Ozone and Aerosol Distribution Measured Above Mexico City with a Differential Absorption Lidar during the MCMA 2003 Field Campaign

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Abstract. Ozone and aerosol vertical distribution and time evolution were measured with a UV DIAL elastic lidar during the MCMA 2003 field campaign. The diurnal evolution of the PBL and ozone were also studied. Formation of a residual layer containing elevated ozone concentrations at nighttime, as well as detachment of the PBL in the late afternoon hours were observed.

Introduction

Mexico City Metropolitan Area (MCMA) is one of the cities in North America most affected by air pollution [Molina]. A high level of Total Suspended Matter (TSP) is the most prevalent form of pollution followed by ozone, sulfur dioxide, and carbon monoxide. A number of factors contribute to the high pollutant levels in the city but the most important are: overpopulation (over 21 million), stagnation of the air because of the U-shaped valley location, high altitude and abundant solar radiation.

As part of the efforts to understand the complex pollution problems of the city, an international field measuring campaign (MCMA 2003) was held in April -May 2003. A number of primary and secondary pollutants, as well as aerosol properties and distribution were measured intensively during the campaign. An important role played lidars since they are the only instruments allowing simultaneous study of boundary layer dynamics and photochemical processes by monitoring the vertical distribution of ozone and aerosols.

The EPFL (Ecole Politechnique Fédéral de Lausanne) team participated with a mobile UV DIAL - 532 nm elastic lidar system. The lidar data together with the results from the ground based instruments, and the synoptic and meteorological information will be used to model the evolution of the pollution episodes in the MCMA.

Experiment

A detailed description of the lidar is given in [Simeonov 2002, Simeonov 2004]. Here we present only the salient features of the system. The transmitter is based on a quadrupled Nd:YAG laser, whose second harmonic (532 nm) is employed for aerosol measurements. DIAL wavelengths are generated from the fourth laser harmonic (266 nm) by Raman conversion in N_2. First (283.4 nm) and second (303.7 nm) N_2 Stokes in combination with the residual 266 nm pump are used for ozone measurements. In the DIAL part of the receiver a dual-telescope configuration is employed to reduce the dynamic range of the signals. A 20 cm Newtonian type telescope equipped with a narrowband IF is used for the aerosol observations at 532 nm.

The campaign was designed to cover the maximum of the annual photochemical season. The atypical for the dry season rain episodes during the afternoons and the evenings at the beginning affected the pollution events and caused discontinuity in the observation. Improved meteorological conditions between 25th and 29th April made possible the realization of continuous measurements during an intensive observation period.

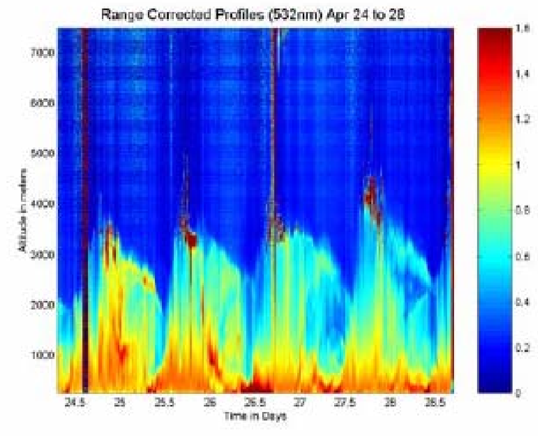

Fig. 1. 532 nm range corrected profiles.

A cloud-topped boundary layer (BL), probably due to high diurnal convection, was the most frequently observed event during this period. Aerosol measurements (Fig. 1) were taken to altitudes higher than 14 km at nighttime and up to 7 km AGL in daytime. The aerosol data at 532 nm shows formation during the evening of an important residual layer (RL) that persisted until noon of the next day when entrainment evolution reached its altitude. The top of the BL showed steady day-to-day evolution, reaching altitudes of up to 4 km, comparable to the altitudes of the surrounding mountains. During the last two days, a detachment of the top of the BL was clearly seen in the time series of the 532 nm range-corrected lidar signal (Fig.1).

Simultaneous ozone measurements were taken during the same period. For altitudes up to 2 km AGL the LIDAR was operated in the “solar blind” region using 266 and 283.6 nm wavelengths. Because of the strong attenuation of the 266 nm radiation, the wavelength pair 283.6/303.7 nm was employed for high altitude observations. The application of the pair was limited to a maximum of 4 km in daytime because of the strong solar radiation and the small Solar Zenith Angle, reaching almost 0° in this period of the year.

High aerosol loading and afternoon clouds were other important constraints for the measurements in the free troposphere. Nighttime measurements to altitudes of up to 6-7 km allowed the recording of background ozone concentrations important for photochemical modeling.

Steady increase in the daily values of ozone concentrations together with the formation of a RL at altitudes above 700-800 m is clearly seen in the time series shown in Fig. 2. The detachment of the BL seen in the aerosol profiles is also observed in the ozone data, especially during the last day of the measurement period. Elevated ozone concentrations measured in the altitude range 2600- 3200 m, and more pronounced on the second and fourth-day profiles are probably caused by the enhanced convection in the early afternoons.

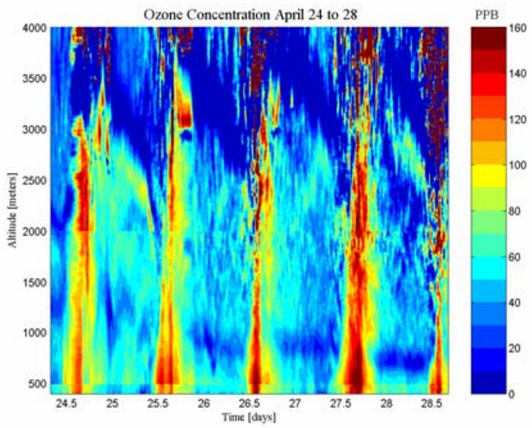

Fig.2. LIDAR time series of the ozone concentration [ppb]

The measurements showed the presence of relatively clean air masses in the altitude region 500-700 m, confirmed by two tethered balloons flown during the period. The balloon data showed significant wind share in this altitude range.

Since the measurements were taken in heavily polluted air with high aerosol loading, most attention was paid to the estimation of the systematic errors caused by differential aerosol extinction and backscatter. The systematic error due to differential SO_2, NO_2, and HCHO absorption was not taken into account in this preliminary study since it is relatively low for the employed DIAL pairs. Nevertheless, their influence will be considered in the final results. Since the Ångström exponent at UV wavelengths is usually lower than the exponent in the visible region, the differential aerosol extinction errors can also be neglected to a first approximation.

The most significant systematic error in regions with inhomogeneous aerosol distribution comes from differential aerosol backscattering at the DIAL wavelengths [Browell]. To define the regions in the ozone profiles with potentially high backscatter error we compared the time series of the derivative of the logarithm of the range-corrected signal at 532 nm (Fig. 3) with simultaneously measured ozone time series. Theoretical analysis can be found in [6]. The range-corrected signal is used instead of a backscatter profile because of the difficulties in inverting the elastic signal in regions with clouds.

The comparison has shown no important change in the ozone concentration in regions with enhanced backscattered gradients, where the most significant errors can be expected.

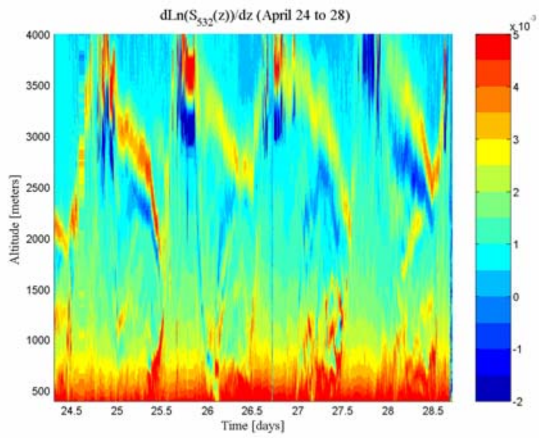

Fig. 3. Derivative of the ln of 532 nm range corrected signal.

Conclusion

Ozone and aerosol measurements were taken during the MCMA 2003 field campaign in Mexico City. Measurements in the BL demonstrate high, well-mixed aerosol and ozone concentrations in daytime. Detachment of the top of the BL and formation of a RL at altitudes above 1000 m were observed both in the ozone and the aerosol data series. The analysis of the aerosol-induced error has shown that it can be neglected to a first approximation. The results will be used to model the evolution of pollution episodes in Mexico City.

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