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## THE CARBON AEROSOL / PARTICLES NUCLEATION WITH A LIDAR: NUMERICAL SIMULATIONS AND FIELD STUDIES.

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### ABSTRACT

In this contribution, we present the results of two recent papers [1,2] published in Optics Express, dedicated to the development of two new lidar methodologies. In [1], while the carbon aerosol (for example, soot particles) is recognized as a major uncertainty on climate and public health, we couple lidar remote sensing with Laser-Induced-Incandescence (LII) to allow retrieving the vertical profile of very low thermal radiation emitted by the carbon aerosol, in agreement with Planck's law, in an urban atmosphere over several hundred meters altitude. In paper [2], awarded as June 2014 OSA Spotlight, we identify the optical requirements ensuring an elastic lidar to be sensitive to new particles formation events (NPF-events) in the atmosphere, while, in the literature, all the ingredients initiating nucleation are still being unrevealed [3]. Both papers proceed with the same methodology by identifying the optical requirements from numerical simulation (Planck and Kirchhoff's laws in [1], Mie and T-matrix numerical codes in [2]), then presenting lidar field application case studies. We believe these new lidar methodologies may be useful for climate, geophysical, as well as fundamental purposes.

### 1. INTRODUCTION

Recent advances in lidar concern both atmospheric traces [4] and aerosols up to three-component particle mixtures involving volcanic ash, desert dust, sea-salt or sulfate particles [5]. However, up to now to our knowledge, no lidar methodology exists that allows a range-resolved specific detection of the carbon aerosol in the atmosphere over several hundreds of meters, despite its key role on human health, as being carcinogenic in urban polluted areas [6], and in radiative forcing, black carbon being the second contributor to global warming after CO<sub>2</sub> [7]. In the literature, the terminology related to the carbon aerosol involves soot particles, carbonaceous

particles, and biomass burning, as well as secondary organic aerosols, to quote only a few. The carbon aerosol light absorbing properties have been addressed through laboratory measurements on soot particles [8] and accurate numerical simulations [9]. While determining the spatial and temporal behavior of the carbon aerosol is essential for developing carbon reduction emission strategies, in this contribution, we perform a field experiment, supported by a numerical simulation, based on coupling the lidar and LII-techniques, to retrieve a range-resolved profile of LII-thermal radiation emitted by the carbon of an urban atmosphere over several hundred meters.

For paper [2], the starting point is the definition of the lidar backscattering coefficient, showing that small-sized particles may strongly contribute to the backscattering coefficient when present in high concentrations, as implied by the detection of molecular backscattering with a lidar. Hence, and as a second point, we here discuss on the optical requirements ensuring a lidar backscattering device to remotely observe new particle formation, as we published in [2]. Considering a dust NPF-event, we applied the Mie and T-matrix numerical codes to simulate the backscattering coefficient of spherical freshly nucleated particles and non-spherical desert dust particles. We hence showed that, to remotely observe such nucleation events promoted by non-spherical particles with an elastic lidar, it should operate in the UV spectral range and be polarization-resolved. Two atmospheric case studies are then proposed, on nucleation events promoted by desert dust or volcanic ash particles.

The paper is organized as follows. In Section 2, we present the numerical simulations and the methodology of our two papers [1, 2] by starting with the carbon aerosol lidar methodology. We then present lidar field application case studies for both papers in Section 3 and then conclude.

## 2. NUMERICAL SIMULATIONS AND METHODOLOGY

We here focus on the optical requirements ensuring a lidar device to be sensitive to the carbon aerosol (Section 2.1, [1]), then to particles nucleation (Section 2.2, [2]).

### 2.1 LII-lidar formalism and numerical simulation

Starting from Planck and Kirchhoff's laws, we address the thermal radiation intensity emitted by a laser-illuminated particle volume at a range R from a lidar laser source. For incandescence occurs during a characteristic time  $\tau$ , the LII-lidar signal takes the mathematical form of a convolution product in which the Planck's distribution is involved. We then addressed the main requirements for operating the atmospheric LII-lidar methodology in the low troposphere by numerically simulating the LII-lidar equation. To observe an LII-lidar signal in the low troposphere, the laser beam divergence should be very low (in the range of 0.05 mrad). The numerical simulation of the LII-lidar signal (see Fig. 1) shows that, even when the carbon aerosol has a simple flat-top vertical profile, the interpretation of the LII-lidar signal is not straightforward. More details are given in [1] where the LII-lidar formalism is set and the LII-lidar equation is established.

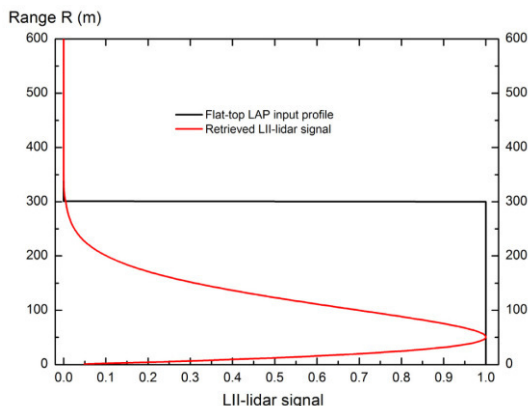


Fig. 1 Numerical simulation of the LII-lidar signal for an input flat-top carbon aerosol vertical profile [1].

### 2.2 Methodology for remotely sensing a NPF-event with a lidar backscattering device

Starting from an observed NPF-event promoted by desert dust particles [10], we numerically simulated the corresponding lidar particles backscattering coefficient  $\beta_{\text{NPF}}$ , by applying the

Mie theory to freshly nucleated sulfuric acid particles and the T-matrix numerical code [9] for non-spherical desert dust particles. We hence retrieved the optical requirement for detecting such a nucleation event (NPF-event) promoted by desert dust particles, for which the lidar backscattering coefficient  $\beta_{\text{NPF}}$  is equal to  $0.7 \text{ Mm}^{-1} \cdot \text{sr}^{-1}$  at the onset of the nucleation process. To highlight the contribution of each particle size to  $\beta_{\text{NPF}}$ , we plotted the integrand of  $\beta_{\text{NPF}}$  in Fig. 2. In the UV spectral range, particles for diameters between 50 and 800 nm contribute to  $\beta_{\text{NPF}}$ , which is hence interestingly increased ( $\beta_{\text{NPF}}$  is four times higher than in the IR-spectral range).

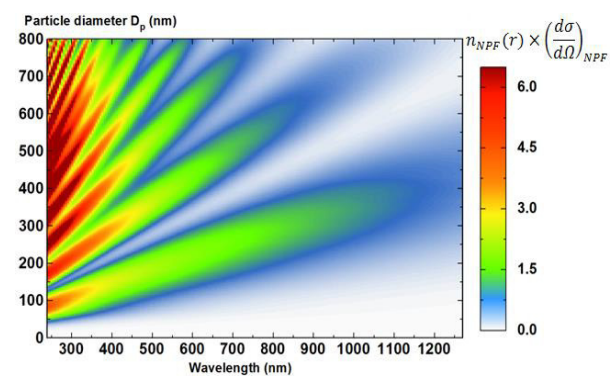


Fig. 2 Numerical simulation of  $\beta_{\text{NPF}}$ 's integrand [2].

Since nucleation events promoted by desert dust particles appear at low dust concentrations [11], the correlative behavior of  $\beta_{\text{NPF}}$  with that of  $\beta_{\text{dust}}$  can be used as optical tracers for remote sensing such NPF-events. Hence,  $\beta_{\text{NPF}}$  can be addressed by using a sensitive and accurate UV polarization lidar. The polarization acts as a particle shape discriminator, allowing backscattering separation of spherical particles from any other non-spherical particle, whatever its size. Besides (see Fig.2), the wavelength acts as a particle size discriminator: the shorter the wavelength is, the more sensitive to fine and ultrafine particles backscattering is.

## 3. FIELD APPLICATION CASE STUDIES

In this section, we report on the first experimental observation [1, 2] of LII-lidar thermal radiation in an urban atmosphere (Section 3.1), of lidar remote sensing of particles nucleation (Section 3.2).

### 3.1 Observation of a LII-lidar vertical profile

Two experimental detector configurations are proposed, allowing either spectrally-resolved LII-

lidar measurements over a range interval or range-resolved LII-lidar measurements at a single detection wavelength (LII-lidar vertical profile). The latter configuration is coupled with an elastic backscattering lidar to address all atmospheric aerosols, either refractory, which may incandesce, or not. Fig. 3a presents the observed LII-lidar signal as a function of the wavelength of the detected radiation. Interestingly, apart from observed Raman vibrational sidebands of water vapor and nitrogen, a continuous signal increase can be adjusted in nice agreement with Planck's law [1]. Fig. 3b presents the corresponding range-resolved LII-lidar signal measured in the Lyon urban troposphere, over several hundred of meters, by using a 1064 nm lidar laser, to avoid superposition with Raman sidebands (down-conversion process).

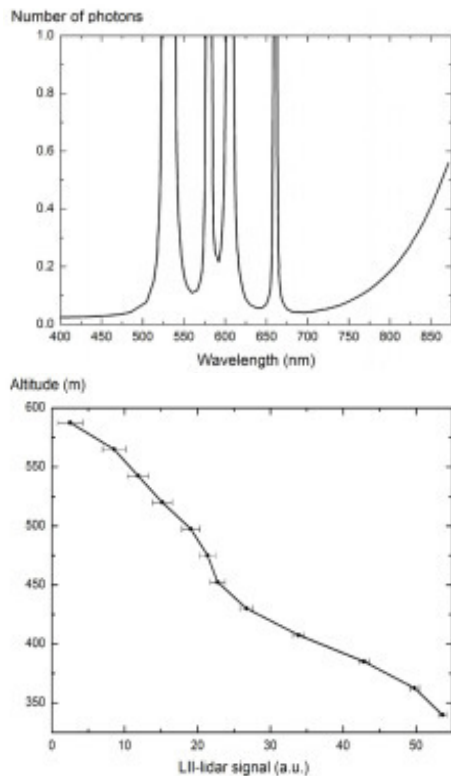


Fig. 3 (a): Measurement of LII-lidar signal spectrum [1], in agreement with Planck's law. (b): Vertical profile of LII-lidar signal in Lyon urban city's troposphere [1].

### 3.2 Lidar observation of a nucleation event

To apply Section 2.2's methodology, a sensitive and accurate UV-polarization lidar is required since the more accurate the polarization analysis

is, the more sensitive to  $\beta_{\text{NPF}}$  the lidar is. We hence operated our home-built UV-polarization lidar [11]. By applying the methodology we published in GRL [12], we obtained the Figure 4 time-altitude maps for the spherical ( $\beta_s$ ) and non-spherical desert dust ( $\beta_{\text{ns}}$ ) lidar backscattering coefficients.

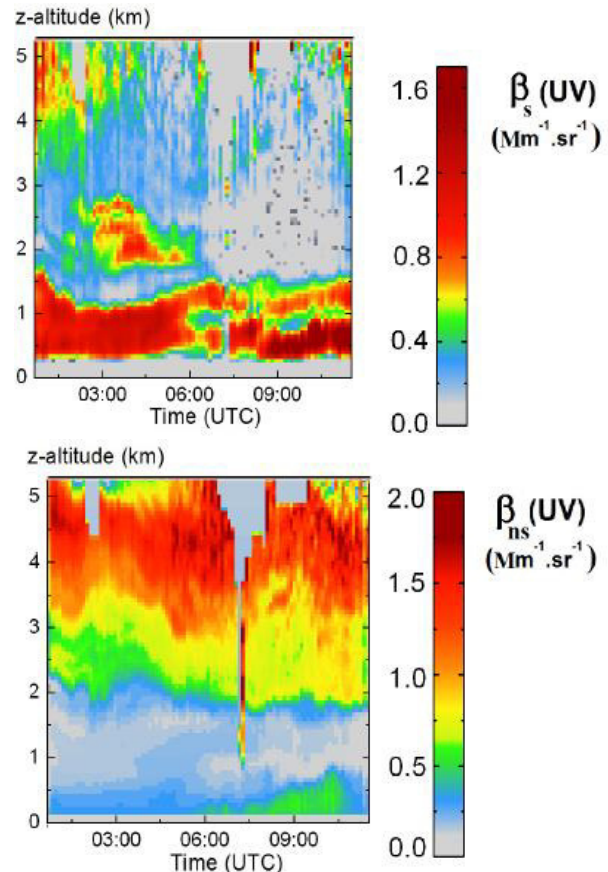


Fig. 4 Time-altitude maps of the spherical and non-spherical lidar particles backscattering coefficients [2].

Interestingly, s-particles are only observed at the border of the dust ns-layer, i.e. a  $\beta_s$ -enhancement only occurs at low dust particles backscattering, in agreement with our numerical simulation [2]. This behavior can a priori result from nucleation, coagulation or condensation on pre-existing surfaces. However, nucleation is the only one leading to such an s-particles backscattering enhancement, occurring only at low dust particles backscattering. Condensation may also occur and for this reason, it is only when  $\beta_{\text{ns}}$  is low that the  $\beta_s$ -increase is indeed observed. Hygroscopic growth may also be responsible for the observed  $\beta_s$ -enhancement but in our case study, the relative humidity remains constant (ECMWF-analysis).

Coagulation might be also invoked, but in this case, photochemistry would not be implied, in contradiction with Dupart et al.'s laboratory experiments [10]. As a result, our numerical simulation and lidar experiments provide strong arguments in favor of the remote lidar observation of a nucleation event promoted by dust particles. Actually, the state-of-the-art atmospheric science does not provide other processes that may induce such an optical behavior of a  $\beta_s$ -enhancement occurring only at low dust concentrations. More details on our newly developed methodology and its limitations are discussed in [2]. In addition, a second case study is proposed on the lidar observation of nucleation events promoted by ashes, released from the Icelandic volcano.

#### 4. CONCLUSIONS

In this contribution, we presented the results of two recently published lidar new methodologies, to detect the carbon aerosol [1] or particles nucleation [2], the latter being awarded as June 2014 OSA Spotlight. More details and outlooks are proposed in the corresponding papers. The oral presentation will focus on the implications of these new methodologies for the lidar community.

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