A METHOD FOR ESTIMATING THE ATMOSPHERIC CONTENT OF SUB-MICROMETER AEROSOL USING DIRECT-SUN-PHOTOMETRIC DATA

Luminiţa Filip and Sabina Ştefan

University of Bucharest, Faculty of Physics

OUTLOOK

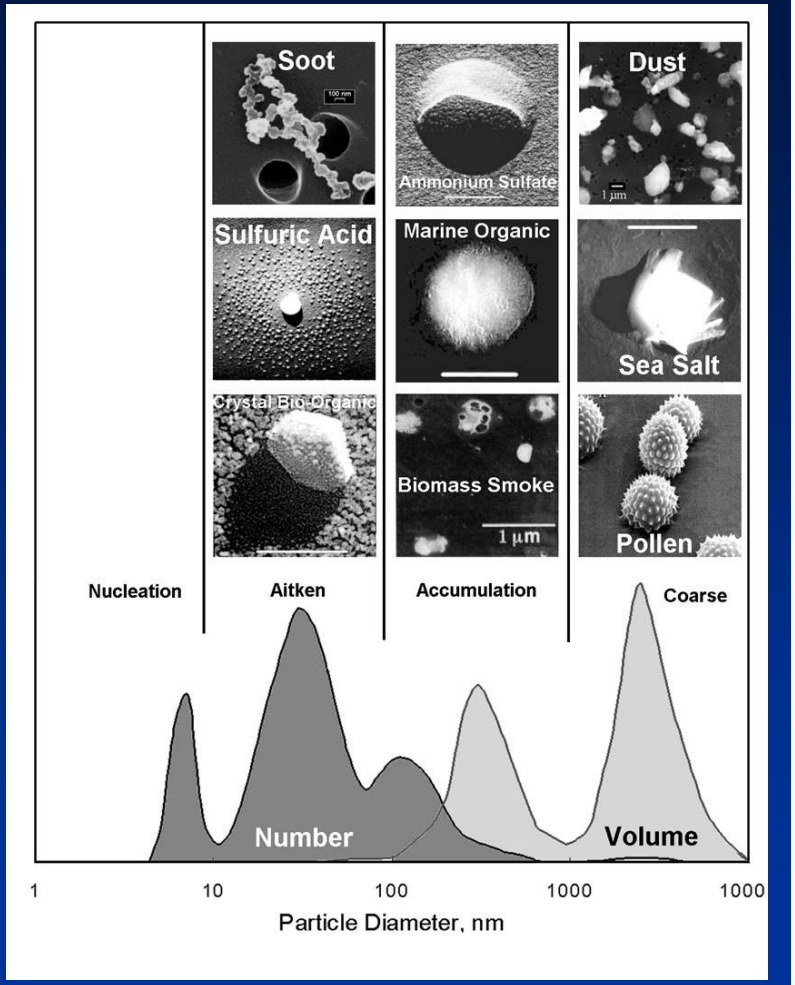
- Motivation & Objectives
- Brief description of the method
- The effects of changing the model parameters
- Validation of the method through comparison to AERONET sun/sky photometry products
- Conclusions

MOTIVATION and OBJECTIVES

It is well known that the aerosol generated by human activity falls in the sub-micrometer range.

⇓

Monitoring the sub-micrometer aerosol on a global scale is being a stringent necessity in protecting the environment



The sun/sky photometry proved a very efficient way for such monitoring activities, mainly when vast networks of instruments (like the Aerosol Robotic Network (AERONET) are used.

Our research aims to find a method for evaluating some characteristics of atmospheric aerosol, based on direct sun-photometric data, minimizing the errors and improving the accuracy.

The method relies **on fitting theoretical AOD spectral diagrams** (computed through Mie theory) to their experimental counterparts.

DESCRIPTION OF THE METHOD

We assume:

•In a widespread convention used also by AERONET, the atmospheric aerosol can be split into *two main dimensional ranges*: the so-called *fine* and *coarse* modes (http://aeronet.gsfc.nasa.gov/index.html).

•The gas filling the space between the particles has a unitary refraction index.

•The radius, *r,* of the target particles is taken as a parameter that varies in a certain range.

•the scattering sphere may have any size and its refraction index may be real or complex.

• Owing to the AERONET convention, the volume particle size distribution, $v(r)$, is introduced instead of $n(r)$, with the following definition:

$$v(r) = \frac{4\pi}{3} r^3 n(r)$$

The general expression of AOD is :

$$\tau_A(\lambda) = \int_{r_m}^{r_M} \pi r^2 Q_{ext}(x,m) n(r) dr$$

where r_m and r_M define the meaningful range of the aerosol radii and $n(r)$ is the size distribution of atmospheric aerosol particles.

The nucleus $Q_{ext}(x,m)$ is usually termed as *extinction efficiency factor* and is a result of Mie theory.

This quantity depends on the complex refraction index of the sphere, usually denoted by m:

$$m = n - i\,k$$

The imaginary part of the refraction index is a characteristic of the absorption of the incident wave in the dielectric of the target sphere.

As such, the separate fine (*f*) and coarse (*c*) components of the AOD can be defined separately:

$$\tau_{f,c}(\lambda) = \frac{3}{4} \int_{r_m^{f,c}}^{r_M^{f,c}} Q_{ext}\left(\frac{2\pi r}{\lambda}, m\right) v_{f,c}(r) \frac{1}{r} dr$$

Under the AERONET standards, the integration limits are the following:

$$r_m^f \equiv r_m = 0.05\ \mu m \qquad r_M^c \equiv r_M = 15\ \mu m \qquad r_M^f = r_m^c \equiv r_s$$

where r_s is the so-called *separation point* between the two ranges that is the radius for which the first minimum of the volume particle size distribution is attained.

The explicit use of previous definitions needs a specific shape for the volume particle size distribution. In this respect, a reasonable approximation is offered by representing each mode's distribution by a lognormal function (O'Neill et al, 2000, 2001, 2003):

$$v_{f,c}(r) = \frac{C_{f,c}}{r\sqrt{2\pi}\,\sigma_{f,c}} \exp\left[-\frac{\ln^2\left(r/R_{f,c}\right)}{2\sigma_{f,c}^2}\right]$$

where $C_{f,c}$ are the modal columnar volumetric aerosol contents (i.e. the total volume of aerosol of each mode, which can be found in the atmospheric column of unit cross-sectional area), $R_{f,c}$ are the modal radii and $\sigma_{f,c}$ are the geometric mean standard deviations describing the spread of each modal distribution .

One may notice at this point that, irrespective of the analytical approximation for each modal volume particle size distribution, the quantities $C_{f,c}$ must appear as distinct factors and this feature readily translates to the modal AOD. Therefore, one may write:

$$\tau_{f,c}(\lambda) = C_{f,c}\,\tilde{\tau}_{f,c}(\lambda)$$

Where $\tilde{\tau}_{f,c}(\lambda)$ **are quantities *independent of the modal columnar volumetric aerosol contents*, which will be further referred as *reduced modal* AODs.**

A parameter that is essential for our discussion is the *volumetric fine mode fraction*:

$$\phi = \frac{C_f}{C_V}$$

where C_V is *total columnar volumetric aerosol content* ,

$$C_V = C_f + C_c$$

In this way, one may readily express the total AOD as:

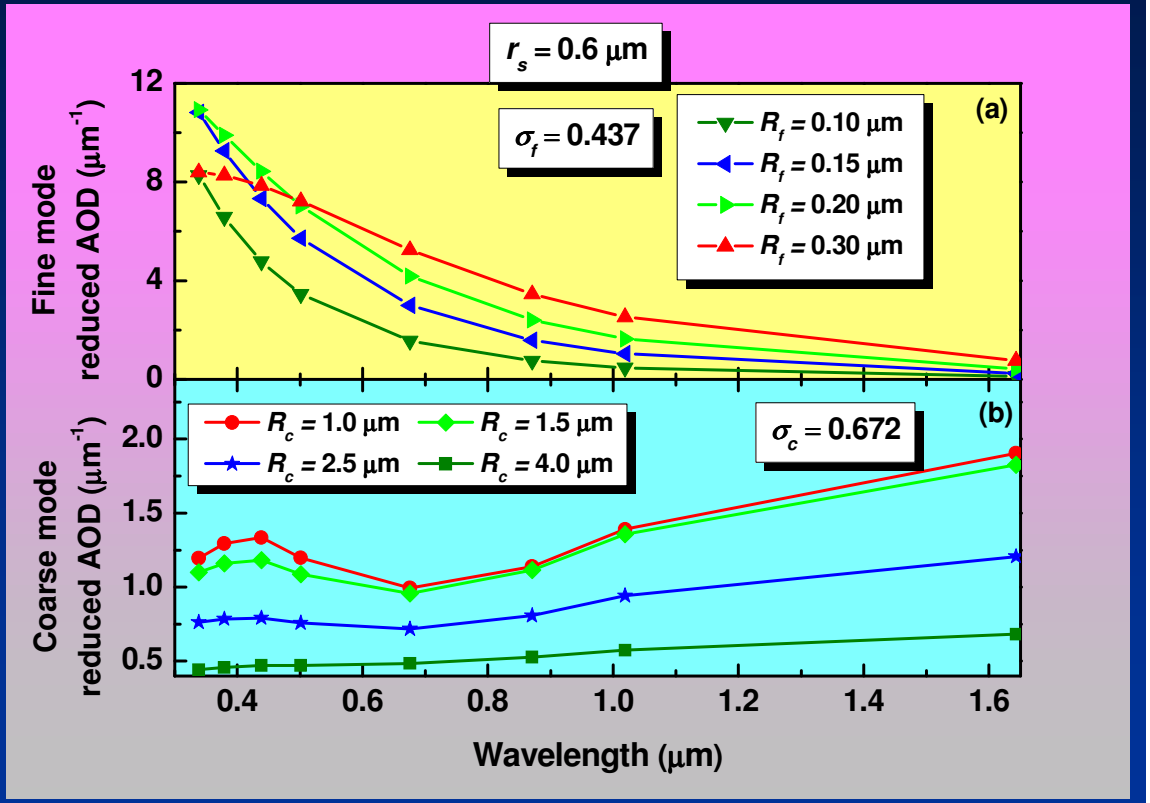
$$\tau_A(\lambda) = C_V \left[\phi \tilde{\tau}_f(\lambda) + (1-\phi) \tilde{\tau}_c(\lambda) \right]$$

The separate spectral variation of the reduced modal AODs differ largely in several respects.

The reduced AOD of the fine mode is about one order of magnitude greater in the visible part of the spectrum ⇒ given a load of fine mode aerosol.

The fine mode reduced AOD shows also a relatively steep decrease over the spectral range normally used in sun-photometry, while that of the coarse mode is almost spectral invariant.

The fine mode reduced AOD is generally *increasing* with the average dimensions of the corresponding aerosol.



On the contrary, the coarse mode reduced AOD is quite steeply decreasing for larger average radii of particles.

The different behavior of the extinctive response of the two main modes of atmospheric aerosol with the variation of the model parameters is a strong indication that the overall effect might be described as a superposition of these two components.

Thus, by varying separately the parameters related to the two main aerosol modes and their abundances, one can expect to fit the experimental spectral AOD diagrams with theoretical curves.

Therefore, the overall outcome of this procedure is the set of parameters' values corresponding to the best fit.

Another interesting quantity related to the fine mode optical response is its *optical ratio* which can be written in terms of ϕ as:

$$\eta_f(\lambda) \equiv \frac{\tau_f(\lambda)}{\tau_A(\lambda)} = \frac{\phi\tilde{\tau}_f(\lambda)}{\phi\tilde{\tau}_f(\lambda)+(1-\phi)\tilde{\tau}_c(\lambda)}$$

This quantity allows direct evaluation of the spectral variation of the relative contribution of the sub-micrometric aerosol particles (often related to anthropic pollution) to the extinction of the incident solar radiation.

In addition, consistent information on the atmospheric aerosol can be obtained from the value of the so-called *Ångström coefficient*, defined as :

$$\alpha(\lambda) \equiv -\frac{d\left(\ln \tau_A\right)}{d\left(\ln \lambda\right)} = -\lambda \frac{\tau'_A(\lambda)}{\tau_A(\lambda)}$$

which is to be distinguished from the usual exponent of the approximate Ångström's power law of extinction.

This quantity essentially represents the slope of the spectral variation of the AOD in logarithmic scale.

While not directly measurable, the practical computation of the Ångström coefficient can be performed by using a significant number of spectral channels in direct-sun photometry to allow for a good precision of the numerical differentiation.

There are two ways to obtain the Ångström coeficient.

On (to be called *direct*) procedure is through numerical differentiation of the spectral data of the total AOD.

The other way (which will be further termed as *composite*) is by splitting the total Ångström coefficient in separate contributions from the two main aerosol modes. Thus, one may readily obtain the modal Ångström coefficients in terms of the reduced modal AODs:

$$\alpha_{f,c}(\lambda) = -\lambda \frac{\tau'_{f,c}(\lambda)}{\tau_{f,c}(\lambda)} = -\lambda \frac{\tilde{\tau}'_{f,c}(\lambda)}{\tilde{\tau}_{f,c}(\lambda)}$$

The total Ångström coefficient may be written as a combination of its modal components:

$$\alpha(\lambda) = \eta_f(\lambda)\alpha_f(\lambda) + \left(1 - \eta_f(\lambda)\right)\alpha_c(\lambda)$$

Numerical computations of $\alpha_{f,c}$ can be performed through straightforward differentiation with a given size distribution.

THE EFFECTS OF CHANGING THE MODEL PARAMETERS

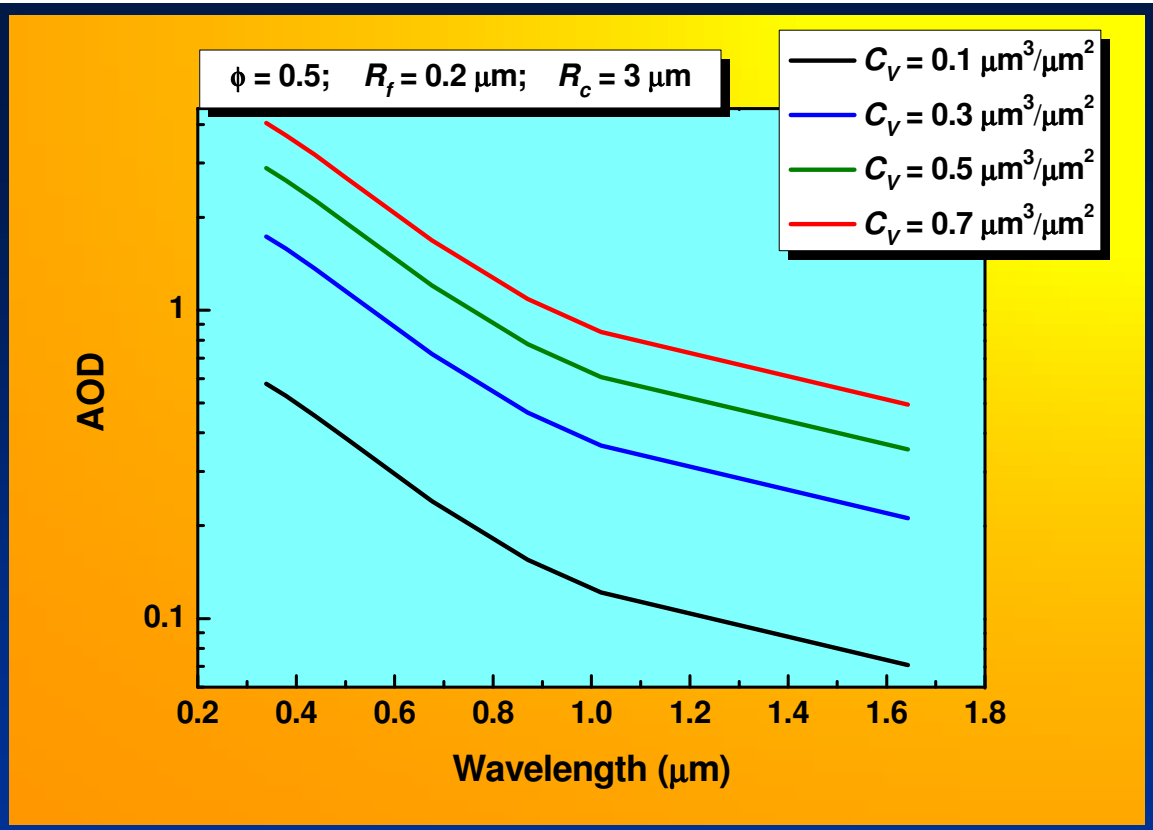
The fitting procedure developed in this research uses a set of nine parameters.

Of this set, only four have been considered as decisive for the computed outcome. Numerical tests have suggested that no significant modifications appear in the total AOD when the other five parameters are modified within the range of their annual values, as obtained from AERONET database

C_V, *the total columnar volumetric aerosol content*;

ϕ, *the volumetric fine mode fraction*;

R_f and R_c, *the modal radii* of the fine and coarse aerosol, respectively.



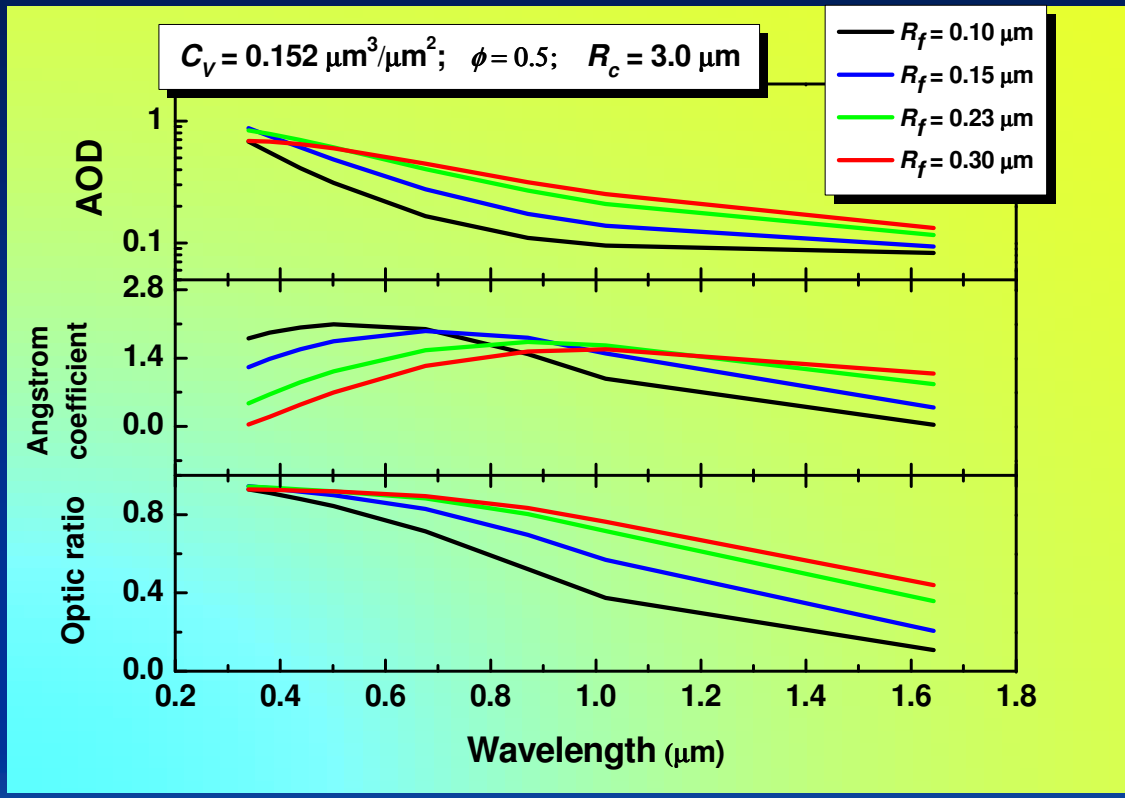
First, one may note that variations in the total aerosol content, C_V, may affect only the total AOD, which, in a logarithmic plot, will be simply shifted along the ordinate axis.

Neither the Ångström coefficient, nor the optical ratio depends on C_V.

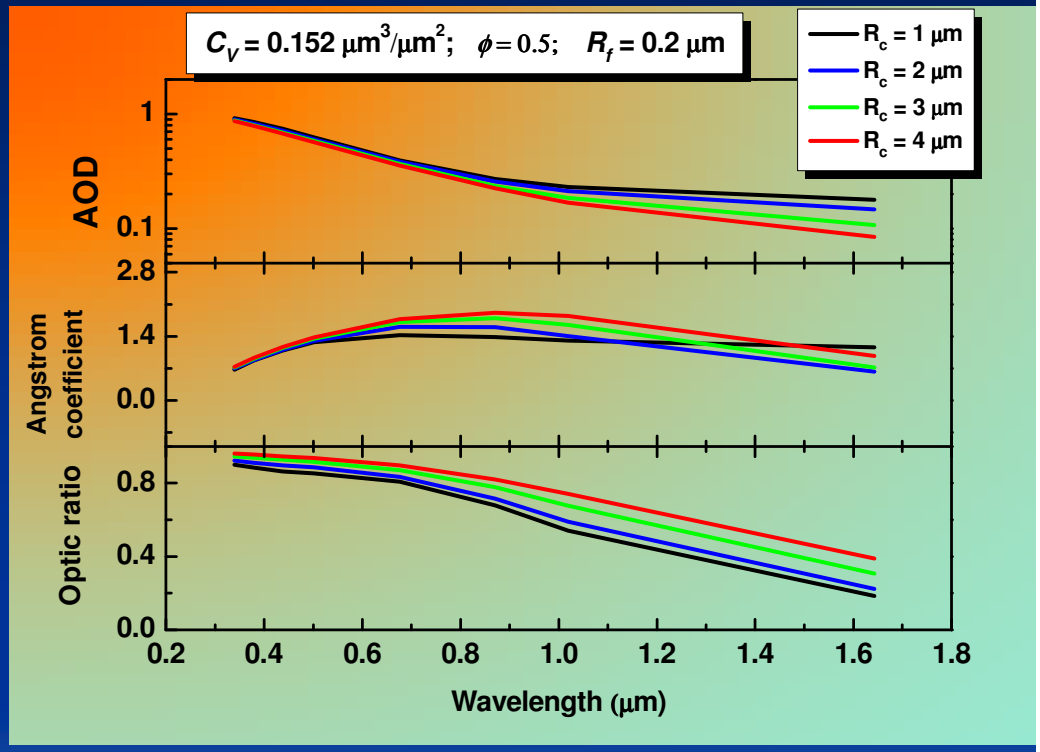
The influence of the variation of the fine mode average radius on the spectral diagram

First, one may note that smaller values of R_f produce a steeper decrease in the spectral variation of AOD.

When the average dimensions of the fine aerosol grow bigger and approach the coarse range the spectral diagrams of AOD become necessarily flatter.



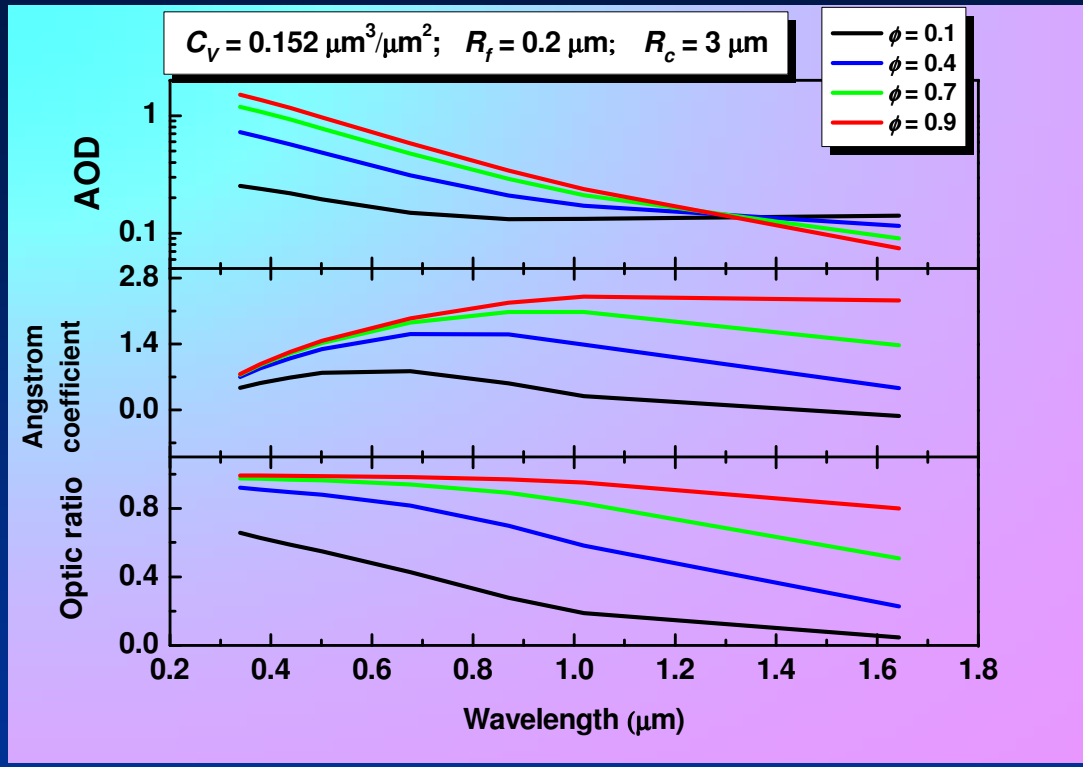
One may further observe that rather large variations in R_c produce no essential changes (with some exceptions at very large wavelengths) in spectral diagram of AOD.



The variation of the fine mode fraction, ϕ involve:

AOD changes greatly in slope, with steeper spectral decrease for cases when fine aerosol dominates.

As the atmosphere becomes more predominantly charged with coarse particles, the spectral diagrams of the AOD become flatter (a fact which is experimentally documented in the literature: Shaw, 1983)

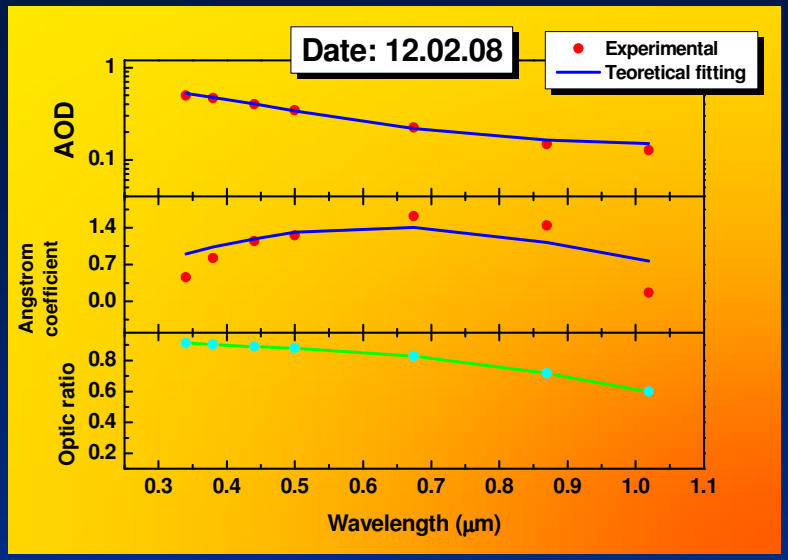


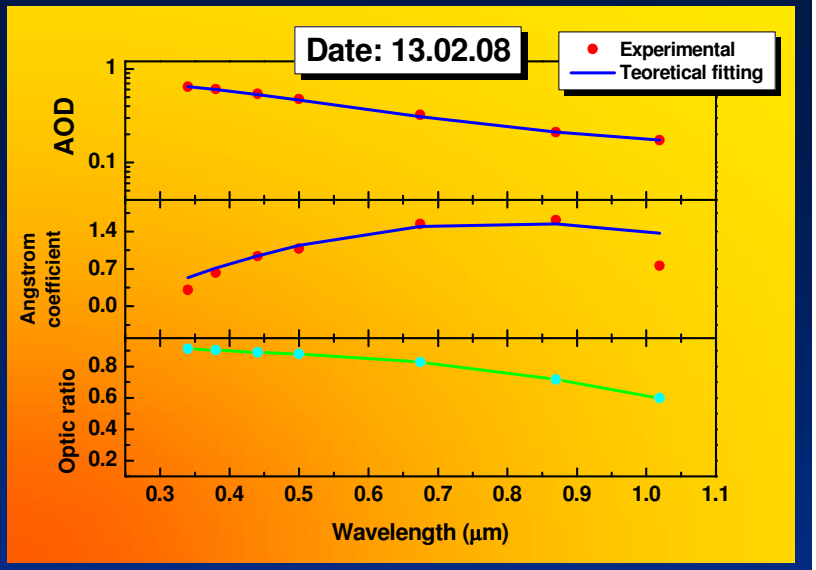
VALIDATION OF THE METHOD THROUGH COMPARISON TO AERONET SUN/SKY PHOTOMETRY PRODUCTS

The method described in the previous sections has been applied on direct-sun data collected during about one hundred days over the year 2008, by two AERONET sun-photometers (#359 and #397) placed in the northern and southern extremities of Bucharest, Romania.

The model diagrams have been compared to the diurnal averages of the measured spectral curves of AOD.

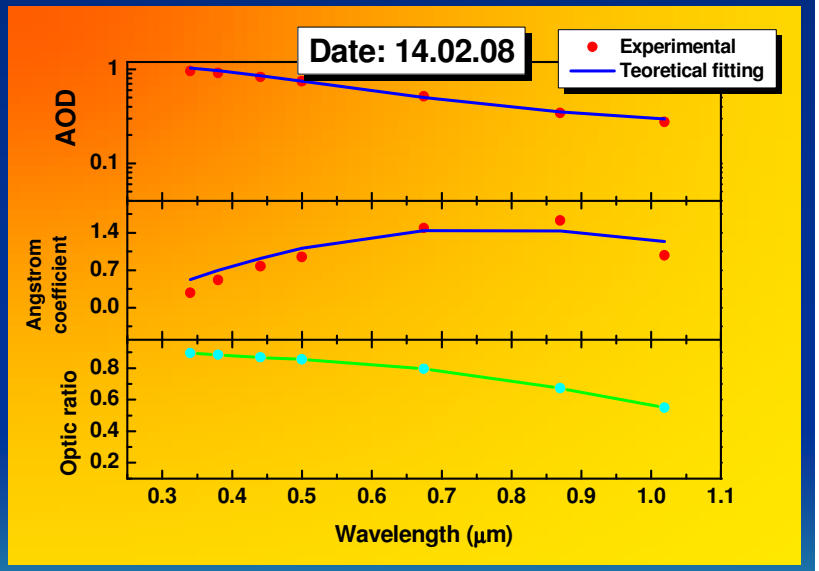
The fixed parameters have been set at the following averaged values: $m = 1.447 - i \cdot 0.008236$, $r_s = 0.59$ μm, $\sigma_f = 0.437$ and $\sigma_c = 0.672$.

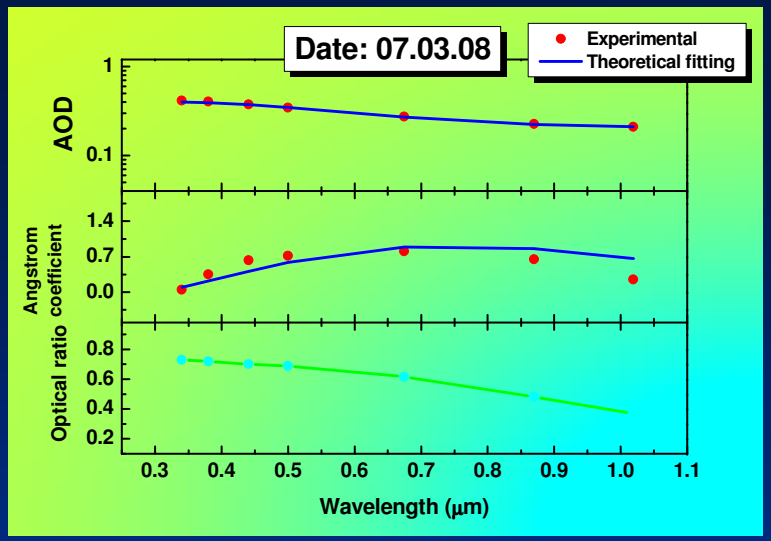


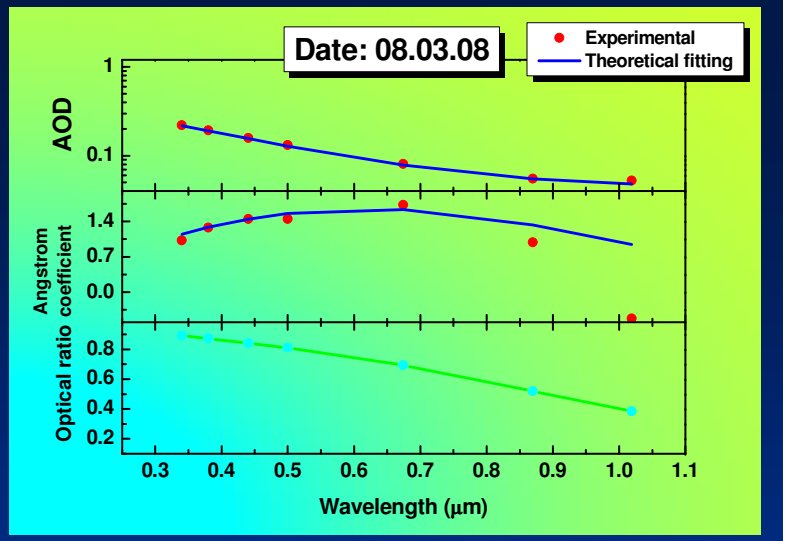


In a first sample series of diagrams one may note the flat shape of the AOD diagrams, with rather high values increasing in time.

By taking into account the obtained small volumetric fractions of fine aerosol during these three days from February (0.28, 0.43 and 0.38, respectively) and the increasing values of the total aerosol content (0.08 $\mu m^3/\mu m^2$, 0.13 $\mu m^3/\mu m^2$ and 0.23 $\mu m^3/\mu m^2$), one may conclude that an intrusion of coarse aerosol (dust) was developing in the region during this period (R_f is between 0.17 μm and 0.22 μm)



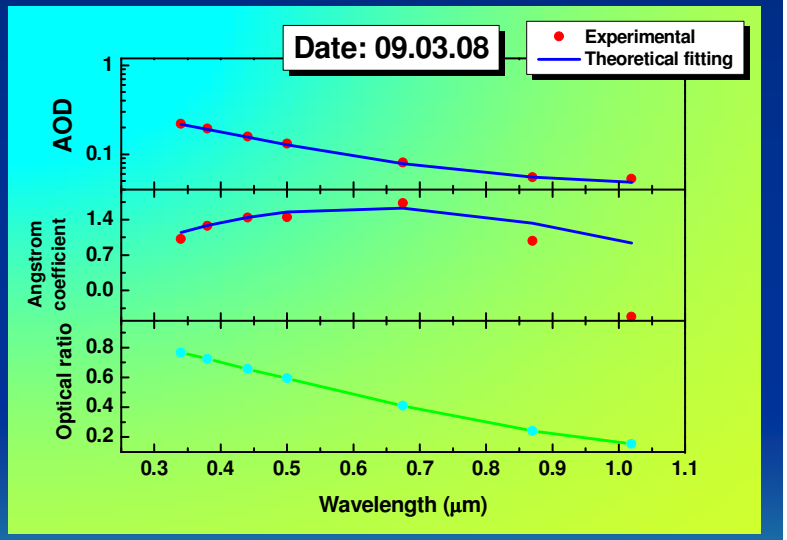




A somehow opposite situation may be observed in the second series of comparisons from the first third of March.

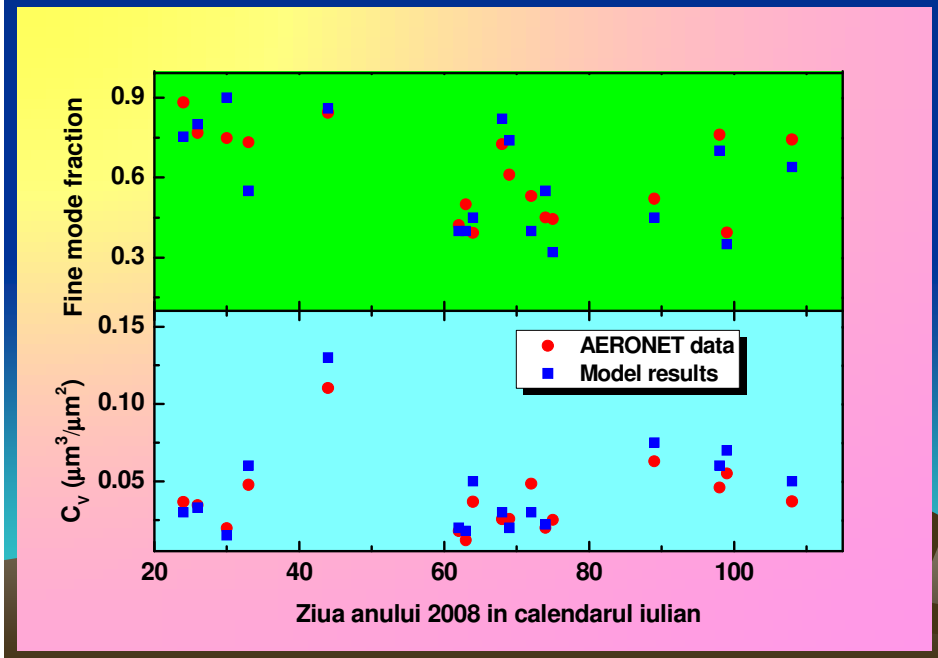
The values of AOD are much lower and decreasing, with an increasing spectral slope. This behavior suggests an advection of clean air in the monitored area (the obtained total aerosol content were 0.13 $\mu m^3/ \mu m^2$, 0.025 $\mu m^3/ \mu m^2$ and 0.025 μm3/ μm2, respectively).

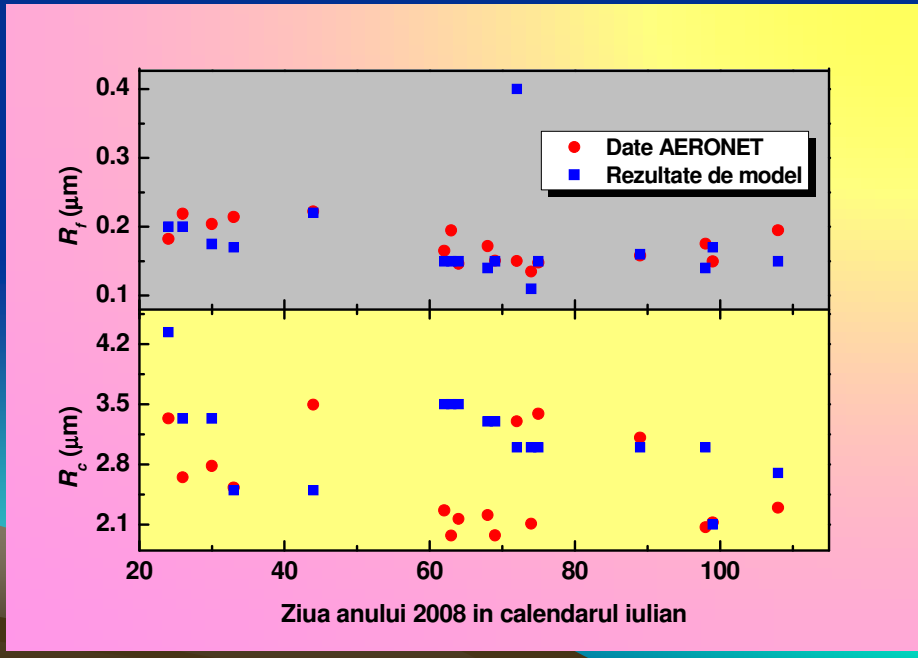
The values of ϕ are similar to those of the previous series (0.17, 0.31 and 0.3) $\Rightarrow$ a constant source of fine aerosol is present in the area (possibly of anthropic origin).



The comparison between the fitting values of some parameters with the corresponding AERONET products, which are obtained independently. The investigated period was of about 100 days of the year 2008.

It may be observed that, excepting the average radius of the coarse mode, which has been demonstrated to have little effect on the fitting procedure, the other fitted parameters keep close values to their counterparts provided by AERONET.





CONCLUSIONS

A simple method to determine some parameters of atmospheric aerosols using direct-sun photometric data existing in related databases.

The proposed method is a consistent alternative to the determination of some local aerosol characteristics.

In addition, the proposed method can readily provide some information that is complementary to the AERONET products (e.g. that contained in the spectral variation of the optical ratio).

The method is validated by applying it to measurements performed in the Bucharest area over an extended period of the year 2008 and by comparing its outcomes to the similar AERONET products.

The proposed method proves thus both reliable and able to expose various atmospheric events that modify the aerosol content

THANK YOU FOR YOUR ATTENTION !