1 **Variability of Lidar-Derived Particle Properties Over West Africa Due to Changes in**

2 **Absorption: Towards an Understanding**

3 Igor Veselovskii[1], Qiaoyun Hu[2], Philippe Goloub[2], Thierry Podvin[2], Mikhael Korenskiy[1],

4 Yevgeny Derimian[2], Michel Legrand[2], Patricia Castellanos[3]

5 *[1]General Physics Institute, Moscow, Russia.*
6 *[2]Univ. Lille, CNRS, UMR 8518 - LOA - Laboratoire d'Optique Atmospherique, Lille F-59000,*
7 *France*
8 *[3]NASA Goddard Space Flight Center, Greenbelt, USA*

9

10 **Abstract**

11 Measurements performed in Western Africa (Senegal) during the SHADOW-2 field

12 campaign are analyzed to show that spectral dependence of the imaginary part of the complex

13 refractive index (CRI) of dust can be revealed by lidar-measured particle parameters. Observations

14 in April 2015 provide good opportunity for such study, because, due to high optical depth of the

15 dust, exceeding 0.5, the extinction coefficient could be derived from lidar measurements with high

16 accuracy and contribution of other aerosol types, such as biomass burning, was negligible. For

17 instance, in the second half of April 2015, AERONET observations demonstrated a temporal

18 decrease of the imaginary part of CRI at 440 nm from approximately 0.0045 to 0.0025. This

19 decrease is in line with a change in relationship between lidar ratios (the extinction-to-

20 backscattering ratio) at 355 nm and 532 nm ($S_{355}$ and $S_{532}$). In the first half of April, $S_{355}/S_{532}$ is as

21 high as 1.5 and the backscatter Angstrom exponent $A_\beta$, is as low as -0.75, while after 15 April

22 $S_{355}/S_{532}=1.0$ and $A_\beta$ is close to zero. The aerosol depolarization ratio $\delta_{532}$ for the whole April

23 exceeded 30% in the height range considered, implying that no other aerosol, except dust,

24 occurred. The performed modeling confirmed that the observed $S_{355}/S_{532}$ and $A_\beta$ values match the

25 spectrally dependent imaginary part of the refractive index as can be expected for mineral dust

26 containing iron oxides. West Africa is also known for significant biomass burning aerosol

27 emissions during the dry season in the Sahel region.

28 The second phase of the SHADOW-2 campaign was focused on evaluation of lidar ratio

29 of smoke and estimates of its dependence on relative humidity (RH). For considered five smoke

30 episodes the lidar ratio increases from 44±5 sr to 66±7 sr at 532 nm and from 62±6 sr to 80±8 sr

31 at 355 nm, when RH varied from 25% to 85%. Performed numerical simulations demonstrate, that

32 observed ratio $S_{355}/S_{532}$, exceeding 1.0 in the smoke plumes, can indicate to increase of the

33 imaginary part of the smoke particles in UV.

34

35

36

## 1. Introduction

37

38  Atmospheric dust provides significant impacts on the Earth's climate system and this

39  impact remains highly uncertain (IPCC report, 2013). In modeling the direct aerosol effect, the

40  vertical profile of aerosol extinction is one of the basic input parameters, and when this profile is

41  derived from the elastic backscatter lidar observations, the knowledge of the extinction-to-

42  backscatter ratio (so called lidar ratio) is essential. Although the desert dust in source regions is

43  sometimes qualified as "pure dust", it is always a mixture of various elements, e.g. iron oxides,

44  clays, quartz and calcium–rich species, which proportions can vary (Sokolik and Toon, 1999;

45  Wagner et al., 2012; Di Biagio et al., 2017, 2019 and references therein). Thus, the dust optical

46  properties, and hence the lidar ratio (S) can vary, depending on relative abundance of various

47  minerals in emission sources. Imaginary part of the complex refractive index (CRI) of different

48  minerals can vary spectrally and often exhibits an increase in UV for dust, containing iron oxides.

49  Therefore, retrieval of the dust extinction profiles from elastic backscatter lidar observation should

50  account for the spectral variation of the lidar ratio.

51  The Raman and HSRL lidars are capable to provide independent profiling of aerosol

52  backscattering and extinction coefficients (Ansmann et al., 1992), and therefore are widely used

53  to measure the lidar ratios of dust from different origins (e.g. Sakai et al., 2003; Papayannis et al.,

54  2008, 2012; Xie et al., 2008; Ansmann et al., 2011; Mamouri et al., 2013; Burton et al., 2014;

55  Nisantzi et al., 2015; Giannakaki et al., 2016; Hofer et al., 2017, 2019). The African deserts are

56  the largest sources of mineral dust and numerous studies have been conducted for quantifying the

57  particle intensive parameters (parameters independent of concentration) during dust transport from

58  this source region to Europe and over the Atlantic Ocean (Mattis et al., 2002; Amiridis et al., 2005;

59  Mona et al., 2006; Papayannis et al., 2008; Preißler et al., 2013; Rittmeister et al., 2017). The dust

60  properties are, however, modified during the transport, experiencing mixing and aging processes,

61  thus characterization of dust properties near the source regions is highly important for evaluation

62  the parameters of "pure dust". The lidar ratios at 355 nm and 532 nm ($S_{355}$ and $S_{532}$) were measured

63  during the SAMUM-1 and 2 experiments in Morocco and Capo Verde respectively (Esselborn et

64  al., 2009; Tesche et al., 2009, 2011; Groß et al., 2011; Ansmann et al., 2011), as well as during the

65  more recent SHADOW-2 experiment in Senegal (Veselovskii et al., 2016, 2018). The lidar ratios

66  $S_{355}$ and $S_{532}$ measured during SAMUM experiments didn't present significant spectral

67  dependence. For example, for SAMUM-2 campaign, the averaged values of $S_{355}$ and $S_{532}$ are

68  53±10 sr and 54±10 sr respectively (Tesche et al., 2011). During SHADOW, however, $S_{355}$

69  significantly exceeded $S_{532}$ in many dust episodes, which was linked to an increase of the

70  imaginary part of CRI of dust at 355 nm (Veselovskii et al., 2016).

71    The dust backscattering coefficient (and so lidar ratio), in contrast to extinction coefficient,

72    is sensitive to the imaginary part of CRI (Perrone et al., 2004; Gasteiger et al., 2011). Thus, it is

73    expected that enhanced absorption in the UV should increase the lidar ratio. In turn, the ratio

74    $S_{355}/S_{532}$ should characterize the spectral variation of the imaginary part of CRI. The latest version

75    of AERONET products (3.0) provides inversions of lidar related properties, including the lidar

76    ratio, from almucantar scans with ground-based sun photometers. For these products, the shortest

77    available wavelength is 440 nm. Despite $Im_{440}$ is lower than $Im_{355}$, AERONET observations still

78    show an increase of absorption at 440 nm in respect to 675 nm that yields a ratio of $S_{440}/S_{675}$ close

79    to 1.4 for Saharan dust (Shin et al., 2018). The goal of this work is to analyze the correlation of

80    variations of $Im_{440}$ from AERONET with measured values from lidar to reveal the effect of dust

81    absorption on lidar-derived aerosol properties. We focus on height and day-to-day variations of

82    the dust intensive properties, such as $S_{355}$ and $S_{532}$, depolarization ratio ($\delta$), as well as the extinction

83    and backscatter Ångström exponents ($A_\alpha$ and $A_\beta$ respectively) measured during several strong dust

84    episodes in April 2015 during the SHADOW-2 campaign.

85    The smoke aerosol particles, typically originated from biomass burning, can also have a

86    pronounced spectral dependence of absorption. This is generally due to presence of carbonaceous

87    particles with organic compounds, so-called brown carbon (BrC) (Sun et al., 2007; Kirchstetter, et

88    al., 2004). The Sahel region is known for seasonal biomass burning caused by human activity on

89    combustion of agricultural waste that can produce an abundant amount of BrC. The smoke can

90    also be mixed with mineral dust during long-range transport or in the emission origin (Haywood

91    et al., 2008). During the SHADOW-2 the observation period included the biomass burning season,

92    thus an additional effort was dedicated to examination of spectral lidar ratio variability of

93    transported biomass burning aerosol under different environmental conditions and presents a

94    supplementary subject of the current study.

95    The paper is organized as follows. Section 2 describes the lidar system and provides the

96    main expressions used for the data analysis. Several strong dust episodes, in April 2015, are

97    analyzed in Section 3. In Section 4, the smoke episodes occurring from December 2015 to January

98    2016, are used to evaluate the variation of the smoke lidar ratio with relative humidity. The paper

99    is finalized with conclusion.

100

101    **2. Experimental setup and data analysis**

102    The observations were performed with LILAS multiwavelength Raman lidar during

103    SHADOW-2 campaign at Mbour, Senegal. Information related to the SHADOW-2 and

104    observation site is presented in Veselovskii et al. (2016). The LILAS is based on a tripled Nd:YAG

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

106     aperture of the receiving telescope is 400 mm. During the campaign, LILAS configuration

107     ($3\beta+2\alpha+1\delta$) allowed the measurement of three particle backscattering ($\beta_{355}$, $\beta_{532}$, $\beta_{1064}$), two

108     extinction coefficients ($\alpha_{355}$, $\alpha_{532}$) and depolarization ratio at 532 nm ($\delta_{532}$). To improve the

109     performance of the system at 532 nm the rotational Raman channel was used instead of the

110     vibrational one (Veselovskii et al, 2015). The measurements were performed at a 47 degrees angle

111     to horizon. The backscattering coefficients and depolarization ratios were calculated with a 7.5 m

112     range resolution (corresponding to 5.5 m vertical resolution), while range resolution of extinction

113     coefficient varied from 50 m (at 1000 m) to 125 m (at 7000 m). Particle extinction and

114     backscattering coefficients at 355 nm and 532 nm are calculated from elastic and Raman

115     backscatter signals, as described in Ansmann et al. (1992). An additional Raman reception channel

116     at 408 nm was setup for profiling the water vapor mixing ratio (WVMR) (Whiteman et al., 1992).

117        The particle depolarization ratio $\delta$, determined as a ratio of cross- and co-polarized

118     components of the particle backscattering coefficient, was calculated and calibrated the same way

119     as described in Freudenthaler et al. (2009). To further the analysis of complex aerosol mixtures,

120     containing dust (d) and smoke (s), we can write $\beta = \beta^d + \beta^s$ and $\alpha = \alpha^d + \alpha^s$. The depolarization

121     ratio of such a mixture is therefore:

122   
$$\delta = \frac{\left(\dfrac{\delta^d}{1+\delta^d}\right)\beta^d + \left(\dfrac{\delta^s}{1+\delta^s}\right)\beta^s}{\dfrac{\beta^d}{1+\delta^d} + \dfrac{\beta^s}{1+\delta^s}} \qquad\qquad (1)$$

123     Here $\delta^d$ and $\delta^s$ are the particle depolarization ratios of dust and smoke components respectively.

124        To characterize the spectral dependence of the extinction ($\alpha$) and backscattering ($\beta$)

125     coefficients, corresponding Ångström exponents are introduced as:

126   
$$A_\alpha = \frac{\ln\left(\dfrac{\alpha_{\lambda_1}}{\alpha_{\lambda_2}}\right)}{\ln\left(\dfrac{\lambda_2}{\lambda_1}\right)} \text{ and } A_\beta = \frac{\ln\left(\dfrac{\beta_{\lambda_1}}{\beta_{\lambda_2}}\right)}{\ln\left(\dfrac{\lambda_2}{\lambda_1}\right)} \qquad\qquad (2)$$

127     Where $\alpha_{\lambda_1}$, $\alpha_{\lambda_2}$, $\beta_{\lambda_1}$, $\beta_{\lambda_2}$ are the extinction and backscattering coefficients at wavelengths $\lambda_1$ and

128     $\lambda_2$. For the mixture of smoke and dust, the extinction Ångström exponent (EAE) can be calculated

129     from the ratio $\dfrac{\alpha_{\lambda_1}}{\alpha_{\lambda_2}}$ :

$$130 \quad \frac{\alpha_{\lambda_1}}{\alpha_{\lambda_2}} = \frac{\alpha_{\lambda_1}^d + \alpha_{\lambda_1}^s}{\alpha_{\lambda_2}^d + \alpha_{\lambda_2}^s} = \frac{\alpha_{\lambda_1}^d}{\alpha_{\lambda_2}^d} \frac{\left(1 + \frac{\alpha_{\lambda_1}^s}{\alpha_{\lambda_1}^d}\right)}{\left(1 + \frac{\alpha_{\lambda_2}^s}{\alpha_{\lambda_2}^d}\right)} = \frac{\alpha_{\lambda_1}^d}{\alpha_{\lambda_2}^d} \frac{\left(1 + \frac{\alpha_{\lambda_2}^s (\frac{\lambda_2}{\lambda_1})^{A_\alpha^s}}{\alpha_{\lambda_2}^d (\frac{\lambda_2}{\lambda_1})^{A_\alpha^d}}\right)}{\left(1 + \frac{\alpha_{\lambda_2}^s}{\alpha_{\lambda_2}^d}\right)} = \frac{\alpha_{\lambda_1}^d}{\alpha_{\lambda_2}^d} \frac{\left(1 + \frac{\alpha_{\lambda_2}^s}{\alpha_{\lambda_2}^d} \left(\frac{\lambda_2}{\lambda_1}\right)^{(A_\alpha^s - A_\alpha^d)}\right)}{\left(1 + \frac{\alpha_{\lambda_2}^s}{\alpha_{\lambda_2}^d}\right)} \quad (3)$$

131    Here $A_\alpha^d$ and $A_\alpha^s$ are the extinction Ångström exponents of dust and smoke. The Ångström

132    exponent of the mixture is obtained from (3):

$$133 \quad A_\alpha = \frac{\ln \frac{\alpha_{\lambda_1}}{\alpha_{\lambda_2}}}{\ln \frac{\lambda_2}{\lambda_1}} = A_\alpha^d + \frac{1}{\ln \frac{\lambda_2}{\lambda_1}} \ln \left[ \frac{\left(1 + \frac{\alpha_{\lambda_2}^s}{\alpha_{\lambda_2}^d} \left(\frac{\lambda_2}{\lambda_1}\right)^{(A_\alpha^s - A_\alpha^d)}\right)}{\left(1 + \frac{\alpha_{\lambda_2}^s}{\alpha_{\lambda_2}^d}\right)} \right] \quad (4)$$

134    The backscattering Ångström exponent (BAE) can be calculated in a similar way. And finally, the

135    lidar ratio of the aerosol mixture is calculated as:

$$136 \quad S = \frac{S^d \beta^d + S^s \beta^s}{\beta^d + \beta^s} = S^d + \frac{\beta^s}{\beta}(S^s - S^d) \quad (5)$$

137    where $S^d$ and $S^s$ are the lidar ratios of dust and smoke.

138

139    **3. Dust observations in March and April 2015**

140        The aerosol over West Africa presents strong seasonal variations. The spring is

141    characterized by strong dust emission, while, during winter season, intense forest fires occurring

142    in the equatorial regions emit smoke particles that are transported over Senegal. The SHADOW-

143    2 campaign included the following periods of measurements: 13 March – 25 April 2015, 8–25

144    December 2015 and 5-24 January 2016, so numerous dust and smoke episodes were observed. In

145    our analysis of lidar-derived aerosol properties, we considered also aerosol columnar properties

146    provided by AERONET (Holben et al. 1998) and aerosol profiles predicted by the Modern-Era

147    Retrospective analysis for Research and Applications, Version 2 (MERRA-2) aerosol reanalysis

148    (Gelaro et al., 2017; Randles et al., 2017). MERRA-2 is the first long-term global reanalysis to

149    assimilate space-based aerosol observations and include their radiative coupling with atmospheric

150    dynamics. MERRA-2 is driven by the Goddard Earth Observing System (GEOS) model version 5

151    that includes the Goddard Chemistry, Aerosol, Radiation and Transport (GOCART) module.

152    GOCART models the sources, sinks, and transformation of the following five aerosol species as

153    external mixtures: dust, organic carbon (OC), black carbon (BC), sulfates (SU) and sea salt (SS).

154    Dust and sea salt are represented by five non-interacting size bins, and have wind-speed dependent

155    emissions. The MERRA-2 reanalysis assimilates AOD observations from the twin Moderate

156    Resolution Imaging Spectroradiometer (MODIS) instruments, MODIS-Terra and MODIS-Aqua,

157    as well as the AERONET ground-based sun photometer network. In addition, the profiles of

158    meteorological variables (P, T, RH), provided by radio-sondes at the Dakar airport, located ~70

159    km from the M'bour site, were also available. The relative humidity (RH) profiles over the M'bour

160    site were calculated from the combination of lidar-derived WVMR and temperature profile from

161    radiosounding.

162         Fig.1 shows the aerosol optical depth at 532 nm ($AOD_{532}$) for March, April and December

163    2015 recalculated from AERONET AOD at 500 nm using 440-675 nm Ångström exponent. The

164    same figure shows the AODs for the five aerosol species used in MERRA-2 model, such as dust,

165    organic carbon (OC), black carbon (BC), sulfates (SU) and sea salt (SS). The optical depths

166    provided by MERRA-2 and AERONET are in a good agreement. Dust is the predominant aerosol

167    component for all three months with the highest values of AOD in April. The contribution of

168    organic carbon (the main component of the biomass burning products) is significant in December,

169    when the forest fire season starts in equatorial regions, though noticeable amount of OC is

170    predicted also for March and for the beginning of April. The contribution of BC and SU to the

171    total AOD is low: the sum of the corresponding AODs is below 0.1 for all three months.

172         The single scattering albedo (SSA) over the M'Bour site in 2015 provided by AERONET

173    at 440 and 675 nm is shown in Fig.2. The $SSA_{675}$ is above 0.97 for March – April period, but at

174    440 nm dust absorption is stronger and, in March, $SSA_{440}$ is about 0.9. However, in the middle of

175    April, $SSA_{440}$ increases up to 0.95, indicating that aerosol becomes less absorbing at shorter

176    wavelengths. We can thus expect that variation of SSA at 355 nm between April and March should

177    be even stronger. In our study we consider two groups of observation. The first group corresponds

178    to the beginning of April, when SSA at 440 nm was lower. The second group covers the second

179    half of April, when SSA at 440 nm increased. By analyzing these two groups we expect to reveal

180    the effect of aerosol absorption, on lidar-derived aerosol properties.

181

182    **3.1. Dust episode on 1 – 4 April 2015**

183         In the beginning of April the dust was transported by Continental trades (Harmattan) from

184    the northeastern/eastern drylands. For period 1 - 4 April, as follows from Fig.1b, the $AOD_{532}$ over

185    Dakar increased up to 1.0. Fig.3 shows spatio-temporal distributions of the aerosol backscattering

186    coefficient $\beta_{532}$, particle depolarization ratio $\delta_{532}$, and water vapor mixing ratio for the nights 1-2,

187    2-3 and 3-4 April 2015. Corresponding back-trajectories, shown in Fig.4, demonstrate that, on 1-

188    2 and 2-3 April, air masses at all heights arrive from the North-East, whereas on 3-4 April the air

189    masses above 2500 m are advected from the East. These air masses are characterized by higher

190    humidity and may contain biomass-burning products. During these three nights, depolarization

191    ratio and WVMR present some evolution. On 1-2 April $\delta_{532}$ exceeds 30% and does not change

192    significantly within the dust layer, even if some decrease is observed above 2000 m after 03:00

193    UTC. By 3-4 April the depolarization ratio above 2500 m decreases below 25%, simultaneously

194    with increase of the WVMR. During the dust episode, the relative humidity did not exceed 20%

195    on 1-3 April, but on 3-4 April it increased up to 40% above 2500 m.

196          Vertical profiles of dust particle properties such as aerosol extinction coefficients $\alpha_{355}$, $\alpha_{532}$,

197    particle depolarization ratio $\delta_{532}$ and lidar ratios $S_{355}$, $S_{532}$ are shown on Fig.5 for the three

198    observation periods on 1, 2-3 and 3-4 April 2015. The corresponding extinction and backscatter

199    Ångström exponents, calculated for 355 and 532 nm wavelengths, are presented in Fig.6. During

200    all three observation periods $A_\alpha$ is slightly negative ($A_\alpha$ = -0.1±0.1) up to 2000 m. For the dust

201    component, MERRA-2 provides value of $A_\alpha$ =-0.14, which agrees with observations. Above 2000

202    m, $A_\alpha$ exhibits some increase, which is most significant on 3-4 April, when $A_\alpha$ reaches 0.3±0.1 at

203    4000 m height. Simultaneous decrease of $\delta_{532}$ indicates to the possible presence of smoke particles

204    above 2000 m. The backscatter Ångström exponent $A_\beta$, in contrast with $A_\alpha$, is sensitive to the

205    spectral dependence of the imaginary part of CRI, thus yielding complicated vertical variability of

206    $A_\beta$. In particular, on 2-3 April $A_\beta$ decreases from -0.5 to -0.7 within 1500–2500 m height range,

207    when $A_\alpha$ remains stable.

208          As follows from Fig.5, on 1 April the lidar ratio $S_{355}$=70±6 sr does not change with height,

209    while $S_{532}$ gradually decreases from 60±5 sr at 1000 m to 50±4 sr at 3000 m height. On sessions

210    that followed (Fig.5b,c) the lidar ratios at both 355 nm and 532 nm decreased. Thus, the range of

211    lidar ratios variation for the dust episode on 1-4 April is 60-70 sr at 355 nm and 45-60 sr at 532

212    nm. The lidar ratios ($S_{355}$ and $S_{532}$) modeled by MERRA-2 for the dust component are also shown

213    on Fig. 5. Corresponding values are of 70 sr and 42 sr respectively and do not vary with altitude

214    as the model optical properties of all dust size bins based on spectral complex refractive indices

215    from the Optical Properties of Aerosols and Clouds (OPAC) tables (Hess et al. 1998) and the

216    spheroidal shape models developed by Meng et al. (2010) are the same and fixed, as dust is treated

217    as homophobic. Modeled value $S_{355}$ is near the top of the range of observed values, while modeled

218    $S_{532}$ underestimates the observations.

219          The gradual decrease of $S_{532}$ with height in Fig.5a,c is however unusual. There are, at least,

220    two possible reasons to explain $S_{532}$ height variation. The first one can be the presence of non-dust

221    particles, for example, smoke. The second reason is that the properties (composition) of dust

222    change with height. If non-dust particles are present, the particle intensive properties, such as S, $\delta$

223    and $A_\alpha$ should vary with height in consistent way. The MERRA-2 modeling reported in Fig.1

224    shows that in the beginning of April the organic carbon is the second main contributor to the AOD,

225    after dust. We should recall, however, that the model can provide a realistic range of OC variation,

226    however not necessarily reproducing the exact spatio-temporal distribution of OC extinction

227    coefficient.

228        In the dust episode considered, the most significant smoke contribution was observed on

229    3-4 April. Fig.7a shows the profiles of measured $\alpha_{355}$ and $\alpha_{532}$ together with MERRA-2 modeled

230    extinction coefficients at 532 nm for five aerosol components. The extinction Ångström exponents

231    measured by lidar and modeled by MERRA-2 for dust component are given by Fig.7b. The same

232    figure shows also the lidar derived water vapor mixing ratio profile together with the relative

233    humidity. At low altitudes (below 2500 m), where aerosol is represented by pure dust, the

234    measured and modeled values of extinction coefficients are close. Above 2500 m the measured

235    value of $\alpha_{355}$ exceeds that of $\alpha_{532}$, indicating the presence of smoke particles, while modeled

236    contribution of OC to the total extinction is very low. The measured extinction Ångström exponent

237    is about -0.1 below 2000 m, which well agrees with modeling results for pure dust. Increase of

238    WVMR and RH above 2000 m coincides with growth of the $A_\alpha$. For the considered case, the

239    model reproduces correctly the dust loading, but underestimates the smoke contribution. At 3500

240    m, the difference between measured and modeled $\alpha_{532}$ is about 0.045 km$^{-1}$ which can be attributed

241    to the smoke contribution.

242        Dust and smoke particles contributions to the total backscattering coefficient can be also

243    separated on the basis of the depolarization measurements, assuming that depolarization ratios of

244    these particles are known (Tesche et al., 2009). The results of such decomposition are presented

245    in Fig.7c, assuming 35% and 7% for dust and smoke depolarization ratio, respectively. The

246    contribution of smoke to the total $\beta_{532}$ at 3500 m is 0.0009 km$^{-1}$sr$^{-1}$. For the smoke lidar ratio of 50

247    sr at 532 nm (validity of this choice will be discussed in section 3.3), the smoke extinction

248    coefficient is about 0.045 km$^{-1}$. This value agrees well with smoke contribution obtained from

249    Fig.7a at 3500 m and thus can be used for estimating the smoke effect on the intensive aerosols

250    properties derived from lidar measurements.

251        The depolarization ratio of the "dust-smoke" mixture, calculated with expression (1),

252    matches the observed value since decomposition in Fig.7c is based on depolarization

253    measurements. The Ångström exponent at 3500 m computed with (4) for $\alpha_{532}^s$ =0.045 km$^{-1}$, $\alpha_{532}^d$

254    =0.147 km$^{-1}$, $A_\alpha^d$ =-0.1 and $A_\alpha^s$ =1.0 yields $A_\alpha$ =0.2, which matches observed value 0.25±0.1.

255    Hence, the observed variation of $A_\alpha$ above 2000 m on 3-4 April is explained by smoke

256    contribution. In a similar way, using (5) we can estimate the smoke lidar ratio ($S_{532}^s$) that would

257 match the observed decrease of $S_{532}$. To explain decrease of the lidar ratio at 3500 m from 50 sr

258 to 45 sr, the smoke lidar ratio should be about 25 sr, which is unrealistically small. Such small

259 lidar ratio could be attributed to the maritime aerosol, but then the lidar ratios at both wavelengths

260 should decrease simultaneously. Recall that on 1-2 April smoke contribution was significantly

261 lower, while decrease of $S_{532}$ is about 10 sr. Thus, smoke particles presence cannot explain the

262 observed decrease of $S_{532}$ and it should be probably attributed to changes of dust composition (and

263 so the imaginary part) with height.

264       Smoke lidar ratio is usually assumed to be higher than that of dust (Burton et all., 2014),

265 meanwhile in Fig 5c the lidar ratio $S_{532}$ is not increased in presence of the smoke particles. It should

266 however be noticed that our results were obtained at low RH. The smoke particles are hygroscopic

267 and the lidar ratio should increase with RH. The way to characterize $S_{532}^{s}$ over Dakar site can be

268 based on the analysis of the lidar measurements during smoke episodes within height range where

269 smoke contribution becomes predominant. The results of such analysis will be discussed later in

270 section 3.3.

271

272       **3.2. Dust episodes on 14 and 24 April 2015**.

273       In the second part of April 2015, dust AOD$_{532}$ exceeded 1.0 (Fig.1b) and contributions of

274 other aerosol components were insignificant. Meanwhile, as follows from Fig.2, SSA$_{440}$ increased

275 after 15 April, thus dust became less absorbing in UV, which should influence the lidar-derived

276 aerosol intensive properties. Fig.8 shows extinction coefficients and lidar ratios at 355 nm and 532

277 nm, together with depolarization ratio $\delta_{532}$ and the Ångström exponents $A_{\alpha}$ and $A_{\beta}$ observed on

278 14 April (00:00 – 05:00 UTC) and 23-24 April (23:00-06:00 UTC). The first case is a "transition

279 day" when SSA$_{440}$ starts to increase. Extinction profiles presented in Fig.8a show that two dust

280 layers can be distinguished. In the first layer (below 2.5 km), aerosol intensive properties are

281 similar to that of 1-4 April with $S_{355}>S_{532}$, slightly negative $A_{\alpha}$ = -0.1 and $A_{\beta}$ as low as -0.35. In

282 the second layer $S_{355}$ and $S_{532}$ coincide and both $A_{\alpha}$ and $A_{\beta}$ are close to zero. The depolarization

283 ratio in the second layer is about 31%, slightly lower than in the first one. Thus, we can assume

284 that increase of the imaginary part in UV in the first layer is more significant, than in the second

285 one. From back-trajectories given in Fig.9, we can conclude that the air masses in the first layer

286 originate from the Northeastern/Eastern drylands, while in the second layer the air masses arrive

287 from the East. After 14 April, $S_{355}$ and $S_{532}$ coincided for the whole height range and results

288 obtained on 23-24 April (Fig.8 c, d) are the example of such observations. Back-trajectories show

289 that the air masses at both 2.0 and 3.0 km height are transported from East. The ratio $S_{355}/S_{532}$ is

290    close to 1.0 within the whole dust layer and both Ångström exponents $A_\alpha$, $A_\beta$ are close to zero.

291    Thus, the results from Fig.8, 9 are indicating that lidar-derived aerosol properties depend on the

292    dust source origin.

293

294    **3.3 Analysis of lidar ratio variations in March – April 2015**

295        Fig.10 summarizes the lidar ratio measurements for period from 29 March to 24 April 2015

296    (first phase of SHADOW ended on 25 April). Here we focus on the properties of the "pure dust",

297    thus do not show results before 29 March, when AOD was lower and the contribution of other

298    aerosol types could be significant (Fig.1). For the Fig.10 we have chosen height intervals, where

299    S value is stable and δ exceeds 30%. For example, on 14 and 24 April lidar ratios are averaged

300    inside 2.7-3.7 km and 2.0-4.0 km layers respectively. For the period considered, $S_{355}$ and $S_{532}$ vary

301    in the ranges 50 sr – 80 sr and 45 sr - 60 sr respectively with a mean values of 62 sr and 51 sr.

302    Enhanced variability of $S_{355}$ compared to $S_{532}$ can be explained by variation of the imaginary part

303    at 355 nm. At the beginning of the 29 March and 8 April dust episodes, $S_{355}/S_{532}$ ratio is as high as

304    1.5 and then gradually decreases. After 14 April, $S_{355}/S_{532}$ ratio becomes close to 1.0, thus S

305    presents no spectral dependence.

306        The day-to-day variation of aerosol column properties, including the spectrally dependent

307    complex refractive index, can be obtained from AERONET (Holben et al., 1998). Fig.11 shows

308    the imaginary part of the refractive index at 440 nm and 675 nm ($Im_{440}$, $Im_{675}$) provided by

309    AERONET for the same period of time as in Fig.10. The $Im_{440}$ strongly decreases after 14 April,

310    correlating with the decrease of $S_{355}/S_{532}$ ratio in Fig.10, which corroborates the suggestion, that

311    variations of $S_{355}/S_{532}$ ratio are related to variation of dust absorption in UV. The retrieved real

312    part (Re) of the complex refractive index oscillates around Re=1.45 and shows no significant

313    spectral dependence. Correlation between enhancement of $Im_{440}$, with in respect to $Im_{675}$, and

314    increase of lidar-derived $S_{355}/S_{532}$ is clearly seen in Fig.12, showing time – series of difference

315    $Im_{440}-Im_{675}$ and $S_{355}/S_{532}$ ratio.

316        To analyze the variations of observed lidar ratios and the Ångström exponents, a simplified

317    numerical simulation has been performed. For a realistic modeling of the dust lidar ratio, various

318    mixtures of different mineral components and particles shapes should be considered. Sensitivity

319    of the modeling results to the dust mixture parameters was demonstrated in study of Gasteiger et

320    al. (2011). Such detailed modeling, however, is out of the scope of the present paper. Here we only

321    intend to evaluate the main impact when the imaginary part of CRI is modified.

322        The lidar ratio depends not only on the complex refractive index but also on the dust

323    particle size distribution (PSD). The PSDs provided by AERONET on 2 and 23 April 2015 (three

324     distributions for each day) are shown in Fig.13. The PSDs are similar and the effective radii for

325     both days are about 0.75 µm, thus, difference in S observed for 2 and 23 April should be related

326     mainly to the complex refractive index. Fig.14a presents modeled $S_{355}$ and $S_{532}$ lidar ratios together

327     with the extinction and backscattering Ångström exponents $A_\alpha$, $A_\beta$ as a function of the imaginary

328     part. Computations were performed for the AERONET derived size distribution on 23 April from

329     Fig.13 using the assembly of randomly oriented spheroids (Dubovik et al., 2006) with the real part

330     Re=1.55. $S_{355}$ and $S_{532}$ increase with the imaginary part and the ratio $S_{355}/S_{532}$ is about 1.1.

331     Extinction coefficient is slightly sensitive to the imaginary part, thus increase of S in Fig.14 is due

332     to decrease of backscattering coefficient with Im. The modeled $A_\alpha$ is about $A_\alpha$=0.1, while $A_\beta$

333     decreases with Im to $A_\beta$=-0.2. To estimate the influence of a spectrally dependent imaginary part

334     $Im(\lambda)$ on $A_\beta$, we have also performed computations assuming a fixed $Im_{532}$=0.002 and only $Im_{355}$

335     is free to vary. Corresponding results are shown in Fig.14a with open stars. Spectral dependence

336     of the imaginary part significantly decreases $A_\beta$: for $Im_{355}$=0.005 ($Im_{355}$ − $Im_{532}$=0.003), $A_\beta$

337     decreases to -0.75.

338          We should recall however, that for the second half of April the observed ratio $S_{355}/S_{532}$,

339     was about 1.0, and both extinction and backscatter Ångström exponents were close to zero. To

340     figure out the kind of PSD that would reproduce those observations, we retrieved the PSD from

341     $3\beta+2\alpha$ measurements, as described in Veselovskii et al. (2002, 2010). For that purpose, data from

342     23-24 April (Fig.8), averaged within 2-3 km layer, were inverted and corresponding PSD is shown

343     in Fig.13 with red line. Inversion was performed for the assembly of randomly oriented spheroids,

344     in assumption of spectrally independent refractive index. Due to the limited number of input data

345     (five) we are able to reproduce only the main features of the PSD. The maximum of this lidar

346     derived PSD is shifted towards larger radii, with respect to the AERONET size distribution, but at

347     the same time, retrieved PSD contains significant contribution from the fine particles. The

348     simulation results for this lidar derived PSD, are given by Fig.14b. The lidar ratios $S_{355}$, $S_{532}$ for

349     all values of the imaginary part are close. The backscatter and extinction Ångström exponents are

350     close to zero, matching the observations of the second half of April 2015. Thus simulation results

351     demonstrate dependence on the PSD chosen, but in both cases these lead to the same conclusion:

352     observed low values of $A_\beta$ can not be reproduced without accounting for spectral dependence of

353     the imaginary part.

354          To compare computations and observations, information upon $Im_{355}$ and $Im_{532}$ values is

355     needed. The recently measured refractive indices of dust, sampled at different regions of Africa,

356     are presented by Di Biagio et al. (2019). In particular, for the countries located North and East of

357     Senegal, the imaginary parts at 370, 470, 520, 660 nm are of 0.0043, 0.0033, 0.0026, 0.0013 for

358    Mauritania and 0.0048, 0.0038, 0.0030, 0.0024 for Mali respectively. The highest values of lidar

359    ratios, observed in our measurements, are about 60 sr and 80 sr at 532 nm and 355 nm respectively.

360    Corresponding imaginary parts of CRI from Fig.14 can be estimated as $Im_{532}$=0.002-0.003 and

361    $Im_{355}$=0.005-0.006, which agrees with results presented by Di Biagio et al. (2019). Assuming

362    $Im_{355}$=0.005 and $Im_{532}$=0.002, the modeled ratio $S_{355}/S_{532}$ is about 1.44 and $A_\beta$ is about -0.75 for

363    both AERONET and lidar derived PSDs, which again reasonably agrees with observations. The

364    modeling performed is very simplified, still it confirms that observed values of $S_{355}/S_{532}$ ratio and

365    $A_\beta$ can be explained by the spectral dependence of the imaginary part of CRI.

366        Thus, based on our measurement results, two types of dust can be distinguished. The first

367    type has high $S_{355}/S_{532}$ ratio (up to 1.5). Such kind of dust is characterized by increase of the

368    imaginary part in UV and it was observed, for example, during 29 March and 10 April episodes.

369    For the second type, the ratio $S_{355}/S_{532} \approx 1.0$, so variation of the imaginary between 532 and 355 nm

370    wavelengths should be smaller than for the first type. Such dust was observed in the second half

371    of April 2015. Both types are characterized by high depolarization ratio, $\delta_{532}$, exceeding 30%, so

372    depolarization measurements at 532 nm are not capable to discriminate between these two types

373    of dust.

374        Difference in the observed dust properties is probably related to the mineralogical

375    characteristics in the source region. From the back-trajectories analysis presented in Figs. 4 and 9

376    one can suppose that the first type of dust was transported from the North–East, while the second

377    type from the East. In order to verify if a difference in the dust emission source region and transport

378    take place, we also analyzed the Infrared Difference Dust Index (IDDI) derived from the Meteosat

379    Second Generation (MSG) geostationary satellite imagery in thermal infrared (TIR). The IDDI is

380    developed by Legrand et al. (1985, 2001) originally for the Meteosat First Generation (MFG) and

381    is based on impact of airborne mineral dust on TIR radiation emitted by terrestrial surface. The

382    physical principle of the IDDI derivation is in thermal contrast between terrestrial surface and

383    atmosphere and the best sensitivity is found at around noon time when the surface temperature is

384    maximal (Legrand et al., 1988). The IDDI product shows that brightness temperature of terrestrial

385    surface observed by satellite can be reduced up to about 50°K in presence of airborne mineral dust,

386    while reduction by about 10°K already indicates a major dust event (Legrand et al., 2001). A direct

387    relationship between the IDDI and aerosol optical thickness in solar spectrum and visibility was

388    also found (Legrand et al., 2001). It should be mentioned here that the IDDI was initially developed

389    for MFG and the absolute consistence with the IDDI values from MSG should be examined due

390    to differences in spatial and spectral resolutions between two sensors. However, the physical

391    principles used for the IDDI determination are the same and a direct application of the MFG IDDI

392    algorithm to MSG was found as possible. Moreover, tests showed that the absolute values of IDDI

393    for a coincident overlapping period of MFG and MSG are very close. Nevertheless, employment

394    of the IDDI from MSG is indeed applicable for the required in the current analysis purpose of

395    solely dust spatial patterns detection.

396          The IDDI calculations, applied to the MSG images taken during the field campaign, clearly

397    show a major dust event in northern and central Africa. The elevated IDDI values over Senegal

398    are also visible. The IDDI images show distinguishable changes in the emission sources and

399    transport features during the different phases of the observations. For instance, Fig. 15 shows that

400    the dust emissions during the first phase of the event are originated in south Algeria, Mauritania

401    and Mali (examples of images from 29 and 30 March 2015). Weeks later, spatial patterns of the

402    elevated IDDI are shifted to south and show source regions in south of Niger (Fig.15c, d). Of

403    course, attribution of emission sources mineralogy to aerosol spectral absorption is a complex task

404    (Alfaro., et al 2004; Lafon et al., 2006; Di Biagio et al., 2017, 2019) and it is difficult to point to a

405    specific source that could clearly explain the observed in this study change in the aerosol absorbing

406    properties. However, the IDDI images clearly suggest a change in the dust transport regime that is

407    consistent with the change in the dust optical properties.

408

409    **4.  Smoke episodes in December 2015 – January 2016**

410          During the SHADOW campaign, we had several strong smoke episodes in December 2015

411    – January 2016, when air mass transported the products of biomass burning from the areas of

412    intensive forest fires in equatorial region. The relative humidity in the advected smoke layers

413    varied from episode to episode, allowing evaluation of the RH influence on the smoke lidar ratios

414    $S_{355}$, $S_{532}$. The spatio-temporal evolution of the particle backscattering coefficient and

415    depolarization ratio at 532 nm, during the 14-15 December 2015 smoke episode, is given in Fig.16.

416    The same figure shows also the water vapor mixing ratio, a convenient tracer to identify wet air

417    mass arrived from the equatorial region. The smoke particles are usually contained in elevated

418    layers, being mixed with dust (Veselovskii et al., 2018). The height ranges where the smoke

419    particles are predominant can be identified by low depolarization ratio and enhanced WVMR. For

420    event considered, the smoke particles are predominant above 1500 m after midnight.

421          The vertical profiles of $\alpha_{355}$, $\alpha_{532}$, $S_{355}$, $S_{532}$, $A_\alpha$, $A_\beta$ together with the water vapor mixing

422    ratio and the relative humidity, for 15 December (04:00 – 06:00 UTC), are shown in Fig.17. The

423    same figure presents decomposition of $\beta_{532}$ to the dust and smoke contributions, based on

424    depolarization measurements (Tesche et al., 2011). The smoke episodes are characterized by

425    different relative humidity within the elevated layer. On 15 December, RH is about 40% in the

426    1500 – 2100 m range and the ratio $\frac{\beta_{532}^s}{\beta_{532}}$ is about of 0.57 at 2000 m. The lidar ratio $S_{532}$ decreases

457    now emitted as a new BrC tracer species that uses $Im_{532}$=0.009 and $Im_{355}$=0.048 values (Hammer

458    et al. 2016). Thus, the spectral behavior of the imaginary part of organic carbon refractive index

459    depends on contribution of the BrC fraction to the primary organic carbon and on the physical-

460    chemical processes in the smoke layer during its transportation. As a result, the spectral

461    dependence of Im can present strong variations. In our study, the computations at 355 nm were

462    performed for four values of the imaginary part of dry particles $Im_{355}$=0.048, 0.03, 0.02, 0.01. At

463    532 nm two values $Im_{532}$=0.005 and 0.009 were considered. The parameters of the dry particle size

464    distribution, the real part of CRI and the hygroscopic growth factor used in computations are given

465    in Veselovskii et al. (2018). The particles are assumed to be homogeneous spheres and an increase

466    of the volume for every RH value (calculated from the growth factor) occurs due to water uptake.

467    Thus both the real and the imaginary part of CRI depend on RH.

468         The results of the simulations, shown in Fig.18, demonstrate strong dependence of the

469    organic carbon lidar ratio on the imaginary part of dry particles and on the relative humidity. For

470    $Im_{355}$=0.048, for all RH, $S_{355}$ is above 95 sr, which strongly exceeds the observed values. For lower

471    $Im_{355}$ the $S_{355}$ (RH) dependence is more pronounced and for $Im_{355}$ within the range 0.01-0.02,

472    computed $S_{355}$ are close to observed values. Computed $S_{532}$ values at low RH exceed the measured

473    ones, but for RH>70% agreement between measurements and GEOS assumed optical properties

474    for OC becomes reasonable.

475         The ratio $S_{355}/S_{532}$ for organic carbon, the same as for dust, is strongly influenced by the

476    spectral dependence of the imaginary part of CRI, hence it can be used as an indicator of Im

477    enhancement in UV. The ratios $S_{355}/S_{532}$, calculated from the results of modeling in Fig.18 for four

478    values of $Im_{355}$ (0.048, 0.03. 0.02, 0.01) and $Im_{532}$= 0.009, are shown in Fig.19. The ratio $S_{355}/S_{532}$,

479    corresponding to $Im_{355}$=0.01, is about 1.1 in the whole range of RH. However for the imaginary

480    part of dry particles $Im_{355}$=0.02 and 0.03 the ratio $S_{355}/S_{532}$ increases up to approximately 1.2 and

481    1.3 respectively for RH in 40%-70% range. Thus, enhanced $Im_{355}$ of dry OC particles should

482    provide increase of $S_{355}/S_{532}$ ratio even high RH and this is how it can be revealed. The measured

483    values of $S_{355}/S_{532}$ are shown on the same figure. As mentioned, observation at 532 nm on 15

484    December (RH=42%) "drops out" of other sessions. For the rest of observations the ratio $S_{355}/S_{532}$

485    is in 1.2 – 1.3 range, and from modeling in Fig.19 the imaginary part of the dry particles at 355

486    nm is estimated to be in 0.02-0.03 range.

487

488    **5. Summary and conclusion**

489         Our study shows the impact of aerosol spectral absorption variation on the lidar-derived

490    aerosol properties. In contrast to extinction, the backscattering coefficient, and so the lidar ratio,

491    are sensitive to the imaginary part of CRI. Hence, $S_{355}/S_{532}$ ratio can be an indicator of the

492    imaginary refractive index enhancement in the UV. Measurements performed during the
493    SHADOW campaign, in dust conditions, show a correlation between the decrease of $Im_{440}$, derived
494    from AERONET observations, and the decrease of lidar-derived $S_{355}/S_{532}$ ratio. Namely, in the
495    second half of April 2015, $S_{355}/S_{532}$ decreased from 1.5 to 1.0, when $Im_{440}$ decreased from 0.0045
496    to 0.0025. Our numerical simulations confirm, that observed $S_{355}/S_{532}$ (ratio close to 1.5) and $A_\beta$
497    (value close to -0.75) can be due to spectral variation of the imaginary part, attributed to iron oxides
498    contained in dust particles. Thus, April 2015 observations suggest the presence of different dust
499    types, characterized by distinct spectral dependence of $Im(\lambda)$. The analysis of backward
500    trajectories and Infrared Difference Dust Index derived from MSG geostationary satellite confirms
501    different air mass and dust particles transport features in the beginning and at the end of April.
502    Hence, the observed variations of $S_{355}/S_{532}$ can be related to the source region mineralogy. During
503    the April, particle depolarization systematically exceeded 30%, therefore no discrimination
504    between different types of dust was possible.

505        The results presented in this study demonstrate also that, for the selected temporal interval,
506    the dust lidar ratios may present significant variation with height. Dust of different size and
507    mineralogical composition can have different deposition rate, hence, complex refractive index can
508    be height-dependent. For instance, on April $1^{st}$, the $S_{532}$ decreased with height from 60 sr to 50 sr
509    within 1000–3000 m range, while depolarization ratio exceeded 30%. The analysis of this episode
510    showed that variation of the lidar ratio is entirely attributed to variations of dust characteristics;
511    the smoke aerosol contribution was insignificant. The data also demonstrate that a seemingly
512    uniform dust layer may have quite a complex height variation. The results therefore suggest the
513    relevance of including a varying mineralogy in radiative and climatic modeling of desert dust
514    impacts.

515        During December – January, the dry season in western Africa, our observations allowed in
516    addition the analysis of biomass burning aerosol properties. These particles are a product of the
517    seasonal forest fires and intensive agricultural waste combustion and can contain a substantial
518    amount of organic compounds, characterized by an enhanced imaginary part in UV (so called
519    BrC). For this aerosol type, the $Im(\lambda)$ dependence should increase the lidar ratio at 355 nm and
520    influence $S_{355}/S_{532}$. The smoke particles can be also hydrophilic and the lidar ratio can therefore
521    exhibit a strong dependence on RH. The numerical simulations performed for organic carbon,
522    which is the main component of smoke in GEOS model, demonstrated that $S_{355}/S_{532}$ is close to 1.0
523    in the absence of spectral variation of the imaginary part; this ratio, however, can be as high as 1.8
524    for dry particles with $Im_{532}=0.009$ and $Im_{355}=0.048$. This $S_{355}/S_{532}$ ratio decreases with RH,
525    however even for high humidity it depends on the $Im_{355}$ value for dry particles. In particular, for
526    $Im_{355}=0.02$ and 0.03 the ratio $S_{355}/S_{532}$ is about 1.2 and 1.3 respectively for RH in 40%-70% range.

527    Thus, observed $S_{355}/S_{532}$ values, exceeding 1.0, could corroborate the enhancement of imaginary

528    refractive index for smoke in UV.

529        Several strong smoke episodes were observed during the SHADOW campaign. While we

530    were able to evaluate the RH profiles, the dependence of the smoke lidar ratio with RH has been

531    estimated. The results obtained should be taken as semi-qualitative only, due to possible variation

532    of smoke particles composition from episode to episode and due to the presence of dust particles.

533    Nevertheless, the results clearly demonstrate an increase of $S_{532}$ from 44±5 sr to 66±7 sr and of

534    $S_{355}$ from 62±6 sr to 80±8 sr, when the RH increased from 25% to 85%. The measured $S_{355}/S_{532}$

535    ratio varied mainly within the range 1.2 - 1.3, so comparison with modeling for OC provides the

536    estimate of $Im_{355}$ of dry smoke particles in 0.02-0.03 range.

537        We would like to conclude that the multi-wavelengths Raman and depolarization lidar

538    measurements in western Africa enabled quite unique and comprehensive profiling of dust and

539    smoke spectral absorption properties. The results demonstrated a high variability of the lidar ratio

540    and the presence of its spectral dependence. Our study is one of the first attempts to track aerosol

541    composition variability using lidar measurements and to understand the mechanism underlying the

542    observed variations.

543

553

554

Atmospheric
Chemistry
and Physics
Discussions

555
556 Table 1. Lidar ratios $S_{355}$, $S_{532}$ for five smoke episodes in December 2015 – January 2016 and
557 corresponding the relative humidity RH. The table provides also the height and temporal interval

558 of observations. The contribution of the smoke particles to the total backscattering $\dfrac{\beta^s_{532}}{\beta_{532}}$ is derived

559 from depolarization measurements.
560

| Date | Height, m | Time, UTC | $\dfrac{\beta^s_{532}}{\beta_{532}}$ | RH, % | $S_{355}$, sr | $S_{532}$, sr |
|---|---|---|---|---|---|---|
| 15 Dec | 2000 | 04:00-06:00 | 0.57 | 42±8 | 67±7 | 44±5 |
| 15 Dec | 1850 | 19:20-20:30 | 0.57 | 25±6 | 62±6 | 50±5 |
| 23 Dec | 2250 | 05:00-07:00 | 0.65 | 65±13 | 76±8 | 56±6 |
| 24 Dec | 3200 | 19:00-23:00 | 0.66 | 75±14 | 76±8 | 62±6 |
| 20 Jan | 4500 | 01:00-07:00 | 0.8 | 85±15 | 80±8 | 66±7 |

561
562

# References

Amiridis, V., Balis, D. S., Kazadzis, S., Bais, A., Giannakaki, E., Papayannis, A., and Zerefos, C.:
Four-year aerosol observations with a Raman lidar at Thessaloniki, Greece, in the framework
of European Aerosol Research Lidar Network (EARLINET), J. Geophys. Res., 110, D21203,
doi:10.1029/2005JD006190, 2005.

Alfaro, S. C., Lafon, S., Rajot, J. L., Formenti, P., Gaudichet, A., and Maille, M.: Iron oxides and
light absorption by pure desert dust: An experimental study, J. Geophys. Res., 109, D08208,
doi:08210.01029/02003JD004374, 2004.

Ansmann, A., Wandinger, U., Riebesell, M., Weitkamp, C.and Michaelis,W., "Independent
measurement of extinction and backscatter profiles in cirrus clouds by using a combined Raman
elastic-backscatter lidar", Appl. Opt. 31, 7113–7131, 1992.

Ansmann, A., Petzold, A., Kandler, K., Tegen, I., Wendisch, M., Müller, D., Weinzierl, B., Müller,
T., Heintzenberg, J.: Saharan Mineral Dust Experiments SAMUM–1 and SAMUM–2: what
have we learned?, Tellus, 63B, 403–429, 2011.

Burton, S. P., Vaughan, M. A., Ferrare, R. A. and Hostetler, C. A.: Separating mixtures of aerosol
types in airborne High Spectral Resolution Lidar data. Atmos. Meas. Tech., 7, 419–436, 2014.

Di Biagio, C., Formenti, P., Balkanski, Y., Caponi, L., Cazaunau, M., Pangui, E., Journet, E.,
Nowak, S., Caquineau, S., Andreae, M. O., Kandler, K., Saeed, T., Piketh, S., Seibert, D.,
Williams, E., and Doussin, J.-F.: Global scale  variability of the mineral dust long-wave
refractive index: a new dataset of in situ measurements for climate modeling and remote
sensing, Atmos. Chem. Phys., 17, 1901-1929, https://doi.org/10.5194/acp-17-1901-2017,
2017.

Di Biagio, C., Formenti, P., Balkanski, Y., Caponi, L., Cazaunau, M., Pangui, E., Journet, E.,
Nowak, S., Andreae, M. O., Kandler, K., Saeed, T., Piketh, S., Seibert, D., Williams, E., and
Doussin, J. – F.: Complex refractive indices and single scattering albedo of global dust aerosols
in the shortwave spectrum and relationship to iron content and size, Atm. Chem. Phys., 19,
15503–15531, 2019.

Dubovik, O., Sinyuk, A., Lapyonok, T., Holben, B.N., Mishchenko, M., Yang, P.,  Eck, T.F.,
Volten, H.,  Munoz, O., Veihelmann, B., van der Zande, W.J.,  Leon, J.-F.,  Sorokin, M.,
Slutsker, I.: Application of spheroid models to account for aerosol particle nonsphericity in
remote sensing of desert dust, *J. Geophys. Res.*, 111, D11208, doi:10.1029/2005JD006619,
2006.

Esselborn, M., Wirth, M., Fix, A., Weinzierl, B., Rasp, K., Tesche, M., and Petzold, A.: Spatial
distribution and optical properties of Saharan dust observed by airborne high spectral resolution

597    lidar during SAMUM 2006, Tellus B, 61, 131–143, doi:10.1111/j.1600-0889.2008.00394.x,

598    2009.

599    Freudenthaler,V., Esselborn, M.,Wiegner, M.,Heese, B., Tesche, M. and co-authors:

600    Depolarization ratio profiling at severalwavelengths in pure Saharan dust during SAMUM

601    2006, *Tellus* 61B, 165–179, 2009.

602    Gasteiger, J., Wiegner, M., Groß, S., Freudenthaler, V., Toledano, C., Tesche, M., and Kandler,

603    K.: Modeling lidar-relevant optical properties of complex mineral dust aerosols, Tellus B,

604    63, 725-741, 2011.

605    Gelaro, R., McCarty, W., Suarez, M.J., Todling, R., Molod, A., Takacs, L., Randles, C.A.,

606    Darmenov, A., Bosilovich, M.G., Reichle, R., Wargan, K., Coy, L., Cullather, R., Draper,

607    C., Akella, S., Buchard, V., Conaty, A., Da Silva, A.M., Gu, W., Kim, G.K., Koster, R.,

608    Lucchesi, R., Merkova, D., Nielsen, J.E., Partyka, G., Pawson, S., Putman, W., Rienecker,

609    M., Schubert, S.D., Sienkiewicz, M., and Zhao, B.: The Modern-Era Retrospective Analysis

610    for Research and Applications, Version 2 (MERRA-2), Journal of Climate, 30, 5419-5454

611    2017.

612    Giannakaki, E., van Zyl, P. G., Müller, D., Balis, D., and Komppula, M.: Optical and

613    microphysical characterization of aerosol layers over South Africa by means of multi-

614    wavelength depolarization and Raman lidar measurements, Atmos. Chem. Phys., 16, 8109–

615    8123, 2016.

616    Groß, S., Tesche, M., Freudenthaler, V., Toledano, C., Wiegner, M., Ansmann, A., Althausen, D.,

617    and Seefeldner, M.: Characterization of Saharan dust, marine aerosols and mixtures of

618    biomassburning aerosols and dust by means of multi-wavelength depolarization and Raman

619    lidar measurements during SAMUM 2, Tellus B, 63, 706724, doi:10.1111/j.1600-

620    0889.2011.00556.x, 2011.

621    Haywood, J. M., Pelon, J., Formenti, P., Bharmal, N., Brooks, M., Capes, G., Chazette, P., Chou,

622    C., Christopher, S., Coe, H., Cuesta, J., Derimian, Y., Desboeufs, K., Greed, G., Harrison, M.,

623    Heese, B., Highwood, E. J., Johnson, B., Mallet, M., Marticorena, B., Marsham, J., Milton, S.,

624    Myhre, G., Osborne, S. R., Parker, D. J., Rajot, J. L., Schulz, M., Slingo, A., Tanre, D., and

625    Tulet, P.: Overview of the Dust and Biomass-burning Experiment and African Monsoon

626    Multidisciplinary Analysis Special Observing Period-0, Journal of Geophysical Research-

627    Atmospheres, 113, 10.1029/2008jd010077, 2008.

628    Hammer, M. S., Martin, R. V., van Donkelaar, A., Buchard, V., Torres, O., Ridley, D. A., and

629    Spurr, R. J. D.: Interpreting the ultraviolet aerosol index observed with the OMI satellite

630    instrument to understand absorption by organic aerosols: implications for atmospheric

631    oxidation and direct radiative effects, Atmos. Chem. Phys., 16, 2507–2523, 2016.

632    Hess, M., Koepke, P., and Schult, I.: Optical properties of aerosols and clouds: The software
633        package OPAC, Bulletin of the American Meteorological Society, 79, 831–844, 1998.

634    Hofer, J., Althausen, D, Abdullaev, S. F., Makhmudov, A. N., Nazarov, B. I., Schettler, G.,
635        Engelmann, R., Baars, H., Fomba, K. W., Müller, K., Heinold, B., Kandler, K., and Ansmann,
636        A.: Long-term profiling of mineral dust and pollution aerosol with multiwavelength
637        polarization Raman lidar at the Central Asian site of Dushanbe, Tajikistan: case studies, Atmos.
638        Chem. Phys., 17, 14559–14577, 2017.

639    Hofer, J., Ansmann, A., Althausen, D., Engelmann, R., Baars, H., Abdullaev, S.F., and
640        Makhmudov, A.N.: Long-term profiling of aerosol light-extinction, particle mass, cloud
641        condensation nuclei, and ice-nucleating particle concentration over Dushanbe, Tajikistan, in
642        Central Asia, Atm. Chem. Phys. Disc. doi.org/10.5194/acp-2019-963.

643    Holben, B.N., Eck, T.F., Slutsker, I., Tanre, D., Buis, J.P., Setzer, A., Vermote, E., Reagan, J.A.,
644        Kaufman, Y.J., Nakajima, T., Lavenu, F., Jankowiak, I., Smirnov, A.: AERONET- A federated
645        instrument network and data archive for aerosol characterization. Remote Sensing of
646        Environment 66, 1-16, 1998.

647    IPCC: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the
648        Fifth Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Stocker,
649        T. F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S. K., Boschung, J., Nauels, A., Xia, Y., Bex,
650        V., and Midgley, P. M., Cambridge University Press, Cambridge, UK and New York, NY,
651        USA, 1535 pp., https://doi.org/10.1017/CBO9781107415324, 2013.

652    Kandler, K., Lieke, K., Benker, N., Emmel, C., Küpper, M., Müller-Ebert,D., Ebert, M.,
653        Scheuvens, D., Schladitz, A., Schütz, L., Weinbruch, S.: Electron microscopy of particles
654        collected at Praia, Cape Verde, during the Saharan Mineral Dust Experiment: Particle
655        chemistry, shape, mixing state and complex refractive index. Tellus 63B, 475-496, 2011.

656    Kirchstetter, T. W., Novakov, T., and Hobbs, P. V.: Evidence that the spectral dependence of light
657        absorption by aerosols is affected by organic carbon, J. Geophys. Res.-Atmos., 109, D21208,
658        https://doi.org/10.1029/2004JD004999, 2004.

659    Lafon, S., Sokolik, I. N., Rajot, J. L., Caquineau, S., & Gaudichet, A.: Characterization of iron
660        oxides in mineral dust aerosols: Implications for light absorption. Journal of Geophysical
661        Research, 111(D21), 2006. https://doi.org/10.1029/2005JD007016

662    Legrand, M., Bertrand, J. J., Desbois, M., Menenger, L., and Fouquart, Y.: The potential of infrared
663        sattelite data for the retrieval of Saharan – dust optical depth over Africa, Journal of Applied
664        Meteorology, 28, 309-319, 1989.

665    Legrand, M., Plana-Fattori, A., and N'Doume, C.: Satellite detection of dust using the IR imagery
666         of Meteosat 1. Infrared difference dust index, Journal of Geophysical Research-Atmospheres,
667         106, 18251-18274, 2001.
668    Mamouri, R. E., Ansmann, A., Nisantzi, A., Kokkalis, P., Schwarz, A., and Hadjimitsis, D.: Low
669         Arabian dust extinction-to- backscatter ratio, Geophys. Res. Lett., 40, 4762–4766, 2013.
670    Mattis, I., Ansmann, A., Müller, D., Wandinger, U., and Althausen, D.: Dual-wavelength Raman
671         lidar observations of the extinction-to-backscatter ratio of Saharan dust, Geophys. Res. Lett.,
672         29, 1306, doi:10.1029/2002GL014721, 2002.
673    Meng, Z., Yang, P., Kattawar, G. W., Bi, L., Liou, K. N., Laszlo, I.: Single-scattering properties
674         of tri-axial ellipsoidal mineral dust aerosols: A database for application to radiative transfer
675         calculations, J. Aerosol Science 41, 501–512, 2010.
676    Mona, L., Amodeo, A., Pandolfi, M., and Pappalardo, G.: Saharan dust intrusions in the
677         Mediterranean area: three years of Raman lidar measurements, J. Geophys. Res., 111,
678         D16203, doi:10.1029/2005JD006569, 2006.
679    Nisantzi, A., Mamouri, R. E., Ansmann, A., Schuster, G. L., and Hadjimitsis, D. G.: Middle East
680         versus Saharan dust extinction-to-backscatter ratios, Atmos. Chem. Phys., 15, 7071–7084,
681         2015.
682    Papayannis, A., Amiridis, V., Mona, L., Tsaknakis, G., Balis, D., Bösenberg, J., Chaikovski, A.,
683         De Tomasi, F., Grigorov, I., Mattis, I., Mitev, V., Müller, D., Nickovic, S., Pérez, C.,
684         Pietruczuk, A., Pisani, G., Ravetta, F., Rizi, V., Sicard, M., Trickl, T., Wiegner, M., Gerding,
685         M., Mamouri, R. E., D'Amico, G., and Pappalardo, G.: Systematic lidar observations of
686         Saharan dust over Europe in the frame of EARLINET (2000–2002), J. Geophys. Res., 113,
687         D10204, doi:10.1029/2007JD009028, 2008.
688    Papayannis, A., Mamouri, R. E., Amiridis, V., Remoundaki, E., Tsaknakis, G., Kokkalis, P.,
689         Veselovskii, I., Kolgotin A., Nenes, A. and Fountoukis, C.: Optical-microphysical properties of
690         Saharan dust aerosols and composition relationship using a multi-wavelength Raman lidar, in
691         situ sensors and modelling: a case study analysis", Atmos. Chem. Phys. 12, 4011-4032 (2012).
692    Perrone, M. R., Barnaba, F., De Tomasi, F., Gobbi, G. P., and Tafuro, A. M.: Imaginary refractive-
693         index effects on desert-aerosol extinction versus backscatter relationships at 351 nm: numerical
694         computations and comparison with Raman lidar measurements, Appl. Opt., 43, 5531 – 5541,
695         2004.
696    Preißler, J., Wagner, F., Guerrero-Rascado, J. L., and Silva, A. M.: Two years of free-tropospheric
697         aerosol layers observed over Portugal by lidar, J. Geophys. Res.-Atmos., 118, 3676–3686,
698         doi:10.1002/jgrd.50350, 2013.

699  Randles, C.A., Da Silva, A.M., Buchard, V., Colarco, P.R., Darmenov, A., Govindaraju, R.,
700  Smirnov, A., Holben, B., Ferrare, R., Hair, J., Shinozuka, Y., and Flynn, J.: The MERRA-2
701  Aerosol Reanalysis, 1980 Onward. Part I: System Description and Data Assimilation
702  Evaluation, J. of Climate, 30, 6823-6850, 2017.

703  Rittmeister, F., Ansmann, A., Engelmann, R., Skupin, A., Baars, H., Kanitz, T., and Kinne, S.:
704  Profiling of Saharan dust from the Caribbean to western Africa – Part 1: Layering structures
705  and optical properties from shipborne polarization/Raman lidar observations, Atmos. Chem.
706  Phys., 17, 12963–12983, https://doi.org/10.5194/acp-17-12963-2017, 2017.

707  Sakai, T., Nagai, T., Nakazato, M., Mano, Y., and Matsumura, T.: Ice clouds and Asian dust
708  studied with lidar measurements of particle extinction-to-backscatter ratio, particle
709  depolarization, and water-vapor mixing ratio over Tsukuba, Appl. Optics, 42, 7103–7116,
710  2003.

711  Shin, S.-K., Tesche, M., Kim, K., Kezoudi, M., Tatarov, B., Müller. D., and Noh, Y.: On the
712  spectral depolarization and lidar ratio of mineral dust provided in the AERONET version 3
713  inversion product, Atm. Chem. Phys., 18, 12735–12746, 2018.

714  Sokolik, I. N., and Toon, O.B.: Incorporation of mineralogical composition into models of the
715  radiative properties of mineral aerosol from UV to IR wavelengths, J. Geophys. Res. 104, D8,
716  9423 – 9444, 1999.

717  Sun, H., Biedermann, L., and Bond, T. C.: Color of brown carbon: A model for ultraviolet and
718  visible light absorption by organic carbon aerosol, Geophys. Res. Lett., 34, L17813,
719  https://doi.org/10.1029/2007GL029797, 2007.

720  Tesche, M., Ansmann, A., Müller, D., Althausen, D., Mattis, I., Heese, B., Freudenthaler, V.,
721  Wiegner, M., Eseelborn, M., Pisani, G., and Knippertz, P.: Vertical profiling of Saharan dust
722  with Raman lidars and airborne HSRL in southern Morocco during SAMUM, Tellus B, 61,
723  144–164, 2009.

724  Tesche, M., Groß, S., Ansmann, A., Müller, D., Althausen, D., Freudenthaler, V., and Esselborn,
725  M.: Profiling of Saharan dust and biomass-burning smoke with multiwavelength polarization
726  Raman lidar at Cape Verde, Tellus B, 63, 649–676, doi:10.1111/j.1600-0889.2011.00548.x,
727  2011.

728  Torres, O., Tanskanen, A., Veihelmann, B., Ahn, C., Braak, R., Bhartia, P.K., Veefkind, P., and
729  Levelt, P.: Aerosols and surface UV products from Ozone Monitoring Instrument
730  observations: An overview, J. Geophys. Res., 112, D24S47, doi:10.1029/2007JD008809,
731  2007.

732    Veselovskii I., Kolgotin, A., Griaznov, V., Müller, D., Wandinger, U., Whiteman, D.: Inversion
733        with regularization for the retrieval of tropospheric aerosol parameters from multi-wavelength
734        lidar sounding, Appl.Opt. 41, 3685-3699, 2002.
735    Veselovskii I., O. Dubovik, A. Kolgotin, T. Lapyonok, P. Di Girolamo, D. Summa, D. N.
736        Whiteman, M. Mishchenko, and D. Tanré, 2010: Application Of Randomly Oriented
737        Spheroids For Retrieval Of Dust Particle Parameters From Multiwavelength Lidar
738        Measurements, J. Geophys. Res., **115**, D21203, doi:10.1029/2010JD014139, 2010.
739    Veselovskii, I., Whiteman, D. N., Korenskiy, M., Suvorina, A., Perez-Ramirez, D.: Use of
740        rotational Raman measurements in multiwavelength aerosol lidar for evaluation of particle
741        backscattering and extinction, Atmos. Meas. Tech., 8, 4111–4122, 2015.
742    Veselovskii, I., Goloub, P., Podvin, T., Bovchaliuk, V., Derimian, Y., Augustin, P., Fourmentin,
743        M., Tanre, D., Korenskiy, M., Whiteman, D., Diallo, A., Ndiaye, T., Kolgotin, A., Dubovik,
744        O.: Study of African dust with multi-wavelength Raman lidar during the "SHADOW"
745        campaign in Senegal, Atm. Chem. Phys. 16, 7013–7028, 2016.
746    Veselovskii, I., P. Goloub, T. Podvin, et al.: Characterization of smoke/dust episode over West
747        Africa: comparison of MERRA-2 modeling with multiwavelength Mie-Raman lidar
748        observations, Atm. Meas. Tech. 11, 949–969, 2018.
749    Wagner, R., Ajtai, T., Kandler, K., Lieke, K., Linke, C., Müller, T., Schnaiter, M., and Vragel, M.:
750        Complex refractive indices of Saharan dust samples at visible and near UV wavelengths: a
751        laboratory study, Atmos. Chem. Phys., 12, 2491–2512, 2012.
752    Whiteman, D., Melfi, S., Ferrare, R.: Raman lidar system for measurement of water vapor and
753        aerosols in the Earth's atmosphere", Appl. Opt. 31, 3068-3082, 1992.
754    Xie, C., Nishizawa, T., Sugimoto, N., Matsui, I., and Wang, Z.: Characteristics of aerosol optical
755    properties in pollution and Asian dust episodes over Beijing, China, Appl. Opt., 47, 4945 – 4951,
756    2008.
757

758
759
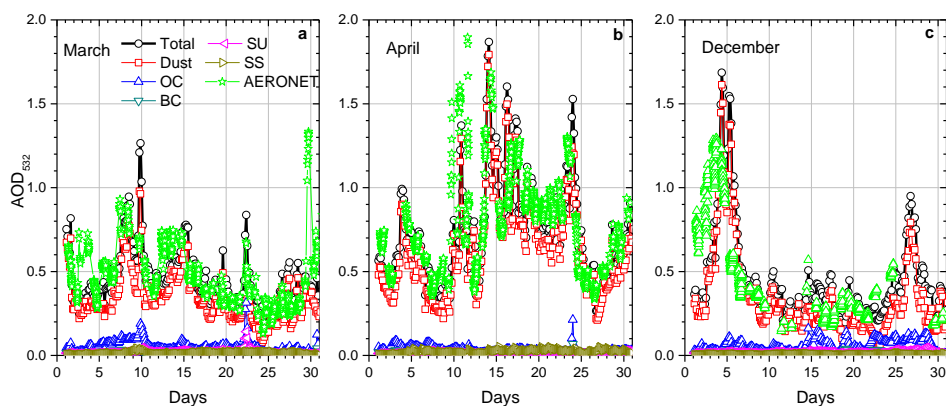


760    Fig.1. The aerosol optical depth (AOD) at 532 nm (open circles) and AODs of the main aerosol
761    components, such as dust, organic carbon (OC), black carbon (BC), sulfates (SU) and sea salt (SS)
762    provided by the MERRA-2 for (a) March, (b) April and (c) December 2015 over Mbour. Open
763    stars show $AOD_{532}$ provided by AERONET.
764
765
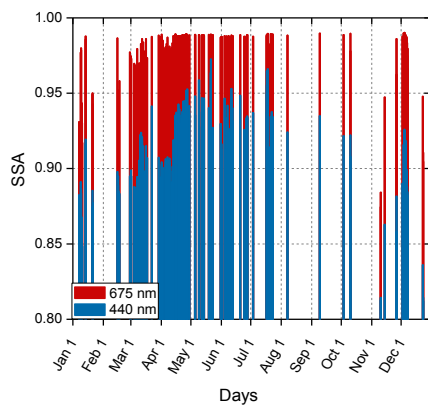


766
767    Fig.2. Aerosol single scattering albedo (SSA) at 675 nm and 440 nm provided by AERONET for
768    M'bour site in 2015.
769

770
771
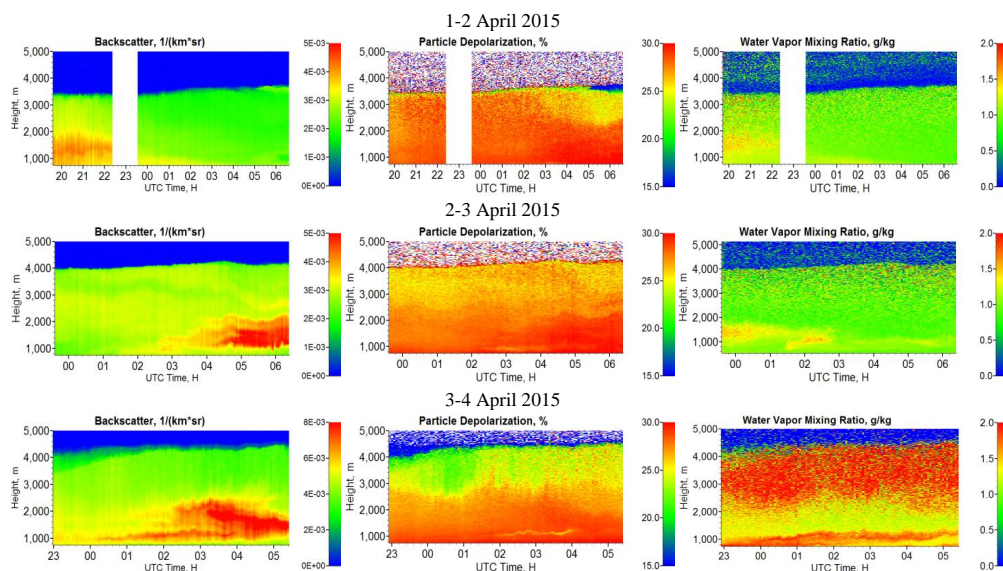


Fig.3. Tempo-spatial distributions of aerosol backscattering coefficient $\beta_{532}$ (left column), particle depolarization ratio $\delta_{532}$ (middle column) and water vapor mixing ratio (right column) for the nights 1-2 April (upper row), 2-3 April (middle row) and 3-4 April 2015 (bottom row).

775

776

777
778
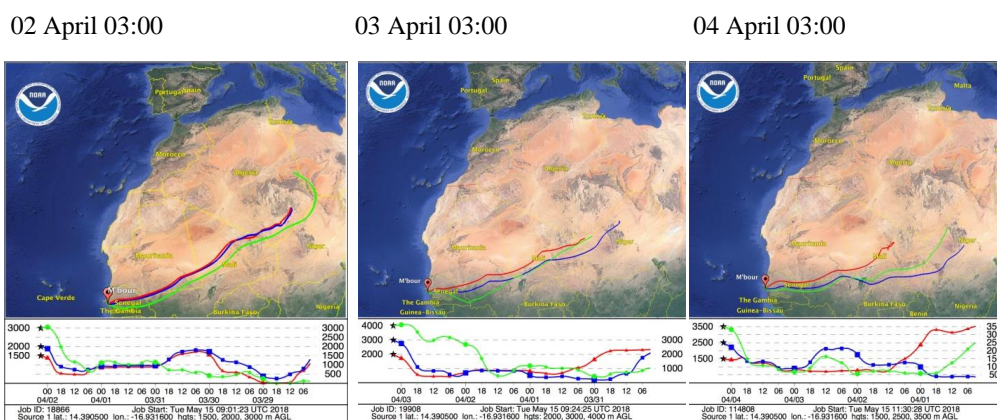


Fig.4. Three-day backward trajectories from the NOAA HYSPLIT model for the air mass in M'bour on 2, 3, 4 April 2015 at 03:00 UTC.

781
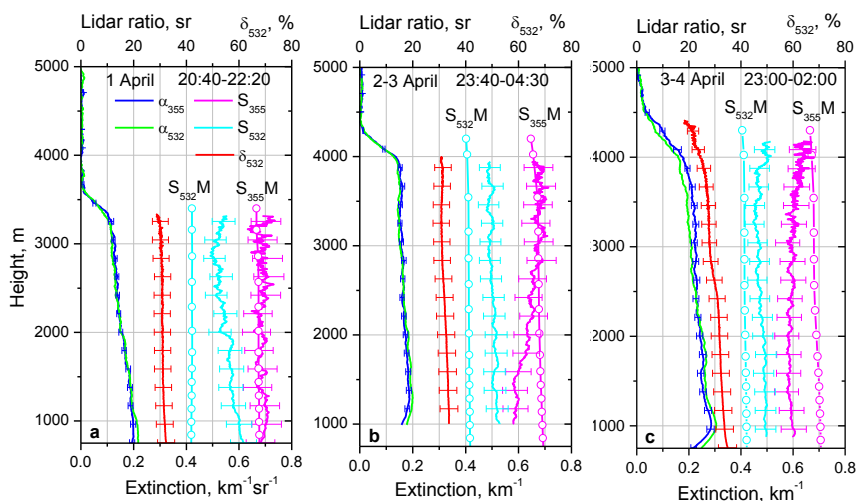
782 Fig.5. Vertical profiles of extinction coefficients ($\alpha_{355}$, $\alpha_{532}$) and lidar ratios ($S_{355}$, $S_{532}$) at 355 nm
783 and 532 nm together with particle depolarization ratio $\delta_{532}$ measured on 1 April (20:40-22:20
784 UTC), 2-3 April (23:40-04:30 UTC) and 3-4 April 2015 (23:00-02:00 UTC). Symbols show the
785 lidar ratios of dust provided by MERRA-2 model ($S_{355}M$, $S_{532}M$).

786
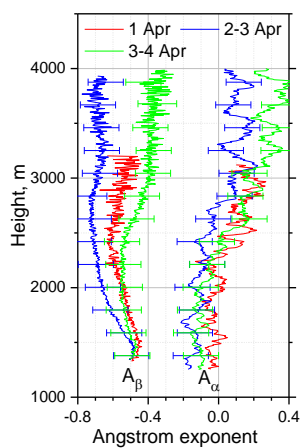


787

788 Fig.6. Vertical profiles of the extinction and backscattering Ångström exponents ($A_\alpha$ and $A_\beta$) at
789 355 – 532 nm for three temporal intervals from Fig.5.

790

791

27

792

793
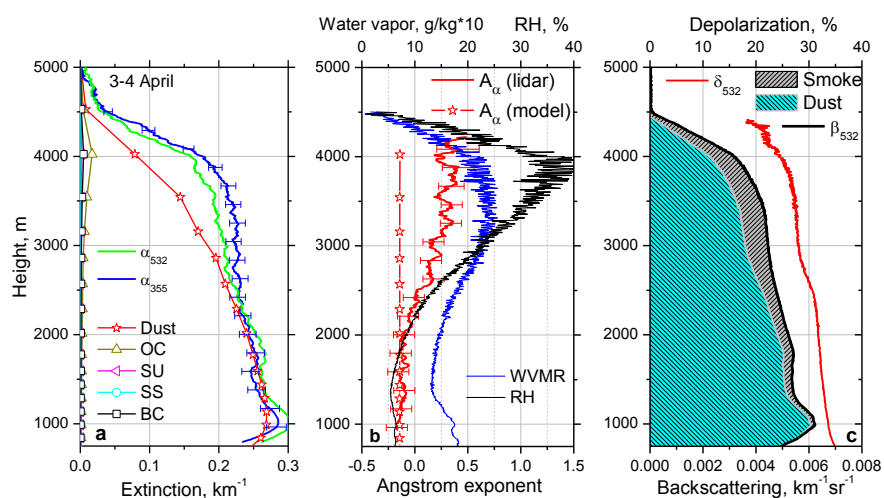


Fig.7. Vertical profiles of (a) extinction coefficients at 355 nm and 532 nm ($\alpha_{355}$, $\alpha_{532}$) measured by lidar (lines) and modeled by MERRA-2 (line+symbol) for five aerosol components at 532 nm; (b) extinction Ångström exponents at 355-532 nm obtained from lidar observations and modeled by MERRA-2 for pure dust (stars) together with water vapor mixing ratio (WVMR) and the relative humidity; (c) contribution of dust and smoke particles to $\beta_{532}$ together with particle depolarization ratio $\delta_{532}$. Values of WVMR are multiplied by factor 10. Lidar measurements were performed on 3-4 April 2015 for period 23:00 – 02:00 UTC. Modeling results are given for 4 April 00:00 UTC.

802

803

804

805

806
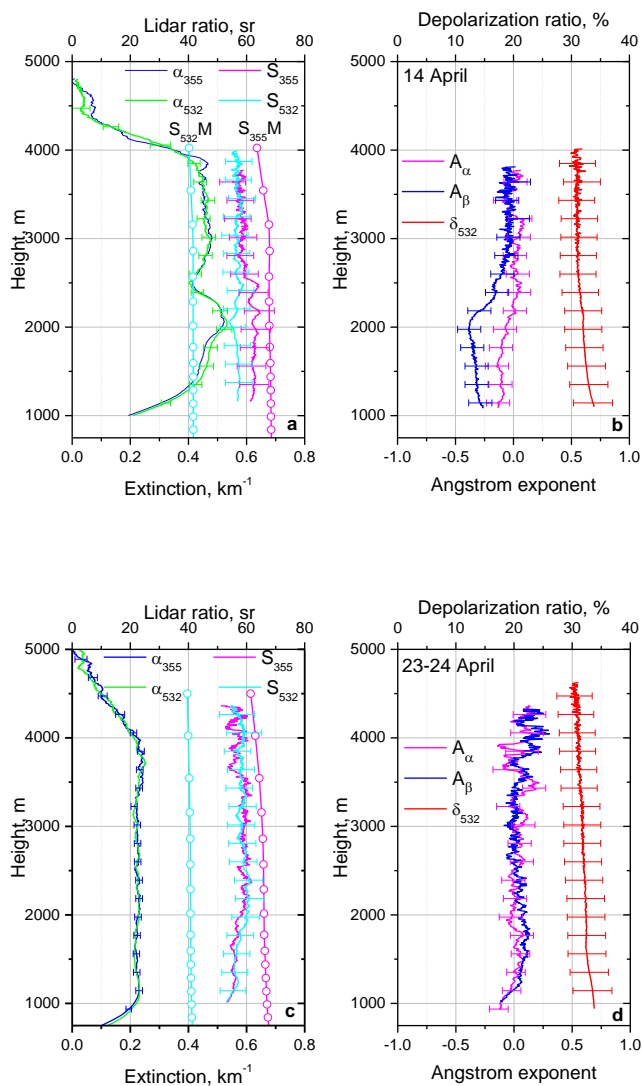


Fig.8. (a, c) Vertical profiles of extinction coefficients ($\alpha_{355}$, $\alpha_{532}$) and lidar ratios ($S_{355}$, $S_{532}$) at 355 nm and 532 nm; together with (b, d) particle depolarization ratio $\delta_{532}$, and extinction and backscattering Ångström exponents ($A_\alpha$, $A_\beta$) measured on (a, b) 14 April 2015 (00:00 – 05:00 UTC) and (c, d) the night 23-24 April (23:00-06:00 UTC). Open symbols on plots (a, c) show the lidar ratios $S_{355}M$ and $S_{532}M$ provided by MERRA-2 model on 14 and 14 April at 00:00 UTC.

812

813

814

815

816

| 14 April | 24 April |
|---|---|



817
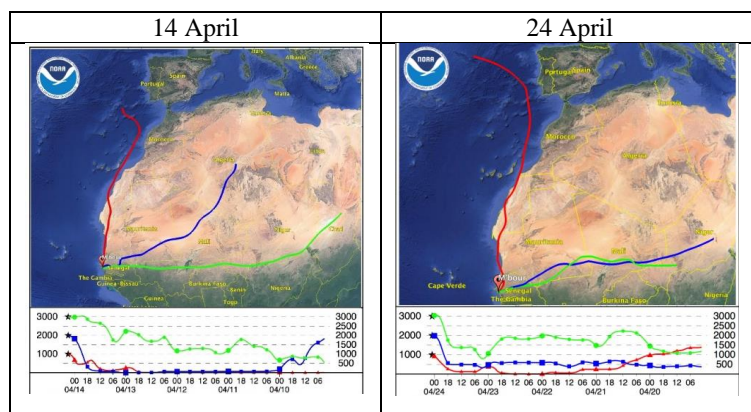
Fig.9. Four-days backward trajectories from the NOAA HYSPLIT model for 14 April (03:00
UTC) and 24 April (00:00 UTC) 2015.

820
821
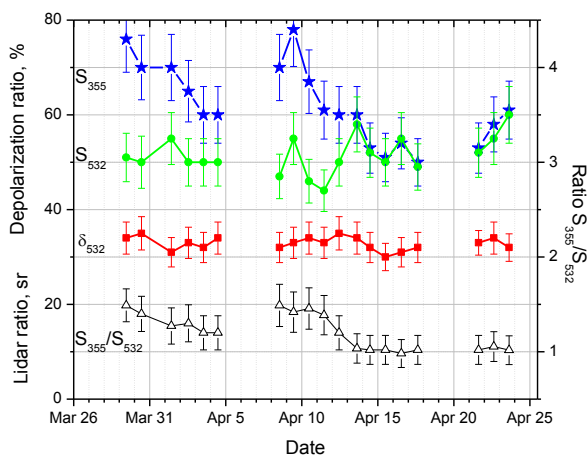
822
823    Fig.10. Lidar ratios $S_{355}$, $S_{532}$ and the particle depolarization ratio $\delta_{532}$ for dust episodes in March
824    - April 2015. Open triangles show the ratio $S_{355}/S_{532}$.
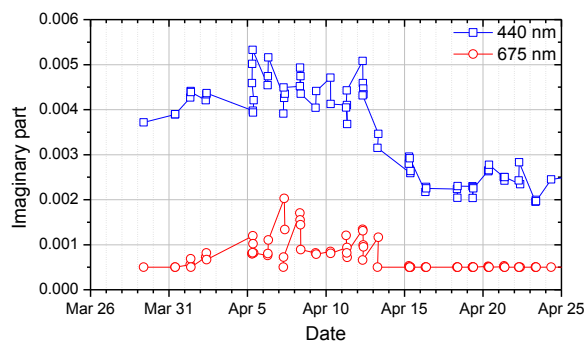


825
826    Fig.11. Imaginary part of the refractive index at 440 nm and 675 nm provided by AERONET in
827    March – April 2015

Atmospheric
Chemistry
and Physics
Discussions

828
829
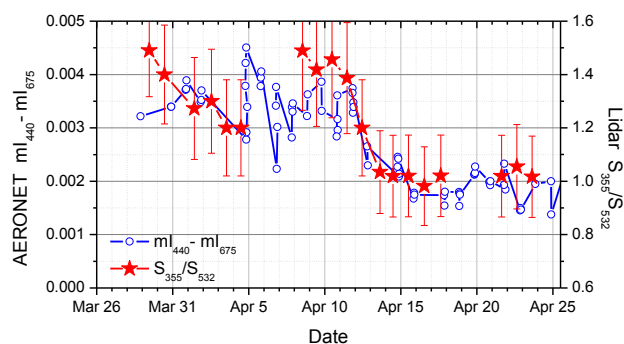


830
831  Fig.12. Difference $Im_{440} - Im_{675}$ from Fig.11 together with lidar measured values $S_{355}/S_{532}$ from
832  Fig.10 for days in April 2015.

833
834
835
836
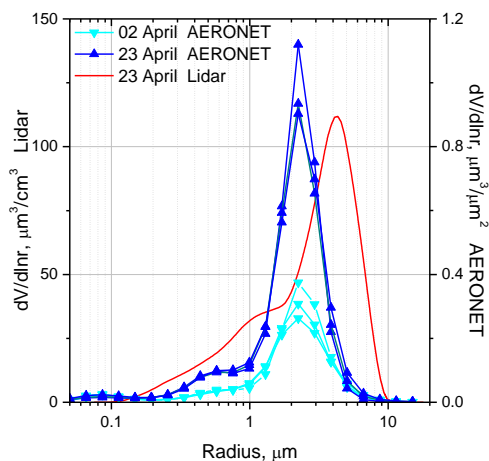


837
838  Fig.13. The particle size distributions provided by AERONET on 2 and 23 April 2015 (three PSDs
839  for each day). Red line shows the PSD derived from 3β+2α lidar measurements on 23-24 April.

840
841

842
843



844
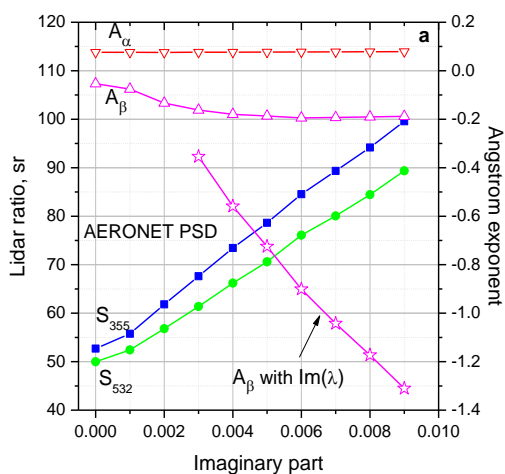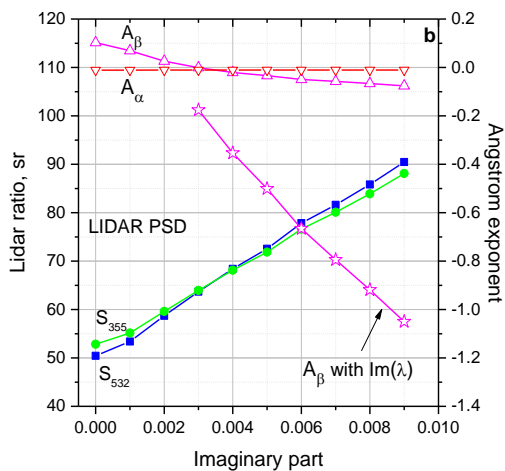


845
846
847    Fig.14. Lidar ratios $S_{355}$, $S_{532}$ together with the extinction and backscattering Ångström exponents
848    $A_\alpha$ and $A_\beta$ calculated for (a) AERONET PSD on 23 April from Fig.13 and (b) lidar derived PSD
849    from Fig.13 as a function of the imaginary part. Open stars show $A_\beta$ for spectrally dependent
850    imaginary part Im($\lambda$), assuming that $Im_{532}=0.002$ is fixed and only $Im_{355}$ is free vary. Computations
851    are performed for the assembly of randomly oriented spheroids with the real part Re=1.55.
852

33
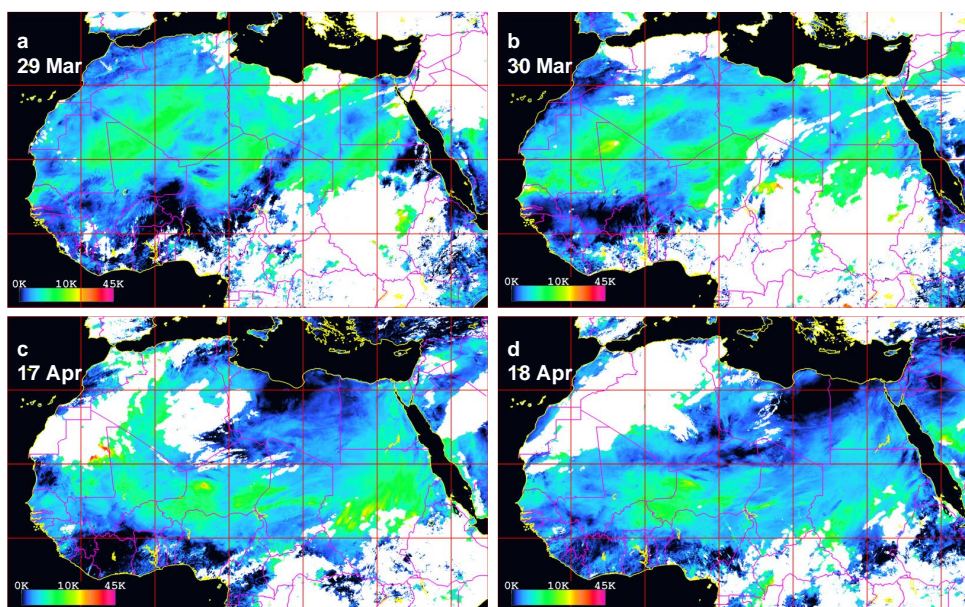
Fig.15. Infrared Difference Dust Index (IDDI) derived from MSG geostationary satellite at noon time. Panels (a), (b) show IDDI elevated values, representing airborne dust emission and transport, over central and northern Sahara on 29, 30 March 2015. The dust transport regime is visibly changed a few days later (17, 18 April 2015, panels (c), (d)); the elevated IDDI values are shifted to the south. The areas in white are cloud screened pixels; the IDDI is derived only over land due to the algorithm physical principle.
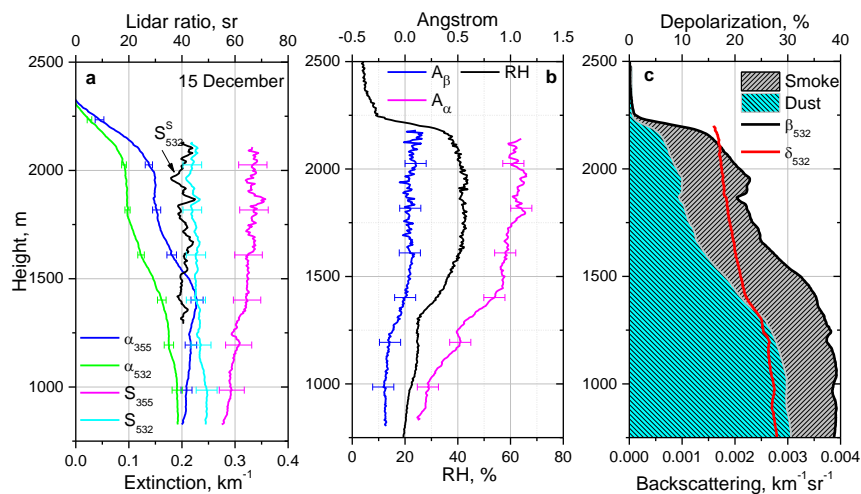
863
864
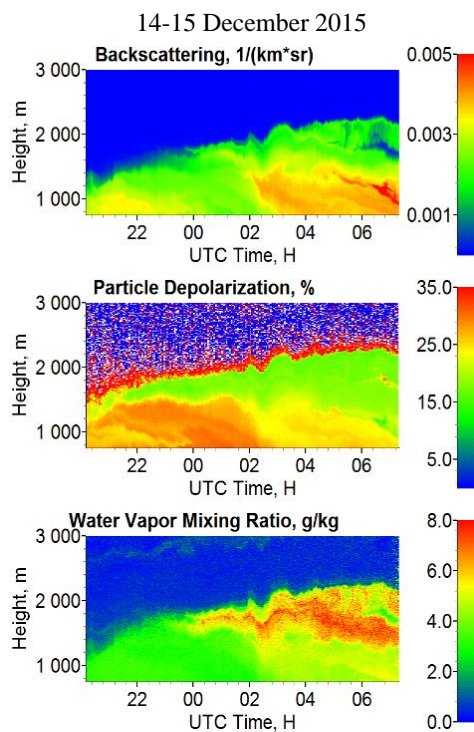


Fig.16. Tempo-spatial distributions of aerosol backscattering coefficient $\beta_{532}$, particle depolarization ratio $\delta_{532}$ and water vapor mixing ratio during smoke episode on the night 14-15 December 2015.

868
869

870



Fig.17. Vertical profiles of (a) extinction coefficients ($\alpha_{355}$, $\alpha_{532}$) and lidar ratios ($S_{355}$, $S_{532}$); (b) extinction, backscattering Ångström exponents ($A_\alpha$, $A_\beta$) at $355 - 532$ nm and relative humidity RH; (c) contribution of dust and smoke to $\beta_{532}$ together with particle depolarization ratio $\delta_{532}$ on 15 December (04:00 – 06:00 UTC). Black line in plot (a) shows the lidar ratio of smoke $S_{532}^s$ calculated from Eq.5.

876
877
878

Atmospheric
Chemistry
and Physics
Discussions
Open Access
EGU
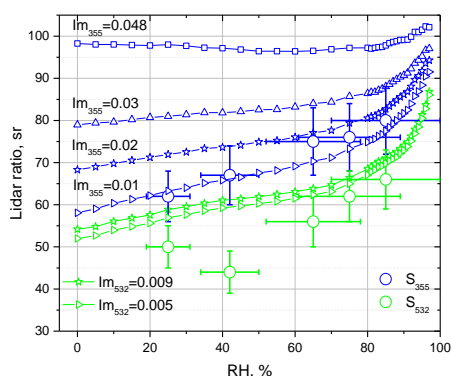
Fig.18. Modeled lidar ratios of organic carbon at 355 nm and 532 nm (line + symbol) as a function of the relative humidity for the particle parameters used in the MERRA-2 model. At 355 nm results are given for four values of the imaginary part of dry particles: $Im_{355}$= 0.048, 0.03. 0.02, 0.01. At 532 nm two values $Im_{532}$= 0.009 and 0.005 are considered. The scattered symbols (circles) show the lidar ratios ($S_{355}$, $S_{532}$) observed during five smoke episodes from Table 1.
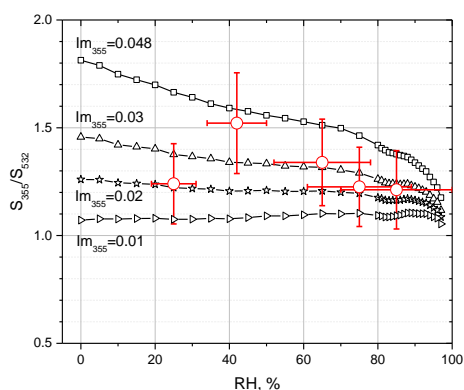


Fig.19. The ratio $S_{355}/S_{532}$ for organic carbon as a function of the relative humidity calculated from modeling results in Fig.18 for $Im_{532}$=0.009 and $Im_{355}$= 0.048, 0.03. 0.02, 0.01. The scattered symbols (circles) show the observed $S_{355}/S_{532}$ values.