



## Soil to Plant Transfer of Sr-90 and Stable Sr in Brazilian Soils

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### 1. Introduction

In many countries, public concerns over environmental protection related to nuclear activities and nuclear waste disposal has increased in recent years due to their worries about potential accidents. The 2011 Fukushima-Daiichi nuclear accident in Japan revived such worries related to the nuclear sector. To assess environmental impact and risks associated with radioactivity, links must be established between the source term and deposition, ecosystem transfer, biological uptake and effects in exposed organisms [1]. Following a nuclear accident there may be social and economic impacts in agricultural area due to contamination of food products. In order to help predict the concentrations of radionuclides in crops and animal fodders growing on contaminated land by, the concept of the ‘transfer factor’ (or concentration ratio) was developed and widely applied in radiological assessment models [2,3,4]. The soil-plant transfer factor ( $F_v$ ) is therefore a critical parameter for dose calculation. The  $F_v$  relates the radionuclide activity concentration in edible parts of the plants ( $\text{Bq kg}^{-1}$  dry weight) to that in soil ( $\text{Bq kg}^{-1}$  dry weight) [5]. Different soil types, plants, and agricultural practices result in large variation of  $F_v$  values for a given element, even for a single plant species. Properties of highly weathered soils can be important in explaining the fate of radionuclides in tropical soils. For instance, Brazilian soils, mostly acid iron/aluminium hydroxides rich soils, have  $^{137}\text{Cs}$   $F_v$  values, one or two order of magnitude higher than those for agricultural soils in temperate climates. In the long-term after contamination, contrary to temperate mineral soils, fixation is low for  $^{137}\text{Cs}$  in some tropical soils [6,7]. The relationship between soil properties and the soil-plant transfer factor for  $^{90}\text{Sr}$  is complex. However, some studies have found correlations between  $^{90}\text{Sr}$   $F_v$  and soil parameters such as exchangeable calcium (Ca) and potassium (K), CEC, pH and organic matter content [8,9,10,11,12]. Most studies on radiostrontium root uptake show that it is influenced by strontium bioavailability in the soil solution, the availability of competing ions in

the soil solution and plant physiology.

In this paper, the investigation of the behaviour of  $^{90}\text{Sr}$  focused in the soil to plant transfer factor and pedological properties. The use of stable Sr data to understand the  $^{90}\text{Sr}$  behaviour in soil - plant systems is also considered.

## 2. Methodology

### *Experimental set-up*

The study was conducted in an experimental area at the Institute of Radioprotection and Dosimetry (IRD; Rio de Janeiro, Brazil) where lysimeters of 1m<sup>3</sup> each, were constructed. These lysimeters were filled, from bottom to top, with 15 cm of sand and gravel (for drainage), 30 cm of soil physically isolated by a nylon-latticed membrane from a further 40 cm of the same soil at the top.

For this study, according to the World Reference Base for Soil Resources- WRB [13], the following class of soils were selected: Ferralsol, Acrisol, and Nitisol. As a consequence of heavy rainfall and high temperatures occurring in tropical regions, soil profiles are deeper (greater than 1.5 m), and the upper layer (A Horizon) is depleted of more complex clay minerals such as illite, montmorillonite and vermiculite.

Ferralsols are strongly weathered soils, with a dominance of sesquioxides and kaolinite, considered to be the 'classic' soils of the humid tropics. They are chemically poor, but physically stable. Due to the large distribution of this soil in Brazil, five lysimeters were filled with the Ferralsol from a savannah area of Brazil, collect at Abadia de Goiás (Goiás, Brazil - 16° 45' 26" S; 49° 26' 15" W), and two lysimeters were filled with Ferralsol, collected at Lavras (Minas Gerais, Brazil - 21° 14' 43" S; 44° 59' 59" W). The former soil, has similar properties to the Ferralsol collected at Abadia de Goiás, but with higher content of iron oxides (between 18% and 36%) responsible for its reddish coloration [14] In this paper, the Lavras's soil will be referred as Ferralsol-Fe and the Ferralsol from the Abadia de Goiás, will be referred as Ferralsol-Al, due to the higher content of aluminium oxides (gibbsite).

The Acrisol, collected at Campos (Rio de Janeiro, Brazil- 21° 45' 15" S; 41° 19' 28" W) have a textural B-horizon (Bt) with low activity clay. The Bt horizon was formed by the long-term process of clay translocation from horizon A to B, and consequently, the horizon A of this soil is usually quite sandy [14]. Ferralsols together with Acrisols represents more than 60% of agricultural soils in Brazil. However, their low nutrient content and acidity restricts their usage for some crops, explaining why just one lysimeter was filled with Acrisol.

The Nitisol was collected from the subtropical south region of Brazil, at the São Borja rural area (Rio Grande do Sul, Brazil - 28° 39' 38" S; 56° 00' 16" W). This class of soil usually presents higher content of nutrients, compared to Ferralsols and Acrisols [14]. This type of soil, despite of its relevance for agricultural purposes, its distribution is secondary at tropical area, compared with Ferralsol and Acrisol, and because of that, just one lysimeter was filled with this soil type.

Contamination of the soils with  $^{90}\text{Sr}$  was conducted by irrigating with a solution containing  $^{90}\text{Sr}$  directly onto the upper layer of the soil in the lysimeter. This method of contamination is not ideal for a homogenous distribution, but as these soils were already in place with other radionuclides representing a historical register, it was decided to apply a new contaminant with a minimal disturbance of the soil profile. Indeed, the mean specific activity for  $^{90}\text{Sr}$  ranged from  $2.65\text{E}+02 \pm 5.56\text{E}+01$  to  $9.88\text{E}+02 \pm 1.08\text{E}+02$  Bq kg<sup>-1</sup> considering all types of studied soils.

Maize (*Zea mays*) and cabbage (*Brassica oleracea*) were selected for this experiment as representative of cereal and leafy crop groups. The experiment started with the sowing of maize. There were two sowings of maize: the first, 6 months after contamination and the second, 1.5 year after contamination. Seeds were sown in lines at a depth of 3 cm, at 7 cm intervals. The distance between lines was 50 cm. Cabbage was sown 3 times: 1, 2 and 2.5 years after contamination. Seeds were sown at a depth of 2 cm in lines at 5 cm intervals (one seed per hole). The distance between lines was 50 cm. All plants were

cultivated and harvested according to regional agricultural practices.

#### *Soil to plant transfer factor*

The soil to plant transfer factor (Fv) was determined as the ratio of the activity in the edible parts of the plant (Bq kg<sup>-1</sup> dry weight) and the activity in the first 20 cm of the soil (Bq.kg<sup>-1</sup> dry weight). All the procedure, including soil contamination, number of crops, agricultural practices and sampling followed the International Union of Radioecologists protocols established for Fv values determinations [15].

#### *Analyses*

Previous to the <sup>90</sup>Sr contamination, samples of soils (1 kg) were collected per lysimeter and sent to the Brazilian Enterprise of Agricultural Research (EMBRAPA-CNPS) for routine pedological analyses. The soil samples were analyzed according to [16]. Soil pH was measured in a 1:2.5 soil: water suspension. Exchangeable Al<sup>3+</sup>, Ca<sup>2+</sup> and Mg<sup>2+</sup> were extracted with KCl 1 mol L<sup>-1</sup>. Ca and Mg were determined by ICP-AES (Perkin-Elmer OPTIMA 8300) and Al by acid-base titration. Exchangeable Na<sup>+</sup> and K<sup>+</sup> were extracted with Mehlich 1 (HCl 0.025 mol L<sup>-1</sup> and H<sub>2</sub>SO<sub>4</sub> 0.0125 mol L<sup>-1</sup>) and determined by photometry. Potential acidity (H<sup>+</sup>+Al<sup>3+</sup>) was extracted with calcium acetate 0.5 mol L<sup>-1</sup> and determined by acid-base titration. Organic carbon was determined by oxidation with dichromate, and total N, with Kjeldahl. Some of the above analyses allowed the calculation of additional parameters such as: cation exchange capacity (CEC), the sum of Ca<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Al<sup>3+</sup> and H<sup>+</sup>. Soil texture was determined by Bouyoucos densimeter after shaking soil vigorously in NaOH 1 mol L<sup>-1</sup>.

After the first maize crop, the soils were sampled as the 0-10 cm and 10-20 cm layers, the effective rooting zone of most cultivated plants, and transferred to the laboratory to be dried (105 °C for 24 h) and sieved (particles greater than 2 mm were discarded), before being placed in appropriate containers to be analysed. An aliquot of each soil sample was sent to the IRD's radiochemical laboratory for the <sup>90</sup>Sr determination.

The determination of Fe, Ca, K, Mg and Sr in soils and plants, was conducted after chemical extraction (Aqua regia + HF, at 60 °C/ 30 min.), and measured by ICP-AES (PE OPTIMA 3000); The <sup>90</sup>Sr radiochemical determination in soil and plant was conducted according to the procedure established by Petrow [17]. The <sup>90</sup>Sr activity was determined by measuring beta activity of Y-90 in a Berthold Low Background Proportional Counter.

#### *Quality Control*

Due to the lack of certified soil samples, to ensure the analytical quality of the sequential extraction protocol used in this work, we summed the extracts from the five phases and compared these to aqua regia extraction of a certified reference material (BCR-701); recovery was generally greater than 75%. For the plant material assays, SRM-1515 (apple leaves) was used as a certified reference recovery was generally above 90%. All soil and plant samples were extracted in duplicate.

Recovery obtained for <sup>90</sup>Sr in reference materials (IAEA Soil 4/2000 and IAEA Vegetation grass 4/2000) based on average of duplicates was above 96% and 85% respectively.

Suprapur grade acids (Merck) and ultrapure water (Millipore ultrapure water system) were used in all laboratory procedures. All containers were soaked in 10% HNO<sub>3</sub> and thoroughly rinsed with deionized water before use.

#### *Statistical treatment of data*

The Student's t-test and all other statistical treatments were performed using Microsoft Excel™.

### **3. Results and Discussion**

### Chemical and physical properties of the soils

According to the soil properties, presented in Table I, it is possible to note that all studied soils are different in properties that may affect the behavior of radionuclides in soils, such as: nutrient content, organic matter content and the cation exchange capacity [18].

The soils contained at lysimeters 1, 2, 3, 5 and 6 are mainly gibbsite rich soil with sandy loam texture, low cation exchange capacity (CEC) and low organic matter (OM) content. The concentrations of exchangeable Ca and also the pH (H<sub>2</sub>O) observed in this soil are higher than that observed in natural conditions since it has received lime. The soils of the lysimeters 4 and 8 are acidic, clayey and has low content of OM, Ca and Mg and low CEC. The clay mineralogy is dominated by hematite and goethite. The soil of the lysimeter 7 has characteristics acidic, clayey and medium organic matter content [19]. This soil presents high CEC and mineralogical analyses indicate the predominance of hematite, goethite and traces of vermiculite in the clayey fraction. The Lysimeter 9 is a moderately acid sandy soil with low nutrient content, low CEC, very low OM content. The clay mineralogy is dominated by kaolinite. For <sup>90</sup>Sr activity concentrations in soil, the mean values reported in Table I were calculated by lysimeter, as recommended by IUR [15], as the mean of values measured in the 0-10 cm and 10-20 cm layers for each soil type. Despite of the type of contamination, spraying the solution at the surface, the final activity remained at the same order of magnitude for all lysimeters. Figure 1 reports the activity concentrations it by layers (0-10 cm and 10-20 cm), measured six months after contamination. This figure demonstrates a variable rate of <sup>90</sup>Sr transport in profile for the same class of soil or the same texture. Indeed, variation on the profile of distribution of <sup>90</sup>Sr was observed for the loamy Ferralsol-Al: lysimeter 1 presented higher concentrations in the 10-20 cm layer, lysimeters 5 and 6, showed highest concentrations on the top and lysimeters 2 and 3, showed a homogenous distribution of <sup>90</sup>Sr in the profile. Lysimeters 4, 7 and 8, all clayey soils, but different classes of soil, presented higher concentration in the 10-20 cm layer, while, lysimeter 9, a sandy soil, the higher concentration was at the superficial layer (0-10 cm). Unfortunately, we do not have information concerning pedological analyses by layers, because all soils used to fill the lysimeter, come from the cultivated layer (0-20 cm) of A horizon. However, considering that our lysimeters had been established about 10 years before the study described here, it is possible that some stratification had occurred for some soil properties. Besides that, soil bulk density is expected to differ in these lysimeters compared to the field, so vertical migration rates may not be the same as for soils under field conditions.

Total stable Sr in the soils ranged from 1 to 10.1 mg.kg<sup>-1</sup> dry mass. These values were lower than values reported in the literature (e.g. 23 mg.kg<sup>-1</sup> for agricultural top soil in Finland and 27 mg.kg<sup>-1</sup> in England & Wales [20]; 45 ± 26 mg.kg<sup>-1</sup> for soils from terrestrial Mediterranean ecosystem [21]). However, in acid soils, strontium is likely to be leached down the profile, while in calcareous soils and in organic matter rich soil-layers, Sr can be concentrated in upper horizons [22]. This observation could explain the lower concentration of Sr in the acid soil from tropical area. Since some of the studied soils received lime and mineral fertilizers, our data for total Ca (table I) were comparable to data reported to soils from terrestrial Mediterranean ecosystem: 1.6E+03 ± 7.6E+02 mg.kg<sup>-1</sup> dw [21].

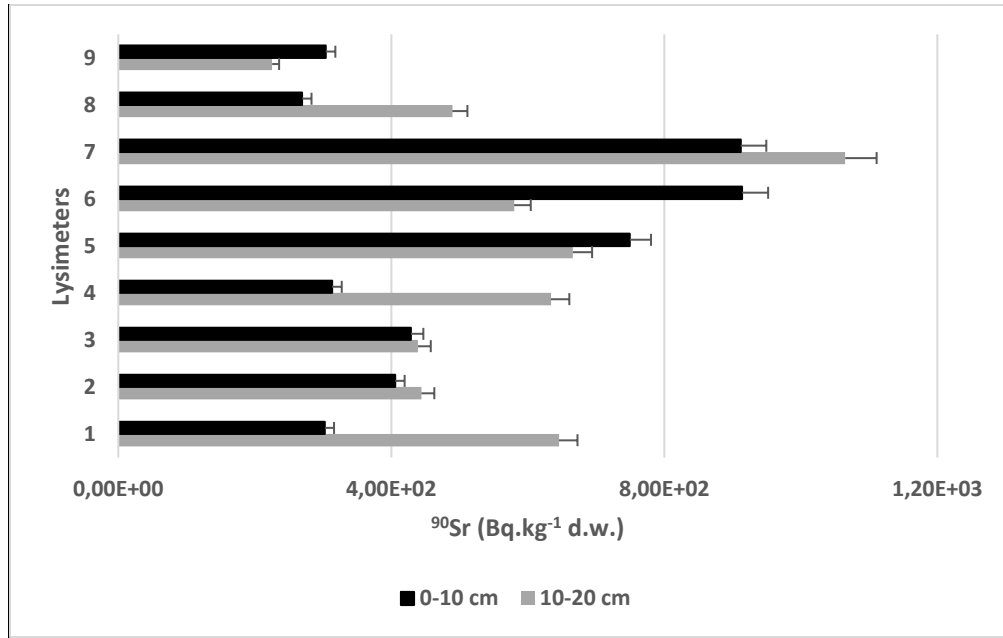
**Table I.** Main properties of Brazilian study soils.

	L1	L2	L3	L4	L5	L6	L7	L8	L9
<b>Exch.* Ca</b> (cmolc kg <sup>-1</sup> )	<b>4.10</b>	<b>3.26</b>	<b>3.53</b>	<b>0.19</b>	<b>4.28</b>	<b>4.39</b>	<b>4.04</b>	<b>0.11</b>	<b>0.70</b>
<b>Exch. Mg</b> (cmolc kg <sup>-1</sup> )	<b>0.79</b>	<b>0.72</b>	<b>0.86</b>	<b>0.08</b>	<b>0.86</b>	<b>0.60</b>	<b>0.77</b>	<b>0.04</b>	<b>0.20</b>
<b>Exch. K</b> (cmolc kg <sup>-1</sup> )	<b>0.14</b>	<b>0.10</b>	<b>0.10</b>	<b>0.09</b>	<b>0.15</b>	<b>0.13</b>	<b>0.18</b>	<b>0.04</b>	<b>0.08</b>

<b>Exch. Sr (mg kg<sup>-1</sup>)</b>	<b>3.61</b>	<b>2.92</b>	<b>3.19</b>	<b>0.24</b>	<b>4.24</b>	<b>4.09</b>	<b>6.66</b>	<b>0.14</b>	<b>1.04</b>
<b>CEC (cmolc.kg<sup>-1</sup>)</b>	<b>5.3</b>	<b>4.5</b>	<b>4.9</b>	<b>5.1</b>	<b>5.8</b>	<b>5.6</b>	<b>10.0</b>	<b>4.9</b>	<b>2.1</b>
<b>pH<sub>water</sub></b>	<b>7.6</b>	<b>7.7</b>	<b>7.5</b>	<b>4.7</b>	<b>7.6</b>	<b>7.9</b>	<b>6.6</b>	<b>4.8</b>	<b>5.8</b>
<b>pH (KCl)</b>	<b>7.2</b>	<b>7.1</b>	<b>7.0</b>	<b>4.1</b>	<b>7.0</b>	<b>7.3</b>	<b>5.6</b>	<b>4.1</b>	<b>4.5</b>
<b>OM (g kg<sup>-1</sup>)</b>	<b>2.26</b>	<b>2.07</b>	<b>2.02</b>	<b>1.60</b>	<b>2.11</b>	<b>2.18</b>	<b>2.14</b>	<b>1.60</b>	<b>0.50</b>
<b>Clay (g kg<sup>-1</sup>)</b>	<b>18.00</b>	<b>22.00</b>	<b>18.00</b>	<b>77.00</b>	<b>20.00</b>	<b>16.00</b>	<b>41.00</b>	<b>77.00</b>	<b>13.00</b>
<b>Sand (g kg<sup>-1</sup>)</b>	<b>71.00</b>	<b>77.00</b>	<b>74.00</b>	<b>19.00</b>	<b>70.00</b>	<b>72.00</b>	<b>10.00</b>	<b>19.00</b>	<b>82.00</b>
<b>SiO<sub>2</sub> (g kg<sup>-1</sup>)</b>	<b>3.30</b>	<b>3.80</b>	<b>3.50</b>	<b>15.60</b>	<b>3.50</b>	<b>3.10</b>	<b>15.70</b>	<b>15.60</b>	<b>3.60</b>
<b>Al<sub>2</sub>O<sub>3</sub> (g kg<sup>-1</sup>)</b>	<b>10.50</b>	<b>11.00</b>	<b>9.20</b>	<b>29.90</b>	<b>10.90</b>	<b>9.70</b>	<b>10.20</b>	<b>29.90</b>	<b>3.90</b>
<b>Fe<sub>2</sub>O<sub>3</sub> (g kg<sup>-1</sup>)</b>	<b>3.40</b>	<b>3.60</b>	<b>3.30</b>	<b>23.40</b>	<b>3.60</b>	<b>3.40</b>	<b>13.30</b>	<b>23.40</b>	<b>1.90</b>
<b>Sr total (mg kg<sup>-1</sup>)</b>	<b>9</b>	<b>6</b>	<b>7</b>	<b>3</b>	<b>10</b>	<b>10</b>	<b>10</b>	<b>1</b>	<b>3</b>
<b><sup>90</sup>Sr (Bq kg<sup>-1</sup>)</b>	<b>474.2 ± 9.7</b>	<b>425.0 ± 3.7</b>	<b>433.9 ± 0.3</b>	<b>473.5 ± 8.9</b>	<b>707.7 ± 2.0</b>	<b>1104.5 ± 29.8</b>	<b>988.4 ± 6.0</b>	<b>379.4 ± 6.0</b>	<b>264.7 ± 2.7</b>
<b>Textural Class (USDA)</b>	<b>Sandy Loam</b>	<b>Sandy Loam</b>	<b>Sandy Loam</b>	<b>Clay</b>	<b>Sandy Loam</b>	<b>Sandy Loam</b>	<b>Silty Clay</b>	<b>Clay</b>	<b>Loamy Sand</b>
<b>Mineral type at clay fraction**</b>	<b>G, K</b>	<b>G, K</b>	<b>G, K</b>	<b>H, G,</b>	<b>G, K</b>	<b>G, K</b>	<b>H, G, V</b>	<b>H, G,</b>	<b>K</b>
<b>Soil Class (WRB)</b>	<b>Ferrasol -Al</b>	<b>Ferrasol -Al</b>	<b>Ferrasol -Al</b>	<b>Ferrasol -Fe</b>	<b>Ferrasol -Al</b>	<b>Ferrasol -Al</b>	<b>Nitisol</b>	<b>Ferrasol -Fe</b>	<b>Acrisol</b>

\*Exchangeable.

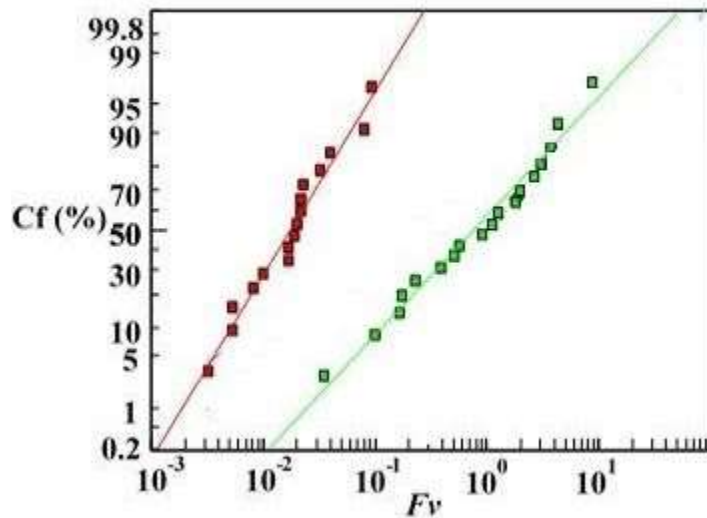
\*\* Clay mineral in order of predominance: G,K: Gibbsite, Kaolinite; H,G: Hematite, Goethite; H,G,V: Hematite, Goethite, Vermiculite; K: Kaolinite.



**Figure. 1.** Distribution of total amount of <sup>90</sup>Sr (Bq.kg<sup>-1</sup> d.w.) in the soil profile (0-20cm) of the studied classes of soil six months after superficial contamination.

#### Preliminary Analysis of Transfer Factor Values

The summary of Fv values obtained in this study and the values of R2 relative to the log-normal adjustments for the set of all values are presented in Table II. These data confirm that Fv values fits better to a lognormal distribution than to a normal distribution. The analysis of the results of the adjustment of the distribution curves, presented in Fig. 2, confirms, through the Student t test at the 95% confidence level, that the different studied vegetables constitute distinct sets, in relation to the <sup>90</sup>Sr Fv, justifying the individual approach of plant species for further discussion. This result is consistent with the physiological differences between these crops and the parts of the plants studied: maize grain is a 'protected structure', where the entry of the elements is through some other structure and subsequent transport to this storage organ (translocation process). According to Andersen [8], during translocations of elements, plants are able to discriminate against Sr, relative to Ca; as consequence, seeds and fruits will likely have lower concentrations.



**Figure 2.** Cumulative frequency of  $F_v$  by crop (maize: red square; cabbage: green square).

**Table II.** Statistical summary for  $F_v$  by crop.

<i>Crop</i>	<i>n</i>	<i>arithmetic mean</i>	<i>Sd</i>	<i>geometric mean</i>	<i>Sd</i>	<i>r2</i>
<b>Cabbage</b>	18	1.8E+0	2.2E+0	7.9E-1	4.4	0.98
<b>Maize</b>	16	2.6E-2	2.6E-2	1.8E-2	2.5	0.97

### Brazilian $^{90}\text{Sr}$ $F_v$ values compared to $^{90}\text{Sr}$ $F_v$ values reported for temperate soils

Table III reports Brazilian  $^{90}\text{Sr}$   $F_v$  values compared to  $^{90}\text{Sr}$   $F_v$  values reported in Technical Reports Series (TRS 472) for soils from temperate areas (IAEA 2010) for each crop; The texture classification was according to classes described at IAEA TEC-DOC 1616 [23]. In this table, the clay type was discriminate, since the mineralogy of clay from humid tropics can differ from the temperate zones [24]. Three Brazilian soils classes showed the dominance of oxy-hydroxides of iron (Fe) and aluminium (Al) in the clay fraction, typical for humid tropical areas. However, one Brazilian soil, the subtropical ones, exhibited traces of vermiculite in the clay fraction, a mineral associated to temperate climate [24]. For maize, all Brazilian soils presented lower  $^{90}\text{Sr}$   $F_v$  values than values observed in temperate soils (TRS-472). Lower  $^{90}\text{Sr}$   $F_v$  values in tropical soil have also previously been reported for maize (Robison, Conrado et al. 2000). However, for cabbage,  $^{90}\text{Sr}$   $F_v$  values showed an absence of a pattern across the three soil textural classes for Brazilian soil compared with TRS-472 values reported for leafy crops.  $^{90}\text{Sr}$   $F_v$  mean value reported for clay texture, for the subtropical Brazilian soil, is more similar to mean value reported for temperate soils, TRS-472. However, the  $^{90}\text{Sr}$   $F_v$  value for the soil with clay mineralogy typical of humid tropics was more than one order of magnitude higher than TRS-472 value reported for temperate soils (clay texture). The  $^{90}\text{Sr}$   $F_v$  mean values reported for the silty Brazilian soil was lower than TRS-472 values (silty texture), however, as these soils had their original properties modified by agriculture practices, changing considerably pH and Ca exchangeable content, properties with effect on  $^{90}\text{Sr}$   $F_v$ , as reported by several authors [e.g.: 9, 12, 25], the discussion related to climate and this textural class (silty) should be avoid for both crops. Anyway, these results show that soil texture is not a good indicator for  $^{90}\text{Sr}$   $F_v$  and seems not able to explain the behaviour of Sr in soils highly weathered. The test T (95%) applied to all data considered  $F_v$  obtained in the 4 types of soils differs among each other.

Consequently, in subsequent discussions, we consider  $^{90}\text{Sr}$  Fv based on soil class and not textural class.

**Table III.**  $^{90}\text{Sr}$  Soil to Plant Transfer Factors (*Fv*) obtained from Brazilian soils reported as a function of soil texture and clay mineral type, compared with TRS-472 [5].

Crop	Soil Texture	Brazilian soils			TRS-472 data for temperate environment		
		$^{90}\text{Sr}$ <i>Fv</i>	Sd	n	$^{90}\text{Sr}$ <i>Fv</i>	Sd	n
<b>Maize</b>	Sand	8.7E-02	6.2E-03	2	5.2E-01	4.0E-02	19
	Silt	1.6E-02	5.5E-03	10	3.6E-01	1.5E-01	13
	Clay (humid tropics)	3.6E-02	3.7E-03	3	-	-	-
	Clay (Subtropical and temperate)	5.8E-03*	2.5E-03	2	6.9E-02	2.0E-03	7
<b>Cabbage</b>	Sand	3.1E+00	1.2E+00	3	1.7E+00	6.4E-02	72
	Silt	6.9E-01	5.4E-01	10	1.2E+00	4.1E-02	84
	Clay (humid tropics)	4.6E+00	2.8E+00	3	-	-	-
	Clay (Subtropical and temperate)	9.2E-01*	8.8E-01	2	1.5E-01	3.9E-03	54

- \* Subtropical: clay fraction dominated by hematite followed by goethite with traces of vermiculite.

#### Transfer of $^{90}\text{Sr}$ to maize

Pearson's correlation coefficients for  $^{90}\text{Sr}$  Fv for maize, in relation to soil properties, were negative and statistically significant for exchangeable Ca, Mg, K and stable Sr, pH and organic matter content (OM) (table IV). Low soil nutrient status and low pH have previously been shown to result in comparatively high radionuclide transfers to plants [18]. Soils with higher exchangeable Ca contents had lower  $^{90}\text{Sr}$  Fv values. The effect of exchangeable Ca on  $^{90}\text{Sr}$  uptake is relatively well know [e.g.: 8, 12, 25] and occurs due to their chemical similarity favoring ionic competition (26, 27). Indeed, for  $^{90}\text{Sr}$  Fv, among exchangeable nutrients, the higher negative correlation occurred with exchangeable Ca ( $r = -0.77$ ;  $\alpha < 0.001$ ;  $n=16$ ) illustrating the relevance of nutritional status of soil on  $^{90}\text{Sr}$  root uptake. Ionic competition among isotopes is also expected for the same reason, being the stable favored by negligible mass of the radioactive Sr. Stable Sr Fv for maize was not correlated with any of soil properties (Table IV). As stable Sr is an endogenous element it will behaves in soil in a different way of that exogenous  $^{90}\text{Sr}$ , being the former, mainly associate to more labile compounds in the soil than the first.

**Table IV.** Pearson's correlation coefficient for  $^{90}\text{Sr}$  *Fv* and stable Sr *Fv* for maize in relation to soil properties.

	$^{90}\text{Sr}$ <i>Fv</i> <sub>maize</sub>	Stable Sr <i>Fv</i> <sub>maize</sub>
<b>Exchangeable Ca</b>	<b>-0.77***</b>	<b>-0.22</b>
<b>Exchangeable Mg</b>	<b>-0.72**</b>	<b>-0.23</b>
<b>Exchangeable K</b>	<b>-0.66**</b>	<b>-0.17</b>
<b>Exchangeable Sr</b>	<b>-0.69**</b>	<b>-0.17</b>
<b>pH<sub>water</sub></b>	<b>-0.58*</b>	<b>-0.13</b>



<b>pH (KCl)</b>	<b>-0.65**</b>	<b>-0.20</b>
<b>CEC</b>	<b>-0.72**</b>	<b>-0.33</b>
<b>OM</b>	<b>-0.97***</b>	<b>-0.50</b>
<b>Clay</b>	<b>-0.05</b>	<b>-0.28</b>
<b>Sand</b>	<b>0.25</b>	<b>0.25</b>
<b>SiO<sub>2</sub></b>	<b>-0.11</b>	<b>-0.20</b>
<b>Al<sub>2</sub>O<sub>3</sub></b>	<b>-0.13</b>	<b>-0.36</b>
<b>Fe<sub>2</sub>O<sub>3</sub></b>	<b>-0.06</b>	<b>-0.26</b>

\*\*\*statistically significant ( $\alpha < 0.001$ ); \*\*statistically significant ( $\alpha < 0.01$ ); \*statistically significant ( $\alpha < 0.05$ ); n=16.

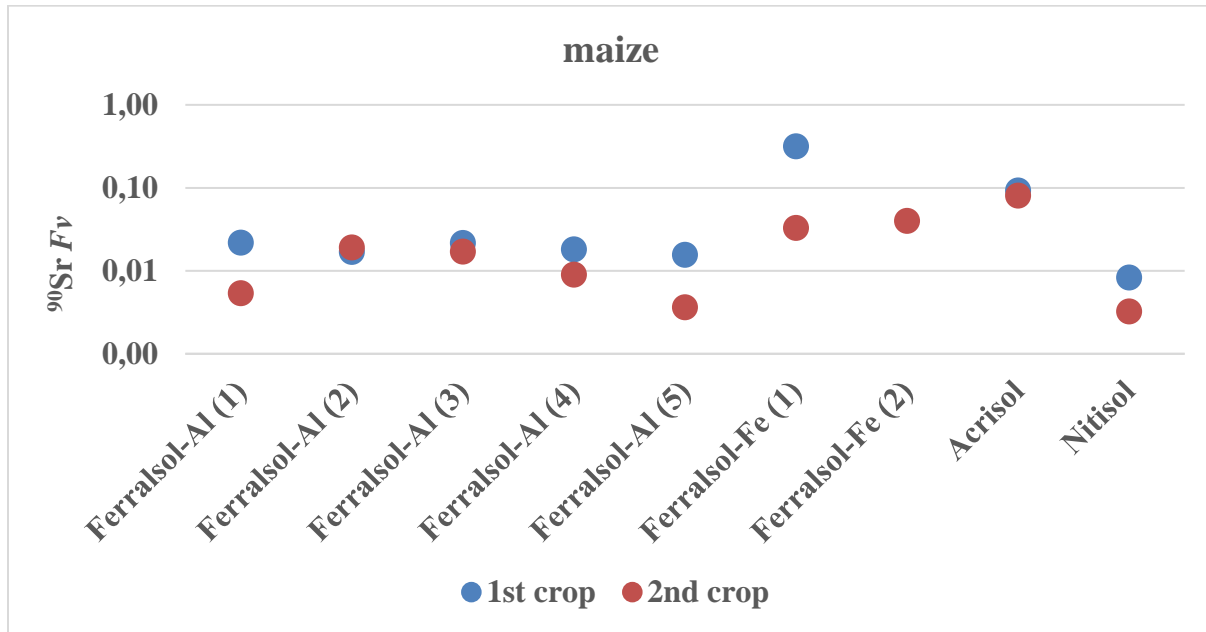
To better understand the <sup>90</sup>Sr uptake by maize, the concentration of <sup>90</sup>Sr in grain was also evaluated in relation to concentration of macronutrients (Ca, Mg and K) and stable Sr. Table V shows that the concentration of stable Sr in maize correlated positively across all soils types with the concentration of the divalent nutrients (Ca and Mg). However, the activity of <sup>90</sup>Sr in maize did not show any correlation with the analysed nutrients. The redistributing of nutrients during seed set, senescence, or periods of deficiency occurs mainly via phloem [28], so the nutrient status of the grain depends on the translocation processes, mainly occurring via phloem. Although many ions are transported in the phloem, divalent cations, (e.g. Ca<sup>2+</sup>, Ba<sup>2+</sup> and Sr<sup>2+</sup>) are relatively immobile [28], what explains the positive correlation found in our study of the stable Sr only with divalent nutrients with. As discussed before, the effect of exchangeable Ca in soil on <sup>90</sup>Sr uptake is well reported [e.g.: 8, 9, 12, 25 ] and it was also verified in this study. Additionally, according to White [28], the phloem composition often reflects the nutrient status of the shoot, it has been speculated that this might provide a signal to align ion uptake and translocation with shoot demand. So, the absence of correlation of divalent nutrients with the <sup>90</sup>Sr, is kind of expected, once the activity concentration of <sup>90</sup>Sr in grain will be affected for several manners, since root uptake, due to competition processes with major nutrients and stable Sr, associated to its negligible mass compared with these elements, till the transport to the grain, due to its low mobility in the phloem, influenced by nutritional needs on translocation processes, that for the radioactive Sr will be negligible also.

**Table V.** Pearson`s correlation values for <sup>90</sup>Sr (Bq. kg<sup>-1</sup>) and stable Sr (mg.kg<sup>-1</sup>) in maize relative to maize nutrient concentrations (mg.kg<sup>-1</sup>).

	<sup>90</sup> Sr (Bq.kg <sup>-1</sup> )	Sr (mg.kg <sup>-1</sup> )
<sup>90</sup> Sr (Bq.kg <sup>-1</sup> )	<b>1.00***</b>	0.38
Sr (mg.kg <sup>-1</sup> )	0.38	<b>1.00***</b>
Ca (mg.kg <sup>-1</sup> )	0.33	<b>0.99***</b>
Mg (mg.kg <sup>-1</sup> )	0.34	<b>0.95***</b>
K (mg.kg <sup>-1</sup> )	0.39	0.69

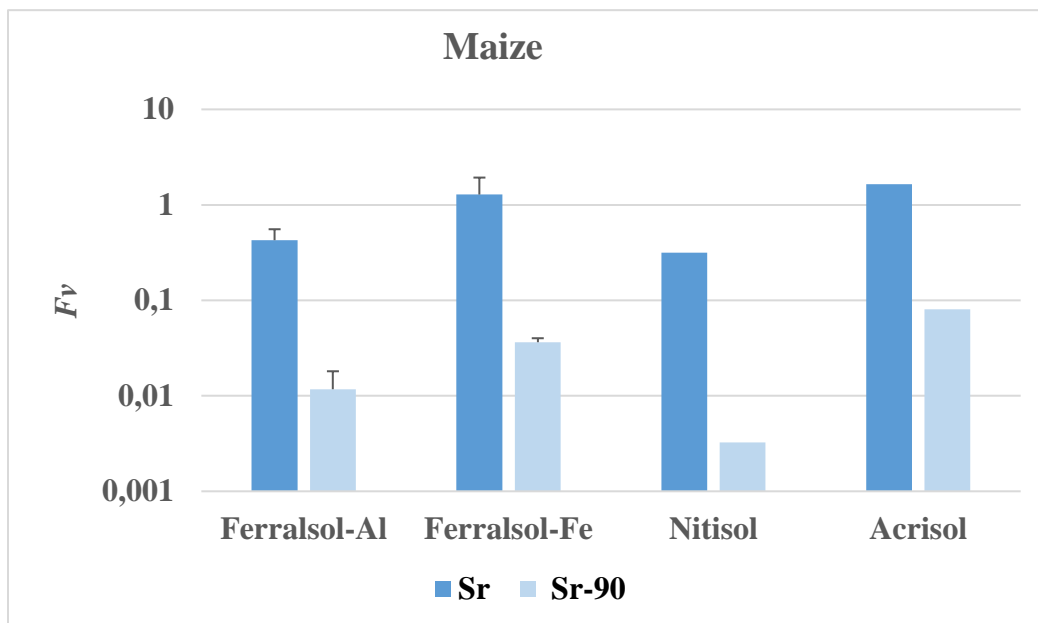
\*\*\*statistically significant ( $\alpha < 0.001$ ); (n=16; n=7 for K).

Fig. 3 present the <sup>90</sup>Sr Fv values for first and second crop obtained for maize in all studied soils. In this figure we can observe that the variation occurred among crops was in average 3 times bigger for the first crop.



**Figure.3.**  $^{90}\text{Sr } F_v$  values for maize in the studied soils.

Fig. 4 shows  $^{90}\text{Sr}$  and stable Sr  $F_v$  values for maize in the studied soils. In this figure, we can see that lowest  $^{90}\text{Sr } F_v$  values occurred in Nitisol (the subtropical soil) and Ferralsol –Al (the limed soil) which are the soil with the higher exchangeable Ca content. The highest  $^{90}\text{Sr } F_v$  values were observed in the most acidic and unfertile soils (Acrisol and Ferralsol-Fe). Stable Sr  $F_v$  were all more than an order of magnitude higher than those for  $^{90}\text{Sr}$  for all soil types; the largest difference being observed for Nitisol (Fig. 4). This result shows that, despite of similarity of processes acting for both (e.g. competition with Ca for root uptake), for radioprotection purpose, the use of Stable Sr  $F_v$  should be avoid for replace  $^{90}\text{Sr } F_v$ , especially for cereals or plants that the organ of concernment are storage organs, due to processes occurring in the phloem, decreasing translocation of the radioactive isotope, with negligible mass, compared with the stable one.



**Figure. 4.**  $^{90}\text{Sr } F_v$  mean values compared with stable Sr  $F_v$  mean values for maize in the studied classes of soils.

The results for  $^{90}\text{Sr}$  in this study are similar to our previous observations for  $^{137}\text{Cs}$  in these same soils: soils with low nutrient availability and low pH had higher  $^{137}\text{Cs}$   $F_V$  for maize [29].

### Transfer of $^{90}\text{Sr}$ to cabbage

Contrary to observations for maize, the concentration of both stable Sr and  $^{90}\text{Sr}$  correlates positively with the concentration of all analysed nutrients; there is also correlation between the two strontium isotopes. However, correlations observed for stable Sr were generally better ( $\alpha < 0.001$ ) than those for  $^{90}\text{Sr}$  ( $\alpha < 0.01 - 0.05$ ) (Table VI). This results seems to corroborate the relevance of phloem via explaining results for maize, since, for leaves, the main route is the xylem, which easily transport ions from the roots to leaves.

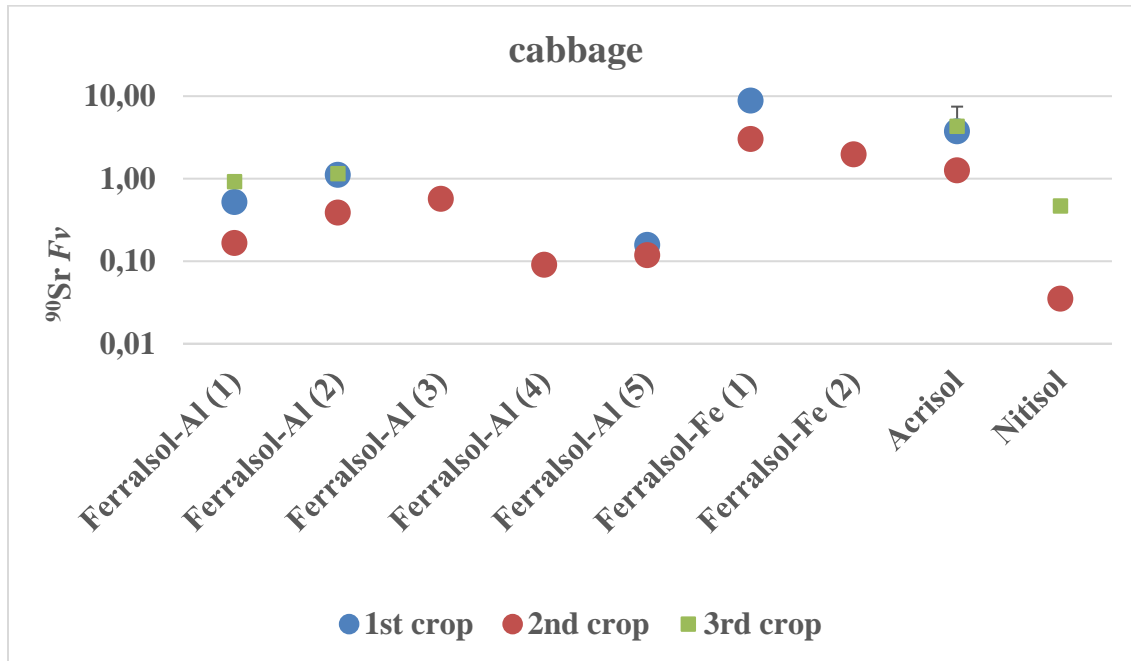
**Table VI.** Pearson's correlation values for  $^{90}\text{Sr}$  ( $\text{Bq.kg}^{-1}$ ) and stable Sr ( $\text{mg.kg}^{-1}$ ) in cabbage relative to the nutrients concentration ( $\text{mg.kg}^{-1}$ ).

	$^{90}\text{Sr}$ ( $\text{Bq.kg}^{-1}$ )	Sr ( $\text{mg.kg}^{-1}$ )
$^{90}\text{Sr}$ ( $\text{Bq.kg}^{-1}$ )	<b>1.00***</b>	<b>0.62**</b>
Sr ( $\text{mg.kg}^{-1}$ )	<b>0.62**</b>	<b>1.00***</b>
Ca ( $\text{mg.kg}^{-1}$ )	<b>0.61**</b>	<b>0.89***</b>
Mg ( $\text{mg.kg}^{-1}$ )	<b>0.56*</b>	<b>0.92***</b>
K ( $\text{mg.kg}^{-1}$ )	<b>0.62**</b>	<b>0.92***</b>

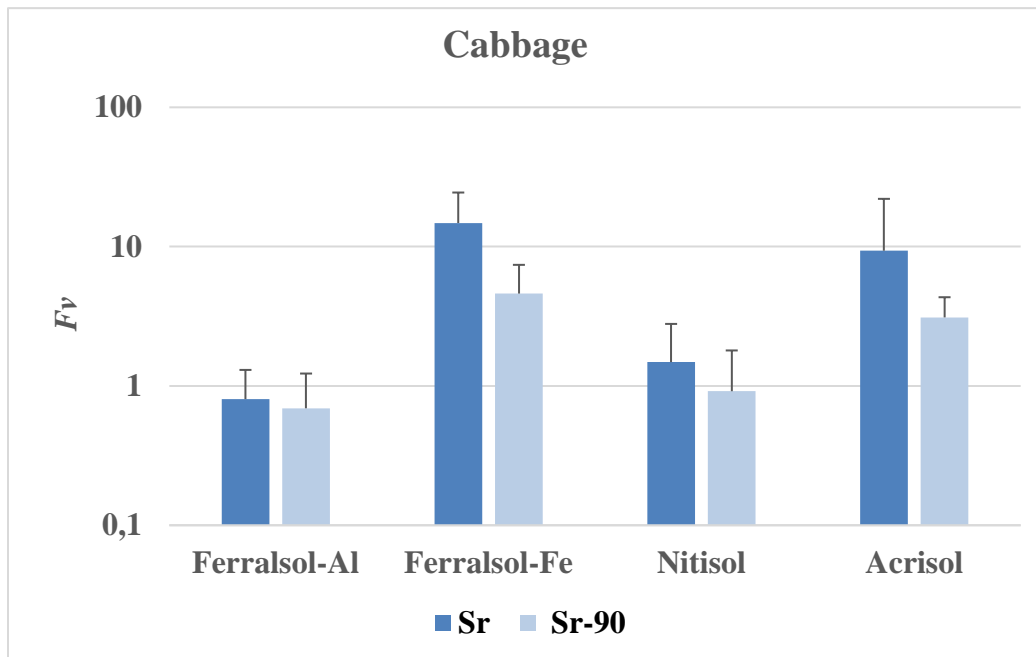
\*\*\*statistically significant ( $\alpha < 0.001$ ); \*\*statistically significant ( $\alpha < 0.01$ ); \*statistically significant ( $\alpha < 0.05$ ); n=17.

Fig. 5 present the  $^{90}\text{Sr}$   $F_V$  values for first and second crop obtained for cabbage in all studied soils. In this figure, as seen for maize, we can observe that second crops was in average 3 times bigger for the first crop. However, the data available for the third crop, they were similar to the first crop. Generally, IUR recommend 3 crops for stablish transfer factor values for a type of soil. These results shows that seasonal variations can be observed in 2 crops for  $^{90}\text{Sr}$ .

Fig. 6 shows  $^{90}\text{Sr}$  and stable Sr  $F_V$  values for cabbage in the studied soils. As seen for maize, that lowest  $^{90}\text{Sr}$   $F_{V\text{cabbage}}$  values occurred in Nitisol and Ferralsol -Al (the limed soil) which are the soil with the higher exchangeable Ca content. Highest  $^{90}\text{Sr}$   $F_V$  values ( $>1$ ) were observed in the most acidic soils (Ferralsol-Fe and Acrisol). The same pattern can be observed for stable Sr  $F_{V\text{cabbage}}$ . The most significant correlations observed for  $^{90}\text{Sr}$   $F_{V\text{cabbage}}$ , were negative and occurred with exchangeable Ca and pH (Table VII), indicating that these soil properties better explain the behaviour of Sr in soil-plant system. The relevance of these parameters has been reported at the literature for several crops [30]. Other significant and negative correlations occurred with exchangeable Mg and Sr, and, at lower level of probability, with OM, clay and  $\text{Fe}_2\text{O}_2$ . The most significant correlations observed for stable Sr  $F_{V\text{cabbage}}$ , were negative and occurred with exchangeable Mg, Ca and pH (Table VII), and, at lower level of probability, with exchangeable Sr and K, and OM, following a similar influence of soil properties observed on  $^{90}\text{Sr}$   $F_{V\text{cabbage}}$ .



**Figure 5.**  $^{90}\text{Sr } Fv$  values for cabbage in the studied soils.



**Figure 6.**  $^{90}\text{Sr } Fv$  mean values compared with stable Sr  $Fv$  mean values for cabbage in the studied classes of soils.

**Table VII.** Pearson's correlation coefficient for  $^{90}\text{Sr } Fv$  and stable Sr  $Fv$  for cabbage in relation to soil properties.

	$^{90}\text{Sr } Fv_{\text{cabbage}} (n=18)$	Sr $Fv_{\text{cabbage}} (n=17)$
Exchangeable Ca	-0.72***	-0.68**
Exchangeable Mg	-0.68**	-0.69**

<b>Exchangeable K</b>	<b>-0.43</b>	<b>-0.55*</b>
<b>Exchangeable Sr</b>	<b>-0.62**</b>	<b>-0.57*</b>
<b>pH<sub>water</sub></b>	<b>-0.71***</b>	<b>-0.67**</b>
<b>pH (KCl)</b>	<b>-0.68**</b>	<b>-0.65**</b>
<b>CEC</b>	<b>-0.29</b>	<b>-0.31</b>
<b>OM</b>	<b>-0.49*</b>	<b>-0.50*</b>
<b>Clay</b>	<b>0.51*</b>	<b>0.44</b>
<b>Sand</b>	<b>-0.29</b>	<b>-0.22</b>
<b>SiO<sub>2</sub></b>	<b>0.40</b>	<b>0.32</b>
<b>Al<sub>2</sub>O<sub>3</sub></b>	<b>0.46</b>	<b>0.37</b>
<b>Fe<sub>2</sub>O<sub>3</sub></b>	<b>0.49*</b>	<b>0.42</b>

\*\*\*statistically significant ( $\alpha < 0.001$ ); \*\*statistically significant ( $\alpha < 0.01$ );

\*Statistically significant ( $\alpha < 0.05$ ).

#### Strontium uptake by plants

For both crop types, the well-weathered soils (Ferralsol-Fe and Acrisol) had higher  $F_v$  values than Nitisol. The lowest  $F_v$  values were observed for Nitisol, which has a comparatively good natural supply of nutrients and contains vermiculite within the clay fraction contributing to more negative permanent charges than typically found in tropical soils that better retains cations; Ferralsol-Fe and Acrisol, which are a typical unfertile tropical soil, have a low retention of cations due to their lower pH and mineralogy dominated by low active clays. Ferralsol- Al, a well-weathered soil, originally with nutritional status similar to Ferralsol-Fe, presented, after liming, lower  $F_v$  values compared to Ferralsol-Fe, but similar  $F_v$  values, compared with Nitisol. In general, it appears that ionic competition for root uptake of macronutrients (most especially Ca and Mg) strongly influences  $^{90}\text{Sr}$   $F_v$ . Because the highly weathered soils, typical for much of Brazil, are depleted in these nutrients, the addition of fertilisers and/or lime will potentially have a bigger impact on  $^{90}\text{Sr}$   $F_v$  as observed for Ferralsol- Al.

For these well-weathered soils, cabbage exhibited higher  $F_v$  values than generic crop values (leafy vegetables) recommended in IAEA [5]. Contrary, maize presented lower  $^{90}\text{Sr}$   $F_v$  values than recommended for this crop in IAEA [5]. Previous studies have demonstrated the difficulty in trying to predict maize nutrient values based upon soil properties (Ferreira, Motta et al. 2012), fact that corroborates the influence of lower mobility of Sr in the phloem, hiding processes occurring on the root uptake due to soil properties. Indeed, our results shows that leaves reflected the influence of soil properties on root uptake, probably due to the transport to leaves being mainly done via xylem.

The results presented in this paper demonstrate the importance of considering both nutrient status and clay mineralogy when trying to understand the behaviour of Sr; this has previously been observed also for Cs [29, 31].

#### 4. Conclusions

Our data showed the relevance of clay mineralogy and nutritional status in explaining the root uptake of stable Sr and  $^{90}\text{Sr}$  in highly weathered soils (where most of the negative charges are pH dependent). For highly weathered soils  $^{90}\text{Sr}$   $F_v$  values differed among species: maize presented  $^{90}\text{Sr}$   $F_v$  lower than values advised by IAEA for temperate soils, whilst cabbage presented generally higher values than recommended

for temperate soils in IAEA [5] For the studied species,  $^{90}\text{Sr}$   $F_v$  for cabbage exhibited the best correlation with soil properties. The lowest  $^{90}\text{Sr}$   $F_v$  values were observed in the subtropical soil (Nitisol) with a mineralogy and nutritional status closer to soils from temperate areas. The highest  $^{90}\text{Sr}$   $F_v$  values were observed in the unfertile highly weathered soils (Acrisol and Ferralsol-Fe). These soils are typical of most Brazilian agricultural soils, suggesting a 'vulnerability' of local soils to  $^{90}\text{Sr}$  contamination, however, being these soils mostly composed by mineral with negative charges depending on pH (iron/ aluminum hydroxides), so, changes in pH promoted by liming reduce  $F_v$  values to comparable levels of temperate or subtropical soils. Our results show that soil texture is not a good indicator of  $^{90}\text{Sr}$   $F_v$ , since soils with the same texture, but different clay mineralogy and properties, presented  $^{90}\text{Sr}$   $F_v$  values which differed by more than an order of magnitude.

Our results suggest that stable Sr may not be a very good analogue of  $^{90}\text{Sr}$  for determining plant  $F_v$  values for maize, since differences are greater than one order of magnitude. The transportation via phloem seems discriminate stable Sr against  $^{90}\text{Sr}$ , explaining higher stable Sr  $F_v$  values for maize. Such behavior was not observed for cabbage, which difference were closer to 3 folds only for unfertile soil, and close to 1 for soils better supplied with Ca. Our results also show a negative correlation between Sr bioavailability and  $^{90}\text{Sr}$   $F_v$  likely demonstrating the importance of competing ions, most especially stable Sr and Ca, in determining root uptake of  $^{90}\text{Sr}$ .

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