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EVALUATION OF CORRELATING FACTORS BETWEEN ^{238}U CONCENTRATION MEASURED IN FINE AND COURSE ATMOSPHERIC PARTICLES

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ABSTRACT

Air quality is ever more important in function of the enormous proportion of human actions that have affected the environment over the last two centuries. Particulate material is one among many pollutants that can cause great risk to human health and the environment. It can be classified as: 1) Total Suspended Particles (TSP), defined simply as particles with less than 50 μm aerodynamic diameter (one group of these particles can be inhaled and may cause health problems, while others may unfavorably affect the population's quality of life, interfering in environmental conditions and impairing normal community activities); and 2) Inhalable Particles (PM_{10}), defined as those particles with less than 10 μm aerodynamic diameter. These particles penetrate the respiratory system and can reach pulmonary alveoli due to their small size, causing serious health damage. The Nuclear Technology Development Center (CDTN) has monitored air quality around its installations since 2000. CDTN's Environmental Monitoring Program (EMP) includes monitoring radioactivity levels contained in atmospheric TSP. In order to optimize its program, CDTN is carrying out a study to estimate the correlation between concentrations of particulate material measured in TSP and those measured in PM_{10}, $PI_{2.5}$ and PI_1, as well as determination of activity concentration for each controlled radionuclide in all parts. The objective of this study is to present preliminary results and report ^{238}U activity concentration results.

1. INTRODUCTION

An increase in atmospheric emissions has been caused by urban population growth and industrialization levels.

Emission of particulates is important among several pollutants from anthropogenic sources, in which finer particles are recognized for having a strong impact in the environment.

Particulate material is a term employed for solid or liquid particles found in the air, originated from immobile or mobile sources. Their chemical and physical characteristics are varied.

Currently, $PI_{2.5}$ aerosol monitoring is more recommended than PI_{10}, due to the fact that these particles present a direct impact on health and their sampling avoids interference of natural particles (<100 μm). However, industrial activity with high emission has a great influence on air quality, due to intense emission of particulates in the 10 – 2.5 μm range[1].

Fine particles ($PI_{2.5}$) contain potentially hazardous chemical species, such as acids, heavy metals, and aromatic polycyclic hydrocarbons. Compared to particles greater than 10 μm in diameter, these particles are directly related to anthropogenic emissions. Size of particles, as well as their physical and chemical properties influences its residence time in the atmosphere [1].

CDTN has operated an Environmental Monitoring Program since 1986, the objective of which is to control and evaluate environmental radiological impact from developed activities [2].

Among several matrices and planned analyses, this program monitors aerosol in order to characterize radioactive material in suspension and concentration of Particulate Material in Suspension.

The objectives of monitoring aerosol are:

- Evaluate conformity and maintenance of air quality standards;
- Provide research data;
- Follow up trends and changes in air quality, due to alteration of pollutant's emission;
- Provide data to support licensing, inspection, and control actions.

Within CDTN's EMP, three permanent monitoring points have been established since 1986, according to their pollution source (within CDTN). Samples are collected with high air volume samplers (Hi-VOL). These points were established through a region climatic study in order to observe the needs for following up trends and changes in air quality due to alterations in pollutant emission.

Radiometric characterization of particles is of great importance within the EMP, which evaluates CDTN contribution regarding a possible increase of regional natural concentrations.

The analysis program is restricted to critical radionuclides from Uranium and Thorium series, i.e. those that probably contribute to a greater annual dose by ingestion, inhalation, or external radiation [3].

In order to optimize its EMP, CDTN is carrying out a study to estimate the existing correlation between measured concentrations of particulate material in TSP (currently monitored) and those measured in PM_{10}, $PI_{2.5}$ and $PI_{1,}$ as well as determination of activity concentration for each monitored radionuclide in every fraction. This study presents preliminary results and ^{238}U activity concentrations.

2. METHODOLOGY

2.1 – Experiment

Aerosol sampling (PI_{10}) followed established USEPA-15 criteria. Data were collected the continuously for 12 consecutive days, using HV, PM_{10}, and Dicotomic samplers.

HV PM10 is a large volume collector for environmental particles smaller than 10 μm (Figure 2). Its working principle is *impacting*, in which particles are directed to a micro-quartz filter for retention (Whatman QM-A – 20.3 cm x 25.4 cm). The device keeps an operational flow of 1.13 m^3 min^{-1} (with 10 % tolerance) in ambient temperature and pressure conditions.

The Dicotomic sampler (PI_{10} and $PI_{2.5}$) separates the environment air particles into two distinct groups: coarse, from 10 to 2.5 μm and fine, less than 2.5 μm. The sampler input allows only particles smaller than 10 μm to enter; it separates them into two groups and collects them on HTTP-type 37 mm, 0.4 μm porosity polycarbonate membrane filters. This sampler operates at a 16.7 L min^{-1} rate with a 10% tolerance. In the coarse particle receptor tube, a 1.67±10% L min^{-1} flow is collected while the fine particles follow through a 15 L min^{-1} flow collection filter.

To determine particulate matter mass concentration in the HV, PM10 and Dicotomic samplers, quartz and polycarbonate fiber filters were utilized, respectively, which were identified and mounted in a weighing chamber before and after collection for a continuous 24 h period in order to avoid hydration of the filters surface following USEPA15 guidelines [4].

The filters were weighed on two analytical scales in order to observe any type of error that may have been made by the operator and/or the equipment itself (Metler Toledo – AG 245) with 0.001 mg precision.

Sampling was carried out simultaneously for a 12-day period at two EMP sampling stations, one considered the blank point (Point 01), that is, unaffected by the facility and the other (Point 2), located in the direction of the prevailing winds. A Hi-Vol sampler was installed in both of the sampling stations for TSP measurements. An HV PM10 sampler was also installed at station 01 and a Dicotomic sampler at station 02.

Concentration of particles suspended in environment air (μg/m^3) was then gravimetrically determined, relating the mass retained in the filter and the volume of air suctioned. In addition, a retarded neutron method ^{238}U analysis was realized in all of the filters.

2.2 – Analytical Methodology for ^{238}U determination by Fission-Retarded Neutron

A 123 cm^2 area was cut from the filter on which the particulate material from the air had been collected. This aliquot was then folded and conditioned in irradiation tubes.

The samples were irradiated in a 100 kW TRIGA MARK-I IPR-RI reactor under a thermal 1.7 x 10^{10} neutron flow. cm^{-2} s^{-1} for the required time. After irradiation and sufficient decay time, fission retarded neutron measure was carried out in a BF_3 detector. The entire sample irradiation, decay and counting process were executed by means of an automated pneumatic system.

The minimum detection limit for fission-retarded neutron ^{238}U analysis is 0.02 mBq/m^3 for a 123 cm^2 aliquot.

2.3 – Statistical Treatment

Correlating factors were determined by applying statistical techniques to the concentration results for suspended particulate material. Data comparison was also carried out for paired data with the objective of verifying equality between the pairs of values of the desired interval, being careful to ensure that the individual values that integrate in the same pair were as similar as possible. The following tests were employed [5,6,7 and 8]:

- T-paired and Confidence Interval tests

There are two samples in a studied pair but each observation of the first sample is paired with an observation from the second sample. As expected, the two observations from the same pair are more probable to be similar and thus aren't considered statistically independent. We can calculate the confidence interval for the **average difference** and test whether average difference is equal to zero or not. Note that in this case we're interested in the **average difference**, while when we have two independent samples, we'll be interested in the **difference of the averages**.

- Kruskal-Wallis Test

In order to verify the existence of statistically significant differences between the size ranges of particulate material among the sample points, a Kruskal-Wallis non-parametric test was utilized. This test evaluates if two or more data sets represent the same population by comparing their medians, calculating a level of significance called "p value". For confidence level 95%, there is a statistically significant difference if "p value" is less than or equal to 0.05 [9].
In this case, all of the results that had a "p value" higher than 0.05 indicated that the data set has the same population and consequently can be considered not to have significant variation of particle concentration regarding the sample point.

3. RESULTS

In order to verify possible errors from operation or calibration of equipment, before the evaluation of the measurements and analyses itself, a comparison between the results obtained for quantity of particulate material was carried out on the two scales utilized.

Measurements from to the two scales for total particulate material, TSP, and PM_{10} at point 01 are shown on Tables 1 and 2 below. Tables 3 and 4 show t test results applied to the data set in order to verify equality between the two measured groups (scale 1 versus scale 2).

Table 1 – Estimated PM_{10} particulate material mass at point 01

Date		*(Scale 1)*			*(Scale 2)*		
Start Date	**End Date**	**Initial (mg)**	**Final (mg)**	**Mass (µg)**	**Initial (mg)**	**Final (mg)**	**Mass (µg)**
18/5/09	19/5/09	2.7402	2.7889	48.7	2.7304	2.7775	47.1
19/5/09	20/5/09	2.749	2.8017	52.7	2.7390	2.7914	52.4
20/5/09	21/5/09	2.7422	2.8188	76.6	2.7309	2.8078	76.9
21/5/09	22/5/09	2.7499	2.8166	66.7	2.7386	2.8108	72.2
25/5/09	26/5/09	2.7414	2.8302	88.8	2.7322	2.8250	92.8
26/5/09	27/5/09	2.7351	2.8407	105.6	2.7261	2.8355	109.4
27/5/09	28/5/09	2.7365	2.8087	72.2	2.7273	2.7994	72.1
28/5/09	29/5/09	2.7534	2.764	10.6	2.7490	2.7544	5.4
1/6/09	2/6/09	2.7626	2.8256	63	2.7580	2.8165	58.5
2/6/09	3/6/09	2.7571	2.8700	112.9	2.7523	2.8645	112.2
3/6/09	4/6/09	2.7605	2.8945	134	2.7562	2.8885	132.3
4/6/09	5/6/09	2.7668					0
8/6/09	9/6/09	2.7779	2.826	48.1	2.7721	2.8203	48.2
9/6/09	10/6/09	2.7444	2.8051	60.7	2.7387	2.8001	61.4

Table 2 – Estimated TSP particulate material mass at point 01

Date		*(Scale 1)*			*(Scale 2)*		
Start Date	**End Date**	**Initial (mg)**	**Final (mg)**	**Mass (µg)**	**Initial (mg)**	**Final (mg)**	**Mass (µg)**
18/5/09	19/5/09	2.7441	2.8347	90.6	2.7341	2.8204	86.3
19/5/09	20/5/09	2.7429	2.8349	92	2.7342	2.8252	91
20/5/09	21/5/09	2.7427	2.8908	148.1	2.7336	2.8808	147.2
21/5/09	22/5/09	2.7416	2.9186	177	2.7317	2.9069	175.2
25/5/09	26/5/09	2.7399	2.8740	134.1	2.7267	2.8688	142.1
26/5/09	27/5/09	2.7301	2.9010	170.9	2.7212	2.8938	172.6
27/5/09	28/5/09	2.7567	2.8850	128.3	2.7499	2.8765	126.6
28/5/09	29/5/09	2.7702	2.9153	145.1	2.7658	2.9069	141.1
1/6/09	2/6/09	2.718	2.8255	107.5	2.7122	2.8161	103.9
2/6/09	3/6/09	2.7582	2.9565	198.3	2.7545	2.9502	195.7
3/6/09	4/6/09	2.741	2.9747	233.7	2.7378	2.9689	231.1
4/6/09	5/6/09	2.7311	2.8195	88.4	2.7256	2.8134	87.8
8/6/09	9/6/09	2.7469	2.8295	82.6	2.7410	2.8249	83.9
9/6/09	10/6/09	2.7673	2.8620	94.7	2.7576	2.8584	100.8

Table 3 – Results for equality test (t test), for TSP between scale 1 and scale 2

	N	Mean	StDev	SE Mean
Scale 1	13	72.35	32.20	8.93
Scale 2	13	72.38	33.42	9.27
Difference	13	-0.023	3.090	0.857

- 95% CI for mean difference: (-1.891; 1.844)
- T-Test of mean difference = 0 (vs not = 0): T-Value = -0.03 P-Value = 0.979

Table 4 – Results for test of equality (t test), for PM_{10} between scale 1 and scale 2

	N	Mean	StDev	SE Mean
Scale 1	14	135.1	46.6	12.5
Scale 2	14	134.7	46.0	12.3
Difference	14	0.429	3.643	0.974

- 95% CI for mean difference: (-1.675; 2.532)
- T-Test of mean difference = 0 (vs not = 0): T-Value = 0.44 P-Value = 0.667

The test results showed the equality between the two groups (scale 01 and scale 02) with a confidence interval of 95 %. Therefore, from these results, the measures obtained from scale 02 came to be utilized to carry out data treatment because it is digital and modern.

In order to identify possible relationships between the parameters analyzed, it was first chosen possible to evaluate the data separately for each sampling station. This related the particulate material concentration results for the various particle sizes considering the different pollution sources from the station locations, since the two stations are located in areas that are affected by differently located mobile and fixed sources.

Particulate material concentration results for PM_{10} and TSP at point 01 and for TSP, PI_{10} and $PI_{2.5}$ at point 02 are presented on Tables 5 and 6.

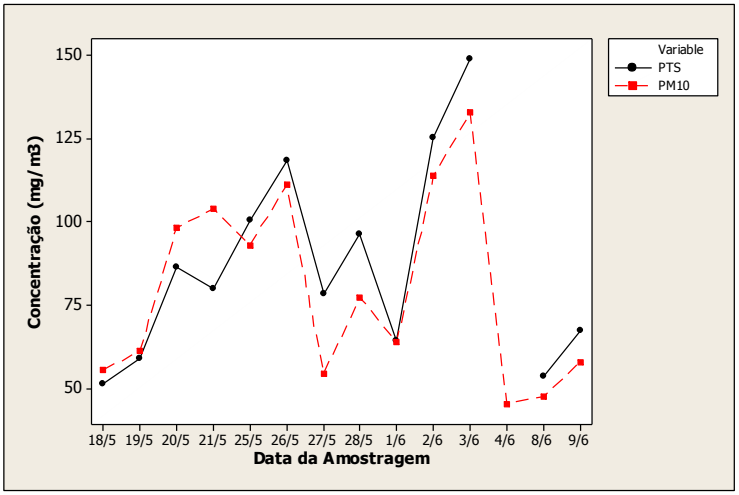
Table 5 – Results of PM_{10} and TSP concentrations. – Point 01

Date		*PM_{10}*		*TSP*	
Start Date	**End Date**	**Sample Volume (m^3)**	**Concentration ($\mu g/m^3$)**	**Sample Volume (m^3)**	**Concentration ($\mu g/m^3$)**
18/5/09	19/5/09	917.22	51.35	1553.48	55.55
19/5/09	20/5/09	890.97	58.81	1483.67	61.33
20/5/09	21/5/09	889.98	86.41	1496.02	98.39
21/5/09	22/5/09	904.45	79.83	1685.38	103.95
25/5/09	26/5/09	922.61	100.58	1529	92.94
26/5/09	27/5/09	923.48	118.46	1551.3	111.26
27/5/09	28/5/09	917.61	78.57	2328.23	54.38
28/5/09	29/5/09	56.08	96.29	1822.28	77.43
1/6/09	2/6/09	907.69	64.45	1628.51	63.80
2/6/09	3/6/09	896.94	125.09	1720.56	113.74
3/6/09	4/6/09	889.16	148.79	1738.12	132.96
4/6/09	5/6/09			1935.35	45.37
8/6/09	9/6/09	900.39	53.53	1763.82	47.57
9/6/09	10/6/09	910.61	67.43	1742.55	57.85

Table 6 - Results of $PM_{2.5}$, PM_{10} and TSP concentrations – Point 02

Data		*TSP*		*PI_{10}*		*$PI_{2.5}$*	
Start Date	**End Date**	**Sample Volume (m^3)**	**Concentration ($\mu g/m^3$)**	**Sample Volume (m^3)**	**Concentration ($\mu g/m^3$)**	**Sample Volume (m^3)**	**Concentration ($\mu g/m^3$)**
18/5/2009	19/5/2009	2319.26	53.51	21.59	20.84	2.40	9.26
19/5/09	20/5/09	2005.75	68.95	21.39	21.03	2.38	9.35
20/5/09	21/5/09	1977.87	69.27	21.03	25.67	2.34	19.02
21/5/09	22/5/09	2047.79	73.59	21.59	33.35	2.40	23.16
25/5/09	26/5/09	442.07	104.96	21.59	50.02	2.40	13.90
26/5/09	27/5/09	2086.24	159.81	21.59	54.19	2.40	13.90
27/5/09	28/5/09	2048.89	73.26	20.01	35.97	2.23	10.00
28/5/09	29/5/09	2170.39	81.41	21.47	37.72	2.39	13.97
1/6/09	2/6/09	2215.87	65.03	21.59	33.35	2.40	9.26
2/6/09	3/6/09	2253.39	92.04	21.60	58.33	2.40	55.56
3/6/09	4/6/09	2218.55	92.63	21.53	50.15	2.40	18.58
4/6/09	5/6/09	2217.09	53.99	21.59	16.67	2.40	4.63
8/6/09	9/6/09	2224.82	49.44	21.59	29.17	2.41	13.90
9/6/09	10/6/09	2256.12	52.08	21.59		2.40	4.63

Graph 1 shows PM_{10} and TSP particulate material concentration results at point 01 and Table 7 shows the test for equality between the groups referring to TSP and PM_{10}, as well as correlation test between the groups.



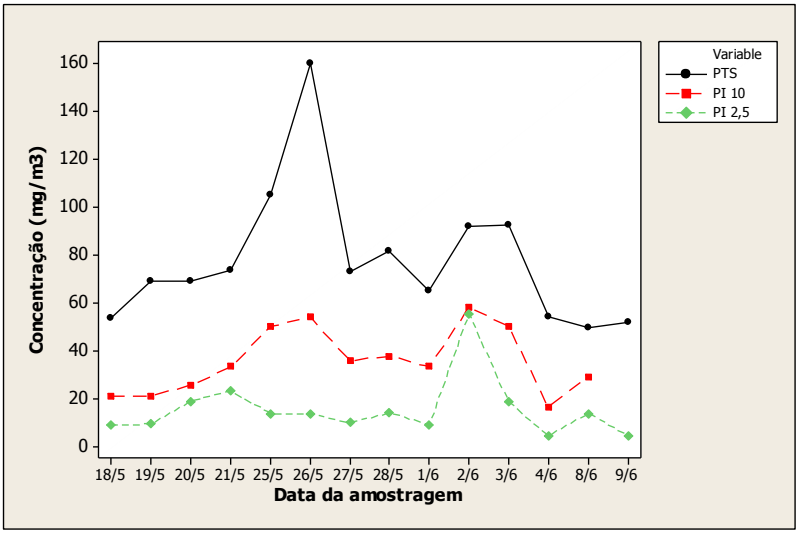
Concentration versus Sample Date

Graph 1 – PM_{10} and TSP particulate material results at Point 01

Table 7 – Results for equality test (t test) and correlation between sample groups regarding TSP and PM_{10} at Point 01

T Test				
	N	Mean	StDev	SE Mean
PM10	13	86.89	29.90	8.29
TSP	13	82.40	27.90	7.74
Difference	13	4.50	13.05	3.62
95% CI for mean difference: (-3.39; 12.38)				
T-Test of mean difference = 0 (vs not = 0): T-Value = 1.24 P-Value = 0.238				
Pearson correlation = 0.900				
P-Value = 0.000				

Graph 2 shows particulate material concentration for TSP, PI_{10} and $PI_{2.5}$ at point 02 and Table 8 shows correlation test results between the two groups.



Graph 2 – Results for concentration of particulate material for TSP, PI_{10}, and $PI_{2.5}$ at Point 02

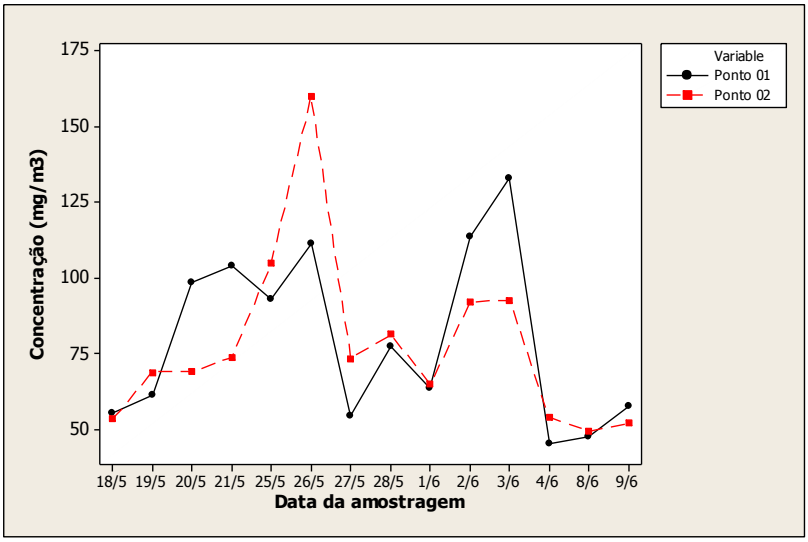
Table 7 - Results for correlation test between the group of samples, regarding TSP, PI_{10}, and $PI_{2.5}$ at Point 02

TSP	PI 10	
PI_{10}	0.778	
	0.002	
$PI_{2.5}$	0.267	0.602
	0.357	0.029
Cell Contents: Pearson correlation		
P-Value		

The results showed that particulate material concentration result values for PM_{10} and TSP at station 01 can be considered equal, that is, there was no significant variation between the concentrations of particles smaller than 10 µm and total particles in suspension.

At point 02, a large difference between the concentrations of different particle sizes could be observed. It was also possible to estimate the correlation that exists between them.

In order to determine the relationship between sampling stations, equality tests were carried out utilizing the Kruskal-Wallis method for non-parametric samples, considering the difference that exists between the sample points. TSP results from stations 01 and 02 can be seen on Graph 3 and the Kruskal-Wallis test on Table 8.



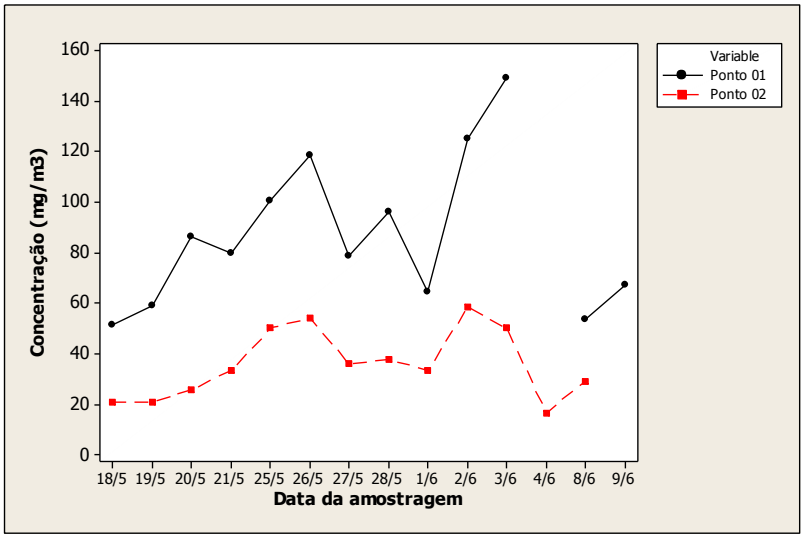
Graph 3 – TSP particulate material results at points 01 and 02

Table 8 - Kruskal-Wallis Test

Kruskal-Wallis Test				
C42	N	Median	Ave Rank	Z
1	14	70.62	14.9	0.28
2	14	71.27	14.1	-0.28
Overall	28		14.5	
H = 0.08 DF = 1 P = 0.783				

The test shows equality between the groups with a confidence level of 95%.

Graph 4 and Table 9 show PM_{10} results obtained for both sampling stations and the Kruskal-Wallis test applied to the equality verification between the groups.



Graph 4 – PM_{10} particulate material results at points 01 and 02

Tabela 8 - Kruskal-Wallis Test

26 cases were used				
2 cases contained missing values				
Kruskal-Wallis Test				
C42	N	Median	Ave Rank	Z
1	13	79.83	19.7	4.13
2	13	33.35	7.3	-4.13
Overall	26		13.5	
H = 17.04 DF = 1 P = 0.000				
H = 17.05 DF = 1 P = 0.000 (adjusted for ties)				

The test showed that the groups weren't equal, but had similar behavior, showing that there is a correlation between the sample stations. Correlation test results are shown on Table 9 below.

Table 9 - Correlations: PM10 (Point 01 and Point 02)

Pearson correlation of C31 and C44 = 0.865
P-Value = 0.000

Below on Tables 10 and 11, ^{238}U concentration results are shown for all of the samples at points 01 and 02.

Table 10 - ^{238}U Concentration – Point 01

Date		*PM_{10}*		*TSP*	
Start Date	**End Date**	**Particulate Concentration (µg/m^3)**	**U-238 Concentration (mBq/m^3)**	**Particulate Concentration (µg/m^3)**	**U-238 Concentration (mBq/m^3)**
18/5/09	19/5/09	51.35	0.01 ± 0.01	55.55	
19/5/09	20/5/09	58.81	0.03 ± 0.01	61.33	0.04 ± 0.01
20/5/09	21/5/09	86.41	0.04 ± 0.01	98.39	0.02 ± 0.01
21/5/09	22/5/09	79.83	0.01 ± 0.01	103.95	0.01 ± 0.01
25/5/09	26/5/09	100.58	0.02 ± 0.01	92.94	0.00 ± 0.01
26/5/09	27/5/09	118.46	0.03 ± 0.01	111.26	0.01 ± 0.01
27/5/09	28/5/09	78.57	0.01 ± 0.01	54.38	0.01 ± 0.01
28/5/09	29/5/09	96.29	0.5 ± 0.1	77.43	0.02 ± 0.01
1/6/09	2/6/09	64.45	0.01 ± 0.01	63.80	0.02 ± 0.01
2/6/09	3/6/09	125.09	0.02 ± 0.01	113.74	0.03 ± 0.01
3/6/09	4/6/09	148.79	0.03 ± 0.01	132.96	0.01 ± 0.01
4/6/09	5/6/09			45.37	0.01 ± 0.01
8/6/09	9/6/09	53.53	0.04 ± 0.01	47.57	0.01 ± 0.01
9/6/09	10/6/09	67.43	0.02 ± 0.01	57.85	0.00 ± 0.01

Table 11 - ^{38}U Concentration – Point 02

Date		*TSP*		*PI_{10}*		*$PI_{2.5}$*	
Start Date	**End Date**	**Particulate Concentration (µg/m^3)**	**U-238 Concentration (mBq/m^3)**	**Particulate Concentration (µg/m^3)**	**U-238 Concentration (mBq/m^3)**	**Particulate Concentration (µg/m^3)**	**U-238 Concentration (mBq/m^3)**
18/5/2009	19/5/2009	53.51		20.84	0.00 ± 0.01	9.26	0.00 ± 0.01
19/5/09	20/5/09	68.95	0.01 ± 0.01	21.03	0.00 ± 0.01	9.35	0.00 ± 0.01
20/5/09	21/5/09	69.27	0.01 ± 0.01	25.67	0.00 ± 0.01	19.02	0.00 ± 0.01
21/5/09	22/5/09	73.59	0.01 ± 0.01	33.35	0.00 ± 0.01	23.16	0.00 ± 0.01
25/5/09	26/5/09	104.96	0.20 ± 0.03	50.02	0.00 ± 0.01	13.90	0.00 ± 0.01
26/5/09	27/5/09	159.81	0.02 ± 0.01	54.19	0.00 ± 0.01	13.90	0.00 ± 0.01
27/5/09	28/5/09	73.26	0.02 ± 0.01	35.97	0.00 ± 0.01	10.00	0.00 ± 0.01
28/5/09	29/5/09	81.41	0.01 ± 0.01	37.72	0.00 ± 0.01	13.97	0.00 ± 0.01
1/6/09	2/6/09	65.03	0.01 ± 0.01	33.35	0.00 ± 0.01	9.26	0.00 ± 0.01
2/6/09	3/6/09	92.04	0.01 ± 0.01	58.33	0.00 ± 0.01	55.56	0.00 ± 0.01
3/6/09	4/6/09	92.63	0.01 ± 0.01	50.15	0.00 ± 0.01	18.58	0.00 ± 0.01
4/6/09	5/6/09	53.99	0.02 ± 0.01	16.67	0.00 ± 0.01	4.63	0.00 ± 0.01
8/6/09	9/6/09	49.44	0.01 ± 0.01	29.17	0.00 ± 0.01	13.90	0.00 ± 0.01
9/6/09	10/6/09	52.08	0.01 ± 0.01		0.00 ± 0.01	4.63	0.00 ± 0.01

The results found for the TSP, PI_{10} and $PI_{2.5}$ filters in the two stations had values very close to the minimum detection limit, thus not being considered significant to the trend or correlation study level.

4. CONCLUSIONS

From the results found and the statistical analyses applied, it can be concluded that:

1- In the case studied, a strong correlation can be seen between the particulate material concentration in the different sampled groups for a specified location. This correlation is specific and exclusive and depends greatly on local characteristics.

2- For total particulate matter, TSP, there wasn't a significant difference between sample stations, thus showing that TSP measured was coarse and can't be characterized or estimated in a generalized way for particles smaller than 10 μm or 2.5 μm.

3- It was verified that the concentration of PM_{10} particulate matter downstream from CDTN is greater than it is upstream. This behavior can be explained by the fact that the downstream point is found in an open area that thus received particulate material from a variety of emission sources (for example: particles from soil erosion, industrial processes, etc.). The other station, despite being in the direction of the prevailing wind is found in an area surrounded by trees that act as a physical barrier.

4- In this study case, it was not possible to establish any correlation between concentrations of ^{238}U in the various filters analyzed due to the low levels found.

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