Planetary Boundary Layer ozone fluxes from combined airborne, ground based lidars and Wind profilers measurements

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Different remote sensing techniques have been used for the analysis of the air pollution in the Milan area. Results and intercomparisons from two lidars are presented, obtained from a ground based and an airborne instrument. Combining them with range resolved wind measurement, horizontal ozone fluxes are obtained, showing a strong vertical stratification of different air masses from different origins.

The PiPaPo (Pianura Padana Produzione di Ozono) experiment took place in the Po Valley, between Milan and the foothills of the Alps, from April 26, 1998 till June 20. It was the first field campaign of the EUROTRAC 2 LOOP (Limitation Of Oxidant Pollution) subproject [1]. Two intensive observation periods (IOPs), during a time with favourable photochemical conditions (nice and warm weather) occurred.

EUROTRAC-2 main objective is to support further development of abatement strategies within Europe by

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providing an improved scientific basis for the quantification of source-receptor relationships for photo-oxidants and acidifying substances. In the LOOP subproject, the aims of the PiPaPo experiment were to obtain a better understanding of the time and spatial evolution of photochemical pollution (Milano plume) and its relative sensitivity on reduction of volatile organic compounds (VOC) and nitrogen oxides (NO_X) emissions. Teams from Italy, Switzerland, France, Germany, Austria and Scotland were installed in a "south to north" line across Milano. The Swiss Federal Institute of Technology in Lausanne (EPFL) was operating a point monitor van, a doas system, an ozone lidar and a wind profiler from the Swiss Meteorology Office in Seregno, 30 km north from Milano in a suburb area. The EPFL also assumed the data treatment for the CNRS airborne lidar.

How do we measure ozone by lidar?

A lidar (light detection and ranging) 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 elastically backscattered due to molecular and particulate scattering, respectively named Rayleigh and Mie scattering. The backscattered light is collected by a telescope, is spectrally resolved, and recorded by a detection unit: this forms the receiving section of the lidar.

The concentration of atmospheric molecules can be selectively measured using two specific wavelengths, one of which being more absorbed by the pollutant of interest than the other. For species which show narrow absorption features, typically one wavelength is tuned on an absorption line (λ_{on}), and the other one is tuned off resonnance (λ_{off}). Two such 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).

The laser beam propagation in a clear atmosphere can be described as:

$$P(\lambda, R) = C(\lambda, R)\, P_L(\lambda) \frac{A_r}{R^2} \Delta R\, T^2(\lambda, R)\, \beta(\lambda, R) \qquad (1)$$

This is the lidar equation, where $P(\lambda, R)$ [W] is the power received at wavelength λ from a distance R from the lidar, $C(\lambda, R)$ [dimensionless] is a system function, $P_L(\lambda)$ [W] is the average laser power emitted to the atmosphere, $\frac{A_r}{R^2}$ [steradians] is the acceptance solid angle of the receiving optics with a collecting area A_r [m^2] (detection surface of the telescope), ΔR [m] is the range resolution of the lidar signal, $\beta(\lambda, R)$ [$m^{-1}\,sr^{-1}$] is the backscattering coefficient and $T^2(\lambda, R)$ [dimensionless] is the round-trip transmittance approximated by the Lambert-Beer law:

$$T^2(\lambda, R) = \exp\left[-2\int_0^R \alpha(\lambda, r)\, dr\right] \qquad (2)$$

where $\alpha(\lambda, r) = [\alpha_{aer}(\lambda, r) + N(R)\,\Delta\sigma(\lambda)]$ [m^{-1}] is the volume extinction coefficient with α_{aer} the extinction due to the aerosols, and $\Delta\sigma = \sigma(\lambda_1) - \sigma(\lambda_2)$ the differential absorption cross section of the target species with concentration $N(R)$.

For DIAL applications, with the two wavelengths λ_{on} and λ_{off} and with the additional approximation of constant system function, we can write:

$$\frac{P(\lambda_{on}, R)}{P(\lambda_{off}, R)} = \frac{\beta(\lambda_{on}, R)\exp\left[-2\int_0^R \alpha(\lambda_{on}, r)dr\right]}{\beta(\lambda_{off}, R)\exp\left[-2\int_0^R \alpha(\lambda_{off}, r)dr\right]}. \qquad (3)$$

By taking the natural logarithm and the derivative at a distance R from the lidar system, we have the concentration of the target species:

$$N(R) = \frac{1}{2\Delta\sigma}\left\{\underbrace{\frac{\partial}{\partial R}\ln\frac{P(R, \lambda_{on})}{P(R, \lambda_{off})}}_{A} - \underbrace{\frac{\partial}{\partial R}\ln\frac{\beta(R, \lambda_{on})}{\beta(R, \lambda_{off})}}_{B} + \underbrace{\left[\alpha(R, \lambda_{on}) - \alpha(R, \lambda_{off})\right]}_{C}\right\} \qquad (4)$$

where term A corresponds to the difference in the slope of the two lidar signals obtained for the on and the off resonance wavelengths, term B is the difference in the backscattering properties of the atmosphere for the two wavelengths, and term C is the difference in the attenuation of the two wavelengths. If interferences from aerosols and other pollutants are neglected, if λ_{on} and λ_{off} are close, and if $\Delta\sigma$ is large, equation (4) can be simplified by taking into account only term A. The ozone absorption spectrum does not exhibit any strong or narrow absorption features in the 250 to 300 nm Harthley band region where most of the DIAL ozone system are operated, and it is needed to use two wavelengths not close enough to allow to neglect terms B and C. This correction is done using atmospheric models and the data from the non absorbed wavelength.

The EPFL ground based lidar

This mobile unit has already been operated in different field campaigns [2–3], and is regularly upgraded [4]. It is an UV DIAL ozone actually based on two quadrupled Nd: YAG laser and two Raman cells, emitting 289 and 299 nm beams as "on" and "off" wavelength sequentially, with a repetition rate of 10 Hz and an average of 4000 shots (or 6 mn 40 s) for each profile. The reception part consists of a 20 cm Newtonian telescope for short range (0.15 – 1 km) detection and a 60 cm Cassegrain telescope for long range measurements (0.8 – 3 km). Ultra compact photomultiplier tubes in analog detection mode are used, with a 12 bits 20 MHz ADC. Daylight operation is allowed by the use of two solar blind filters and an holographic band pass filter for each telescope. Real time raw ozone profiles are displayed during operation but post processing of the data is needed.

The airborne CNRS/SA ALTO lidar

The French Research Airplane [5] took part to the PiPaPo experiment, from May 30rd to June 6th with a total of 9 flights. The aircraft was instrumented with point measurements apparatus as well as an ozone DIAL in a down looking configuration. Flights in the upper part of the planetary boundary layer (PBL) were devoted to point measurements, and the lidar was not operated. Typical lidar flights were done at higher altitude, around 2500 m above sea level (asl). This ozone DIAL is operated at 266, 289 and 316 nm, these three wavelength being generated by a quadrupled Nd:YAG laser working with a 20 Hz repetition rate and a Raman cell. The receiving telescope is a 40 cm Cassegrain, and both analog and photon counting detection modes are used. Ozone profiles are displayed in real time onboard, raw data are stored with the aircraft flight information and *in situ* results.

The Lidar observations during PiPaPo

Ground based lidar and balloon measurements

On June the 4th, two ozone radiosonde balloons were launched from Seregno for comparison with the ground based lidar measurements.

In figure 1, we show an intercomparison between relative humidity and ozone profiles obtained with the radiosonde balloon launched from Seregno, and lidar data obtained from the short range and the long range telescope for both the backscatter and the ozone concentration. This experiment was performed in a day of very hazy conditions, as it can be already seen on the humidity profile: typically above 1100 m asl, very strong gradients in the water vapor mixing ratio are essentially preventing our ozone lidar instrument from any reasonable ozone measurements. Thus in this case, ozone lidar values are obtained only up to this altitude, with a resolution of 90 m and for an averaging period of 15 mn. The error bars are smaller on the long range telescope due to a better signal to noise ratio with the large telescope. On the contrary, if we use only the less absorbed wavelength λ_{off}, the backscattered coefficient is obtained for both the short range and the long range telescope with an excellent overlap between these two sets of measurements from approximately 600 to 1800 m asl, and a resolution of 22 m. The backscatter coefficient underlines again the presence of very dense aerosols layers, and in particular shows a clear decay at the top of the mixing layer where a large fraction of the suspended particulate matter are trapped in the inversion layer. Hence the daily evolution of the PBL over

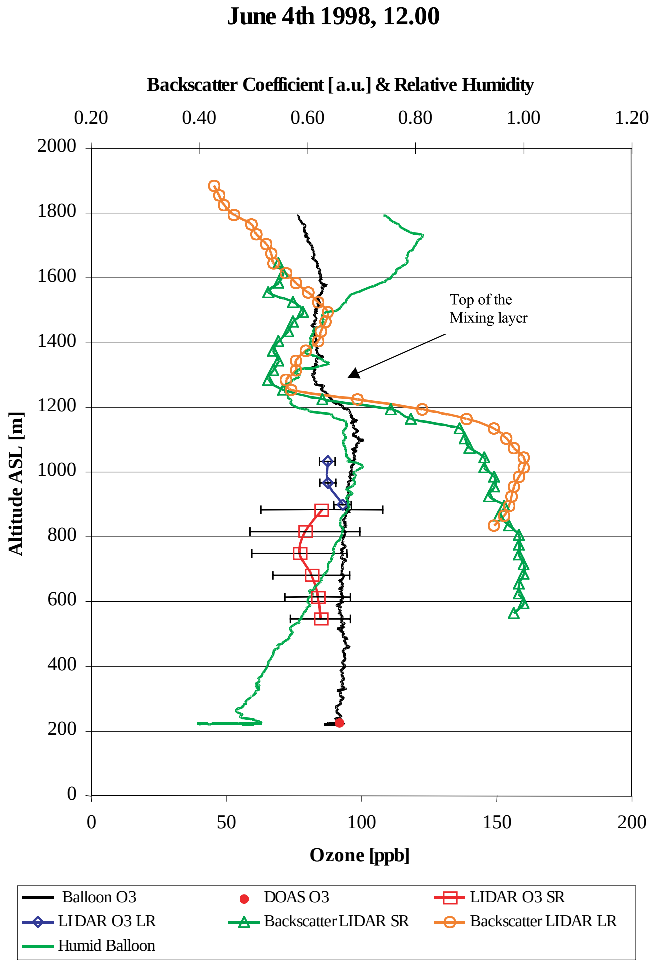

Figure 1. Ozone and humidity radiosonde and lidar measurement, with doas value at ground level. Lidar backscattering coefficient is an useful indicator of the height of the PBL, as could be the relative humidity or the ozone content measured by balloon.

Seregno is derived from the lidar data set. This information is also of importance for the analysis of the Radon measurements done in Seregno by the university of Milan.

Ground based and airborne lidar

Among different airborne lidar flights during PiPaPo, a typical flight patterns is presented in figure 2, from a flight track covering approximately 70 km north to south, extending 20 km south of Milan, and 125 km west to east centred over Milano, while figure 3 presents an intercomparison between this airborne and the ground based lidar. The airborne profiles are given with a measurement each 75 m, but the effective vertical resolution is 300 m, and the profiles are recovered from an horizontal averaging of approximately 3.5 km (averaging time is 50 s, with a flight speed of 70 m/s). No errors bars are drawn on the profile, they are

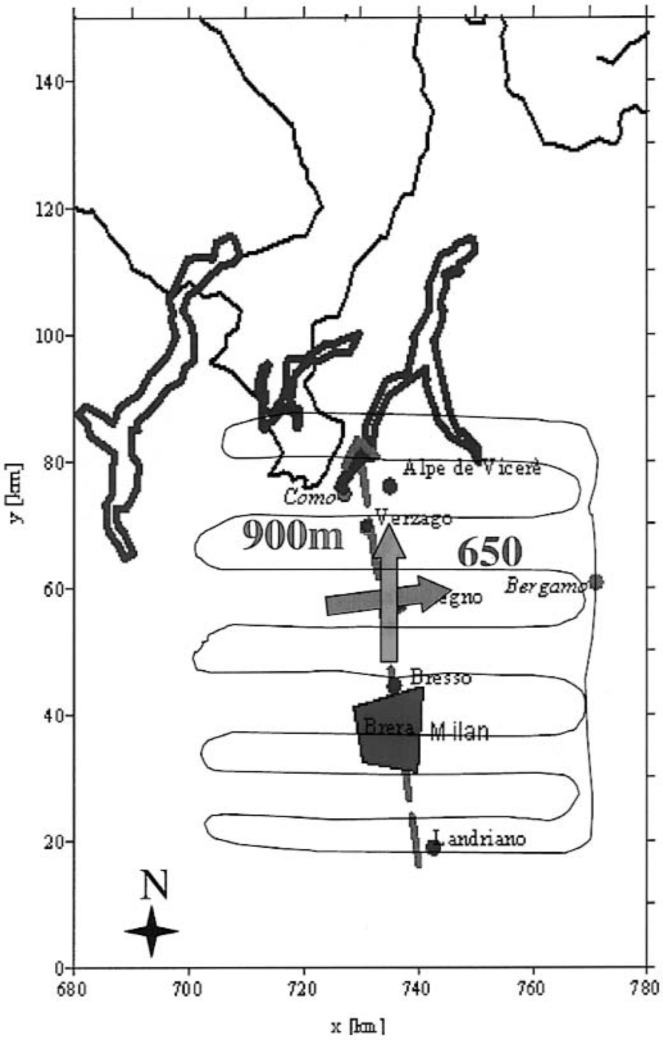

Figure 2. Schematic view of the area where was the PiPaPo experiment held. A typical flight pattern is presented. The two arrows represent the ozone flux direction at altitude of 650 m and 900 m asl on May 13th, around 16h30.

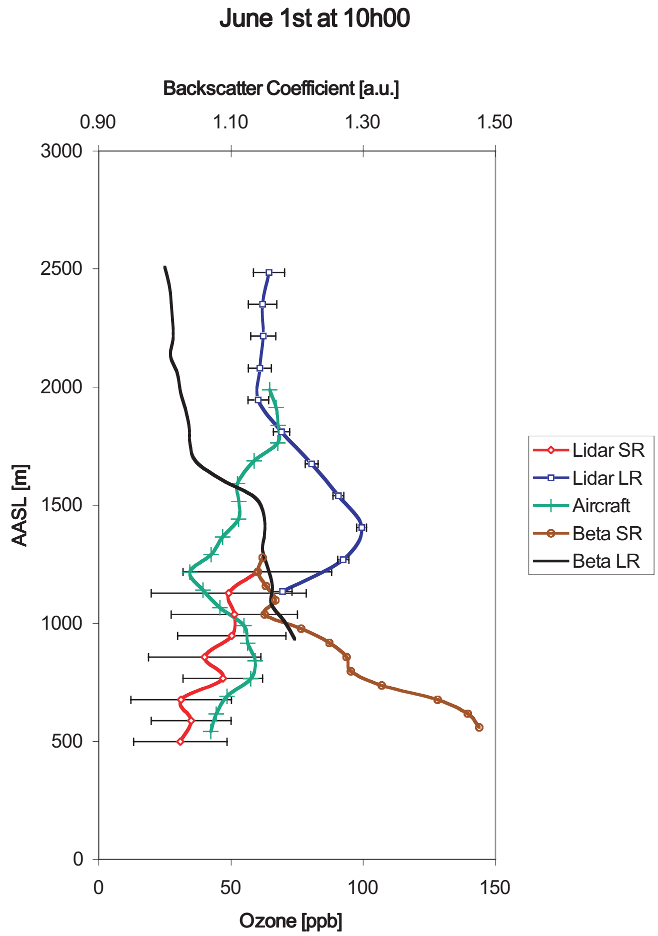

Figure 3. Airborne and ground based lidar ozone measurement, with the backscatter coefficient measured by the EPFL lidar, in arbitrary unit. Ozone values between 1000 and 1800 m asl are strongly affected by the aerosols gradient.

discussed in [5]. This intercomparison is performed on June 1st, a day with a better visibility than the results presented in figure 1. In this case, ground based lidar ozone observation are achieved up to 2500 m asl. Nevertheless between 1100 m and 1800 m asl, we have again strong variations in the backscatter ratio observed with the two telescopes, and in this range, the estimated ozone values can be affected by large systematic errors. On the data points, only statistical error is presented. This optical bias is by evidence also affecting the airborne lidar data and it is only above or below this strong aerosol layer that both lidar observations agree within 10 – 15 ppb. Part of this discrepancy is also caused by inhomogeneity in the ozone field sampled by the aircraft.

It must be pointed out that the ground based lidar system can follow during some days the temporal evolution of the ozone content in 2D in the vertical axis, while the airborne lidar is able to perform a "freezed" cartography of the ozone repartition in 3D. Hence both the temporal and the spatial evolution of a photochemical smog event is obtained using those two instruments.

A typical example of the daily ozone evolution is presented in figure 4. The ground value (altitude of 220 m asl) is defined by the DOAS, and is linearly fitted with the DIAL values. The ozone data affected by the influence of the aerosol layer have been removed considering the backscattering values, as discussed previously. The corresponding Wind profile evolution is presented. A clear increase of the ozone content is seen after the solar warm up, associated with an altitude increase of the high ozone content layer linked with convection process and thermal heating by the ground. At night, low altitude ozone is destructed while an

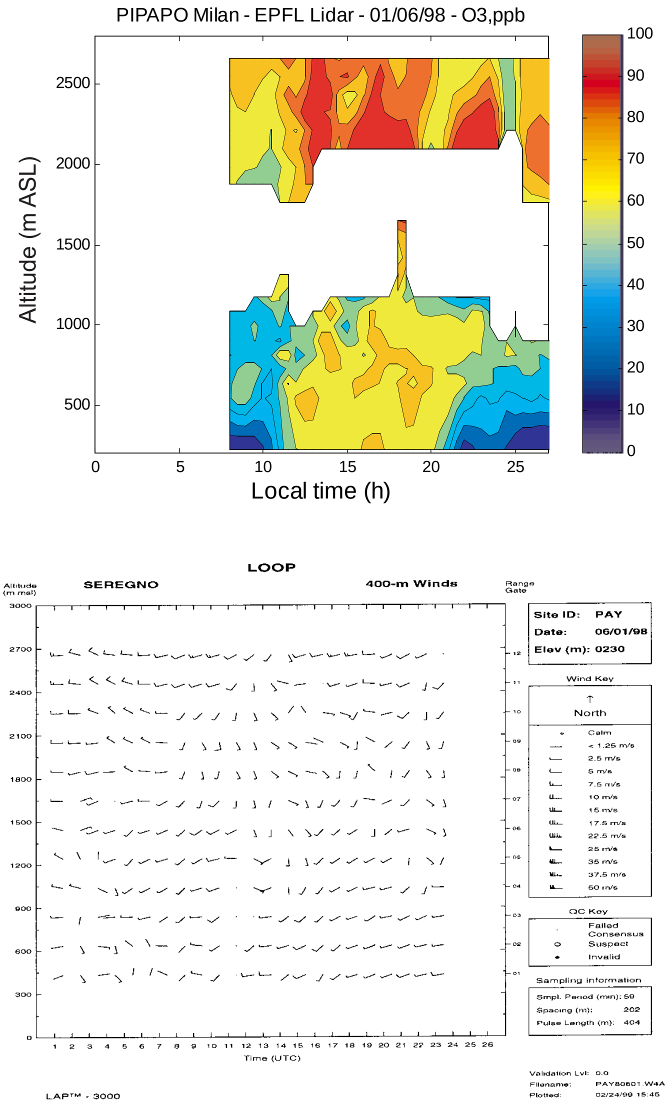

Figure 4. Daily ozone evolution as measured by the ground based EPFL lidar the 1st of June, and the corresponding wind speed and direction as measured by the wind profiler. The altitude scale is above sea level, the ground based ozone value (225 m AASL) is taken from the DOAS system and is linearly fitted with the lidar data. The two altitude ranges corresponds to the short range and long range telescope. Data affected by the aerosol layer or by a poor signal over noise ratio have been removed.

ozone reservoir is formed around 2000 m and is slowly consumed during the next early morning. This graph clearly shows the unique ability of a lidar to follow the temporal evolution of ozone.

Determination of ozone fluxes

The horizontal wind speed and direction are obtained from a wind profiler located close to the lidar system in Seregno

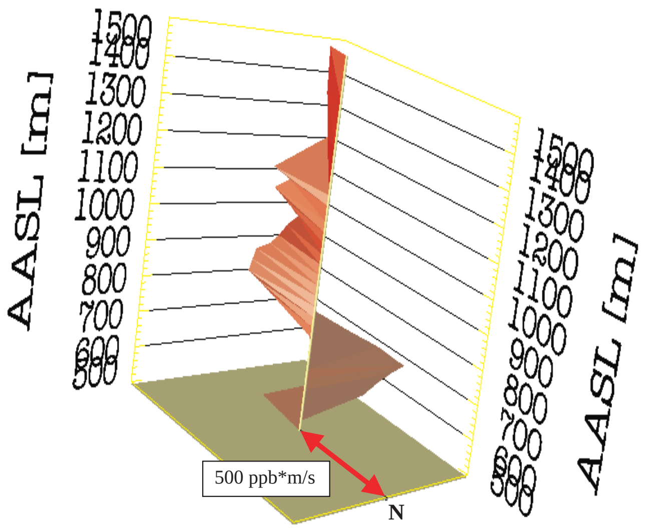

Figure 5. Ozone flux measured in Seregno, on May 13th, 16h30. North direction is indicated by the “N”. The arrow represent a 500 ppb*m/s scale.

(see Ruffieux, this issue). When we combined the ozone lidar profiles with the wind informations, we obtain a first estimate of the horizontal ozone fluxes in ppb*m/s at different altitudes. Figure 5 shows a typical result for May 13th at 4.30 pm. N shows the North wind direction while the red arrow gives the scale for the ozone flux (full scale: 500 ppb*m/s). At 650 meter asl, ozone flux comes from the east whereas at higher altitudes, the flux comes from the south with rather constant intensity. This effect is also shown on the map in figure 2 where a first arrow is showing the ozone flux at 650 m asl (E-W) and a second at around 900 m asl (S-N) with a higher altitude extension. Even if at this period of the day, ozone is measured with well mixed conditions at these altitude, the result clearly shows two different sources of ozone with strong stratification. In this case, the plume of Milano is not affecting the lower part of the profile, thus showing all the complexity for the modelling approach and the need of Eulerian models. This result proves all the advantage of a simultaneous use and coupling of an ozone lidar and a wind profiler. Together with the “freezed” 3-D representation of the ozone content given by the airborne lidar, a clear identification of the ozone dynamics will be gained.

Conclusion

Airborne and ground based ozone lidar are intercompared and also checked against balloon measurements. The use of an airborne lidar gives a “photography” of the ozone field at a given time, while the temporal evolution in one point of the domain is tracked by the ground based instrument.

Using a wind profiler together with the lidar reveals the complexity of the processes involved, clearly highlighting a stratification in the ozone transport.

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References

1. For information about Loop; see: http://www1.psi.ch/~loop/
2. Schoulepnikoff, L.; Van Den bergh, H.; Calpini, B.; Mitev; V. "Tropospheric air pollution monitoring, lidar", Encyclopedia of Environmental Analysis and Remediation, John Wiley and Sons, 1998.
3. Calpini, B.; Simeonov, V; Jeanneret, F.; Kuebler, J.; Sathya, V.; van den Bergh, H. "Ozone LIDAR as an Analytical Tool in Effective Air Pollution Management: The Geneva 96 Campaign", *Chimia* 51 (CHI), 700-704, Neue Schweizerische Chemische Gesellschaft, 1997.
4. Simeonov, V.; Jeanneret, F.; Quaglia, P; Van den Bergh, H.; Calpini, B. "The EPFL UV Ozone DIAL: results and upgradings", Proc., 19th ILRC, Annapolis USA, 1998.
5. Ancellet, G.; Ravetta, F. "Compact airborne lidar for tropospheric ozone: description and field measurements", *Appl. Optics* **1998**, 37(24), 5509-5521.