Atmósfera (1993), 6, pp. 39 - 49

Some urban and meteorological effects on the production of cloud condensation nuclei in Mexico City

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(Manuscript received February 26, 1992; Accepted in final form June 17, 1992)

RESUMEN

Se llevaron a cabo mediciones de la concentración de núcleos de condensación de nube (CCN) al nivel del suelo, a intervalos de una hora, en una zona cercana al centro de la ciudad de México durante 19 días del verano de 1985. Para los muestreos, se utilizó una cámara cilíndrica de difusión térmica, operada a una sobresaturación fija de 0.75%, localizada a una altura de 2 m sobre el nivel del piso. Estas mediciones fueron analizadas en términos de su relación con los niveles de contaminación (NO, NO_2 y SO_2), concentraciones de partículas de aerosol y condiciones meteorológicas imperantes, haciendo uso de datos proporcionados por diversas fuentes. Los resultados indican que las fuentes antropogénicas, así como las condiciones meteorológicas locales, determinan los patrones diarios de la concentración de CCN en la ciudad de México.

ABSTRACT

Series of ground-based measurements of cloud condensation nuclei (CCN) concentrations were performed at one-hour intervals near downtown Mexico City during nineteen days in the summer of 1985. The samplings were made with a cylindrical, thermal diffusion chamber operating at a fixed supersaturation of 0.75% and located 2 m above ground level. These data were analyzed in view of concurrent contaminant levels (NO, NO_2, and SO_2), aerosol particle concentrations and meteorological conditions, using information gathered by diverse agencies. The results indicate that the anthropogenic sources, along with the local meteorological conditions, determine the daily patterns of CCN concentrations in Mexico City.

1. Introduction

It has been recognized for some time that anthropogenic sources for the production of aerosols play an important role in inadvertent weather modification (Changnon, 1968). In particular, several investigations have shown that air pollution causes modifications to chemical and microphysical characteristics of clouds and precipitation (Hobbs *et al.*, 1970; Hegg *et al.*, 1984; Pueschel *et al.*, 1986). These changes are due in part to the generation of cloud condensation nuclei (CCN) (Warner, 1968) and ice nuclei (Telford, 1960) by pollution sources, mainly through direct production, coagulation, gas-to-particle conversion and other mechanisms. These are complicated by other urban- and topographic-related effects (Huff and Changnon, 1986).

Mexico City, located at a height of 2240 m ASL, is confined to a valley formed by a great mountain chain of volcanic origin with an average height of 3000 m ASL. Every day, about 2.5 million vehicles and 35 thousand factories and small business establishments burn about 14 million liters of gasoline, 4 million liters of diesel, 3 million liters of natural gas and 6.5 million liters of combustoleo, a very poorly refined fuel oil. These emissions are added to hundreds of tons of particles that the wind carries from nearby eroded zones. These characteristics make Mexico City's atmosphere one of the most polluted in the world.

As part of a longer term program oriented toward understanding the influence of air pollution on the weather and climate of Mexico City and, in particular, its possible effects on the production of CCN, measurements of CCN concentrations were carried out between 22 July and 9 August, 1985. These measurements were analyzed in view of the meteorological conditions present during the sampling periods and compared to data on gaseous contaminants and aerosol particle concentrations gathered by diverse agencies. The results of these analyses are reported in what follows.

2. Intrumentation and methodology

The CCN samplings were made with a cylindrical, horizontal thermal diffusion chamber of the type reported by Gagin and Terliuc (1968). Specific details on the characteristics and operation of the chamber have been described by Herrera and Castro (1988). The CCN concentrations were determined using a semi-automatic optical scattering device (Montañez and Castro, 1989), with a photodetector placed at an angle of 157° with respect to a He-Ne laser beam used to illuminate the sampling section. Two-minute long samplings, performed 2 m above ground level with the chamber operating at a fixed supersaturation of 0.75%, were carried out at one-hour intervals between 0800 and 2000 LST on the grounds of the Subdirección de Investigación y Tecnología de Apoyo (SITA) of the Water Resources Ministry. The neighborhoods surrounding the SITA, which is located in a densely populated zone near downtown Mexico City, are residential and office areas with practically no industrial activity, though automobile traffic is often congested on nearby highways.

Simultaneous data on gaseous contaminants produced by the burning of fuel oil products[1] (NO, NO_2 and SO_2) were obtained with an NO_x Beckman analyzer (Model 952) and a Monitor Labs fluorescent SO_2 analyzer (Model 8850). These compounds were chosen for this study due to their characteristics as sources of aerosols by way of gas-to-particle conversion mechanisms. The combustion of gasoline in motor vehicules is considered to be the main source of NO_x, while SO_2 is mainly produced by the refining of crude oil and the combustion of heavier fuels (see, for example, Friedlander, 1977). Measurements of aerosol particle concentrations[2] carried out with an aerosol size analyzer (Thermo Systems Inc., TSI Model 3030 Electrical) were also available for the study period, with samplings being taken one, two or three times a day, except on 7 and 8 August when seven measurements were made on each day. The data set includes particle concentrations resolved in nine size-intervals, ranging from 0.003 to 0.5 μm in radius. These were grouped into two categories for analysis purposes: from 0.003 to 0.09 μm (Aitken nuclei) and from 0.09 to 0.5 μm (lower half-range of large nuclei). Both aerosol and gaseous contaminants samplings were performed on the main campus of the National University of Mexico, located 9 km to the south of SITA.

Meteorological data, which include radiosonde observations at 0600 LST from Mexico City International Airport and precipitation records from Tacubaya Observatory (located 3 km to the west of the CCN sampling site), were provided by the National Meteorological Service of Mexico.

Two points should be mentioned. First, the CCN, aerosol particles and gaseous contaminants data sets, as well as the meteorological records, were not all obtained at the same sampling site, so some caution should be exercised when interpreting the relationships found between them.

[1] Data provided by the Environmental Pollution Section of the Centro de Ciencias de la Atmósfera, Universidad Nacional Autónoma de México (UNAM).

[2] Data provided by the Solar Radiation Observatory of the Instituto de Geofísica, Universidad Nacional Autónoma de México (UNAM).

All three sites, though, have similar general characteristics in the sense that local anthropogenic aerosol sources are mainly vehicular traffic and commercial activities. Industrial activity takes place mainly in the northern end of the city, from which the dominant winds blow, roughly crossing the valley in the north-south direction. The general trends in the data are then expected to be very similar at the different sites considered. Second, in order to better understand CCN production due to human activities in the Valley of Mexico, it would have been desirable to obtain samplings upwind, inside and downwind of the city. Since this kind of data was not available, an average background value of CCN concentrations for air masses of continental origin was assumed for comparison purposes. This point will be further discussed in the following section.

3. Results and discussion

3.1 Emission Aspects

The results obtained throughout the study period showed concentrations ranging from 1,100 to 6,500 cm^{-3} for CCN, from 23,900 to 202,200 cm^{-3} for Aitken nuclei, and from 78 to 4,500 cm^{-3} for large nuclei. Concentrations of NO, NO_2 and SO_2 ranged from 0.01 to 0.137, 0.01 to 0.08 and 0.027 to 0.067 ppm, respectively. Measurements on 8 August demonstrate observed typical trends of the hourly variation in concentrations on a given day and are shown in Figure 1. Both CCN and large nuclei concentrations reached, in all cases, a maximum sometime between 0800 and 1100 LST, followed by a sharp decrease by midday. CCN concentrations reached a second, less pronounced maximum between 1700 and 2000 LST. For the case of large nuclei, it was not possible to observe the afternoon behavior in concentration due to the general lack of data in that part of the day. Two points should be stressed. First, it was generally observed that large nuclei concentrations were smaller than those of CCN at a given time. Second, the afternoon CCN maximum was not observed on three days over the study period, this being related to the rainfall events that occurred at the time (see Section 3.2).

In order to estimate the importance of urban-related effects on CCN production, the results obtained here can be compared with concentrations observed for different geographical areas. For example, Twomey and Wojciechowski (1969) report an average concentration between 200 and 800 cm^{-3} for long distance flights over continental areas. Thus, values above the latter can be considered here a result of anthropogenic emissions. Observations by other investigators in both urban and rural areas, made at ground level and for similar or even larger values of supersaturation, indicate that average CCN concentrations range from 800 to 5,300 cm^{-3} (Alkezweeny and Green, 1970; Sax and Hudson, 1981; Alofs and Liu, 1981; Hudson and Frisbie, 1991) and are dependent on seasonal, wind speed and other factors. Average concentrations reported here (see Table 1) fall within the higher end of the above-mentioned range. On the other hand, short interval (one minute to one hour) samplings by Hudson (1991) and Hudson and Frisbie (1991) show maximum concentrations between 5,000 and 20,000 cm^{-3}. It is surprising that, given the emission characteristics in Mexico City mentioned in the Introduction, maximum CCN counts observed in the present study were not even higher. This, along with the fact that minimum counts seemed fairly high, suggests that the sampling technique used here may require sample-dilution in order to prevent vapor depletion in the cloud chamber. Nevertheless, the typical diurnal variation of CCN concentrations shown in Figure 1 is in very good agreement with results obtained at the same sampling site by Herrera and Castro (1988), but for a smaller data set and in the spring season.

Figure 1 also shows that both NO and NO_2 presented maxima in concentration in the morning and afternoon hours. The morning peak was generally observed to occur earlier than that of

CCN, whereas the afternoon one occurred, in most cases, simultaneously. It is interesting to note that a very similar behavior for both NO and NO_2 has been reported in urban areas for *averages* over so-called "days of eye irritation" (Leighton, 1961). As for the SO_2, its concentrations showed an initial rise in the morning, then remaining fairly constant throughout most of the day, and slowly decreasing towards the late afternoon.

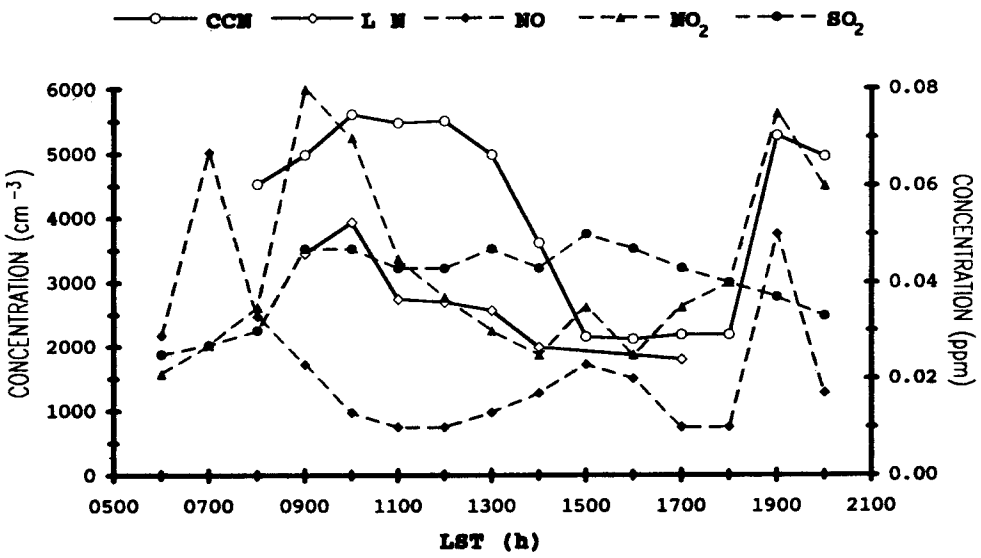

Fig. 1. Diurnal variation of concentration on 8 August 1985 in Mexico City: for CCN activated at 0.75% supersaturation and for large nuclei (LN) between 0.09 and 0.5 μm in radius (in cm^{-3}, left scale); and for NO, NO_2 and SO_2 (in ppm, right scale).

It is very difficult to distinguish between the effects on diurnal variations of concentration due to the atmospheric chemistry, in particular photochemical reactions, and those due to factors such as the varying rate of contributions of different sources of pollutants, air motions of the atmosphere, short-term variations in solar radiation and so on. Such a detailed analysis would go beyond the scope of the present investigation. However, some insight can be gained from the hourly variation of trace gases concentrations presented in Figure 2 for the period 7-9 August. For all three gases, the increase in concentration starts at about 0500 LST following an overnight low trend. Thus, the morning peaks can be related to the beginning of most of the industrial and vehicular activity, as well as to the high degree of atmospheric stability which is promoted by the topography of the city and prevents the transport of contaminants outside the valley (see Section 3.2). Although no overnight data are available for CCN and large nuclei, the previous assertion may also apply to their behavior, specially if it is considered that typical urban CCN are produced immediately by anthropogenic processes (Hudson, 1991). The midday and early afternoon decrease in concentrations is related to convection generated by surface heating and by an increase in wind speed at this time of the day (Hudson and Frisbie, 1991), thus promoting the transport of aerosols to higher levels in the atmosphere, and may also be aided by a decrease in vehicular traffic. In particular, the rapid depletion of NO and the corresponding buildup of NO_2 right after sunrise are also closely related to nitric oxide-hydrocarbon photolysis. The afternoon peaks can be associated with a new rise in vehicular activity due to rush-hour. It should be noted that, occasionally, a night peak in NO and/or NO_2 concentration, for which no explanation is given, was also observed. On the other hand, the daily behavior of SO_2 seems to better follow industrial activity trends, given that the average nocturnal concentration is consistently smaller than the diurnal one.

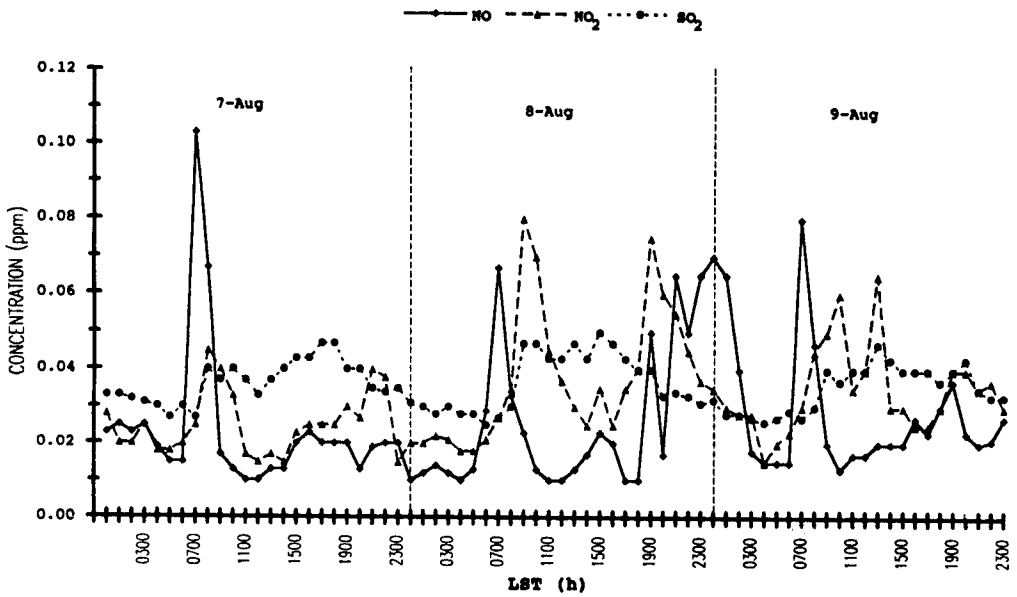

Fig. 2. Hourly variation in concentration for trace gases (NO, NO_2 and SO_2) for the period 7-9 August 1985 in Mexico City.

The relationships discussed above are further supported by detailed observations of the trends of hourly variations of vehicular traffic in the city. Figure 3 shows this for a particular case of one of the main highways in Mexico City, the Anillo Periférico, that crosses the valley in a roughly northwesth-southeast direction. It can be observed how the automobile flux at a given point sharply increases during the early hours of the day until reaching a maximum by 0900 LST, then slowly decreases and peaks up again by the afternoon rush-hour at about 1800 LST. It is interesting to note how well this diurnal trend follows that of the variations in concentration (Figure 1), in particular for CCN and NO_2.

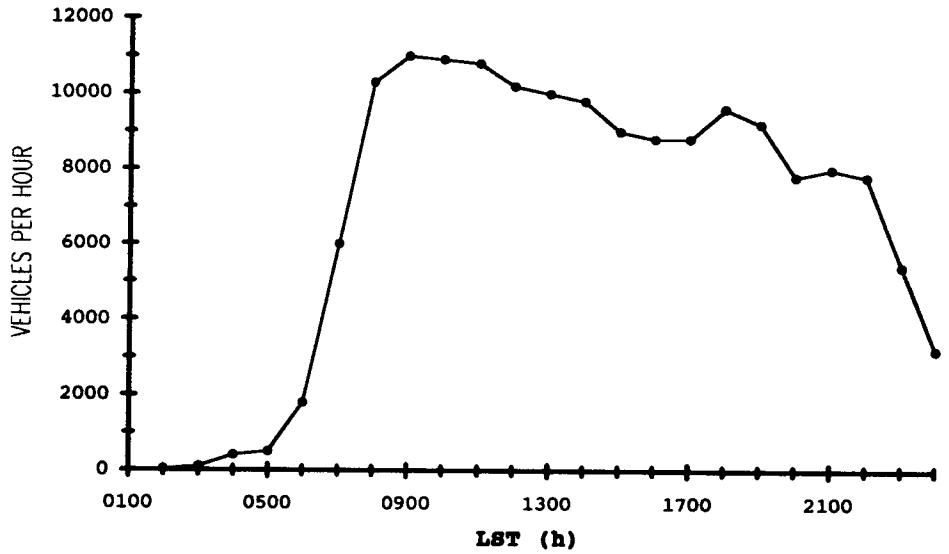

Fig. 3. Typical week-day hourly variation of vehicular traffic in the Anillo Periférico of Mexico City. The particular data shown correspond to 17 April 1987. (After J. I. C. A., 1988).

The hourly behavior for Aitken nuclei concentrations was observed to be more variable on a day to day basis and, for this reason, the corresponding data were not included in Figure 1. Nevertheless, the *average* diurnal variation of concentration for samples taken over the whole study period also seems to indicate a double-peak trend (Fig. 4), with the first maximum occurring by noon. Due to the lack of data (as in the case of large nuclei), it is not possible to establish at what time occurs the afternoon peak. Also, there is an apparent lag between the occurrence of the morning maxima for CCN and Aitken nuclei concentrations for which no plausible explanation has been found. This is in agreement with Hudson and Frisbie's (1991) observation that Aitken nuclei concentrations cannot be used as a surrogate for CCN concentrations. However, the observations give an indication that Aitken nuclei decrease in the afternoon and that this, as previously stated for CCN, may be due to increasing convection and wind speed and decreasing vehicular traffic.

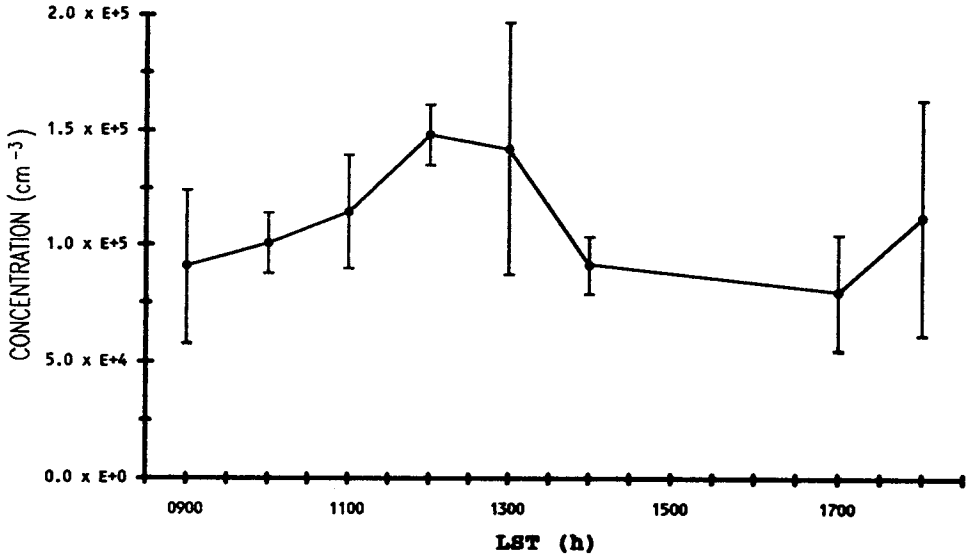

Fig. 4. Average diurnal variation of Aitken nuclei (particles with radii between 0.003 and 0.09 μm) concentration in Mexico City during 22 July to 9 August 1985. Error bars correspond to one standard deviation in the average.

The observed particle size distribution for the whole data set is shown in Figure 5. According to Junge (1969), the average size-spectrum for atmospheric aerosol particles larger than 0.1 μm in radius can be described by the following functional form:

$$\frac{\Delta N}{\Delta r} = cr^{\alpha}, \tag{1}$$

where $\Delta N/\Delta r$ (in $\mathrm{cm}^{-3}\ \mu\mathrm{m}^{-1}$) is the particle concentration per size interval, r is the particle radius (in μm), and c and α are constants. A least-squares fit performed on Eq. 1 for $r \geq 0.09$ μm (Fig. 5) indicates that Junge's functional form satisfactorily describes the present data set, including values of the radius down to 0.05 μm. Thus, Eq. 1 can be written in a more standard form as:

$$\frac{\Delta N}{\Delta r} = n_o \left[\frac{r_o}{r}\right]^{\alpha}, \tag{2}$$

where N (in cm^{-3}) is the concentration of particles larger than r, r_o (= 0.05 μm) is the minimum value of r for which Eq. 2 holds, and n_o (= 198,789 cm^{-3} $\mu m^{-1} \pm 10\%$) is the aerosol particle concentration at r_o. The value found for α (= 3.0 ± 0.1) from the curvefit indicates that, in the present study, the emission sources for larger particles are dominant. Typical average values of α for air masses of continental origin are 4, while for very clean tropospheric air masses are approximately 5. For values of $r < 0.05$ μm, the concentration increases at a slower, non-uniform rate as the particle size decreases. This is in general good agreement with average spectra reported in the literature (see, for example, Junge, 1952) except for particles in the size-interval from 0.003 to 0.005 μm, whose concentrations seem to be consistently too large. This deviation from the reported average spectrum may be related to a suspected overcounting of the aerosol size analyzer in that particular channel.

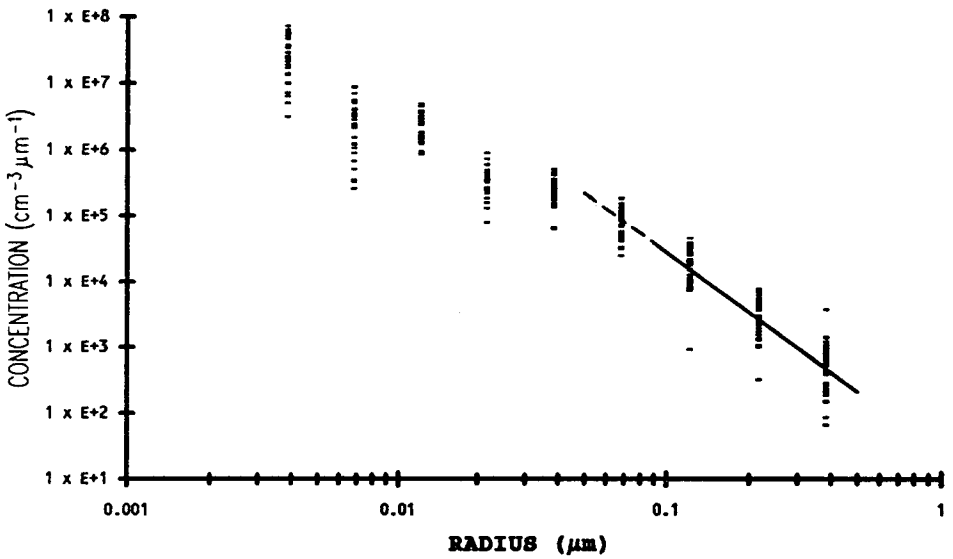

Fig. 5. Aerosol size distribution in Mexico City during 22 July to 9 August 1985. The continuous and dashed lines correspond to the curvefits given by Eqs. 1 and 2, respectively.

The concentrations of large nuclei ($r \geq 0.09$ μm) at a given time were found to be consistently lower than the corresponding ones of CCN (Fig. 1). Although not all large nuclei are expected to be CCN on a one-to-one basis, it must be remembered that, in the present data set, large nuclei only include particles between 0.09 and 0.5 μm in radius. Assuming that Eq. 2 holds for all values of $r \geq 0.09$ μm, it can be calculated that the contribution of particles in the range $0.09 \leq r \leq 100$ μm is not enough to even match the observed CCN concentrations. Thus, it is concluded that some of the smaller aerosol particles must also be CCN at the supersaturation of 0.75% considered here.

A plot of CCN concentration for the whole data set as a function of the aerosol particle concentration in the range of values where Junge's power law was found to be valid ($0.05 \leq r \leq 0.5$ μm) is shown in Figure 6. With the aim of having a better understanding of the general trends, and in view of possible future applications for modelling purposes, it was decided to perform a power law curvefit to the data (also plotted in Fig. 6) as follows:

$$N_{CCN} = bN_{AP}^{\beta}, \tag{3}$$

where N_{CCN} and N_{AP} are, respectively, the CCN and aerosol particle ($0.05 \leq r \leq 0.5$ μm) concentrations in cm^{-3}, and b (= 20.1) and β (= 0.6) are parameters of the fit (linear-correlation

coefficient of 0.51). A better understanding of this problem would, however, require an investigation on the detailed atmospheric chemistry, including reaction times, for the particular conditions of high altitude of Mexico City.

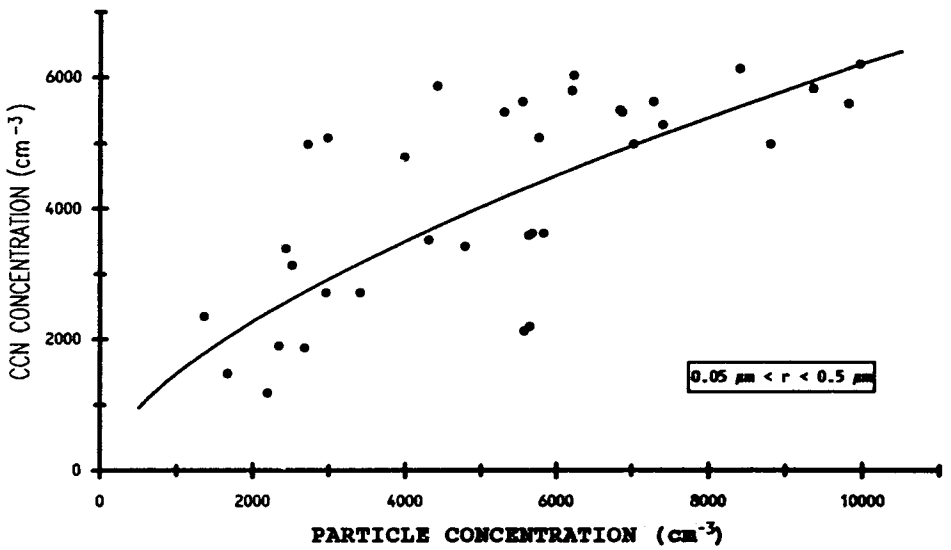

Fig. 6. Concentration of CCN activated at 0.75% supersaturation as a function of concurrent aerosol particle ($0.05 \leq r \leq 0.5$ μm) concentration in Mexico City during 22 July to 9 August 1985. The continuous line corresponds to the curvefit given by Eq. 3.

3.2 Meteorological Aspects

As mentioned in Section 3.1, there exist certain meteorological aspects which have important effects on the high concentrations of both pollutants and CCN found in Mexico City. In particular, it is well known that thermal inversions in the valley, along with high emmision rates, are directly responsible for critical pollution episodes in the city. Thermal inversions detected from radiosonde data in Mexico City International Airport were reported at 0600 LST on four

Table 1. Concentrations of CCN activated at 0.75% at 0800 LST on the four days when thermal inversions were reported at Mexico City International Airport (2240 m ASL) during the sampling period. Note that three out of the four days the inversion bottom was located at ground-level. The average CCN concentration at 0800 LST for the rest of the study period was (4849 ± 555) cm^{-3}.

DATE	CCN	INVERSION TOP		INVERSION BOTTOM	
	CONCENT. (cm^{-3})	HEIGHT (m ASL)	TEMP. (°C)	HEIGHT (m ASL)	TEMP. (°C)
26 July	6193	2360	13.6	2240	13.4
29 July	5997	3150	8.4	2880	8.0
6 August	6258	2330	12.6	2240	11.6
9 August	6095	2330	14.4	2240	13.2
All other	4849				

days during the sampling period. Concentrations measured at 0800 LST (the first sampling on a given day) on those days were compared against the average concentration in the remaining

days at that same time. The results indicate that limitations in the vertical displacement of the air masses allow contaminants to accumulate in the atmosphere near the ground. In particular, CCN concentrations were found to be up to 30% higher on inversion days than on the average of the rest of the sampling period (Table 1).

On the other hand, precipitation was observed to have the opposite effect of thermal inversions. Nine days with rainfall events occurred over the sampling period. In all nine cases a decrease in CCN concentration was noticed when rain was present, in three of which the afternoon maxima were suppressed (see Section 3.1). A typical case is shown in Figure 7. The anticorrelation of CCN with rainfall is evidence for the scavenging of the former via several mechanisms associated with the formation and development of clouds and precipitation (nucleation, coalescence and direct impaction scavenging) and related meteorological conditions (relative humidity, wind, etc.).

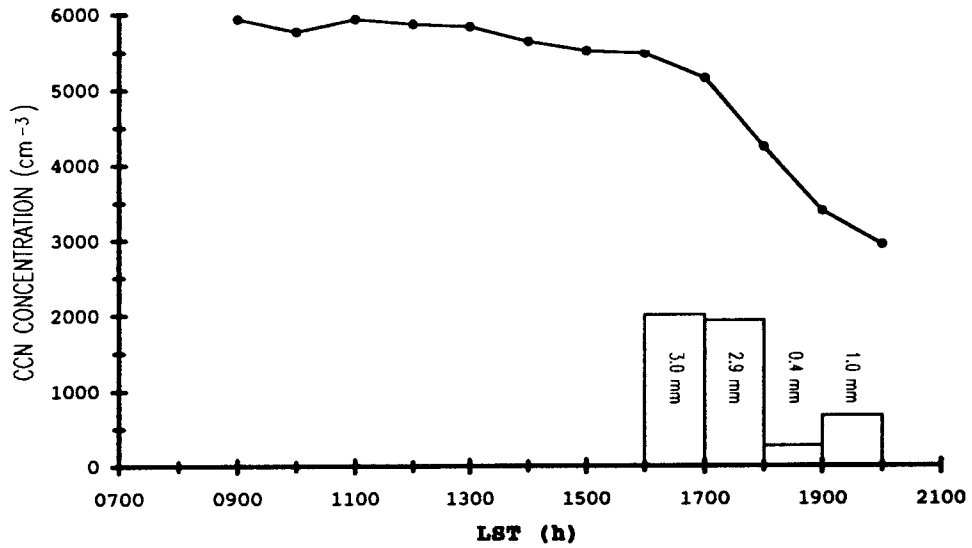

Fig. 7. Diurnal variation of concentration of CCN activated at 0.75% supersaturation on 30 July 1985 in Mexico City. Vertical bars represent concurrent, hourly precipitation recorded at Tacubaya Observatory.

4. Conclusions

Ground-level measurements of CCN, aerosol particles and gaseous contaminants were carried out in Mexico City during nineteen days in the summer of 1985. The main purpose was to understand the impact of pollution and some meteorological factors on the production of CCN in the city. The most important results can be summarized as follows:

1. The diurnal variations of CCN, large and Aitken nuclei, NO and NO_2 concentrations are found to present a behavior which is related to vehicular and industrial activities. The observed persistent morning maxima are also an effect of the high degree of atmospheric stability in the valley, whereas the sharp decrease in concentration around midday is aided by surface heating and an increase in wind speed which promote convection and the transport of aerosols to higher levels in the atmosphere. In particular, the rapid depletion of NO and the corrresponding buildup of NO_2 right after sunrise are also closely related to nitric oxide-hydrocarbon photolysis. For the case of CCN, NO and NO_2, the observed afternoon peaks can be associated with a new rise in vehicular activity due to rush-hour. The daily

behavior of SO_2 seems to better follow industrial activity trends.

2. The behavior of CCN concentration with time of the day is similar to that found in previous observations made in Mexico City during the spring season (Herrera and Castro, 1988).
3. The observed CCN concentrations are much higher than *background* values reported in the literature for air masses of continental origin, and similar to averages found for polluted regions at ground level.
4. Thermal inversions promote an increase in CCN concentration, whereas an opposite effect is observed during precipitation events.
5. The aerosol particle size-distribution found here is satisfactorily described by Junge's power law (Eq. 2) for a polluted atmosphere.
6. A power law relationship (Eq. 3) is proposed between concurrent CCN and aerosol particle ($0.05 \leq r \leq 0.5$ μm) concentrations.

The results presented in this paper indicate the interplay between the production of CCN and the local urban effects and meteorological conditions in Mexico City. A better understanding of the problem should include simultaneous observations upwind, inside and downwind of the city, as well as detailed analyses of the CCN chemical composition and the atmospheric chemistry, including the important effects that high altitude has on the different processes. This, in turn, would help to better determine the repercussions that air pollution may have on the weather and climate of the valley, besides the already disturbing problem of human health.

Acknowledgements

The authors thank Dr. H. Bravo for supplying the contaminants data, and Ms. S. Salazar and Mr. A. Muhlia who supplied the aerosol data. Drs. C. Gay and J. G. Hudson made comments which helped to greatly improve the manuscript. This study was partially supported by the Subdirección de Investigación y Tecnología de Apoyo (at present Instituto Mexicano de Tecnología del Agua of the Comisión Nacional del Agua).

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