

## Study of ozone smog episodes by Lidar 3D measurements in Lyon and Paris during summer 1999

A.THOMASSON<sup>®(1,3)</sup>, D.MONDELAIN<sup>(1)</sup>, J.YU<sup>(1)</sup>, J.P.WOLF<sup>(1)</sup>,  
E.FREJAFON<sup>(2)</sup>, P.VISCARDI<sup>(2)</sup>, P.RITTER<sup>(2)</sup>, T.MENARD<sup>(3)</sup>, Y.GODET<sup>(3)</sup>, D.WEIDAUER<sup>(4)</sup>,  
R.FABIAN<sup>(5)</sup>, V.SCHMIDT<sup>(5)</sup>, D.MOELLER<sup>(5)</sup>

- (1) Laboratoire de Spectrométrie Ionique et Moléculaire (LASIM)  
UCBL1, UMR 5579, bât.205, 43 Bd du 11 nov. 1918, 69622 Villeurbanne Cedex, FRANCE  
(2) Ville de Lyon, 35 rue Bossuet, 69006 Lyon, FRANCE  
(3) Institut National de l'Environnement Industriel et des Risques (INERIS)  
Parc Technologique ALATA, BP n°2, 60550 Verneuil-en-Halatte, FRANCE  
(4) ELIGHT Laser Systems GmbH, Warthestraße 21, D-14513, Teltow, Berlin, ALLEMAGNE  
(5) Brandenburg Technical University Cottbus, PO Box 10 13 44, D-03013, Cottbus, ALLEMAGNE

### ABSTRACT

Every summer, ozone smog episodes systematically take place in large agglomerations. In order to prevent them, a better understanding of formation dynamics is needed using numerical models. These models must, however, be validated. Lidar is a unique tool for this task since it provides 3D measurements, for example combining 2D spatial measurements with time in an "animation movie". We present here two recent examples of such ozone Lidar measurement campaigns: the first over Lyon, was mainly used to evaluate a UAM-V type photochemical model and obtain ozone inter comparison data between ground level monitors and Lidar results. The other was performed in Paris and dedicated to validating the Lidar measurements themselves. Very good comparison has been achieved until 300 m altitude, with fixed ozone sensors on the Eiffel Tower. Moreover, since both campaigns took place during an ozone accumulation episode in summer 99, the Lidar results will be compared and discussed.

### 1. Introduction

Ozone is one of the most encountered pollutants around urban areas in summer. Economic impacts on agriculture and public health issues are major concerns for new enforcement, which in turn requires adequate modelling. Forecast of summer ozone episodes is in this respect an important challenge.

Extensive work has been dedicated in the last decade to modelling ozone smog formation using predictive photochemical models. However, most of the validation procedures are only made of comparison with ground based spot monitors, which data are highly dependent on their location and restricted to the surface. They moreover are not representative of air volumes on the same spatial scale (numerical grid cells are usually around 1kmx1 km). Validating numerical models with 3D concentration profiles over large distances is therefore a very important task of Lidar/DIAL systems [1],[2]. However, the calibration of Lidar systems themselves has also to be made, and adequate comparison schemes with spot measurements have thus to be defined as well. These concerns motivated the realization of the two campaigns in summer 1999 presented here.

The first, which has been performed in June 1999 in Lyon, aimed to evaluate a deterministic photochemical model (UAM-V type) and to obtain ozone inter-comparisons between ground level monitors and Lidar results. Besides the 2 Lidar sys-

tems involved (one for aerosols and the other for ozone measurements), numerous instruments were used for complementary data such as meteorological stations, gas analyzers from the city network, a mobile laboratory unit, data from 3 Sodars, etc..

Work on the modelling side is in progress (performed within the campaign by the ELF Research Center) and first data will be soon available for comparison. The investigated episode covered 3 consecutive days without interruption (June 21<sup>st</sup>-23<sup>rd</sup>). The Lidar measurements provided the spatially resolved ozone temporal evolution (concentrations as a function of altitude, direction and time). Comparison and correlation with meteorological data and fixed detectors results are shown. We also present Lidar measurements of aerosols, which will be used in a second phase for improving the model and applying a correction to the ozone Lidar results (a question of the moment).

The main purpose of the second campaign, in July 1999 in Paris, was to validate ozone Lidar measurements in urban conditions with proper inter-comparison schemes. For this, the Lidar was vertically scanning 5 angles above the "Champ de Mars", in the direction of the "Eiffel Tower", which was equipped with ozone analyzers at different levels.

During this campaign, the urban ozone episode was recorded during three consecutive days. Correlation with wind and temperatures is presented, and very good agreement was observed with the standard monitors from the ground to 300 m altitude.

<sup>®</sup> alex@lasim.univ-lyon1.fr

## 2. Campaign in Lyon (June 1999)

Since 1998, in France, each large city has the obligation to develop an air quality management by making continuous environmental analysis but also by giving some elements for a reduction strategy, in case of forecasting pollution episodes.

At the City of Lyon, the air quality network (COPARLY) is based on 150 monitors (implanted since 20 years for some of them), 4 meteorological stations and, since 1999, a new mobile Laboratory which includes a DIAL, a Sodar, and a meteorological station. This mobile DIAL/Sodar system (model 510M) is manufactured by ELIGHT Laser Systems GmbH (Germany), and is able to provide 3D-mappings of  $O_3$ ,  $NO_2$ ,  $SO_2$ , Toluene, and Benzene, as well as wind field profiles [3].

During the ELO (Etude Lyonnaise sur l'Ozone) campaign, a large set of input and output data was collected, through a collaboration between the City of Lyon (sensors network, ozone Lidar, Sodar and meteorological data), ELF Research Centre (photochemical model, emissions inventory, VOC speciation), LASIM (aerosol Lidar) and Météo France (meteorological data, balloon sounding, fixed Sodar).

This intensive campaign took place during an ozone accumulation episode which reached the public information level (2nd level: mean ozone concentration above  $180 \mu g \cdot m^{-3}$ ). We present here the Lidar results obtained during this episode, such as the temporal evolution of the vertical ozone profile (shown in Fig. 1), and 2D maps (over  $180^\circ$  angular scans) recorded at day and night (e.g. at 7.00 a.m. and 18.30 p.m., in Fig. 2). It has to be noticed that the actual data set covers the whole time period and can be presented as a movie, but because of the mode of presentation in paperform only two images are shown.

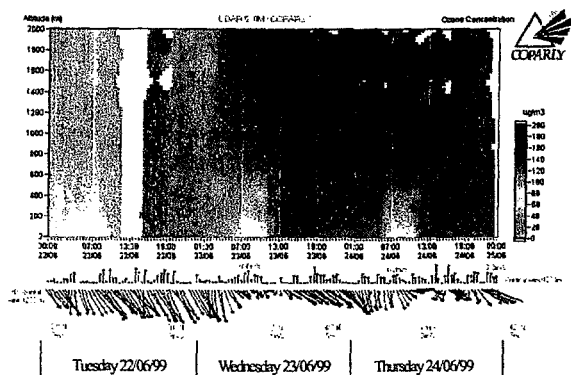


Fig. 1: Vertical Lidar ozone profile, temporal evolution within 3 days.

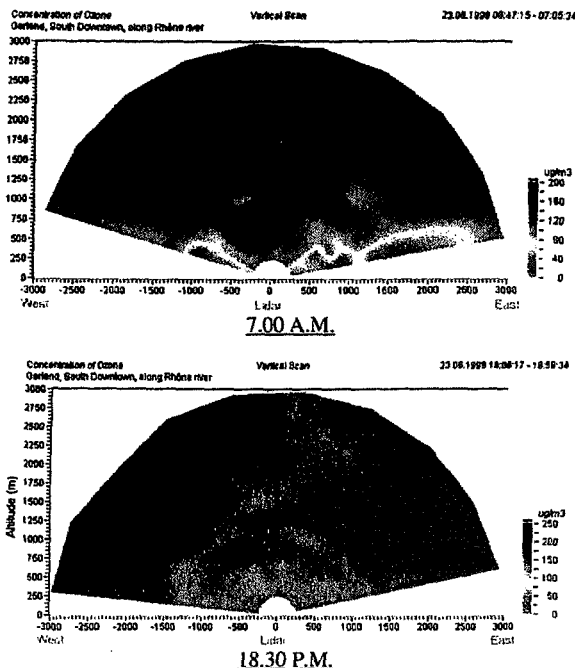


Fig. 2: 2D vertical Lidar map, at different time of the day (7.00a.m. and 18.30p.m.).

The in-homogeneity of the vertical ozone profile is very well observed between 4a.m. and 10a.m. It is mainly due to the heavy morning traffic (ozone is consumed by  $NO$  at lower altitudes). This daily low ozone concentration layer is about 500 m thick, which altitude might represent the urban boundary layer. After 10 a.m., the layers mix and the ozone concentration increases, due to convection and a rise in temperature and solar radiation. All the Lidar scans have been compiled in a "GIF animation", which continuously gives an access to the dynamic processes (3D temporal view).

The model will be evaluated by comparing the obtained 3D Lidar profiles and the network data, to the model's outputs. The impact of urban aerosols in photochemical processes altering the ozone concentration will be considered in the model in a second phase. For this purpose, LASIM measured aerosol profiles using a second eye-safe Lidar and new optical diffusion models. These Lidar data were inverted to retrieve quantitative concentration profiles of urban particles with a method previously described [4],[5]. Figure 3 shows the vertical and temporal aerosols concentration.

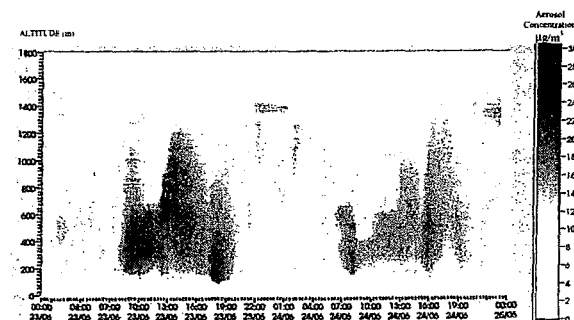


Fig. 3: vertical quantitative aerosols Lidar measurements, temporal evolution.

The final step which is in progress, is to inject these results both in the model and in the ozone Lidar profiles. For the modelling part, the aerosol concentration will be implemented in heterogeneous chemistry reaction, while they will be used to correct Mie-scattering errors in the ozone DIAL-profiles.

### 3. Campaign in Paris (July 1999)

Since 1996, INERIS (Institut National de l'Environnement Industriel et des Risques) is in charge in France of calibrating and validating optical measurement techniques including DOAS and Lidar. In particular, INERIS evaluated the Lidar 510M of Lyon (according to the French Norm AFNOR X20-300) for  $\text{SO}_2$ ,  $\text{NO}_2$  and  $\text{O}_3$  [6],[7]. From July 12 to July 20 1999, a campaign was organised in collaboration with AIRPARIF (the air quality network of Paris) intended to study the response of the Lidar 510M in real urban conditions [9],[10].

A large area in the heart of Paris has been selected for this study : the "Champ de Mars". The Lidar, located in front of the "Military School", was scanning over 5 angles in a vertical plan, in the direction of the "Eiffel Tower". Hence, 2D mappings, as shown below, and comparison with AIRPARIF's standard ozone monitors (located at different levels of the "Eiffel Tower") could be achieved.

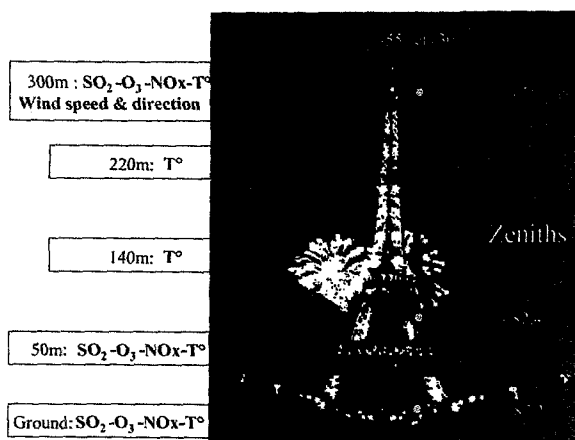


Fig. 4: AIRPARIF detectors' location and Lidar scanning plan.

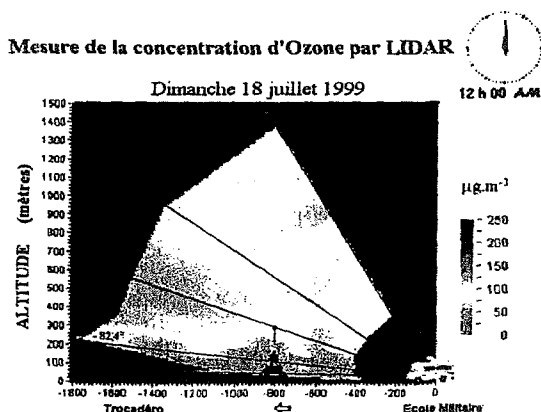


Fig. 5: Lidar location and typical mapping of 2D measurements.

During the campaign, based on the weather forecasts, high concentrations of ozone could be

expected. A 72h continuous measurement was performed in order to monitor the evolution. Results within the most interesting 36 hours are presented here: from Sunday, July 18<sup>th</sup>, at midnight (when the main episode really took place) to Monday, July 19<sup>th</sup>, in the morning. The clearest presentation of such an evolution is again the temporal vertical profile, from the "Eiffel Tower" position (Fig. 6). Wind data from the top floor and temperatures from the ground to the top are reported as well.

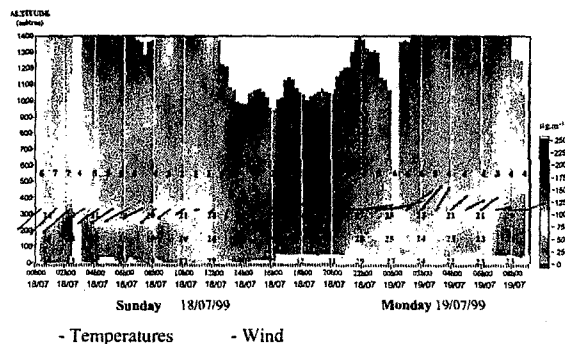


Fig. 6: vertical and temporal ozone profile correlated to meteorological data (local time: TU+2h).

Correlations between the meteorological situation and the episode evolution from the Lidar data can be clearly observed and summarized as follows: at night, without solar radiation, the concentrations decrease significantly at lower altitudes (below 300m), but at higher, an ozone storage reservoir is left. The episode formation has obviously taken benefit from this reservoir to take place, but also from the wind behaviour in terms of wind shears within 24h and a resulting average speed decrease. The layers mixed at about 10 a.m. as the convection started to transport the nitrogen dioxide created at the ground towards higher altitudes, and the solar radiation photo-generated ozone. This day, the 2nd level of pollution has been reached (public information above  $180 \mu\text{g}\cdot\text{m}^{-3}$ ). Until 10 p.m. temperatures remained rather high, and the ozone concentrations at ground level consequently did not clearly decrease before 4 a.m. the next morning.

Comparisons with the fixed standard analyzers measurements on the "Eiffel Tower" are presented:

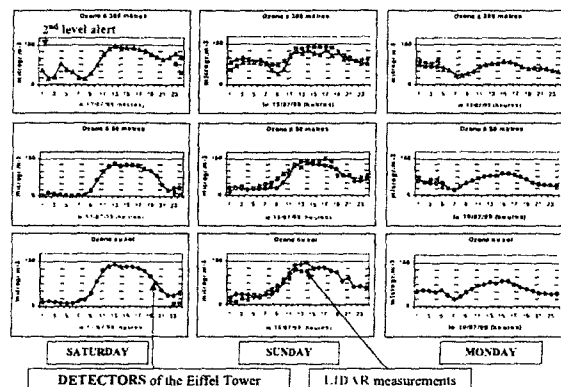


Fig. 7: Comparison of standard analyzers (Eiffel Tower) and Lidar measurements, at the ground level, 50m and 300m altitude, from Saturday, July 17<sup>th</sup>, to Monday, July 19<sup>th</sup>, (abscissa in TU).

A good agreement (within  $30 \mu\text{g.m}^{-3}$ ) is found most of the time ( $50 \mu\text{g.m}^{-3}$  in the worst case). The sensors' incertitude is at least 20% ( $35 \mu\text{g.m}^{-3}$  for a  $180 \mu\text{g.m}^{-3}$  measurement) and the range resolution of the Lidar measurements corresponded to a concentration accuracy of  $20 \mu\text{g.m}^{-3}$ . Although only a rough and uniform Mie correction of  $10 \mu\text{g.m}^{-3}$  was applied, the agreement is excellent.

Hence, this campaign could successfully validate the ozone Lidar measurements of the used instrument in urban conditions.

#### 4. Conclusion

These two campaigns followed different aims, but within them, two ozone smog episodes could be measured during a 3 days period with the DIAL technique. In both cases, a computer movie representing the ozone concentration evolution could be created, allowing an ozone urban dynamic comparison, and a study of general trends. Indeed, it stands out that from 4 a.m. to 10 a.m., the ozone concentration decreases in a layer of a few hundred meters high (thickness of 300 m to 500 m), due to the traffic ( $\text{NO}_2$  generation). Above it, an ozone reservoir is formed, with a relatively homogeneous concentration (about  $100 \mu\text{g.m}^{-3}$ ). After 10 a.m., depending on the atmospheric conditions, the ozone concentration increases from this background level, and quickly reaches the critical level of  $180 \mu\text{g.m}^{-3}$ .

In summary, Lidar instruments are shown as extremely valuable for urban ozone episode forecasting studies.

#### 5. Acknowledgements

The authors wish to acknowledge COPARLY and AIRPARIF (air quality networks, respectively of Lyon and Paris), CRES (ELF Research Center, Lyon), ADEME (Agence de l'Environnement et de la maîtrise de l'Energie) and MATE (Ministère de l'Aménagement du Territoire et de l'Environnement) for their collaboration in these campaigns. LASIM especially thanks Gaëlle DUFOUR and Bénédicte HERVE for their help during the 3 days and nights of aerosols Lidar measurements.

#### 6. References

- [1] E.FREJAFON & al. "Laser applications for atmospheric pollution monitoring", Eur.phys.J.D 4, 231-238, (1998)
- [2] ELIGHT GmbH, "Lidar study on the tropospheric ozone dynamics", private communication (Spain, summer 1996)
- [3] J.P.WOLF "UV DIAL technics for air pollution monitoring", J.Wiley & Sons (Chapter 4), NY 2000
- [4] E.FREJAFON & al., "3D-analysis of urban aerosols by use of a combined Lidar, SEM & X-Ray microanalysis", Appl.Opt. 37 (12) 2231-37 (1998b)
- [5] J.KASPARIAN & al., "Characterization of urban aerosols using SEM, X-Ray and Lidar measurements", Atm.Env. 32 (17) 2957-67 (1998)
- [6] T.MENARD & al., "Calibrage et évaluation de systèmes Lidar", Poll.Atm., 105-119, oct.-nov. 1998
- [7] D.WEIDAUWER & al., "Lidar calibration with internal gas cells", Environsense Symposium communication (Munich, June 1998)
- [8] T.MENARD & al., "Evaluations du Lidar 510M" (1<sup>er</sup> rapport :  $\text{SO}_2$ , 2<sup>ème</sup> rapport:  $\text{O}_3$ ), rapports INERIS - ADEME (déc. 1996, avril 1998)
- [9] Y.GODET & al., "Etude relatives aux analyseurs à long parcours optique: Lidar", rapport INERIS - Loi sur l'Air (déc.1999)
- [10] A.THOMASON & J.P.WOLF "Lidar et techniques optiques utilisées pour l'étude de la pollution atmosphérique" communication INSU Météo-France (Toulouse, France, oct.1999)