



Assessment of the Particulate Matter Behavior According to Synoptic Situations and Source Regions. Case of Study, Rainless Season in InSTEC

Avaliação do Comportamento de Material Particulado de Acordo com as Condições Sinóticas e Regiões de Origem. Estudo de Caso, Estação sem Chuva em INSTEC

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Abstract

The influence of synoptic situations and source regions on heavy metals and radioisotopes are evaluated. The studied area consists of a 5 km radius around the Higher Institute of Technologies and Applied Sciences (InSTEC), where concentrations of PM10 were measured with a high-flow sensor using quartz filters Staplex 20x25 cm Staplex® Type TFAQ810. Levels of heavy metals (Cu, Ni, Cd, Co, Zn, Hg and Pb) were evaluated using the Emission Spectrometry with Inductively Coupled Plasma (ICP-AES) on a VHR JY38 technique, while radioisotopes (⁷Be, ⁴⁰K, ²¹²Pb, ²¹⁴Pb and ²⁰⁸Ti) were measured in a low background gamma camera. Furthermore, analysis of meteorological factors was performed using the Lapinel classification of the Synoptic Situations Types more frequent in Cuba. Subsequently, a statistical analysis was performed. Concentrations of heavy metals and radioisotopes showed differences in the presence of synoptic situations, however, only the ⁷Be showed statistically significant differences in the presence of low situations and extratropical cyclones. Sources Regions of air masses were determined according to Gomez (2011) and was obtained that the PM10 presents statistically significant differences in concentrations when Zone III, located in the center of the North American continent, provides the air masses. Regarding the weekly differences only the ²¹⁴Pb showed variation with statistically significant, showing that the contributions are different in magnitude between weekdays and weekends.

Keywords: particulate matter; synoptic situations; source regions

Resumo

A influência das situações sinóticas e regiões de origem sobre os metais pesados e radioisótopos são avaliadas. A área de estudo consiste em um raio de 5 km em torno do Instituto Superior de Tecnologias e Ciências Aplicadas (em inglês INSTEC), onde as concentrações de PM10 foram medidas com um sensor de alta vazão usando filtros de quartzo Staplex 20x25 cm Staplex® Tipo TFAQ810. Os níveis de metais pesados (Cu, Ni, Cd, Co, Zn, Hg e Pb) foram avaliados usando a Espectrometria de Emissão com Plasma Acoplado por Indução (em inglês ICP-AES) utilizando a técnica VHR JY38, enquanto os radioisótopos (⁷Be, ⁴⁰K, ²¹²Pb, ²¹⁴Pb and ²⁰⁸Ti) foram medidos em uma câmara gama de baixo-fundo. Além disso, a análise dos fatores meteorológicos foi realizada utilizando a classificação Lapinel dos tipos de situações sinóticas mais frequentes em Cuba. Posteriormente, uma análise estatística foi realizada. As concentrações de metais pesados e de radioisótopos apresentaram diferenças em função das situações sinóticas, no entanto, somente o ⁷Be mostrou diferenças estatisticamente significante na presença de condições de baixa pressão e ciclones extratropicais. A Região de origem das massas de ar foram determinadas de acordo com Gomez (2011) e foi identificado que as concentrações de PM10 apresentam diferenças estatisticamente significante quando as massas de ar são provenientes da Zona III, localizada no centro do continente norte-americano. Com relação às diferenças semanais, apenas a concentração do ²¹⁴Pb apresentou variação com significância estatística, mostrando que as contribuições são diferentes em magnitude entre os dias úteis e fins de semana.

Palavras-chave: material particulado; situações sinóticas; regiões de origem

1 Introduction

The presence of particle matter in the air exceeding certain limits generates enormous concern to scientific community as well as the general population. Without any doubt, the meteorological factors have a remarkable influence on the pollutant concentrations in the atmosphere, because it can increase or reduce its dispersion. This is the reason why the evaluation of the relation between meteorological variables, synoptic and meso scale situations, with the pollutant behavior is becoming so important, Fernandez & Ternero (2004). This can be archived using air quality simulation models: Gómez & Cuesta (2011); Strong *et al.* (2010) or the Lapinel classification of Synoptic Situation Types Lapinel (1988), Cuesta (1995), Collazo (2011) and Cremata *et al.* (2014). This last one considers that the pollutants move along with the atmospheric circulation; hence, it groups the typical meteorological systems that have influence on this circulation, such as the Subtropical Anticyclones and cold fronts.

On the other hand, one of the more noxious air pollutants is the heavy metals. They are group together due to its density and potential harms to human health. The other kinds of pollutants that are highly controlled are the radioactive isotopes.

During the rainless season multiple cases of respiratory diseases appeared, mostly because meridian wind transport, which carries down several pollutant from the North American continent Gómez (2010); Marcelo & Garcia (2010). However, not all the synoptic situations are involved in this issue. An investigation that establishes the main

relations between atmospheric weather systems and pollutants' transport is the first step that contribute to an air quality model, focused in this aspect.

Therefore, the analysis of the influence of the synoptic situations in the heavy metals and radioactive isotopes concentrations give a valuable knowledge for the air quality protection and it is necessary because of the harmful consequences on the ecosystems and human's health due to these pollutants.

For all those reasons, the main objective of this work is to assess the influence of synoptic situations on the concentrations of heavy metals and radioactive isotopes, within the particle matter less than 10 μm collected in urban areas surrounded by vegetation during the rainless season. The Higher Institute of Applied Sciences and Technologies (InSTEC in Spanish) will be the case of study.

2 Methods and Data set

2.1 Sampling Location

The particles were collected in the Environmental Analysis Laboratory located in the InSTEC (23° 7' North Latitude and 82° 22' West Longitude). Given the multiple influences of sources and the most likely origin of the several pollutants a 5 Km radio area was set (Figure 1).

There are two seasons that dominate the area, rainy (May to October) and not rainy (November to April). The predominant wind direction is from East and East-Northeast all year long, there are exceptions in the presence of any meteorological system, such as cold fronts or extratropical lows (Figure 2).

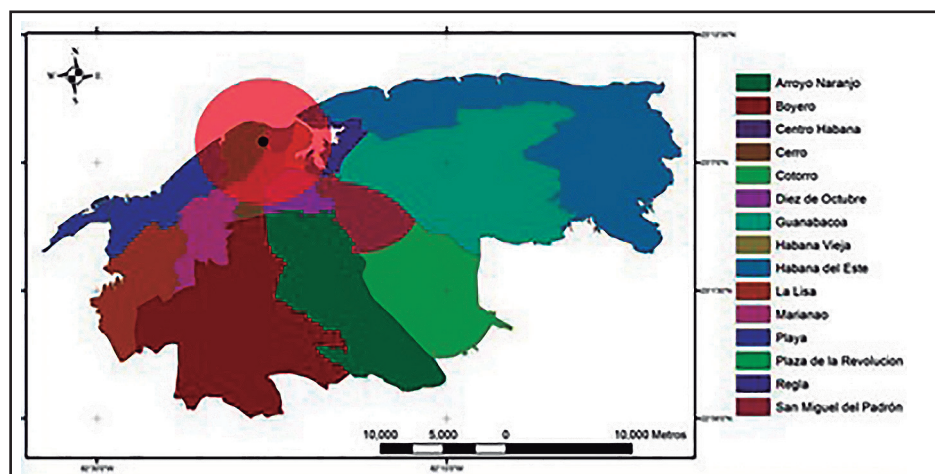


Figure 1 Area of study.

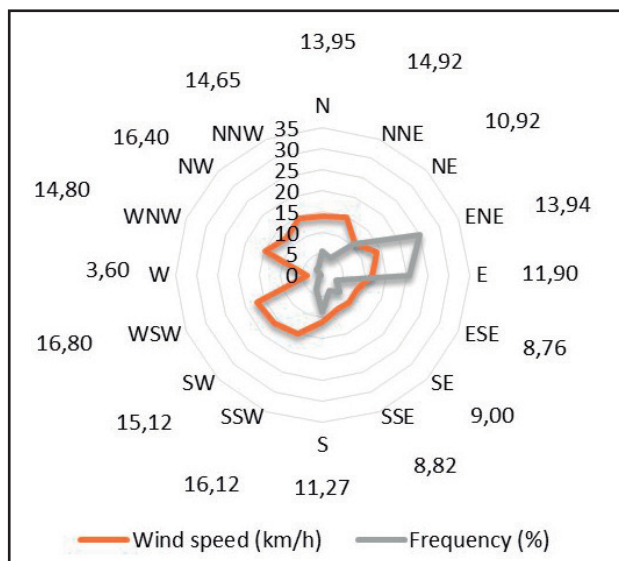


Figure 2 Wind rose from the sampling site.

The location receives also influences of different air masses along the year. In the rainless season the air masses are classified as polar, cold and dry air from the North American continent. On the other hand, in the rainy season the main source is tropical, warm and moist air from the Caribbean Sea and the Atlantic Ocean.

The Synoptic Situation Types classification from Lapinel (1988) takes into account these previous air masses. Given that atmospheric circulation is extremely complex and changing, similar circulations are group as types, with the intention of studying weather processes using a genetic criterion. This is the best classification of synoptic situations that have been made for Cuba, and it is widely used for applied meteorology studies.

2.2 Meteorological Data

As mentioned before, the Synoptic Situation Types (SST) classification from Lapinel (1988) was used, and hence completed for the 2009 year (Table 1). Because of the atmospheric circulation is very complex and changing, to the point where one can't never find the same, it is very useful this classification, for it groups similar systems into "types". The main criteria in this grouping are the similarities in the pressure and wind field, along with the source area, which can be oceanic or continental. The predominant synoptic situation during the sampling period was classified, analyzing the surface maps starting in the 12 UTC until the 12 UTC of the next day.

SST	SSS
I. Atlantic Anticyclone	1. Predominant wind direction from first quadrant
	2. Predominant wind direction from second quadrant
VI. Extratropical Cyclone	3. Near and direct influence from the low pressure center. Warm sector
	4. Near and direct influence from the low pressure center. Cold sector
	5. Previous area to the front. With squall line
	6. Previous area to the front. Without squall line
V. Convergence zones	7. Within the east flow
	8. Others
VIII. Migratory Anticyclones	9. On the continent
	10. On the East of United States, over the ocean and in rapid tropical transition
	11. In the Mexican Gulf or over Cuba
II. Widespread Anticyclone	12. Typical zones of the widespread anticyclone (without tropical wave)
	13. Typical zones of the widespread anticyclone (with divergent zones)
VII. Cold Front Systems	14. Classic cold front
	15. Wayward cold front
	16. Stationary cold front
	17. Dissipative cold front
III. Weak pressure gradients	18. Generally associated to tropical waves
IV. Cyclonic situations	19. Cyclonic depressions, cyclones and hurricanes
IX. Other situations	20. Not identifiable

Table 1 Synoptic Situations Types (SST) and Subtypes (SSS) classification by Lapinel (1988).

2.3 Backward Trajectory Model and Source

The source regions of several pollutants can be identified by the analysis of backward trajectories by HYSPLIT model. These are grouped by Gomez (2011) withdraw classification (Figure 3b). Some parameters, as the starting time, the total run time and the trajectory levels, where chosen according to the classification of source regions.

First, it was chosen Run HYSPLIT Trajectory Model. Afterwards, in the menu TrajectoryModel it was selected the Compute Archive Trajectories, due to the trajectories were from previous days. Then, it was necessary to enter the coordinates, 23.12°N and

depending on each other and equally distributed, are obtained from the two populations. On the contrary, t test for dependent samples consist typically in a sample of values with similar statistical units, or a group of units that have been evaluated in two different occasions. Considering all of this, a correlation analysis is proposed to accomplish first.

The criteria to accept or reject the null hypothesis that the two means are equal $H_0: \bar{x}_1 = \bar{x}_2$ is that $t < t_{tabulated}$.

The t tests for independent and dependent samples are summarized in table 2.

In equation 1 \bar{x}_1 and \bar{x}_2 are the groups mean values, n represents the amount of values, S_1^2 and S_2^2 are the variances, (1) Monday to Friday group and (2) weekends group.

In equation 2 \bar{x}_1 and \bar{x}_2 represent the mean concentrations from Monday to Friday (1) and weekends (2); n_1 and n_2 are the number of items in each group (1 and 2); S^2 is the variance, which it is calculated using the next formula:

$$S^2 = \frac{(n_1-1) \cdot S_1^2 + (n_2-1) \cdot S_2^2}{n_1 + n_2 - 2} \quad (3)$$

Where S_1^2 and S_2^2 denotes the variances in group 1 and 2 respectively.

The equation 4 is also known as t Welch and it is only used when one assumes the variances are different, however, the sample sizes may be equal or not. As a result, they should be separately estimated.

In the equation 5 the difference for all pares must be calculated. The mean is X_D and the standard deviation is S_D , while n represents the amount of values. In case one wants to prove the means are different then $\mu_0 \neq 0$, and $\mu_0 = 0$ if vice versa.

2.4.2 Relation Between Synoptic Situations Types and Particulate Matter Concentrations

The variance analysis (ANOVA) was conducted in order to establish if the observed differences between two means were random or due to synoptic situations or source regions.

Null hypothesis: The Synoptic Situations Types / Source Regions do not have a significant

Case of study	Formula
Equal sample size and equal variances	$t = \frac{\bar{x}_1 - \bar{x}_2}{\sqrt{\frac{2}{n} \times \sqrt{\frac{1}{2}(S_1^2 + S_2^2)}}}$ (Equation 1)
Different sample sizes and equal variances	$t = \frac{ \bar{x}_1 - \bar{x}_2 }{\sqrt{S^2 \cdot \left(\frac{1}{n_1} + \frac{1}{n_2}\right)}}$ (Equation 2)
Unequal sample sizes and unequal variances	$t = \frac{ \bar{x}_1 - \bar{x}_2 }{\sqrt{\frac{S_1^2}{n_1} + \frac{S_2^2}{n_2}}}$ (Equation 4)
The t test for dependent samples	$t = \frac{X_D - \mu_0}{S_D / \sqrt{n}}$ (Equation 5)

Table 2 Formulas for the t test.

influence on the concentrations of the particulate matter elements $H_0: \bar{X}_i = \bar{X}_j \dots = \bar{X}_k$.

Alternative hypothesis: The Synoptic Situations Types/Source Regions do have a significant influence on the concentrations of the particle matter elements $H_1: A \text{ different } \bar{X}_i \text{ was found.}$

The statistic number F_0 represents the quotient between the series variance and the self-series variance. If $F_0 > F_{1-\alpha}^{(k-1; n-k)}$ the null hypothesis is rejected and the sample variances are due to synoptic characteristics with a confident level of 95 % (hence $1 - \alpha = 0.05$). The steps for this analysis are summarized in the next table. Firstly, one should calculate the variance between groups and inside groups, each one with the freedom degree. Afterwards, one have to determine the mean square, for the variance between groups and inside groups. To conclude, one obtain F_0 and compare it with $F_{1-\alpha}^{(k-1; n-k)}$. This last one is a tabulated number available on the Fisher table.

2.5 Summary of the Sampling and Chemical Techniques

The particulate matter was collected with a Staplexhigh flux impactor, using quartz filters of 20x25 cm Staplex® Type TFAQ810 to collect the PM10 fraction. This impactor was set on the roof of the InSTEC, at a 12 m height from the ground

Source of variance	Square sum	Freedom degree	Mean square	Statistic number
Between groups (SC_E)	$\sum_{i=1}^k n_j(\bar{y}_i - \bar{y})^2$	$k - 1$	$\frac{SC_E}{k - 1}$	$F_0 = \frac{SC_E^2}{SC_D^2}$
Inside groups (SC_D)	$\sum_{i=1}^k \sum_{j=1}^{n_i} n_j(\bar{y}_{ij} - \bar{y}_j)^2$	$n - k$	$\frac{SC_D}{n - k}$	
Total	$\sum_{i=1}^k \sum_{j=1}^{n_i} (\bar{y}_{ij} - \bar{y})^2$	$n - 1$		

Table 3 The formulas of the variance analysis.

and 1.5 m from the base of the equipment to the sampling bell. The area chosen for the sampling is a little bit unpopulated and it is surrounded by tropical vegetation.

The sampling schedule can be observed in the Figure 4. A total amount of fourteen samples were collected.

The filters were dried at 400°C prior the sampling, during 16 hours, put into a drier for 24

hours and were weighed. The filters were put in the Staplex PM10 impactor, from United States, over 48 hours, with a constant flux of 1.13 m³/min. after the sampling period, the filter was put into the drier for 24 hours and the weighted. The PM10 mass deposited was obtained by subtracting the masses before and after the sampling.

The particle matter concentrations were calculated by dividing the deposited mass to the quotient of the next factors: sampling flux (1.13 m³/

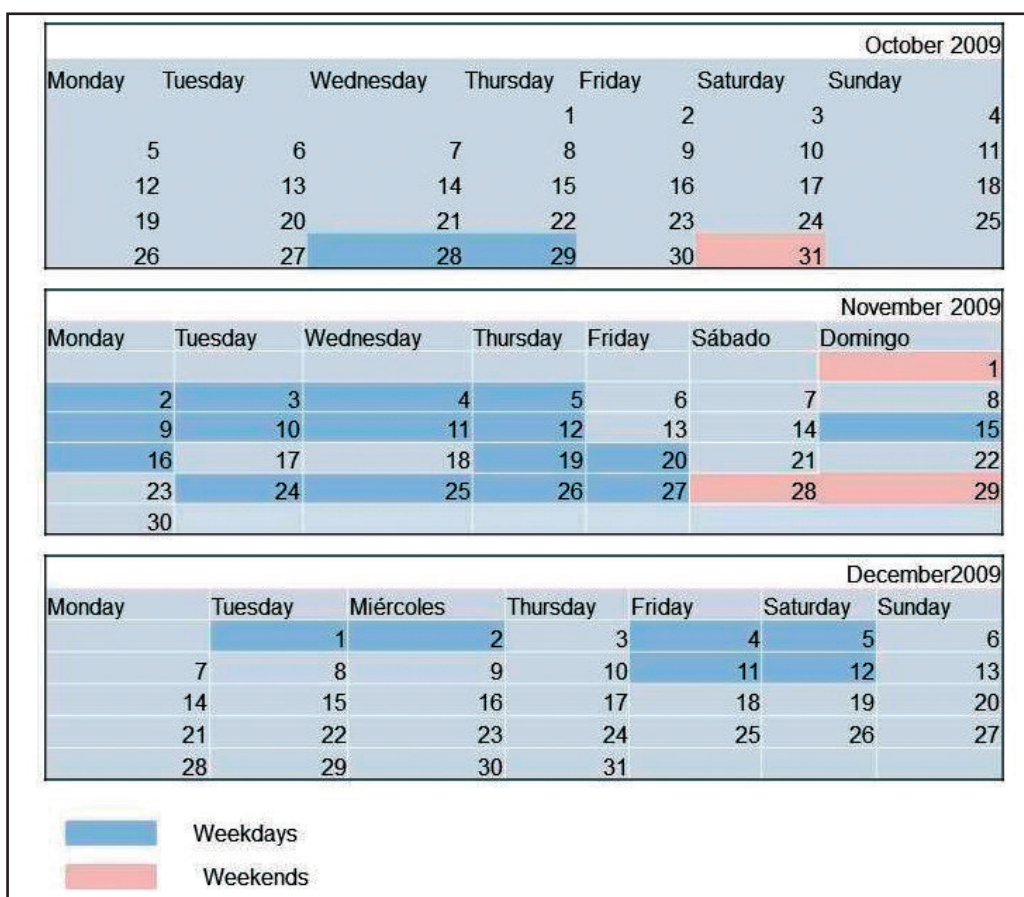


Figure 4 Sampling days highlighted in blue (weekdays) and orange (weekends).

min), period of sampling (48 hours) and minutes within an hour (60).

Subsequently, the white parts of the filters (portions where no particle matter was deposited) were cut. Next, these filters were put into the drier for 24 hours and the weighted and cut in four equally pieces, each one for individual determination of heavy metals, radioisotopes, organic compounds and mercury. These pieces were kept into the drier for further analysis.

The chemical techniques employed in the mercury assessment were a determination system by atomic absorption spectrophotometry and generation of cold vapor flow injection in the Environmental Analysis Laboratory.

The determination of heavy metals Cu, Ni, Cd, Co, Zn, Hg and Pb in the resulting solutions was performed using digestion technique Emission Spectrometry with Inductively Coupled Plasma (ICP-AES) on a JY38 VHR. Calibration solutions with Cu, Cd, Zn, Pb, Co and Ni elements were prepared from the concentration patterns Merck 1000 mg / L.

For determination of the environmental interest radionuclides (^7Be , ^{40}K , ^{212}Pb , ^{214}Pb and ^{208}Tl) present in the filters it is not necessary pretreatment of the samples. The filters are placed in the capsule holder of the equipment and are measured for 16 h in a low background Gamma camera. Hyperpure Germanium detector was used for measurements.

3 Results and Discussion

3.1 Synoptic Situation

Types (SST) and Subtypes (SSS)

The period of time analyzed in this study covers random days of three months in the rainless season of the 2009. The SST classification was completed for those specific days and it was calculated the frequency for each month. However, this number should not be taken as representative unless a further statistical analysis of this matter support that the amount of samples is enough to describe the normal behavior of the PM₁₀, heavy metals and radioisotopes studied, along with the synoptic situations.

A first approach for the behavior of the synoptic situations is to compare the one obtained in this investigation with another one that has analyzed it for a larger amount of time, close to thirty years. This case of Marcelo & Garcia (2011) analyzed the synoptic situations for the Western Region from 1981 to 2008. The results are explain below.

According to Marcelo & Garcia (2011) the typical behavior of the synoptic situations in October is the presence of the migratory anticyclone (SST VIII, subtypes 9 and 10), and less frequent, the weak pressure gradients (SST III, subtype 18) and the widespread anticyclone without tropical waves (SST II, subtype 12). This is an expected scenario because October is the last month of the rainy season and therefore a transition occur from the tropical systems to the extratropical ones. On the other hand, in November and December, the synoptic situations that prevail are only the migratory anticyclone (SST VIII, subtypes 9 and 10).

In the month of October two types were presented: the Synoptic Situation Types named extratropical low and migratory anticyclones. The first one is not as usual at this month, but it's possible that extratropical lows formed in the United States or the Gulf of Mexico have an effect over the western region of Cuba.

Furthermore, in November a predominance of migratory anticyclones and frontal systems was observed, and in December the influence of this first is reduced, showing that a slight disagreement with the typical behavior of those (Figure 5).

3.2 The Element Concentration

Behavior Regarding Synoptic Situations Types and Subtypes

Before performing the statistical analysis, a description of the maximum and minimum concentration according to the synoptic situation was made. This gives a panoramically background, conversely, it can agree or not with the subsequent results, because the differences may not be due to the synoptic situations. It is necessary to remember that the only factor analyzed was the synoptic situations.

The average maximum concentrations of PM₁₀ (Figure 6) occurred with cyclonic situations

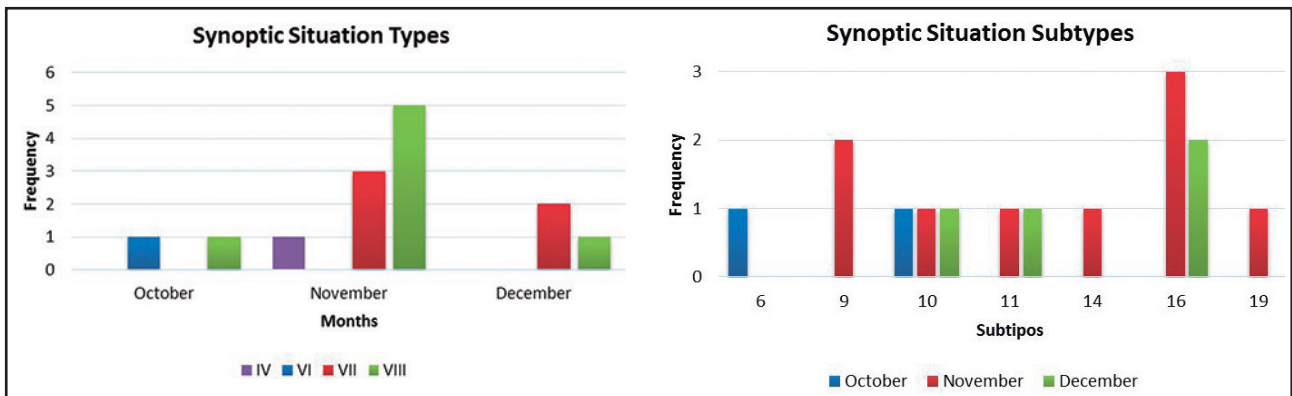


Figure 5 Frequency of the Synoptic Situation Types and Subtypes in the studied months

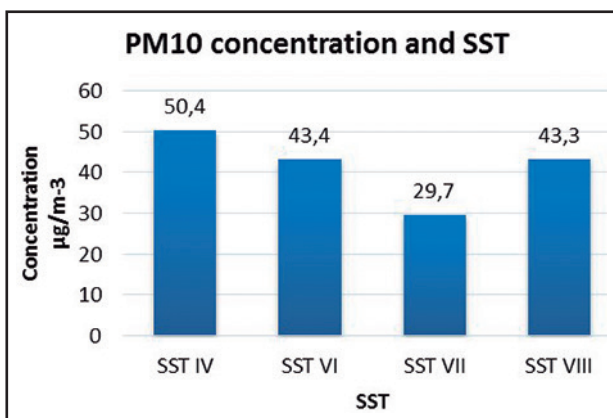


Figure 6 Average maximum PM10 concentrations with the different SST.

(SST IV) and migratory anticyclones (SST VIII), however concentrations were lower with the front systems (SST VII).

The elements of heavy metals more numerous were copper (Cu), zinc (Zn) and lead (Pb). Conversely, heavy metals behave a little bit different, because the highest concentrations of Zn, Pb and cadmium (Cd) were with the front systems (SST VII) and migratory anticyclones (SST VIII). The minimum concentrations with cyclonic situations (SST IV). Nickel (Ni) reached a higher level with the SST VII and the concentration decreased to migratory anticyclones.

The concentrations of cobalt (Co) and mercury (Hg) behaved similarly for all synoptic situations. Finally, the maximum concentration of copper was with extratropical low (SST VI) and the minimum, with the presence of cyclonic situations (SST IV). All was plotted in Figure 7.

On the other hand, the main causes for the presence of radionuclides in PM10 in InSTEC Manduca *et al.* (2011) are:

- The direct product of radioactive nuclear tests in the 60s of last century, releases by nuclear power

plants and the Chernobyl accident, especially for artificial radionuclides and long-lived precipitation.

- Land dust suspension and its associated radionuclides.
- The natural radioactivity (cosmogonic).
- Radionuclides in fossil fuels used for energy (mainly cars and industrial activities).

The values determined on the filters in the low background gamma camera are relatively low (figure 8). Nonetheless, it stands out a maximum concentration of ⁷Be with low extratropical synoptic situation.

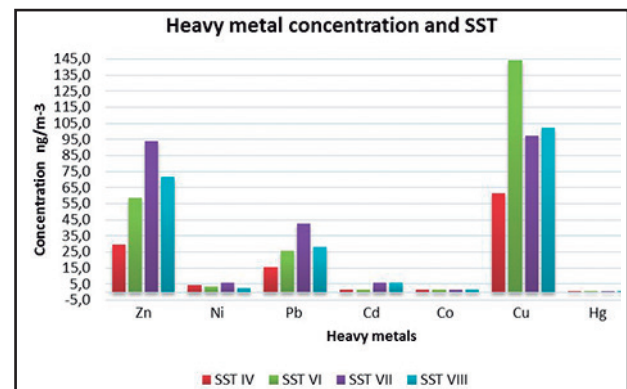


Figure 7 Heavy metals concentration.

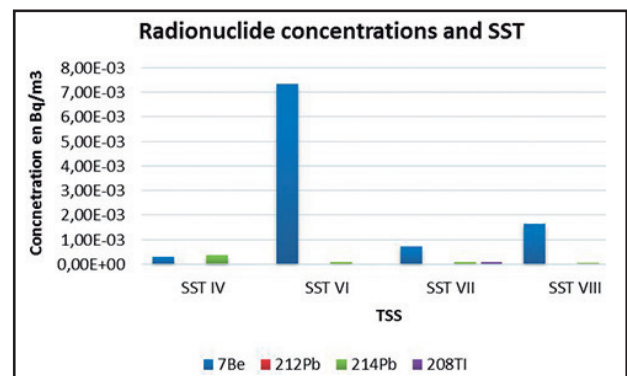


Figure 8 Radionuclide concentrations.

3.3 ANOVA Results for Synoptic Situation Types

Statistical analysis showed that variations in heavy metals sample concentrations and radionuclides are not attributable to the synoptic situations, except for ⁷Be. The *F* statistic represents the ratio of the variance between series and the variance within the series. If $F_{1-\alpha}^{(k-1;m-k)} < 3,708$ then the null hypothesis is accepted and it is considered that the variations in the samples are not due to synoptic features with a confidence level of 95%.

The null hypothesis is overruled for cobalt and beryllium 7 as they are greater than $F_{1-\alpha}^{(k-1;m-k)}$. Regardless this, for cobalt the measurements were at very close range from the detection limit of the instrument and concentrations are almost equals, therefore the variance between groups is zero and the result is not valid. For beryllium 7, $F_0 = 18,010$ and $F_{1-\alpha}^{(k-1;m-k)} = 3,708$.

In order to determine which one or which ones of these situations are the statistically different for the ⁷Be analysis is necessary to use a confidence interval for the mean, with this, the situations which means are statistically different were found. They are those who do not find themselves within the abovementioned interval. In the following table the situations indicated in red color are those in which the means of the ⁷Be concentrations are significantly different.

SST	Mean (Bq/m ³)
IV	0,00029
VI	0,00736
VII	0,00073
VIII	0,00167
Confidence interval	0,00062 < \bar{x} < 0,00253

Table 4 Synoptic Situations Types were the means are statistically different.

The concentrations of ⁷Be are different statistically in the presence of cyclonic situations (SST IV) and extra-tropical lows (SST VI).

3.4 Source Regions

The source regions of air masses were classified according to Gómez (2010) using the HYSPLIT backward trajectories, and its frequency

for each level (500 m, 1500 m y 3000 m) was calculated. The 500 m level is representative for the Planet Boundary Layer (PBL), which is where the atmospheric turbulence mixed up all the contaminants. The 1500 m level is considered the average height of the PBL, which is where the exchange between the upper and lower levels of the atmosphere is observed. Finally, the 3000 m level is where the pollutants move freely, that is, without the influence of the surface roughness.

Due to Cuba's geographic position, in the south-west of the Subtropical Anticyclone, the Zone I was the source region most frequent (Figure 9), as the trade winds transport most of the pollutants. For more information, see Figure 2.

The source regions corresponding to Zone II, III and IV represent the northerly flux, which is predominant in the not rainy season. However, that does not exclude the fact that the trade winds influence was bigger in the studied period.

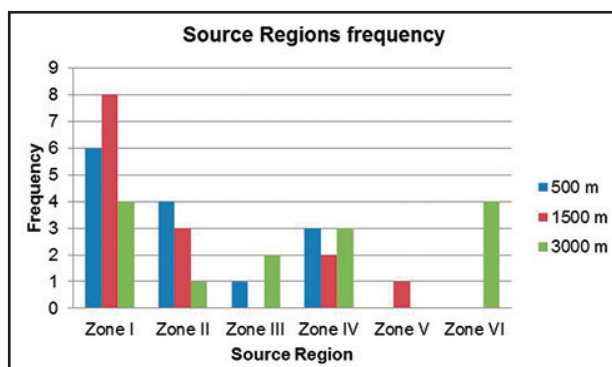


Figure 9 Source regions frequency in the studied period.

3.5 ANOVA Results for Source Regions

Taking into account that the 500 m level is the closest to the surface therefore the air masses that included the measured concentration of the pollutants were those in this level. In this level the Zone V and VI were not present. The statistical analysis showed that the variation of heavy metals and radionuclide concentration does not depend on the source region of the air mass, except in the PM10 concentrations, where $F_0 = 8,830$. The $F_{1-\alpha}^{(k-1;m-k)} = 3,708$.

As well as in the synoptic situation analysis if $F_{1-\alpha}^{(k-1;m-k)} < 3,708$ the null hypothesis is accepted

and it is considered that the variations in the samples are not due to the source regions with a confidence level of 95%.

Following the same path, the confidence level for the mean was estimated and the concentration means which were not within this interval were the statistical different.

As the Table 6 indicates the concentrations of PM10 are statistically different when the Zone III, corresponding to the center or the North American continent, provides the air masses.

3.6 Variation of the Samples, Results of t-Student

First, the relationship between samples from weekends (Saturday and Sunday) and weekdays (Monday to Friday) was determined. The differences are significant for a confidence level of 95%, in some elements.

The values of the Pearson correlation coefficient showed that the strongest relationships between weekly samples and the weekend are in the PM10, Hg and ²¹²Pb (0.81; 0.86; -0.87 respectively). Subsequently, they were analyzed as dependent samples.

The following step, the t-Student test, was performed according to the explanation given at the statistical methods section.

For all elements, except ²¹⁴Pb where $t = 0.75$ and $p = 0.46$, the null hypothesis was accepted, then concluding that the samples weekly and weekend of this radionuclide have not statistical differences.

In the Figure 10 it shows that none of the maximum concentrations was recorded on weekends, marked with a red dot. Peak concentrations occurred during the week and with the presence of a stationary cold front.

4 Conclusions and recommendations

Concentrations of PM10, heavy metals and radioisotopes showed differences in the presence of synoptic situations, as the average maximums were different for each element. Nevertheless, only in the case of the ⁷Be variations were statistically significant for the 95 % confidence level. It was established

Source regions	Mean (µg/m³)
I	36,467
II	34,225
III	70,8
IV	43,8
Confidence interval	$34,158 < \bar{X} < 45,541$

Table 5 Confidence intervals for source regions.

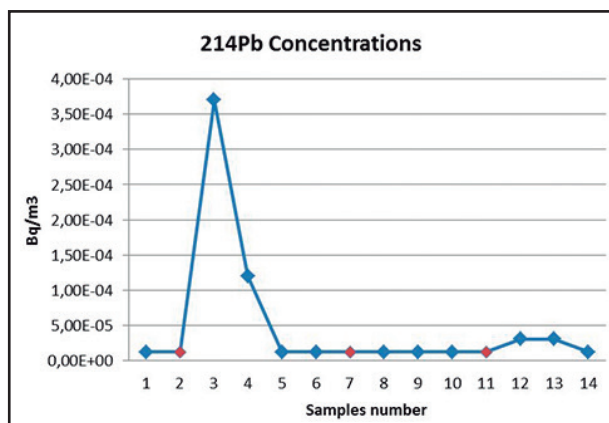


Figure 10 Graphic of the ²¹⁴Pb concentrations.

that the cyclonic situations and the extra-tropical lows were the synoptic situations that influenced the concentrations during the sampling period.

What's more, the concentrations of PM10 showed statistical differences according to the air masses source regions, specifically the Zone III, placed in the center of the North American continent. This zone characterizes for agricultural activities and for having big supplies of coal and steel metal. This showed that the synoptic situations and the air masses source regions are not the only factors regarding the concentration behavior of the studied elements, and therefore it is recommended to investigate the other sources of variation.

Regarding weekly variation, the statistically significant differences for was only obtained with ²¹⁴Pb, showing that the contributions are different in terms of magnitude between weekdays and weekends.

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