

## Aerosol optical properties in Mediterranean basin under Saharan dust outbreaks

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Four measurement campaigns held in three different sites of the Mediterranean basin and in different years revealed the intrusion of mineral dust coming from North Africa, as confirmed even by the HYSPLIT back-trajectories analysis. Two of the sites are located in South Italy, the third one is in South Spain.. One data-set was obtained by a Monolight spectrometer (400 nm – 800 nm, resolution 3 nm), the other by means of an Avantes USB2000 spectrometer (400 nm – 900 nm, resolution 1.5 nm). A fitting procedure and a non-parametric inversion technique were applied to the measured AODs to retrieve, respectively, the Ångström parameters  $\alpha$  and  $\beta$ , and the Aerosol Size Distributions. Each site is characterised by different size distributions (bi-modal and Junge functions) but, independently from these functions, all the measurements affected by Saharan dust showed higher particles density in the radii range  $0.43\mu\text{m} \leq r \leq 3.0\mu\text{m}$ . The second result is the strong correlation between  $\alpha$  and  $\ln(\beta)$  for dust data points obtained in all sites, suggesting that mineral particles properties predominate over the background ones. Finally, Aerosol Size Distributions have been simulated, modifying their log-normal parameters and fixing, time by time, the refractive index values, to reproduce the experimental  $\alpha$  vs.  $\ln(\beta)$  behaviour. In this way, an “equivalent refractive index” common to the dust data has been retrieved.

### INTRODUCTION

Dust particles contamination modify both aerosols microphysical properties, as in Di Iorio et al. (2003), and radiative processes, as in Miller and Tegen (1998), in a way depending on the areas they cross and on the background particles they meet. This means that variations of dust particles content in the atmosphere strongly influences the Earth's radiation budget, while particles transported over very long distances can represent good tracers of the air-masses circulation on a large scale. These reasons, along with the need of developing regional and global circulation models as shown by Tegen, (2003), push scientists in studying optical and physical properties variation of atmospheric aerosols.

Mineral aerosol properties have been studied by using different radiometers such as CIMEL from the AERONET network, PREDE, MFRSR, EKO MS-120: measures performed in sites located in or very close to desert areas, generally highlight the increase of the Aerosol Optical Depth values and the dominance of the coarse mode particles over the fine one as in Smirnov et al. (1998), Eck et al. (1999), Tanré et al. (2001), Kubilay et al. (2003), Masmoudi et al. (2003), Xiangao et al. (2004), Ogunjobi et al. (2004).

In this work, ground-based radiometric measurements performed with two different high-resolution radiometers and collected in two sites in South Italy (Tito Scalo, July 2001, July 2004 and Pollino Mount, June 2003) and one site in South Spain (Tabernas Desert, INDALO campaign, September 2003) were analysed. All the sites are located in the Mediterranean area, with different background aerosols featuring different overall aerosol characteristics when mixed with dust.

Direct solar irradiance was measured with two radiometers covering, respectively, the range 400 nm - 800 nm, resolution 3 nm, and the range 400 nm - 900 nm, resolution 1.5 nm. The analysis has been supported by the HYSPLIT back-trajectories calculation tool that, along with the Aerosol Optical Depth data analysis, contributed to identify Saharan dust outbreaks. The retrieved aerosols size distributions were both bi-modal and Junge functions, depending on the site and the conditions under analysis. Finally, according to Halthore et al., 1992, the correlation between  $\alpha$  and  $\ln\beta$  has been looked for. The surprising result is that a high correlation ( $R^2= 0.94$ ) has been found for dusty data, independently on the site.

## 1. Measurements sites and experimental equipment description

As mentioned above, the sites involved in this analysis are located in the Mediterranean area: two of them in South Italy, the third one in South Spain.

Tito Scalo (40.60° N, 15.72° E, 750 m a.s.l.) is a rural area, on which few small factories insist, while Pollino Mount is a massif whose highest peak is at 2267 m a.s.l., The measurement site Piano Ruggio (40.00° N, 16.10° E, 1550 m a.s.l.), surrounded by higher peaks, is about 28 km far away from the Tirrenian Sea and 40 km from the Ionian Sea and it is characterised by grassy plateau and woods.

Finally, Tabernas Desert, in South Spain (37.09° N, 2.36° W, 505 m a.s.l.) is an extended semi-arid area covered by spotted vegetation at about 20 km from the sea and characterized by Easterly winds. This site was chosen for the INDALO (FIELD CAMPAIGN FOR DETERMINING ATMOSPHERIC AEROSOL PROPERTIES IN SEMIARID REGIONS) campaign.

The instruments used were a Monolight radiometer equipped with a rotating diffraction grating and a silicon detector, and an Avantes AVS-USB 2000 equipped with a one-dimensional linear CCD array. Both instruments are provided with a focusing system, whose field of view is  $1^\circ$ , thus avoiding solar diffuse light contamination. The measurements last, respectively, 15 seconds and 1 second. All spectra were collected every 15 minutes, from the sunrise to the sunset.

## 2. Measurements analysis: Saharan dust detection

### 2.1 Aerosols Optical Depths and Ångström parameters

Atmospheric attenuation of the solar radiation can be well described by the Lambert-Beer law, while the Langley calibration procedure allows the Aerosol Optical Depth estimation in spectral ranges not affected by gaseous absorption.

In this analysis, the ozone contribution to the optical depth, due to the Chappuis band 450 nm – 700 nm, is taken into account by using the columnar daily data of the ozone content (available on the TOMS site [toms.gsfc.nasa.gov/eptoms/ep.html](http://toms.gsfc.nasa.gov/eptoms/ep.html)).

The Ångström turbidity parameters  $\alpha$  and  $\beta$  are estimated by a fitting procedure and a modified non-parametric inversion technique, described in details in Esposito et al. (2004) allows the Aerosol Size Distributions retrieval. This retrieval is strongly reliable over the radii range  $0.1 \mu\text{m} \div 3 \mu\text{m}$ , using an a priori refractive index  $n_r = 1.45 + 0.0i$ .

This approximation is supported by the studies of Gonzalez and Ogren (1996) and Alados-Arboledas et al. (2003) showing that aerosol size distributions are not very sensitive to the imaginary part of the refractive index. The wavelength dependence of the AOD collected in Tito, Pollino and Tabernas is shown in fig. 1, where each panel reports, for each site, the AODs spectral dependence for dusty and no-dusty data: different spectral ranges, resolutions and error bars amplitude, depend on the radiometers used.

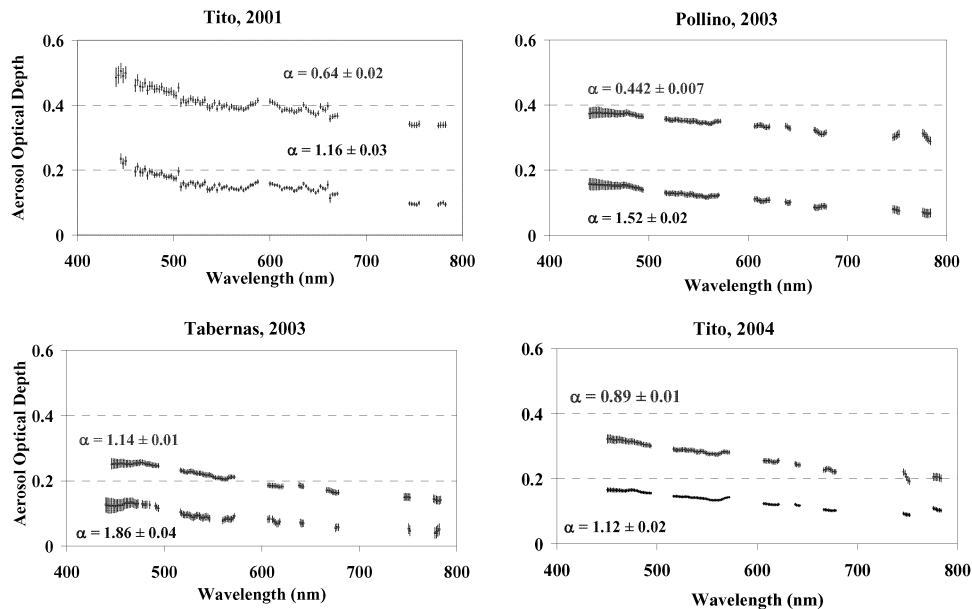


Fig.1: AOD spectral behaviour with and without mineral dust aerosol.

In figure 1 it is clear the influence of dust particles on the AOD amplitude. The lowest AOD values, obviously, correspond to the background aerosols: Tabernas optical thicknesses are lower than those measured in Tito and on Pollino mount. In this last location, the higher peaks protected the measurements site from winds, avoiding particles removal; on the contrary, the Spanish site is a wide plateau frequently swept by Easterly winds. AOD data affected by dust particles are sensibly higher: in fact mean  $\tau_{480}$ , increases of 142% for Tito, 146% for Pollino and 100% for Tabernas, mean  $\tau_{650}$  increases of 176%, 277% and 192%. These variations imply that the AOD as wavelength function becomes flat, determining the  $\alpha$  values decrease caused by the influence of larger particles, as in Smirnov et al. (1998).

The HYSPLIT model, available at <http://www.arl.noaa.gov/ss/models/hysplit.html>, web site was used to calculate the air-masses back-trajectories for turbid days, namely with high AOD and low  $\alpha$  values and trajectories originated in North-Africa were found.

The variation ranges of the alpha values for the four dust cases are the following: Tito (2001)  $0.56 \leq \alpha \leq 1.11$ , Pollino  $0.35 \leq \alpha \leq 0.88$  and Tabernas  $0.95 \leq \alpha \leq 1.58$ , Tito (2004)  $0.29 \leq \alpha \leq 1.1$ . These values are quite spread, depending on the mix of different

background aerosols and mineral particles; however, a good agreement is found, for example, with Lyamani et al. (2005). On the other hand, these wide ranges are common in literature, as reported in Dubovik et al. (2002), for AERONET measurements performed in Bahrain – Persian Gulf with  $0 \leq \alpha \leq 1.6$  or in Masmoudi et al. (2003) in Tunisia with  $-0.127 \leq \alpha \leq 1.78$ . In particular, it should be noticed that, although Tabernas is a desert site, the corresponding  $\alpha$  values are higher than the other two sites, suggesting the influence of smaller particles: in fact, its height is the lowest (505 m a.s.l.) and it is not far from the coast, from which easterly winds can carry small particles produced from anthropogenic sources, such as urban aerosols or biomass burning.

## 2.2 Aerosol Size Distributions

Aerosols size distributions are influenced by mineral particles in a way that is different, according to the different background particles. In Tito and Tabernas both Junge and bimodal functions dominated by the accumulation mode were found ( $0.1 \mu\text{m} \leq r \leq 0.42 \mu\text{m}$ ), while on Pollino mount simple Junge distributions were retrieved.

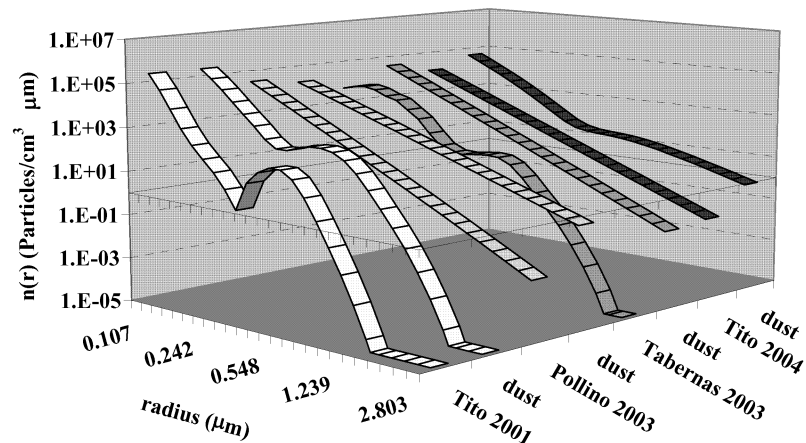


Fig.2: Comparison of aerosol size distributions with and without mineral dust aerosol.

The figure above shows, for each measurements campaign, the variation of the size distributions due to the arrival of mineral particles. As can be easily seen, the effect changes from time to time: in fact during Tito, 2001, the size distributions remain bi-modal functions, with a particles number increasing over the whole radii range, while during Tito, 2004 the background Junge function becomes a not well defined bi-modal distribution. On Pollino mount, the size distributions maintain their Junge function shape: the presence of mineral dust produces, even in this case, an increase of particles concentration both in the accumulation and in the coarse mode. For Tabernas, the most unexpected variation is registered: the bi-modal size distribution (second mode centred at  $0.72 \mu\text{m}$ ) of the no-dusty day becomes a Junge function. In fact, the particles concentration increases in the accumulation mode ( $0.27 \mu\text{m} - 0.55 \mu\text{m}$ ) and in the coarse mode ( $r \geq 1 \mu\text{m}$ ) and the two-mode shape of the no-dusty day disappears. This last result

is in agreement with Lyamani et al. (2005), where an increased particles concentration in the range  $0.4 \mu\text{m} \leq r \leq 2 \mu\text{m}$  is found, but disagrees with Dubovik et al., 2002 where the coarse mode maximum corresponds to  $2 \mu\text{m}$ . It has to be noticed that the sites considered in this latter study are very close to desert areas, such as Cape Verde or Saudi Arabia, where it is likely to have a strong dominance of larger particles.

### 2.3 Correlation between particles concentration and Ångström parameter

The Ångström parameters  $\alpha$  and  $\beta$  are strictly related to the aerosols physical properties: the lower is  $\alpha$ , the larger are the particles contributing to the AODs and the lower is  $\beta$ , the lower is the particles concentration.

Halthore et al., 1992 considered correlations between  $\alpha$  and  $\ln\beta$  for atmospheric aerosols having different origin, thus verifying that measured data were fitted by lines depending on the air masses past history. In the present study this correlation has been investigated for dusty and no-dusty measurements obtained during all the four campaigns under examination, as shown in plot 3.

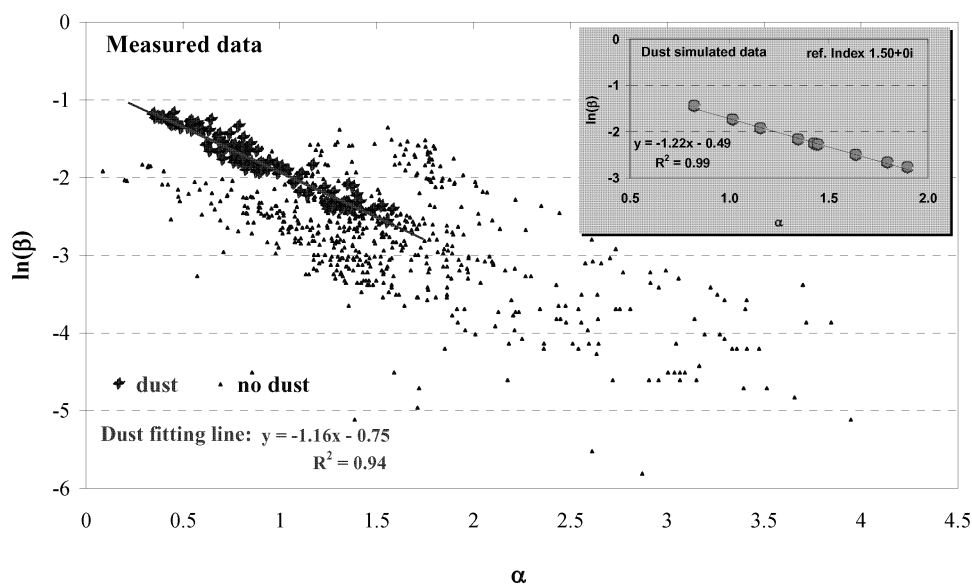


Fig.3: Correlation between  $\alpha$  and  $\ln\beta$ . The large plot shows experimental dusty (stars) and no dusty data (circles). The small plot shows simulated data with a complex refractive index  $1.50 + 0i$ .

Stars represent dusty data, while small circles are all the other measurements. It is really surprising to find all data affected by mineral particles, even if measured in three sites far each other and in different periods, to be strongly correlated ( $R^2 = 0.94$ ). In some sense, it is as if mineral aerosols properties prevail on background aerosols ones, which vary site by site.

To explain this high correlation, synthetic AODs were generated considering bi-modal aerosol size distributions (a sum of a Junge and a log-normal function) and changing their corresponding parameters, once fixed the refractive index value. These parameters are: Junge exponent, log-normal maximum and width. The simulated AODs were used to estimate the corresponding  $\alpha$  and  $\beta$  values and to build on a plot the couples  $\alpha$ ,  $\log \beta$ . Once fixed the refractive index value the simulated points, obtained changing only the log-normal parameters, describe a straight line. In this way the experimental slope corresponding to dusty data is reproduced by changing the refractive index value and this has been depicted on the small panel in fig. 3. The simulated fitting line best reproducing the experimental slope corresponds to a refractive index  $n_r$ , equal to  $1.5 + 0i$ : this value describes shared optical properties of many background particles mixed with dust mineral aerosol. It is relevant to find an index of refraction value which is in good agreement with those obtained in the framework of AERONET by measuring spectral sky radiance as in Dubovik et al., 2002, where a variation range of the real part is in the range 1.48 – 1.56, while the imaginary part can assume values between 0.0006 and 0.003.

## Conclusions

This work has been focused on the detailed analysis of high spectral direct solar irradiance measurements obtained during four campaigns, performed in three different sites of the Mediterranean basin. Two of them are located in South Italy, while the third one is in South Spain and all of them are not close to urban areas. These measures, with the support of the HYSPLIT back-trajectories, allowed the identification and characterization of aerosols physical and optical properties variation during Saharan dust outbreaks. The parameters analyzed are: Aerosol Optical Depth, Ångström turbidity parameters and aerosol size distributions.

The main results can be synthesized as follows:

- 1) all days affected by mineral airborne particles show higher values of the Aerosol Optical Depths, if compared with the values obtained during "clean" days. Correspondingly, the Ångström  $\alpha$  parameter values go down, due to the prevailing large particles, but in different ways site by site. On the other hand, these variations are in agreement with data reported in literature.
- 2) Aerosol size distributions, expressed as particles concentration per radii unit, show an increased particles number with a large particles predominance and, again, they are modified in different ways, depending on the site. In particular on Pollino mount Junge functions change only their slope, in Tabernas distributions not strongly bi-modal become Junge type and in Tito bi-modal functions change their magnitude or Junge functions become "light" bimodal distributions.
- 3) The plot  $\alpha$  vs  $\ln \beta$ , derived considering all identified dusty points, highlights a strong correlation ( $R^2 = 0.94$ ) among them: mineral particles optical properties dominate over different background aerosol optical properties. Finally, a set of bi-modals aerosol size distributions with different geometrical parameters and fixed refractive index were generated, by which AODs were simulated. The corresponding  $\alpha$  and  $\ln \beta$  couples of values describe, on a plot, a straight line with a high correlation coefficient, whose slope changes if the complex refractive index changes. In particular the synthetic slope is equal to the experimental one for a value of the refractive index ( $n_r = 1.5 + 0i$ ) which can be thus considered as an "equivalent refractive index" and is in good agreement with values found in literature for dust.

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