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SAMIR ZEGHBIB

SAMIA BOUZID

ABDELHAFID CHAABI

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Quantitative characterization of atmospheric desert particles via multispectral LiDAR method (UV to IR), using genetic algorithms and compared with Chomette's mode

Samir ZEGHBIB^{1,*}, Samia BOUZID², Abdelhafid CHAABI¹

¹University of Constantine, Faculty of Engineering, Institute of Electronics

Semiconductor and Ultra High Frequency Laboratory, Constantine 25000, Algeria

²University of Constantine, Faculty of Medicine, Research Unit, Laboratory of Lasers and Applications

PB 125, Chalet des pins, Constantine 25000, Algeria

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Abstract: Size distribution of desert aerosols was measured via LiDAR multi-spectral method using laser wavelengths from UV to mid-IR $\lambda = 0.193, 0.694, 1.064, 3.370$ and $10.60 \mu\text{m}$, and applying statistical Markov chains and genetic algorithms. Our results are compared later with the well-known Chomette's desert model.

Key words: Desert aerosols, LiDAR, LiDAR via impaction, Klett inversion, Mie theory, Chomette's desert model, genetic algorithms.

1. Introduction

Monitoring of air quality necessarily involves the quantitative knowledge of gaseous agents, solids, as well as organic and bacteriological agents in our environment, together with the associated physical chemistry.

This knowledge must be resolved in time and space in order together to answer public concerns in terms of health and to reach decision on policy. An interesting approach is the use of optical techniques, particularly LiDAR technique. This method allows three-dimensional maps of the main pollutants in the atmosphere, particularly atmospheric aerosols which play an important role in the balance of terrestrial radiation.

While gas pollution is now beginning to be well measured, aerosols are very difficult to measure in a meaningful way. Indeed, the parameters needed to characterize aerosols are much more demanding than for gaseous pollutants. Furthermore, to understand the evolution of concentration over time we must know the size distribution and nature of the aerosol particles present at each point. This information is essential to predict the effect of aerosols on atmospheric physical chemistry and subsequently health.

Among the components of tropospheric aerosol, the desert aerosol emitted by arid surfaces of the sphere represents the main components, with a proportion of approximately 43% [1] of the total mass of aerosol produced per year, including natural and anthropogenic sources.

The contents of atmospheric aerosols found in desert regions are more variable and complex. Properties and abundance of the particles can vary largely in space and in time. This great diversity and variability are from several theoretical models which describe radiative and physicochemical parameters of the aerosols

*Correspondence: sept7_sam@yahoo.fr

(absorption, diffusion and extinction coefficients, etc.). Among these, the most well-known is Chomette's model [2].

Determination of air quality as a function of particulate suspension requires knowledge of state of the atmosphere throughout the distribution and over time. For that, various instruments and systems are used, principle of which is the optical LiDAR method (Light Detection and Ranging). An original LiDAR method using a single wavelength has been developed. It employs the technique of micro analyzing X-rays of impacted filters. This method allows access to the distribution of particle size, but it can characterize the air's contents only at location of sampling. To gain access to the geographical distribution of aerosols spatially and temporally, and to distinguish between large and small particles, we use a multi-spectral LiDAR optical system with spectrum IR to UV. This method has great potential to determine the distributions of aerosol size and refractive indices from LiDAR measurements at several wavelengths without making assumptions a priori about the shape of the distribution, which was the case for other methods. The choice of wavelengths used is very important because the simulation algorithm is significantly affected as the particle size approaches that of the wavelength.

Our objective was the study of desert particles in which we performed a detection of their size distributions using the multispectral LiDAR method with fixed wavelengths $\lambda = 0.193, 0.694, 1.064, 3.370$ and $10.60 \mu\text{m}$. The extinction coefficients used in the LiDAR profiles in this study are taken from the model of Chomette. We must apply these profiles, which we consider as measurements, via statistical methods in order to extract the size distribution of aerosols. Genetic algorithms are the optimization search method used.

1.1. Desert model of Chomette

Chomette has studied the cycle of desert aerosols, from which he constructed a mesoscale model [2]. Chomette explored the phenomena of transport, deposit and influence on the microphysics and radiative properties of desert aerosols. The extent of desert aerosol distribution is very little influenced by the mineralogical composition [3]. The transport process, even over long distances, does not affect the mineralogical composition of the aerosol as long as it occurs in a dry atmosphere [4], which implies a weak evolution of the complex index of refraction over time.

In order to model desert aerosols, a trimodal lognormal distribution was considered. On the other hand, only one complex index of refraction characterizes the totality of particle mixtures. These indices are taken from Volz [5] and Patterson [6] in the infrared and from Grams [7] in the visible.

During transport in dry atmosphere, the granulometry of desert aerosol evolves via sedimentation (per the effect of gravity), affecting mainly large particles. As one moves away from sources the granulometric spectrum evolves then to the finest particles. Two granulometries obtained from in situ measurements [8] were selected in order to describe this behaviour. The distribution of *wind carrying dust* (WCD) characterizes the desert aerosol transported above the continent following uprising. Desert aerosols away from the sources are characterized by the distribution *background* (BG), a fine granulometry with few large particles compared to the distribution WCD.

A second granulometry source included in this modelling, denoted as A1, comes from the results from Alfaro [3] on the suspension of mineral particles ejected by blower, using different soil types (Sahelian and Saharan origins). To complete this granulometric description, a second distribution (A2) is deduced from A1 by modelling the sedimentation of the large particles of A1 during transport away from sources.

2. LiDAR principal

Aerosol Cloud LiDAR (ACL) is an atmospheric remote sensing instrument that allows measurement and characterization of the optical characteristics and microphysics of clouds and aerosols present in the atmosphere [9]. A laser directs and sends pulses of light into the air; subsequent backscatter is collected and measured as a function of time (see Figure 1). The laser light is both diffused and absorbed by the atmosphere and embedded aerosols. Extinction increases with aerosol density and molecular concentration, while a fraction of light is backscattered towards the light source.

In order to increase the solid angle of reception, a telescope and the laser are mounted coaxially. The laser is pulsed and thus allows ranging of a measurement via the relationship $z = ct/2$, where z is the distance, t is the round-trip time of the light and c is the speed of light in the air.

The backscattered light intensity, $I(z, \lambda)$, which is a function of wavelength λ and distance z , is expressed in the case of elastic diffusion (neglecting multiple diffusion) [10, 11] as:

$$I(z, \lambda) = I_0(0, \lambda) \frac{A_0}{z^2} \beta(z, \lambda) \Delta z \chi(z, \lambda) \exp\left(-2 \int \alpha_{ext}(z, \lambda) dz\right), \quad (1)$$

where $I_0(0, \lambda)$ is the light intensity emitted with the wavelength λ by the laser; A_0/z^2 is the solid angle of acceptance of the optical receiver (A_0 is, for example, the effective surface detection of the telescope); $\beta(z, \lambda)$ is the total coefficient of backscatter volume; $\Delta z = c\Delta\tau/2$ with c is speed of light and $\Delta\tau$ is the impulse duration of the laser; $\chi(z, \lambda)$ is the detection effectiveness; $\alpha_{ext}(z, \lambda)$ is the total coefficient of extinction volume; and $\exp(-2 \int \alpha_{ext}(z, \lambda) dz)$ expresses the Beer-Lambert law on the light propagation between 0 and z .

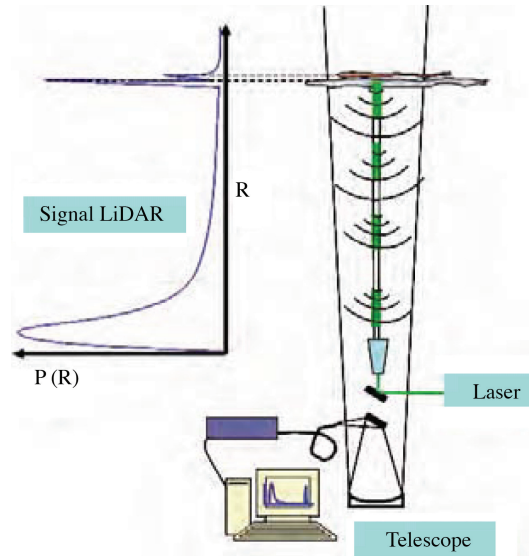


Figure 1. General diagram illustrating a LiDAR system.

3. Measurement by LiDAR

In order to give detailed information on the distribution of particles size, their concentration and composition, we give three methods.

3.1. LiDAR + sample acquisition by impaction

Frejafon et al. [12] have used LiDAR together with scanning-electronic microscopy (SEM) and X-ray micro-analysis to characterize atmospheric aerosols. This method enables us to have a normalized distribution and an analysis of the concentration and the composition of the particles on the ground, but it does not give access to the geographical distribution of aerosols.

3.2. LiDAR measurement at distance with one wavelength

This monospectral optical method allows us to know the geographical repartition of aerosols as well as their dynamics over distance and time within an atmospheric layer [13]. It allows cartographic mapping in three dimensions the concentration in atmospheric aerosols and follow its temporal evolution. The coefficients of extinction and backscatter can be rewritten as:

$$\alpha_{ext}^{Aero}(\lambda) = \int \sigma_{ext}^{Aero}(\lambda, r) \rho(r) dr = \int \pi r^2 Q_{ext} \rho(r) dr, \quad (2)$$

$$\beta_{back}^{Aero}(\lambda) = \int \sigma_{back}^{Aero}(\lambda, r) \rho(r) dr = \int \pi r^2 Q_{back} \rho(r) dr, \quad (3)$$

where, α_{ext}^{Aero} and β_{back}^{Aero} are, respectively, the coefficients of extinction and backscatter. They are calculated by using the theory of Mie [14]:

$$\sigma_{ext}^{Aero} = \frac{2\pi}{K^2} \sum_{p=1}^{+\infty} (2p + 1) Re(a_p + b_p), \quad (4)$$

$$\sigma_{back}^{Aero} = \frac{2\pi}{K^2} \left| \sum_{p=1}^{+\infty} (2p + 1) (-1)^p (a_p + b_p) \right|^2, \quad (5)$$

where, diffusion coefficients a_p and b_p are defined by the Ricatti-Bessel and Hankel functions; Q_{ext} and Q_{back} are, respectively, the effectiveness of extinction (ext) and backscatter (back); $\rho(r)$ is the existing distribution of atmospheric particles which can be written as an unspecified function of distribution. For the aerosols, the distributions suggested are the Djermendjian distribution [15] or lognormal distribution [16]. But with this traditional LiDAR technique using only one wavelength it is not possible to scale with the concentration of the various types of atmospheric aerosols. For that, it is necessary to use multispectral LiDAR systems, which as more difficult to implement.

3.3. Method using multiple wavelengths

This method is an inversion technique employing methods of genetic algorithms; it is used to determine the size distribution of aerosols and the distribution of refraction indices via LiDAR measurements over multiple wavelengths, without an a priori hypothesis about the distribution form, unlike the method described in Section 3.2. In this method, $\rho(r) = N_0 \cdot g(r)$, where N_0 is whole particle density and $g(r)$ is a function of the normalized probability distribution. If $g(r)$ is supposed to be limited inside a defined interval ($r_{\min} \leq r \leq r_{\max}$), integral equation (2) can then be approximated by a sum on a discrete number M of rays:

$$\alpha_{ext}^{Aero}(\lambda) = N_0 \sum_{i=1}^M \overline{K_{ext}(r_i, \lambda)} f(r_i), \quad (6)$$

where

$$\overline{K_{ext}(r_i, \lambda)} = \frac{1}{\delta r} \int_{r_i - \delta r/2}^{r_i + \delta r/2} K_{ext}(r, \lambda) dr, \tag{7}$$

is the average of the function on the interval of width δr centred on r_i and $f(r_i)$ is the fraction of the particles in the same interval. Also,

$$K_{ext} = \pi r^2 Q_{ext}^{Aero}(r, \lambda). \tag{8}$$

In the same way, the coefficient of backscatter can be written as:

$$\beta^{Aero}(\lambda) = N_0 \sum_{i=1}^M \overline{K_{back}(r_i, \lambda)} f(r_i), \tag{9}$$

$$\overline{K_{back}(r_i, \lambda)} = \frac{1}{\delta r} \int_{r_i - \delta r/2}^{r_i + \delta r/2} K_{back}(r, \lambda) dr, \tag{10}$$

$$K_{back} = \pi r^2 Q_{Back}^{Aero}(r, \lambda). \tag{11}$$

The number M of intervals used to solve equations (6) and (9) depends on the number N of wavelengths employed during measurement. It is maximally equal to double this number. Associated to each wavelength are two parameters: α_{ext} and β . The difficulties in inverting equations (6) stem from the multiple values for the nucleons of diffusion and solutions which are not singular. In order to stabilize the solutions for areas specific of data, we impose that the solutions be positive.

In order to solve (6), we express it as a linear (matrix) system:

$$\begin{bmatrix} \alpha_1 \\ \alpha_2 \\ \alpha_3 \\ \cdot \\ \cdot \\ \cdot \\ \cdot \\ \alpha_N \end{bmatrix} = N_0 \cdot \begin{bmatrix} K_1^1 & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & K_1^M \\ \cdot & & & & & & & \\ \cdot & & & & & & & \\ \cdot & & & & & & & \\ \cdot & & & & & & & \\ \cdot & & & & & & & \\ \cdot & & & & & & & \\ K_N^1 & \cdot & \cdot & \cdot & \cdot & \cdot & \cdot & K_N^M \end{bmatrix} \cdot \begin{bmatrix} g_1 \\ \cdot \\ \cdot \\ \cdot \\ \cdot \\ \cdot \\ \cdot \\ g_M \end{bmatrix} \tag{12}$$

Experimental data (α_{ext} and β) and the first calculation of functions (7) allow one to solve this matrix system. A statistical approach is used based on genetic algorithms [17], which can be an effective method among problems of optimization in that it allows limited freedom from the direct treatment of errors. The initial population is a set of vectors of size distribution $[\rho_i]$, where each individual ρ_i is a distribution of discrete size on M different sizes of particles. The uniform initial population is generated in a random way and the number of individuals should not remain fixed throughout the procedure. The genetic algorithm advances from population to population by using standard genetic generators until an optimization criterion is satisfied.

4. Results and discussion

The objective of this study is the use of multispectral LiDAR method UV-Vis-IR for retrieving the size distribution of desert particles. This LiDAR method is based on a statistical method involving genetic algorithms and, unlike other monospectral methods, has great potential to determine the distributions of aerosol size and refractive indices from measurements at several wavelengths without making assumptions a priori about the shape of the distribution.

4.1. Profile of extinction coefficients

In order to determine the coefficients of extinctions at different wavelengths, rough LiDAR signals are pre-treated by multiplying them by z^2 (distance), then by taking the Napierian logarithm, i.e. $\ln(z^2)$. Subsequent values are then processed in the Klett inversion [18]. With respect to the present work, we take the profile of the extinction coefficients directly from the Chomette model as a function of wavelength [2].

Wavelengths should be well chosen so that they may be close in dimensions to particles under observation; this will enhance the sensitivity and optimisation of the results. The use of a wide interval of wavelengths in LiDAR measurements allows characterisation of a great number and size variation of detected particles. The wave lengths used are: $\lambda = 0.193, 0.694, 1.064, 3.370$ and $10.60 \mu\text{m}$.

4.2. Effectiveness factors of extinctions

Figures 2 and 3 show, respectively, the extinction effectiveness factors and extinction cross-sections in a function of distance for each type of particle, using Mie diffusion theory. For this, we used only one set of refractive indices according to Chomette's model. These indices of Chomette's model are borrowed from Volz [5] and Patterson [6] in the infra-red and from Grams [7] in the visible.

The first results of the size distribution of the aerosols by multispectral LiDAR data via genetic algorithms have been very satisfactory. In addition, we can extract two important points. First, for small particles, their effectiveness of extinction has values near zero for all wavelengths of the laser. However, optical parameters increase sharply when particle size is similar to the wavelength. The effectiveness of extinction can be linked to destructive interference that arises between particle-diffracted and non-diffracted light. The wavelength at which this occurs is related to the refractive indices. Second, the data exhibit a trend in which the extinction effectiveness factor tends to 2, suggesting the larger particles attenuate twice as much energy than expected of their cross sections.

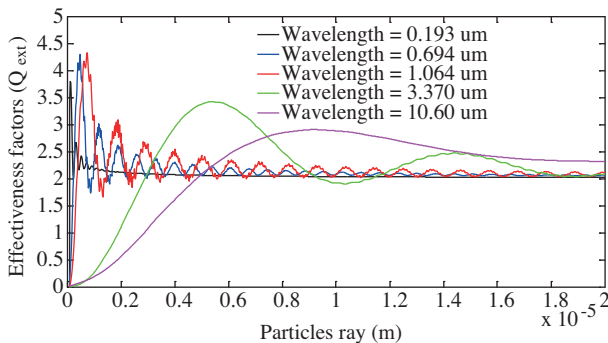


Figure 2. Extinction effectiveness factors for desert particles calculated via the theory of Mie, for a set of wavelengths $\lambda = 0.193, 0.694, 1.064, 3.370$ and $10.60 \mu\text{m}$.

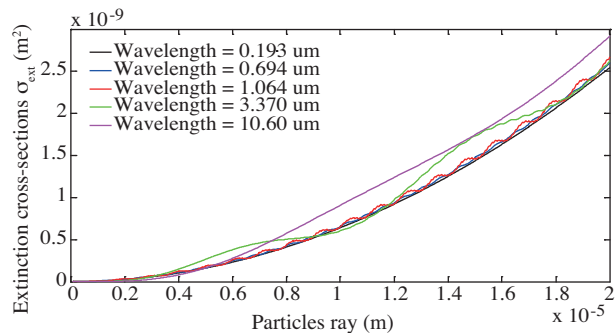


Figure 3. Extinction cross-sections of Mie, calculated versus particle ray and at several wavelengths by using Mie diffusion theory.

4.3. Size distribution of desert particles

Our multispectral LiDAR system, employing a range of five wavelengths, allows with ease precision analysis of principle aerosol particle size. Figure 4 shows a comparison of distribution sizes obtained by our multispectral LiDAR with the theoretical model developed by Chomette; we note the results show a good correlation. Both curves were fit to a Gaussian distribution, and the peak indicates the particles have a mean radius of $0.1 \mu\text{m}$.

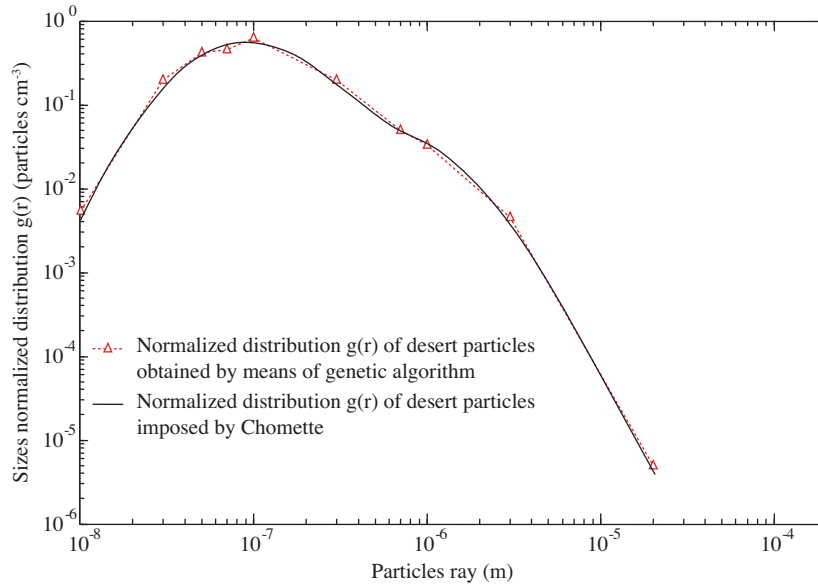


Figure 4. Comparison of size normalized distribution $g(r)$ of desert aerosols, imposed by the Chomette’s model (continuous black line) and obtained by means of genetic algorithms (red dotted line).

Finally, we validate our results by comparing in Figure 5 the extinction coefficient as a function wavelength for Chomette’s model against extinction coefficient extracted from our model. Although there is a visual difference between the two profiles, it is acceptable and further confirms the validity of our results.

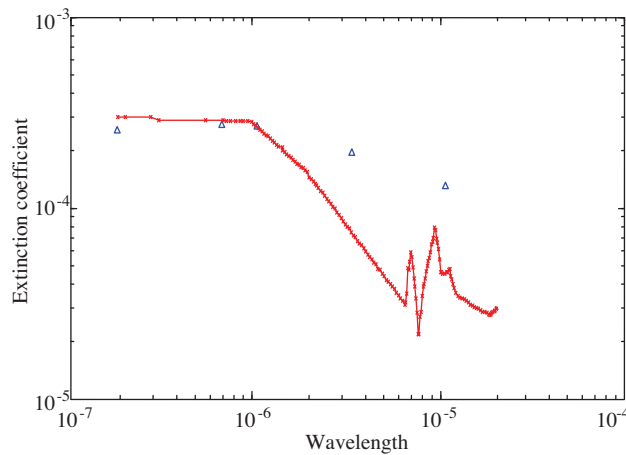


Figure 5. Extinction coefficient given by Chomette’s model (red line) and calculated (blue triangles) from the distribution of size (shown in Figure 4) as given by the genetic algorithms solution.

5. Conclusion

The LiDAR method allows quantitative and compositional characterization of gaseous pollutants as well as urban and desert aerosols together with their size distribution and concentration.

Throughout research, several methods of measuring desert aerosols by LiDAR have been implemented. One is a method using a single frequency LiDAR coupled with x-ray microanalysis of impacted filters. It provides detailed information on concentration and composition of atmospheric desert aerosols. But the measurements do not give geographical distribution. They only cover the location at which they were acquired.

To improve quality of the data obtained by LiDAR and refine the optical size distribution of desert particles we used a multi-spectral LiDAR system whose extinction coefficient profile in terms of wavelengths has been taken from Chomette's model. The wavelengths used were: $\lambda = 0.193, 0.694, 1.064, 3.370$ and $10.60 \mu\text{m}$.

To characterize the distribution of desert particle size by multispectral LiDAR, a method of optimization was developed via the method of genetic algorithms, in order to find a set of unknowns parameters whose number was fixed by ten elements (twice the number of wavelengths). These parameters characterize the radius of particles existing in the atmosphere.

Our result is compared with Chomette's well-known desert distribution model. The comparison did not give a perfect conformity due to errors due that arose from the diversity of parameters over which optimisation was conducted.

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