Response letter to the comments of Reviewer 1:

First of all, it should be acknowledged that the authors have attempted to answer most of the reviewers questions and remarks. However, there are still a few major issues which have to be clarified and mentioned in the revised manuscript before it can be published in ACP.

1. The knowledge of the dry state (here the dry particle extinction coefficient) is an inevitable requirement when studying the hygroscopicity of aerosols. I understand that this is not a simple task for this kind of measurement technique, but it should have been more thoroughly discussed. This is especially important for the conclusions of this work since the majority of the results are based on this crucial assumption. In the revised version it states that 'Various measures were taken to ensure this precondition', however, it is highly questionable that backward trajectories and shorter time periods are applicable to ensure a constant dry extinction coefficient.
The authors state that these effects will be small and state an uncertainty of 20-30 %, however, this is pure speculation and most likely a significant underestimate. Looking at the PBL dilution effect alone, the authors already show in their first case study that the extinction coefficient decreased by 30% after the PBL height increased from 1300 to 1900 m. In addition to the variation of the PBL, other aerosol effects will have a large influence on the light extinction coefficient and thus increase the here stated uncertainty above 30 %. For example, changes in local emissions, gas-to-particle partitioning effects, new particle formation and other photo-oxidation processes. Again, this has to be clearly discussed and mentioned especially in the conclusions.

We want to thank the reviewer again for his efforts. After careful considerations of this comment, we now include an additional section in the paper. We are aware that this discussion is only an approximation of the true error, but hopefully the major critical point or drawback of the method now becomes clearer to the reader.

Considerations on the uncertainty of ¥ (Chapter after 3.6)

The fitting parameter ¥ depends on the knowledge of the initial dry particle extinction coefficient. Unfortunately this parameter is not easily available for our measurement technique. Although various measures were taken to ensure constant dry conditions (see section 3.6) certain errors can arise from the assumption of a constant dry particle extinction. Not only dilution effects within the PBL but also changes in local emissions, gas-to-particle partitioning effects, new particle formation, and other photo-oxidation processes could contribute to the error.

For our data from 2009 corresponding (hourly averaged) in-situ data (TDMPS + APS) which coincide with the cases used for the calculation of ¥ were available at TROPOS. These in-situ size spectra were used to determine the dry extinction coefficient in a simplistic manner (Mie scattering, λ=550 nm, n=1.53 + 0.01i, c.f. Fig. 9). In order to estimate an error that occurred from our assumption of a constant dry extinction coefficient we calculated the relative change of the dry extinction from the moment of highest and lowest RH, respectively, within our fit periods. On average of 54 cases from 2009 the dry extinction was indeed lower by 33.5% (with a standard deviation of 35.7%) during low RH conditions (usually later during the day) with respect to the time of high RH (usually in the morning).

The error propagation of Eq. 3 with respect to the uncertainty of the dry particle extinction $\Delta b_{p,e}(f=0)$ results to:

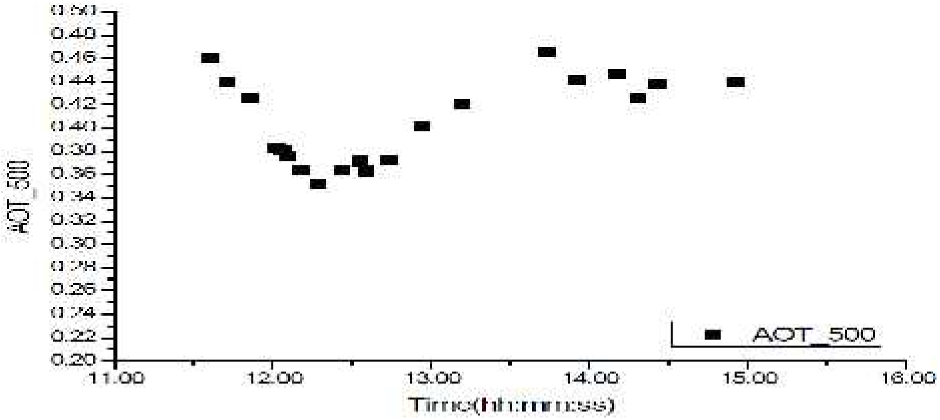
$$\Delta ¥ = \ln(1-f)^{-1}\ \Delta b_{p,e}(f=0) / b_{p,e}(f=0)\ \ \sqrt{N}^{-1}$$

where N is the number of data points in a fit (usually between 10 and 20). Typically, we performed ¥ fits in the RH range from 40-95%. Following these considerations, on average an overestimation of ¥ between 0.17 and 0.03 could be a result of the unknown dry particle extinction coefficient.

We also included the uncertainty in the conclusions: "*However, this precondition is not necessarily always valid and an overestimation of ¥ of 0.17 to 0.03 cannot fully be ruled out*."

2. Similar to the point above, I find it surprising that the AOD in the second case study (27 August 2009) stays more or less constant while the extinction coefficient at the surface decreases significantly. So if the loading is constant, as the authors state, why does the decreasing surface extinction (from approx. 0.4 to 0.1km^{-1}) not cause the AOD to decrease as well? For this particular day most likely really both the PBL dilution and the hygroscopicity were impacting the magnitude of the particle extinction coefficient measured at the surface.

The level 2 AOD data (500 nm) of the AERONET photometer only give measurements from 11:30 UTC (see Figure). The significant decrease of the extinction coefficient is measured between 0330 and 0800 UTC. In this case study we cannot really compare the photometer and the SAEMS measurements regarding the hygroscopic influence.



But in addition, the lidar measurements of Leipzig (*http://polly.tropos.de/?p=bilder&lidar=PollyXT_IfT&lambda=1064&Jahr=2009&Monat=8&Tag=27&Ort=Leipzig#bildanker*) show that an aerosol layer up to 2.5 km was present during this day. Hence, most of the column AOD was not affected by the hygroscopic surface layer variations. The typical nocturnal BL (where radiative cooling and humidity increase occurs during the night) is mostly only 0.2 - 0.3 km deep, so that the surface layer contains only 10-15% of the total columnar aerosol. A reduction of the extinction coefficient from 0.45 to 0.15 km^{-1} would thus only reduce the total AOD by 0.075.

To clarify this we included in sec 3.2:
"It should be noted that because of the present 2-km deep aerosol layer the decrease of the extinction coefficient in the shallow surface layer did not affect the column AOD significantly."

3. Since in the revised version only Eq. 3 is being used, I would suggest to remove Eq. 4 and 5 and the description of it.

Thanks for this remark. We have removed these equations as considered.

4. In the reply letter the authors state that only the spectral data from 2009 to 2010 was quality assured. Why is the 550nm channel 'more robust'? The authors need to justify this statement if the data for only this channel is to be shown for the entire 4 years. I ask because there is a clear difference between the 2009-2010 and 2011-2012 data (Fig. 7).

The detection unit of our system consists of a 2-channel spectrometer and a large photodiode with a 550 nm filter (Skupin et al. 2014). We use an automatic adjustment procedure to direct the transmitted light onto the retro reflectors and to receive the maximum light intensity onto the photodiode. The wide-viewing-field setup of the photodiode ensures to receive all of the light while the fibre-coupled spectrometer only receives a small amount of the returned light. In 2009 and 2010 we have studied the spectral extinction coefficient in detail and therefore regularly readjusted the optical fibre.

Since 2011 SAEMS runs automatically without continuous supervision. Hence often the spectral data did not receive enough light. The 550-nm data from the photodiode however are very robust to these misalignments. Thus we are very confident in the data of Fig 7d. (Also, a theoretical misalignment would always result in a loss of signal intensity, which in contrary would show as an increased extinction coefficient. But for the years 2011-2012 the values are reduced.)

We will mention this issue in the theoretical part of our paper:
“In 2009 and 2010 we have studied the spectral extinction coefficient in detail and carefully observed the alignment of the spectral channels on a daily basis. Since 2011 SAEMS did run automatically without continuous supervision and thus occasionally experienced data loss within the spectrometer. However, the 550-nm data were obtained by a large-area photodiode with an automatic adjustment system (Skupin et al. 2013) and were not affected by misalignments. Thus we present the quality assured spectral data only for 2009 and 2010 while the 550-nm data are used for the entire measurement period from 2009 to 2012.”

Some more minor issues:
_ Page 3: There is a lonely ’?’ before Draxler and Hess, 1998
_ Page 3: I would suggest to replace ’particle optical depth’ by ’aerosol optical depth’
_ Page 2, right column, third sentence: Remove ’us’
_ Fig. 7: There is a Latex error in the caption.

Thank you, we have fixed those issues.