and to the decision making process after an accidental contamination, that need to be based on the knowledge of environmental

processes governing the long term behavior of radionuclides at a specific type of environment.

This is particularly important to tropical areas because of the different soil classes used for agricultural purposes, as compared to temperate climate conditions, for which most literature data has been historically developed.

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