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Pb-210 DEPOSITION MEASURED IN RAINFALL IN SÃO PAULO, SP – BRAZIL

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ABSTRACT

Pb-210 ($T_{1/2}$ = 22.3 y), a natural radionuclide from U-238 serie can be found in the atmosphere, as a product of ^{222}Rn decay that emanates from the ground, where its atoms become rapidly fixed to aerosols and return to the earth as dry fallout or are washed out in the rain. This natural radionuclide has been widely used as an atmospheric tracer, to determine the aerosol residence time as well as chronometers in the environment. Pb-210 was measured during a period of two years, 2011 to 2013, in samples of rainfall in all the rainy events that ocurred at the Instituto de Pesquisas Energéticas e Nucleares (IPEN) campus (23°33'59.24" S - 46°44'15.63" O at 760 m above sea level) which is located in the city of São Paulo, in the state of São Paulo, Brazil. Pb-210 concentration was measured in a total of 123 rainy events by beta gross counting in a low background gas flow proportional detector, after radiochemistry procedure. The results obtained were correlated to seasons and rainfall. The concentrations of ^{210}Pb in rainfall varied from the minimum detectable activity, 4.9 mBq L^{-1} to 1408 ± 43 mBq L^{-1}. The highest concentrations were obtained in the months of winter and the lowest in summer. The monthly depositional flux of ^{210}Pb, varied from 4.03 Bq m^{-2} $month^{-1}$ to 46.4 Bq m^{-2} $month^{-1}$ presenting a strong correlation with the amount of precipitation and hence showing seasonal trends.

1. INTRODUCTION

The natural radionuclide ^{210}Pb ($T_{1/2}$ = 22.3 y) has been widely used as tracer of soil erosion, transport processes in watershed and as chronometers in the environment. It is present in the atmosphere as decay products of the gas ^{222}Rn. The ^{222}Rn is replenished in the atmosphere by emanation from the surface of the earth where it is produced by decay of ^{226}Ra in geological materials. ^{210}Pb return to the earth as dry fallout or are washed out in rain. As ^{222}Rn is a noble gas that emanates predominantly from the Earth's continental crust, the activity of ^{210}Pb in the air is longitudinal dependant, and its activity will decrease with elevation from the Earth's surface due to a decrease in the activity of ^{222}Rn [1-5].

^{210}Pb is scavenged by aerosols and subsequently removed from the atmosphere, primarily by precipitation. In areas where the amount of rainfall is low, the dry fallout can be a significant component of the bulk depositional flux while in areas where there is a relatively higher amount of rainfall, the wet fallout is the major component (>90%) of the bulk depositional flux. In general, the amount of rainfall controls the seasonal variations of the depositional fluxes of ^{210}Pb, while precipitation-normalized depositional fluxes during spring are higher in the middle latitudes [6-10].

From measurements of the activities of ^{210}Pb in rainfall, it is possible to estimate its annual flux, which is useful in understanding sediment chronology. The behavior of ^{210}Pb provides knowledge on the behavior of other chemical species in atmospheric aerosols and knowledge of site-specific atmospheric fluxes is also crucial to studies of the impact of atmospherically delivered pollutants on terrestrial ecosystems. This isotope has also been used to determine aerosol residence time and removal rates of aerosols.

The chemical composition of rainfall is a combination of the chemical composition of the droplets that form clouds and substances that incorporate the raindrops during precipitation. Thus, rainfall, somehow, depicts characteristics of air mass with particles and water soluble gases, which cross the raindrops during rainfall. An evidence of this is the change in chemical composition of rainwater in relation to weather. Another evidence is the inverse relationship between the total amount of dissolved ions and the quantity of rainfall, suggesting that the majority of the present ions in the rainfall are incorporated to the water during the precipitation, process known as below-cloud removal.

The atmospheric deposition is a major mechanism of cycling and redistribution of various chemicals on the planet's surface, exerting, therefore, a key role in continental and coastal biogeochemical processes. The qualitative and quantitative knowledge of atmospheric deposition is relevant to the understanding of biogeochemical cycles of elements and the influence of human activities on these processes [11].

In the city of São Paulo, the first studies of chemical characterization of rainfall were conducted between November 1983 and February 1985 using manual collection in a single sampling point, with determinations of free acidity and total acidity. In this period, the average pH of rainfall recorded was equal to 4.5, which characterized the phenomenon of acid rain [12].

There is limited data on the depositional fluxes of ^{210}Pb in the precipitation in Brazil, compared with data from Northen Hemisphere [4, 6, 8-10, 13-17]. The aim of this paper is to present preliminary results of ^{210}Pb concentrations in rainfall collected in the city of São Paulo - Brazil to provide the baseline data for the bulk depositional fluxes of ^{210}Pb. For a period from April 2011 to June 2013 (26 months), rainfall samples were collected and measured and the resulting seasonal ^{210}Pb data were correlated to seasons and precipitation. It was also possible to estimate the depositional flux of this natural radionuclide in the city of São Paulo.

2. MATERIALS AND METHODS

2.1. Site Description

The sampling site is located at IPEN – Instituto de Pesquisas Energéticas e Nucleares, FIG. 1, which is approximately 10 km west from downtown São Paulo. The city of São Paulo is situated on a plateau in southeastern Brazil, at latitude 23^{o}33’59.24” S and longitude 46^{o}44’15.63” O, having an average altitude of 760 m above sea level. Climate in the area is temperate tropical with dry period in winter and rainy in summer. The annual average temperature is 19.1^{o}C, showing minimum and maximum of 15.3^{o}C and 24.9^{o}C, respectively. The annual rainfall in the city, for the studied period, averaged from 1590 mm to 2081 m. The winds in IPEN campus are predominantly from the SSE sector.

Figure 1: Brazil and São Paulo map and IPEN aerial view.

2.2. Sampling

Rainfall was collected in each rainy event from April 2011 to June 2013, in a polyethylene container of 0,132 m^2, totaling 123 rainy events. It was only considered precipitation values grater than 5 mm, because this volume is used to characterize different radionuclides deposition process [13, 18]. At the end of each rainy event and when the sample was collected, the container was rinsed with 300 mL of 1M HNO_3 and 200 mL super pure water, which were added to the sample. The amount of precipitation in mm was obtained from a rain gauge installed at the sampling point and the pH was measured in this rainfall collected. Since the rainfall container was exposed to the atmosphere continuously, the total deposition was collected, that is, the wet and dry fallout. The entire collected sample was acidified with HNO_3 at pH lower than 2, in order to avoid losses due to adsorption on the concentration process, and then concentrated to a final volume of 100 mL.

2.3. ^{210}Pb determination

^{210}Pb concentration was determined by radiochemical procedure that consists of an initial precipitation of Pb with 3M H_2SO_4, dissolution of the precipitate with nitrilo-tri-acetic acid at basic pH, and precipitation of $^{210}PbCrO_4$ with 30% sodium chromate. The ^{210}Pb concentration was determined through its decay product, ^{210}Bi, by measuring the gross beta activity of the $^{210}PbCrO_4$ precipitate. The chemical yields were determined by gravimetric analysis. The methodology minimum detectable activity, MDA, is 4.9 mBq L^{-1}. ^{210}Pb was determined in a low background gas flow proportional detector [19].

3. RESULTS AND DISCUSSION

Table 1, Table 2 and Table 3 present the sampling date, the amount of rainfall in mm, pH values, ^{210}Pb concentration in mBq L^{-1}, and ^{210}Pb flux in Bq m^{-2}, for each rainy event that occurred from April to December 2011, in 2012 and from January to June 2013, respectively.

The amount of precipitation for the study period ranged from 5 to 160 mm of each rainy event; in September of 2011 and August of 2012 there was no precipitation.

The pH values of each rainy event ranged from 4.30 to 7.42, characterizing the rainfall from lightly acid to lightly basic.

^{210}Pb concentrations ranged from the minimum detectable activity, 4,9 mBq L^{-1} obtained in rainfall of 12/18/2011, to 1408 ± 43 mBq L^{-1} and the ^{210}Pb flux results ranged from 0.13 Bq m^{-2} to 22.77 Bq m^{-2}.

Table 1: Sampling date, volume (mm), pH, ^{210}Pb concentration (mBqL^{-1}) and ^{210}Pb flux (Bq m^{-2}) for the samples collected in 2011.

Sampling 2011	V mm	pH	^{210}Pb mBqL^{-1}	^{210}Pb flux Bq m^{-2}	Sampling 2011	mm	pH	^{210}Pb mBqL^{-1}	^{210}Pb flux Bq m^{-2}
02/04	52	4,61	35 ± 4	1,59	11/10	116	5,25	86 ± 9	9,10
08/04	8	4,72	211 ± 19	1,40	26/10	13	7,03	250 ± 20	2,79
12/04	65	nm	73 ± 6	4,43	29/10	24	6,60	130 ± 16	2,79
21/04	22	5,72	294 ± 20	5,46	12/11	126	5,58	30 ± 3	2,99
27/04	16	5,86	131 ± 18	1,74	22/11	8	6,73	1044 ± 88	7,15
01/05	6	5,90	42 ± 4	0,23	29/11	20	6,20	122 ± 13	1,77
15/05	12	nm	378 ± 24	3,80	01/12	10	6,87	44 ± 5	0,38
07/06	25	6,50	444 ± 16	9,32	07/12	18	6,84	158 ± 7	2,26
09/06	25	nm	144 ± 10	2,89	08/12	30	6,88	22 ± 1	0,55
26/06	23	5,50	272 ± 19	5,26	10/12	18	6,38	74 ± 4	0,92
30/07	6	6,30	1408 ± 43	7,08	14/12	10	6,47	160 ± 16	1,79
20/08	25	6,48	314 ± 18	6,68	15/12	25	6,80	122 ± 19	2,72
31/08	36	5,44	400 ± 12	13,34	18/12	5	7,42	< MDA	-
02/10	20	6,44	1275 ± 20	6,20	24/12	20	6,83	115 ± 8	1,48
08/10	13	6,40	500 ± 48	5,44	30/12	58	6,59	83 ± 5	3,83

nm: not measured
- MDA: 4.9 mBq L^{-1}

Table 2: Sampling date, volume (mm), pH, ^{210}Pb concentration (mBqL^{-1}) and ^{210}Pb flux (Bq m^{-2}) for the samples collected in 2012.

Sampling 2012	mm	pH	^{210}Pb mBqL^{-1}	^{210}Pb flux Bq m^{-2}	Sampling 2012	mm	pH	^{210}Pb mBqL^{-1}	^{210}Pb flux Bq m^{-2}
05/01	14	5,84	197 ± 20	2,28	12/05	14	6,70	359 ± 27	3,79
10/01	34	6,19	69 ± 2	0,47	25/05	23	6,22	264 ± 14	4,78
12/01	28	6,49	96 ± 6	1,32	01/06	5	6,74	637 ± 68	0,58
14/01	22	5,90	86 ± 4	1,44	07/06	120	5,69	93 ± 6	11,69
17/01	30	6,17	117 ± 13	2,68	19/06	62	6,21	185 ± 7	8,41
18/01	30	5,87	127 ± 13	2,86	20/06	30	6,12	118 ± 11	2,85
20/01	48	5,92	11 ± 1	0,43	07/07	15	6,49	93 ± 6	1,11
24/01	66	5,27	105 ± 7	5,36	12/07	7	6,84	612 ± 53	3,01
11/02	160	5,21	43 ± 3	6,12	16/07	90	5,91	363 ± 34	22,77
14/02	15	4,84	1145 ± 24	13,10	19/09	5	6,41	1049 ± 82	3,66
17/02	15	4,54	342 ± 24	4,02	25/09	12	5,71	91 ± 90	0,65
19/02	22	5,27	207 ± 26	3,14	11/10	16	6,55	244 ± 6	3,79
22/02	6	6,00	213 ± 33	1,05	17/10	15	6,37	285 ± 43	4,38
23/02	5	6,70	505 ± 56	1,34	19/10	5	6,32	774 ± 11	2,76
24/02	12	5,81	487 ± 72	2,80	22/10	14	4,96	349 ± 30	5,10
27/02	10	4,35	232 ± 8	1,95	27/10	28	5,18	239 ± 9	6,52
02/03	14	7,02	244 ± 21	1,92	08/11	7	5,90	109 ± 8	1,78
10/03	50	6,32	161 ± 6	6,77	10/11	94	6,08	80 ± 5	7,53
13/03	8	6,17	501 ± 28	3,23	20/11	22	5,85	155 ± 11	2,33
15/03	12	6,20	100 ± 6	0,87	24/11	5	5,75	142 ± 6	0,61
22/03	54	6,14	86 ± 4	3,87	26/11	19	5,30	43 ± 3	1,17
24/03	8	6,30	126 ± 10	0,70	01/12	27	5,51	205 ± 27	1,56
27/03	10	6,55	133 ± 6	1,20	05/12	10	5,60	274 ± 10	5,68
08/04	8	6,83	264 ± 18	1,64	13/12	15	5,97	129 ± 20	1,87
11/04	70	5,04	302 ± 7	19,72	15/12	76	5,83	60 ± 2	1,73
12/04	18	5,45	101 ± 2	1,48	17/12	5	5,30	260 ± 20	4,80
21/04	30	6,56	152 ± 16	3,43	19/12	30	5,93	160 ± 7	1,71
26/04	38	6,03	41 ± 6	1,55	20/12	21	5,80	134 ± 13	1,05
27/04	70	6,28	73 ± 4	4,13	27/12	18	5,60	112 ± 7	4,09
					29/12	62	4,98	135 ± 4	2,45

Table 3: Sampling date, volume (mm), pH, ^{210}Pb concentration (mBqL^{-1}) and ^{210}Pb flux (Bq m^{-2}) for the samples collected in 2013.

Sampling 2013	mm	pH	^{210}Pb mBqL^{-1}	^{210}Pb flux Bq m^{-2}	Sampling 2013	mm	pH	^{210}Pb mBqL^{-1}	^{210}Pb flux Bq m^{-2}
08/01	6	6,21	334 ± 27	2,20	07/03	13	4,72	173 ± 9	1,87
12/01	38	5,92	49 ± 9	1,61	08/03	85	5,08	126 ± 12	11,33
14/01	46	5,38	72 ± 3	3,19	11/03	6	4,98	171 ± 9	0,74
16/01	25	5,67	6 ± 1	0,13	13/03	6	6,00	164 ± 19	1,34
19/01	5	5,78	242 ± 14	0,83	16/03	25	6,01	42 ± 6	0,85
26/01	16	6,7	141 ± 13	2,28	20/03	13	6,10	164 ± 6	1,80
01/02	20	6,19	114 ± 10	2,14	24/03	10	6,20	205 ± 12	1,86
06/02	5	6,27	341 ± 21	1,78	26/03	22	5,87	89 ± 6	2,19
07/02	12	5,42	109 ± 14	1,35	05/04	7	5,80	188 ± 9	1,95
08/02	25	5,91	180 ± 12	3,61	06/04	5	5,90	221 ± 17	1,16
14/02	95	5,04	162 ± 6	12,15	12/04	30	6,20	80 ± 3	1,48
15/02	28	6,10	226 ± 13	5,05	22/05	12	6,80	234 ± 18	2,96
18/02	44	5,93	233 ± 7	6,80	28/05	6	6,80	214 ± 12	1,67
19/02	34	4,3	236 ± 3	7,51	01/06	26	6,23	162 ± 16	3,68
23/02	7	4,91	802 ± 14	3,34	11/06	9	5,28	213 ± 16	1,68
26/02	14	6,03	175 ± 20	2,40	16/06	16	5,69	177 ± 14	2,79
					24/06	94	5,57	229 ± 6	20,99

The monthly amount of precipitation for the studied period is plotted in Fig. 1. Comparing the amount of precipitation among the 26 months of study, it is possible to indentify the climate of São Paulo, temperate tropical, with dry periods in winter and rainy in summer only in 2011, where the winter months presented the lowest values of rainfall.

In 2012 the months of June and July presented high indexes of rainfall, which can be comparable to the summer months of January, February and March. In 2013 the months of June and July also presented high indexes, like in 2012.

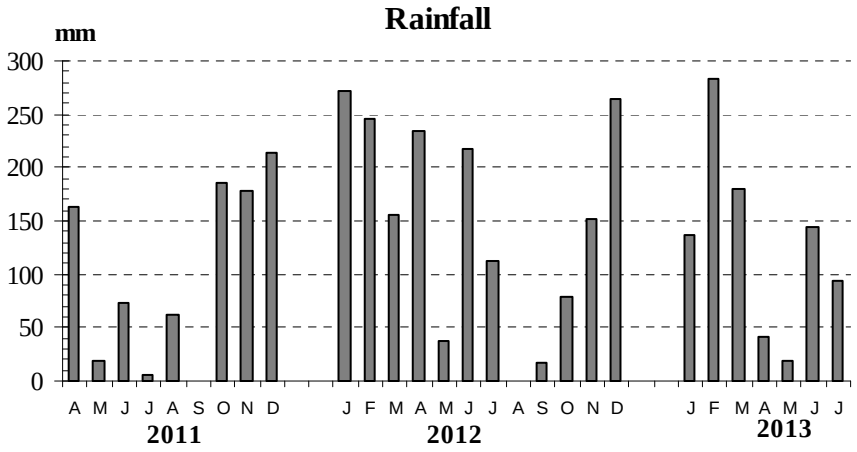

Figure 1: Monthly precipitation in the city of São Paulo during the sampling period (April 2011 to June 2013).

The Radiological Environmental Monitoring Program of IPEN - Programa de Monitoração Radiológica Ambiental - PMRA, which was established in 1992 [20], has data of rainfall (measured inside of IPEN campus) since 1993, Fig. 2; as it can be seen, in almost 20 years of measuring and up to present time, the year of 2012 had the highest rainfall recorded [21].

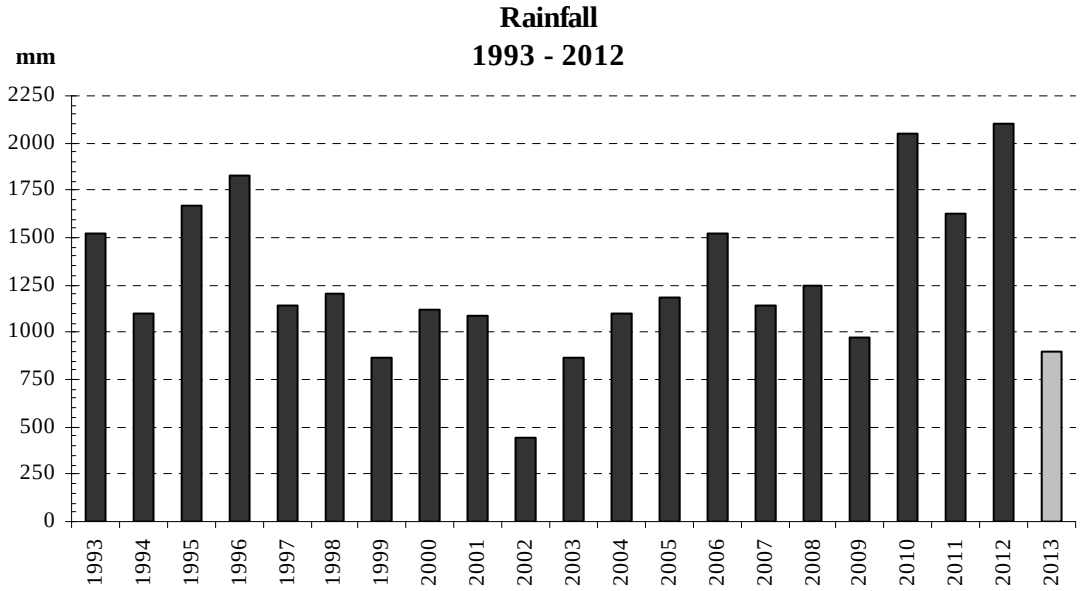

Figure 2: Annual rainfall measured by PMRA, IPEN since 1993 (2013 - from January to July).

Fig. 3 presents the rainfal volume, mm, and the pH values for the samples collected in 2011. It can be observed, for the studied period of 2011, that the months with higher rainfall volume were the months of spring followed by autumn and that the pH values ranged from 4.61 to 7.42

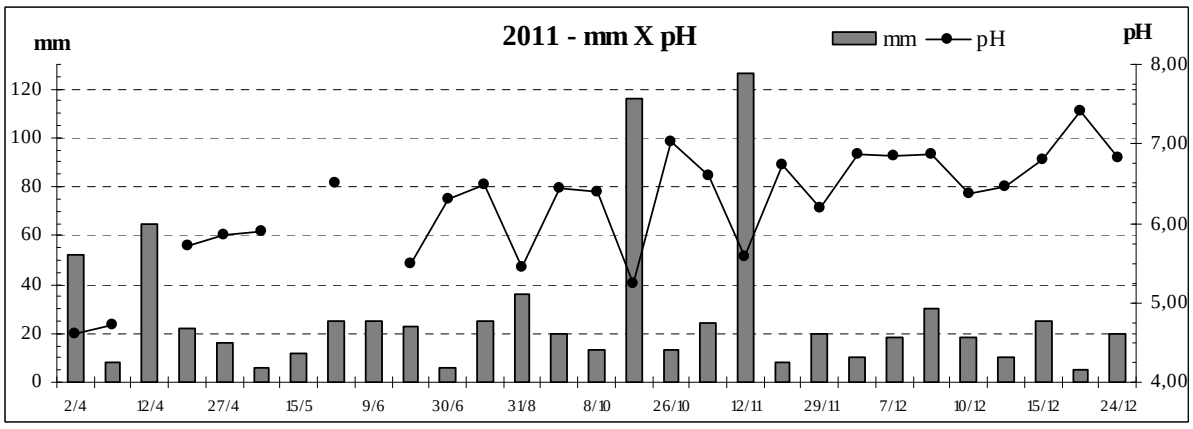

Figure 3: Rainfall volume, mm, and pH values for the samples collected in 2011.

Fig. 4 presents the rainfal volume, mm, and the pH values for the samples collected in 2012. It can be observed, for the year of 2012, that the months with higher rainfal volume were the months of spring followed by autumn and that the pH values ranged from 4.35 to 7.42.

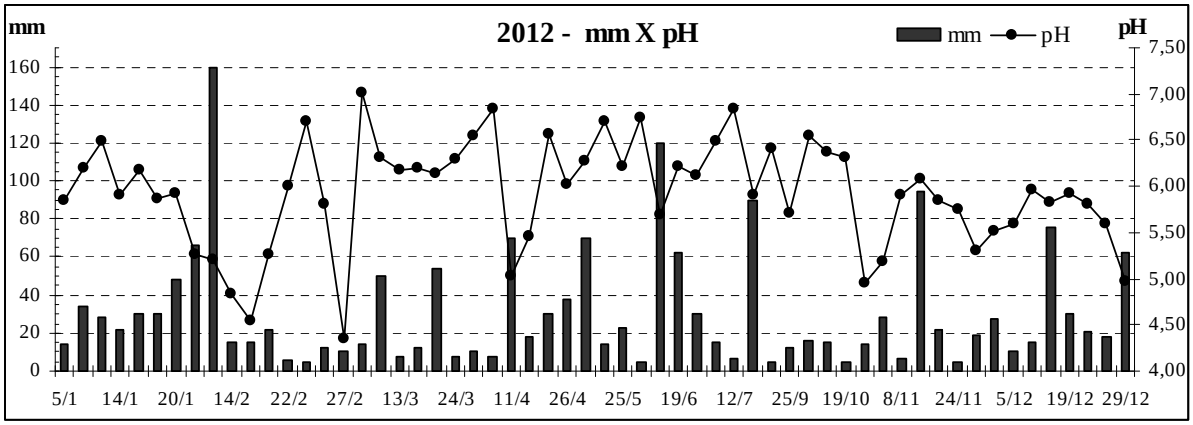

Figure 4: Rainfall volume, mm, and pH values for the samples collected in 2012.

Fig. 5 presents the rainfal volume, mm, and the pH values for the samples collected in 2013. It can be observed, for the studied period of 2013, that the months with higher rainfal volume were the months of spring followed by autumn and that the pH values ranged from 4.35 to 6.80.

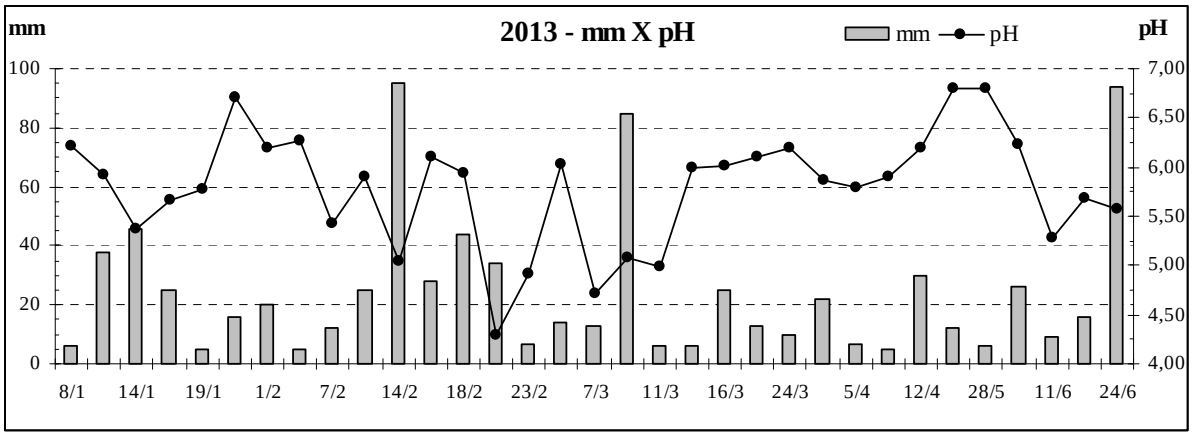

Figure 5: Rainfall volume, mm, and pH values for the samples collected in 2013.

Fig. 6, 7 and 8 present the ^{210}Pb concentrations, mBq L^{-1}, and the rainfall in mm, for 2011, 2012 and 2013, respectively. For the whole period studied it was found that the highest concentrations of ^{210}Pb were obtained in the winter months in which smaller amounts of rainfall occur. Moreover, large amounts of rainfall cause the sample's dilution and hence lower concentrations of ^{210}Pb.

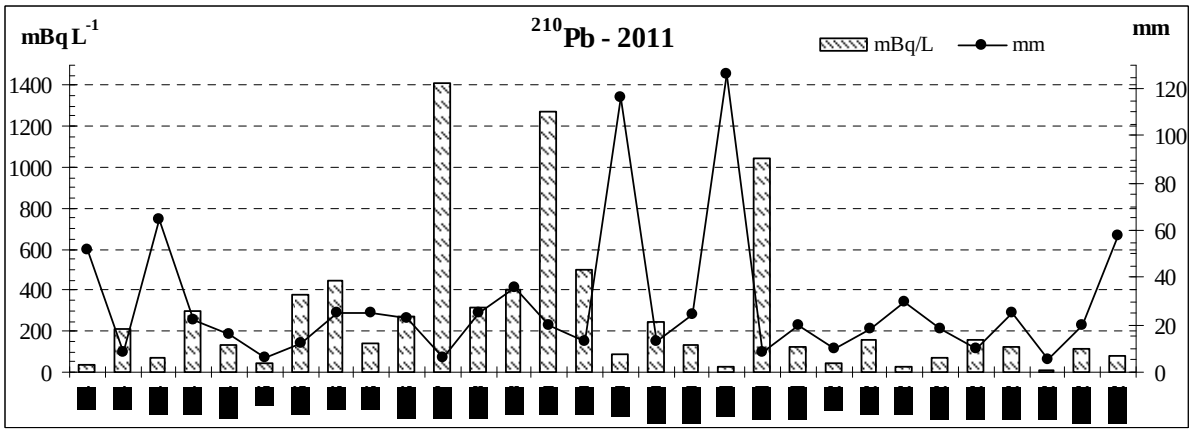

Figure 6: ^{210}Pb concentrations, mBq L^{-1}, and rainfall, mm, for 2011

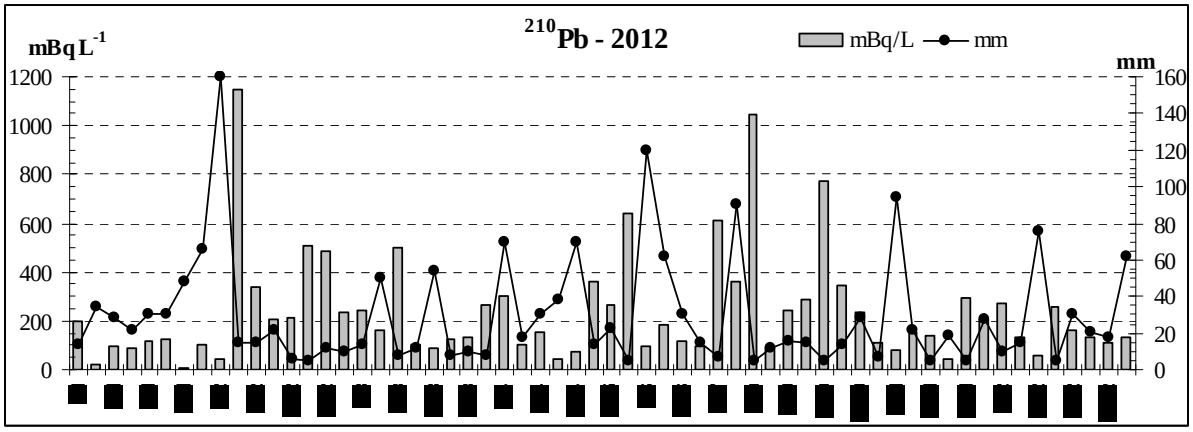

Figure 7: ^{210}Pb concentrations, mBq L^{-1}, and rainfall, mm, for 2012

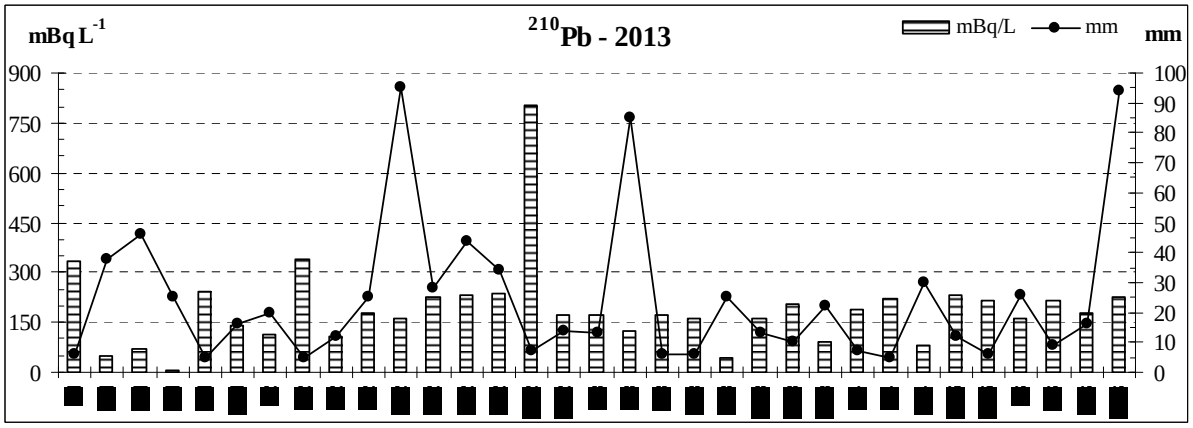

Figure 8: ^{210}Pb concentrations, mBq L^{-1}, and rainfall, mm, for 2013.

Fig. 9 presents the monthly depositional flux of ^{210}Pb, Bq m^{-2} month^{-1}, and the rainfall, mm, for the studied period and Fig. 10 the relationship between the ^{210}Pb flux per season and the

amount of rainfall. The monthly flux varied from 4,03 Bq m^{-2} $month^{-1}$ to 46,4 Bq m^{-2} $month^{-1}$, the highest flux was obtained in the month where the highest rainfall occurred, February 2013, followed by February and April of 2012, summer and fall. The high values obtained for ^{210}Pb flux could be due to higher release rates of ^{222}Rn from the continent due to warmer temperatures in summer.

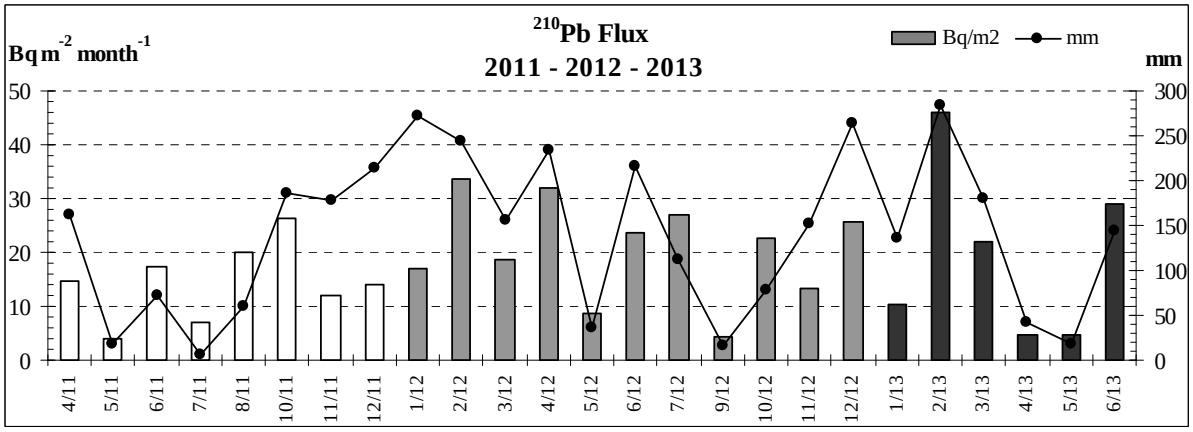

Figure 9: Monthly depositional flux of ^{210}Pb, Bq m^{-2} $month^{-1}$, and rainfall, mm, for the studied period

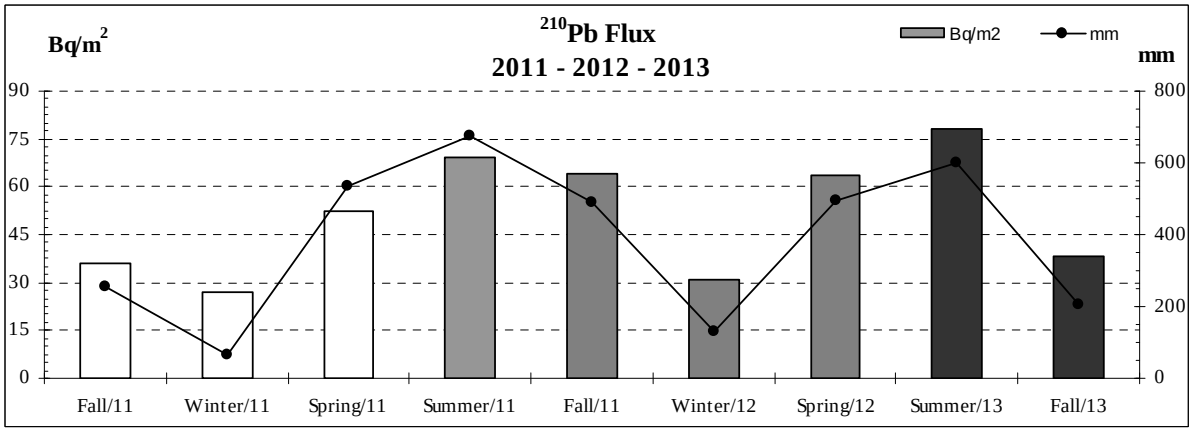

Figure 10: Relationship between the ^{210}Pb flux, Bq m^{-2}, per season and the amount of rainfall, for the studied period

4. CONCLUSIONS

This paper presets the preliminary results for ^{210}Pb concentration measured in all the rainy event that occurred from April 2011 to June 2013, at IPEN, São Paulo, SP, Brazil in order to provide the baseline data for the bulk depositional fluxes of ^{210}Pb in Brazil and southern hemisphere and to understand the atmospheric behavior of this radionuclide.

The concentrations of ^{210}Pb in rainfall varied from the minimum detectable activity, 4.9 mBq L^{-1} to 1408 ± 43 mBq L^{-1}. The highest concentrations were obtained in the months of winter and the lowest in summer.

The monthly depositional flux of ^{210}Pb, during the studied period, did not remain constant, but varied from 4.03 Bq m^{-2} $month^{-1}$ to 46.4 Bq m^{-2} $month^{-1}$ presenting a strong correlation with the amount of precipitation and hence showing seasonal trends.

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