Aerosol properties retrieved from sky radiance at the Principal Plane for non-spherical particles

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Abstract — **Due to the proximity, the Iberian Peninsula is a gateway for the air masses coming from the Saharan desert, being a crucial stage for the study of optical and physical properties of mineral particles. Many authors have used sky radiance measurements in the almucantar configuration, in combination with direct irradiance measurements, to retrieve the columnar aerosol size distributions, single scattering albedo and asymmetry parameter using sun-photometric techniques; but very few authors have retrieved these aerosol properties by the inversion of sky radiance measurements using the principal plane configuration. Here, we present the columnar aerosol properties obtained during the Saharan dust events observed over southern Iberian Peninsula during 2005-2008. The aerosol optical depth (AOD), Angström exponent (α), volume size distributions, single scattering albedo (SSA) and asymmetry parameter were simultaneously retrieved from sun photometric measurements, according to the method described by Olmo et al. (2008). Measurements were performed in the AERONET station at Granada (37.18 ºN, 3.58 ºW, 680 m a.m.s.l), and were obtained by a CIMEL CE 318-1 sun-photometer. Monthly and annual daily averages have been obtained from AOD and α parameter. The aerosol volume size distributions showed a bimodal structure. The fine mode showed the highest peak at (0.19±0.03) μm for all seasons. Coarse mode showed the highest peak at (1.95±1.27) μm for spring. The SSA increases with the wavelength in agreement with desert dust properties, showing lower values in spring and summer due to a mixture with more absorbent local anthropogenic particles.**

Keywords — **Dust Desert, Aerosol properties, Principal Plane, Non-spherical particles.**

I. INTRODUCTIÓN

Mineral dust particles originated over deserts like Sahara play an important role on Earth´s climate system. Dust particles interact with the solar and thermal radiation modulating Earth´s radiative budget. The Sahara desert is the most important source of mineral dust in the northern hemisfere. Mineral aerosols and dust are a common type of suspended particulate matter that characterize arid regions. North African dust is injected into the atmosphere through resuspension processes at the sources areas, and it is then transported to different altitudes (from sea level up to 4-6 km), being the maximum dust transport in summer when large quantities of dust are carried across the Mediterranean basin to Europe and Middle East and across the Atlantic ocean to the Caribbean, the southeastern United States and the mid-latitude western Noth Atlantic. In winter, there is also considerable transport when large amounts of dust are carried toward south America and sporadically to Western Europe. Mineral dust is responsible for aproximately one third of global extinction optical depth (Tegen *et al.*, 1997), and absorbs significantly in the blue and ultraviolet wavelengths due to iron oxide impurities (Sokolik *et al.*, 1999). It has been shown that the extend of solar attenuation is sensitive to the dust particles´s source region, because their optical properties vary depending on the source and on the chemical and physical processes ocurring along the trayectory.

Analyses of labotatory measurements and in situ data using scanning electron microscopes reveal that the shapes of dust particles are exclusively irregular. Especially, a detailed theoretical study on the optical properties of dust particles, based on a spheroidal polydispersion model (Mishchenko *et al.*, 1997), has shown that the nonspherical effect of dust particles can be significant in comparison with the Lorenz-Mie solution for spheres. Nonspherical dust particles have substantially different scattering phase functions, asymmetry parameters, optical depths, and single-scattering albedos, as compared with those of the volume-equivalent spheres. Retrievals of the scattering phase function, including scattering angles larger than 90º, are important, since this angular particle scattering range determines the aerosol effect on climate and is used for remote sensing. On the other hand, particle shape affects aerosol scattering at large angles 100-140º. The backscattering of non-spherical particles is usually less dependent on the scattering angle than for spherical particles. The difference between nonspherical and spherical scattering is near maximun at an angle of 120º.

Many works obtained optical and microphysical properties of desert dust using measurements of sky radiance in the almucantar configuration. However, few authors have used the sky radiance in the principal plane configuration. Olmo *et al.* (2008) analyzed Saharan dust events in the city of Granada using sky radiance in the principal plane configuration. This last measurements configuration allows us to extend the aerosol parameter retrieval to smaller zenith angles than that used in the retrieval from almucantar geometries, showing the feasibility of extending the retrieval of atmospheric aerosol optical properties along the day, not just for large solar zenith angles (Olmo et *al.*, 2008).

In this paper, aerosol microphysical properties have been computed using the sky radiance measurements and the Nakajima code, adapted by Olmo *et al.* (2006, 2008), taking into account the principal plane configuration and nonspherical particles.

This study focuses on the Saharan dust outbreaks detected at Granada from January 2005 to December 2008.

The site of measurements and the instrumentation used are described in section II. The experimental method is sumamarized in section III. The analysis of the columnar aerosol properties of the Saharan dust events, based on aerosol optical depth (AOD) and Angström exponent (α) values, are explains section IV. Finally the conclusions are described in section V.

II. Experimental site and instrumentation

Measurements have been perfomed in the urban area of Granada (37.18ºN, 3.58ºW and 680 m a.m.s.l.). Granada, located in south-eastern Spain, is a non-industrialized, medium-sized city with a population of 300000. The city is situated in a natural basin surrounded by mountains with altitudes over 1000 m. The near-continental conditions prevailing at this site are responsible for large seasonal temperature differences, providing cool winters and hot summers. The study area is also at short distance, about 200 km away from the African continent, and aproximately 50 km away from the western Mediterranean basin.

For this work we use solar extinction (direct irradiance) and diffuse sky radiance at the solar principal plane configuration, both measured with a CIMEL CE-318-4 sun-photometer included in the AERONET network (Holben *et al.*, 1998). This sun-photometer makes direct sun measurements with a 1.2º full field of view at 340, 380, 440, 670, 870, and 1020 nm. The full-width at half-maximun of the interference filters are 2 nm at 340 nm, 4 nm at 380 nm and 10 nm at all other wavelenghts. The sky radiance measurements are carried out at 440, 670, 870, and 1020 nm by means of almucantar and principal plane configurations. Together with the AERONET calibration procedures, Langley plots at high location in Sierra Nevada Range (2200 m a.m.s.l.) have been made regularly, at least twice a year, to determine the spectral extraterrestrial voltage for this instrument. On the other hand, an integrating sphere has been used to calibrate the instruments for radiance measurements (Alados-Arboledas *et al.*, 2004). The total uncertainty in AOD and sky-radiance measurements is about ± 0.01 and ±5%, respectively (Holben *et al.*, 1998).

III. Methodology

We focus this work on Saharan dust events that have been confirmed by CALIMA, which belongs to a network of the Spanish Ministry of Rural Environment and Marine (www.calima.ws). The HYSPLIT.4 model has been used to calculate five-days backtrajectories of air masses coming to Granada at six different altitudes above ground level (a.g.l.) using the GDAS database (Global Data Assimilation System, ftp://www.arl.noaa.gov/pub/archives/ gdas1/). The retrieved information from sky radiance at large scattering angles requires accurate correction for the effects of multiple scattering and for the contribution of light reflected from the Earth's surface and scattered downward in the atmosphere. Nakajima *et al.* (1996) developed and applied an inversion scheme for spherical particles that includes accurate radiative transfer modeling to account for multiple scattering (Skyrad.pack code), and Olmo *et al.* (2006, 2008) improve this method to take into account nonspherical particles. The method uses specified wavelengths, selected outside the gas absorption bands, in order to reduce the radiative transfer problem to a pure scattering problem. The inversion procedure uses the normalized sky radiance (almucantar or principal plane configuration) and the AOD measured by means of a method that requires absolute calibration.

From the sun-photometer direct Sun measurements, the aerosol optical depth (AOD) at selected spectral chanels has been derived, applying the Lambert-Beer-law after the subtraction of the Rayleigh and gas contributions (Alados-Arboledas *et al.*, 2003; Lyamani *et al.*, 2005). Considering the error sources in derived AOD, the AOD uncertainty is dependent on the wavelength and is about 0.01 (Alados-Arboledas *et al.*, 2003). Moreover, we have first removed the measurements contaminated by clouds using the cloud screening methods by Smirnov *et al.* (2000). Furthermore, to assure the clear sky conditions, the difference between principal plane radiances from both sides of the sun (symmetry) has been used as a check for the homogeneity of the sky conditions during the measurement process. We have discarded asymmetrical situations (differences>10%) associated with inhomogeneous atmospheric conditions or the presence of some clouds. Using the principal plane configuration and the non-spherical inversion code, we computed a large set of columnar aerosol parameters: volume size distribution, mean radius, effective radius, volume concentration (for total, fine and coarse modes), refractive indices (real and imaginary parts), single scattering albedo, phase function and asymmetry parameter.

IV. RESULTS

As discussed in Section III, we used the CALIMA data base to ensure that the Saharan dust events have been recorded at ground level.

Over the whole dataset, from 2005 to 2008, 46.69% of the days with inversion outputs were characterized by the presence of dust events. 54 African dust episodes were identified accounting for a total of 234 days, with a mean duration of 4.33 days per episode. This represented a mean of 14 episodes per year with a maximun of 18 in 2008, and minimun of 12 in 2005.

The highest number of days with African episodes was recorded in 2008 (68 days), and the lowest number of days was recorded in 2005 (47 days).

The frequency was highest in summer (58.85%), followed by autumn (54.72%), and was lowest in winter (23.59%). In this period (2005-2008), dust events whose time duration were 3 to 4 days occurred 11 times. Dust events of 2 days duration were repeated for 9 times, and events of 1 day 7 times.

During the Saharan dust events, the increase in AOD values was accomplished by a decrease in the α parameter. Spring had the largest mean value of AOD-440 nm (0.36) and the lowest mean value of α (0.46). 2005 and 2006 had the highest annual mean value of AOD-440 nm (0.32). Meanwhile, 2005 and 2008 had the lowest α mean value of 0.5.

The daily AOD-440 nm was mostly in the range 0.3 to 0.5 in spring, and in the range 0.2 to 0.3 in summer and autumn. In winter the most common value AOD-440 nm ranged from 0.1 to 0.2. Values of α were more or less uniformly distributed between 0.1 and 0.4 in spring, and between 0.3 and 1.1 in winter. In summer and autumn α follow a normal distribution with a maximun in the range 0.5 to 0.6. The largest daily AOD-440 nm value was registered on February 25th, 2008 (1.41), corresponding to the dust event ocurred from February 23rd to 29th, 2008. The lowest daily value of α was obtained on March 20th, 2005, with an average value of -0.02.

The monthly evolution of AOD-440 nm and α is shown in figure 1 during the period 2005-2008.

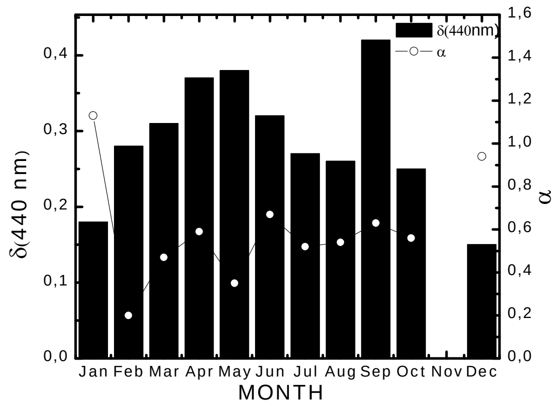

Fig. 1 Monthly distribution of AOD-440 nm and α during the period 2005-2008.

In November there was no desert dust events. The months of January, February, and December were characterized as having limited data available due to the cloud conditions. The monthly high values of AOD-440 nm were registered between March and June in the range 0.31 to 0.38, however we find the maximun mean value of AOD-440 nm in September, reaching a value of 0.41. The lowest values of α were registred between February and May, ranging from 0.2 to 0.35. The highest value of α was reached in January, with a mean value close to 1.13.

It should be noted a desert dust event that took place for 15 days and whose AOD-440 nm mean value for the entire period was 0.22, and α mean value was 0.54. The event took place from July 14th to 28th, 2007.

Escudero *et al.* (2005) analyzed African dust events over eastern Spain during the period 1996-2002. They find, for this period, that Saharan dust events reaches Spain in 15% of the days, in about 16 episodes per year, with peaks in May, followed by June, July, and August. The average duration of events was 3.3 days. In our study the average duration was 3.7 days.

Aerosol Size Distributions and Physical Parameters

Radiative impact of atmospheric aerosol particles depends not only on aerosol concentration but also on their size and chemical composition. Compared to anthropogenic sulfate, desert dust is generally larger in size and more absorbing at UV-VIS and infrared wavelenghts. This results in increased atmospheric heating along with decreased incident solar radiation at ground and trapping of outgoing thermal radiation.

Table 1 shows the annual averages of optical and microphysical properties during Sahara dust events observed during 2005-2008. The particle size distributions always showed bimodal size distribution with fine particle mode (r <0.5 µm) and coarse particle mode (r >0.5 µm). The fine mode had an average value for the entire period of (0.19±0.03) µm, while the coarse mode had an average value of (2.87±1.63) µm. The lowest average value for the fine mode corresponded to 2007 (0.18±0.03) µm, while the highest value corresponded to 2008 with an average value of (0.21±0.03) µm. Contribution to the fine mode (submicrometric particles) was due to the influence of local anthropogenic aerosol. For coarse mode, we find a modal radius value between (2.4±1.4) µm for 2005 and (3.21±1.6) µm for 2007. Dust particles were responsible for the micrometric mode. The ratio V_c/V_f indicated the prevalence of large particles compared to small particles, which is characteristic of desert dust. These values were consistent with those obtained by authors like Dubovik *et al.* (2002).

Table 1 Annual average values of optical and microphysical properties

	2005	2006	2007	2008
δ(440nm)	0.32±0.14	0.32±0.15	0.28±0.14	0.28±0.14
α	0.50±0.27	0.53±0.26	0.64±0.26	0.50±0.29
ω_0(440nm)	0.89±0.03	0.88±0.02	0.88±0.02	0.91±0.03
ω_0(670nm)	0.90±0.03	0.89±0.03	0.88±0.02	0.92±0.03
ω_0(870nm)	0.91±0.03	0.90±0.03	0.89±0.03	0.93±0.03
ω_0(1020nm)	0.92±0.03	0.90±0.03	0.90±0.03	0.94±0.03
g(440nm)	0.69±0.02	0.71±0.02	0.70±0.02	0.70±0.02
g(670nm)	0.67±0.02	0.67±0.02	0.67±0.02	0.67±0.02
g(870nm)	0.67±0.02	0.66±0.02	0.66±0.02	0.66±0.02
g(1020nm)	0.67±0.02	0.67±0.02	0.67±0.01	0.67±0.02
r_{eff} (µm)	0.89±0.42	0.99±0.36	1.11±0.44	1.06±0.47
r_c (µm)	2.4±1.4	2.64±1.43	3.21±1.64	2.60±1.61
V_c ($\mu m^3/\mu m^2$)	0.18±0.21	0.19±0.10	0.19±0.09	0.17±0.09
r_f(µm)	0.19±0.03	0.20±0.03	0.18±0.03	0.21±0.04
V_f ($\mu m^3/\mu m^2$)	0.017±0.007	0.018±0.007	0.018±0.007	0.015±0.006
V_c/V_f	10.3±6.3	10.54±4.97	11.93±5.37	11.79±6.93

We expect a predominance of coarse mode during dust desert conditions, and more or less severe changes in this predominance depending on the influence of anthropogenic aerosols, which mainly contribute to fine mode (Eck *et al.*, 1999). The volume size distributions seasonal averages during 2005 to 2008 are shown in figure 2, being bimodal. The fine mode showed a modal radius around (0.19±0.03) µm for all seasons. Particles of anthropogenic origin contributed to the fine mode and its value shows the same trend for all seasons. The coarse mode showed the maximum value at radius (1.95±1.27) µm in spring and at radius (2.62±1.35) µm in other seasons.

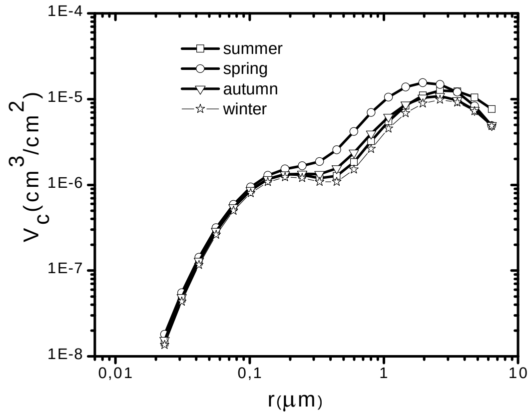

Fig. 2 Seasonal average of volume size distributions during the period 2005-2008.

The fine mode mean radius not change significantly whereas the coarse mode mean radius was a good indicator of the dust desert arrival. These values agree with those obtained by other authors for desert dust particles (Kim *et al.*, 2004; Dubovik *et al.*, 2002a).

The single scattering albedo (SSA) and the asymmetry parameter are key parameters for the estimation of the direct radiative impact of aerosol particles. The SSA is defined as the ratio of the scattering coefficient and the extinction coefficient. It depends on the relative source strengths of the various aerosol substances and on aging during transport. Absorption of solar radiation by atmospheric particles results mainly from elemental carbon originating from anthropogenic activities, biomass burning and from mineral dust. The asymmetry parameter gives an indication of the angular distribution of the scattered radiation.

In this paper we follow the recommendations of Dubovik *et al.* (2000): only for AOD-440nm>0.4 one can rely on the accuracy on the retrieved SSA. Figure 3 shows the annual evolution of the single scattering albedo at 670 nm. We find a decrease in SSA for dry season. In these months the solar radiation on surface increases. This caused the mixed layer rise and the local anthropogenic particles were mixed with desert dust causing a decrease in SSA values. Analyzing the seasonal average we find that the SSA showed a low dependence on wavelength for all seasons except for spring where increase slightly, with a range of 0.89-0.91 for 440-1020 nm.

The maximum values of SSA for 2005 were 0.89±0.03, 0.90±0.03, 0.91±0.03 and 0.92±0.03, and for 2008 were 0.91±0.03, 0.92±0.03, 0.93±0.03 and 0.94±0.03, at 440, 670, 870, and 1020 nm, respectively.

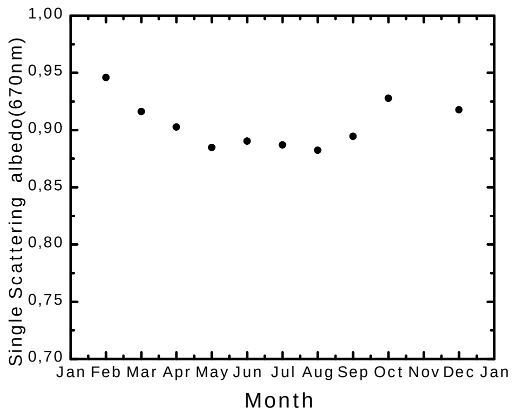

Fig. 3 Annual average of SSA at 670 nm during the period 2005-2008.

As we observe in figure 4, the SSA values increase with wavelength for all AOD ranges. As increasing aerosol loading increases the SSA.

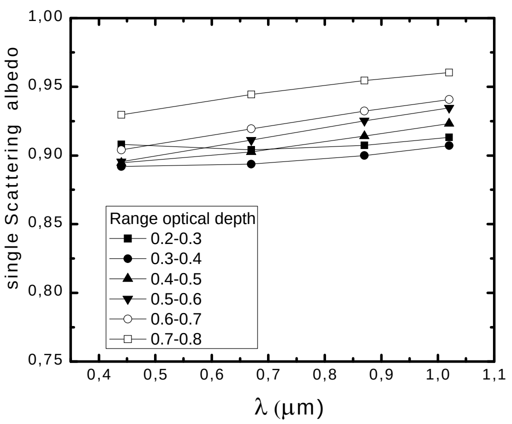

Fig. 4 SSA versus wavelength for different AOD ranges during the period 2005-2008.

The asymmetry parameter is indicative of the rate of radiation scattered forward, and is related to the particles size. During our analyzed period, we find an annual average value of 0.69±0.02 at 440 nm, and ranges from 0.65 to 0.71. The smaller values were found at January and December, being 0.65±0.02 and 0.67±0.02, respectively. These last values indicate more load of small particles due to the anthropogenic contribution in cool season. The highest value was found at May, being 0.71±0.01. This coincides with the higher values of AOD-440 nm and the higher coarse mode concentration during spring season, indicative of predominance of coarse mode particles. The asymmetry parameter seasonal averages at 440, 670, 870, and 1020 nm are shown in figure 5.

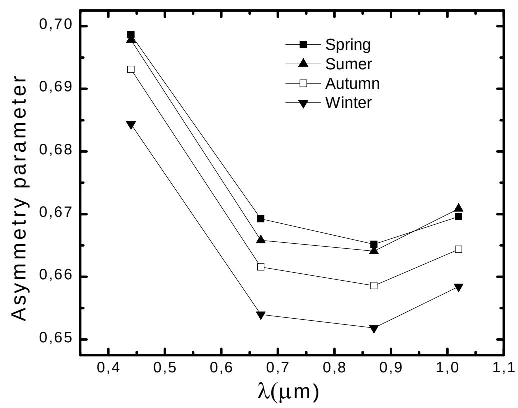

Fig. 5 Asymmetry parameter seasonal average at 440, 670, 870, and 1020 nm during the period 2005-2008.

Figure 5 shows a decreasing trend with wavelengths (440-870 nm) in all seasons, and an increasing trend at 870-1020 nm. The asymmetry parameter averages were found larger during spring and summer respect to autumn and winter, and ranges from 0.65±0.02 to 0.69±0.02 for all wavelengths. The asymmetry parameter increase was possible due to the higher contribution of large particles.

V. SUMMARY

This paper present the columnar aerosol properties obtained during desert dust events registered at Granada during 2005-2008. A total of 491 daily average values of AOD and α were used in the analysis. 234 days were influenced by desert dust according to the CALIMA database. The results show that 46.69% of the days were influenced by desert dust events, with average values for

AOD-440 nm and α of 0.29±0.14 and 0.55±0.27, respectively.

The aerosol volume size distributions showed a bimodal structure. The fine mode showed the highest peak at (0.19±0.03) μm for all seasons. Due to the local sources, this followed the same trend for the whole measurement campaign. Coarse mode showed the highest peak at (1.95±1.27) μm for spring, in agreement with the greater loading of AOD for this season.

During the analyzed period, the SSA increases with the wavelength, in agreement with desert dust properties. However, during the spring and summer the SSA is lowest for all wavelengths, probably due to a mixture of desert dust particles with more absorbent local anthropogenic particles.

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