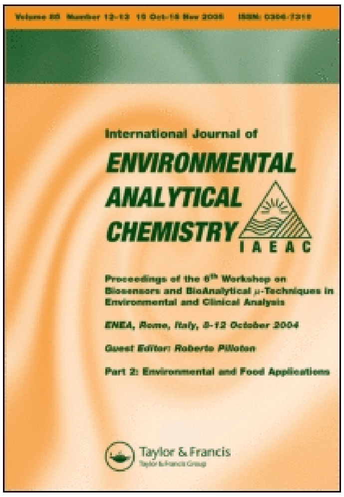

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A Study of the Atmospheric Lead Pollution in Seville, Spain. Influence of Meteorology and Traffic, and Relationship with other Traffic-Generated Pollutants

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A study of the atmospheric pollution by suspended particulate lead in the city of Seville, Spain, was carried out during the period between March 1983 and February 1984. The results obtained from seven sampling stations allowed us to study the site to site variations; there are two locations with average annual concentrations higher than $2\,\mu g \cdot m^{-3}$. In one station, the meteorological and traffic effects and relationship with other traffic-related pollutants, were investigated. The regression analyses performed on the average monthly data have shown an inverse correlation with wind speed and temperature and a direct correlation with carbon monoxide and smoke concentrations, but no significant correlation was found with the lead deposition levels.

KEY WORDS: Atmospheric lead pollution, seasonal pollution, traffic pollution, carbon monoxide pollution, smoke pollution.

INTRODUCTION

The atmospheric lead particulate concentration has increased significantly during the last decade, due, fundamentally, to the generalized usage of lead alkyls as anti-knock additives in petrol. Due to this fact, several countries have adopted a series of measures which tend to reduce the content of lead in petrol, however in Spain, these measures have not yet been adopted.

The city of Seville, with a population of nearly one million inhabitants, is the most important urban nucleus in the southern region of Spain. The number of vehicles has increased nearly 20% during the last five years and the maximum traffic flows are registered in the circumvallation roads of the old part of the town, with an average value of 60,000 to 70,000 vehicles per day. The global growth of the urban traffic is estimated as 30% during the last five years. On the other hand, its climatic characteristics (minimum high temperatures and low wind speeds) aids the appearance of thermic inversion phenomena which obstruct the dispersion of the pollutants. Also, the pollutants effects of the vehicles are accentuated by the existence of a great number of narrow circulatory roads, above all in the old part of the city.

Due to the facts mentioned and to the increasing environmental degradation, an extensive research programme has been carried out, for several years, in this Department in order to evaluate the actual state of the atmospheric pollution of the city of Seville. Two papers on the pollution by setteable particulates were reported earlier.[1,2]

In the present paper, it has been intended:

a) To carry out a comparative study of the lead levels in different parts of the city of Seville, in order to learn the actual entity of the problem, the most affected zones and the conditioning factors.

b) To determine the influence of several factors, especially those of meteorology and traffic. According to the bibliography, both factors are fundamental in the levels of the pollutants associated with the traffic of vehicles, apart from other additional factors (topography, distance between the sampling point and the circulatory roads, etc.).

c) To establish the possible relationship between the atmospheric lead concentrations and the concentrations of other pollutants generated by the traffic of vehicles, fundamentally carbon monoxide, smoke, and setteable particulate lead.

EXPERIMENTAL

Sampling

The period of sampling is between March 1983 and February 1984.

In order to carry out a comparative study of the lead levels in the city of Seville, seven sampling points were chosen: one in the old part of the city (Plaza Nueva, point 1), three distributed along the circumvallation roads round the mentioned old part of the city within the limits of the Western Section (Marqués de Paradas, point 2), Northern Section (Resolana, point 3) and East Section (Recaredo, point 4), two in important radial roads in densely populated sections (Eastern Section, Luís Montoto, point 5 and Southeastern Section, República Argentina, point 6) and the final one in a residential zone in the Southern Section. Its localization is shown in Figure 1.

The sampling units were situated on terraces at heights of 3–5 meters above ground level and near the circulatory roads.

The sampling of Total Suspended Particulates (TSP) were collected with high volume samplers, Sierra 305–2,000, on 20 × 25 cm

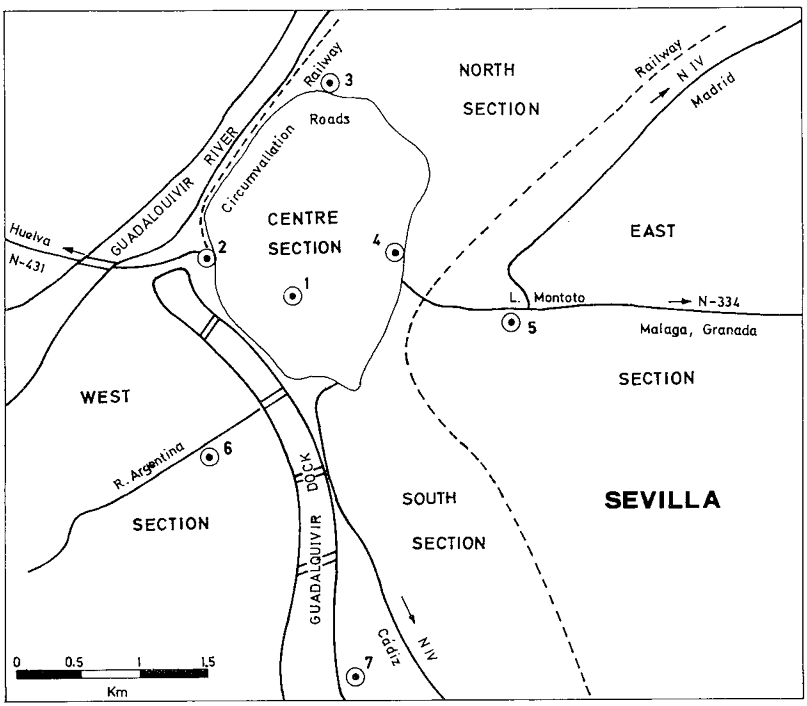

FIGURE 1 Location of sampling stations.

filters Whatman 41, and with sampling periods of 24 hours. The air volumes sampled were of 800–900 m^3.

From each of the mentioned sampling points, 12 samples, homogeneously distributed within a year, were collected.

In order to study the meteorological and traffic effects, and the relationship with other pollutants, the sampling station No. 5 (Luís Montoto) was chosen for its ideal characteristics and for being situated in a representative section of the city.

To establish the metereological influence and the relationship with other pollutants, a total of 72 daily samples of TSP were collected, homogeneously distributed within the year, with a similar method and device to that previously described. Simultaneously, smoke, carbon monoxide and setteable particulates were collected. The daily smoke samples were collected with an apparatus for sulphur dioxide and smoke, Tec-Lab SP8, on 5 cm filters Whatman 1. The carbon monoxide samples were taken and analyzed continually by means of an Aoriba 8310 analyzer, equipped with a data treatment unit, Monitor Lab. 9300 and a calibration unit. The consecutive data given by the analyzer were averaged in order to find the daily average. Finally, the setteable particulates samples were collected monthly with a collecting equipment designed according to the specifications of the British Standard Method.

To establish the traffic influence, the TSP samples were collected with a device similar to that previously described, but with sampling periods of 3 hours. A total of 48 samples were collected, homogeneously distributed during the day and in different months of the year.

Analysis

The filters were weighed before and after sampling to determine total suspended particulate matter. Then, the filters were wet ashed with a mixture of nitric and perchloric acids 1:1. After the filter was completely ashed, the solution was boiled down until fumes of perchloric acid appeared, and diluted with distilled water, in a volumetric flask, to a convenient volume. The lead concentration was determined by Atomic Absorption Spectrometry using a Perkin-Elmer 2280 spectrophotometer with an air–acetylene flame at 283.3 nm.

For the calibration, the standard solutions containing lead underwent a similar procedure to that followed by the samples. A blank solution was prepared by treating an empty filter in the same way as the samples and the standards.

The smoke concentrations were determined reflectometrically, according to the British Standard Sampler (BSS) method.

The carbon monoxide concentrations were determined as it has previously been indicated, in an automatic analyzer.

For the determination of the setteable lead concentrations, an aliquot of the samples, which also contained the rain-water, was evaporated to dryness at 110°C. The residue, once weighed, was digested with a mixture of nitric and perchloric acids in the same way as for the TSP samples. The lead concentration was determined by Atomic Absorption Spectrometry at 283.3 nm.

The meteorological data were supplied by the National Institute of Meteorology, San Pablo Airport.

The traffic flows data were measured with gauging pneumatic tubes, equipped with counters and recorders.

RESULTS AND DISCUSSION

Comparative study of the atmospheric lead concentrations in the city of Seville

The annual mean concentrations of lead, and its corresponding intervals, in the seven locations studied are shown in Table I. The lead percentages in the TSP and the traffic parameters are also included, as well as other characteristics.

It can be observed that the highest average annual lead concentrations (superior to $2\,\mu g \cdot m^{-3}$, which is the maximum value recommended by the European Common Market) are registered in Marqués de Paradas and Resolana. Both points are characterized by their high traffic flow, while Marqués de Paradas has a minor vehicle speed due to the existence of complicated cross-roads in the area. This last fact accounts for its higher lead levels, in spite of its minor value of traffic flow.

The lowest concentrations have been measured in Reina Mercedes. These values can be considered as the background levels of the city,

TABLE I

Annual mean concentrations of lead in different zones of the city of Seville during March 1983–February 1984

Locations	Air concentrations ($\mu g.m^{-3}$)	Percentage in TSP	Traffic parameters: Flow traffic ($vehicle \cdot day^{-1}$)	Traffic parameters: Vehicle speed	Observations
Plaza Nueva	1.30 (2.30–0.34)	0.51	23,300	Slow	Narrow streets
Marqués de Paradas	2.78 (4.75–0.88)	0.70	50,120	Very slow	—
Resolana	2.05 (4.03–0.72)	0.55	60,030	Rapid	—
Recaredo	1.04 (2.01–0.67)	0.50	58,430	Rapid	—
Luís Montoto	1.10 (1.93–0.58)	0.48	43,450	Medium	—
República Argentina	1.45 (2.26–0.76)	0.55	28,550	Slow	High buildings
Reina Mercedes	0.41 (0.96–0.14)	0.27	—	—	—

since this sampling point was situated in a residential zone, with wide open-spaces and far enough from important circulatory roads.

For the rest of the locations, the measured concentrations were between 1 and 1.5 $\mu g \cdot m^{-3}$. It should be noted that although Recadero has a high traffic-flow the measured concentrations were lower than were expected due mainly to the traffic speed and to the longer distance between the sampling-point and the circulatory roads. The measured concentrations in the Plaza Nueva and República Argentina were higher than those expected due to the topographic characteristics of the roads (little width in the former and high buildings in the latter).

The average lead percentages in the TSP are around 0.5% in five of the seven locations studied. The most polluted point (Marqués de Paradas with 0.7%) and the least polluted (Reina Mercedes with 0.27%) are excluded from this interval. In the first case, this is due to the slowness of the traffic which causes the lead content in the particulates emitted by the vehicles to increase. In the second case, the low lead percentages are explained by the existence in the area of other sources of suspended particulates different to the vehicles.

Meteorological influence

Seasonal variation The variation of monthly mean atmospheric lead concentrations during the twelve months of study, in station No. 5, is shown in Figure 2.

It can be observed that the highest concentrations appear during autumn and winter, reaching a maximum in January, and the lowest appear in spring and summer, reaching a minimum in July.

This seasonal variation must be intimately related to the variation of some meteorological parameters. In order to be able to study these variations, in Figure 2 the variation of the inverse of the monthly mean values of wind speed and temperature at seven o'clock in the evening is shown. This temperature has been considered in order to bear in mind the inversions produced by the cooling of the surface of the earth during the last hours of the evening, especially on clear days. On the other hand, since the city of Seville does not possess termic stability data, it has not been possible to represent the variation of the mentioned variable.

It can be observed that a parallel evolution exists between the lead concentrations and the inverse values of the meteorological parameters considered. The high lead concentrations during the winter coincide with low wind speeds and low temperatures, while the low concentrations during the summer coincide with high values of the mentioned parameters. These conclusions agree with those described by various investigators[3–6] in other locations.

Linear correlations The relationship between the meteorological parameters considered and the lead concentration can be better evaluated by carrying out a regression lineal analysis. The regression equations obtained, which relate the lead concentration with the inverse of the wind speed and temperature, by pairs and conjunctly, are shown in Table II. The high correlation coefficients obtained should be noted.

The comparison of the monthly mean concentrations calculated using the conjunct equation with the observed values indicates fair coincidence between the two (Table III).

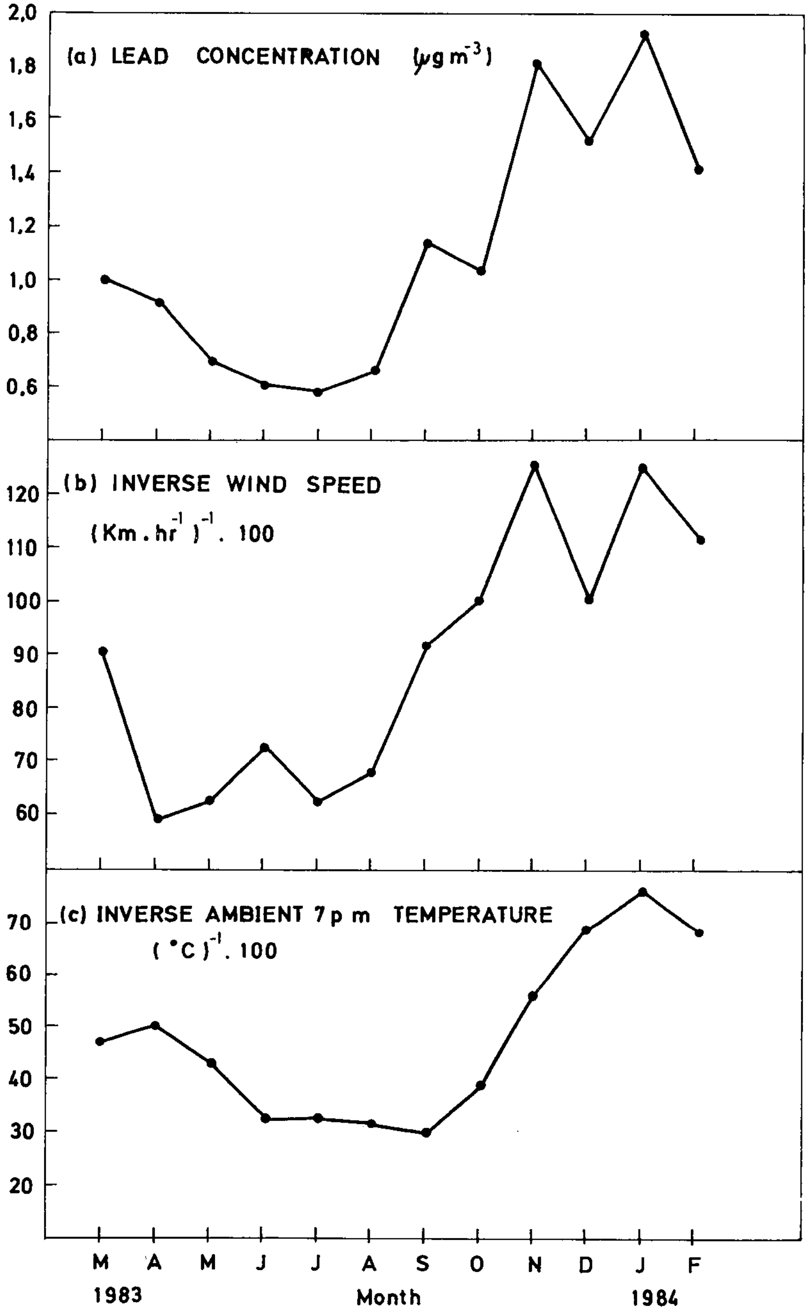

FIGURE 2 Seasonal variation of monthly mean atmospheric lead concentrations and meteorological parameters at Luís Montoto (sampling point No. 5).

TABLE II

Regression equations relating atmospheric lead concentrations to wind speed and ambient 7 p.m. temperature at Luís Montoto (sampling point No. 5)

Variables	Equation	Correlation coefficient (r)
$Pb-v^{-1}$	$Pb=17.76v^{-1}-0.46$	0.92
$Pb-t_7^{-1}$	$Pb=23.54t_7^{-1}-0.02$	0.82
$Pb-v^{-1}-t_7^{-1}$	$Pb=12.96v^{-1}+10.30t_7^{-1}-0.54$	0.95

Pb = Monthly mean atmospheric lead concentrations, in $\mu g \cdot m^{-3}$
v = Monthly mean wind speed, in $km \cdot hr^{-1}$
t_7 = Monthly mean ambient 7 p.m. temperature, in °C

TABLE III

Comparison of observed and calculated monthly mean concentrations at Luís Montoto (sampling point No. 5)

Month	Lead concentration observed $\mu g \cdot m^{-3}$	Lead concentration calculated $\mu g \cdot m^{-3}$
March	0.99	1.12
April	0.92	0.73
May	0.68	0.71
June	0.61	0.71
July	0.58	0.60
August	0.66	0.65
September	1.15	0.95
October	1.02	1.15
November	1.81	1.65
December	1.50	1.47
January	1.93	1.86
February	1.41	1.60

Traffic influence

The atmospheric lead concentrations are directly related to various vehicle traffic parameters such as their flow and their speed, apart from the meteorology, topographic characteristics of the circulatory roads and the distance from the sampling point. Because of this and due to the diversity of factors, it is difficult to clearly establish the traffic influence on the atmospheric lead concentrations.[7–9]

In this work, sampling point No. 5 was chosen for being situated at an altitude of 3 meters above ground level and near an important circulatory road. Sampling periods of 3 hours were adopted in order to compare the lead levels at different times of the day with different traffic flow which were measured simultaneously to the sampling. The traffic speed was considered constant during the sampling and was not measured.

Since the meteorological parameters change during the day, both the temperature and the wind speed were measured during the sampling.

The average annual values of the lead concentrations and traffic flow for each of the 8 periods of an established 3 hours are shown in Table IV. The mean annual values of the temperature and wind speed measured at 2 a.m., 8 a.m., 2 p.m. and 8 p.m. also appear.

It can be observed that during the night (2 a.m. to 8 a.m.), when the atmospheric stability is high, the lead concentrations follow a variation similar to that of the traffic flow. Likewise, the maximum lead concentrations are obtained during the first hours of the morning (8 a.m. to 11 a.m.) coinciding with the maximum traffic flow. However, during the rest of the day (11 a.m. to 8 p.m.), although the traffic maintains a certain steadiness with various fluctuations, the lead levels gradually decrease. This fact can only be explicable if the increase in the wind speed and in the temperature during the day are taken in mind, which aids the dispersion of pollutants. At the end of the day (8 p.m. to 11 p.m.) the decrease of the atmospheric instability results in a slight increase of the lead concentration, which will later rapidly decrease as a consequence of the diminution of the traffic flow.

Relationship with other traffic generated pollutants

The variation of the mean monthly values of the atmospheric concentrations of lead, carbon monoxide and smoke during the twleve months of study in sampling point No. 5, are shown in Figure 3.

It can be observed that the seasonal variation in the carbon monoxide and smoke levels is very similar to that previously discussed for those of lead. The equations obtained from the regression analysis carried out appear in Table V. The high correlation coefficients obtained (0.91 for carbon monoxide and 0.82 for

TABLE IV

Annual mean values of flow traffic, wind speed, ambient temperature and atmospheric lead concentrations over three hours periods, at Luís Montoto (sampling point No. 5)

	Time (hours)							
	2 a.m.–5 a.m.	5 a.m.–8 a.m.	8 a.m.–11 a.m.	11 a.m.–2 p.m.	2 p.m.–5 p.m.	5 p.m.–8 p.m.	8 p.m.–11 p.m.	11 p.m.–2 a.m.
Flow traffic (vehicle · hr^{-1})	305 (240–434)	890 (429–1035)	2670 (2543–2744)	2538 (2409–2696)	2507 (2387–2630)	2256 (2139–2359)	2198 (2140–2338)	1150 (735–1598)
Wind speed (Km · hr^{-1})	7.4–6		6–12.7		12.7–14.1		14.1–7.4	
Temperature (°C)	15.7–13.8		13.8–23.9		23.9–23.1		23.1–15.7	
Lead concentration ($\mu g \cdot m^{-3}$)	0.35 (0.20–0.42)	0.63 (0.38–0.76)	1.26 (0.55–1.99)	0.67 (0.49–1.14)	0.69 (0.43–1.28)	0.61 (0.37–0.98)	0.77 (0.30–1.12)	0.28 (0.22–0.34)

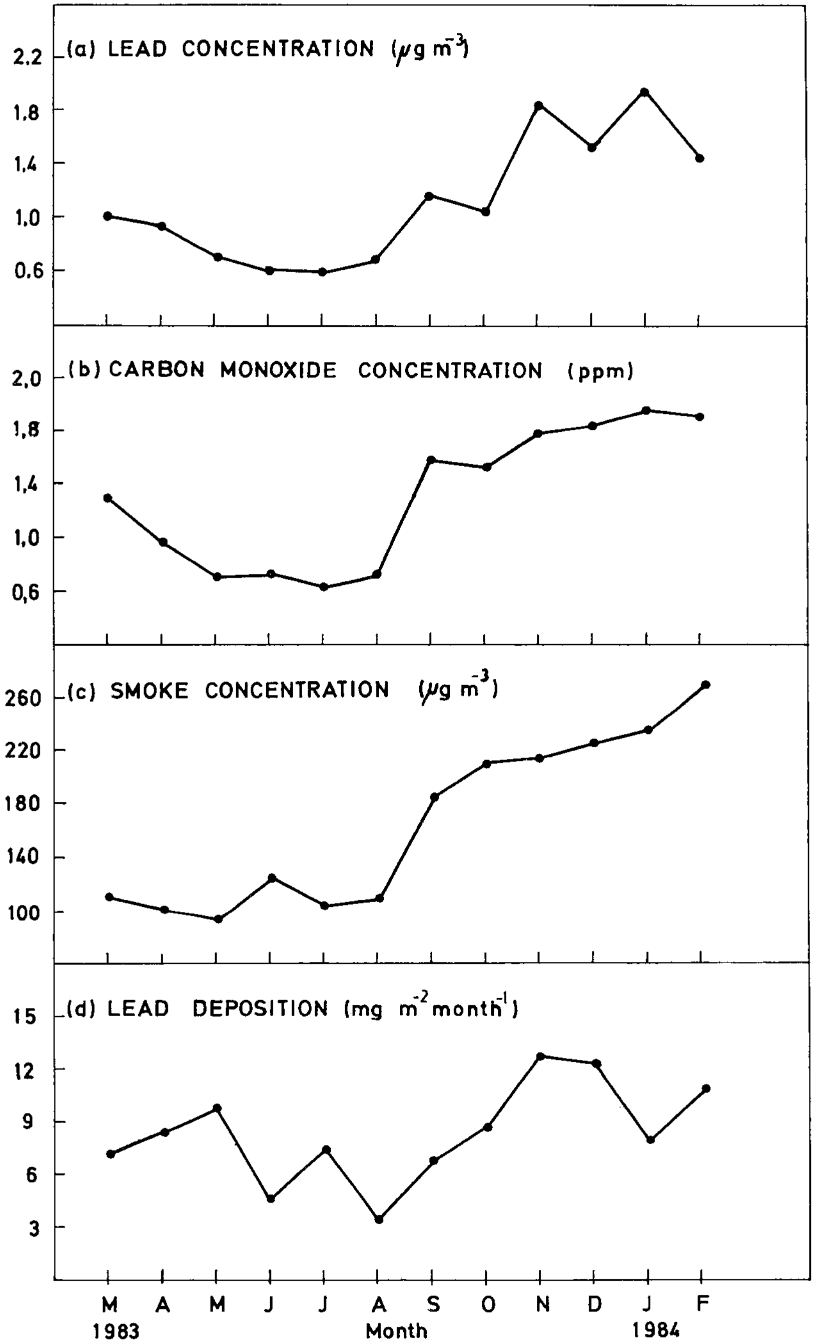

FIGURE 3 Monthly mean atmospheric concentrations of lead and other traffic-generated pollutants.

TABLE V

Regression equations relating atmospheric lead concentrations to carbon monoxide, smoke and setteable lead concentrations at Luís Montoto (sampling point No. 5)

Variables	Equation	Correlation coefficient (r)
Pb–CO	$Pb = 0.14 + 0.72\,CO$	0.92
Pb–BSS	$Pb = 0.11 + 0.006\,BSS$	0.82
Pb–Pb_{set}	$Pb = 0.24 + 0.10\,Pb_{set}$	0.64

Pb = Monthly mean atmospheric lead concentration, in $\mu g \cdot m^{-3}$
CO = Monthly mean atmospheric carbon monoxide concentration, in ppm
BSS = Monthly mean atmospheric smoke shade concentration, in $\mu g \cdot m^{-3}$
Pb_{set} = Monthly mean atmospheric lead deposition in $mg \cdot m^{-2} \cdot month^{-1}$

smoke) should be noted, indicating the existence of a common source (vehicles) for the three pollutants. The existence of a good relationship with the carbon monoxide level agrees with the results obtained by other investigators.[10] However, the relationship between the smoke concentrations has hardly been studied. The existence of a good correlation in the city of Seville can be explained by the point of view that Seville is a scarcely industrialized city, and that due to its climatic characteristics, domestic heating installations are few in number. Due to all this, the smoke emissions are fundamentally associated with the vehicles, the contribution of the stationary sources being relatively small.

With respect to the setteable lead levels it can be observed in Figure 3 that its seasonal variation is not similar to that observed by the suspended lead levels. The regression analysis carried out (Table V) indicates a correlation coefficient appreciably less to that obtained with the previous pollutants. These results are explicable, since the fluctuations in the setteable lead levels obey other meteorological parameters (rainfall fundamentally) and besides, only a small part of the lead emitted by the vehicles deposits. The main sources of setteable particulate lead are of an industrial type.

Finally, it must be noted that the correlations studies carried out in this work will allow the reduction in the number of determinations in an air quality network of traffic pollution.