Retrieval of optical and physical properties of African dust from multi-wavelength Raman lidar measurements during the "SHADOW" campaign in Senegal

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1 ABSTRACT

2 West Africa and the adjacent oceanic regions are very important locations for studying 3 dust properties and their influence on weather and climate. The SHADOW (Study of SaHAran 4 Dust Over West Africa) campaign is performing a multi-scale and multi-laboratory study of 5 aerosol properties and dynamics using a set of in situ and remote sensing instruments at an 6 observation site located at IRD (Institute for Research and Development) Center, Mbour, Senegal (14⁰N, 17⁰W). In this paper, we present the results of lidar measurements performed 7 8 during the first phase of SHADOW which occurred in March-April, 2015. The multiwavelength 9 Mie-Raman lidar acquired $3\beta+2\alpha+1\delta$ measurements during this period. This set of measurements 10 has permitted particle intensive properties such as extinction and backscattering Ångström 11 exponents (BAE) for 355/532 nm wavelengths corresponding lidar ratios and depolarization ratio 12 at 532 nm to be determined. The mean values of dust lidar ratios during the observation period 13 were about 53 sr at both 532 nm and 355 nm, which agrees with the values observed during the 14 SAMUM 1 and SAMUM 2 campaigns held in Morocco and Cape Verde in 2006, 2008. The 15 mean value of particle depolarization ratio at 532 nm was 30±4.5%, however during strong dust 16 episodes this ratio increased to 35±5%, which is also in agreement with the results of the SAMUM campaigns. The backscattering Ångström exponent during the dust episodes decreased 17

to ~-0.7, while the extinction Ångström exponent though being negative, was greater than -0.2. Low values of BAE can likely be explained by an increase in the imaginary part of the dust refractive index at 355 nm compared to 532 nm. The dust extinction and backscattering coefficients at multiple wavelengths were inverted to the particle microphysics using the regularization algorithm and the model of randomly oriented spheroids. The analysis performed has demonstrated that the spectral dependence of the imaginary part of the dust refractive index may significantly influence the inversion results and should be taken into account.

8

9 **1. INTRODUCTION**

10 The impact of desert dust emitted into atmosphere on the Earth's radiation budget is the 11 subject of intense research (Sokolik and Toon, 1996; Balkanski et al., 2007; Mahowald et al., 12 2010; Formenti et al., 2011, 2014). Due to the wind patterns involved, dust can be transported far 13 away from the main source regions in Africa and Asia allowing dust to be distributed in varying 14 amounts all over the globe. North Africa is the largest source of dust in the world and several 15 field campaigns have been conducted to evaluate dust particle microphysical properties over 16 Western Africa and to study long range transport of Saharan dust (Reid et al., 2003; Tanre et al., 17 2003; Redelsperger et al., 2006; Haywood et al., 2008; McConnell et al., 2008). During these 18 campaigns, dust particles were studied via aircraft, ground sampling and using sun photometer 19 measurements. However, vertical distribution of dust has received little attention even though 20 dust vertical structure is critical for an improved understanding of dust advection, transport and 21 dust-cloud interactions. The commonly used instrument to evaluate the height profile of dust 22 particle properties is the aerosol lidar. The numerous measurements performed in Europe, 23 America and Asia with multiwavelength Raman and HSRL lidar systems have resulted in a 24 significant amount of information about the vertical distribution of dust intensive properties, 25 such as depolarization, lidar ratios, extinction and backscattering Ångström exponents (Sakai et 26 al., 2003; De Tomasi et al., 2003; Shimizu et al., 2004; Mona et al., 2006; Papayannis et al., 27 2008; Xie et al., 2008; Ansmann et al., 2012; Burton et al., 2014; Nisantzi et al., 2015). However 28 these measurements were mostly performed at a significant distance from the source area, so the 29 dust particles were aged due to mixing with local aerosols and coating with soluble aerosol 30 species (Li et al., 2009) and may not have well represented the characteristics of the dust upon initial emission. 31

1 To analyze the properties of pure dust measurements near the source regions are needed. 2 Such measurements of Saharan dust were performed during the SAMUM1 and SAMUM2 3 experiments using the assembly of Raman and HSRL lidars (Ansmann et al., 2011). During 4 those measurements the dust episodes and more complicated events, when the dust and smoke 5 layers occurred simultaneously, were studied (Tesche et al., 2009a,b; 2011; Esselborn et al., 2009). However, for the estimation of aerosol radiative forcing not only the particle intensive 6 7 parameters, but also their microphysical properties, such as size, concentration and the complex 8 refractive index (CRI) are needed. An estimation of the vertical distribution of particle 9 microphysics can be achieved, for example, by combining lidar and sun photometer 10 measurements; a review of such studies can be found in a recent publication (Binietoglou et al., 11 2015). However in these retrievals the mean radii and refractive indices of particles in the fine 12 and the coarse mode are assumed to be height independent, and only particle volume in each of 13 the modes is permitted to vary. Such assumptions may become invalid when aerosol layers of 14 different origins occur.

15 The alternative approach to evaluating the vertical distribution of dust properties is to 16 estimate the particle properties from lidar measurements only. Raman (or HSRL) 17 multiwavelength lidars based on a tripled Nd:YAG laser are able to provide three particle 18 backscattering and two extinction coefficients (so called $3\beta+2\alpha$ dataset). Different techniques 19 have been considered to invert these measurements into particle microphysics (Ansmann and 20 Müller, 2005), but the main issue is small number of input measurements (typically five), 21 compared to the numerous parameters needed for describing the aerosol microphysical 22 properties. This implies that the inverse problem is underdetermined and that numerous solutions 23 may reproduce the input measurements with similar accuracy. This family of solutions can be 24 localized by applying constraints to the "search space", i.e. limiting the range of particle radii and 25 refractive indices considered. The additional assumption usually made is that the refractive index is spectrally independent and identical over the whole size range (Müller et al., 1999; 26 27 Veselovskii et al., 2002). Such an approach has proved to be efficient for aerosol particle size 28 distributions (PSD) with a predominant fine mode as, for example, in the case of biomass 29 burning aerosols (Müller et al., 2005; Veselovskii et al., 2015). However, in the case of dust the 30 inversion of lidar measurements becomes more challenging since the dust PSD contains a strong 31 coarse mode with particle radii extending up to $\sim 15 \,\mu m$ and the estimation of properties for such

1 big particles is less accurate when measurements are only performed in the wavelength range of 2 355-1064 nm. Moreover, dust particles are of irregular shape and Mie theory is thus not 3 applicable for computations of their scattering properties. Also, the imaginary part of the 4 refractive index (RI) of dust is spectrally dependent, with a strong enhancement of the absorption 5 in the UV region (Patterson et al., 1977). And finally, particles in the fine and coarse mode may 6 have different origin, so the size dependence of the refractive index should also be considered. 7 The complexity of the problem outlined above demands the use of assumptions and 8 simplifications in the retrieval algorithms.

9 A widely used model for treating irregularly shaped particles is the one used in the 10 operational AERONET algorithm that mimics dust scattering properties with an assembly of 11 randomly oriented spheroids (Mishchenko et al., 1997; Dubovik et al., 2006). For typical dust 12 PSDs the AERONET model provides lidar and depolarization ratios which agrees reasonably 13 well with observed values (Wiegner et al., 2009). The first attempts to invert lidar dust 14 measurements into particle microphysics using the spheroids model were recently made 15 (Veselovskii et al., 2010; Di Girolamo et al., 2012; Papayannis et al., 2012) but were applied to 16 lofted layers of aged dust over Europe. The only test of the spheroidal model relevant to pure 17 dust was performed by using the data acquired during the SAMUM-1 and SAMUM-2 campaigns 18 (Müller et al., 2013). Results indicate that the effective radii derived from lidar measurements are in reasonable agreement with the values provided by AERONET and airplane sampling, while 19 20 differences are significant for the refractive index.

21 The application of spheroids to the analysis of lidar dust observations is an important step 22 forward when compared to the spherical particle approximation of Mie theory. Still we should 23 keep in mind that spheroid model was not specifically designed for lidar applications where 24 scattering in the backward direction is considered. For instance, as previously discussed (Gasteiger et al., 2011; Müller et al., 2013) the spheroidal model has difficulty in reproducing 25 26 depolarization ratios (δ) greater than 30%, values that are representative for pure dust. When 27 using the spheroidal model, such high depolarization ratios can only be obtained when the real 28 (m_R) and imaginary (m_i) parts of the refractive index are less than 1.5 and 0.005, respectively 29 (Dubovik et al., 2006), even though coincident in situ measurements of dust report higher values (Kandler et al., 2011). To investigate these issues, more measurements near the dust origin 30 31 source and more tests of suitable inversion schemes are needed.

1 West Africa and the adjacent oceanic regions are very important locations for studying 2 dust properties and their influence on weather and climate. The SHADOW (Study of SaHAran 3 Dust Over West Africa) campaign is performing a multi-scale and multi-laboratory study of 4 aerosol properties and dynamics using a set of in situ and remote sensing instrumentation (multi-5 wavelength Raman LIDAR, Wind-LIDAR, nephelometer, aethalometer, sun/lunar photometer, 6 airborne sunphotometer, optical particle counter) in the framework of the CaPPA (Chemical and 7 Physical Processed in The Atmosphere) project (http://www.labex-cappa.fr/). The site is located at IRD (Institue for Research and Development) Center, Mbour, Senegal (14⁰N, 17⁰W). The 8 9 objective of the experiment is to report the optical, chemical and physical properties of the 10 aerosols as well as the source apportionment in a location where aerosol loading can be very 11 large and aerosol type depends on the season. Two enhanced observing periods of 7 weeks are 12 considered: March-April 2015 when dust due to the Harmattan, which is a dry trade wind that transports dust laden air from the Sahara to West Africa (Schwanghart and Schütt, 2008) is 13 dominant, and December 2015-January 2016 when dust and carbonaceous aerosols resulting 14 15 from fire activities are in variable proportion and transported at different altitudes. Other types of 16 aerosols can also be present such as sulfates from nearby urban areas or maritime aerosols 17 depending on the air mass flow. The mixed state of these various chemical components results in 18 different radiative properties of the aerosols.

We hereinafter focus our study on multiwavelength Mie-Raman lidar measurements 19 20 performed during the first phase of the SHADOW campaign for the period 8 March - 24 April 2015. During this period approximately 40 day- and night-time measurement sessions were 21 22 performed and numerous strong dust episodes were observed. Those lidar observations are used 23 for the analysis of the vertical distribution of the dust intensive and microphysical properties. In 24 section 2 we describe the lidar equipment and in section 3 we provide examples of joint 25 measurements of wind and Raman lidars. Section 4 presents day-to-day variation of dust 26 properties and examples of vertical distribution of dust intensive parameters. The results of 27 inversion of lidar measurements into particle microphysics are given in section 5.

28 29

30 2. LIDAR EXPERIMENTAL SET

Data from three lidar systems were available during SHADOW campaign. These systems
 are: Doppler wind lidar and multiwavelength Mie-Raman lidar.

3

Doppler lidar

4 The wind field within the lower troposphere (<5 km) was measured by an eye safe 5 scanning wind lidar (Windcube WLS 100) manufactured by the LEOSPHERE company (www.leosphere.com). This pulsed Doppler lidar operates at 1543 nm with a repetition rate of 10 6 7 kHz and uses a heterodyne technique to measure the Doppler shift of laser radiation 8 backscattered by aerosols. Simultaneous measurements of radial wind speed and aerosol 9 backscatter provides information on both aerosol layer stratification and the dynamics of the 10 lower troposphere (Thobois and Soderholm, 2015). More technical details are given by (Kumer 11 et al., 2014; Ruchith and Ernest Raj, 2015).

12 During this experiment, continuous monitoring of the wind field in the range from 100 m 13 to 5 km with 50 m range resolution was performed. The total scanning cycle included two 180° scans in the vertical plane along East/West and South/North axes with 1° resolution, 360° 14 15 azimuthal scan with 2° resolution at 5° elevation angle, and line of sight (LOS) profiles at 75° 16 elevation in the four cardinal directions. The duration of the total cycle was approximately 10 17 minutes. The combination of LOS sequences is used in order to determine the three components 18 of the wind vector vertical profile relying on the Doppler Beam Swinging (DBS) technique 19 (Browning and Wexler, 1968).

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Multiwavelength Mie-Raman lidar

21 The LILAS multiwavelength Mie-Raman lidar is based on a tripled Nd:YAG Spectra Physics

INDI laser with a 20 Hz repetition rate, and pulse energy of 90/100/100 mJ at 355/532/1064 nm.

23 The backscattered light is collected by a 40-cm aperture Newtonian telescope, which is inclined

at an angle of 47 degrees to the horizon. Measurements were performed from inside the IRD

25 building through a window and 47 deg to horizon was the maximal angle possible.

26 . The outputs of the detectors (R1924, R9880 PMTs for 355 nm-532 nm spectral range 27 and APD for 1064 nm) are recorded at 7.5 m range resolution using Licel transient recorders that 28 incorporate both analog and photon-counting electronics. The full geometrical overlap of the 29 laser beam and the telescope FOV is achieved at 800 m -1400 m range depending on FOV used. 30 The system is designed for simultaneous detection of elastic and Raman backscatter signals and 31 thus provides three particle backscattering and two extinction coefficients along with

1 depolarization ratio at 532 nm (so called $3\beta + 2\alpha + 1\delta$ set). For the calibration of depolarization 2 measurements, the so called $\pm 45^{\circ}$ method, (Freudenthaler et al., 2009) was used. The relative 3 uncertainty of depolarization measurements due to calibration is estimated as $\pm 15\%$. Acquiring 4 Raman backscatter at 408 nm permits profiling the water vapor mixing ratio (WVMR) (Whiteman et al., 1992). For calibration of the WVMR, radiosonde launches from the Dakar 5 6 airport, located ~70 km from Mbour, were used. The large separation between the lidar and 7 radiosonde locations prevented an accurate calibration of the WVMR so the WVMR data were 8 used mainly to monitor the relative change of the water vapor content. To improve the system 9 capability for particle extinction measurements at 532 nm, rotational Raman (RR) scattering was 10 used instead of vibrational nitrogen Raman scattering at 608 nm (Veselovskii et al., 2015). For 11 each profile, 4000 laser pulses were accumulated so the temporal resolution of the measurements 12 was approximately 3 minutes. The backscattering coefficients and depolarization ratio were 13 calculated with range resolution 7.5 m (corresponding height resolution 5.5.m). Resolution of 14 extinction measurements varied with height from 50 m (at 1000 m) to 125 m at (at 7000 m).

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3. TROPOSPHERE STRATIFIACATION AND DYNAMICS

17 The aerosol layer stratification over the observation site was mixed-up and difficult to 18 analyze. To demonstrate the advantage of the joint use of wind and Raman lidar measurements, 19 we provide an example of observations performed on the night of 15-16 April. The transport 20 paths of different stratified air masses have been studied by using back trajectories from the 21 NOAA HYSPLIT model (http://ready.arl.noaa.gov/HYSPLIT.php).

For period from 23:00 UTC to 7:00 UTC on 15-16 April 2015 night, the time-height 22 23 sections of the logarithmic range corrected signal (LRCS) is shown in fig.1 while fig.2 shows the 24 horizontal wind speed (color scale) and direction (arrow) deduced from wind lidar and the sonic 25 anemometer wind measurements near the ground. Back trajectories of the air masses ending in 26 Mbour on 16 April 2015 at 2500 m (02:00 UTC, 06:00 UTC), at 900 m (00:00 UTC) and at 700 27 m (06:00 UTC) are reported in Fig.3. These figures reveal complex stratification and dynamics 28 of the lower troposphere on 15-16 April: we can distinguish four layers (A-D) from 100 m to a 29 height of approximately 3000 m. In parallel, the wind field highlights the appearance of multi-30 layered wind structure mainly consisted of a northerly wind (downward arrow) prevailing near 31 ground, which changes to an easterly wind (leftward arrow) with height (fig. 2).

Layer A, located between 1000 m to 3000 m (at 00:00 UTC), is associated with a small northerly wind speed (< 5 m/s) in the lower part of the layer, and a slightly larger easterly wind speed (> 5 m/s) above 2000 m. Layer A can be considered to be a continentalized maritime trade (CMT) wind which is one of oceanic origin that has been progressively altered by continental trade (CT), as follows from the back trajectories shown in fig.3. Therefore, this layer is characterized by a mixture of maritime and continental air.

- Layer B located between 400 m 800 m at the beginning of the study period rises
 progressively up to 700 m 1000 m by the end of the dataset. This layer is characterized by
 northeasterly winds and high aerosol loading. According to the back trajectories shown in
 fig. 3, this air mass was transported from a continental area (Mali) and was mainly advected
 by a southeasterly continental wind (CW).
- Layer C is a nocturnal low-level jet (LLJ). The jet core height is between 250 and 400 m
 with a maximum jet speed exceeding 15 m/s. The LLJ was observed throughout the night
 with a thickness that progressively increased with time perhaps being the causative
 mechanism for the corresponding increase in height of layer B (fig.1). The LRCS values
 within layer C decrease progressively up to the end of the observation period, perhaps due to
 dilution of the aerosol loading.

Finally, layer D corresponds to the nocturnal boundary layer (NBL) characterized by high
 LRCS values and by small northerly or northwesterly wind speed (< 5 m/s). The NBL top
 can be deduced from the LRCS profile discontinuity (Seibert et al., 2000) and is estimated as
 approximately 200-300 m during the night.

22 Fig.4 shows the particle extinction at 532nm (4-a), water vapor mixing ratio (4-b), lidar 23 ratio (4-c) and depolarization ratio (4-d) both at 532 nm for the same time-height section as in 24 fig. 1 and 2. The water vapor can be used as a convenient tracer to separate dry continental air 25 masses from oceanic air masses that are characterized by higher vapor content. Due to the 26 geometrical overlap factor, the LILAS minimum height of the measurements shown in fig. 4 is 800 m. Still, layer B (CW) is well observed starting at 03:00 UTC (fig.4a) due to the increase of 27 28 the layer height. The particle extinction α_{532} in layer A increases after 03:00 UTC while the 29 mixing ratio is decreasing (fig.4b). This may indicate that continental air mass advected by CT 30 has become dominant. The lidar ratio LR₅₃₂ of the particles associated with CT is about 55 sr

1 while for CMT as observed during the first part of the observation period, it is lower (about 45 2 sr). The depolarization ratio δ_{532} is about 30% in layer A and shows a small enhancement up to 3 35% for layer B. We should mention also, that for air masses which we consider as 4 "continentalized maritime" the particle depolarization ratio is in excess of 25%, implying a 5 significant amount of dust even in this layer.

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7 4. DUST PARTICLE PROPERTIES DERIVED FROM RAMAN LIDAR

8 **OBSERVATIONS**

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4.1. Day-to- day variation of particle intensive parameters

10 One of the goals of the SHADOW campaign was to study the dust particle intensive parameters such as extinction and backscattering Ångström exponents together with lidar and 11 12 depolarization ratios. During March-April 2015 about 40 measurement sessions, including both 13 day and night time periods, were performed. In the analysis presented below only night time 14 measurements are considered, and for every session all lidar signals measured during the night 15 are temporally averaged. Moreover, for an evaluation of day-to-day variations of the particle 16 parameters we use only extinction and backscattering coefficients averaged within 1500 - 2000 17 m height layer, where a high dust concentration is frequently observed. For such extensive averaging the uncertainties of derived parameters are mainly due to the systematical errors. Thus 18 19 we estimate the uncertainty of extinction and lidar ratio calculation to be below 10% and 15% 20 respectively for both wavelengths. Uncertainties of extinction and backscattering Ångström exponents derivation are estimated to be below ± 0.2 . 21

22 To give an overview of the variation in aerosol loading, the aerosol optical thickness (AOT) at 440 nm together with the extinction Ångström exponent (EAE) $A^{\alpha}_{380/500}$ measured with 23 24 Cimel sun photometer is reported in Fig.5 for the 10 March-23 April 2015 period. The AOT was 25 relatively low (mainly below 0.4) for 17-28 March, but increased after 28 March reaching values 26 up to 2.0. The high AOTs are associated with low values of the extinction Ångström exponent indicating numerous dust episodes. Fig.6 shows the particle extinction α_{532} together with 27 extinction (EAE) and backscattering (BAE) Ångström exponents $A^{\alpha}_{355/532}$, $A^{\beta}_{355/532}$ derived from 28 29 the lidar measurements for the same time period. During 28 March – 15 April several strong dust 30 episodes occurred as indicated by averaged over night particle extinction values as high as 0.5

km⁻¹. The insert in fig.6 provides the frequency distribution of observed EAE and BAE values.
EAE varies in -0.15 - 0.3 range, while BAE presents stronger variation, because it is more
sensitive to the change of complex refractive index (CRI) and decreases to as low a value as 0.55 during dust events.

The day-to-day variation of the lidar ratios at 355 nm and 532 nm together with particle depolarization ratio at 532 nm is shown in fig.7. The lidar ratios at both wavelengths vary in the 40-65 sr range and the frequency distribution for the ratio LR_{355}/LR_{532} is given by insert in fig.7. In 60% of the cases the ratio LR_{355}/LR_{532} is close to 1, but during dust events this ratio increased up to 1.4. The mean values of lidar ratios are close: $LR_{355}=54\pm8$ sr and $LR_{532}=53\pm8$ sr. The mean value of particle depolarization ratio is $30\pm4.5\%$, however during the dust events depolarization ratio could increase up to $35\pm5\%$.

12

4.2. Vertical distribution of particle intensive properties

The vertical distribution of particle intensive properties is strongly influenced by the origin of the air masses which during the SHADOW measurement period were coming either from ocean or continental regions. In this section, we present the results for three days (13, 29 March and 10 April) characterized by different types of air masses.

17 *13 March*

18 As follows from fig.8, on 13 March at 21:00 UTC the air masses at the 3 heights (1500, 2500 and 3500m) were transported mainly over the ocean, but the back trajectory at 1500 m 19 20 presents a "loop" over continent, so the corresponding air masses may contain more dust 21 compared to other heights. The relative humidity measured with meteorological sonde in Dakar 22 was below 38% inside 1000 m – 2600 m height range and it increased up to 75% above 3350 m. Fig.9 shows the vertical profiles of $3\beta+2\alpha$ measurements together with lidar ratios LR₃₅₅, LR₅₃₂, 23 depolarization ratio δ_{532} , and Ångström exponents $A^{\alpha}_{355/532}$, $A^{\beta}_{355/532}$ on 13 March 2015 averaged 24 25 over the 20:30-21:30 time period. The aerosol layer extended up to 3500 m but the extinction 26 coefficient α was relatively small; at both 355 and 532 nm wavelengths α did not exceed 0.16 km⁻¹. The particle depolarization ratio at 532 nm was approximately 31±4.5% inside the dust 27 layer (up to ~2750 m) and decreased to less than 15% at 3250 m. Likewise, the $A^{\alpha}_{355/532}$ and 28 $A_{355/532}^{\beta}$ are close to zero up to 2750 m, but start to increase above indicating the presence of 29

1 smaller particles. The lidar ratios LR_{355} and LR_{532} are approximately 53 ± 8 sr inside the dust 2 layer. Above 2750 m the values of LR are more noisy but do not seem to change.

3

29 March

4 The backtrajectories from the night of 29-30 March associated to a strong dust case are 5 shown in Fig.10. The air masses at low altitude were transported over the continent and were 6 strongly loaded with dust. The relative humidity was below 19% in 600 m - 1500 m height range 7 Fig.11 presents the vertical profiles of the same particle parameters as in fig.10 but for 29 March. The extinction coefficient α inside the dust layer (below 1500 m) is greater than 0.6 km⁻¹ for 8 9 both wavelengths. The backscattering coefficient β_{355} nm inside the dust layer is lower than β_{532} 10 which is consistent with the lidar ratio R larger at 355 nm than that at 532 nm with values as large as 65 sr. The $A_{355/532}^{\beta}$ (BAE) is negative and gets near -0.8, while EAE is still close to 0 as 11 12 observed on 13 March (Fig.9). The negative values of BAE can result from the spectral 13 dependence of the imaginary part of the dust refractive index (RI) which is larger at 355 than at 14 532 nm (e.g. Patterson et al., 1977; Ansmann et al., 2011).

15 The ground based measurements performed during the SAMUM campaign demonstrated that the imaginary part of the dust RI could vary from $m_1=0.005$ at 532 nm to $m_1=0.02$ at 355 nm 16 17 (Ansmann, et al., 2011). Such a strong enhancement of m_I may lead to a decrease of the 18 backscattering coefficient (Veselovskii et al., 2010). To estimate the impact of the m_I 19 enhancement at 355 nm on the values of EAE and BAE at 355/532 nm wavelengths, numerical 20 simulations were performed. Extinction and backscattering Ångström exponents were calculated 21 using the model of randomly oriented spheroids as described in (Veselovskii et al., 2010) for a 22 bimodal particle size distribution:

23
$$\frac{dn(r)}{d\ln(r)} = \sum_{i=f,c} \frac{N_i}{(2\pi)^{1/2} \ln \sigma_i} \exp\left[-\frac{(\ln r - \ln r_i)^2}{2(\ln \sigma_i)^2}\right].$$
 (1)

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where $N_{f,c}$ is particle number density in the fine (*f*) and the coarse (*c*) mode. Each mode is represented by a lognormal distribution with modal radius $r_{f,c}$ and dispersion $\ln \sigma_{f,c}$. For the fine mode, values of $r_{f}=0.1 \ \mu\text{m}$ and $\ln \sigma_{f}=0.4$ were used. For the coarse mode $r_{c}=1.0 \ \mu\text{m}$ and three values $\ln \sigma_{c}=0.4$, 0.5, 0.6 were considered. The three size distributions expressed in volume are reported in the insert of fig.12. The ratio N_c/N_f in all cases was 0.01, and the real part of CRI was 1.55 for all wavelengths. The imaginary part was fixed at 0.005 for 532 nm while it varied within the 0.005 – 0.05 range at 355 nm. Values of EAE and BAE as a function of m_I at 355 nm are given by fig.12. The EAE shows no significant sensitivity to changes in m_I , but BAE decreases rapidly as a function of m_I at 355 nm. The present sensitivity study is limited but illustrates the importance of accounting for the right spectral dependence of $m_I(\lambda)$.

6

10 April

7 On April 10, the air masses were coming from continental regions and particle 8 parameters showed large variation with height. We selected measurements during the period 9 0:00-2:00 UTC for which the backward trajectories at 1:00 UTC are shown in fig.13. The air 10 masses at 2000 m and 3000 m originate from the dust-laden continental region (Barren or 11 sparsely vegetated areas), while at 4500 m the air masses come from regions covered by grass 12 lands and savannas. Fig.14 shows profiles of the $3\beta+2\alpha$ measurements together with particle 13 intensive parameters. The particle extinction increases with height reaching a maximum value of around 0.2 km⁻¹ for both wavelengths at a height of approximately 3000 m and then decreases up 14 15 to 5 000m. The EAE is approximately zero up to 3000 m and then it increases to 1.0 at 4500 m. The BAE below 3000 m is smaller with minimum value $A_{355/532}^{\beta} \approx -0.5$, but increase up to 4500 m 16 17 where EAE and BAE are approximately equivalent. The depolarization ratio is around 30% in 18 the 2000-3500 m range, and decreases for higher altitudes. The lidar ratio at 355 nm inside 2000-19 3500 m layer exceeds that at 532 nm (averaged values are 60 ± 9 sr and 53 ± 8 sr correspondingly) 20 and for higher altitudes the lidar ratios at both wavelengths are close. So we can identify 21 different aerosol layers with different properties: mostly pure dust layer within the 2000-3500 m 22 altitude range and mixed aerosols above it.

23 The relative humidity on 10 April was higher than on 13, 29 March, which could impact 24 the particle properties. Fig.15 shows the estimated profile of water vapor mixing ratio (WVMR) 25 obtained from the lidar measurements. WVMR is less than 3 g/kg within the dust layer and 26 increases above 3500 m reaching approximately 5.5 g/kg at 4000 m. The WVMR and the 27 relative humidity measured in Dakar at 0:00h using a radiosounding is reported on Fig.15 for 28 comparison. Both WVMR's measured by sounding and lidar are in agreement between 3000m 29 and 5000m (note that there is no sounding data between 4620 m and 3880 m). There are clearly 30 two distinct layers. If the derived properties of aerosols within the lower layer are representative

1 of dust, the air mass above 4000m brings another particle type. Particles, characterized by lower 2 depolarization ratio, are smaller since the EAE is increasing, and the layer is more humid since 3 the RH is increasing. Based on the analysis of the satellite data quick-looks (see for instance 4 http://earthobservatory.nasa.gov/GlobalMaps/), the back-trajectories reporting in Fig. 13 show 5 that the air mass at 4500m is coming from regions where fires were active during several days, 6 which can result in emission of smoke particles transported over M'Bour few days later. The 7 derived properties of aerosols within the 4000-5000m layer are consistent with this hypothesis; 8 the assumption of the air-mass origin is also consistent with the RH increase.

9

10 5. INVERSION OF RAMAN LIDAR OBSERVATIONS TO THE PARTICLE 11 MICROPHYSICS

12 The lidar $3\beta+2\alpha$ and $3\beta+2\alpha+1\delta$ observations analyzed in the previous sections can be 13 inverted into microphysical properties using regularization algorithm. As previously mentioned, 14 in the case of irregularly shaped dust particles such inversion is more complicated compared to 15 other aerosol types that may be well handled by spherical particle assumptions. In an earlier 16 study, a model of randomly oriented spheroids for dust was used (Veselovskii et al., 2010). This 17 model handles the dust particles as a mixture of spheres and spheroids, so an additional unknown 18 parameter, spheroids volume fraction (SVF), appears. The SVF in principle can be determined in 19 the process of inversion of $3\beta+2\alpha+1\delta$ measurements thanks to the use of depolarization ratio as 20 input parameter. However, for the dust layers, in a first guess, we assume a value of SVF=100% 21 to decrease the number of retrieved parameters. In the process of inversion we used algorithm from (Veselovskii et al., 2010) and the "search space" parameters similar to those described in 22 23 (Müller et al., 2013). The boundary of the inversion window has been set to minimum and 24 maximum particle radii of 0.075 and 15 µm, respectively. The real part of RI was allowed to 25 vary in the range 1.35 - 1.65, while the imaginary part varied in the range 0 - 0.02. The refractive 26 index was assumed to be spectrally independent. The effects of a possible spectral dependence of 27 the imaginary part of RI will be considered at the end of this section.

Fig. 16 shows the particle volume density retrieved from $3\beta+2\alpha$ measurements on 13 March, 29 March and 10 April, which were discussed in the 4.2 section. The profiles of particle volume are given together with corresponding extinction coefficients at 532 nm. The volume – extinction ratio V/ α_{532} for these days is also reported as an insert. Inside the dust layer this ratio 1 varies within the range $(0.50-0.65)*10^{-6}$ m, while outside the dust layer, the V/ α_{532} ratio 2 decreases. An overview of observed values of the volume – extinction ratio for dust, obtained 3 from in situ, AERONET and lidar measurements is presented in Ansmann et al., 2012 and 4 provides V/ α_{532} varying within the range $(0.60-1.29)*10^{-6}$ m. Thus our results are near the low 5 boundary of these previously published results.

6 The profiles of the effective radius and the real part of RI are shown in fig.17. The 7 inverted effective radius inside the dust layer is between 1.05 and 1.25µm (1.15±0.3 µm) and 8 similar for the 3 days. The AERONET retrievals provided column integrated values that are in 9 the same range and agrees within the uncertainty. On 30 March early morning, when the dust 10 contribution to the AOT is prevailing, the effective radius $r_{eff}=1.36 \mu m$ and it varies between 11 0.918 and 1.70 depending on the days and time. The lidar retrievals indicate that the real part of 12 the CRI in the dust layer varied from 1.51±0.05 to 1.57±0.05, which is quite typical for desert 13 dust (Patterson et al., 1977), while the AERONET retrievals yield values between approximately 14 1.46 and 1.58 depending on the days. Outside of the dust layer the retrieval of m_R is not reliable 15 because the assumption of SVF=100% is not fulfilled and, as a result, the retrieved values of m_R 16 are overestimated (Veselovskii et al., 2010).

The values of the imaginary part of the CRI retrieved from lidar measurements are approximately 0.007 inside the dust layer. However, the retrieved value is associated to average value over spectral range and characterized by high uncertainty (Müller et al., 2013). Still, this uncertainty does not affect significantly the retrieved values of volume and effective radius (Veselovskii et al., 2013.

22 The regularization approach estimates also the particle volume size distribution (PSD), 23 allowing to conclude, which of the modes (fine or coarse) is predominant. Fig.18 shows the 24 PSDs derived from lidar measurements on 10 April for four height layers of 150 m width 25 centered at 1940, 3150, 4070, 4370 m heights. For the layers with strong dust loadings (1940, 26 3150 m) the coarse mode is dominant, at higher altitude outside the dust layer (4070, 4370 m), 27 the fine mode (around 0.15µm) prevails. For comparison, the column integrated PSD obtained 28 from AERONET level 1.5 data on 9 April at 18:00 UTC is also reported. The coarse mode looks shifted toward larger particles when compared to the lidar retrievals but the difference can be due 29 to the spectral dependence of the imaginary part of m_I, as it will be discussed further in this 30 31 section.

1 Depolarization measurements provide additional information about particle properties 2 that can be used in the inversion algorithm as long as the forward model can compute the particle 3 depolarization ratio with sufficient accuracy (Veselovskii et al., 2010; Müller et al., 2013). Hereinafter, we compare the retrieved aerosol parameters using $3\beta+2\alpha$ or $3\beta+2\alpha+1\delta$ 4 observations. To perform such a comparison we calculated the ratio of the effective radii (R_s^r) 5 6 derived from $3\beta+2\alpha+1\delta$ and $3\beta+2\alpha$ sets. Fig.19 shows the profiles of R_{δ}^{r} for the same three days 7 (right part associated with bottom x-axis); a value of 1.0 would mean that the additional input 8 has no impact on the retrieval. Inside the dust layer the ratio is about 1.15 for the measurements 9 taken on 13 and 29 of March. On 10 April, the ratio is noisier and more oscillating, but the average is still close to the results obtained for 13 and 29 March. Let us mention that the ratio of 10 the particle volumes R_{δ}^{V} is very close to R_{δ}^{r} , so it is not shown in the figure. The increase of the 11 12 effective radius (and volume) retrieved from $3\beta+2\alpha+1\delta$ measurements compared to $3\beta+2\alpha$ 13 occurs simultaneously with a decrease of the real and imaginary parts of CRI (Veselovskii et al., 14 2010; Müller et al., 2013). m_R and m_I decrease to values less than 1.45 and 0.005, respectively 15 and are lower than expected based on in situ measurements (Müller et al., 2013). It may suggest 16 that the spheroidal model has difficulty to reproduce high depolarization measurements. On March 13 and April 10, the depolarization ratio δ is decreasing above 2500m and 3700m (Figs 9 17 and 14 respectively) and we can notice that the value R_{δ}^{r} is then close to 1. Assuming that results 18 19 obtained using $3\beta+2\alpha$ data are more representative of the actual values, it means that the spheroidal model cannot reproduce high depolarization ratios reasonably well. Additional 20 21 information brought by the depolarization ratio is so not suitable in such conditions.

22 The inversion results presented in fig.16, 17 are obtained assuming a spectrally 23 independent refractive index while the imaginary part of CRI of dust is expected to increase in 24 the UV spectral region. To test the effect of a spectrally dependent imaginary part $m_{I}(\lambda)$ on the 25 retrieval, we now assume that the imaginary parts at 1064 nm and 532 nm wavelengths are the same $m_1(532) = m_1(1064)$, while $m_1(355) = 4m_1(532)$. Such an enhancement of m_1 at 355 nm is 26 quite typical for Saharan dust (Ansmann et al., 2011). The $3\beta+2\alpha$ measurements for the same 27 28 three days are so inverted assuming this $m_I(\lambda)$ spectral dependence as described in (Veselovskii et al., 2010). Fig.19 (left part associated with top x-axis) shows profiles of R_{ml}^{r} , which is the 29

1 ratio of the effective radii retrieved under the assumption of spectrally dependent and spectrally independent imaginary part of RI. Again, the corresponding ratios R_{ml}^{V} for the volumes are close 2 to R_{ml}^r and we do not report them. As expected, the effect of $m_I(\lambda)$ is more pronounced inside the 3 4 dust layer, and on 29 March and 10 April (days characterized by negative BAE), the value of R_{ml}^r is up to 1.5. Accounting for $m_I(\lambda)$ dependence increases also the real part of RI, thus e on 29 5 March m_R increases from approximately 1.52 to 1.56 inside the dust layer. These model 6 7 computations demonstrate that accounting for the spectral dependence of the imaginary part of 8 RI in the dust layers may significantly increase the retrieved values of the effective radius and 9 particle volume. In particular, it may explain smaller radii of the coarse mode particles retrieved 10 from lidar measurements inside the dust layer (fig.19) when compared to AERONET results. 11 The effects of spectral dependence of m_1 can be to some extent corrected by using $m_1(\lambda)$ model 12 based on in situ measurements and implying it in retrieval algorithm as described in (Veselovskii et al., 2010). 13

14

15 CONCLUSION

16 The lidar measurements performed in March-April 2015 during the first phase of the 17 SHADOW campaign in Senegal has provided a significant amount of information about dust 18 particle parameters. The use of rotational Raman scattering in the LILAS for 532 nm observations improved the α_{532} measurements and, as a result, the calculation of lidar ratio and 19 20 extinction Ångström exponent were improved as well. The mean values of lidar ratios of pure 21 dust were about 53 ± 8 sr for both 532 nm and 355 nm wavelengths, which agrees with the values 22 observed during SAMUM 1 (Morocco) and SAMUM 2 (Cape Verde) campaigns. The mean 23 value of particle depolarization ratio at 532 nm was 30±4.5%, however during strong dust 24 episodes this ratio increased up to 35±5%, which is also in agreement with the results of SAMUM campaigns. The backscattering Ångström exponent at 355/532 nm wavelengths during 25 26 the dust episodes decreased to \sim -0.7, while the EAE values, though being negative, were higher 27 than -0.2. Low values of BAE may be a result of enhanced dust absorption at 355 nm.

The inversion of $3\beta+2\alpha$ measurements to particle microphysics in the case of dust is more challenging than other types of aerosols. The use of pure dust observations somewhat simplifies this task, because the contribution of the particles in the fine mode (which may have different

1 origin) is insignificant. Moreover, in the retrieval of pure dust properties we don't need to 2 consider the mixture of spheres and spheroids and can assume SVF=100%. The use of the 3 spheroids model for the inversion of $3\beta+2\alpha$ measurements leads to values of effective radius in 4 reasonable agreement with AERONET observations and yields reasonable values of the real part 5 of RI. However, the use of depolarization measurements $(3\beta+2\alpha+1\delta)$ in the inversion for pure dust, which is characterized by a depolarization ratio δ_{532} exceeding 30%, leads to values of 6 7 effective radius and volume exceeding the corresponding values obtained from $3\beta+2\alpha$ 8 measurements by a factor up to 1.15. At the same time, the values of $m_{\rm R}$ are decreased. These 9 issues have already been discussed so at the current time we prefer to not use the depolarization 10 ratio in the input data set for the inversion of dust particle parameters. We hope also that these 11 discussions will stimulate development of the forward model accurately describing polarization 12 properties of laser radiation backscattered by dust particles.

13 The analysis performed here demonstrates the importance of the spectral dependence of 14 the imaginary part of RI in the UV spectral region. Model simulations demonstrate that including 15 $m_I(\lambda)$ dependence may increase the values of effective radius and volume by a factor as large as 16 1.5. Thus, at the moment, dust particle microphysical properties obtained by inversion of lidar 17 measurements may contain significant biases. Further research is needed to develop techniques 18 correcting these biases in order the uncertainty of the estimates of r_{eff} and V to remain below 19 30%, which is a typical value when particles with prevailing fine mode are considered.

20 In addition to aerosol properties, the LILAS system provided profiles of the water vapor 21 mixing ratio, which, being a conserved quantity, was frequently a convenient tracer that 22 indicated the boundary between dry air masses transported over the continent and moist air 23 masses transported over the ocean. The episodes considered in this paper were characterized 24 mainly by low values of RH and the effects of the particles hygroscopic growth were not 25 considered. Still, we have significant amount of the measurements in the condition of high RH, 26 accompanied by formation of water and ice clouds near the dust layers. We plan to present these 27 results in a separate publication.

During phase 1 of SHADOW the aerosols other than dust didn't occur in significant amount inside the height range available for the measurements, so this publication is focused mainly on the pure dust study. However in phase 2 the episodes of dust-smoke mixture were

- observed and such episodes provide opportunity to test the algorithm ability to profile the layers
 with different particle types. Such analysis is in progress and will be presented separately.

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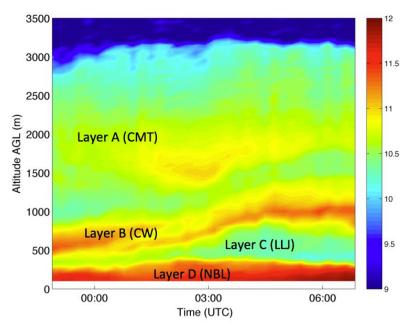




Fig.1. Time-height section of the logarithmic range corrected lidar signal (in arbitrary units) deduced from the Doppler lidar measurements during the 15-16 April 2015 night at Mbour. The stratification is represented by four layers: (A) continentalized maritime trade (CMT), (B) Layer advected mainly by a continental wind (CW), (C) low-level jet (LLJ) and (D) nocturnal boundary layer (NBL).

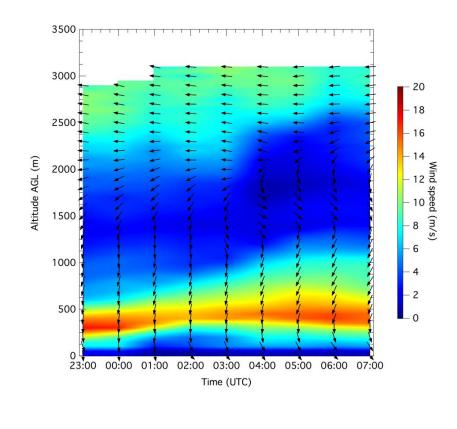


Fig. 2. Time-height section of horizontal wind direction (arrows) and wind speed (color map)
deduced from Doppler lidar during 15-16 April. Leftward and downward arrows represent,
respectively, easterly wind and northerly wind

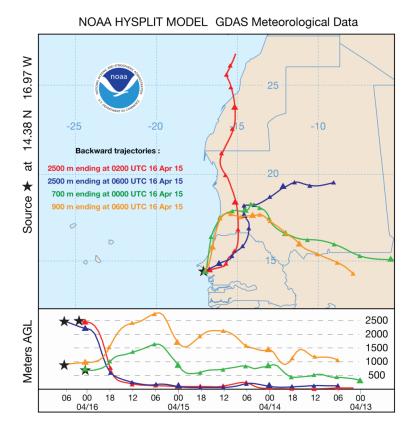
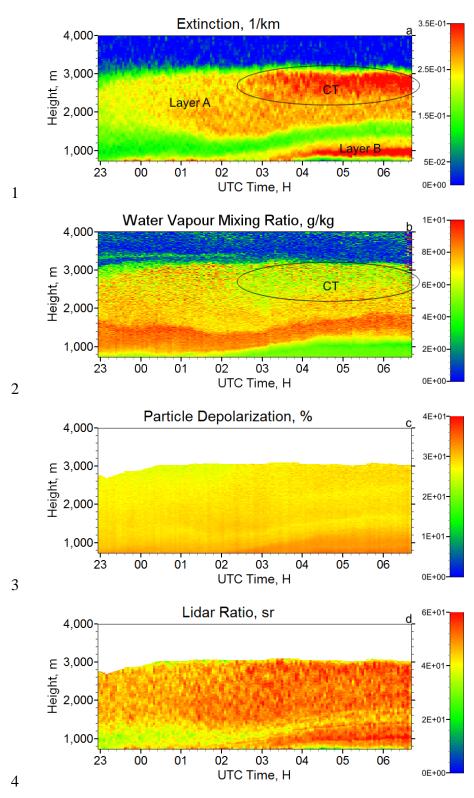
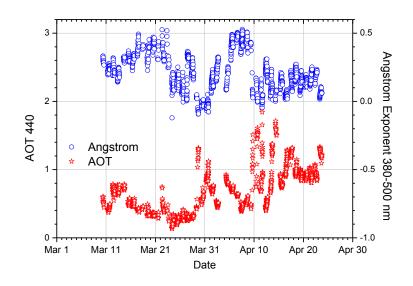


Fig.3. Back trajectories of the air masses ending in Mbour on 16 April 2015 at 2500 m (02:00
UTC, 06:00 UTC), at 700 m (00:00 UTC) and at 900 m (06:00 UTC). First two back trajectories
correspond layer A from fig.1, while last two back trajectories correspond layer B from the same
figure.



5 Fig.4. Height-temporal distribution of particle characteristics: (a) extinction α_{532} , (b) water vapor 6 mixing ratio, (c) particle depolarization and (d) lidar ratio R₅₃₂ measured during the 15-16 April 7



4 Fig.5. Aerosol optical thickness (AOT) at 440 nm and the extinction Ångström exponent at 380-

5 550 nm wavelengths provided by AERONET in Mbour for March – April 2015 period.

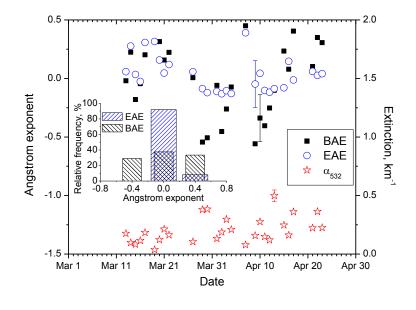
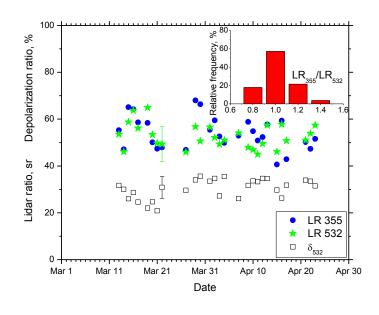


Fig. 6. Particle extinction at 532 nm together with backscattering and extinction Ångström
exponents derived from lidar measurements within 1500 m - 2000 m layer for period MarchApril 2015. The insert shows the frequency distributions of BAE and EAE.

5



6

7 Fig. 7. Lidar ratios at 355 nm and 532 nm together with particle depolarization ratios derived

8 from lidar measurements within 1500 m – 2000 m layer for period March-April 2015. The insert 9 shows the frequency distribution of the ratio LR_{355}/LR_{532} .

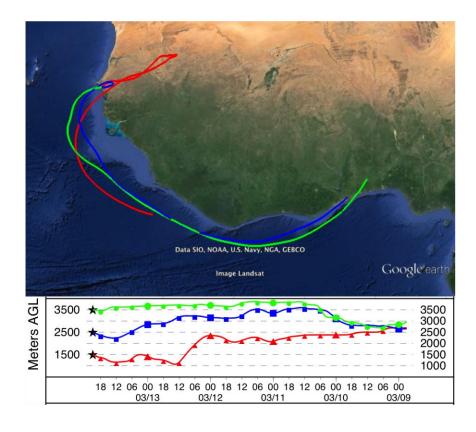


Fig.8. Five-day backward trajectories for the air mass in Mbour at altitudes 1500 m, 2500 m,
3500 m, on 13 March 2015 at 21:00 UTC.

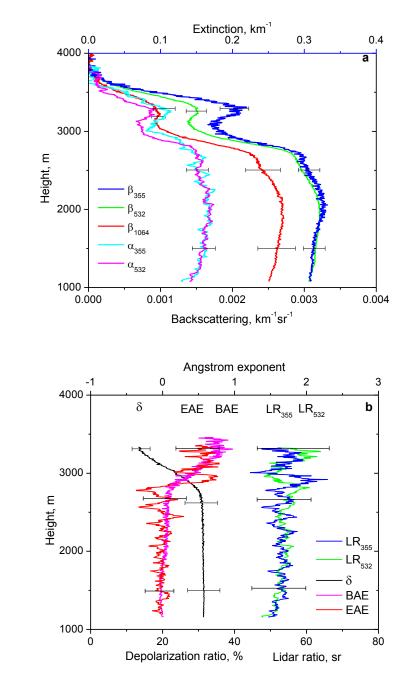


Fig.9. Vertical profiles of (a) backscattering and extinction coefficients and (b) lidar ratios,
depolarization ratio, backscattering and extinction Ångström exponents at 355/532 nm measured
on 13 March 2015 for period 20:30-21:30 UTC.

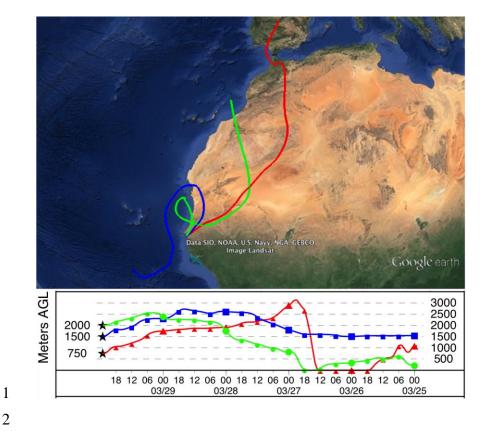
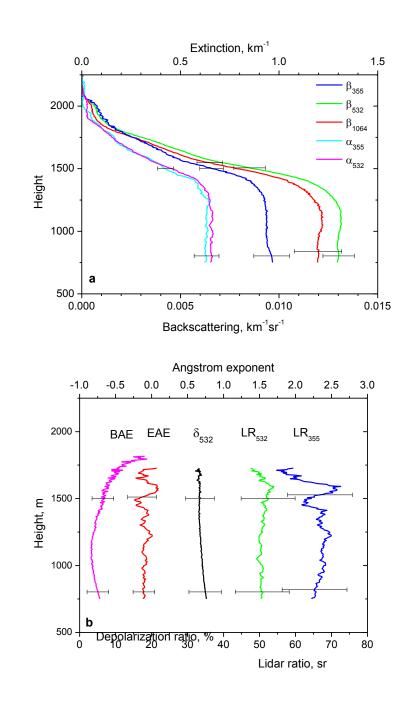




Fig.10. Five-day backward trajectories for the air mass in Mbour at altitudes 750 m, 1500 m, 2000 m on 29 March 2015 at 23:00 UTC.







4 Fig.11. Vertical profiles of (a) backscattering and extinction coefficients and (b) lidar ratios,
5 depolarization ratio, backscattering and extinction Ångström exponents measured on 29 March
6 2015 for period 22:00-23:30 UTC.

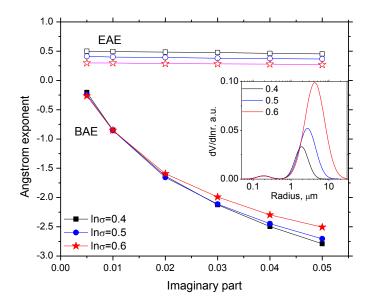




Fig.12. Extinction and backscattering Ångström exponent for 355/532nm wavelengths as a function of the imaginary part of the refractive index at 355 nm. The CRI at 532 nm was kept m=1.55-i.005. Computations were performed using the model of randomly oriented spheroids for three bimodal PSDs shown in the insert.

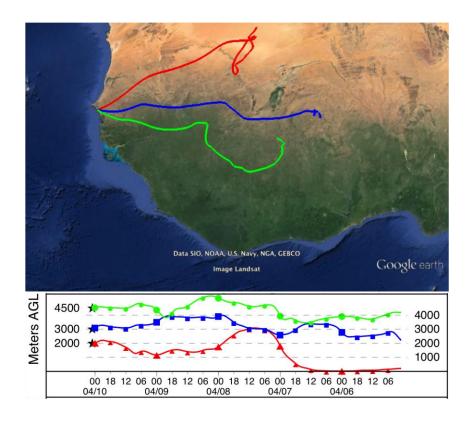


Fig.13. Five-day backward trajectories for the air mass in Mbour at altitudes 2000 m, 3000 m,
4500 m on 10 April 2015 at 01:00 UTC.



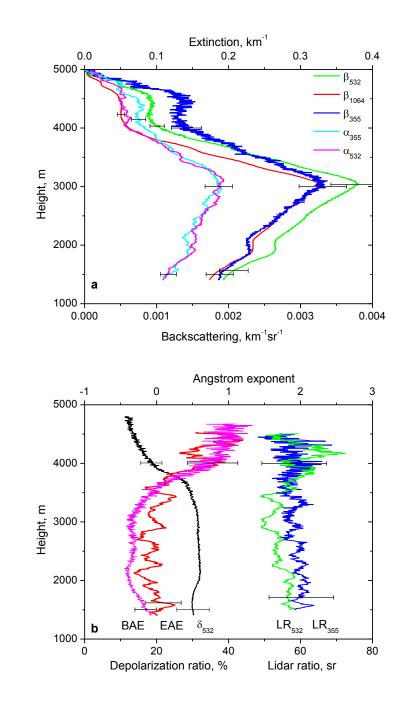
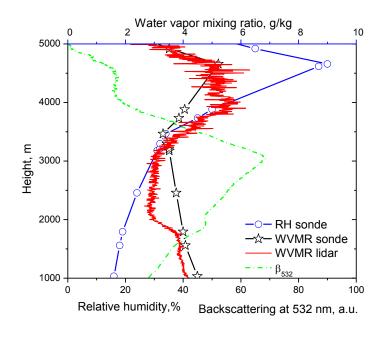
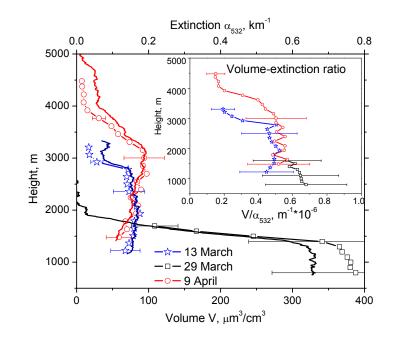


Fig.14 Vertical profiles of (a) backscattering and extinction coefficients and (b) depolarization
ratio, backscattering and extinction Ångström exponents measured on 10 April 2015 for period
00:00-02:00 UTC.



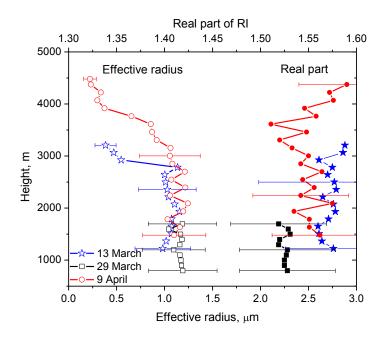
- 2 Fig.15. Vertical profile of water vapor mixing ratio (WVMR) measured with Raman lidar. The
- 3 symbols show WVMR and the relative humidity (RH) measured with radio sonde in Dakar on 10
- 4 April at 00:00 UTC. Green dash-dot line shows backscattering coefficient at 532 nm.







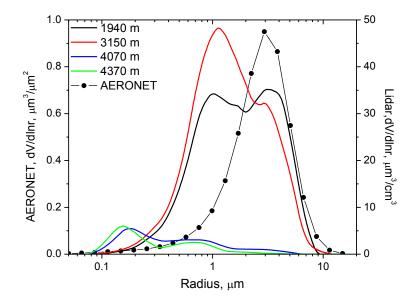
4 Fig.16. Vertical profiles of the particle volume density V retrieved from $3\beta+2\alpha$ measurements on 5 13 March, 29 March and 10 April (symbols). Solid lines indicate the profiles of extinction 6 coefficient at 532 nm. The insert shows the volume – extinction ratio V/α₅₃₂ for the days 7 considered.





2 Fig.17. The profiles of (open symbols) the particle effective radius and (solid symbols) the real

3 part of RI retrieved from $3\beta+2\alpha$ measurements on 13 March, 29 March and 10 April.





2 Fig.18. Particle size distributions retrieved from the measurements on 10 April for four height

layers 1940, 3150, 4070, 4370 m. Symbols show the PSD provided by AERONET on 9 April at

4 18:00 UTC, inversion level 1.5.

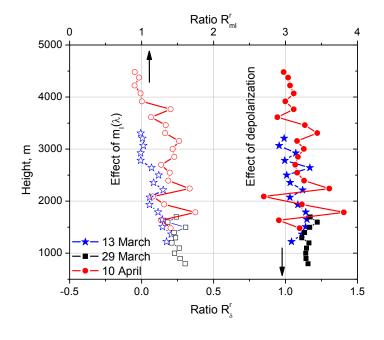


Fig. 19. Enhancement of retrieved effective radius due to using the particle depolarization ratio in input data set (R_{δ}^{r}) and due to accounting for the spectral dependence of the imaginary part of RI (R_{ml}^{r}) .Shown are results for the measurements on 13 March, 29 March and 10 April 2015.