EVOLUTION OF LEAD CONCENTRATION IN THE PARTICULATE MATTER OF SANTIAGO, CHILE, SINCE 1978

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Abstract. Santiago is a city of over 4.5 million inhabitants and about 415000 motor vehicles using leaded gasoline. Twenty-four-hour samples were taken of total and fractionated particulate matter using a low-volume system and two Andersen cascade impactors, between 1978 and 1989 at different sites of the city of Santiago. Lead concentrations were determined by atomic absorption spectrophotometry. The results show that Pb concentrations vary greatly depending on the site selected. Higher values are always detected near the city center. An annual variation with the highest value during the autumn-winter period (May-July) is clearly observed. Lead concentrations versus size are clearly biased to particles smaller than 2 μm. A correlation is observed between Pb concentrations and the suspended particulate matter under 3 μm at different sites of the city. The curves of Pb concentration versus size of the aerosols emitted by the gas exhaust of motor vehicles using leaded gasoline are similar to those of the atmospheric aerosols.

1. Introduction

Lead is a pollutant that is extremely widely disseminated in the environment. On a global scale the annual emissions of anthropogenic lead represent 3 to 100 times the natural ones.

A high percentage of anthropogenic emissions polluting the air correspond to lead compounds (Viala *et al.*, 1973; Hofscheuder and Harssema, 1983; Boiteau *et al.*, 1983; Vostal *et al.*, 1983). They also pollute homes (Raunemaa *et al.*, 1983) and food (Hofscheuder and Harssema, 1983). Other sources, like lead pipes, paints, and different industries (Petrilli *et al.*, 1983) may be, or may have been not long ago, locally prevalent.

The lead incorporated in gasoline is emitted to the atmosphere essentially as mineral salts, oxides, halides, and mixed compounds. Some organic lead compounds have also been detected near traffic areas (Kleinman, 1980). Of the Pb emitted, 95% is included in microspheres of aerodynamic diameter less than 0.5 μm (Lee *et al.*, 1971). Of these microspheres 60–90% remains suspended in the air and can reach long distances when they are emitted (Raunemaa *et al.*, 1983), or can remain near their sources when they are emitted in the cities.

Particles of < 0.5 μm in diameter are 100% respirable and, according to the AIEA (1978), this means that they can reach the non-ciliate part of the lungs, where the gaseous exchanges with the blood occur.

A regulation for Pb alkyls has economic and technical consequences; nevertheless, the risks are so important that some European countries and the United States

have taken some control measures; as a consequence, the Pb concentration in the air of rural and urban areas, as well as in the soils near the highways or roads of those countries (Eisenreich *et al.*, 1986; Byrd *et al.*, 1983), is decreasing.

Mexico is a Latin-American country that is reducing Pb in gasoline. It has been reported that this reduction has caused an increase in the tropospheric ozone concentrations (Bravo *et al.*, 1989).

The purpose of this study is to integrate all of the information produced by our research group since 1978, in order to quantify the evolution of Pb concentration in the air of Santiago during this period, and to establish the curves of Pb concentration versus diameter of the particles in the atmospheric aerosols and in the emissions of the gasoline motor vehicles of Santiago, and in this way be able to evaluate the eventual effects on the health of the population of the city.

2. Experimental

The low-volume system uses a vacuum pump of about 1.77 $ft^3\ min^{-1}$ and a cellulose filter.

One of the Andersen impactors has five stages with aerodynamic diameter: 2.84 μm, 2.04 μm, 1.40 μm, 0.80 μm, and 0.41 μm (corrected by the Cunningham factor), operating at a sample flow rate of 4.9 $ft^3\ min^{-1}$; the other impactor operates at a flow rate of 1 $ft^3\ min^{-1}$ and has five stages with aerodynamic diameter: 3.3 μm, 2.2 μm, 1.1 μm, 0.7 μm, and 0.40 μm (corrected by the Cunningham factor). A Millipore filter of 0.8 μm diameter pore size was used as a back-up filter for both impactors.

The first three years (1978 to 1980), the 24-h, low-volume sampling lasted a whole week each month for five sites located about 3 km north (north site: north campus of the Faculty of Chemical and Pharmaceutical Sciences, located near the foothill of the San Cristobal Hill), 5 km south (south site), 4.5 km east-southeast (east site), and 6 km west (west site) of the central site located at the central campus of the Faculty of Chemical and Pharmaceutical Sciences, University of Chile (central site; about 1.8 km east-southeast of downtown). Figure 1 shows the location of the different sites mentioned in this paper.

During 1980, 1981, and 1985, the sampling with the Andersen cascade impactor of 4.9 $ft^3\ min^{-1}$ was run at the north site. This sampling considered different seasons of the year with 24-h samples during 1 to 2 weeks each time. The concentrations of Pb night (9 PM to 8 AM) and day (8 AM to 9 PM) were also obtained using the low-volume system.

During 1983 and 1984 the sampling with the cascade impactor was extended to seven other sites in the city, including some suburban sites located between 14 and 27 km from the central site. These sites are: La Florida, La Granja, Maipú, Pudahuel, Apoquindo, Grecia and Providencia. Downtown corresponds to the central site that was moved in 1983 about 2.1 km northwest. The sampling of the north site during 1988 and 1989 was reinforced by a simultaneous sampling of the

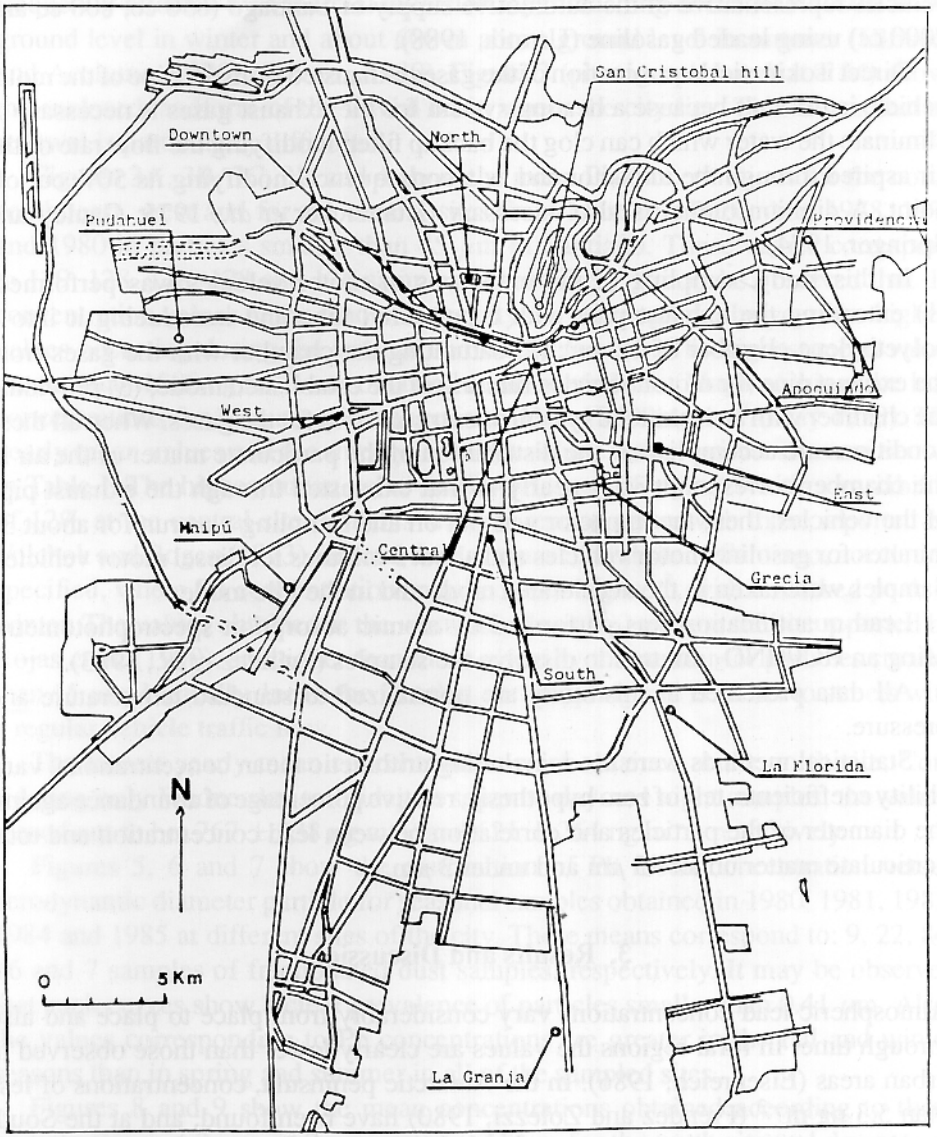

Fig. 1. Scheme of the city showing the different sites sampled.

atmospheric aerosols above the thermal inversion layer, with the second Andersen cascade impactor, located at the top of the San Cristobal Hill (about 860 m above sea level).

The Andersen cascade impactor was also adapted to the sampling of three motor vehicles representative of the automotive supply of Santiago (650 cc, 800 cc and 2000 cc) using leaded gasoline (Urrutia, 1988).

Direct isokinetic determination of the gases exhausted from the pipe of the motor vehicle is difficult because a heating system for the exhaust gases is necessary to eliminate the water which can clog the backup filter, modifying the flow rate of the air aspired through the impactor and as a consequence modifying its 50% cut-off point. A dilution tunnel is also necessary (Huntzicker *et al.*, 1975; Gauley and Springer, 1974).

In this study, sampling of the emissions of motor vehicles was performed: (1) extending the exhaust pipe with a polyvinyl tube and introducing it into a polyethylene chamber of 1.2 m^3; (2) saturating the chamber with the gases from the exhaust pipe for motor vehicles running in the established mode; (3) providing the chamber with an isokinetic system of entry and exit for the gases. When all these conditions are accomplished, the distribution of the particulate matter of the air in the chamber corresponds very nearly to that exhausted through the exhaust pipe of the vehicles; then, the impactor was put on and sampling was run for about 30 minutes for gasoline motor vehicles and about 5 minures for diesel motor vehicles. Samples were taken in the accelerated mode and in the idle mode.

Lead quantification was performed by atomic absorption spectrophotometry, using an HCl/HNO_3 mixture to dissolve the samples (Préndez *et al.*, 1983).

All data presented in this study are normalized to standard temperature and pressure.

Statistical methods were used, including arithmetic mean concentrations, variability coefficients, test of zero hypothesis, relative percentage of abundance against the diameter of the particles and correlation between lead concentration and total particulate matter under 45 μm and under 3 μm.

3. Results and Discussion

Atmospheric lead concentrations vary considerably from place to place and also through time. In rural regions the values are clearly lower than those observed in urban areas (Eisenreich, 1986). In the Antarctic peninsula, concentrations of less than 5.3 ng m^{-3} (Préndez and Zolezzi, 1980) have been found, and at the South Pole, values of less than 0.3 ng m^{-3} have been reported (Zoller *et al.*, 1974). In large cities in the U.S.A. and Europe, the values range from 500 to 10000 ng m^{-3} (Viala *et al.*, 1983), with great daily and monthly variations. In many of these places these values have tended to decrease in recent years (Gällipurghart, 1990).

In relation to Santiago, there is information about some specific places (Préndez *et al.*, 1980; Préndez and Garrido, 1981; Rojas *et al.*, 1990; Trier and Silva, 1987)

and sites that were part of a Latin-American monitoring network (Volchok and Bogen, 1973).

Santiago, Chile, is located in the subtropical region, about 33°30′ S lat. and 72°20′ W long. and it is affected by the Pacific anticyclone. For this reason, it is frequently affected by a thermal inversion layer reaching about 300–400 m above ground level in winter and about 600 m above ground level in summer (Ulriksen and Aceituno, 1975; Ulriksen, 1980). Figure 2 (photograph Landsat T M) shows the enclosed landform of the fluvial basin where the city lies. This means that air renewal is particularly difficult.

Figures 3A, 3B, 3C, 3D and 4 show the mean Pb concentrations for the north, south, east, west and for the central sites from March to December, 1978, 1979 and 1980 in particles smaller than 45 μm in Santiago. These values correspond to 139, 128, 128, 128 and 136 samples, respectively. It may be observed that the concentration values in the whole of the city vary throughout the year, the highest values occurring during the autumn and winter months. The highest mean value, greater than 1500 ng m^{-3} was obtained at the central site (May); at this place lead concentrations for 24-h samples range from 450 to 2800 ng m^{-3} (May 1978). The yearly mean values and the three year mean values at the various sites are shown in Table I. The highest mean value for 78–80 was 705 ng m^{-3} with a variability of 12% at the central site. For this site our results agree with those reported by Volchok and Bogen (1973) for total dust at some central site in Santiago, not clearly specified, where Pb concentrations reached 1000 ng m^{-3}, with a marked peak in winter. The values obtained at the west site, about 800 m from the site reported by Rojas *et al.* (1990), are higher than those given by this author for particles smaller than 15 μm, basically due to the fact that our sampling site is closer to streets with a regular vehicle traffic flow.

The lowest yearly mean concentrations were obtained at the north site. Considering only Wednesday to Friday in separate day and night samples, the values turned out to be: 767 ± 238 ng m^{-3} and 621 ± 264 ng m^{-3}, repectively.

Figures 5, 6 and 7 show the distribution of Pb mean concentration versus aerodynamic diameter particle for seasonal samples obtained in 1980, 1981, 1983, 1984 and 1985 at different sites of the city. These means correspond to: 9, 22, 84, 16 and 7 samples of fractionated dust samples, respectively. It may be observed that such curves show a clear prevalence of particles smaller than 0.41 μm. Also, the values corresponding to Pb concentrations are greater in the fall and winter seasons than in spring and summer in all of the sampled sites.

Figures 8 and 9 show the mean concentrations obtained according to three particle sizes: < 3 μm, < 2 μm and < 0.41 μm for the north site and downtown, respectively. The figures for Pb concentrations at the north site for the years 1980, 1981, 1983, 1985 and 1989 once again confirm that the highest values are obtained in the colder fall and winter seasons but there is no evidence of a rise in the concentrations along the years. At downtown, the Pb concentration values for the years 1983 and 1984 show that the seasonal peak varies from fall to winter

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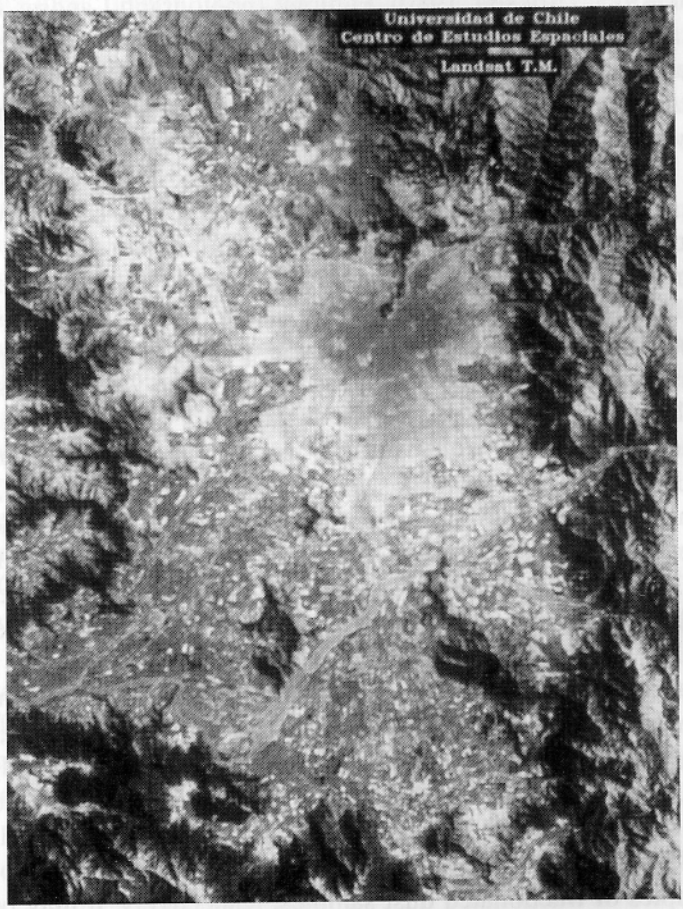
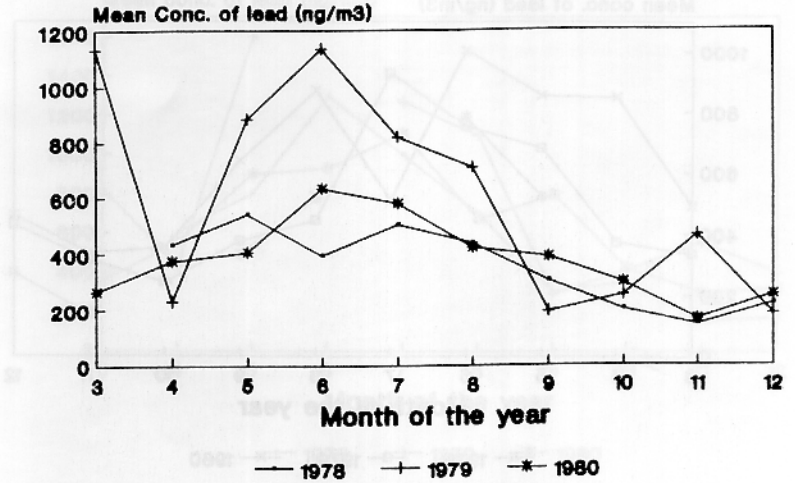
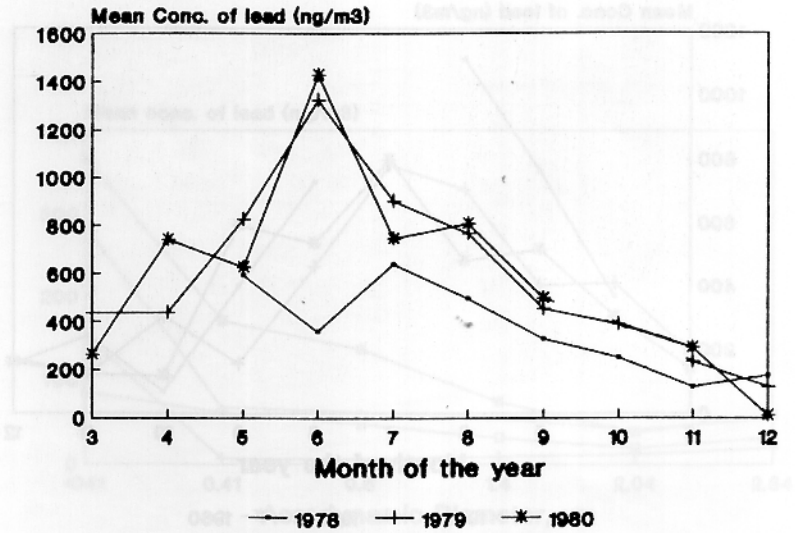


Fig. 2. Fluvial basin of Santiago. Photograph from Landsat.

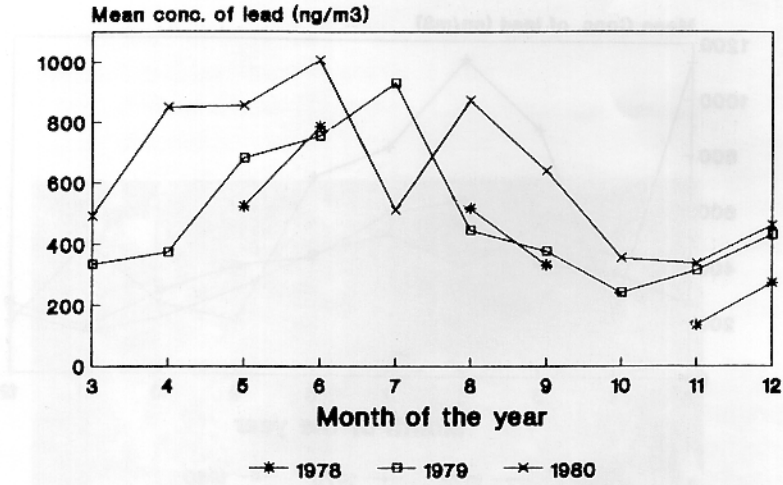


A) North site

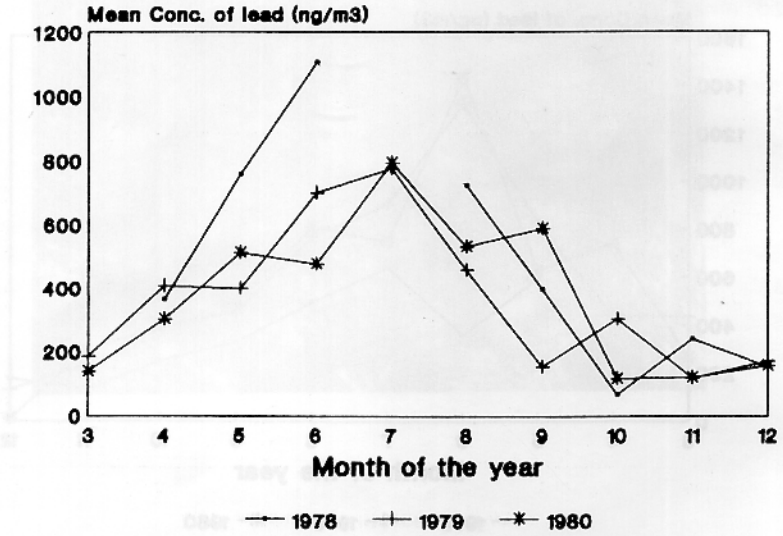


B) South site

Fig. 3. Mean concentration of Pb in particles smaller than 45 μm for different sites of the city of Santiago, Chile, in the months of March through December 1978, 1979 and 1980: (A) North, (B) South, (C) East and (D) West.



C) East



D) West site

Fig. 3. *Continued.*

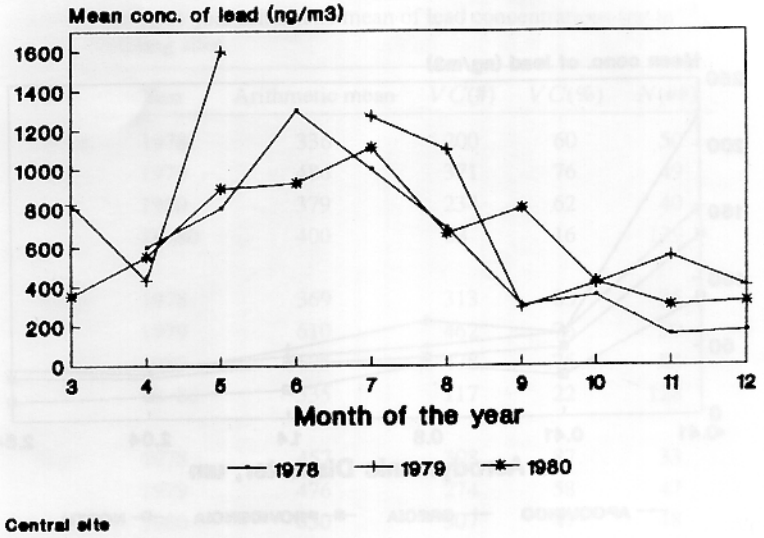


Fig. 4. Mean concentration of Pb in particles maller than 45 μm for the central site, Santiago, Chile, in the months of March through December 1978, 1979 and 1980.

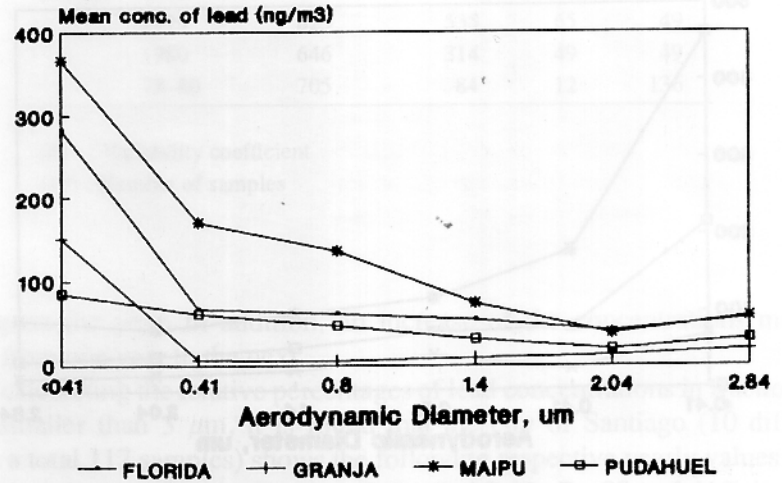


Fig. 5. Mean concentration of Pb as function of the particle size, for different sites of Santiago, Chile, during Fall and Winter.

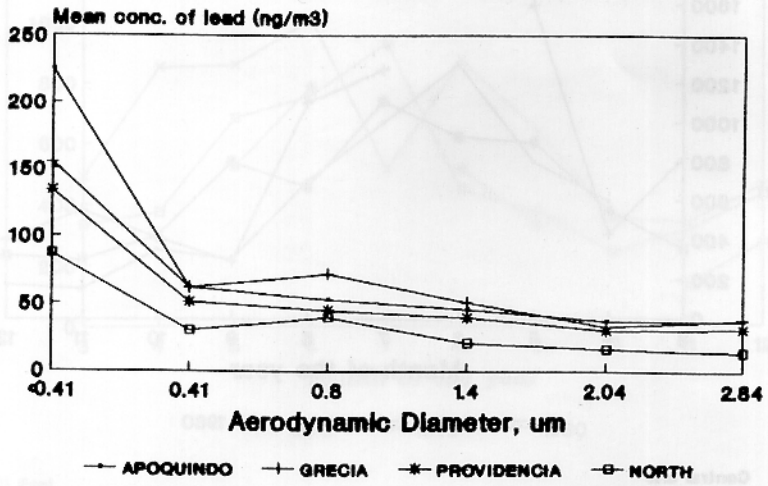


Fig. 6. Mean concentration of Pb as function of the particle size, for different sites of Santiago, Chile, during Spring and Summer.

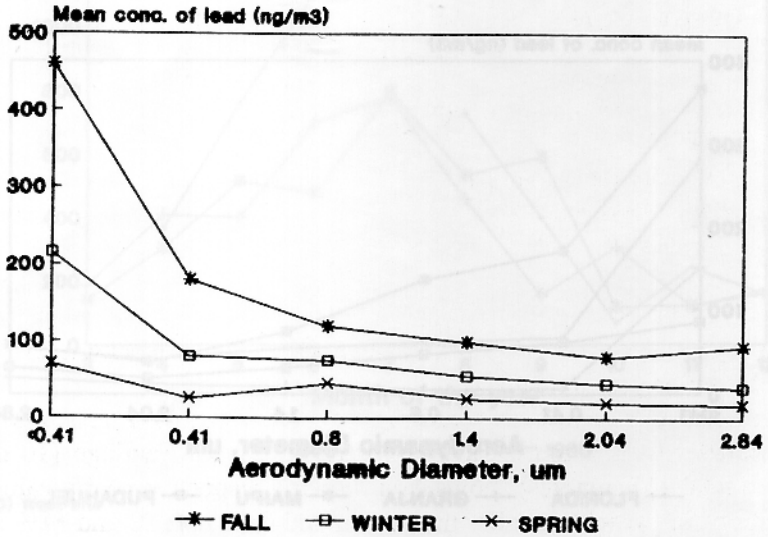


Fig. 7. Mean concentration of Pb as function of the particle size, for downtown Santiago, Chile, in Fall, Winter and Spring.

TABLE I
Annual and three year arithmetic mean of lead concentrations (ng m^{-3}), at five sampling sites.

Site	Year	Arithmetic mean	$VC(\#)$	$VC(\%)$	$N(\#\#)$
North	1978	336	200	60	50
	1979	486	371	76	49
	1980	379	234	62	40
	78–80	400	63	16	139
South	1978	369	313	85	35
	1979	610	462	76	50
	1980	625	478	75	43
	78–80	535	117	22	128
East	1978	457	398	87	33
	1979	476	274	58	47
	1980	650	307	47	48
	78–80	528	87	16	128
West	1978	562	426	76	34
	1979	377	300	79	44
	1980	374	300	80	50
	78–80	438	88	20	128
Central	1978	645	564	87	38
	1979	823	535	65	49
	1980	646	314	49	49
	78–80	705	84	12	136

(#) : Variability coefficient
(##) : Number of samples

depending on the year. In addition, an increase of the concentrations may be observed from one year to the next.

When calculating the relative percentages of lead concentrations in fractionated particles smaller than 3 μm, it is found that the city of Santiago (10 different sites with a total 117 samples) shows the following respective yearly values of Pb concentrations in particles smaller than 2 μm and 0.41 μm: 88 and 41% in 1980, 86 and 44% in 1981, 85 and 47% in 1983, 86 and 56% in 1984, and 81 and 43% in 1985. From this information a mean value of 85% and 46% may be calculated for Pb concentration in particles smaller than 2 μm and in particles smaller than 0.41 μm, respectively, for city atmospheric aerosols smaller than 3 μm.

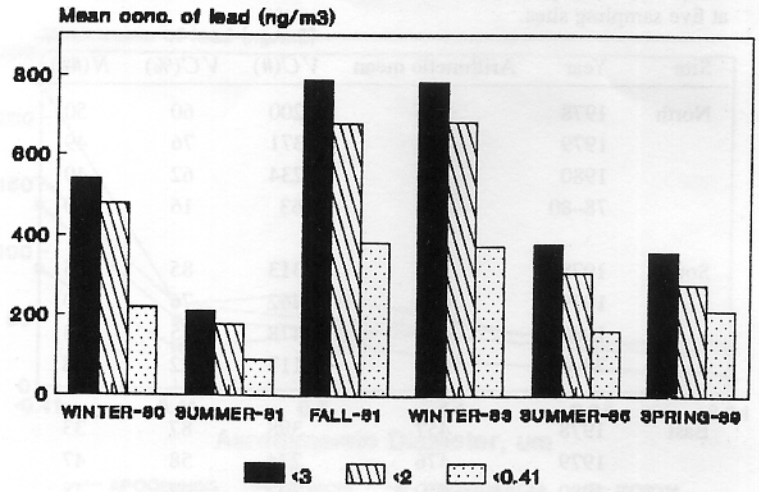


Fig. 8. Bar diagram for the average concentration of lead in aerosols < 3, < 2 and < 0.41 μm in the site North of the city of Santiago, Chile.

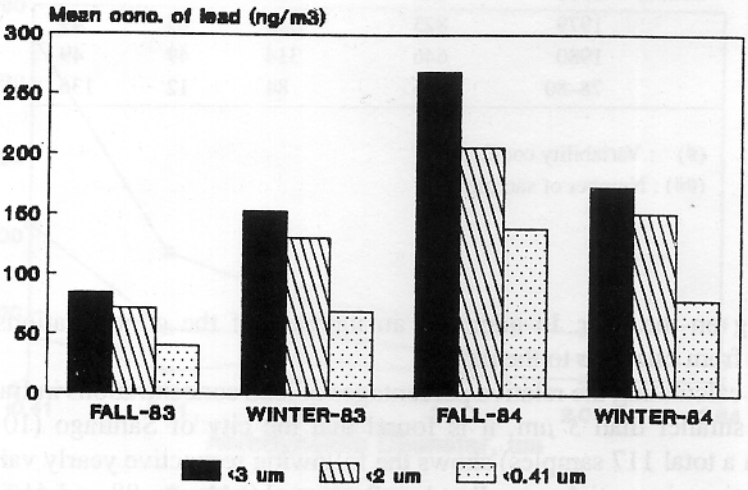


Fig. 9. Bar diagram for the average concentration of lead in aerosols < 3, < 2 and < 0.41 μm in downtown Santiago, Chile.

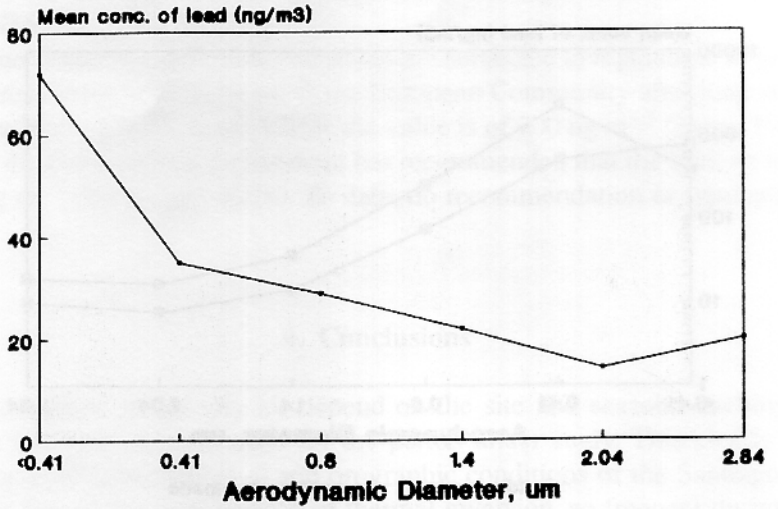


Fig. 10. Mean concentration of Pb as function of the particle size for atmospheric aerosols in the site North of Santiago, Chile, above the thermal inversion layer, in Spring 1988 and 1989.

Figure 10 shows the curve of mean concentration of Pb versus particle size for the atmospheric aerosols at the north site above the thermal inversion layer during the spring season in 1988 and 1989 (45 samples). Such a curve also shows the peak for particles smaller than 0.41 μm. The values for relative percentages of Pb concentrations in fractionated particulate matter smaller than 3 μm for particles smaller than 2 μm and smaller than 0.41 μm, are respectively 79% and 36% (1988), and 86 and 40% (1989). These results differ from those obtained for particles smaller than 0.41 μm at the same site below the thermal inversion layer, 79% and 60% respectively, as sampled with the 1 ft^3 min^{-1} impactor, but they are similar to those of other sites of the city, sampled with the 5 ft^3 min^{-1} impactor. Part of the Pb added to gasoline is retained in lubricant oil, in the oil filter and in the exhaust pipe; only 54% is eliminated to the atmosphere through the exhaust pipe (Huntzicker *et al.*, 1975).

The curves of Pb concentration versus particle diameter for aerosols emitted by motor vehicle exhaust as the result of the use of leaded gasoline are shown in Figure 11. The idea underlying the experiments conducted in this study was just to compare the shape of the curves of mass-size distribution versus diameter of the particles smaller than 3 μm, between the aerosols present at the urban air and those exhausted to the same air by motor vehicles, in order to evaluate the impact on the citizen inhaling it. It may be observed that the curves corresponding to idle mode are similar to those obtained for the atmospheric aerosols with the peak for particles

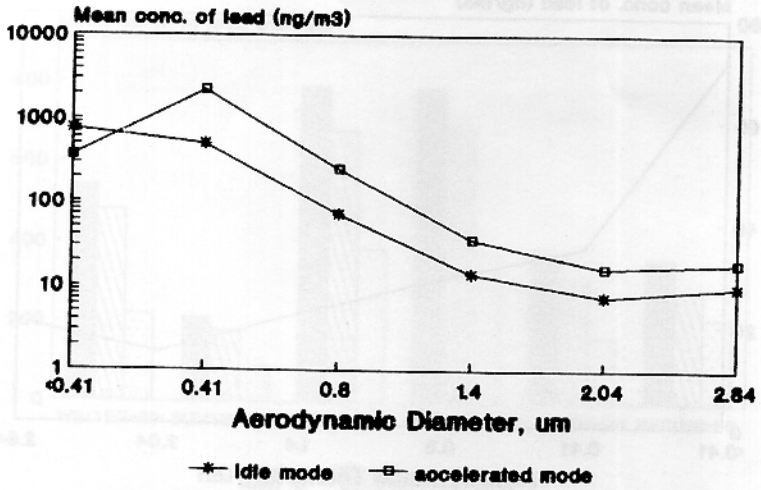


Fig. 11. Mean concentration of Pb as function of the particle size for aerosols emitted from motor vehicle exhaust, when engine uses leaded gasoline in idle and accelerated mode.

smaller than 0.41 μm. In the case of accelerated model the peak concentration is observed for particles with an aerodynamic diameter of 0.41 μm. The unimodal curves for Pb have also been observed in recent years in Puerto Rico (Infante and Acosta, 1991).

These results are also supported by the information provided by Cass and McRae (1983), who associated Pb halogenated compounds with fine particles, and by Buerki *et al.* (1989), who related the particle fraction with an aerodynamic diameter of 0.18 μm to vehicle traffic emissions, and by Dzubay *et al.* (1988) who found that more of 72% of Pb in the fine fraction of the dichotomus sampler can be attributed to vehicle exhaust.

Pair connections between Pb concentration and total particulate matter obtained for each stage of each sampling with the cascade impactors showed a 95% and 99% significant correlation at the sites most closely associated with a large vehicle traffic flow, such as the Central, Downtown, Providencia, Grecia and North sites. At this last site the correlation turned out to be 99% significant for urban aerosols collected above and below the thermal inversion layer, in agreement with the finding of Infante and Acosta (1991). For total dust samples collected with the low-volume, the correlation between total dust and Pb concentration turned out to be 99% significant.

Considering the total daily emissions, the distance (km) run, the number of motor vehicles using leaded gasoline, and estimating that between 54% and 80%

of the lead added to the gasoline is thrown out to the atmosphere, Urrutia (1988) calculated that between 319 and 472 tons of Pb were expeled to the air of Santiago during 1986.

In some countries air Pb concentrations are subjected to regulation. In the USA the EPA regulation is 1500 ng m^{-3}; the European Community after long analyses settled on 2000 ng m^{-3}. In the USSR the value is of 700 ng m^{-3} (Stern, 1984). In Belgium the Environment Department has recommended that the limit be reduced to 400 ng m^{-3} (De Regge, 1978). To date, no recommendation or standard exists in Chile.

4. Conclusions

Pb concentrations in the city air depend on the site and season, reaching 2800 ng/m^3 in fall and winter months for the period under study. This clearly shows the influence of meteorological and orographic conditions of the Santiago basin, especially that of the phenomenon of thermal inversion, so frequent during those seasons.

The concentration of Pb in air is greater in the central area and decreases towards the periphery, which agrees with the greater vehicle traffic flow in the central area. These results show a close association of the presence of Pb in atmospheric aerosols of the city, above and below the thermal inversion layer, with the emissions of the motor vehicles that use leaded gasoline.

Pb concentrations are frequently above the threshold value recommended or established in Belgium, the USSR, and the USA.

The greatest Pb concentrations are found in the finest particles, equal or smaller than 0.41 μm, which are 100% respirable for atmospheric aerosols smaller than 3 μm and thus may have an impact on the health of the Santiago population.

In Chile, the used lubricating oil is principally burned as complementary oil for furnaces and boilers, but it is also used on the soil to prevent dust, or it may be simply eliminated through the sewage system, thus polluting air, soil and water. So, considering the effects on health produced by the inhalation of lead, especially in children, the characteristics of the atmospheric aerosols containing Pb and their concentrations (greater than those accepted in other countries), the information revealed in this paper must be an indication to the official national authorities to establish the necessary controls for Pb in the urban air of Santiago.

Note

This paper was sent to EMA on October 1990 and accepted in October 1991. In 1991, the Chilean authorities decided to eliminate Pb in gasoline for the new motor vehicles of 1992.

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