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PARTICULATE CARBON, A SIGNIFICANT CONTRIBUTOR TO THE VISIBILITY REDUCTION OF MEXICO CITY

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RESUMEN

Se determinaron las concentraciones de carbono elemental total (C_t) en partículas suspendidas (PST), incluyendo las fracciones de carbono orgánico volátil (C_{ov}) y carbón libre (C_b) "carbón negro" en 92 filtros muestreados durante 1982, utilizando un muestreador de altos volúmenes (HI-VOL), de acuerdo con un programa de muestreos de 24 hrs. Simultáneamente, se muestreó la fracción fina de las PST (CFPS) utilizando un muestreador Andersen modificado. El sitio de muestreo fue "El Casco de Santo Tomás", localizado cerca del Centro de la Ciudad de México. El rango de concentraciones de C_t en las PST varió de 14.8 a 98.7 $\mu g/m^3$. La concentración promedio de C_t, C_{ov} y C_b en las PST fueron 49.87, 32.8 y 16.9 $\mu g/m^3$ respectivamente, con una correlación estadística de C_t y C_b en PST de 0.81 y 0.9; C_{ov} resulta de $C_{ov} = C_t - C_b$. La concentración promedio de CFPS fue 154 $\mu g/m^3$ (mediana, del diámetro de masa= 0.6μm), sin embargo, no se determinaron especies de carbono en ellas.

Usando procedimientos para estimar coeficientes de absorción y dispersión de luz reportados por diversos autores, así como de datos de sulfatos y nitratos en PST obtenidos en la estación de muestreo del Observatorio de Tacubaya, a 5.6 km de "El Casco de Santo Tomás" se estimó que la contribución a la reducción de visibilidad por carbono elemental total es cercana al 50%. Usando la ecuación de Koschmieder y el coeficiente de extinción resultante en este estudio ($b_{ext} = 7.64 \times 10^{-4} m^{-1}$), la visibilidad estimada para la Ciudad de México es de 5.10 km. La visibilidad promedio reportada por el Observatorio de Tacubaya para ese mismo período fue de 6.3 km.

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ABSTRACT

Total particulate elemental carbon concentrations (C_t) including organic volatile (C_{ov}) and black carbon (C_b), were determined in 92 stored high-volume sampled filters collected during 1982 in a 24 hr sampling period with an alternative schedule. Coarse and fine particulate mass (CFPM) were sampled at the same schedule and site using a modified Anderson sampler. The sampling site "El Casco de Santo Tomás", is located near the Mexico City's downtown area.

The results show that C_t ranged from 14.8 to 98.7 $\mu g/m^3$. The total suspended particulate (TSP) mean concentration was 204 $\mu g/m^3$. The C_t, C_{ov} and C_b mean concentrations content in the TSP were 49.87, 32.3 and 16.9 $\mu g/m^3$ respectively. The statistical analysis of C_t and C_b on TSP data shows a correlation coefficient of 0.81 and 0.9 respectively; C_{ov} results from: $C_{ov} = C_t - C_b$. The CFPM mean concentration was 154 $\mu g/m^3$ (0.5 μm mass median diameter), but C_t was not determined.

Using both specific light-absorption and light-scattering coefficients estimating procedures reported by several authors, and sulfate and nitrate data from the Tacubaya Observatory sampling station, 5.6 km from the first one, the contribution of the elemental carbon to the estimated visibility reduction was deduced to be near 50%.

The estimated visibility using the Koschmieder relationship and the extinction coefficient ($b_{ext} = 7.64 \times 10^{-4} m^{-1}$) obtained from the data was 5.10 km. The average visibility reported by the Tacubaya Observatory Station was 6.3 km for the same period.

INTRODUCTION

The Mexico City Metropolitan Area is located in the Southwest of an elevated basin, 2240 m above sea level (Fig. 1) at a latitude of 19°26'13" North. It has a population of 18 million inhabitants, close to 2 million motorized vehicles and 21% of the total industrial activity of Mexico.

One of the most important effects due to atmospheric pollution in Mexico City is the visibility reduction. Figure 2 represents the comparison of visibility frequency less than 2 km performed at the Tacubaya Observatory Station in Mexico City for the years 1937 and 1966 (Jáuregui, 1971).

The total suspended particle concentration (TSP) in the Mexico City Metropolitan Area is high and increasing (SEDUE, 1986). The TSP concentration is variable for each zone. Currently, the Northern area has the highest values, the Southern area has the smallest, and the downtown area has an intermediate value (Salazar *et al.*, 1981; Espinoza, 1982; Sigler *et al.;* Bravo *et al.*, 1984). Typical fine particle mass median diameter for Mexico City ranges from 0.45 to 0.65 μm (Bravo *et al.*, 1982; Sigler *et al.*, 1982; Bravo *et al.*, 1984).

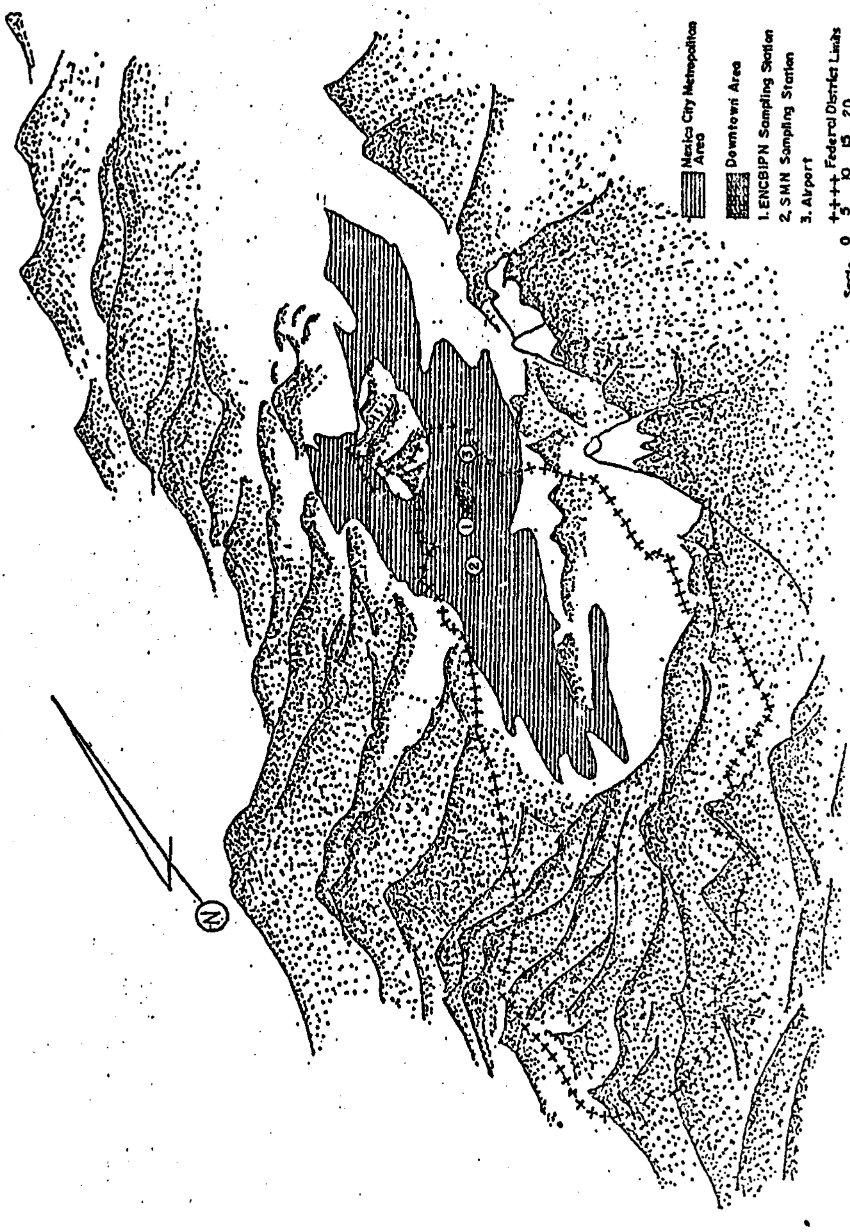

Fig. 1. Schematic representation of the Mexico City Metropolitan Zone in the Mexican Basin.

The mean visibility range, as measured visually from the Tacubaya Observatory station (10 km from downtown area) ranges from 5 to 7 km, depending upon the season of the year (SMN, 1970-1983).

Species such as Sulfates (SO_4^{-2}) and Nitrates (NO_3^-) had been evaluated for the TSP of Mexico City and they have shown to have an unimportant influence on visibility reduction (Bravo *et al.,* 1981). However, the elemental carbon content (including organic volatile and black carbon) in the TSP, shows good agreement with the visibility reduction in Mexico City.

THEORETICAL BACKGROUND

During the past few years, the potential importance of particulate elemental carbon (PEC) has been recognized because of its role in the visibility reduction (Wolff *et al.,* 1982). The presence in the atmosphere of particulate elemental carbon can be deduced from:

1) Source of PEC exist chiefly in the form of combustion of carbon-based materials and fuel,
2) under atmospheric conditions, PEC is inert to oxidation and modifitacion of its usual graphitic molecular structure, and it is removed by scavenging processes, and
3) it is observed that filter samples are gray in rural sites and black in urban areas, due to the fact that in Metropolitan areas a great percentage of the fossil fuels is consumed (Charlson and Ogren, 1982).

The consequences of the blackness or light-absorption properties are several:

(Wolff *et al.,* 1982; Reck, 1974; Wolff, 1983).

1) The elemental carbon is the most efficient visibility reducing particulate,
2) the heating or cooling of the atmosphere will be sensitive to the amount of elemental carbon because of the light-absorption properties of this particulate specie, and
3) under certain conditions, light-absorption and light-scattering have an additive effect on reducing visibility. The light-absorption efficiency for elemental carbon has been estimated to be between 4.9 to 13 m^2/g. Adding this to the ele-

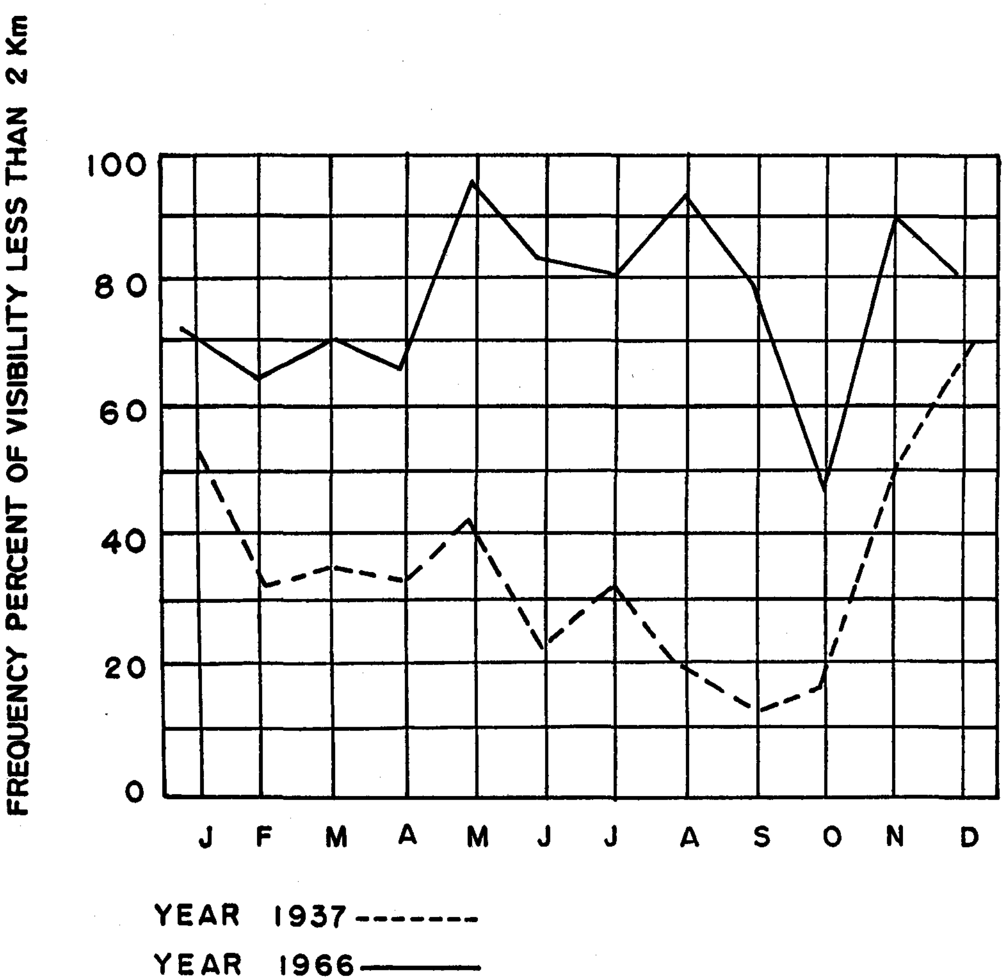

Fig. 2. Comparison of visibility frequency less than 2 km for the years 1937 and 1966 in the Mexico City Metropolitan Zone.

mental carbon light-scattering efficiency mean value of 3.2 m^2/g, brings the total light-extinction efficiency of elemental carbon to 8.1 - 17.2 m^2/g.

The airborne carbon particles consist principally of organic material accompanied by black nonvolatile soot components which have a chemical structure similar to impure graphite (Rosen and Novakov, 1978). Polycyclic aromatic hydrocarbons which are adsorbed onto soot particles show mutagenic activity, and hence are of public health concern (Wei *et al.*, 1980).

METHODOLOGY

In 1981, the Centro de Ciencias de la Atmósfera at the National University (CCA-UNAM) and the Escuela Nacional de Ciencias Biológicas of the Polytechnic Institute (ENCBIPN), with the technical aid of the Climatological Observatory Station of Tacubaya (SMN), organized a study aimed to understand the causes of the Mexico City visibility reduction. The sampling sites (Fig. 3) were located in the urban area of Mexico City. The sites were typically downwind of most significant sources. Consequently, the pollutant samples collected at the ENCBIPN and the SMN sites are thought to be representative of the TSP concentrations in the Metropolitan central area.

Particle suspended matter was collected at both sites. Two types of samplers were operated, the high-volume sampling system (a General Metal Works Hi-Vol sampler) with a collection medium of 8 x 10 inches glass fiber filters, and the modified cascade impactor (a Research Appliance Co., five stages and back-up filter). The instruments were calibrated at Mexico City conditions (2 240 m altitude).

The location of the samplers were on the roofs of the buildings. No taller buildings were nearer than 250 m (Fig. 4).

The first study period was from March to August 1981 on the roof of the SMN station. Sampling duration was 24 hr beginning at 08:00 hr (local standard time) for each event. Currently, range visibility and meteorological determinations are being done at the same SMN station on a hourly schedule. The inorganic analyses performed to the 41 samples collected with the Hi-Vol, were for sulfate and nitrate. The purpose of analyses of the 41 fractionated samples collected with the cascade impactor was the mass median diameter determination.

The second study period was from January to September 1982 at the ENCBIPN station. The analyses performed to the 92 samples collected with the Hi-Vol, were for PEC, Pb, Fe, Cu and Zn. The 92 fractionated samples collected with the cascade impactor were analyzed for the mass median diameter, Pb, Fe, Cu and Zn. However, metals in TSP and CFPM were not included in the visibility reduction analyses.

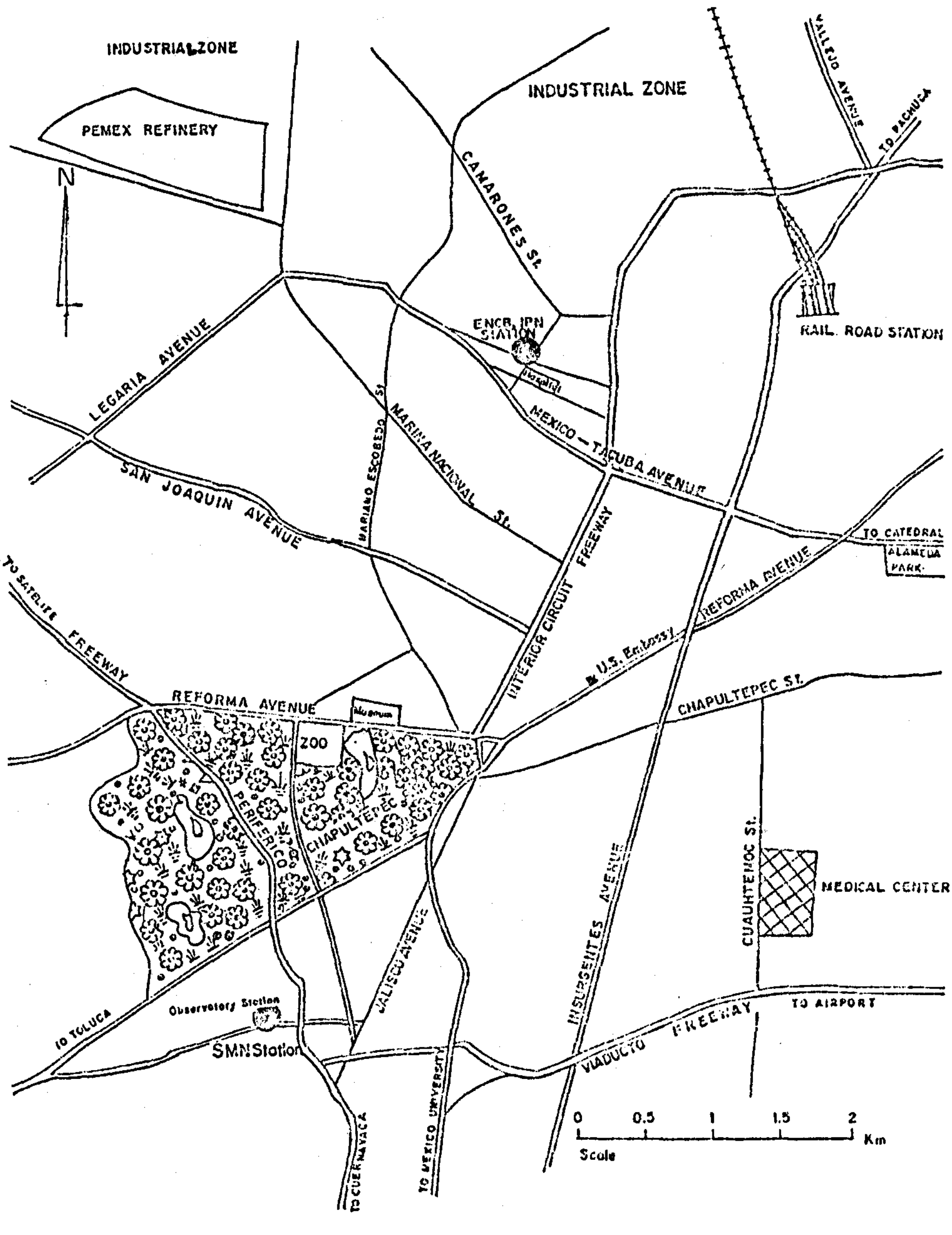

Fig. 3. Location of the ENCBIPN and SMN sampling sites in the Mexico City Central Area.

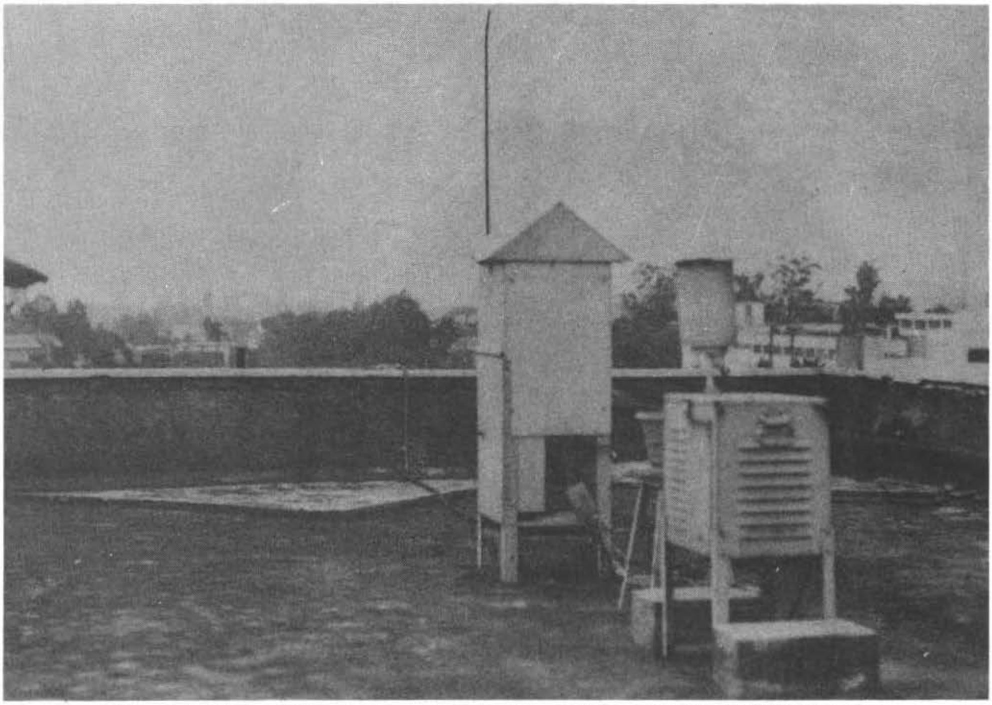

Fig. 4. Hi-Vol and Cascade Impactor samplers on the roof of the ENCBIPN station.

For the total elemental carbon content (C_t) in the TSP, the analytical procedure followed was the Walkley and Blank method for determination of carbon in soils (Walkley and Blank, 1934).

In this method the samples suffer a digestion with a $K_2Cr_2O_7$ titled solution in a H_2SO_4 medium. The carbon present is converted to CO_2 and H_2O. After the digestion, the $K_2Cr_2O_7$ in excess is titrated with a standard ferrous sulfate solution.

A filter blank not sampled was analyzed in each run.

However, the separation of the organic carbon from the absorbing soot component is very dangerous and difficult, so, just five selected sampled filters were analyzed for organic volatile material (C_{ov}) and black nonvolatile soot (C_b). The selection criteria for filters was to choose those which fall exactly on the curve resulting from the statystical adjust in the C_t and PST data (C_t vs. TSP correlation was: $r^2 = 0.81$, see Figure 5).

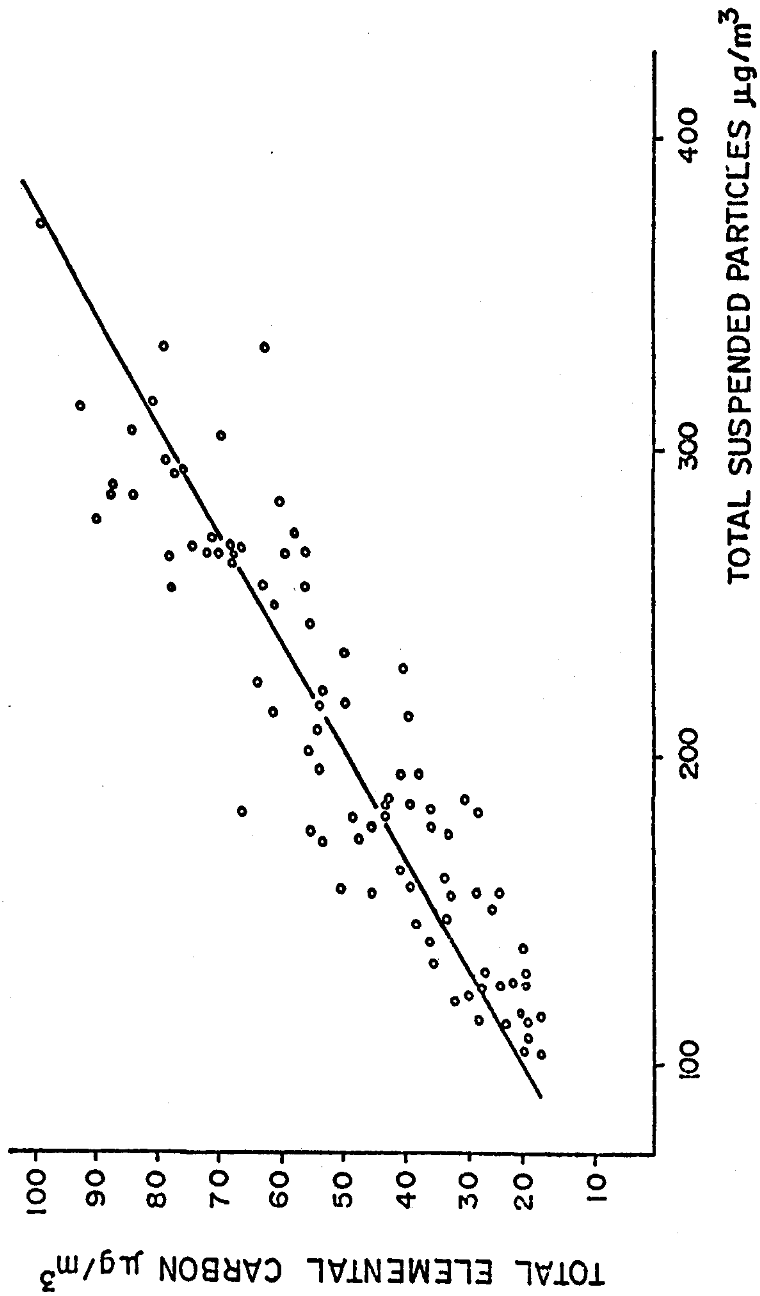

Fig. 5. Plotting of data from C_t and TSP at the ENCBIPN station.

The procedure consists of a nitric acid digestion for 24 hour (McCarthy and Moore, 1952). Filter samples were treated with 70% nitric acid and heated to destroy the organic matter. The residue which contains the free carbon and insoluble inorganic matter is collected in a porcelain filter crucible, dried, and weighted, free carbon (carbon black) then determined by the loss of weight of the crucible on ignition.

Table 1 shows the summary of the parameters measured at the SMN and ENCBIPN stations.

VISIBILITY BUDGET

An accurate calculation of the contribution to visibility of the various chemical components of the aerosol requires detailed data on the particle size and chemical composition distribution as well as simultaneous optical measurements.

The correlation of Pratsinis *et al.* (1984) is a correction of the correlation of Groblicki *et al.* (1981) and was used here to set up a visibility budget for particles at the urban area of Mexico City. The visibility (visual range) V is related to the light-extinction coefficient, b_{ext}, by the Koschmieder relationship:

$$V = \frac{3.912}{b_{ext}} \tag{1}$$

The extinction coefficient is the sum of the contributions from light scattering and absorption by gases and particles at the wave length of interest:

$$b_{ext} = b_{sg} + b_{sp} + b_{sw} + b_{ag} + b_{ap} \tag{2}$$

where b_{sg}, B_{sp} and b_{sw} are the scattering contributions from gases, dry particles and water in the aerosol phase, respectively. The absorption contribution from gases and particles is denoted b_{ag} and b_{ap} respectively.

The molecular (Rayleigh) scattering for the visual wave length (λ = 560 nm) at 20°C and 2 240 m above sea level obtained from Wagooner *et al.* (1981) is:

$$b_{sg} = 0.0758 \times 10^{-4} m^{-1} \tag{3}$$

The scattering coefficient for dry aerosol particles as given by Pratsinis *et al.* (1984) is:

$$b_{sp} = 0.047 \{(NH_4)_2SO_4\} + 0.020 \{NH_4NO_3\} + 0.031 \{1.2\, C_{ov}\} + 0.023 \{C_b\} + 0.012 \{Remainder\} - 0.12 \tag{4}$$

Table 1

Summary of parameters measured in the Mexico City Visibility Reduction Study

Parameter	Symbol	Frequency of measurements	Instrumental or Analytical method	Reference
Total particulate mass	TSP	24 hrs	High-Volume Sampler	EPA (1976)
Coarse and fine particulate mass	CFPM	24 hrs	Modified Andersen Fractionating Sampler	Lee and Goranson (1972)
Sulfates in TSP	SO_4^{-2}	24 hrs	Turbidimetric	U. S. Department of Health, Education and Welfare (1965)
Nitrates in TSP	NO_3^-	24 hrs	Spectrophotometric	U. S. Department of Health, Education and Welfare (1965)
Total carbon in TSP	C_t	24 hrs	Wet Chemical Method	Walkley and Blank (1934)
Organic and black carbon in TSP	C_{ov}, C_b	24 hrs	Nitric Acid Digestion	McCarthy and Moore (1952)
Nitrogen dioxide	NO_2	24 hrs	Saltzman Method	EPA (1976)
Relative humidity	RH	1 hr	Hygrotermograph	WMO (1983)
Visibility range	Vr	1 hr	Visually	WMO (1983)

where C_{ov} and C_b represent the amount of volatile and black carbon respectively in $\mu g/m^3$. The coefficient $\{1.2\}$ in the volatile carbon entry is used to account for the associated oxygen and hydrogen. The term Remainder represents the difference between the measured total mass and the sum of the chemical components.

$(NH_4)_2 SO_4$ and NH_4NO_3 are in $\mu g/m^3$.

The relative humidity and the concentration of hygroscopic salts $(NH_4)_2SO_4$ and NH_4NO_3 determine the extent of the light scattering due to aerosol water b_{sw} (in $10^{-4}m^{-1}$). The relationship (5) is an updated version of one originally proposed by Cass (1979) and developed by Pratsinis *et al.* (1984).

$$b_{sw} = \frac{0.0124}{1-\mu} \left\{(NH)_2SO_4\right\} + \frac{0.0105}{1-\mu} \left\{NH_4NO_3\right\} \quad (5)$$

where μ is % humidity/100. It should be noted that this formula does not have a physicochemical basis, but results from application of a regression model to empirical data.

Light absorption (λ = 550 nm) by gases is attributed to NO_2 (in $10^{-4}m^{-1}$) according to the relationship reported by Hodkinson (1966).

$$b_{ag} = 3.3 * [NO^2] \quad (6)$$

where NO_2 is the nitrogen dioxide concentration in p.p.m.

Groblicki *et al.* (1981) reported an absorption coefficient for black carbon of 11.8 m^2/g, while Wolfe *et al.* (1983) suggests the value of 11.24 m^2/g, so the intermediate value of b_{ap} (in $10^{-4}m^{-1}$) is:

$$b_{ap} = 0.116\ m^2/g*[C_b] \quad (7)$$

where C_b is the black carbon concentration in $\mu g/m^3$.

To apply equation (4) to our data set, some conversions and assumptions are necessary. The measured sulfate and nitrate concentrations were converted to their ammonium salts. Since sulfate and nitrate data were not available at the ENCBIPN station, the data obtained from the sampling site at Tacubaya were used (SMN station).

The equation (4) was developed originally for fine particles, so the results must be considered just as an approximation to explain the Mexico City visibility problem, although the CFPM comprises more than 75% of the PST for the ENCBIPN station.

RESULTS

Results from the parameters of interest in the present study are shown in Table 2 and Table 3 for the SMN and the ENCBIPN stations respectively. Table 4 shows statistical analyses from carbon and TSP data.

The coefficients contributions to light extinction coefficient after applying relationships (2) through (7) to our data are shown in Figure 6. Figure 7 shows the contribution of each component to the light extinction coefficient.

The carbon compounds contribute 46.3% (organic volatile carbon 15.1% and black carbon 31.2%). The other compounds such as sulfate and nitrate ammonium and water, contribute 30% and 5% respectively.

Table 5 shows a summary of light extinction budgets compiled by Latimer *et al.* (1985) in which results for Mexico City are included.

Table 2

Average results for the determinations performed at the Observatory Station of Tacubaya during March-August 1981

Parameter	Average result*
TSP	201.1 $\mu g/m^3$
CFPM	171.4 $\mu g/m^3$
Mass median diameter	0.5 μm
SO_4^{-2}	20.6 $\mu g/m^3$
NO_3^-	6.4 $\mu g/m^3$
Visibility range	7.1 km
Relative humidity	55%
NO_2	0.07 ppm

* Atmospheric pressure was 580 mn and 20°C temperature.

Table 3

Average results for the determinations performed at the ENCBIPN Station during January-September, 1982

Parameter	Average result*
TSP	204 $\mu g/m^3$
CFPM	154 $\mu g/m^3$
Mass median diameter	0.45 μm
C_t	49.87 $\mu g/m^3$
C_b^1	16.9 $\mu g/m^3$
C_{ov}^2	32.0 $\mu g/m^3$
Visibility range	6.3 km
Relative humidity	58%

[1] After adjust of data with regression line forced through zero between TSP and C_b.

[2] After equation $C_t = C_b + C_{ov}$.

* Atmospheric pressure was 580 mmHg and 20°C temperature.

Table 4

Summary of statistical analyses to data at the ENCBIPN station

Parameters[1]	Least squares fit	r^2
C_t vs. TSP	$C_t = 8.727 + 0.288$ TSP	0.81
C_b vs. TSP	$C_b = 1.0086 + 0.08819$ TSP	0.86
C_{ov}^2 vs. C_t	$C_v = 5.705 + 0.7573\ C_t$	0.80
C_{ov}^2 vs. TSP	$C_v = 3.1798 + 1673$ TSP	0.94

[1] all parameters in $\mu g/m^3$

[2] $C_{ov} = C_t - C_b$.

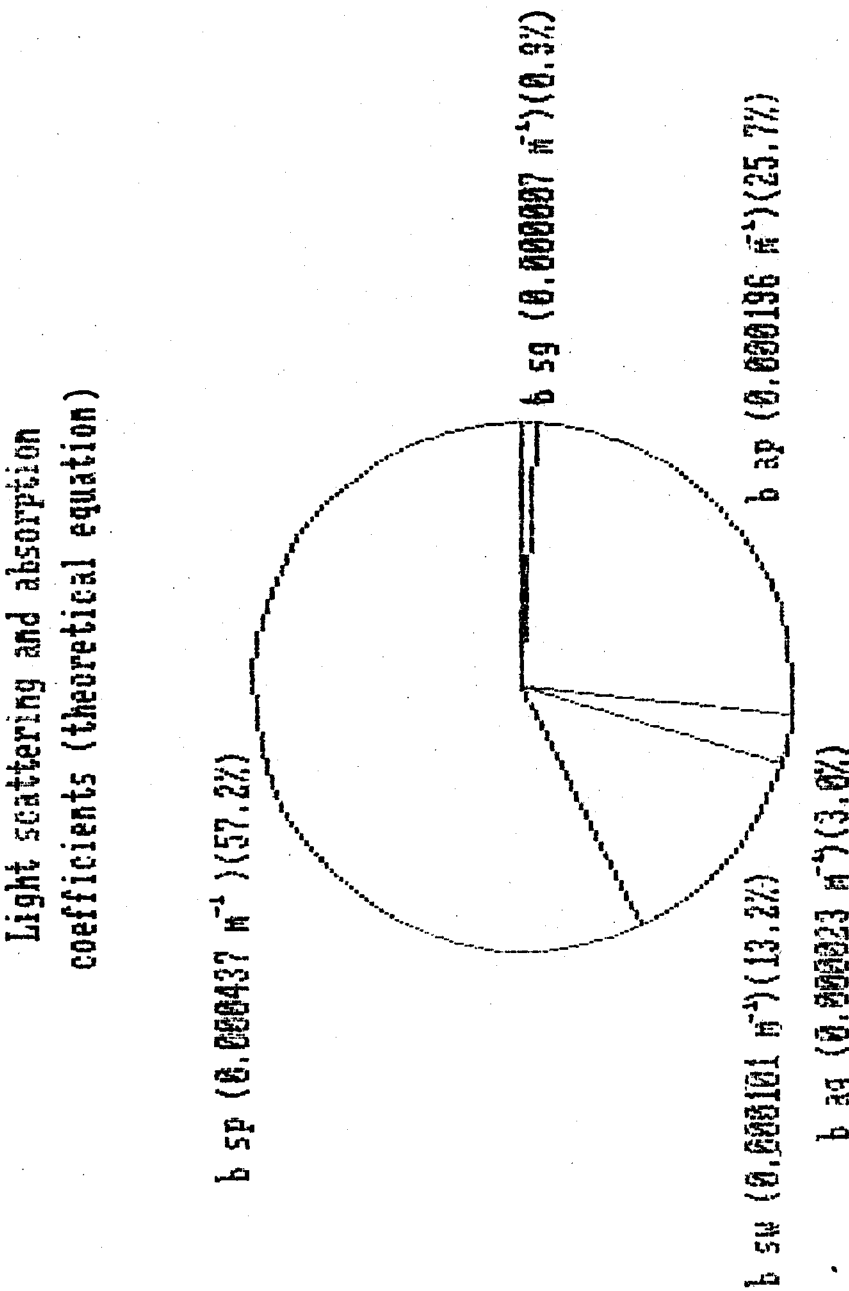

Fig. 6. Estimated coefficients contributions to light extinction coefficient in the Mexico City Metropolitan Zone.

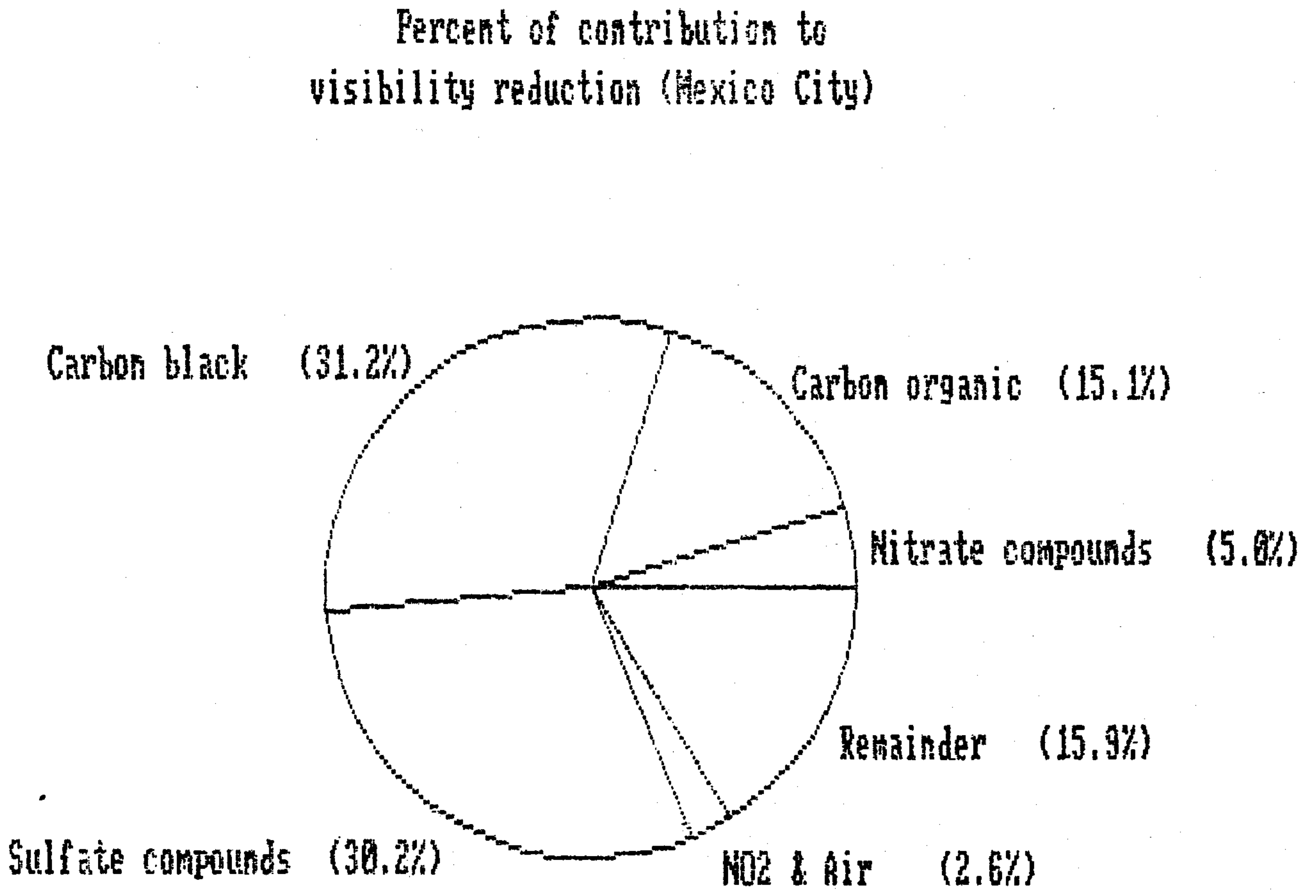

Fig. 7. Estimated contributions by chemical species to light extinction coefficient at the ENCBIPN station in the Mexico City Metropolitan Zone.

Table 5

Summary of light extinction budgets (percent contributions of various species) adapted from Latimer *et al.* (1985)

Species	Denver Winter haze (1978)[a]	California sites (1982)[b]	Lennox California (1980)[c]	Duarte California (1980)[c]	Downtown Los Angeles (1982)[d]	Nonurban Arizona (1981)[c] Clear day	Nonurban Arizona (1981)[c] Hazy day	Downtown Mexico City*
Fine particles	12	6	3	3	—	—	—	—
Sulfate	20	30	53	31	31	11	33	30
Organics	13	—	13	23	19	20	30	15.1
Elemental carbon (soot)	38	20	14	21	31	11	12	31.2
Nitrate	17	36	4	1	13	0	0	5.0
Crustal	—	—	—	—	7	4	2	—
Others	—	—	6	15	—	30	8	15.9
Coarse particles	—	—	—	—	—	24	15	—
Nitrogen dioxide	—	8	7	6	—	—	—	2.5

* Estimated by equation developed by Pratsinis *et al.* (1984) upon determination in Hi-Vol sampled filters.
a. Groblicki, Wolff and Countess (1981).
b. Appel *et al.* (1983)
c. Pratsinis *et al.* (1984)
d. Gray *et al.* (1984)
e. Macías, Zwicker and White (1984)

CONCLUSIONS

The most significant finding in this study is the role of C_t on visibility reduction. This result explains in a rough way the early find from Bravo *et al.* (1981) with regards to the low correlation between sulfates and nitrates on visibility in the Mexico City area.

The total carbon content in TSP is well correlated ($r_2 = 0.81$) following the relationship:

$$C_t = -8.727 + 0.288 \, TSP$$

The organic volatile carbon contents in C_t is near 67% and the black carbon contents in C_t is near 33%. The particulate elemental carbon contribute 46.3% to reduction of visibility in Mexico City.

The b_{ext} estimated empirically was $7.64 \times 10^{-4} m^{-1}$.

An updated study which will include C_t content in the fine fraction, as well as trends from 1982 through1987 for the visibility problem in Mexico City will be available from the authors in the spring of 1989.

ACKNOWLEDGEMENTS

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