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Chimia 51 (1997) 700-704
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 ISSN 0009-4293

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Bertrand Calpini*, Valentin Simeonov, François Jeanneret, Jérôme Kuebler, Vijay Sathya, and Hubert van den Bergh

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1. Introduction: Why do We Need a 3D Real-Time Measuring Technique for Air Quality Studies?

It is recognized that three-dimensional air quality models are one of the most powerful tools for identifying effective strategies to improve air quality. For example, the Eulerian mesoscale chemical transport model developed at the EPFL-LPAS is used to simulate pollutant dynamics over regions like Geneva, Athens Greece, canton Obwalden, and the Swiss Plateau to provide technical guidance to air quality management agencies. The domain covered typically ranges in the order of 100×100 km horizontally, and up to 6 km vertically. The grid resolution can range from below 1-5 km horizontally, with a vertical resolution of some tens of meters for the lowest layer of the model up to 500 m for the top layer. Before the results can be exploited with confidence, it is crucial to validate the model's ability to simulate the dynamics of air pollutants in an area.

Evaluation of air quality models is not as easy as it may seem. The traditional air

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Evaluation of air quality models is not as easy as it may seem. The traditional air

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pollutant measurements are ground based, and are only taken at single points. Such point measurements are often influenced by local sources, and are not truly representative of the surrounding area. On the other hand, air quality models predict pollutant dynamics averaged over rather large volumes. This, and the ability to provide air pollutant data well above ground level, are the strength of using lidar methods. LIDAR has proven to be a very powerful technique for measuring ozone, a criteria pollutants which is a focal point of studies with both a regional and global importance. On a regional scale, in the planetary boundary layer (PBL), ozone is a major component of photochemical smog, and can be harmful, both to humans and as plants. Globally, it is found that man's activities are leading to a rise in background ozone levels, which may in the near future have consequences in terms of greenhouse effect.

Due to the nonlinearity of the phenomena of photochemical air pollution, mathematical modelling is essentially the only scientifically acceptable way to design strategies of abatement of pollution. The experimental data needed to test the mathematical models must be 3D, with a time and spatial resolution comparable to that of the models, as well as a capability for sustained measurement over larger periods in time which is financially not possible with airplanes. Hence, LIDAR may well be an optimal instrument in air pollution measurement campaigns. As shown in a recent study in the Athens Basin [1], surface monitoring of ozone concentrations does not provide all the desired information for the modelling.

2. How to Measure Ozone in 3D?

A lidar instrument is composed of a transmitting and a receiving section. The laser beam is emitted in the atmosphere by the transmitter. The beam interacts with atmospheric constituents upon propagation, and light is then either backscattered due to phenomena like *Rayleigh*, *Mie*, and *Raman* scattering or fluorescence. The backscattered light is collected by a telescope, is spectrally resolved, and recorded by a detection unit: the latter form the receiving section of the lidar instrument.

The concentration of atmospheric molecules can be selectively measured by using two specific wavelengths, one of which is more absorbed by the species than the other. For species which show narrow absorption features, typically one wavelength is tuned on an absorption line

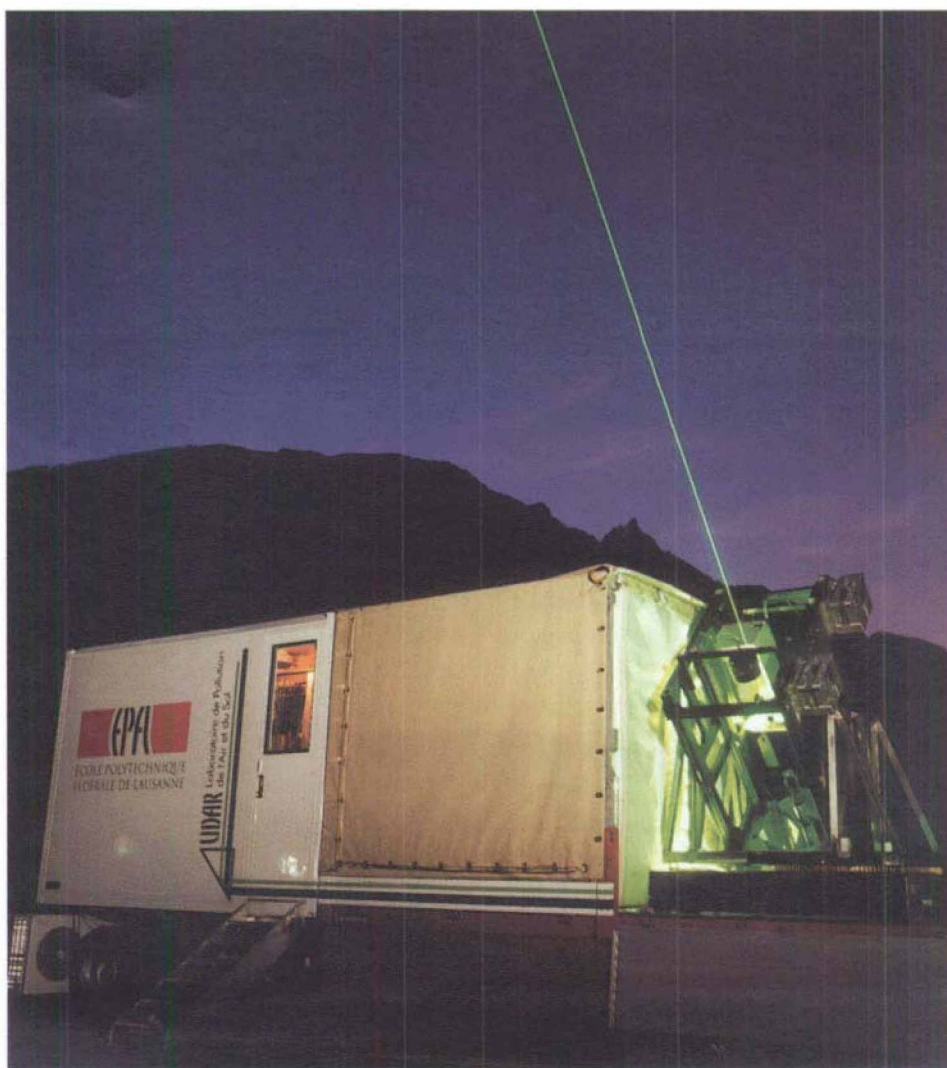


Fig. 1. Night-time picture of the EPFL-LIDAR in Alpach, July 1997

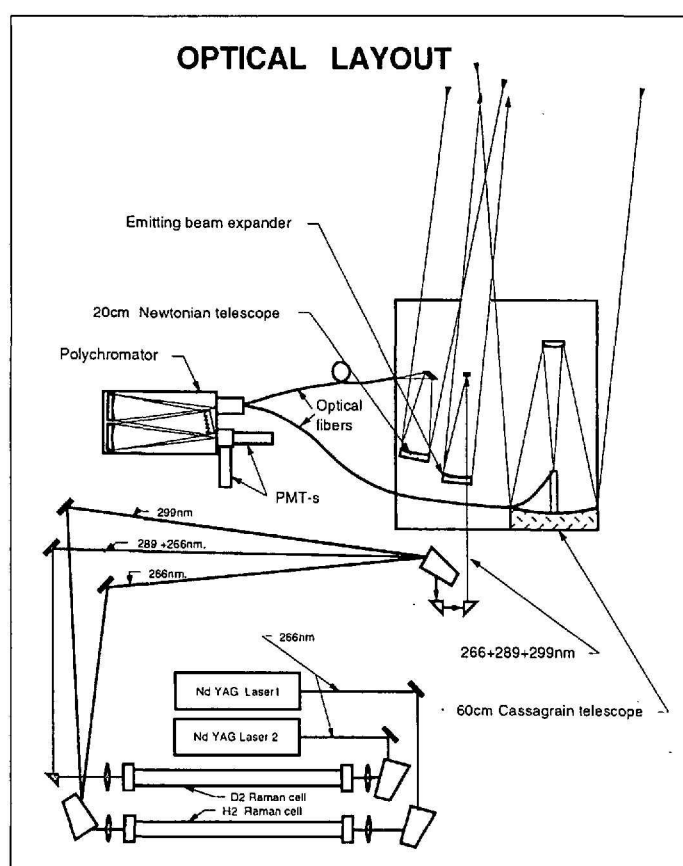


Fig. 2. Optical layout of the ozone DIAL system

(λ_{on}), and the other one is tuned off the line (λ_{off}). Two wavelengths rather than a single one are needed in order to strongly reduce the effect of the extinction in the atmosphere (mostly due to scattering) and to correct for the instrument calibration constant. Such a lidar is called **Differential Absorption Lidar (DIAL)** [1][2].

For ozone DIAL measurements, the ozone absorption spectrum does not exhibit any strong or narrow absorption features in the 250–300 nm *Hartley* band region where most of the ozone DIAL systems are operated. In this case, the attenuation and the backscattering properties of the atmosphere at the two wavelengths can strongly be affected by the presence of aerosol layers in the atmosphere and a so-called '*Mie* correction' must be applied for the ozone retrieve [3].

3. The EPFL-LPAS Ozone DIAL System

3.1. The Basic Concepts

The picture in *Fig. 1* shows the LIDAR trailer, with the emitter/receiver box at an elevation angle of 50° with the green light at 532 nm emitted by the 2nd harmonic generation of a NdYAG solid-state laser. This picture was taken at Alpnach Airport during the July 97 field campaign in Obwalden, and the Pilatus skyline can be seen behind the LIDAR system.

Fig. 2 shows the basic optical layout of the system. Its transmitting section consists of two separate solid-state NdYAG laser sources which emit sequentially at 266 nm (NdYAG 4th harmonic generation) with a delay of 100 μ s, an energy per pulse of 100 mJ and a 10 Hz repetition

rate. Each laser pulse is transmitted into a *Raman* cell using a *Pellin Broca* prism. The *Pellin Broca* prisms are chosen so to avoid *Raman* returns back to the laser head and to prevent any damage to the internal optics of the laser. In this configuration, one cell is filled with hydrogen (3 bars) as *Raman* active gas, and the second one with deuterium (2 bars). The first hydrogen *Stokes* line is *Raman* shifted to 299 nm and is emitted together with the remaining 266 nm pumping beam at the output of the hydrogen cell, with a conversion efficiency to the first H₂ *Stokes* wavelength of up to 50%. The 266 nm and the 299 nm beams are separated by another *Pellin Broca* prism, after minimizing their divergence with a lens system consisting of two lenses of 75 cm focal length. From the high pressure deuterium cell, the D₂ first *Stokes* line is shifted to 289 nm and is kept colinear with the 266 nm pumping beam at the output of the cell. The *Raman* conversion efficiency in D₂ is lower than in H₂ but can be close to 40%. This three wavelengths emitter offers the possibility to optimize the choice of the two DIAL wavelengths needed for different air quality conditions (ozone smog episode, low visibility) as well as for different ranges and elevation angles of the measurements [4].

Three dichroic mirrors combine the 266, 289, and 299 nm beams at the entrance of another *Pellin Broca* prism, under very accurate angles of incidence so as to make the beams colinear at the exit of this polarizing prism. The three beams are on the same axis after the prism and they are aligned on both the azimuthal and the elevation axis of rotation of the emitting/receiving box, which contains the last emitting beam expander (magnification $\times 10$) as well as two receiving telescopes (see below). This box can be tilted towards any direction in the space for the 3D measurements.

The receiving section of this LIDAR is based on the use of two telescopes operated simultaneously: a 'short range' *Newtonian* telescope with a primary mirror diameter of 20 cm and a 60 cm focal length to measure species at short distance, typically between 50 and 600 m, and a 'long range' *Cassegrain* telescope with a primary mirror diameter of 60 cm and a 460 cm focal length for measurements from 500 m up to the free troposphere (typ. up to 3 km AGL). This is a new configuration which permits to measure vertical ozone profiles with a range resolution similar or better than the vertical resolution of the layers of the LPAS *Eulerian* mesoscale model. At the focal point of each telescope, the light is injected into two similar all UV silica

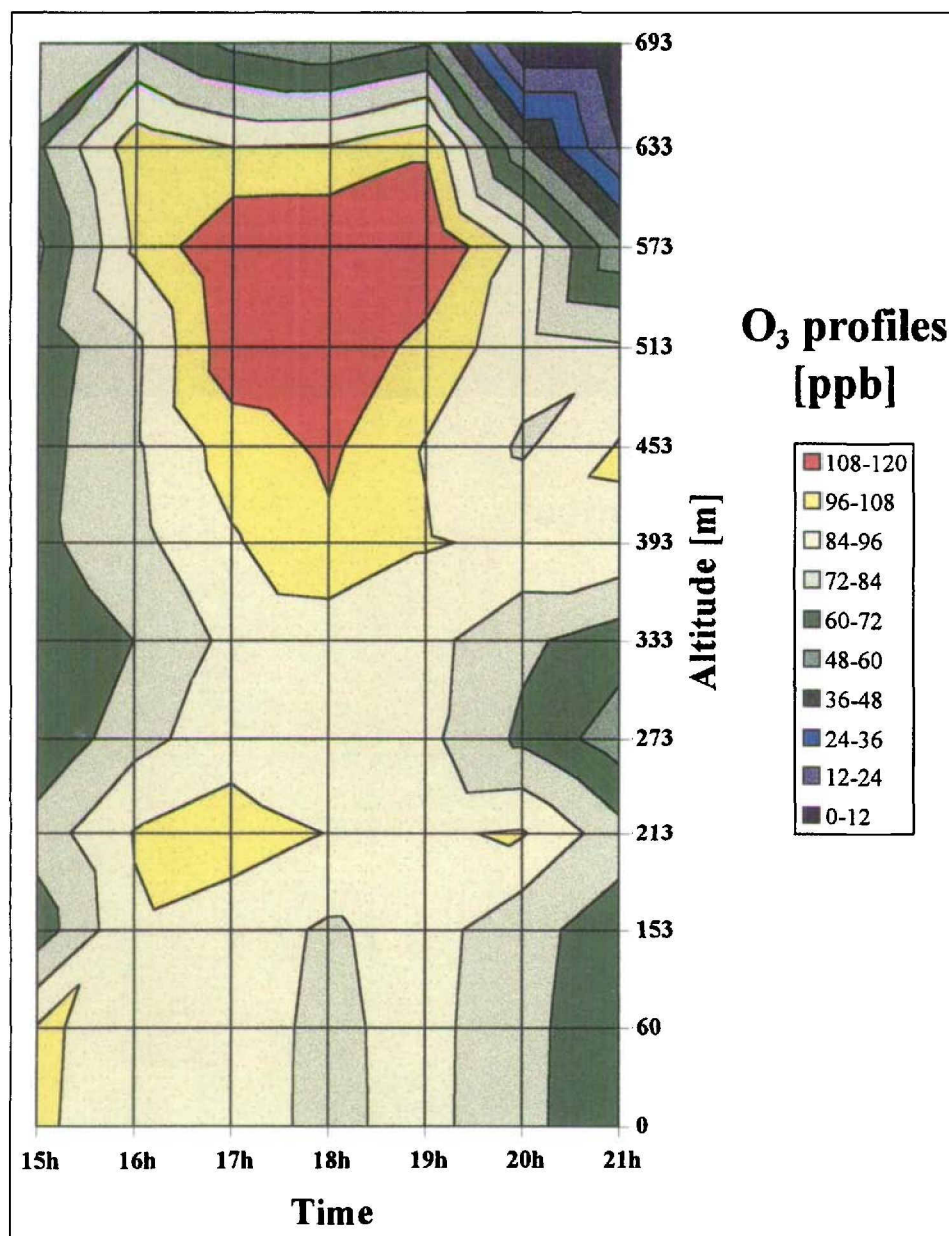


Fig. 3. A time series of ozone vertical profiles for July 18th 1996 in Geneva (Pont de l'île), from 3 to 9 pm, and from the ground level to ≈ 700 m. The ozone data at the ground level are measured by the ECOTOX-Geneva DOAS system.

fibers with a 1.9 mm core diameter, thus corresponding to a 3 mrad acceptance angle for the short range telescope and 2 mrad for the long range telescope. The fibers are connected to a 500 mm optical path polychromator so as to select the backscattered light at the different laser wavelengths, and to reject the solar background (rejection ratio 1:10⁵). At 299 nm, the remaining solar noise is still in the photon counting regime, even at the maximum of the solar radiation. The two PMT's at the exit of the polychromator detect, respectively, the light from each telescope, and the analog signals from the PMT's are preamplified, and captured with an 8-bits 500 MHz transient digitizer with a special shot per shot electronics [5].

3.2. Results and Discussion

As a first example, Fig. 3 shows a time series of ozone vertical profiles measured at the Pont de l'île in the center of Geneva on July 18th 1996 in the afternoon. The ozone concentrations are presented from the ground level up to 700 m, between 3 and 9 p.m. The ground level DOAS measurements taken by the Geneva ECOTOX service are shown together with the DIAL ozone vertical profiles obtained with the short range telescope only. The emitter/receiver of the DOAS system is located near the LIDAR trailer with the retroreflector at ca. 200 m distance along the Rhône river. Relatively high ozone concentrations, around 80 ppb, are measured in the center of Geneva, with a well-mixed layer before 4 p.m. over the range presented here, thus revealing essentially constant ozone concentrations from the ground level up to 700 m. From 5 to 8 p.m., this situation changes rapidly and a strong increase in ozone concentration is measured between 450 and 620 m above ground, with values reaching 120 ppb. Around 9 p.m., this episode is over and the ozone concentrations return to ca. 70 ppb, as measured by DOAS at ground level, and with maximum values around 90 ppb at higher altitudes. This example shows the potential of a DIAL system, with measurements obtained in this case for 6 h in a row, at an altitude and location (Geneva Center) for which airborne measurements are essentially not possible. One should note that above 650 m, these measurements with the smaller of the two telescope systems are hampered by an insufficient signal to noise ratio for the more absorbed wavelength λ_{on} .

For the profiles presented in Fig. 4, the LIDAR system was located in Changins near Nyon during the first phase of this Geneva air quality study, at the NE border

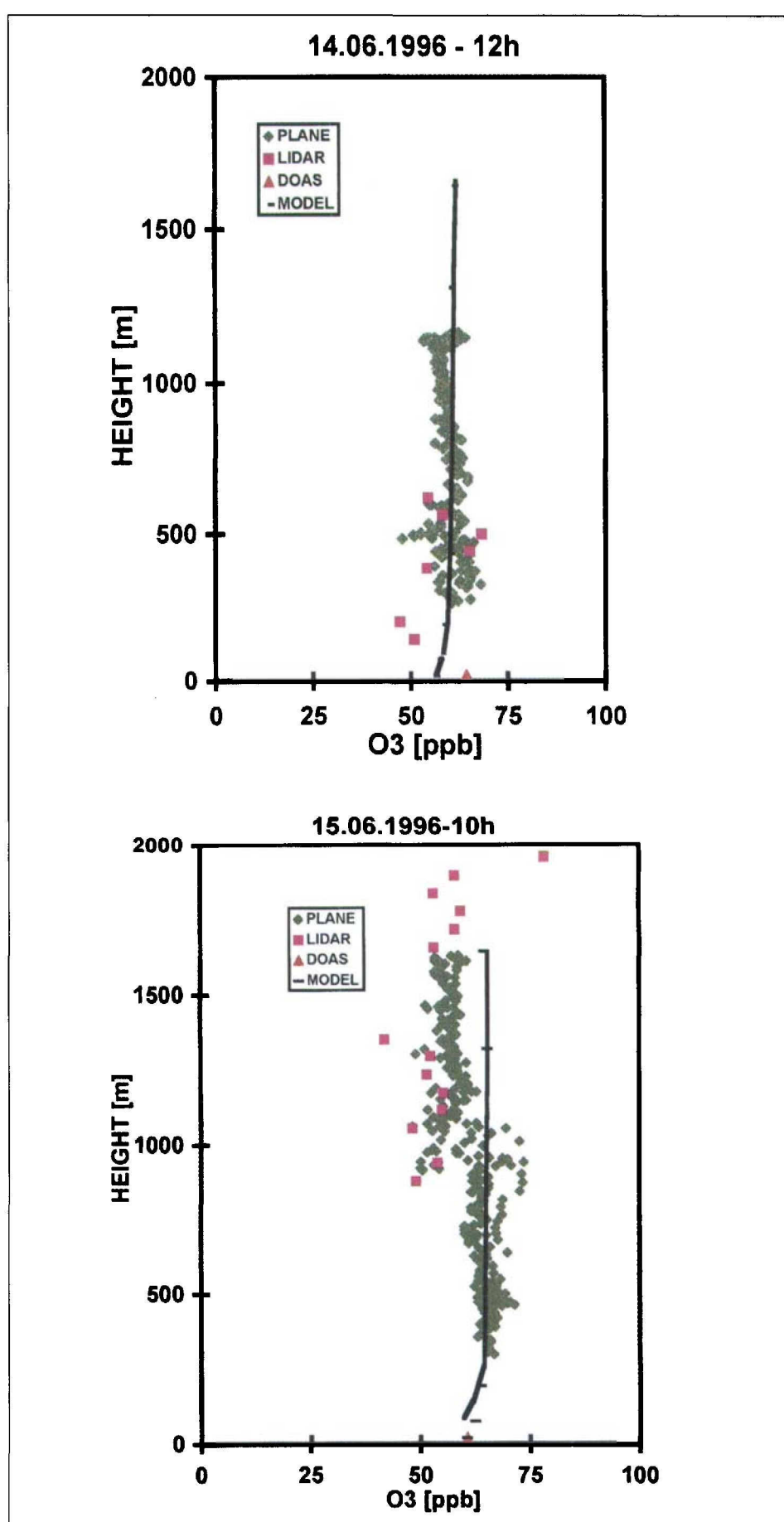


Fig. 4. Ozone vertical profiles over Changins/Nyon obtained by the METAIR motorglider (PLANE), the DIAL measurements (LIDAR) and the DOAS from IUL-Liebefeld at ground level. These measurements are compared with the simulated results obtained by the CIT-EPFL Eulerian model. For July 14th at 12 UTC (Fig. 4, a), the DIAL data are measured with the short range Newtonian telescope (primary mirror diam 20 cm), and for July 15th at 10 UTC (Fig. 4, b), with the long range Cassegrain telescope (primary mirror diam 60 cm). See text for further details.

of the model domain. This position was selected to assess the input boundary conditions of the model for NE wind and in particular the ozone flux into the model domain. At the same time, airborne measurements were performed by METAIR with a motorglider equipped with various chemical analyzers. During the flights, detailed information throughout the model domain was obtained, and in particular ozone was monitored with an UV differential absorption analyzer. The ozone values measured in a radius of 4 km around the LIDAR vertical axis are plotted here, ranging from 300 m to 1 200 m above the ground level in Fig. 4, a and from 350 to 1 600 m for Fig. 4, b, for a time window of 1 h around noon on July 14th and respectively 10 a.m. on July 15th. Ozone DIAL data are presented in Fig 4, a for the short range telescope, and in Fig 4, b with the long range telescope. The airborne data gives an accurate local value of the ozone, while the DIAL data are averaged over an optical path of 60 m and with an accuracy of ca. $\pm 10\%$ [6]. Such a comparison is often carried out with monitoring systems that are not probing exactly the same volume of air at the same time. To compare the different results, one has to make the assumption of a homogeneous ozone distribution in space and time, which is not always realistic. Ozone data are compared here with the model results obtained with the CIT-EPFL model. The model was applied for the episode between July 13th and July 16th. Results of the model show good agreement with the measurements (Fig. 4). Nevertheless, the model result at ground level slightly overestimates the effective ozone concentration measured by the DOAS for the 15.06 at 10 p.m, while the top of the mixing layer is well defined around 900 m AGL by the airplane and the LIDAR data, but is not retrieved on the simulated profile because it was assumed in the calculations that there was no change in the ozone concentration below and above the top of the mixing layer.

4. Conclusions

In the past, ground based measurements were the primary, if not the only, data to be used for comparison with the air quality model results. Such measurements have significant limitations since they can be influenced by local sources while the model results are averaged values over the entire 'box'.

On the contrary, LIDAR provides volume averaged measurements of many pol-

lutants of interest, such as ozone, NO_2 and others. The data obtained by LIDAR are particularly well suited for tropospheric air quality field studies. Mesoscale air quality models are based on predefined volumes of ca. 1–5 km horizontally and tens or hundreds of meters resolution vertically, and the total height in most modeling studies generally include the top of the PBL and the lower part of the free troposphere. Both of these characteristics are similar to the range of the LIDAR data. Thus LIDAR data, which can be obtained within minutes for one laser beam position, provide a very valuable data base for evaluating the model's results, and for further understanding of the dynamics of pollutants in the atmosphere. Aircraft based instruments give further useful complementary 3D air quality measurements, with the advantage of giving information on a larger domain. They, however, suffer from the limitation of relatively short flight durations, and the fact that it is difficult to work at night or at low altitudes. Furthermore, the LIDAR technique does not only provide data with the right spatial range, but it also provides essentially continuous measurements in time. In air pollution studies, such as those for Geneva, Athens and elsewhere, the importance of LIDAR measurements is being further demonstrated.

We gratefully acknowledge the help of Prof. A.G. Russell during the final preparation of the manuscript, and we thank the *Federal Office for Science and Education (OFES)* for the funding support of the COST 615 Geneva project.

Received: August 4, 1997

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