TENTATIVE TO RETRIEVE AEROSOL COMPLEX REFRACTIVE INDEX FROM A SYNERGY BETWEEN LIDAR AND IN SITU MEASUREMENTS

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ABSTRACT

The LISAIR (Lidar pour la Surveillance de l'AIR) experiment devoted to a better understanding of the exchanges of particulate pollutants between surface (streets) and the planetary boundary layer (PBL) took place in Paris during May 2005. Dedicated active remote sensors (lidar) as well as ground-based in situ instrumentation (nephelometer, aethalometer and particle sizers) were used to highlight the interest of such a synergy to follow the evolution of aerosols during the day, thus function of human activity. The investigation of urban aerosol optical properties in the PBL Paris area is presented and the results are discussed.

1. INTRODUCTION

Air quality monitoring in urban and suburban areas has become a major health issue. High pollution levels in big cities areas owing to concentrated anthropic activity require on behalf of policy makers to improve predictive capabilities on pollution events and better understand processes driving pollutant concentrations. Studies have consequently been carried out in various megacities, such as Mexico-city [1] or Athens [2]. The role of aerosols is particularly investigated in urban environments: their limited lifetime in the low troposphere, the specific character and various origins of their sources naturally lead to surveys conducted from local to global scales.

Air quality survey over Paris is investigated by means of surface networks dedicated to measurements of critical pollutant concentrations. Nonetheless, pollutants do not remain trapped onto surface and can be transported through the PBL. Lidar are then the most efficiency systems to follow the spatiotemporal evolution of the anthropogenic aerosol in the urban PBL and aloft. An important three-year project, called ESQUIF (Etude et Simulation de la QUalité de l'air en region Ile-de-France), dedicated to the study of the processes leading to pollution events had already taken place in Paris. However this program covers the whole Paris area [3].

Particulate pollutant exchanges between the streets and the PBL, and their daily evolution linked to human activity were studied in the framework of the LISAIR experiment combining in situ measurements and lidar observations. This program lasted from 10 to 31 May 2005. Two lidar systems were used. The first one worked at the wavelength of 532 nm with polarized channels. It was onboard the Mobile aerosol station [3]. The second one was eye safe and operated at the wavelength of 355 nm onboard a car to follow the aerosol dispersion in the street and through the PBL around the ring of Paris.

This work describes the reciprocal contributions of both the fixed and mobile lidar to retrieve optical properties of aerosol in the atmospheric column.

2. LIDAR FOR SPATIOTEMPORAL ANALYSIS

The mobile lidar system has been used to study the spatiotemporal variability of the aerosol trapped in the urban PBL. Measurements were performed on 25 May from town hall place, Champs Elysées Boulevard and around the Paris ring.

Lidar data have been inverted using a well-known method, based on Bernoulli's differential form of the propagation equation. Two situations have been studied before and during the morning traffic jam (Fig. 1 and Fig. 2, respectively).

A specific mean lidar profile is also given in Fig. 1 and 2. The traffic is highly enhanced between the two periods, which leads to a significant increase of the aerosol extinction coefficient in the lower part of the PBL. The relative increase is close to 50% and it reaches 100% in the centre of Paris. On both figures, a residual layer of pollutants can be observed between 0.4 and 0.8 km above the ground level (agl). As shown from previous studies [4], the aerosol present above the Paris area is mainly emitted from car traffic with a significant part of black carbon component [5]. Such a proportion can significantly affect the absorption properties of the anthropogenic aerosol and then the single scattering albedo and the lidar ratio defined to be the product of the single scattering albedo by the normalized backscatter phase function. These parameters are functions of the size distribution, structure and chemical composition of the aerosols, and so are expressed against the complex refractive index of particles.

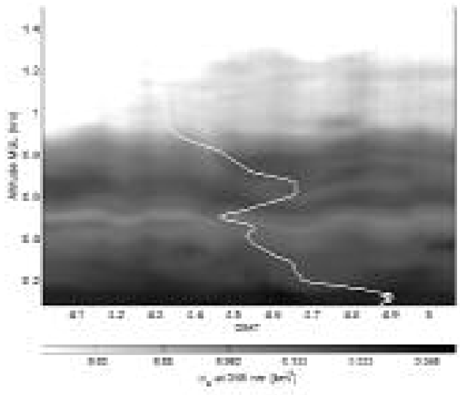

Fig. 1. Temporal evolution of the vertical profile of aerosol extinction coefficient at 355 nm above Paris on May 25, 2005 during the early morning before the traffic jam. The Paris ring is between 4.3 and 5 GMT. The white line is the shape of the mean lidar profile in arbitrary unit.

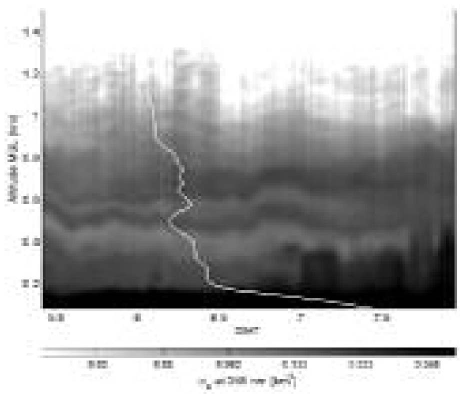

Fig. 2. Temporal evolution of the vertical profile of aerosol extinction coefficient at 355 nm above Paris on May 25, 2005 during the morning traffic jam. The Paris ring is between 5.5 and 7.3 GMT. The white line is the shape of the mean lidar profile in arbitrary unit.

3. AEROSOL CLOSURE USING LIDAR AND IN SITU MEASUREMENTS

This part describes the contribution of our observations to a synergy between fixed lidar at 532 nm and in situ measurements above town hall place to retrieve the aerosol properties: complex refractive index, simple scattering albedo, optical thickness, backscatter-to-extinction ratio (*BER*). The lidar is associated with an overlap factor close to 1 at 100 m agl. After correction, we retrieved the lidar signal until ~50 m agl within an relative error close to 20%. The representativeness of in situ measurements of the aerosol aloft is ensured if the relative humidity does not significantly vary between the surface layer and the mixed layer.

The lidar equation is underconstrained and requires considering an external constraint. Sun photometer measurements in Paris from AERONET network (http://aeronet.gsfc.nasa.gov/) were used to constrain the lidar inversion. It was thus possible to determine *BER* with an iterative procedure, while varying *BER* between 0.005 and 0.055 sr^{-1} at the wavelength of 532 nm As described in [6], this method lies on the hypothesis that the part of the atmospheric column sounded by the lidar system is representative of the entire atmospheric column, since no aerosol layer has been supposed to be observed above 1.8 km agl. A statistical analysis was developed to prevent from inversing the measurements on an individual profile basis, source of noise. The procedure is convergent when the variation between sun photometer and lidar derived optical thicknesses is lower than 10^{-4}. The histogram of the aerosol backscatter-to-extinction ratio assessed from daytime lidar measurements, when the procedure has been convergent, is reported in Fig. 3. The mean value calculated for the significant values (low rate of relative humidity) of *BER* is close to 0.0175 sr^{-1} with a standard deviation of 0.006 sr^{-1}.

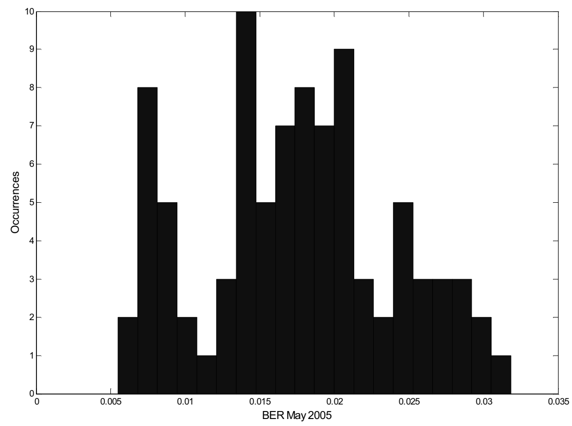

Fig. 3. Histogram of the aerosol backscatter-to-extinction ratio assessed from lidar measurements during the LISAIR program over Paris area in May 2006.

These results can be confronted to those obtained in various experiments. The BER values lie between 0.0125 and 0.025 sr^{-1} on Autralian coast (marine boundary layer), 0.011 and 0.02 sr^{-1} in smoke layers in Southern Hemisphere, 0.0105 and 0.018 sr^{-1} in a polluted boundary layer over Leipzig in Germany, 0.011 and 0.02 sr^{-1} in the polluted lower troposphere above Indian Ocean, 0.0143 and 0.0166 sr^{-1} on the United States northern coast during continental flow, and between 0.0141 and 0.03 sr^{-1} in the polluted centre of the United States. Those results are referred in [7]. A *BER* close to 0.018 sr^{-1} for anthropogenic aerosols as retrieved in our study, reminds the presence of a predominant fine mode in size distribution mainly due to automobile traffic sources. This *BER* is consistent with the value of about 0.014 sr^{-1} found in Paris area at 532 nm within the framework of the ESQUIF program where airborne lidar measurements were performed [4].

To calculate the aerosol optical properties, one needs to know the complex refractive index of these particles during the measurement period. Different techniques enable to calculate urban aerosols refractive indices, for example through a partial molar fraction approach [8]. In this paper, we use the synergy between lidar and in situ measurements.

The aerosol scattering coefficient derived from the three wavelength nephelometer (manufactured by TSI, USA) is a good constraint to assess the real part of the complex refractive index. Lidar is a useful additional constraint because it makes possible an assessment of the imaginary part of the aerosol complex refractive index using the *BER*.

We have used the wavelength of 550 nm for the nephelometer to be close to the one of the lidar system. To determine the real part of the complex refractive index a look-up table has been built from the aerosol scattering cross-sections at 550 nm. Various complex refractive indexes between 1.3 and 1.9 for the real part, between 10^{-8} and 0.2 for the imaginary part have been considered as input values of the look-up table. Calculations have been performed considering spherical aerosols and using the Mie theory.

The aerosol size distribution has been calculated using an approach well described in Randriamiarisoa et al. [5] from the measurements of the particles sizers. During the LISAIR program we used an Electrical Low Pressure Impactor (ELPI manufactured by Dekati) and a Condensation Particle Counter (CPC, manufactured by TSI). Mainly a bimodal size distribution has been retrieved including the nucleation and accumulation modes. The contribution of a coarse mode was not significantly observed.

The aerosol scattering cross section retrieved from in situ measurements on May 18 is shown in Fig 4. In the likely range of the imaginary part of the complex refractive index (between 10^{-8} and 5.10^{-2}), the real part can be assessed to be close to 1.63 with a standard deviation of ~0.01. The computations have been performed for measurements on May 18 since this day is representative of the mean condition of the LISAIR campaign in terms of in situ and remote sensing measurements.

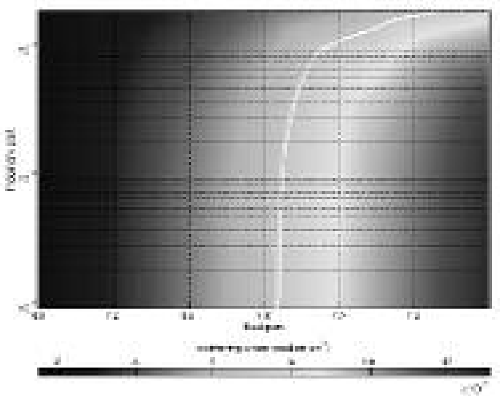

Fig.4. Aerosol scattering cross-sections calculated for various real and imaginary parts of the complex refractive index (look-up table) at the wavelength of 550 nm. The mean value of the scattering cross section (7.08 10^{-11} cm^2) is also given in white for the 18th of May.

The lidar/sunphotometer-derived *BER* is independent parameters of aerosol size distribution and scattering coefficient. The mean value of the aerosol backscatter-to-extinction ratio assessed from lidar measurements on May 18 is close to 0.0156 sr^{-1} with a standard deviation of 0.0058 sr^{-1}. The determination of the imaginary part of the aerosol complex refractive index lies on a comparison between the previous *BER* used to invert lidar data and different values of *BER* calculated from size distribution with a real part of the complex refractive index equal to 1.63. As a result, the imaginary part of the complex refractive index has been assessed to be ~0.006, and the single-scattering albedo is ~ 0.97.

The AERONET website does not provide any data over Paris in May 2005 for the single scattering albedo nor for the complex refractive indices. Values obtained from that network at 440 and 670 nm thus arise from measurements performed in June 2005. Averages and standard deviations are 1.41±0.008 and 0.020±0.002 for the real part and the imaginary part of the aerosol complex refractive index, respectively. The single scattering albedo is then ~0.84±0.02.

During the ESQUIF program in July 2000, Chazette et al. [4] found a single scattering albedo at 550 nm exhibiting a mean value ranging from 0.85 to 0.92. This value was close to the mean AERONET value of 0.87±0.068. The uncertainty on the single scattering albedo has been assessed to be close to 0.06.

We have also used the synergy between in situ and lidar/sunphotometer measurements in the framework of the POVA (Pollution dans les Vallées Alpines) experiment that took place in the alpine valleys of Chamonix in summer 2003 [9]. The same in situ and remote sensing measurements were performed during this campaign. The main aerosol sources are similar than for Paris and due to the traffic. Calculations have given 1.48±0.05 and 0.042±0.01 for the real and the imaginary parts of the aerosol complex refractive index, respectively. The single scattering albedo has been found between 0.75 and 0.85.

4. CONCLUSION

Both ground-based in situ and active/passive remote sensor measurements were performed in Paris to study the anthropogenic aerosol survey above such a megacity. We have presented here a first assessment on the aerosol complex refractive index using the synergy between lidar, sunphotometer and in situ measurements. For a specific day, the retrieved single scattering albedo seems to be larger than the value obtained from the AERONET data and from the previous campaign around the Paris area. Nevertheless, the same method applied in the Chamonix valley provides smaller values of the single scattering albedo for similar aerosol sources due to traffic.

This new approach using the lidar measurements offers new perspectives for aerosol pollution studies above megacities. The knowledge of the aerosols properties in the urban PBL will be very useful to best understand the climate variability in the big cities due to their pollutant emissions.

5. ACKNOWLEDGMENTS

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