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The impact of megacity pollution on local climate and implications for the regional environment: Mexico City

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Abstract

We present calculations to estimate potential changes to the local climate and photochemistry caused by pollutants (gases and particles) produced in Mexico City, and the implications for the regional scale when pollutants are exported to surrounding regions. Measured aerosol optical properties are used in a 2-stream delta-Eddington radiative transfer model (Slingo and Schrecker, 1982. Quarterly Journal of the Royal Meteorological Society 108, 407–426) to estimate net radiative fluxes and heating rates, while photolysis rates for nitrogen dioxide and ozone are estimated from a much more detailed model (Madronich, 1987. Journal of Geophysical Research 92, 9740–9752). The presence of highly absorbing aerosols in Mexico City leads to a 17.6% reduction in solar radiative flux at the surface when an optical depth of 0.55 is considered. Photolysis rates for nitrogen dioxide and ozone are reduced between 18 and 21% at the surface, while an increase of between 15 and 17% is predicted above the boundary layer, for local noon calculations.The non-uniform vertical structure of aerosol concentrations observed (Pérez Vidal and Raga, 1998. Atmosfera 11, 95–108) plays a significant role in determining localized regions of heating, i.e. stabilization at the top of the boundary layer that results in a temperature increase of 0.4K h^{-1} at that level. The presence of a 200 m-deep aerosol layer at the top of the boundary layer results in vertical profiles of the photolysis rates that are significantly different from the case where the aerosols are uniformly distributed in the mixed layer. At the bottom of the aerosol layer (about 1 km above the surface), the rates are about 28% lower than when there is a uniform aerosol distribution in the boundary layer. Finally, there is also an enhancement of photolysis rates at the top of the boundary layer that may lead to increased ozone production compared to the non-aerosol case.

Keywords: Mexico City air quality; Aerosol optical properties; Regional impact of megacity pollution; Aerosol–photochemistry interactions

1. Background

Atmospheric pollution in Mexico City has been a significant problem for more than a decade, due to the unrestricted growth of population and industry. The photochemical smog combined with high levels of primary particles are the result of emissions by vehicles (about 3.4 million), industry and five thermoelectric plants located within the elevated basin (2.2 km above sea level), as discussed in PROAIRE (1996). Recent studies by Edgerton et al. (1999), Raga et al. (1999a, b) and Baumgardner et al. (2000) have started to provide some insight on the composition and microphysical properties of fine particles within Mexico City, which appear to be quite different than those present in other North-American cities.

There is increased evidence that pollutants emitted or photochemically produced in large urban areas can be transported out of the source regions into surrounding, less urbanized areas (NAS, 1991; Central US, Ball and Robinson, 1982; Los Angeles pollution along the California coast, Strawbridge and Hoff, 1996; EPA, 1996). The

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transported pollutants can increase background concentrations in areas that would have been considered relatively pollution-free.

Due to Mexico City's tropical location the height of the boundary layer may reach up to 2 km above the surface, higher than the mountain passes that surround the basin. It is therefore likely that pollutants emitted within the city will be able to move out of the basin. Moreover, numerical simulations by Fast and Zhong (1998) and Whiteman et al. (2000) indicate that there are several paths by which the basin cleans itself daily. Passive tracers included in the simulations can be found 200–300 km away from the basin at 5 km above sea level.

With the use of a radiative transfer model we study the role of aerosol particles in changing tropospheric stratification and determine photolysis rates for nitrogen dioxide and ozone for a variety of aerosol characteristics. Furthermore, we attempt to estimate the impact of pollution exported from Mexico City into the regional scale and to assess its role in possible changes to the regional climate.

2. Measurements

The urban area that includes Mexico City and neighboring counties (considered a megacity, with about 18 million inhabitants) currently covers approximately 1200 km^2. A total of 5567 ton yr^{-1} of PM-2.5 (particulate matter with sizes smaller than 2.5 μm) are emitted within the megacity (IMADA, 1998), with 4704 ton yr^{-1} (84%) linked to vehicle emissions. Analysis of composition obtained during an intensive campaign carried out in February–March 1997 (Edgerton et al., 1999), indicated that 15–20% of the mass corresponds to elemental carbon and 25–30% to organic carbon. Assuming that these particulate emissions can be uniformly distributed in space over the urban area as well as in time, an average concentration of 8.5 μg m^{-3} can be estimated for a 1.5 km mixed layer height.

A pilot project was conducted during a two-week period in November 1997 to characterize the microphysical and optical properties of small particles and their relationships to other gaseous pollutants (Raga et al., 1999a, b; Baumgardner et al., 2000). This was part of an ongoing collaborative effort between Universidad Nacional Autónoma de México (UNAM) and the National Center for Atmospheric Research (NCAR) to study aerosol particles in Mexico City. The measurement site was located on the Southwest edge of the city, within an ecological reserve (19°15′N, 99°11′W), at an elevation of 440 m above the mean basin level of 2240 m. Measurements were made of the following gaseous pollutants: carbon monoxide (CO), sulfur dioxide (SO_2), nitrogen oxides (NO, NO_y) and ozone (O_3). Maximum concentrations for these pollutants vary significantly during the 2-week observing period, and for example, O_3 daily maxima ranged from 90 to 270 ppbv (Baumgardner et al., 2000).

Airborne particles were measured with a variety of instruments to characterize their size distribution, including cascade impactors and automatic counters. The cascade impactors included a 10-stage micro orifice uniform deposit impactor (Moudi), for sizing particles from 18.0 down to 0.056 μm (in aerodynamic diameter) and a 7-stage Andersen impactor, for sizes between 10.0 and 0.45 μm. Samples from the Moudi were analyzed for carbon content, while those from the Andersen were analyzed for inorganic ions. A differential mobility analyzer (DMA) and an active scattering aerosol spectrometer probe (ASASP) were used to sample size distributions automatically in the range from 0.01 to 3.0 μm (Baumgardner et al., 2000). Particle optical properties were determined with a 3-wavelength nephelometer and a soot photometer and measurements were also made of visible and ultraviolet irradiance. Fig. 1 shows the extinction coefficient (C_{ext}) and the single scattering albedo (ω_0) for the 2-week period, for a wavelength of 0.5 μm. The extinction coefficient shows large variability with a few instances of very low values (12–14 November). The single scattering albedo also shows very large variability during the same period. An overall average value of $\omega_0 = 0.8$, indicates the large fraction of absorbing material present in particulate matter in the city. These measurements are consistent with observations of elemental carbon in particles collected in filters (Edgerton et al., 1999; Baumgardner et al., 2000). When the 12–14 November period is excluded, then the average

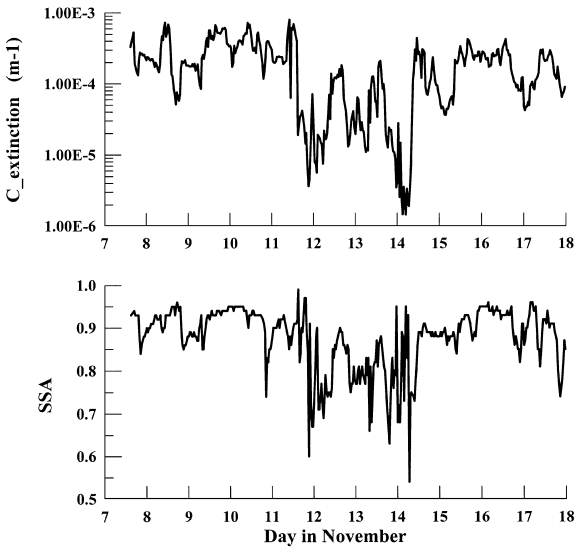

Fig. 1. Time series of (a) the extinction coefficient (C_{ext}) at a wavelength of 0.5 μm and (b) the single scattering albedo (ω_0) at the same wavelength, for the 2-week period in November 1997.

ω_0 is closer to 0.9. These values highlight the differences in particle characteristics between Mexico City and other polluted areas in North America (such as the Los Angeles basin or the Eastern US seaboard, where ω_0 is ~ 0.95 or higher). These gas and particle measurements are used as input in the numerical calculations described in the following section.

3. Results

3.1. Effect of particles on local climate

We use a multiple scattering radiative transfer model in which aerosol layers are included to analyze the optical impact of these aerosols on the climate of regions immediately downwind. The code uses a two-stream delta-Eddington approximation to solve the radiative transfer problem in the atmosphere, and includes Rayleigh scattering and absorption by water vapor and ozone, in the range from 0.25 to 4.0 μm (Slingo and Schreker, 1982).

The observed optical properties at the surface for 15 November 1997 were used as initial conditions for the simulations. In particular, the observed aerosol scattering coefficient, ranged from a minimum of $3.12 \times 10^{-5}\,m^{-1}$ to a maximum of $3.98 \times 10^{-4}\,m^{-1}$, with an average value for the 2-week period of 1.42×10^{-4}, while the corresponding values for the absorption coefficient, $\sigma_{a,}$ were 0.48×10^{-5}, 5.89×10^{-5} and $2.23 \times 10^{-5}\,m^{-1}$, respectively. These values translate into a range of ω_0 from 0.346 to 0.928. Since no measurements of the vertical profiles of particle optical properties have been made in Mexico City, we used the surface values throughout the boundary layer. Fig. 2 shows the vertical profiles of extinction coefficient, water vapor and ozone concentrations used in the calculations. Two distinct cases were studied, one with a uniform aerosol layer in the boundary layer and the other with an aerosol layer at the top of the boundary layer, consistent with observations presented by Perez Vidal and Raga (1998). The boundary layer was kept constant for the simulations, with a height of 1.2 km. The vertical profiles of ozone and water vapor correspond to values observed from an instrumented aircraft during a field campaign to study the ozone problem in Mexico City (EGCA/MARI, 1994). The profile used corresponds to the aircraft ascent on 13 February 1991 at 8:45 a.m. (local time).

Table 1 shows some of the cases that were simulated. The optical depth, τ, ranged from 0.04 to 0.55, corresponding to a wavelength of 0.5 μm and was considered to be independent of wavelength in the simulations. These values are much smaller than spectral optical depths measured in Mexico City during June–July 1992

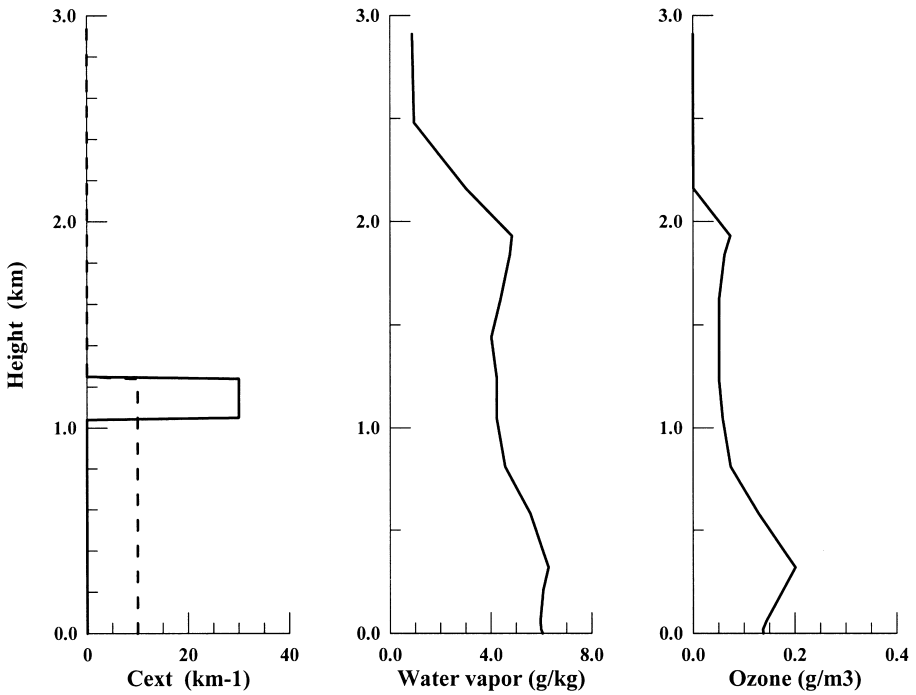

Fig. 2. Vertical profiles of (a) extinction coefficient (C_{ext}), (b) water vapor mixing ratio and (c) ozone concentrations used in the simulations. The profiles shown in (b) and (c) were obtained from an aircraft ascent in Mexico City on 13 February 1991 (8:45 a.m., local time).

Table 1
Extinction coefficient (σ_{ext}), single scattering albedo (ω_0) and optical depth (τ) used in the different cases simulated. The asymmetry factor was 0.176 for all cases. Runs were also performed with a value of 0.035, but no significant differences were observed from the results presented here

Case	σ_{ext}	ω_0	τ
(I) Uniform	0.33×10^{-4}	0.864	0.04
(II) Uniform	1.64×10^{-4}	0.864	0.20
(III) Uniform	4.58×10^{-4}	0.871	0.55
(IV) Only BL top	2.75×10^{-3}	0.871	0.55
(V) No aerosol	—	—	—

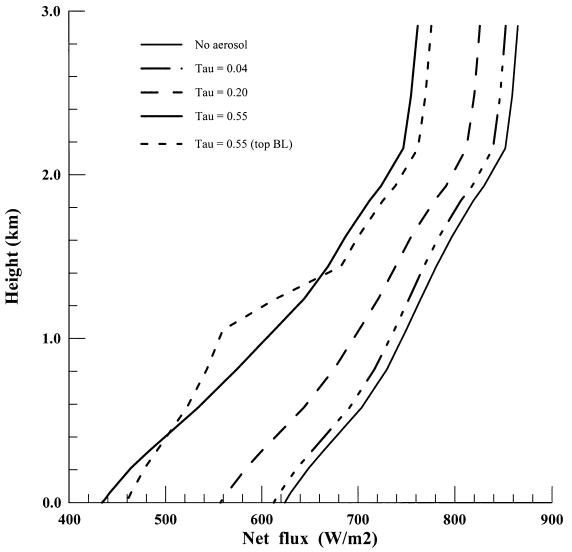

Fig. 3. Vertical profiles of the predicted net flux for an aerosol-free case (light solid line) and 4 different cases of uniformly distributed aerosols within the boundary layer ($\tau = 0.04$, 0.20 and 0.55). The last case shows the results when the aerosol layer is concentrated in the top 200 m of the boundary layer ($\tau = 0.55$).

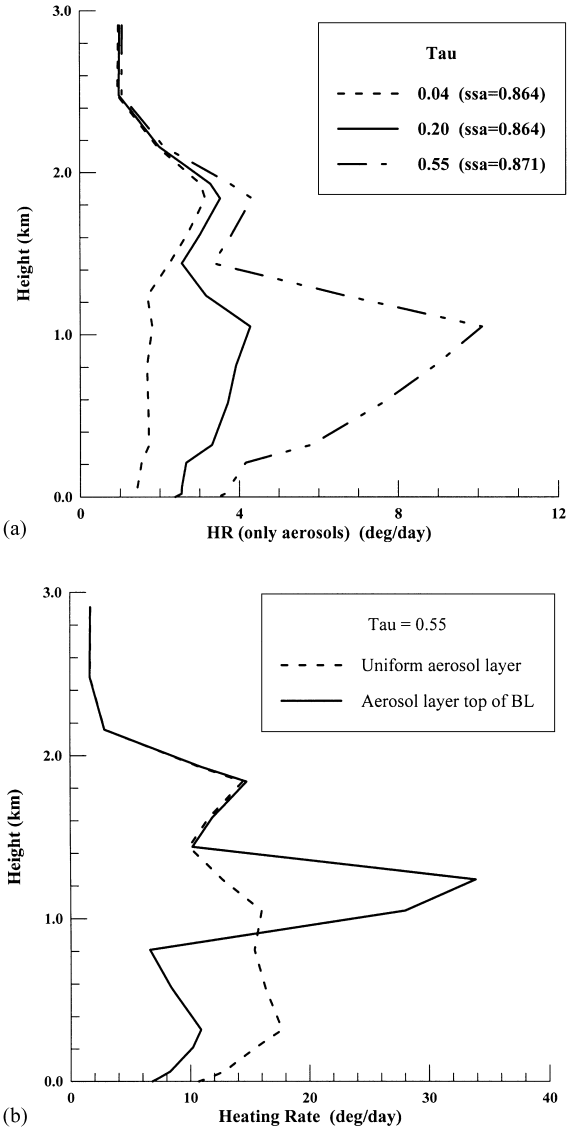

Fig. 4. Vertical profiles of the predicted heating rate, (a) due only to the presence of aerosols and (b) due to aerosols and gases, comparing the cases of uniform vertical distribution and concentrated aerosol layer at the top of the boundary layer.

(Vasilyev et al., 1995), which present values up to 1.0 for wavelengths near 0.4 μm.

Fig. 3 displays the calculated net fluxes as a function of height for τ increasing from 0 to 0.55 and aerosols distributed uniformly in the boundary layer. One example is also given of the case where the aerosol layer is only at the top of the boundary layer. It is interesting to note that for the same aerosol optical depth, the concentrated layer at the top of the boundary layer, results in an 8.8% reduction of the net flux just below the aerosol layer, but a slight increase in the surface value. For the case of an optical depth of 0.55, a 30.4% reduction at the surface is observed when compared to the aerosol free case. A slightly less reduction (26.4%) is observed when the aerosol layer is only 200 m deep at the top of the boundary layer.

Fig. 4 presents the vertical profile of the heating rate for several simulations. The aerosol contribution to the heating rate (Fig. 4a) can reach up to 8K day^{-1} at the top of the boundary layer for the case of 0.55 optical depth. This is due only to aerosols, uniformly distributed in the vertical. In Fig. 4b, the heating rate profiles are the result of gases (H_2O and O_3) as well as uniformly distributed aerosols. A concentrated heating results from the presence of an aerosol layer at the top of the boundary layer.

While the integrated vertical rate is similar in both cases, the elevated aerosol layer has a more pronounced effect on the stabilization of the boundary layer capping inversion. This localized heating corresponds to a temperature increase of 0.4K at the top of the mixed layer after 1 h ($\tau = 0.55$). The increased stability at this level tends to inhibit the penetration of turbulent eddies through it, leading to a subsequent decrease in mixed layer growth rate. This effect was first suggested in the numerical simulations by Zdunkowski et al. (1976), Ackerman (1977) and more recently by Jacobson (1998). An increased stability at the top of the inversion increases the lifetime of particles and creates the potential for farther transport from the city than would occur otherwise. While remaining at that level, particles would also have more chance to interact with solar radiation and potentially affect photochemistry, as discussed in the following section.

3.2. Effect of particles on local photochemistry

The increased actinic flux within an aerosol layer at the top of the mixed layer promotes faster photochemical reactions at that level compared to near the surface. We test this hypothesis with the model developed by Madronich (1987), which uses a very detailed radiative transfer calculation to determine the photolysis rates for NO_2 and O_3, in the presence of an aerosol layer. Fifteen cases were studied that covered the observed range of C_{ext} and ω_0, resulting in optical depths between 0.04 and 0.55. As discussed above, aerosols were either assumed to be uniformly distributed in the vertical or in a more concentrated layer (200 m in thickness) at the top of the boundary layer. The surface albedo was set to 0.10, consistent with measurements made in different locations within the city in 1995 (Castro and Mar, personal communication). The calculations were carried out for noon conditions on 15 November and ozone concentrations observed that day were also used as input for the model. The results presented in Fig. 5, are in agreement with measurements of J-NO_2 made earlier (Ruiz-Suarez et al., 1993; Castro et al., 1997) and, as expected, indicate that the presence of absorbing aerosols decreases the actinic flux reaching the surface. A decrease at the surface of 18.6% in NO_2 photolysis rate is observed when $\tau = 0.55$ is included, compared to the non-aerosol case. This observed τ can be considered somewhat conservative for UV wavelengths, based on earlier measurements by Vasyliev et al. (1995). Similarly, a 21.7% decrease in the O_3 photolysis rate is calculated for the same case. These modeled reductions in NO_2 and O_3 photolysis rates at the surface would result in a decrease in O_3 formation at the surface, which has been also discussed by Dickerson et al. (1997) and Jacobson (1998). The presence of aerosols in the boundary layer results in increased values of the photolysis rates by 16.9 and 14.9% at the top of the boundary layer (NO_2 and O_3, respectively for local noon), as was pointed out by Madronich (1987) in the case of a cloud layer. The differences observed in the photolysis rates when the vertical structure of the aerosol layer is considered (uniform within the boundary layer vs. a concentrated layer at the top with the same τ), is restricted to the boundary layer. Both cases result in the same increase in the actinic flux at the top of the boundary layer and decrease at the surface compared to the non-aerosol case. On the other hand, the vertical profiles within the layer are very different, with photolysis rates much lower (27.4 and 28% for NO_2 and O_3, respectively) at the bottom of the aerosol layer than when the aerosol is distributed uniformly.

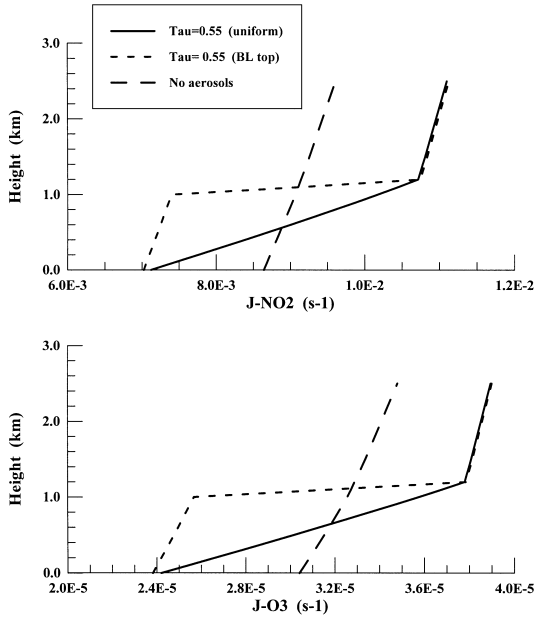

Fig. 5. Vertical profiles of the predicted photolysis rates of (a) NO_2 and (b) O_3, for an aerosol-free case (long dash line), as well as a uniformly distributed aerosol layer (solid line) and a concentrated layer at the top of the boundary layer (short dash line).

4. Discussion

In this study, we have presented radiative transfer calculations applied to conditions observed in Mexico City. The range of optical depths considered can be used as a surrogate for the polluted air mass as it leaves the basin and is diluted with cleaner air at the regional scale. The presence of an aerosol layer can have a significant impact on the vertical profile of the heating rate that leads to a stabilization of the thermodynamic profile. Elevated aerosol layers, as have been observed in Mexico City in the past (Perez Vidal and Raga, 1998), could

strengthen the boundary layer capping inversion, slow its growth and further concentrate pollutants. The calculations indicate that a heating rate of 0.4K h^{-1} would be consistent with the presence of an aerosol layer at the inversion level. This strengthens the lapse rate at the inversion, with an associated reduction of the turbulent exchange between the boundary layer and the free atmosphere. Since these heated aerosols would tend to remain at the inversion level longer, exchange of pollutants between the boundary layer and the free atmosphere would still occur. As mentioned above, Mexico City is located at 2.2 km above sea level and mixed layers extend to 2 km, even during the winter. Therefore, pollution is introduced at altitudes that are considered free troposphere only 300 km away. Thus, it is reasonable to assume that gases and particles will have longer lifetimes in this environment as they move out from the city than if they had been introduced by cities at sea level. The dry season (typically from November to May) is characterized by a westerly component of the upper level winds such that Mexico City pollutants most likely are carried towards the East. There is some evidence from satellite-derived aerosol optical depths (Husar et al., 1997) that the pollution plume from Mexico City appears as a local maximum over the Gulf of Mexico. A fairly complex wind circulation is observed within the Mexico City basin, as described in Doran et al. (1998). Numerical results by Whiteman et al. (2000) indicate that pollution generated in Mexico City reaches the coasts. Our calculations quantify the effects of aerosol absorption on the local climate; however, it is clear that the impact of exported pollution would be on a regional scale, as also discussed by Gaffney and Marley (1998). The fate of Mexico City pollutants will be very different during the rainy season, when deep convection occurs daily and the upper level winds are predominantly Easterly. Deep cumulus clouds would transport pollutants from the boundary layer up to 10 km very rapidly. The interactions between gases and particles and cloud droplets are very complex and different processes can take place. We are currently starting to investigate these interactions using a detailed cloud microphysical model that also includes some chemical reactions.

The effect on local ozone formation of an aerosol layer was investigated using a detailed radiative transfer code (Madronich, 1987) that predicts the NO_2 and O_3 photodissociation rates. The presence of aerosols results in a decrease of the actinic flux at the surface and decreases in the photolysis rates of up to 21% compared to the non-aerosol case. Similarly, an increase in the rates of up to 17% at the top of the boundary layer was also predicted. The effect of the aerosol layer concentrated only at the inversion level (200 m in depth) is to modify significantly the vertical profiles of the photolysis rates and decrease these rates to $\sim 27\%$ at the bottom of the aerosol layer when compared to the case of uniformly distributed aerosol. These vertical profiles affect ozone production rates, which will not be uniform within the layer and have a maximum that occurs higher in the layer, as indicated by Raga and Raga (2000).

Finally, there are significant implications for photochemistry in the regional scale. Once pollutants move out of the city (where the main sources are located) there is potential for ozone formation in the lower nitrogen oxides regime (Madronich, personal communication). The ozone photodissociation leads to formation of hydroxyl radicals involved in the oxidative decomposition of most trace gases in the troposphere. Gaffney et al. (1999) made measurements of peroxyacyl nitrates (PANs) in Mexico City and concluded that the daily "clean-up" of the basin would necessarily imply increased concentrations in the surrounding areas. Using a very simple model, they estimate about 750 ton day^{-1} of PANs and 3000 ton day^{-1} of O_3 exported from Mexico City. These regions surrounding the city would be clearly influenced by these (and other) pollutants and there is a need for improved measurements in the regional scale to determine the true impact of the exported pollutants. As a first step, a field campaign will take place in Mexico City in February 2000 to further characterize aerosol properties and to measure the actinic flux. A follow-up campaign is planned for January–February 2001 to measure pollutants in the regional scale, which will help answer some of the issues raised in this study.

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