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Pump-probe differential Lidar to quantify atmospheric supersaturation and particle-forming trace gases

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Abstract We propose a pump-probe differential Lidar technique to remotely map the ability of the atmosphere to undergo particle condensation, which depends on the concentration of both pre-existing nanoparticles and condensable species or their precursors. Besides its interest for improving short-time, local weather forecast, this technique could provide information on atmospheric parameters such as the concentration of condensable species and physical parameters including the temperature and relative humidity.

1 Introduction

The atmospheric physico-chemistry belongs to the most complex natural systems, owing on the very large number of both physical parameters and chemical species at play, including a bunch of highly reactive ions, radicals, and molecules present at the trace level. Among the atmospheric processes, aerosol particle nucleation and growth are particularly complex, governed by many parameters, including temperature (T), relative humidity (RH), the availability of condensation nuclei, as well as condensable trace gases like mineral or organic acids [1, 2].

Such complexity makes even short-term weather forecast based on modeling the atmospheric physico-chemistry a challenging task. Such dynamics is strongly influenced by pre-existing natural nanometer-sized particles and many gaseous trace species of high reactivity, as well as by gaseous precursors of these condensable species, that could be activated chemically or photo-chemically by the sunlight and/or by the laser light. Besides their key relevance in the atmospheric processes, the high reactivity of many of those species keeps their concentration low, making it difficult to detect and quantify them.

Acknowledging the impossibility to control, or even measure, all atmospheric parameters relevant to condensation, and hence to model this process satisfactorily, we propose an alternative pragmatic approach. We probe the ability of the atmosphere to locally give rise to particle formation by observing the yield of laser-induced condensation [3, 4] based on the filamentation of ultrashort laser pulses [5–9]. This ability is strongly influenced by its contents in pre-existing nanometric particles, as well as in particle-forming trace gases and their precursors.

As we recently showed that the effect of the laser mainly consists in enhancing natural phenomena, as opposed to opening fully new condensation pathways [10], the local yield in laser-induced particles provides a relevant probe into the natural potential of the probed air mass to produce condensation under appropriate conditions. As such, it can be considered as bearing composite information on all physical and chemical parameters influencing photochemically new particle formation.

Furthermore, we propose to use this signal to extract physical or chemical parameters (T, RH, pre-existing nanoparticles, volatile organic compound (VOC)

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concentration…) that influence laser-induced condensation [10, 11].

More specifically, we evaluate the potential of remotely probing the laser-induced condensation by pump-probe Light Detection and Ranging (Lidar) [12]. Lidar is a powerful optical remote sensing technique analogous to optical radar [13, 14]. It consists in emitting light pulses and detecting the light backscattered by the atmosphere as a function of the light time-of-flight, i.e., of the measurement distance z. The retrieved signal is proportional to

$$S(z) \propto \frac{\xi(z)}{z^2}\beta(z)e^{-2\int_0^z \alpha(z')dz'} \tag{1}$$

where $\xi(z)$ is the geometrical overlap term between the volumes covered by the light emitter and the field of view of the collection system, and α and β are the extinction and backscattering coefficients, respectively. They stem from scattering and absorption (resp., backscattering), of both molecules and aerosol particles, therefore bearing information on the atmospheric composition.

This technique was already used in the first demonstration of the effect of the laser in the atmosphere [3]. Here, instead of measuring the aerosol particle production to characterize the effect of the laser, we use the latter, which has been much better documented since then, to characterize the atmosphere in the volume illuminated by the laser, as illustrated in Fig. 1.

We evaluate the feasibility of such pump-probe Lidar, based on numerical simulations. Furthermore, we discuss the system sensitivity to noise, in order to assess for its practical usability in applications. We also briefly discuss potential improvements brought by multiwavelength systems, for improved precision as well as multiparameter detection.

2 Methods

We simulated a pump-probe Lidar detecting laser-induced particles by using Eq. (1), where α and β have been evaluated based on the Rayleigh–Mie theory for each considered aerosol particle size distribution.

We then calculated the differential signal by substracting the Lidar signal immediately following the pump (filamenting) pulse (S_{ON}) and the reference conditions (S_{OFF}) that correspond to different aerosol particle size distributions and particle densities:

$$\begin{aligned} \Delta S(z) = S_{ON}(z) - S_{OFF}(z) \propto \frac{1}{z^2}\big(\beta_R(z) + \beta_{M,back}(z) \\ +\beta_{M,laser}(z)\big)e^{-2\int_0^z \left(\alpha_{M,back}(z)+\alpha_{M,laser}(z)\right)dz'} \\ -\frac{1}{z^2}\big(\beta_R(z) + \beta_{M,back}(z)\big)e^{-2\int_0^z \alpha_{M,back}(z)dz'} \end{aligned} \tag{2}$$

where the subscripts R, M,*back*, and M,*laser* stand for the Rayleigh, background Mie, and laser-induced Mie contributions, respectively, and the Rayleigh contribution to the extinction has been neglected [13]. The Mie contribution depends on the concentration and size distribution of the atmospheric particles, both from the background and laser-generated. They depend in turn on T and RH, as well as on the concentration of condensable species in the atmosphere.

For each temperature and relative humidity, the Rayleigh contribution β_R to the backscattering was calculated from the density of air molecules, while the aerosol contribution to α and β has been computed based on the size distributions and particle densities measured previously [4], assuming spherical transparent particles with a refractive index of $n = 1.33$ and using the Mie theory [15] to compute their optical properties.

Having simulated the differential Lidar signal, at one or several wavelengths, we inverted it using nonlinear least-

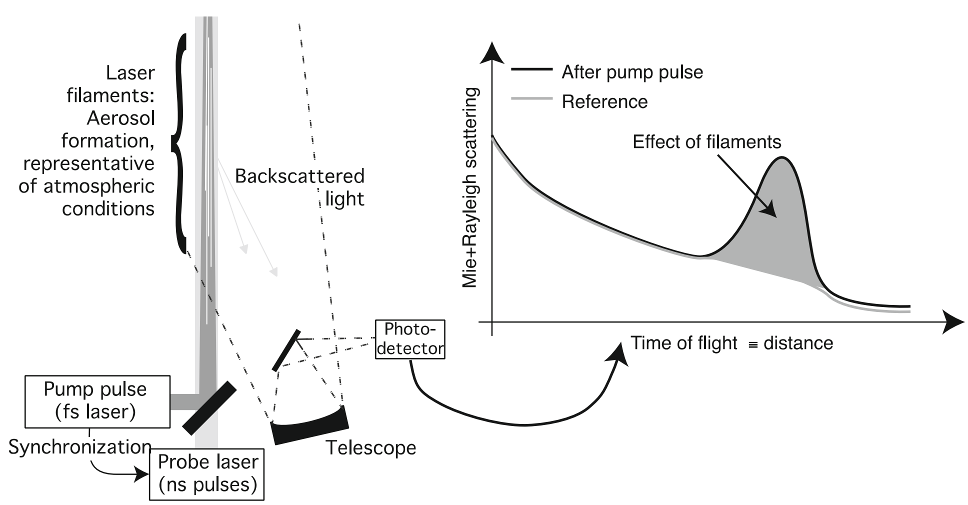

Fig. 1 Principle of the pump-probe Lidar

squares curve fitting to recover the particle number concentration produced by the laser. This laser-induced particle concentration is used as a proxy for the local concentration of condensable trace gases. The robustness of the technique was tested by adding a white noise to the simulated Lidar signal before its inversion, and assessing the influence of this noise on the retrieved signal.

Based on the strong influence of T and RH on laser-induced particle formation, we also investigated the feasibility of multiparameter remote sensing of the atmosphere by addressing the simultaneous measurement of T and RH. In that purpose, T and RH profiles were postulated. The associated size distribution in the presence and absence of laser filaments was extracted and linearized from field experiment data [4], and the differential Lidar signal was simulated as specified above. It was then inverted, and its robustness was tested against noise using a similar procedure as for the single-parameter case.

3 Results and discussion

Figure 2 illustrates the power of pump-probe Lidar. The initially postulated laser-induced particle concentration (Panel b) and the differential Lidar signal (Panel c) are perfectly retrieved by the simulated pump-probe Lidar experiment. Simulations at 1,064, 532, and 366 nm all yield an error on the particle concentration below 0.01 %.

However, the main limitation for the practical realization of such experiment is not the mathematical inversion of the Lidar equation. Rather, it resides in the sensitivity to the noise intrinsic to any Lidar measurement. This is especially important at longer distances where the $1/R^2$ dependence drastically decreases the signal-to-noise ratio. As shown in Fig. 3a, the retrieved laser-induced particle density remains within 6 % of the input one over 650 m for a noise level of 1 % of the peak signal, typically corresponding to a realistic noise of 0.1–1 mV in an actual experiment (Fig. 3a). Our simulations therefore suggest that pump-probe differential Lidar could efficiently probe the atmospheric condensability within the km range.

Beyond the condensability of the atmosphere, our technique may even allow to estimate atmospheric parameters such as T, RH, or the concentration of trace species like α-pinene having a strong influence on laser-induced condensation [10]. Furthermore, multiwavelength experiments could even allow to get information about the size distribution of the laser-generated particles, and hence on the atmospheric status itself. We illustrate this possibility in Fig. 4, where a typical two-wavelength Lidar

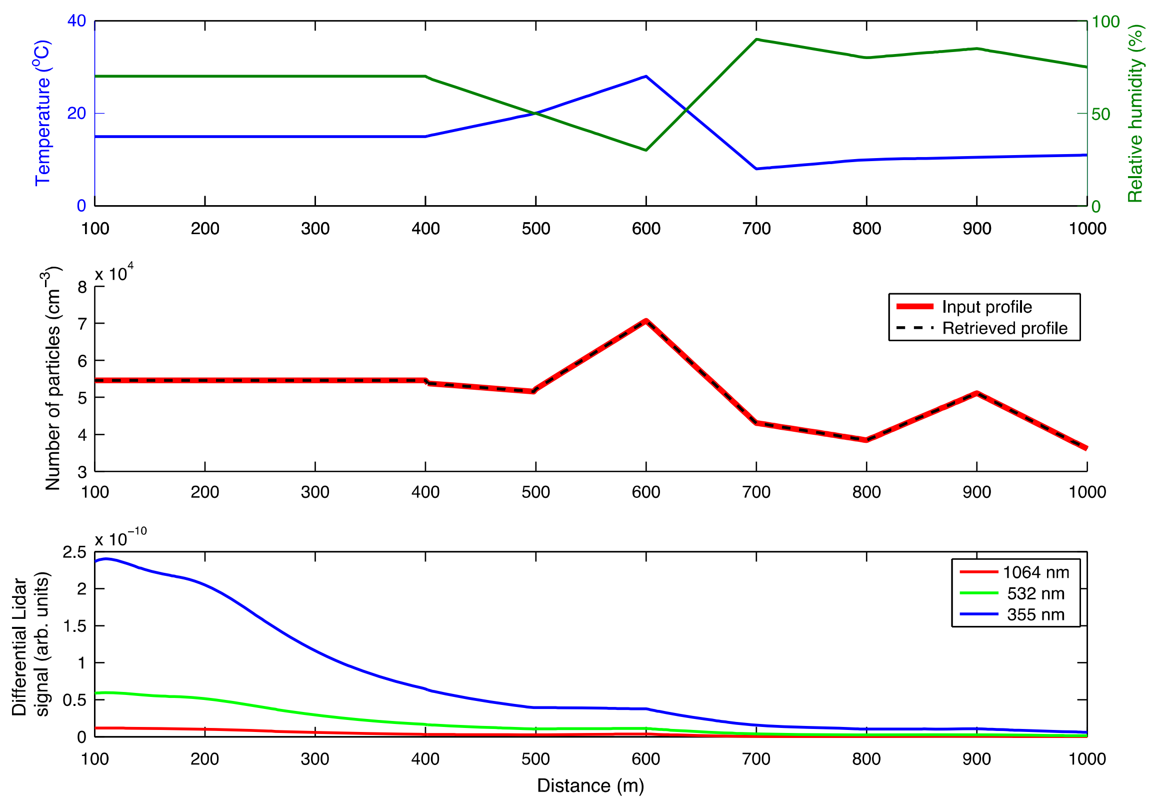

Fig. 2 Pump-probe Lidar for atmospheric condensability. **a** Input temperature and relative humidity used in the calculation; **b** corresponding input and retrieved profiles of atmospheric laser-induced condensation; **c** corresponding simulated differential Lidar signals

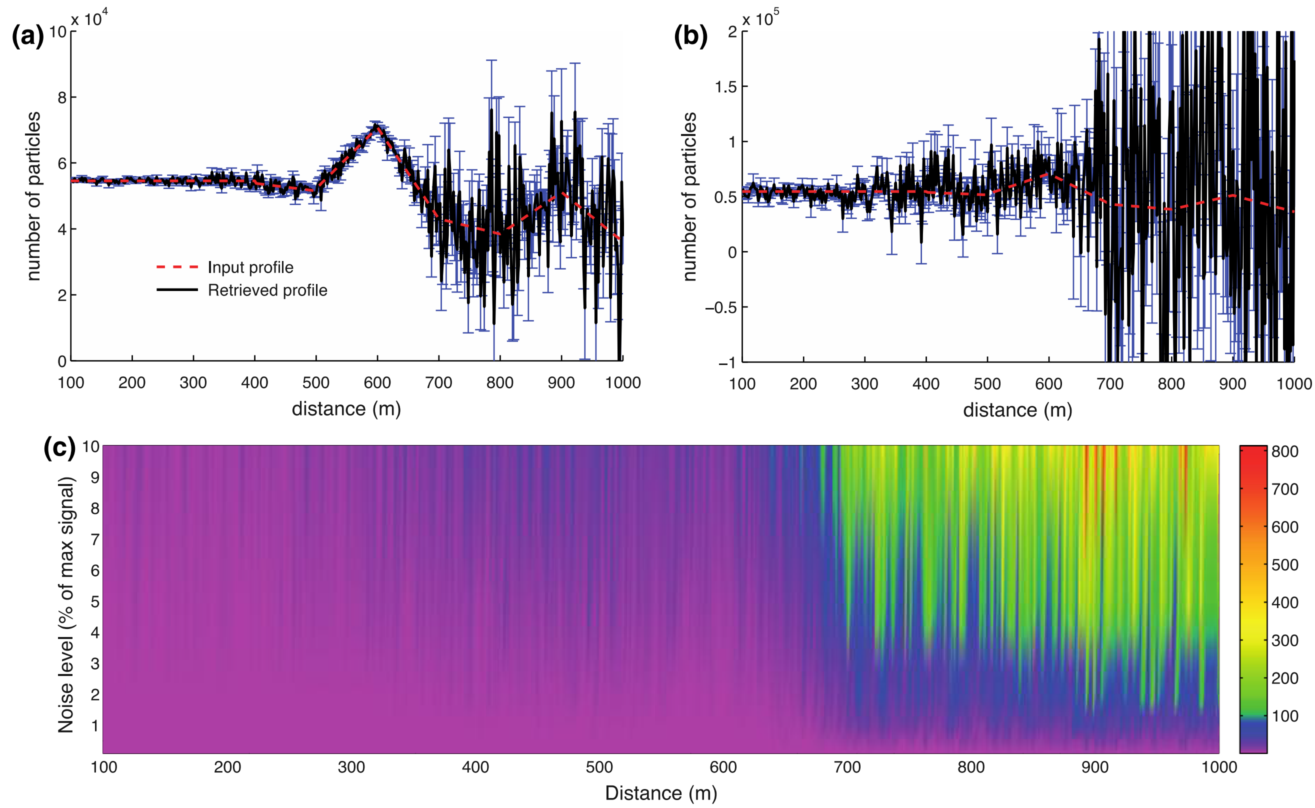

Fig. 3 Influence of noise on the inversion of differential pump-probe Lidar. **a**, **b** Average and standard deviation of the condensability profile retrieved from the simulated differential Lidar at 1,064 nm over 20 realizations with an added white noise of **a** 1 % and **b** 10 % of the maximum Lidar signal. **c** Relative error (standard deviation normalized to the original value) in the retrieval as a function of distance and noise level (relative to the maximum of simulated Lidar signal)

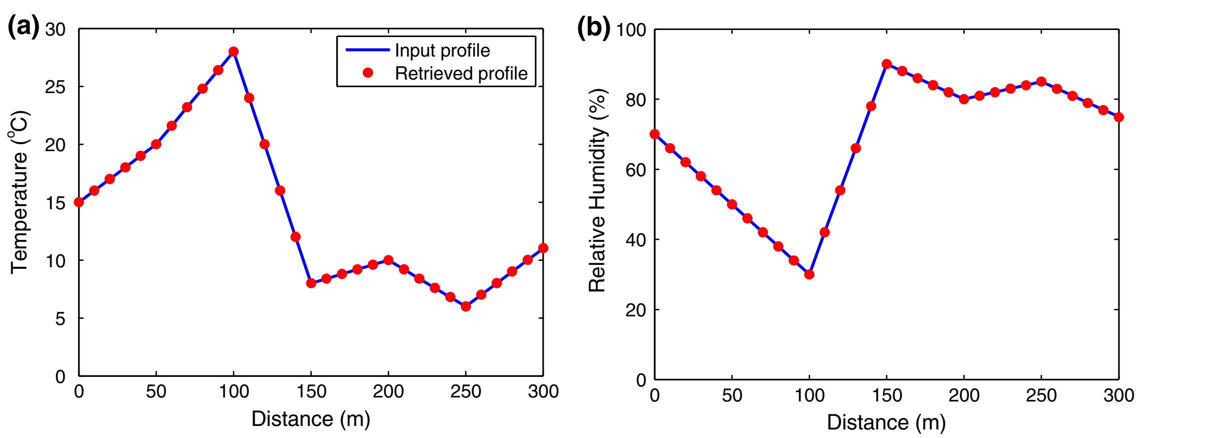

Fig. 4 Simulation of a pump-probe Lidar measurement of T and RH via the two-wavelength (532 and 355 nm) measurement of the atmosphere ability to give rise to particle condensation. **a** Temperature; **b** relative humidity profiles

inversion is used to retrieve arbitrary T and RH profiles, based on the procedure described above. Due to the different influence of T and RH on the laser-induced particle size distribution, and to the different spectral dependence of each particle size, it is indeed possible to retrieve both parameters over several hundreds of meters. In spite of the very rough variations of the input T and RH, the profiles are retrieved within 0.3 % over 1 km distance.

This technique is, however, pretty sensitive to noise, as illustrated in Fig. 5. Adding a random white noise of 0.1 % of the signal maximum onto the whole Lidar signal before performing the data inversion does not affect the inversion

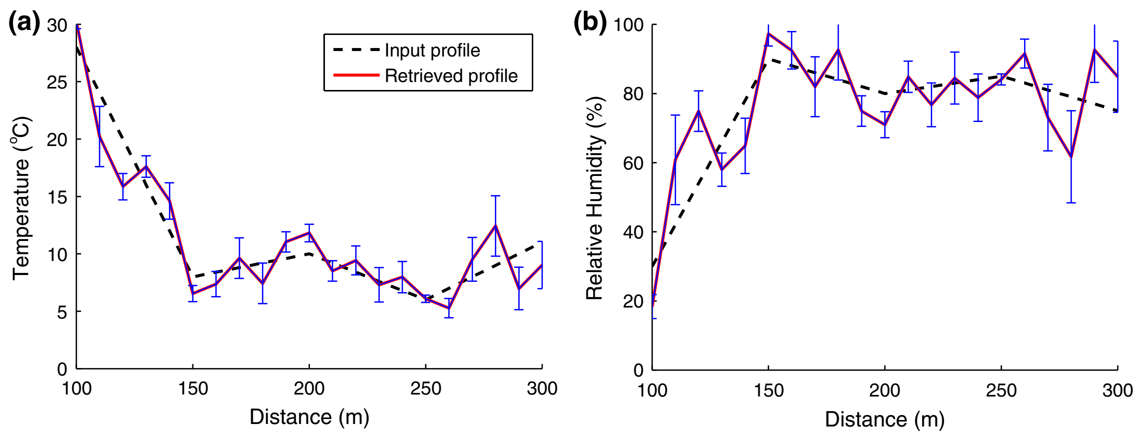

Fig. 5 Simulated retrieval of **a** temperature and **b** relative humidity profiles by two-color (532 and 355 nm) pump-probe differential Lidar, with the addition of a white noise of 0.1 % of the peak intensity

significantly. However, with 1 % noise, the error on temperature reaches up to 10 °C, while the 40–50 % error on RH virtually jeopardizes the relative humidity estimation. This sensitivity to noise is due to the fact that T and RH influence much more the laser-induced particle number than the size distribution, especially in the particle size range that mostly contributes to scattering (~2.5 µm) [4]. As a consequence, inverse changes in T and RH partially compensate each other over a wide range.

A more precise knowledge of the dependence of the size distribution of laser-induced particles with T, RH, or other atmospheric parameters including pre-existing nanoparticles or the concentration of individual VOC species or families would open the perspective of more specific measurements. Since the scattering properties of particles are strongly size- and wavelength-dependent, their impact on different probe wavelengths will be different. One can therefore expect that multiple species can be measured simultaneously if the pump-probe Lidar proposed here is generalized to the use of several wavelengths. This has been exemplified with the measurement of T and RH. Such measurement remains possible provided the noise measurement is kept low.

4 Conclusion

As a conclusion, we have proposed a pump-probe Lidar technique to measure the new particle formation potential of the atmosphere. It opens perspective to gain pragmatic overall information about an important aspect of the atmospheric state without a need to characterize individual physicochemical parameters. Furthermore, if associated with adequate calibration of the influence of each atmospheric parameter on the laser-induced particle size distribution, it may open appealing prospects for retrieving information on some of these individual atmospheric parameters such as the abundance and size of pre-existing nanometric aerosols, the concentration of condensable trace gas or their precursors, temperature, or relative humidity. Prospects for multiparameter measurements are also proposed, based on a multiwavelength probe, although such approach is much more sensitive to noise. Such concepts could improve the atmospheric diagnostics.

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