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ENVIRONMENTAL ASSESSMENT OF NUCLEAR INSTALLATIONS USING ACCUMULATED LITTERFALL CYCLING.

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ABSTRACT

For 25 years the Nuclear and Energy Research Institute – IPEN/SP processed uranium oxide to produce the fuel element. Even with major care in the handling of uranium hexafluoride and uranium compounds, there is the probability of small fractions are dispersed into the atmosphere. Due to this fact, it was proposed a study of these compounds in the environment, aiming at the biomonitoring of toxic substances originating from the fabrications process of fuel element, as well toxic metals. The litterfall it´s consisted of fragments of organic vegetable, including leaves, flowers, fruits, branches, twigs and animal waste. The objective of this study was to determine the production and seasonality of litterfall in the gardens of IPEN, establish a correlation between the compartment leaves, wood and reproductive parts and evaluate the chemical composition of leaves originated of litterfall through chemical analysis. Was installed 10 litterfall collectors to determinate the production . The determination of chemical elements was realized by X-ray fluorescence for dispersion of wavelength (WDXRF). The production of dry litterfall during the period was 5.86 Kg m² $_{-1}$. The elements analyzed were Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, Mn, Fe, Ni, Cu, Br, Rb, Sr, Zr, Th and U. The major constituents of the composition of leaf Ca, Si, and K (1.8%, 0.5% and 0.6% respectively). The results allowed us to conclude that the installations used in the nuclear fuel cycle earlier, as well as the installations in operation, actually didn´t affect the biogeochemical cycle of plants.

1. INTRODUCTION

At IPEN, the early development of the fabrication technology of fuel elements is very old. The work was started yet in the 60s when it was designed and installed a pilot plant for purifying uranium. Next in 1974, began the operations of Pilot Plant Production of uranium tetrafluoride. In the middle of 80’s began production of thorium nitrate in pilot scale. The main objective was to manufacture fuel for research reactor ARGONAUTA, the Institute of Nuclear Engineering CNEN. In the 80's, IPEN dominated almost every step of the Nuclear Fuel Cycle1.

After 25 years processing uranium oxides for the fabrication of fuel element, strategic changes and the rationalization in the processing of uranium line, led to the decommissioning of the plant, and the reinstatement of the building for others purposes [2].

In the uranium processing installations, even major care in the handling of uranium hexafluoride and uranium compounds, there is the probability of small fractions are dispersed into the atmosphere, along with ^{234Th} and ^{234}Pa. The uranium hexafluoride reacts immediately with moisture in the air, joining with the dust particles existing and subsequently falling on the surrounding ecosystems to the installation[3]. Even in the intermediate stages of manufacture of the fuel element always exists the possiblity of possible fumes from metals to the environment.

Thus it was necessary a more detailed study of the cycle of these compounds in the environment both for the biomonitoring of toxic substances from the manufacturing process of fuel element then toxic metal.

1.2 Biomonitoring through the litterfall

The bioindicators can be defined as organisms or group of organisms that respond to environmental pertubations through changes in their vital functions or chemical composition, can be used to assess the extent of changes in their environment[4].

In summary, the techniques of biomonitoring complement the instrumental procedures, and could be represent the only alternative for assessment of pollution in remote areas or deprived of technological resources.

The litterfall it´s consisted of fragments of organic vegetable, including leaves, flowers, fruits, branches, twigs and animal waste that fall on the ground by means of any processes[5]. The concentration and proportion of elements can be change significantly the process f senescence, decomposition and mineralization of litterfall[6].

The deposition and decomposition of litterfall are important mechanisms of the flow of nutrients and organic material in areas of vegetation[7]. Additionally, decomposition and mineralization of litterfall are given mainly by a limited number of elements, be of great importance some essential nutrientes and some potentially toxic metals[6].

Several studies of mineral cycling were conducted in forest areas to obtain information about the contribution of litterfall and nutrients. This studies are limited for the distribution of chemical elements such as Ca, K, Mg, Si, P, Fe, Mn, B, Cl, Mo, Zn, Na, U, Th and Co[8,9,10]. However, few studies have been made with respect to toxic elements and features in no native species for environmental monitoring.

The knowledge and evaluation of fractions of trace elements and their correlations can contribute to the understanding of the impact of man in the compartment ecological studied.

1.4 Wavelength dispersive X-ray fluorescence (WDXRF)

The X-ray fluorescence is classified as an atomic emission technique, based on the photoelectric effect. When an atom is subjected to an irradiation process using a source of X-ray (X-ray tubes, particle-induced, natural radioisotopes, synchrotron light and others), one electron can be ejected from the electron shell innermost of atom. For stabilization of this excited state, electrons from the outer layers quickly occupy the vacancies generated by releasing the energy difference between the two energy levels, the radiation emitted for each transition is characteristic for each element present in the sample. Thus, the energy or wavelength of radiation can be directly used in the qualitative determination of an element, as well as the intensity of radiation emitted can be used in the quantification of such species[11, 12, 13].

The WDXRF system has advantages which we can cite the adaptability for automation, non destructive tests and also the detection limit was considered statistically suitable for biological samples.

2. OBJECTIVE

The objective of this study was to know the distribution and behavior of chemical elements, stable, in the biotic compartment of the ecosystem formed by gardening around the nuclear installations and radioactive from Nuclear and Energy Research Institute – IPEN.

3. MATERIALS AND METHODS

3. 1. Study Area

This area chosen for this study is located in Nuclear and Energy Research Institute (IPEN), in regions to the gardening close to activities of the nuclear fuel cycle, developed in the decades of 1970 – 1990, or under development.

In FIG 1 is represented the map of IPEN and in detail the location of the study area: the garden of CQMA and around.

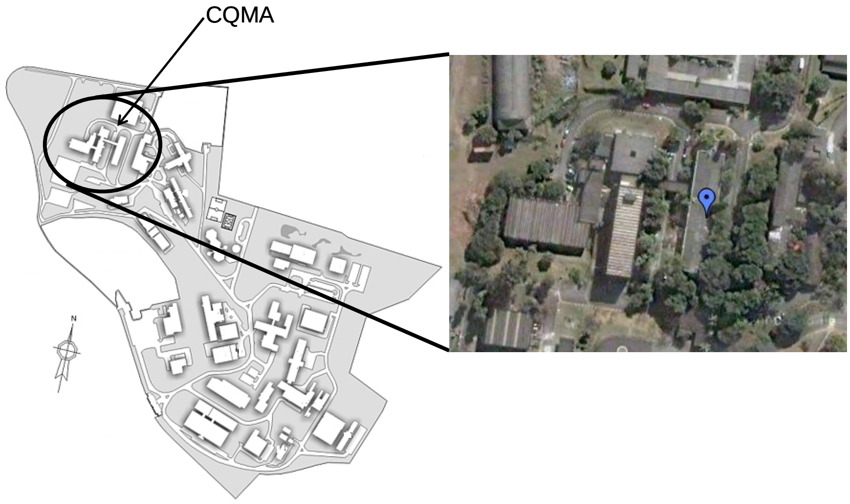

FIGURE 1 – Map of IPEN and in detail the study area, the garden of Environmental Science and Technology Center (CQMA)
***Source: Google Earth, 2009*.**

A collection points, called control point ("Branco"), located in the rural town of Andradas/MG was also monitored by way of comparison.

3.1 Species identification

For species identification, samples of branches of each tree with leaves, Wood, flower and fruit (when possible) and made presses of cardboard with string. The branches were collected and identified, have been dried on an oven at 35 ºC (Celcius), for at one week and later mounted in folders with ID chips.

These identifications carried in the Santa Cecília University Herbarium, with the help of a taxonomist and curator of the Herbarium Msc. Paulo Sales Penteado Sampaio, were identified genus or species.

3. 2. Litterfall Collection

To determine the production of litterfall were collected monthly deposition from collectors over a period o one year, from Febuary 2010. For purposes of comparison two collection of litterfall samples were taken as control point ("branco"), free from the urban activities and facilities of IPEN influence, these collections were among July at December 2010. Each sample collected was separate in fractions, leaves, twigs, flowers and fruits, and dried at 70°C for 72 h, and after made their weights (precision ± 1g). All information was plotted and carried the descriptive and statistical analysis, mean, median, standard deviation and correlation (r²), with assistance of ECXEL 2008. As a measure of interpretation of Pearson's coefficient, or correlation coefficient, the methodology developed by Shimakura (2006)[14], which establishes a classification system for this coefficient by assigning a qualitative value to the numerical values of coefficient found (Table 1).

Table 1 – Pearsons's coefficient rating

Value of (+ our -)	Interpretation
0.00 - 0.19	A very weak correlation
0.20 - 0.39	A weak correlation
0.40 - 0.69	A moderate weak
0.70 - 0.89	A strong correlation
0.90 - 1.00	A very strong correlation

(Source: Adapted from Shimakura, 2006)

3. 3 X-ray fluorescence spectrometry – Method of Fundamental Parameters (FP)

The determination of chemical elements constituents from the leaves of litterfall was carried by wavelength dispersive X-ray fluorescence (WDXRF),using a spectrometer RIGAKU Co., model RIX 3000, 1996.

The technique of X-ray fluorescence uses several methods for quantitative chemical analysis. Between the methods of algorithms, the fundamental parameters method (FP), allows to calculate the analytical composition of a sample without the use of similar standards, from the measured intensity of the emission line of the analito and the tabulated values of the major fundamental parameters as: primary spectral distribution (source), absorption coefficient (photoelectric and mass), fluorescence income and others[12,14].

In this work, the method of fundamental parameters was applied the determination of metals and macro and micro-constituents elements in samples of diagnoses leaf litterfall. The method was validated by statistical tools, using certified reference materials, Peach leaves - NIST 1547 and Mixed Polish Herbs - INCT-MPH-2 (MPH). The methodology was evaluated for the biological matrix (leaves), as certified, to the elements, Mg, Al, S, Cl, K, Ca, Cr, Mn, Ni, Zn, Br, Rb e Sr. For the other elements Si, Cr, Th, U, Zr, Ti, F, Na, V, Co, Ga, Ge, As, Y, Zr, Nb, Mo, Pd, Ag, C, In, Sn, Sb, Te, I, La, Ce, Pr, Nd, Sm, Gd, Tb, Dy, Yb, Hf, Ta e W, were used the limits detection of the methodology.

3. 3 1 Procedures for preparation of leaf samples for analysis by dispersive X-ray fluorescence (WDXRF)

The samples of leaves, after then dried, were pulverized (diameter>0.8 mm), with the aid of a porcelain mortar and pestle, previously washed with Extran and distilled water.

For the analysis of samples by the technique of wavelength dispersive X-ray fluorescence (WDXRF), were prepared pressed pellets with double-layer. The dust obtained after pulverization was compressed in a hydraulic press (HERZOG).

The methodology proposed of sample preparation has the advantage of being simple, and inexpensive, with no need for prior chemical treatment, such as digestion processes[15].

4. RESULTS

4.1. Species Identification

The species identification was performed by means of specific literature and consultation with specialist. The list of taxons was organized in order of collectors and is in the genus and species (Table 2).

Table 2– Species identified with scientific name (genus/species) and popular

Collector	Genus/Species	Popular name
1	*Magifera indica*	Mangueira
2	Tecoma stans	Ipê de jardim
3	*Psidium guajara*	Goiabeira
4	*Jacaranda mimosifolia*	Jacarandá mimoso
5	*Psidium guajara*	Goiabeira
6	*Morus Migra*	Amoreira
7	*Ulmaceae*	—
8	*Chromolucuma baehniana*	—
9	*Eugenia dysenterica*	Cagaita
10	Leucaena leucocephala	Linhaça

4.2 Litterfall Dry Production

The dry litterfall production during the period of development of the research was 5859.84 g year^{-1} (Goiabeira, *Psidium guajara*). The leaf fraction had a production of 420.44 g year^{-1}, followed by the wood fraction with 338.80 g year^{-1} and the parts reproductive fraction with only 97.80 g year^{-1}. The collector with lower production was the collector 8 with a productivity of 121.50 g year^{-1} divided enters the leaf fraction with 61.66 g year^{-1} and wood fraction with 59.84g year^{-1} (Table 3).

Table 3. Dry litterfall production (dry weight: g m^2 year^{-1}) during the development of research

	Compartment (dry weight (g m^2 year^{-1})			
COLLECTOR	**Leaves**	**Wood**	**Parts Reproductive**	**Total**
1	384.70	140.92	123.22	648.84
2	257.60	171.34	33.27	462.21
3	420.44	97.80	338.80	857.04
4	153.14	239.01	98.26	490.41
5	492.75	203.38	66.70	762.83
6	214.60	102.75	60.08	377.43
7	283.41	131.71	129.40	544.52
8	61.66	59.84	0.00	121.50
9	501.27	178.92	88.80	768.99
10	342.92	247.12	236.03	826.07
TOTAL	3112.49	1572.79	1174.56	**5859.84**

The anthropic forest around CQMA/IPEN, had an average annual production of litterfall of 5.86 T ha^{-1}, that's deposition it's considered normal according the literature[16].

By measuring the deposition of litterfall material by fraction, we could establish a relationship between these fractions and prevalence of each fraction.

The proportion of dry litterfall equivalent to proportion of humid litterfall, and the leaf compartment was predominant with 53.12% followed by wood compartment with 26.84% and reproductive parts compartment with 20.04% of total dry litterfall (FIG 2).

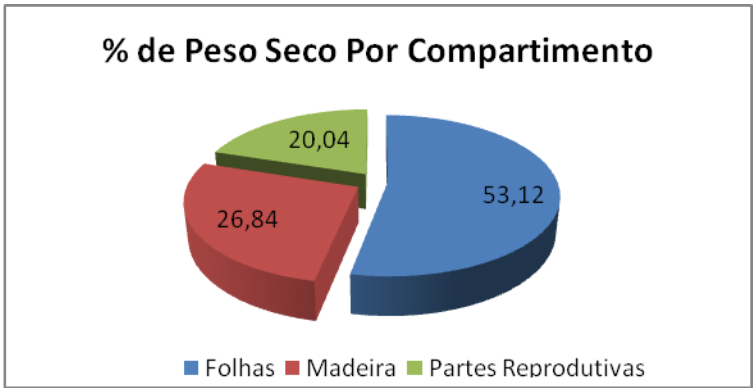

Figure 2– Percentage of dry litterfall per compartment

In the Figure 3 we could see the seasonal variation of litterfall deposition, of the compartments, Leaf, Wood and Reproductive Parts in the anthropic Forest around the CQMA/Ipen.

It's observed that most of the deposition of litterfall occurred between the months of June to September.

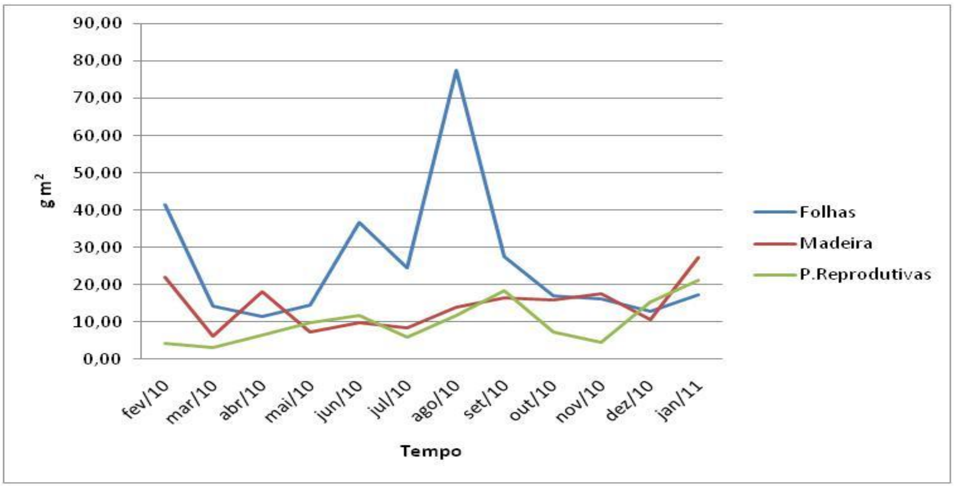

Figure 3 – Seasonal variation of litterfall production for the three compartments in g m² months^{-1}, for the anthropic forest around CQMA/Ipen

The values of Pearson correlation calculated between the compartments studied Leaves – Wood and Leaves – Reproductive Parts was 0.06, wich according to the classificatio[17], is considered very weak. The correlation between the compartments Wood – Reproductive parts (0.33) as established by the same classification, was considered only like a weak correlation.

The Pearson correlations calculated between the three compartments are shown in Table 4.

Table 4 – Pearson correlation values between the compartments studied

Correlação	(r^2)
Leaves x Wood	0.06
Leaves x Reproductive parts	0.06
Wood x Reproductive parts	0.33

4.3 Determination of metals and trace elements by X-ray Fluorescene

To check the reliability of the methodology were applied statistical tests with the certified reference materials Peach leaves - NIST 1547 and Mixed Polish Herbs - INCT-MPH-2 (MPH). The DPR (%) obtained for these elements was satisfactory (0.9 at 41.5) demonstrating the repeatability of tests. The accuracy (relative error) obtained was satisfactory and Z-test too. Therefore, the methodology could be applied in this study.

The Table 5 presents the results of the determinations of the concentrations of elements in mg kg^{-1} for the diagnosis leaves samples except for the macro constituents Ca, Si and K, expressed in (%). The values represent the average of the results of twelve monthly collections (n=12), in the sampled points.

Table 5 – Concentration, Average and Standard deviation for the elements determined in the compartments leaves, referring to twelve collections per collector (n=12)

Collector (number)	Average and Standard deviation								
		(%)				(ppm)			
	Ca	Si	K	P	Fe	Cl	Ni	Sr	Zn
1	2.3 ± 1.2	1.3 ± 1.0	0.4 ± 0.1	385 ± 137	305 ± 146	476 ± 481	8 ± 10	15 ± 12	22 ± 9
2	1.1 ± 0.4	0.1 ± 0.1	0.5 ± 0.1	788 ± 207	394 ± 151	373 ± 185	5 ± 8	6 ± 3	17 ± 3
3	1.5 ± 0.6	0.4 ± 0.2	0.8 ± 0.3	736 ± 286	259 ± 99	976 ± 750	3 ± 8	13 ± 6	22 ± 9
4	0.9 ± 0.3	0.2 ± 0.2	0.2 ± 0.1	711 ± 314	517 ± 268	184 ± 173	9 ± 7	5 ± 3	22 ± 9
5	1.2 ± 0.5	0.3 ± 0.1	0.6 ± 0.3	592 ± 208	231 ± 137	770 ± 700	7 ± 7	9 ± 5	51 ± 18
6	3.6 ± 1.0	1.9 ± 1.0	1.1 ± 0.4	1130 ± 480	539 ± 220	1591 ± 989	4 ± 7	36 ± 15	29 ± 13
7	3.0 ± 1.0	0.3 ± 0.1	0.7 ± 0.3	407 ± 169	359 ± 176	752 ± 614	3 ± 6	37 ± 15	12 ± 8
8	0.5 ± 0.4	0.2 ± 0.1	0.7 ± 0.4	785 ± 387	247 ± 119	203 ± 160	9 ± 6	2 ± 2	20 ± 7
9	1.7 ± 0.6	0.1 ± 0.1	0.7 ± 0.1	675 ± 325	233 ± 67	438 ± 357	14 ± 5	11 ± 6	44 ± 12
10	2.8 ± 0.8	0.2 ± 0.1	0.7 ± 0.3	918 ± 310	549 ± 273	553 ± 253	1 ± 4	24 ± 8	27 ± 9
Branco	2.0 ± 0.0	2.2 ± 0.1	0.2 ± 0.2	248 ± 107	164 ±36	219 ± 223	7 ± 11	36 ± 13	11 ± 3

Collector (number)	Average and Standard deviation (ppm)									
	Cu	Th	U	Mn	Al	Ti	Na	Mg	S	Br
1	7 ± 7	<15	<17	905 ± 384	792 ± 433	<15	79 ± 99	1222 ± 397	880 ± 366	<15
2	15 ± 3	<15	<17	35 ± 16	851 ± 454	<15	27 ± 64	1742 ± 504	1383 ± 338	<15
3	20 ± 8	<15	<17	54 ± 19	963 ± 509	<15	47 ± 89	1658 ± 470	3233 ± 1209	<15
4	13 ± 5	<15	<17	12 ± 15	1790 ± 984	<15	18 ± 58	1410 ± 335	1230 ± 356	<15
5	17 ± 5	<15	<17	38 ± 13	840 ± 625	<15	112 ± 125	1410 ± 265	2375 ± 836	<15
6	9 ± 9	<15	<17	140 ± 57	859 ± 407	<15	47 ± 87	2208 ± 814	3392 ± 2531	<15
7	10 ± 7	<15	<17	600 ± 194	1005 ± 807	<15	13 ± 44	1397 ± 1052	982 ± 294	<15
8	11 ± 6	<15	<17	79 ± 109	573 ± 289	<15	97 ± 110	3425 ± 1066	1221 ± 417	<15
9	17 ± 6	<15	<17	187 ± 89	593 ± 170	<15	199 ± 169	2692 ± 825	1200 ± 479	<15
10	14 ± 7	<15	<17	34 ± 9	1345 ± 1273	<15	23 ± 54	1667 ± 975	2500 ± 756	<15
Branco	-	<15	<17	898 ± 286	457 ± 308	<15	-	918 ± 116	820 ± 57	<15

The average of concentration of elements found in all collectors had the following that's order: Ca > K > Si > Mg > S > Al > P > Cl > Fe > Mn > Na >Zn > Sr > Cu > Br > Ti >Ni > Th = U.

5 CONCLUSIONS

The production of litterfall on the forest around the nuclear facilities from CQMA/IPEN was 5.0 kg m^2, being represented by 53.12% of leaves, 26.84% of wood and 20.04% of reproductive parts. The average annual production of litterfall was 5.86 t ha^{-1}, which is considered a normal deposition according to the literature[16].

The correlation between Leaves – Wood and Leaves – Reproductive parts (0.06) was considered very weak and between compartments Wood – Reproductive parts (0.33) was considered weak, according to the methodology proposed[17].

The concentrations of elements in the leaves of the litterfall in this study had following decreasing order: Ca > K > Si > Mg > S > Al > P > Cl > Fe > Mn > Na >Zn > Sr > Cu > Br > Ti >Ni > Th = U.

According to the results obtained can be observed that the toxic metals were not detected in diagnosis leaves samples made in this study, whose limit f detection for Ba, Cd, Cr, Ni, Pb, U, Th were de 10; 32; 7; 8; 12; 17; 15 mg Kg^{-1}, respectively. In literature has not found reference data of the maximum concentration of these metals in leaves of the litterfall similar to those evaluated.

This study showed no contamination in the diagnosis leaves through atmospheric deposition or increased through growth from litterfall in the forest around CQMA-IPEN-CNEN/SP, in relation to toxic metals analyzed. The nuclear facilities existing or disabled not provide increases in the concentrations of Ba, Cd, Cr, Ni, Pb, U, Th in the diagnosis leaves for species analyzed.

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