Referee 1

1. “The paper needs only technical revision. See attached annotated file.”

We are very grateful to Referee 1 for careful reading of the manuscript and numerous improvements. Corresponding changes are introduced in the revised version of manuscript.

2. Fig.7. “a general comment is on the value of beta (1064 nm) below 2 km height. Why it is bigger than the one of 355 nm. There should be a process error, there!”

At low altitudes the dust backscattering coefficient at 355 nm can be lower than at 1064 nm, because of higher value of the imaginary part of RI.

3. “the word "backscattering" should be replaced by "backscatter" through out the whole paper.”

Expression “backscattering coefficient” is commonly used in lidar publications. So we would prefer to use it also.

Referee 2

First of all, we would like to thank the referee for careful reading of the manuscript. His comments helped us to improve the manuscript in the process of revision. Below we provide respond to his comments.

“Main question/comment: In the introduction the authors indicate that the aerosol retrievals that use only the multiwavelength lidar data can have significant uncertainties, especially in the refractive index and single scattering albedo, because the inversion from the 3 backscatter + 2 extinction inversion is underdetermined. They indicate that additional information should be used in the retrievals to improve the accuracy of the retrieved products and indicate that aerosol transport models can be one source of additional information by providing an initial guess for the inversion scheme. While this may be true, wouldn’t this then preclude the lidar+model retrieval results from being used to assess the accuracy of the models since the retrieval results would depend to some degree on the model parameters used as input? Perhaps a more pertinent question is how can the model parameterizations, optical properties, aerosol speciation, etc. be improved by the measurements and retrievals described here?”

This is an important question. In this work, we are using the lidar observations to assess the accuracy of the aerosol transport model independently. Our motivation for the work is to show that the aerosol transport model has sufficient skill to serve as additional constraint on the 3 backscatter + 2 extinction inversion. Thus, an inversion of lidar data using GEOS-5 model constraints would probably reduce the uncertainty of the retrievals. The second step will be assimilation of lidar measurements (not retrievals) in the aerosol transport model or even joint processing of lidar and model data. However, this task is very complicated, and we are not ready to move forward with it yet.

1. “Abstract, line 23. How close were measured and modeled extinctions?”

In the manuscript we provide the statistics for the deviation of modeled extinction from observations, but in Abstract we just mention that these are close, because for other episodes the numbers can be different.
2. “Abstract, line 28. What was the simulated lidar ratio?” It is 40 sr. To make it more clearly we modified the sentence: “…the simulated lidar ratio (about 40 sr) is lower than measurements (50±7 sr).”

3. Page 3, line 28. Note that the Buchard et al. (2017) validated MERRA-2 extinction profiles with independent airborne HSRL extinction profiles, as well as AOD and PM2.5. Mentioning of comparison with HSRL is added

4. “Page 4, line 8. Note that Buchard et al. (2017) and Randles et al. (2017) both used airborne HSRL data for model validation.” Mentioning of comparison with HSRL is added.

5. “Page 4, line 26. What wavelength does the Doppler lidar use?” It is 1543 nm. Added to the text.

6. “Page 5 (line 1). Are these nighttime only Raman lidar measurements? Page 13 mentions Raman lidar measurements of layer AOD so it’s not clear which measurements are nighttime only and which measurements are made both during day and night.” Continuous night and day time Raman measurements were performed only for selected episodes. In particular, such measurements are available for the smoke episode considered. However in day time, the Raman measurements at 532 nm were possible only up to 2-3 km height.

7. “Page 5 (line 9). Range or height resolution of extinction?” It is height resolution.

8. “Page 7 (line 28). The blue line is hard to see, but it looks like it also passes over the ocean extensively.” Yes, the most of the dust is concentrated below 1000 m (red line). At 1500 m (blue line) the concentration of dust is lower (masses are partly transported over the ocean). Corrections are introduced in the text.

9. “Figure 2. What parameter and units are shown in Figure 2? The color bar has no label or units.” These are arbitrary units. Added to the capture.

10. “Figure 5 does not add much to the paper and can be omitted.” By this figure we want to show that increase of the wind speed in LLJ is accompanied by increase of backscattering, so LLJ transports the dust. We would prefer to keep this figure.

11. “Figure 10 and discussion on page 10 (lines 21). The dust lidar ratios are 55 and 70 at 532 and 355 nm, respectively. There seems to be a significant wavelength difference in these ratios as compared to previous reports (see for example Müller et al., 2006, JGR) who reported $S_{355}/S_{532} = 1$. Any thoughts on this? Figure 12 shows there could be some smoke in this lower layer; perhaps this contributes to a larger wavelength dependence of the lidar ratio?” Yes, it is important point. In our measurements, backscattering of dust at 355 nm is lower than at 532 nm, though extinctions coincide. We think that the reason is the spectral dependence of the
imaginary part of RI of the dust. Observed behavior agrees with simulations, when typical spectral dependence of Im is considered. We aware, that the same tendency is observed in some recent measurements of other groups. But the effect depends on the dust origin. Measurements of Muller et al., 2006 didn’t report the spectral dependence of the lidar ratio, so the dust at SAMUM campaign had probably different origin.

12. “Page 12, line 19. What are the model lidar ratios for these various components? Are these values for the dry contributions of these components and how do these vary at higher RH? (Figure 6c shows RH~70-80% in upper layer).”

Lidar ratios of aerosol components (except dust) depended on the relative humidity (RH). The modeled water vapor profiles are close to the lidar measurements, especially in the elevated layer (fig.17), so the modeled RH profiles are also realistic. The growth factors given in Appendix allow to calculate the size distributions and RI of components for any RH. From these values the lidar ratios are obtained. We didn’t show lidar ratios for separate components, because these are height dependent. The values of modeled lidar ratios of organic carbon at the top of the elevated layer are given in Discussion section (71 sr and 66 sr at 355 and 532 nm respectively).

13. “Figure 16 shows pretty much the same information as figures 14 and 15 so I suggest keeping 16 and removing 14 and 15. “

Yes, we agree. The figures are removed.

14. “Page 15, line 6. If the model represented the depolarization closer to the value measured by the lidar (35% instead of 22%), would the model backscattering profiles at 532 nm be in better agreement with the lidar measurements in the dust layer?”

The model underestimates depolarization because it uses smooth particles (ellipsoids), while real dust particles have sharp faces. It is difficult to say how the presence of sharp faces will influence the backscattering (probably will increase). However, backscattering depends strongly also on the imaginary part of the refractive index. Actually for 355 nm agreement the model and measurements is good. So we think that by fine adjustment of the model parameters (fine particle contribution, mI(532)) better agreement with measurements can be obtained.

15. “Page 15, Inversion section. Just to be clear, the inversions discussed in this section did not use any model information as a first guess, is that correct?”

Yes, no model information is used.

16. “Page 16, line 23. In the dust layer (800-2000 m), the modeled effective radius (blue) seems to agree more with the spherical model (red) overall than the spheroidal model (black). Why? Likewise for volume concentration. This is contrary to what is discussed as a motivation for using the spheroidal model. Likewise for volume concentration.”

There are several reasons for such behavior. First of all, inside the height range 1500-2000 m the depolarization ratio is below 30%, so dust particles are mixed with biomass burning products. Thus the use of only spheroids in retrieval will slightly underestimate the effective radius and volume. Second, we don’t consider spectral dependence of mI in retrieval, which may underestimate the volume and radius up to ~20% (Veselovskii et al., 2016). Actually the volume concentration and effective radius obtained spheres and spheroids differ not so much (keeping in mind that uncertainty of retrieval is ~30%), the reason for this was discussed in (Veselovskii et
The use of spheroids is critical for retrieval of the complex refractive index (CRI), because the use of spheres, in particular, strongly underestimates the real part of CRI.

17. “Figure 23. The spheroidal profiles have error bars but the spherical profiles do not. Why? What are the uncertainties in the retrievals that use spherical aerosols?”

Errors bars are the same for spherical particles. We didn’t show these not to overcomplicate the figure.

18. “Figure 24. What are the uncertainties in the lidar retrievals shown in Figure 24?”

Error bars are added.

19. “Figures 23 and 24. The description indicates that the spherical model should be used for the smoke layer and the spheroidal model should be used for the dust layer inversions. What is the criterion (or criteria) for choosing when to use spherical vs. spheroidal? Particulate depolarization? If so, what value of particulate depolarization?”

In principle, the mixture of spheres and spheroids should be considered and the algorithm itself should find the spheroids volume fraction (SVF) for every height. In practices, however, retrieval of SVF at a moment is not stable enough. So, we presented retrievals for spheres and spheroids separately. Criterion for the model choice is the particle depolarization. From our experience for depolarization below 15% the spherical particles can be used.

20. “Page 17, line 28. This indicates that this Raman lidar provides profiles of particulate backscattering and extinction with uncertainties below 10%. Are these random uncertainties, systematic uncertainties, or combined uncertainties? Are the sub-10% uncertainties achieved with the 3 minute temporal, 50-125 m vertical resolutions discussed on page 5 or were different resolutions needed to get 10% uncertainties?”

The 10% is the total uncertainty for presented profiles. But to get it at least 30 minutes signal averaging is needed for the vertical resolution specified. The comment is added to the manuscript.
Referee 3

1. “General: The paper is appropriate for AMT and clearly worthwhile to be published. It brings together state-of-the-art lidar-derived aerosol products with state-of-the-art aerosol modeling. And the agreement between the model and observational results is very good. However, the paper is very long and should be shortened to make it more attractive. Furthermore, I would emphasis the synergy (complementarity) of lidar-derived optical and microphysical aerosol properties and the complementary model-derived product (chemical composition, mixing state, aerosol components), rather than highlight the good agreement. Sure, good agreement is needed, before one can focus on synergy. All this is given in the paper, just need to be re-arranged.”

We are grateful to the Referee 3 for praise of our results and for numerous important suggestions. We tried to follow his recommendations and shortened the manuscript. Five figures are removed.

2. “Abstract should be short: goals, methods used, strategy, main results. Not more. Just facts.”

Abstract is shortened

3. “P2, L8-13: The first sentence should be improved, better separation of the aerosol effects on radiation, clouds, environment, and we need better observations as a function of height to characterize aerosols in terms of optical properties, size distribution, chemical composition (aerosol mixture, components, etc): should be the message.”

It was changed. Now it is:

“Atmospheric aerosols are an important factor influencing the Earth’s radiative budget, though its impact is still highly uncertain due largely to the complicated mechanisms of aerosol – cloud interaction. Aerosol particles serve as cloud condensation nuclei and ice-nucleating particles, providing strong impact on cloud and precipitation formation. However different aerosol types differ significantly in their ability to initiate drop and ice crystal nucleation. There is thus a clear need for a better knowledge on vertically resolved optical, physical and chemical aerosol properties.”

4. “P3, L 5-11: In the discussion of inversion problems, one should not forget the shape parameterization of dust particles as a significant error source.”

Added: “The irregularity of the particles shape can be also a significant error source.”

5. “P3, L24: I do not believe that MERRA-2 can help to improve lidar backscatter modeling, when even optical models usually fail to properly simulate dust backscatter coefficients. I would not concentrate on backscatter. This is a quantity only lidar people are interested in. But sure, the lidar ratio is an important aerosol typing parameter.”

We agree that extinction profiles are more important for users. Still we think that considering of backscattering is also useful. For low aerosol extinction the uncertainty of lidar ratio calculation is high, while backscattering still can be derived and compared with the model. Actually we observed good agreement between measured and modeled dust backscattering coefficient (at least at 355 nm).
6. “P6, L5: At the end of section 2.1 one should briefly describe the inversion technique and the retrievable products (shown in Fig 23 and 24).”

Paragraph with brief description of inversion is added.

7. P6, L21: Visit the papers from Kemppinen et al., 2015 (ACP or AMT, JQSRT) and you will see how complicated the simulation of dust backscatter as retrieved from lidar really is.

Yes, Kemppinen et al. studies point to the importance of internal structure consideration. But such computations are extremely time consuming and can’t be done for big particles. So we have to use simplified ellipsoids/spheroids models at a moment.

Result section:

8. “First of all, I would like to make the following suggestions concerning figures. To my opinion one can reduce the number of figures significantly without losing information. Here are my suggestions:”

Five figures are removed.

“Figure 1 is ok.”

“Figure 2 is not needed. Figure 3 is sufficient so that Fig.2 can be removed.”

Fig.2 shows continuous monitoring of the dust and smoke layers for 4 days. We would like to show this long-term evolution, so would prefer to keep this figure.

“Figure 4 and 5 are not needed.”

Fig.4 and 5 show the wind measurements. We think that it is important to show, that the wind in the dust and in the smoke layers has different direction and that the low level jets appearance is accompanied by the increase of the dust backscattering. So we would prefer to keep these figures.

“Figure 6 is ok.”

“Figure 7 should be combined with Figure 8 to get a complete overview in just one figure.”

Done

“Or combine Figures 8 with 9, and leave out water vapor here. It is not need at this time, later it is shown. That is sufficient. Or even Figure 7, 8, and 9 could be combined to one overview figure.”

Fig.8 is removed

“Figure 10 is ok, but why are not all three time periods (in Figure 7) shown? The x-axis value ranges should be the same for all curves. It is confusing when the ranges are different and one wants to compare the different profiles.”

The scales are modified. Now these are the same. As mentioned in the text, the uncertainties of lidar ratio calculation below 2000 m for other two temporal intervals are two high (due to high gradients). So the results are not very informative and we didn’t show these.

“Figure 11 is fine.”

“Figure 12 and Figure 13 should be combined (to see the synergy we can have).”
Unfortunately the figure becomes overcomplicated, so we would prefer to keep these separate.

“Figure 13: To my opinion, the backscatter coefficients are not needed. The model uses given lidar ratios (40sr for dust, 80sr (?) for smoke) to convert extinction into backscatter: : : is my feeling.”

We think that backscattering coefficients are still useful. In the layers with low extinction coefficient the uncertainty of lidar ratio calculation is high, while backscattering coefficients still can be calculated and compared with model.

“I would remove Figures 14 and 15, because Figure 16 is sufficient. We should add 532nm solutions in Figure 16. Then this figure is very nice and convincing: : :. And shows the synergy of observations with models, together with Figure 13.”

We agree. The Fig.14,15 are removed and extinction at 532 nm is added.

“Figures 17 and 18 are not needed, to my opinion.”

We agree. Figures 17 and 18 are removed.

“Figure 19 is fine, brings some new results, and shows good agreement, a basic requirement for the next step: synergy!”

“Figure 20 can be removed. Who is interested in backscatter coefficient comparison?”

As discussed above, at low extinction coefficient the uncertainty of lidar ratio calculation is high, while backscattering coefficients still can be used. So we prefer to keep the figure.

‘Figure 21 nicely illustrates that the spheroidal model fails when the particles are nonspherical. But this is not discussed. Should be improved.”

Actually we discuss, that spheroids model fails to reproduce the depolarization of pure dust. But modeling of backscattering coefficient, in principle, can be improved. In particular, model reproduces backscattering at 355 nm quite well.

“Figure 22 shows the water vapor comparison. This is sufficient for the entire paper.”

“Is Table 1 needed?”

We think it is useful to keep all these values in one table.

9. “After rearranging the remaining figures one needs to update the discussion. To say it again, one should focus on the synergy aspect, i.e., that observations and the MERRA-2 model contribute in a complementary way. The synergy of observations and models is a key to improve the characterization of aerosols. However, in step 1, one has to demonstrate how well observations and models agree and describe the same aerosol scenario.”

We introduced changes in the discussion in revised manuscript.

10. “P10 L3-18: The Angstrom values for dust are not alwaysl negative, as suggested, when I am looking at all the field campaign data in the SAMUM and SALTRACE special issues. One should study the lidar papers in these special issues. Also, I miss again the discussion of the impact of the irregular shape of the dust particles as a significant error source.”

We have added comment, that backscattering Angstrom exponent strongly depends on the dust origin and no negative values of BAE were reported during SAMUM campaigns.
11. “P11, L25-26: There is a paper of Nisantzi et al. 2015 on the topic of depolarization increase by soil dust during fires”

Added to references

12. “P12, L5-9: I would not show a new period (for 3 UTC, Fig.13), not needed.”

We tried to show with this new period that lidar and model data are complimentary. Low depolarization ratio at ~2000 m height observed in the measurements can be understand from modeling results, pointing to the increase of organic carbon contribution at this height.

13. “P12, L22: The simulation of the backscattering coefficient is more challenging: : : As already mentioned, I would leave out the backscattering part. MERRA-2 cannot simulate backscatter, as so many other models, including all the optical models focusing on dust. They always fail, it is simply too complicated, see Kemppinen papers.”

Yes, it is challenging, still we think it is useful to show what model provides. And actually for 355 nm the model reproduces backscattering pretty well.

14. “P13, all the discussion should be shortened after re-arranging the figures starting from P13 and then on the following pages. Provide an attractive and short discussion. It is lengthy at the moment. What are the goals, should be the motivating question? And I believe, the demonstration of agreement (first step) is important but more important is the demonstration of synergy (second step)”.

After figures removal we introduced changes in the text and discussions, trying to follow reviewer recommendations.

15. “P16, L2: So both the lidar inversion method and the model are based on the same ‘wrong’ spheroidal model. So, no surprise when the lidar and model products agree here.”

Not exactly. Model provides the mass concentrations without considering any light scattering. So agreement actually demonstrates, that lidar inversion works not so bad.

16. “Conclusions, summary and abstract must be updated after finishing the updated result section.”

Updated

17. Congratulations to the nice results. The paper will be a significant contribution to the literature.

Thanks!
Characterization of smoke/dust episode over West Africa: comparison of MERRA-2 modeling with multiwavelength Mie-Raman lidar observations

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Abstract

Observations of multiwavelength Mie-Raman lidar taken during the SHADOW field campaign are used to analyze a smoke/dust episode over West Africa on 24-27 December 2015. For the case considered, the dust layer extended from the ground up to approximately 2000 m while the elevated smoke layer occurred in the 2500 m – 4000 m range. The profiles of lidar measured backscattering, extinction coefficients and depolarization ratios are compared with the vertical distribution of aerosol parameters provided by the Modern-Era Retrospective analysis for Research and Applications, Version 2 (MERRA-2). The MERRA-2 model simulated the correct location of the near-surface dust and elevated smoke layers. The value of modeled and observed aerosol extinction coefficients at both 355 nm and 532 nm are also rather close. The model predicts significant concentration of dust particles inside the elevated smoke layer, which is supported by an increased depolarization ratio of 15% observed in the center of this layer. The modeled at 355 nm the lidar ratio of 65 sr in the near-surface dust layer is close to the observed value (70±10) sr. At 532 nm however, the simulated lidar ratio (about 40 sr) is lower than measurements (55±8 sr). The results presented demonstrate that the lidar and model data are complimentary and the synergy of observations and models is a key to improve the aerosols characterization.
1. Introduction

Atmospheric aerosols are an important factor influencing the Earth’s radiative budget, though its impact is still highly uncertain due largely to the complicated mechanisms of aerosol – cloud interaction. Aerosol particles serve as cloud condensation nuclei and ice-nucleating particles, providing strong impact on cloud and precipitation formation. However, different aerosol types differ significantly in their ability to initiate drop and ice crystal nucleation. There is thus a clear need for a better knowledge on vertically resolved optical, physical and chemical aerosol properties. Lidar is a recognized instrument for vertical profiling of aerosol properties, and the possibility to invert lidar observations at several wavelengths to aerosol microphysical properties has been extensively studied both theoretically and experimentally over the two past decades (e.g. Müller et al., 1999; 2016; Veselovskii et al., 2002; Böckman et al, 2005). These studies revealed the importance of using Raman or HSRL (high spectral resolution lidar) systems, which allow independent measurements of aerosol extinction and backscattering coefficients to be made. At present, the most practical configuration of Raman (HSRL) lidar is based on a triple wavelength Nd:YAG laser. Such a lidar provides the so called $3\beta+2\alpha$ set of observations, including three backscattering (355 nm, 532 nm, 1064 nm) and two extinction (355 nm, 532 nm) coefficients.

However, the problem of inversion of $3\beta+2\alpha$ observations is underdetermined (Chemyakin et al., 2016; Alexandrov and Mishchenko 2017). As a result, instead of a unique solution, a family of solutions should be considered, leading to an increase in retrieval uncertainties. Still, the estimation of volume density ($V$) and effective radius ($r_{eff}$) with uncertainty below 30% is possible, especially when the fine mode in the particle size distribution (PSD) is predominant (e.g. Veselovskii et al., 2004; Müller et al., 2005; 2016; Pérez-Ramírez et al., 2013). The refractive index (RI) can be also estimated from the measurements, although the uncertainty of such estimation is significant: for the real part ($m_R$) of RI the uncertainty is normally about ±0.05 and for the imaginary part ($m_I$) it is about 50% when $m_I>0.01$ (Veselovskii et al., 2004; Müller et al., 2016). Proposed improvements of inversion schemes were considered in recent publications (Chemyakin et al., 2014; Kolgotin et al., 2016), still these improvements are not able to resolve the fundamental issue: the information content of $3\beta+2\alpha$ observations is insufficient to support exact solution of the problem and additional information should be used in
retrievals to improve the accuracy of the retrieved products (Veselovskii et al., 2005; Burton et al., 2016; Kahnert and Andersson, 2017; Alexandrov and Mishchenko 2017).

We should recall also that in the inversion schemes considered, the refractive index is normally assumed to be spectrally and size independent, which is generally not the case in the atmosphere. The irregularity of the particles shape can be also a significant error source. Moreover, the volume density and effective radius obtained from $3\beta+2\alpha$ observations are attributed to the whole size distribution, which is of limited practical use, because of the importance of characterizing the particle properties separately for the fine and coarse modes. Considering these issues makes the inverse problem even more underdetermined, emphasizing the need for additional input information.

One opportunity to get this additional information is by combining the lidar observations with aerosol transport models (Kahnert and Andersson, 2017). Models provide the vertical distribution of mass mixing ratios of chemical aerosol components, which can be used as “initial guess” in the inversion scheme. MERRA-2 offers a unique opportunity to provide such an “initial guesses” of the vertical structure of aerosol chemical composition. MERRA-2 is produced with NASA’s global Earth system model, GEOS-5 (Goddard Earth Observing System version 5) (Gelaro et al 2017) and includes an online coupling with the Goddard Chemistry, Aerosol, Radiation and Transport model (GOCART), which allows for assimilation of aerosol optical depth (AOD) from space borne and surface instruments such as MODIS, AVHRR, MISR, and AERONET (Randles et al. 2017). The fundamental data that MERRA-2 provides are vertical profiles of the mass mixing ratios of five aerosol components: dust, sea salt, black and organic carbon, and sulfate aerosols. The main optical parameters related to lidar measurements, such as aerosol extinction and backscattering coefficients can be calculated basing on these data. The principal question arising, however, is how well the reanalysis reproduces independent observations, and thus can provide a realistic initial guess for a lidar inversion scheme. Buchard et al. (2017) and Randles et al (2017) extensively validated MERRA-2 with independent surface and aircraft observations of particulate matter (PM2.5) and AOD, as well as space-based observations of absorption aerosol optical depth and aerosol index. The extinction profiles derived from airborne HSRL measurements were also compared with modeling, finding generally good agreement between the observations and MERRA-2.
For global validation of the aerosol vertical distribution, the modeled profiles of attenuated backscatter were compared to spaceborne Cloud–Aerosol Lidar with Orthogonal Polarization (CALIOP) observations (Winker et al., 2009), and a good consistency between simulations and observations was reported (Nowottnick et al., 2015, Buchard et al., 2017). Additional opportunities for model validation are provided by ground based multiwavelength Raman or HSRL systems. Such lidars by their nature have limited spatial coverage but are well suited for the characterization of the vertical distribution of particle properties at a chosen location.

In our paper, we consider Raman lidar observations taken during a smoke/dust episode over West Africa in December 2015 during the SHADOW campaign (Veselovskii et al., 2016), and compare the vertical profiles of particle parameters with MERRA-2. The simultaneous presence of dust and smoke layers in the atmosphere provides an opportunity to test the ability of the model to reproduce the vertical structure of aerosol properties over the observation site.

2. Measurement setup and data analysis

2.1 Observation site

The observation site is located at the Institute for Research and Development (IRD) Center, Mbour, Senegal (14°N, 17°W). Information about the SHADOW (study of SaHAran Dust Over West Africa) campaign and instruments at the IRD site can be found in the recent publication by Veselovskii et al. (2016). During the SHADOW campaign data from three lidar instruments were available:

- Cimel CE-370 micropulse lidar (www.cimel.fr) operated 24 hours per day at 532 nm allowing real-time monitoring of aerosol and cloud layers.
- Doppler lidar Windcube WLS 100 (www.leosphere.com) provided continuous monitoring of the wind field in the range from 100 m to 5 km with 50 m range resolution at 1543 nm wavelength.
- Multiwavelength Mie - Raman polarization lidar LILAS (LIlle Lidar AtmosphereS), allowed simultaneous detection of elastic and Raman backscatter signals and thus provides 3β+2α observations along with depolarization ratio at 532 nm.

LILAS measurements were performed from inside a laboratory building through a window at an angle of 47 deg with respect to the horizon. Acquiring Raman backscatter at 408
nm also permits profiling of the water vapor mixing ratio (WVMR) (Whiteman et al., 1992). For calibration of the water vapor channel, radiosonde launches from Dakar (about 70 km away from Mbour) were used. The large separation between the lidar and radiosonde locations prevented an accurate calibration, so the WVMR data were used mainly to monitor the relative change of the water vapor content. The temporal resolution of the measurements was approximately 3 minutes. The backscattering coefficients and depolarization ratio were calculated with range resolution 7.5 m (corresponding to a vertical spatial resolution of 5.5 m). The spatial resolution of the extinction coefficient measurements varied with height from 50 m (at 1000 m) to 125 m (at 7000 m).

The particle extinction ($\alpha_p$) and backscattering ($\beta_p$) coefficients at 355 nm and 532 nm are calculated from elastic and Raman backscatter signals, as described in Ansmann et al. (1992). Backscattering coefficients at 1064 nm ($\beta_{1064}$) were calculated by the Klett method (Klett, 1981).

In the data analysis both volume ($\delta_v$) and particle ($\delta_p$) depolarization ratios are considered. These ratios are defined as

$$\delta_v = \frac{\beta_p^v + \beta_m^v}{\beta_{\perp}^v + \beta_{\parallel}^v} = \frac{\alpha_p}{C P}$$

(1)

$$\delta_p = \frac{\beta_p^\perp}{\beta_{\parallel}^v}$$

(2)

Here $P$ is the power of the elastic backscatter signal. Superscripts “p” and “m” indicate particle and molecule backscattering, while subscripts “$\perp$” and “$\parallel$” indicate cross- and co-polarized components, $C$ is the calibration constant. Particle depolarization is calculated as suggested by (Freudenthaler et al., 2009):

$$\delta = \frac{(1 + \delta_m) R - (1 + \delta^v) \delta_m}{(1 + \delta^v) R - (1 + \delta^\perp)}$$

(3)

Here $\delta_m$ is the molecular depolarization ratio and $R$ is the aerosol scattering ratio:

$$R = \frac{\beta_p^v + \beta_m^v}{\beta_{\parallel}^v}$$

(4)

For further convenience we will use the notations $\beta = \beta_p^v + \beta_{m}^v$, and $\alpha = \alpha^v$. To characterize the spectral dependence of $\beta$ and $\alpha$, the backscattering and extinction Ângström exponents (BAE and EAE) for wavelengths $\lambda_1$ and $\lambda_2$ are calculated as:
The lidar-derived backscattering and extinction coefficients can be inverted to the particle microphysical properties, as described at Veselovskii et al., (2002). The only constraints on the permitted refractive index and the particle size distribution are that the refractive index is considered to be wavelength independent and that the concentration of the particles with radii below some $r_{\text{min}}$ and above some $r_{\text{max}}$ is zero, where the values of these radii are found in the process of inversion.

\[
A^b = \ln\left(\frac{\beta_\alpha}{\beta_\lambda}\right), \quad A^\alpha = \ln\left(\frac{\alpha_\lambda}{\alpha_\beta}\right) \quad (5)
\]

2.2 MERRA-2 aerosol reanalysis

The MERRA-2 simulations of aerosol properties over the observation site were made using the GOCART model (Chin et al., 2002) integrated within GEOS-5. The model includes representations of dust, sea salt, black and organic carbon, and sulfate aerosols. The aerosol components are assumed to be externally mixed. The optical properties of these aerosol components are summarized in Appendix 1. Sulfate and carbonaceous aerosols are both assumed to be in the fine mode. Sea salt and dust are both represented by five size bins spanning 0.1 – 10 microns radius for dust and 0.03 – 10 microns dry radius for sea salt, allowing for simulation of both the fine and coarse fractions of each. A more complete description of how GOCART is implemented in GEOS-5 is provided in Colarco et al. (2010), which also includes a detailed evaluation of the model with respect to MODIS, MISR, and AERONET aerosol optical depth observations.

The aerosol optical properties are primarily based on Mie calculations using the particle properties as in Colarco et al. (2010) and Chin et al. (2002), with spectral refractive indices from the Optical Properties of Aerosols and Clouds (OPAC, Hess et al. 1998) database. However, for dust, non-spherical optical properties derived from an offline database are used (Colarco et al. 2014). For sea salt, sulfate, and the hydrophilic portion of carbonaceous aerosol, hygroscopic growth is considered following Chin et al. (2002), with growth factors from OPAC (Gerber, 1985). The refractive index for organic carbon is based on the 100% brown carbon case from Hammer et al. (2016) and it is implemented as described in Colarco et al. (2017).
The sources of aerosols in the model include wind-speed based emissions of dust and sea salt, fossil fuel combustion, biomass burning, biofuel consumption, biogenic particulate organic matter, and oxidation of di-methyl sulfide (DMS) and SO$_2$, which includes volcanic sources. Aerosol sinks include convective scavenging, dry deposition, and wet removal, where aerosol hygroscopic growth is considered in the calculation of particle fall velocity and deposition velocity. The model resolution is 0.5° × 0.625° latitude by longitude with 72 hybrid-eta layers from the surface to 0.01 hPa. Additional details of the simulation can be found in Randles et al. (2017) and Buchard et al. (2017).

In MERRA-2, aerosol and meteorological observations are jointly assimilated within GEOS-5. Aerosols are assimilated by means of analysis splitting and the local displacement ensemble (LDE) methodology (Buchard et al. 2015, 2016). The system assimilated MODIS, AVHRR, MISR, and AERONET 550 nm AOD. AERONET measurements are interpolated to 550 nm using the Angström relationship and the closest available channels, generally 500 and 675 nm. The assimilation determines an AOD increment, which corrects the model AOD in a way that minimizes the differences between the model and observations. The AOD increment both corrects for misplaced aerosol plumes, and scales the aerosol mass mixing ratio to match the observations. The 2D AOD increment does not contain enough information to correct either the vertical distribution of aerosols or the aerosol composition. Thus, the model determines the aerosol speciation, optical properties, and vertical distributions, while the AOD increments modulate the aerosol mass. Thus, the assimilated aerosol distributions and physical and optical properties arise from the forecast model assumptions and the formulation of the aerosol data assimilation algorithm.

3. Experimental results

The smoke layers from forest fires near the equator were regularly observed over the instrumentation site during the wintertime measurement sessions made in December 2015 – January 2016. In our study we will focus on a strong smoke episode that occurred on 24-27 December 2015. Air mass back trajectories over Mbour on 25 December 2015 at 04:00 UTC are shown in Fig.1 together with map of fires on 20 December 2015 (https://worldview.earthdata.nasa.gov).
The air masses below 1000 m (red line) are transported over the desert and are strongly loaded by dust, while air masses at 3000 m (green line) arrive from the South and pass over the regions of forest fires, and thus can transport smoke particles. The Cimel MPL operated continuously through the period of 24-27 December and thus monitored the arrival and evolution of the smoke layer, as shown in Fig.2. An elevated smoke layer appears on 24 December around 00:00 UTC. The aerosol layer becomes thicker during the day but remains confined to the height interval of 2.5 km– 4.0 km and stays well separated from the dust layer, which extends from the ground to approximately 2.0 km. Such structure of the layers is preserved throughout 25 December, as well. Cirrus clouds appear at 08:00 UTC on 24 December at a height of 8 km, soon after the smoke layer arrival (Fig.2a), and persist throughout the smoke episode. After 12:00 UTC on 24 December the clouds start descending and by 7:00 UTC on 25 December the cloud base is below 6 km (Fig.2b). On 26 December strong precipitation of ice particles occurs (Fig.2c) and finally, on 27 December the cloud is located at the top of the smoke-dust layer (Fig.2d).

Multiwavelength Raman lidar observations are available for the 23-25 December period only. The height – temporal evolution of the particle backscattering coefficient $\beta_{532}$, depolarization ratio $\delta_{532}$ and water vapor mixing ratio $w$ measured by Raman lidar on the nights 23-24 and 24-25 December 2015 are shown in Fig.3. Due to the geometrical overlap factor the extinction data can be processed starting from approximately 750 m, thus plots of all parameters start at this height. The depolarization ratios of pure dust observed during SHADOW are in the 30-35% range (Veselovskii et al., 2016), while the depolarization ratio of smoke at 532 nm normally is below 10% (e.g. Tesche et al., 2011, Burton et al., 2015). Hence depolarization measurements provide a convenient way to separate the aerosols into dust and smoke components.

On the night of 23-24 December the dust layer extends up to 2500 m, but high depolarization ratio (>30%), which is usually associated with pure dust, is observed only below 1000 m, meaning that in 1000 m – 2500 m range the dust is probably mixed with smoke. The optical depth of the elevated smoke layer is rather small on 23-24 December (0.1 at 5:00 UTC), but on 24-25 December it increases up to 0.25 making possible the calculation of extinction coefficients from the Raman lidar signals. For analyzing the vertical distribution of smoke and dust particle parameters, we focus on the nighttime measurements of 24-25 December 2015.
Fig. 4 shows the horizontal wind direction and speed measured by the wind lidar on 24-25 December. The range corrected signal of the wind lidar can be evaluated starting from 100 m height, and the corresponding height–temporal image is shown in Fig. 5. The wind speed was measured in the dust layer (<1500 m) for the whole period, however inside the smoke layer the backscatter signal is lower, so the measurements were possible only in the period of 16:00-22:00 UTC on 24 December. During 24-25 December 2015, the wind in the low troposphere (<1500 m) is mainly dominated by the easterly Harmattan continental trades. Deceleration and acceleration of the lower part of the Harmattan (<1000 m) are observed, respectively in the beginning of the afternoon and during the night. The vertical profile of the wind speed demonstrates the presence of a Low Level Jet (LLJ) where the maximum wind speed (Jet speed) is located at a height of 350 m (LLJ height) at 1:00 UTC. LLJs are known to contribute to regional horizontal aerosol transport and to increase vertical mixing. Indeed, the LLJ occurrence at 1:00 UTC increases the aerosol loading by transporting desert dust. The corresponding increase of backscattering due to the LLJ at 1:00 UTC on 25 December can also be seen in Fig. 3.

The vertical profiles of temperature T, potential temperature $\Theta$, wind direction and speed together with relative humidity RH and water vapor mixing ratio (WVMR) from radiosonde launched from Dakar at 00:00 UTC on 25 December 2015 are shown in Fig. 6. The profile of wind speed and wind direction obtained from the sonde confirms that the LLJ observed with lidar at Mbour is not a local phenomenon, because it is also observed at Dakar. The vertical profile of the potential temperature suggests that the Nocturnal Boundary Layer (NBL) top corresponds to the LLJ height. Above 3000 m, the lidar and sonde depict southerly winds which transport the smoke plume. The water vapor mixing ratio increases above 2500 m; as a result the RH in the smoke layer reaches 75% while in the dust layer RH is below 30%.

To quantify the vertical distribution of particle parameters, Fig. 7 shows the profiles of backscattering ($\beta_{355}, \beta_{532}, \beta_{1064}$), extinction ($\alpha_{355}, \alpha_{532}$) coefficients and the particle depolarization ratio ($\delta_{532}$) derived from Raman lidar measurements for three temporal intervals on the night of 24-25 December: 19:00-23:00 UTC, 1:00-4:00 UTC and 4:00-7:00 UTC. For the profiles presented, the uncertainty of both $\beta$ and $\alpha$ computations is estimated to be below 10% for the Raman technique and to be below 20% for $\beta_{1064}$ computation by the Klett method. The relative uncertainty of depolarization measurements is below 15%. The extinction and backscattering Ångström exponents $A_{355/532}^\alpha, A_{355/532}^\beta, A_{1064/532}^\delta$ are given by Fig. 8.
For the first temporal interval (Fig.7a) dust and smoke layers are well separated. Extinction coefficients $\alpha_{355}$ and $\alpha_{532}$ differ in the smoke layer ($\alpha_{355} > \alpha_{532}$), but inside the near-surface dust layer (below 1750 m) the extinction values are nearly the same. The depolarization ratio is $\delta_{532} = 35\pm5\%$ at 750 m and it gradually decreases with height to $27\pm4\%$ at 1750 m. Above that height $\delta_{532}$ decreases quickly, indicating an increase in the contribution of smoke particles.

For the second and third temporal intervals the dust and smoke layers appear to mix leading to layering in the backscattering coefficient in the 1000 m – 2000 m range. The EAE in this range is increased up to 0.5 (Fig.8c) indicating that these layers may contain significant amounts of smoke.

The EAE of pure dust observed during SHADOW is slightly negative $A_{532/355}^\alpha = −0.1$ (Veselovskii et al., 2016). In Fig.8a the EAE below 1500 m is about 0.2±0.2, so the dust likely contains some amount of smoke. Values of EAE close to zero are observed in Fig.8b,c below 1000, where the depolarization ratio increases up to 35±5%. Inside the dust layer $\beta_{355} < \beta_{532}$, so the corresponding backscattering Ångström exponent is negative. The negative values of $A_{532/355}^\beta$ have been already reported by Veselovskii et al., (2016), where negative BAE was attributed to an increase of the imaginary part of the complex refractive index at 355 nm compared to 532 nm.

In the center of the elevated layer at 3100 m $\delta_{532} = 14\pm3\%$, while at the top of this layer $\delta_{532}$ decreases to $6\pm1.5\%$ (fig.8a), indicating a possible presence of dust particles in the center of elevated layer. The loading of elevated layer with dust particles is supported also by the profiles of $A_{532/355}^\beta$: for all three temporal intervals $A_{532/355}^\beta$ demonstrates the dip in the center of elevated layer, while $A_{532/355}^\alpha$ and $A_{532/1064}^\beta$ do not decrease in the 2500-4000 m range. As mentioned, for pure dust $A_{532/355}^\beta$ is negative, so presence of dust in the center of smoke layer should decrease the backscattering Ångström exponent. The presence of dust in the smoke layer is not surprising, because upwelling airflows in forest fires region can lift a significant amount of dust together with biomass burning products (Nisantzi et al., 2014). We should mention also, that the spectral dependence of the imaginary part (and thus $A_{532/355}^\beta$) depends on the dust origin. In particular, no negative values of $A_{532/355}^\alpha$ of dust were reported during the SAMUM campaign (Tesche et al., 2011).

Lidar ratio profiles at 355 nm and 532 nm, for the same temporal intervals as in Fig.7, are shown in Fig.9. The lidar ratios in the dust layer at 532 nm and 355 nm for 19:00-23:00 UTC
period are LR$_{532}$=55±8 sr and LR$_{355}$=70±10 sr, respectively. At the top of the elevated layer, where the smoke particles are predominant, the lidar ratios for the same period are higher: LR$_{532}$=65±10 sr and LR$_{355}$=75±11 sr. Due to the presence of dust in the center of the elevated layer, the height dependence of lidar ratios shows a decrease, with a minimum at approximately 3000 m for all three temporal intervals. The decrease is more pronounced at 532 nm, because the difference between smoke and dust lidar ratios is larger at this wavelength. The lidar ratios below 2000 m at 01:00-04:00 and 04:00-07:00 UTC become strongly oscillating because of high gradients of backscattering and extinction coefficients at low altitudes and are not shown due to high uncertainties.

Fig.10, shows the dependence of the particle depolarization ratio $\delta_{532}$ on the extinction Ångström exponent derived from data in Fig.7,8. The depolarization ratio monotonically decreases while EAE rises from 0 to 0.9. Thus observed high values of the depolarization ratio are attributed to big dust particles with EAE close to zero, while small smoke particles are characterized by low depolarization (below 10%). If depolarization ratios of smoke $\delta^s$ and dust $\delta^d$ are known, the contributions of smoke and dust particles to the total backscattering can be separated $\beta = \beta^s + \beta^d$ (Sugimoto and Lee, 2006; Tesche et al., 2009; Miffre et al., 2012; David et al., 2013, Burton et al., 2014). Assuming that the depolarization ratios of dust and smoke particles do not change with height the contributions $\beta^d$ and $\beta^s$ can be calculated as suggested by Tesche et al. (2009):

$$\beta^d = \beta (\delta^d - \delta^s) (1+\delta^d) (1+\delta^s)$$

and

$$\beta^s = \beta - \beta^d.$$ (6)

In our computations we used values $\delta^d = 35\%$ and $\delta^s = 7\%$.

The results of the decomposition of $\beta_{532}$ for $\beta^s_{532}$ and $\beta^d_{532}$ components for the same three temporal intervals as in Fig.7 are shown in Fig.11. This figure presents the total backscattering coefficient $\beta_{532}$ together with the particle depolarization ratio $\delta_{532}$. The dust contribution to backscattering is marked with magenta, while the residual backscattering $\beta_{532} - \beta^d_{532}$ is attributed to the smoke and is marked with grey. For the height regions with low backscattering the uncertainty of $\beta_{532}$ is high, so the decomposition for these regions is not shown. The dust is predominant below 1700 m for 19:00-23:00 UTC period, however even the elevated layer contains a significant amount of dust: at 3100 m $\beta^d_{532} \approx 0.3\beta_{532}$. After 01:00 UTC the smoke
layers descend (Fig.3) and their contribution to backscattering becomes significant down to 1000 m height.

4. Comparison of lidar measurements with MERRA-2

MERRA-2 provides the vertical distribution of mass mixing ratios of five aerosol components, so for each of these components the extinction, backscattering coefficients and depolarization ratios can be calculated. The vertical profiles of extinction coefficient of dust, black carbon (BC), organic carbon (OC), sea salt (SS) and sulfates (SU) together with total extinction $\alpha_{532}$ are shown in Fig.12 for 03:00 UTC and 21:00 UTC on 24 December 2015. At 03:00 UTC the aerosol is localized below 3000 m. Dust extinction is predominant, but contribution of OC to the total extinction coefficient rises with height reaching maxima at 2250 m. The presence of a significant amount of OC agrees with the low values of depolarization ratio above 1500 m for this temporal interval in Fig.3.

At 21:00 UTC an elevated layer with a maximum of extinction at 3150 m is observed (Fig.12b). In this layer OC and dust provide similar contributions to extinction (about 40% at 3150 m height). From the results shown in Fig.11a we can estimate the contribution of dust to $\alpha_{532}$ in the center of elevated layer as 30% (by assuming the dust lidar ratio $LR_{532}=55$ sr), so the measured and simulated dust contributions are in good agreement. Below 1750 m the dust is the main contributor to the extinction coefficient providing 88% of $\alpha_{532}$ at 1000 m (Fig.12b). The observed dust contribution to $\alpha_{532}$ at the same height is about 90% (Fig.11a), which again shows good agreement between the model and measurements. Total contribution of BC and SU to extinction is below 20% in the elevated smoke layer, and in the near-surface dust layer their contribution is negligible. The extinction coefficients can be recalculated to the backscattering using model lidar ratios of the aerosol components. Fig.12c shows the profiles of backscattering coefficients at 532 nm computed for the same temporal interval as in Fig.12b. The simulation of the backscattering coefficient is more challenging than that of extinction, because backscatter depends more strongly on the particle morphology and refractive index. A detailed comparison of measured and modeled profiles of backscattering coefficients will be performed later in this section.

As mentioned, the comparison of model and observed values is more straightforward for extinction coefficients. Fig.13 shows the time-series of extinction profiles at 355 and 532 nm.
modeled for the night of 24-25 December 2015 at 18:00, 21:00, 00:00, 03:00, 06:00 UTC. The profiles are shifted relative to each other by 0.2 km\(^{-1}\). For comparison, the same figure presents the profiles of extinction coefficients derived from Raman lidar measurements. The model reproduces well the location of the elevated smoke layer, as well as the top of the near-surface dust layer. However, the model does not resolve the oscillations of extinction profile below 2000 m at 03:00 and 06:00 UTC on 25 January.

To quantify the difference between the measured (\(\alpha^{\text{meas}}\)) and modeled (\(\alpha^{\text{mod}}\)) extinction coefficients the difference \(\Delta \alpha = \alpha^{\text{meas}} - \alpha^{\text{mod}}\) was calculated. The statistical analysis of the frequency distribution of \(\Delta \alpha\) for all five profiles in Fig. 13 shows that at 355 nm the mean value of \(\Delta \alpha\) is -0.01 km\(^{-1}\) and standard deviation of 0.042 km\(^{-1}\). With typical values of extinction coefficient in elevated smoke layer and near-surface dust layer being on the order of 0.2 km\(^{-1}\), the relative difference of modeled and measured extinction is estimated to be below 25% for the time period considered. The results of statistical analysis for \(\alpha^{532}\) nm are similar.

To analyze how well the model reproduces the temporal variations of aerosol optical depth, Fig. 14 presents AODs at 355 nm on 23 – 24 December 2015 for two height intervals: 750 – 2000 m and 2500 m – 4500 m. The first interval corresponds to the near-surface dust layer, while the second interval corresponds to the elevated smoke layer. The AOD is calculated from the Raman backscatter channel, and in the daytime measurements could be processed only in the dust layer, due to enhanced background noise. Thus, daytime measurements in the elevated smoke layer are not plotted. The time of the appearance of the smoke layer is well represented in the model results (about 00:00 UTC on 24 December), however the lidar derived AOD of this layer increases rapidly from the first appearance of the layer, while in the model the rapid increase in AOD growth starts approximately 5 hours later. The model predicts that the maximum value of AOD in the smoke layer (0.27) is reached at 20:00-24:00 UTC interval, which reasonably agrees with observations: mean value of measured AOD for this interval is 0.23±0.02. After midnight the modeled AOD of the smoke layer decreases quickly, while lidar measured AOD stays about 0.25. The measured AOD of the near-surface layer agrees with the model. The observed AOD exceeds the model values in the beginning (at 00:00 UTC on 24 December measured and modeled AODs are 0.24 and 0.175, respectively), but after 10:00 UTC, the values are in better agreement. Thus, we can conclude that the model reproduces the temporal variability of AOD in the dust and smoke layers.
The agreement between modeled and observed extinction profiles provides an opportunity to test how well the backscattering coefficients can be modeled. Simulation of backscattering coefficients is especially challenging for dust for several reasons. First of all, we are not confident in the accuracy of the presumed scattering phase function in the backward direction. Second, the backscattering coefficient strongly depends on the particle refractive index, in particular on the imaginary part, which may vary over a wide range depending on dust origin. The in situ ground measurements in West Africa, performed during the SAMUM field campaign, demonstrate that the mean value of $m_i$ for dust episodes is about 0.003 at 532 nm and 0.02 at 355 nm. However deviation from these mean values for every individual measurement can be significant (Müller et al., 2009; Kandler et al., 2011; Ansmann et al., 2011). The imaginary part of RI of dust in the model is assumed to be 0.007 at 355 nm, following previous OMI data analysis (Torres et al., 2007) and 0.0025 at 532 nm.

Fig. 15 shows measured and modeled backscattering coefficients at 355 nm and 532 nm for the same five temporal intervals as in Fig. 13. At 355 nm the modeled and measured values agree for both the smoke and dust layers. However at 532 nm the aerosol backscattering coefficients agree only inside elevated layer, while below 1750 m the modeled $\beta_{532}$ significantly exceeds the measured values. As mentioned, the modeled lidar ratio $LR_{532}$ for the mixture is close to 40 sr at 1000 m, while the measured lidar ratio in the near surface dust layer is 55±8 sr. The reason for this disagreement could be that the assumed imaginary part of the refractive index for dust (0.0025 at 532 nm) is too low. Recall however, that we cannot determine the imaginary part of the refractive index for dust by simply adjusting the modeled lidar ratio to the measured one, because the lidar ratio depends on several factors besides $m_i$ such as the particle size distribution and the aspect ratio of the ellipsoids used in the model. It is possible that the particle size distribution in the model is too much weighted toward fine mode dust.

The modeled and measured particle intensive parameters, such as extinction $A_{532}^\alpha$ and backscattering $A_{532}^\beta$ Ångström exponents together with the particle depolarization ratio $\delta_{532}$ are shown in Fig. 16. The measurements are averaged over 19:00 – 23:00 UTC interval while modeled values are given for 21:00 UTC. The model reproduces well the observed vertical distribution of $A_{532}^\alpha$, both in the dust and the elevated layer. As follows from Fig. 7a, inside the dust layer $\beta_{535} < \beta_{532}$, so the corresponding $A_{535}^\beta$ is negative with a minimum value of about -
The model predicts values of $A_{532}^β$ as low as -1.4. The modeled BAE is sensitive to the choice of the imaginary part of RI at 355 nm and 532 nm and, as mentioned, the chosen $m_0(532)=0.0025$ may be too low for this episode. In the elevated layer the modeled $A_{532}^β$ is close to the observed one. The modeled BAE has no minimum in the center of elevated layer, because the modeled ratio of dust and OC aerosol concentrations shows only a small variation throughout the elevated layer.

The model reproduces reasonably well the depolarization in the elevated layer, but inside the dust layer the modeled $δ_{532}$ is significantly lower than what is observed (22% compared to 35%). This problem is well known: the spheroidal model underestimates the depolarization ratio when typical dust PSD and complex refractive index are used (Veselovskii et al., 2010; Wiegner et al., 2009; Müller et al., 2013; Nowottnick et al., 2015).

One of the MERRA-2 data products is the water vapor mixing ratio (WVMR), which helps to identify atmospheric parcels, is critically important for determining atmospheric stability and serves as the source of water for aerosol hygroscopic growth. Fig.17 shows five model profiles of WVMR together with the results of Raman lidar measurements for the same temporal intervals as in Fig.13. The model reproduces rather well the WVMR profile inside the elevated layer (2500 m – 4500 m) on 24 December, though on 25 December the modeled values in this range are lower than the observations. In the near-surface dust layer, the deviation of modeled values from the measurements is larger. Statistical analysis of the deviation of modeled values from lidar measurements for all five profiles, shows that mean difference is 0.04 g/kg with standard deviation of differences of 1.6 g/kg. Thus in the elevated layer, where WVMR is approximately 8 g/kg, the agreement is quite good, however in the dust layer, which is characterized by low water vapor content (below 4 g/kg), the difference may be up to 40%.

5. Inversion of lidar measurements to particle microphysical properties

In the previous section, as validation of the model output we compared the modeled aerosol optical parameters, such as extinction, backscattering coefficients, and depolarization ratio with the values derived from lidar measurements in a straightforward way. The comparison of particle microphysical properties such as volume, effective radius and complex refractive index, however, is not straightforward, since it needs inversion of the measurements and requires
additional assumptions. In the case of dust particles the inversion becomes especially challenging, because:

- The size distribution of dust contains a strong coarse mode with particle radii extending up to ~ 15 µm, and the estimation of properties for such big particles is difficult since measurements are only performed in the wavelength range 0.355-1.064 µm;
- The inversions have to consider the refractive index as spectrally independent. In fact, the imaginary part of the dust RI is spectrally dependent with a strong enhancement at 355 nm compared to 532 nm;
- The dust particles are not spherical so that the application of Mie formulas for the forward modeling results in errors in computing the scattering phase function.

Regarding the shape issue, one of the ways to mimic the scattering properties of dust particles is to use the model of randomly oriented spheroids (Mishchenko et al., 1997; Dubovik et al., 2006). The implementation of this model for inversion of dust lidar measurements is described in Veselovskii et al., (2010, 2016), and Müller et al., (2013). This algorithm was used also for inversion of our 3β+2α observations. The range of particle radius in the inversion has been set to a minimum and maximum of 0.075 and 15 µm, respectively. The real part of RI was allowed to vary in the range 1.35 - 1.65, while the imaginary part varied in the range 0 - 0.02. The refractive index was assumed to be spectrally independent. The effects of a possible spectral dependence of the imaginary part of RI were considered in Veselovskii et al., (2016).

Profiles of the effective radius, volume density, and real part of the refractive index retrieved from optical measurements in Fig.7a are shown in Fig.18. The inversion was performed for two cases, with the assumption of all spherical particles or all spheroids. A realistic solution (for the mixture of spherical and non-spherical particles), should be closer to spheroids in the dust layer, while in the elevated layer it should be closer to the results obtained with spheres. The model results provided by MERRA-2 are shown on the same plot. The effective radius and volume density obtained in assumption of spherical particles are always higher than the values obtained with spheroids. The modeled effective radius at 1000 m height is 1.1 µm, which is close to \( r_{\text{eff}} = 1.0\pm 0.3 \) µm obtained from lidar measurements using the spheroids model. Lidar derived effective radius in the elevated layer at 3000 m is approximately 0.4 µm and 0.5 µm when spheroids and spheres are used respectively, while the modeled value is 0.3 µm. The reason for the lower value of modeled effective radius is the contribution of black carbon, which is
characterized by small size and relatively low hygroscopic growth. Recall that in the inversion of lidar measurements, the smallest radius considered is 0.075 μm. Modeled values of the volume density agree well with lidar retrievals in both dust and elevated layers.

The estimation of the real part of RI from lidar measurements is sensitive to the type of kernel functions chosen for retrieval. In the regularization algorithm the treatment of dust particles as spheres strongly underestimates $m_R$ (Veselovskii et al., 2010), so results obtained with spheres in the dust layer are not shown in Fig.18c. At 1000 m the $m_R$ retrieved with spheroids is 1.52±0.05, which agrees well with the modeled value. Inside the elevated smoke layer, where fine mode particles predominate, the application of spheroids overestimates $m_R$. The lidar derived real part of RI at 3000 m is 1.43±0.05 for spheres and 1.51±0.05 for spheroids, so we expect that the true value would lie within this. The simulated value of $m_R=1.50$ in the elevated layer is quite high, which is again the result of BC contribution.

The single scattering albedo (SSA) is one of the key parameters to be retrieved and conclusions about the potential of the multiwavelength lidar method strongly rely on its ability to profile SSA. Fig.19 shows SSA at 355 nm, 532 nm and 1064 nm. As mentioned, the spectral dependence of $m_I$ was not accounted for and the algorithm retrieves an average value of the imaginary part over the interval of 355 nm – 1064 nm. In particular, for dust and OC the imaginary part is underestimated at 355 nm and overestimated at 532nm and 1064 nm. As a result, in the dust layer the retrieved SSA exceeds the model values at 355 nm, while at 532nm and 1064 the situation is opposite. Still at a height of 1000 m, the difference between modeled and lidar derived SSAs is below 0.04 for all wavelengths. In the elevated layer, where the spectral dependence of $m_I$ is less pronounced, the simulated and retrieved SSA agree well with a corresponding difference of less than 0.02.

6. Summary and conclusion

The synergy of lidar observations with the aerosol transport model has a great potential to improve the characterization of aerosols properties, and as a first step in such synergy one has to demonstrate how well observations and models agree and describe the same aerosol scenario. For that we have considered a smoke/dust episode over West Africa to compare the vertical profiles of particle parameters modeled by MERRA-2 and retrieved from Raman lidar measurements. In the case selected, the simultaneous presence of the dust and smoke layers...
resulted in significant height variation of particle parameters, providing a good opportunity to test the models’ capability to reproduce complicated vertical structure. Modeled and observed vertical profiles of $\alpha_{355}$ and $\alpha_{532}$ show good similarity: MERRA-2 provides the correct location of both the near-surface and elevated layers.

The modeling of the dust lidar ratio is challenging due to irregularity of the particles shape and due to the spectral dependence of the imaginary part of the refractive index. $m_i$ can change significantly for dust of different origin and this variability may be accounted for in future model developments. The modeled at 355 nm the lidar ratio of 65 sr in the near-surface dust layer is close to the observed value (70±10 sr). At 532 nm however, the simulated dust lidar ratio (about 40 sr) is lower than measurements (55±7 sr). This discrepancy may be an indication that $m_i$ of dust during the episode considered is higher than the value assumed in the model. Another possible explanation is that the model particle size distribution is too much weighted toward fine mode dust. The measured lidar ratios at the top of the elevated layer, where smoke particles are predominant, are $LR_{355}=75±11$ sr and $LR_{532}=70±10$ sr, which is close to the corresponding model values for organic carbon of 71 sr and 66 sr, respectively.

MERRA-2 predicts the existence of a significant amount of dust in the elevated smoke layer, and the high values of observed depolarization ratio agree with this prediction. The existence of minima of $A'^{d}_{355/532}$ in the center of elevated layer, characterized by the highest $\delta_{532}$ also supports this finding. Moreover, the lidar ratios at both 355 and 532 nm also have a minima in the center of the layer because the lidar ratio of dust is lower than that of smoke. The contributions of dust and smoke particles to the aerosol backscattering and extinction coefficient at 532 nm evaluated from particle depolarization ratio agree with the values provided by the model.

Of course an analysis of only one episode is not sufficient for broad conclusions regarding how well the model reproduces the vertical distribution of particle properties. More measurements at different locations are needed. However, the results presented here demonstrate that observations and the MERRA-2 model contribute in a complementary way allowing to separate the contributions of different chemical component of the aerosol mixture. Analysis of different measurement sessions along with the use of modeled particle parameters in lidar retrieval schemes are in our future plans.
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APENDIX

Optical properties of aerosol components in MERRA-2 model.

Table 1 summarizes the main characteristics of five aerosol components: dust, sea salt, black carbon, organic, carbon and sulfates used in MERRA-2 model. For dust and sea salt five size bins are considered. All values are given for the relative humidity RH=0. Thus OC, BC and SU with the effective radii of 0.09 µm, 0.04 µ and, 0.157 µm respectively are presented by the fine fraction only, while dust and sea salt contribute to both fine and coarse fractions.

The dust particles are assumed to be hydrophobic, but other aerosol components may present significant hygroscopic growth. To account for the effect of relative humidity, the growth factor $g$, which is the ratio of particle radius at current RH to the dry particle radius, is introduced. Fig.A1 shows dependence of the growth factor of different aerosol components on relative humidity (RH). For sea salt the results are given for five size bins from Table 1. Each bin has different growth factor: $g$ increases with increase of particle radius. Relative humidity modifies also the particle complex refractive index (CRI). Dependence of the real and the imaginary part of particle components on relative humidity is shown in fig.A2. For dry sea salt particles RI is supposed to be the same for all size bins. However in the process of hygroscopic growth the RI of different bins behaves differently: both $m_r$ and $m_i$ decrease with bin number (radius) increasing.
Table 1. Parameters of the aerosol components, such as minimal radius \( r_{\text{min}} \), maximal radius \( r_{\text{max}} \), effective radius \( r_{\text{eff}} \), real \( m_R \) and imaginary \( m_I \) part of the refractive index at 355 nm, 532 nm, 1064 nm used in MERRA-2 model. For dust and sea salt five size bins are considered. All values are given for RH=0.

<table>
<thead>
<tr>
<th>Component</th>
<th>Bin</th>
<th>( r_{\text{min}} ) µm</th>
<th>( r_{\text{max}} ) µm</th>
<th>( r_{\text{eff}} ) µm</th>
<th>( m_R \text{355} )</th>
<th>( m_R \text{532} )</th>
<th>( m_R \text{1064} )</th>
<th>( m_I \text{355} )</th>
<th>( m_I \text{532} )</th>
<th>( m_I \text{1064} )</th>
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<td>0.1</td>
<td>1.0</td>
<td>0.64</td>
<td>1.53</td>
<td>1.53</td>
<td>1.53</td>
<td>0.007</td>
<td>0.0026</td>
<td>0.0022</td>
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<tr>
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<td>1</td>
<td>1.5</td>
<td>1.32</td>
<td>1.53</td>
<td>1.53</td>
<td>1.53</td>
<td>0.007</td>
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<td>1.53</td>
<td>1.53</td>
<td>1.53</td>
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<td>0.0026</td>
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<tr>
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<td>3.0</td>
<td>7.0</td>
<td>4.17</td>
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<td>1.53</td>
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<tr>
<td></td>
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<td>7.0</td>
<td>10.0</td>
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<td>1.53</td>
<td>1.53</td>
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<td>0.08</td>
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<td>1.50</td>
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<td>0.27</td>
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<td>1.2E-8</td>
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<tr>
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<td>1.53</td>
<td>1.52</td>
<td>0.048</td>
<td>0.009</td>
<td>0.016</td>
</tr>
<tr>
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<td>1.75</td>
<td>1.75</td>
<td>0.46</td>
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<tr>
<td>SU</td>
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<td>0.157</td>
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<td>1.42</td>
<td>1E-8</td>
<td>1E-8</td>
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Fig. A1. Dependence of the growth factor of organic carbon, black carbon, sulfates and sea salt on relative humidity (RH) used in MERRA-2. For the sea salt the results are given for five size bins from Table 1. The growth factor increases with increase of bin number.
Fig.A2. Dependence of the real and imaginary part of the refractive index of organic carbon, black carbon, sulfates and sea salt on relative humidity (RH) used in the MERRA-2 model. For the sea salt the results are given for five size bins from Table 1. Both $m_R$ and $m_I$ decrease with bin number increasing.
Fig. 1. Five-day backward trajectories for the air mass in Mbour at altitudes 750 m, 1500 m, 3500 m, on 25 December 2015 at 04:00 UTC together with the map of forest fires on 20 December 2015.

Comment: Figures 8, 14, 15, 17, 18 we removed from original manuscript.
Fig. 2. Range corrected lidar signal (in arbitrary units) of Cimel MPL for 24 - 27 December 2015.
Fig. 3 Height-temporal distributions of the backscattering coefficient and particle depolarization ratio at 532 nm together with the water vapor mixing ratio derived from the Raman lidar measurements on the nights 23-24 (left column) and 24-25 December 2015 (right column).
Fig. 4. Time-height section of horizontal wind direction (arrows) and wind speed (color map) deduced from Doppler lidar during 24-25 December 2015. Leftward and downward arrows represent, respectively, easterly wind and northerly wind.

Fig. 5. Time-height section of the logarithmic range corrected lidar signal (in arbitrary units) deduced from the Doppler lidar measurements during the 24-25 December 2015 night.
Fig. 6. Vertical profiles of (a) temperature $T$, potential temperature $\Theta$, (b) wind direction and speed and (c) relative humidity RH and water vapor mixing ratio (WVMR) measured by the radiosonde in Dakar at 00:00 on 25 December 2015. Solid line in plot (a) shows the aerosol backscattering coefficient at 532 nm in arbitrary units measured by the Raman lidar at 21:00 on 24 December.
Fig. 7. Vertical profiles of the aerosol backscattering ($\beta_{355}$, $\beta_{532}$, $\beta_{1064}$) and extinction ($\alpha_{355}$, $\alpha_{532}$) coefficients together with the particle depolarization ratio ($\delta_{532}$) for three temporal intervals: 19:00-23:00, 01:00-04:00 and 04:00-07:00 UTC on 24-25 December 2015. The values of $\delta_{532}$ are multiplied by factor 0.02.

Comment [i13]: Was modified.
Fig. 8 Extinction \( A_\text{355/532}^\alpha \) and backscattering \( A_\text{355/532}^\beta, A_\text{532/1064}^\beta \) Ångström exponents together with backscattering coefficient \( \beta_{352} \) for the same three temporal intervals as in Fig. 7.
Fig. 9. Lidar ratios at 355 nm (solid lines) and 532 nm (dash lines) for three temporal intervals from Fig. 7.
Fig. 10. Particle depolarization ratio as a function of the extinction Ångström exponent derived from data shown in Fig. 7, 8.
Fig.11. Contributions of dust and smoke to the total backscattering coefficient $\beta_{532}$ together with particle depolarization ratio $\delta_{532}$ for three temporal intervals on 24-25 December 2015. Magenta and grey regions correspond to dust and smoke contribution to total scattering $\beta_{532} = \beta_{532}^{d} + \beta_{532}^{s}$. 
Fig. 12. Vertical profiles of extinction coefficients at (a) 03:00 UTC, (b) 21:00 UTC and (c) backscattering coefficients at 21:00 UTC on 24 December 2015 from MERRA-2 model at 532 nm. Profiles are given for five aerosol components: dust, black carbon (BC), organic carbon (OC), sea salt (SS), sulfates (SU) together with total extinction $\alpha_{532}$ and backscattering $\beta_{532}$. 
Fig. 13. Comparison of extinction profiles at (a) 355 nm and (b) 532 nm derived from Raman lidar measurements (line) and modeled by MERRA-2 (line + symbols) on the night 24-25 December 2015. Model profiles are provided at 18:00, 21:00, 00:00, 03:00, 06:00 UTC. The lidar measurements are given for temporal intervals centered at: 19:00, 21:00, 00:00, 03:00, 06:00 UTC. For each profile 2 hours of measurements are averaged. The profiles are shifted relatively to each other by 0.2 km\(^{-1}\).

Comment [i15]: Plot b was added.
Fig. 14. Aerosol optical depth at 355 nm on 23 – 24 December 2015 obtained from MERRA-2 (line + symbols) and from the Raman lidar measurements (solid lines). The results are given for two height intervals: 750 m – 2000 m (red) and 2500 m – 4500 m (black). Zero of time scale corresponds to 00:00 UTC on 24 December.
Fig. 15. Backscattering coefficients at (a) 355 nm and (b) 532 nm measured by Raman lidar (solid line) and modeled by MERRA-2 (line + symbols) on the night 24-24 December 2015. Profiles are shifted relatively to each other by 0.0025 km$^{-1}$ sr$^{-1}$. The temporal intervals are the same as in Fig. 13.
Fig. 16. Extinction ($A_{355/532}^\alpha$) and backscattering ($A_{355/532}^\beta$) Ångström exponents together with the particle depolarization ratio $\delta_{532}$ obtained from lidar measurements (line) and from MERRA-2 modeling (line + symbols). Lidar data are averaged over 19:00 – 23:00 UTC period while model data are given for 21:00 UTC.
Fig. 17. Water vapor mixing ratio derived from Raman lidar measurements (solid line) and obtained from the model (line + symbols) on the night 24-25 December 2015. Temporal intervals are the same as in Fig. 13. The profiles are shifted relatively each other by 5 g/kg.
Fig. 18. Profiles of (a) effective radius, (b) particle volume and (c) real part of the refractive index on 24 December 2015 retrieved from 3β+2α lidar measurements shown in Fig. 7a (solid symbols) and provided by MERRA-2 for 21:00 UTC (open symbols). Inversion of lidar measurements was performed in assumption of spherical particles (s) and using the model of spheroids (ns).
Fig. 19. The single scattering albedo at 355 nm (blue), 532 nm (green) and 1064 nm (red) on 24 December 2015 retrieved from 3β+2α lidar measurements shown in Fig.7a (solid symbols) and provided by the MERRA-2 model for 21:00 UTC (line + open symbols). For inversion of lidar data the spheroids (ns) were used below 2000 m and spheres (s) above 2000 m.