

We thank the reviewers for their suggestions, which certainly helped us improving the manuscript. We tried to account for many of the points which are listed below in the text. In the following the reviewer's comments are presented in italics, the author answer with normal letters in blue and the modifications on the manuscript with bold blue letters.

Anonymous Referee #1

General points

In Section 2 the authors report about trace gas measurements and show a global distribution in Figure 1. These measurements and Figure do not have an impact on the further analysis and inversion of the lidar data and should be omitted to make space for some other analysis which are missing in this paper, for example a case study of urban/industrial aerosols and more information about the mixed biomass burning layers.

In section 2.4, paragraph 4, we report on trace gas measurements. These measurements were actually used to classify the origin of the aerosols in each of our case study of table 1. The trace gas measurements are part of our analysis on the categorization of urban/industrial aerosol type. In particular, as clearly mentioned in the manuscript '15 min data were averaged for the extent of measurement time for each of the measurements periods (Table 1). For instances where the combined use of trajectory analysis and fire hotspots did not indicate the presence of biomass burning aerosols we checked whether the measured NO_x, SO₂ or H₂S concentrations were higher than the seasonal mean values of that measured for the entire period of the EUCAARI campaign. These seasonal mean values are presented in Laakso et al. (2012). In addition, when the trace gases concentrations were lower than the mean seasonal values measured during the EUCAARI campaign and biomass-burning activity or desert dust advection were absent, we checked if the daily concentration of the trace gases exceeded the mean critical values.'

Figure 1 show the global map of long-term average tropospheric NO₂ column derived from SCIAMACHY data from August 2002 to March 2012. We agree with the reviewer that this figure do not have an impact on further data analysis but this figure is used to demonstrate the distribution and intensity of urban / industrial aerosols in the region. It is used in the section 2.1 which is the description of measurement site and we believe that helps (together with Figure 2) the reader to understand the existence of the 2 dominant aerosol sources. For this reason we prefer to keep Figure 1.

The authors state that the uncertainties of the extinction coefficient are in the order of 10-30%. Later they make assumptions of differences in the size of the particles mainly indicated by differences of the Angstroem exponent. How are these assumptions and the Angstroem exponents affected by the general uncertainties of the extinction coefficient?

We thank the reviewer#1 for the comment. In section 2,2 we briefly state the errors in backscatter, extinction, depolarization and lidar ratio, but we missed to state the error in the Ångström exponent. We now clearly state the error also in the Ångström exponent. **'The overall relative errors of the lidar-derived aerosol properties range between 5%-15% for the backscatter coefficients, 10%-30% for the extinction coefficients, 20%-40% for the Ångström exponents, 15%-40% for the lidar ratios and approximately 5%-10% for the linear particle depolarization ratio (Hänel et al., 2012).'** Also a reference ('Wagner et al., 2008') is cited in the revised manuscript where the error in Ångström exponents and how these errors are influenced by errors in aerosol optical depths, is discussed in detail.

The authors should report more about their uncertainties; what do they consider for their analysis of the uncertainties. Which parameters are not considered? If possible they should do an error analysis for all reported and considered measurement cases.

A detailed error analysis for FMI-Polly^{XT} systems has not been done, and it would take up an entire new publication including the error propagation formalisms, the separation of statistical and systematic errors, Monte-Carlo approach and known uncertainties for all of the channels. However some discussion on the error

analysis are already made on *Baars et al. 2016* and on *Engelmann et al., 2016* and these two publications are cited in the revised manuscript.

Baars, H., Kanitz, T., Engelmann, R., Althausen, D., Heese, B., Komppula, M., Preißler, J., Tesche, M., Ansmann, A., Wandinger, U., Lim, J.-H., Ahn, J. Y., Stachlewska, I. S., Amiridis, V., Marinou, E., Seifert, P., Hofer, J., Skupin, A., Schneider, F., Bohlmann, S., Foth, A., Bley, S., Pfüller, A., Giannakaki, E., Lihavainen, H., Viisanen, Y., Hooda, R. K., Pereira, S. N., Bortoli, D., Wagner, F., Mattis, I., Janicka, L., Markowicz, K. M., Achtert, P., Artaxo, P., Pauliquevis, T., Souza, R. A. F., Sharma, V. P., van Zyl, P. G., Beukes, J. P., Sun, J., Rohwer, E. G., Deng, R., Mamouri, R.-E., and Zamorano, F.: An overview of the first decade of PollyNET: an emerging network of automated Raman-polarization lidars for continuous aerosol profiling, *Atmos. Chem. Phys.*, 16, 5111-5137, doi:10.5194/acp-16-5111-2016, 2016.

Engelmann, R., Kanitz, T., Baars, H., Heese, B., Althausen, D., Skupin, A., Wandinger, U., Komppula, M., Stachlewska, I. S., Amiridis, V., Marinou, E., Mattis, I., Linné, H., and Ansmann, A.: The automated multiwavelength Raman polarization and water-vapor lidar PollyXT: the neXT generation, *Atmos. Meas. Tech.*, 9, 1767–1784, doi:10.5194/amt-9-1767-2016, 2016.

In this study we limited ourselves to the general description of the error sources of FMI-Polly^{XT} and to those we can actually specify. Thus we include a small paragraph in the revised manuscript:

The uncertainties affecting the retrieval of extinction and backscatter coefficients, and thus the calculation of lidar ratio and Ångström exponents are mainly due: to the statistical error due to signal detection, the systematic error associated with the estimation of the atmospheric molecular number density from the pressure and temperature profiles, the systematic error associated with the evaluation of the aerosol scattering wavelength dependence, the systematic error for overlap function, the errors introduced by operation procedure such as signal binning (smoothing) and averaging accumulating lidar returns.

The authors classify their aerosol types mainly based on trajectory analysis. How these classifications are connected with lidar based classification schemes (Burton et al., 2012, Gross et al., 2013, or at 355 nm: Gross et al., 2015, Illingworth et al., 2015)? Please add this in your publication.

The aerosol type identification as described in section 2.4 (Aerosol classification) is based on three tools.

1. Backward trajectories (paragraph 2 of section 2.4)
2. Modis fire hot spots (paragraph 3 of section 2.4)
3. Trace gas measurements criteria (paragraph 4 of section 2.4)

We would like to make clear at this point that the purpose of this study is not to classify our measurements based on the post-processing of lidar data products but to pre classify our layers and then calculate and present our averaged intensive properties of the different types of aerosol layers in the region of South Africa. Also, at the last part of our study we compare our results with other studies (also those proposed by the reviewer#1).

How is the assumption of less absorption for biomass burning conform with the lidar ratio of 52 sr compared to 92 sr for industrial/urban aerosols?

In section 3 a case study of biomass burning aerosols is presented and discussed. In the layer between 1090m and 1900m, high values of the lidar ratio of 96 ± 5 sr at 355 nm and 89 ± 5 sr at 532 nm are calculated which indicate that the smoke particles inside this layer were most likely highly light-absorbing. The single scattering albedo for this particular layer was 0.86 at 532 nm which also indicates relatively strong-absorbing aerosols.

In Table 2 and 3 we also present the mean aerosol properties for the three aerosol types. The biomass burning particles were found to be larger and slightly less absorbing compared to urban/industrial aerosols. Our results for lidar ratios and single scattering albedo in general were within the same range of previously reported values. The slightly higher values of single scattering albedo for biomass burning aerosols (0.90 ± 0.06) compared with the single scattering albedo of urban/industrial aerosols (0.87 ± 0.06) is caused by the lower imaginary part of the retrieved complex refractive index of biomass burning aerosols ($0.016 (\pm 0.011)i$) compared with that of

urban/industrial aerosols (0.021 ± 0.010)_i). The values are presented in Table 3. Here the retrieved single scattering albedo is used as a metric of the absorption of the aerosols (and is not an assumption as the reviewer suggested). The lidar ratio is a parameter that is affected by the absorption, but it is also depending on the size of the particles. Smoke particles were found larger (effective radius 0.17 ± 0.04) than urban/industrial aerosols (effective radius 0.10 ± 0.03) which have an impact on backscatter efficiency and thus on lidar ratio values. Taking these into account, together with the standard deviation of the single scattering albedo (± 0.06) we think that our results are reasonable.

How do you calibrate your depolarization measurements? How does this calibration method affect your results? Please report in you publication.

Depolarization measurements at 355 nm are performed. The Rayleigh calibration method was applied within the data analysis under the assumption of pure Rayleigh depolarization in an aerosol-free height range (Behrendt and Nakamura, 2002). However, we should note here that the FMI Polly^{XT} system has been upgraded and we now perform measurements at 532 nm with the $\Delta 90^\circ$ -calibration (formerly known as $\pm 45^\circ$ -calibration method (Freudenthaler et al., 2009)).

How do the uncertainties in the single measurement parameters affect the result of the inversion algorithm?

The effect of the uncertainties of the single measurement parameters to the results of the inversion algorithm is discussed in detail in Müller et al., 2001 and is briefly reported in section 2.3: **'A minimum of three backscatter coefficients (355, 532, and 1064 nm) and two extinction coefficients (355 and 532 nm), with measurement errors less than 30%, are required as input in order to obtain microphysical results that have reasonably low uncertainties (Müller et al., 2001). The selection of the individual inversion solutions is based on the concept that the back-calculated optical data should agree with the original data within the limits of the measurement errors, and that a pre-selected discrepancy level, which is an output parameter of the inversion algorithm (Müller et al., 1999a), is not exceeded.'**

Are the +/- values the mean uncertainties or the standard deviation? Please add this information in your publication.

The +/- values are the standard deviation. The information is already provided in the text and the captions of the Tables 2 and 3 as well as in the Figure 7 (previously Figure 8).

Instead of showing the trace gas measurements the authors should show a figure with AOD, extinction coefficient or backscatter coefficient for one day prior to one day after their biomass burning case study as that seems to be an important point and mentioned in the text.

We would like to keep Figure 1 because we think that helps the reader to understand the dominant aerosol sources in the region. In Figure 4 (previously Figure 5) we now include the mean backscatter coefficient at 532 nm for one day prior and one day after the biomass burning case study as suggested by the reviewer. We do not include all three wavelengths to keep the figure as clear as possible.

A case study showing a urban/industrial aerosol case and a mixed biomass burning case is missing. Especial important would be to see the differences in transport way, extinction coefficient or AOD, and layering for the different cases.

We thank the reviewer for the comment. In this study we present our results on 38 aerosol layers, from which 17 are referring to urban/industrial, 14 to biomass burning and 7 to mixed of biomass burning with desert dust aerosols. We think that the measurement example is a good and common way to show the typical products of our system and to demonstrate the methodology used to derive the optical aerosol properties. Differences in transport way, extinction and backscatter coefficients as well as layering structures are observed at all cases, also within one cluster of aerosol types. Some information on the aerosol structure for each of the layer analyzed is already given in the Table 1 (bottom and top of each layer observed). Mean extinction coefficient at 355 nm and 532 nm for each of the layer observed are now add in the Table 1 of the revised manuscript as

suggested by the reviewer#1. The respective aerosol optical depths at 355 nm and 532 nm can now be easily retrieved from the mean extinctions coefficients and geometrical information provided in the Table 1.

The authors should give more evidence that the mixed biomass burning cases are not miss-classified. The measurements presented (e.g. in Figure 5) show almost the same values as what is classified as aged biomass burning aerosols in Illingworth et al., 2015. Furthermore Amiridis et al., 2009 reported that the optical properties of biomass burning aerosols alter during aging.

It is also not clear to me what really should happen with the dust particles. How would this affect their shape and optical properties? The authors should give more references and evidence for their assumption.

Thank you the reviewer for the comment. Illingworth et al., 2015 among other types, studied the intensive properties of aged boreal biomass burning aerosols and found depolarization values at 355 nm between 10 – 11 %. This range value is indeed very similar with our values for mixed biomass burning / desert dust particles. However, the lidar ratio values given by Illingworth et al., 2015 for aged boreal biomass burning (35 – 50 sr) is much smaller than the values reported in this study (59 – 90 sr).

Amiridis et al., 2009 reported a wide range of lidar ratio values at 355 nm for biomass burning aerosols and our values are within this range. They also studied the relation between backscatter related Ångström exponent and the age of carbon monoxide from the emissions. Our results on the intensive parameters (both for biomass burning and mixed aerosols) are in agreement with those reported by Amiridis et al., 2009. Although we should note that we report Ångström exponent related to extinction and not related to backscatter. The main difference between the biomass burning and biomass burning missed with desert dust is the depolarization ratio values and this is a parameter that is not reported in Amiridis et al. 2009.

According to our trajectory analysis there is certainly evidence of the transport of biomass burning aerosols in the measurement site. The smoke is relatively fresh (less than 3-day-old smoke plume). A relation between travel time and Ångström exponent was not found in our dataset, and we believe that the travel time is relatively short and thus don't make possible to see large differences in the intensive properties. The transport paths are different even in one cluster of aerosol types and thus we do not provide such a Figure.

We agree with the reviewer that the evidence of desert dust transport in our study is not so clear and thus we changed the name of this aerosol type to mixed aerosols. The mixing state of the aerosols is possible from desert dust but also industrial aerosols can not be excluded, especially in this region.

Is the assumption of mixture of dust and biomass burning also conforming in the lidar ratio? The authors report quite low lidar ratios compared to the referenced studies which they use as hint for their assumption. How these assumptions do are supported by results of optical modelling (e.g. Gasteiger et al., 2011 for the referenced measurements)?

Considering the last points a detailed case study should be added. This case study should also include information (satellite / reports) of dust activation and a connection to trajectory analysis including the mixing layer height and trajectory height.

A new publication including the mixing of desert dust, urban aerosols and biomass burning aerosols during biomass burning period will follow in the near future. The contribution of each of these aerosol types will be quantified using the information of particle depolarization ratio. In the present study we have changed the mixture of dust and biomass burning to a mixed state of aerosols, since there is not yet enough proof of dust activation. However, the larger depolarization values found for this mixture type cannot be explained by the age of the smoke plume as explained in the previous answer.

The lidar ratio values is very well compared with the mixture type as shown in Figure 7. The lower depolarization values and larger Ångström exponent values compared to the literature values can be explained both from the different kind of dust and smoke as well as from the different (less) contribution of the dust to the mixing state of aerosols.

Specific comments:

Abstract:

Change 'proper ties' to 'properties'.

done

Change 'single scattering, albedo' to 'single scattering albedo'.

done

Why not give also the lidar ratio at 532 nm?

done

AE for biomass burning is not consistent with Table 2.

Thank you the reviewer for the comment. The reviewer is right. In the new manuscript the AE for biomass burning is consistent with Table 2

Section 2:

Which is the range of full overlap?

Usually the overlap function is equal to 0.7 at heights between 300-500 m. In this study, we only report aerosol layers in the range of full overlap.

Section 3:

Please constrain your assumption of 'fresh smoke'; give references.

The travel time of air masses studied here is less than 3-day-old

Section 4:

Do you really mean anthropogenic here? Give references for this assumption.

Thank you for the reviewer's comment. It was not cleared in the manuscript that we were referring to our results and not to the literature. In the revised manuscript we have replaced the sentence 'Anthropogenic aerosol layers are characterized by lower lidar ratios in the range between 41 and 59 sr' with '**Urban / Industrial aerosol layers were found to have lower lidar ratio values in the range between 41 and 59 sr at 355 nm**'.

Figure 2:

Add 'of fire' to indicate which hot spots you mean.

done

Figure 3:

How does a quicklook with 15 km height resolution go conform with a reported vertical resolution of 30 km?

Thank you for the reviewer's comment. The vertical resolution is 30 m and not 15 m. In the revised manuscript the correct height resolution is reported in section 2.2. Figure 3 is deleted in the revised version as reviewer #3 has proposed.

Figure 4 / Section 3:

I cannot see that airmasses are coming either from northeasterly or northwesterly direction. A more detailed trajectory analysis including also the mixing layer height and trajectory height along the way would give more evidence at which part of the transport aerosol uptake took place.

Thank you the reviewer for the comment. In the new manuscript we have change Figure 4. In Figure 3a (previously Figure 4) we present the fire hot spots, and in Figure 4b we present the four day back-trajectories along with the trajectory height and mixing layer height as suggested by the reviewer. The discussion of the trajectory has also changed in the revised paper: '**MODIS fire hotspots product reveal that several fires were active during the period 28th of September 2010 – 1st of October 2010 as shown in Figure 3 (a). In Figure 3 (b), four-day backward trajectories arriving at Elandsfontein on 1st of October 2010 at 00:00 are presented. The trajectories are computed for arrival heights of the bottom, center and top of the observed layer. The trajectory**

analysis along with MODIS fire hotspots reveals that the air masses are highly possible to carry smoke particles at Elandsfontein on the day of the measurement. '

Figure 5:

How do you explain the increase of the backscatter ratio at 355 nm with height?

Thank you the reviewer for the comment. There is indeed an increase of backscatter coefficient at 355 nm. This is partially caused by the vertically smoothing applied in our analysis. This increase is small and inside the order of the error bars. In our analysis we don't take into account the lower part of the profile. Only the mean values of optical properties of the aerosol layer (grey region) were used and looking also the vertical distribution of lidar ratio and Ångström exponent there is only a small effect in the averaged values of the layer taken into account.

What is the vertical resolution of this data?

The vertical resolution of the data is 30 m as reported in section 2.2. We have applied a smoothing of 9 points (270 m) in the specific case study.

Figure 7:

Change your labeling from 'Depolarazation' to 'Depolarization'.

Thank you the reviewer for the comment. Done

Figure 8:

The labeling is not readable. Please change.

Thank you the reviewer for the comment. Done