Response letter to the comments of Reviewer 1

We would like to thank the referee for his comments. At first the comments were pretty tough but we think we could really improve the manuscript based on the suggestions. To some extent we see the concerns the reviewer has with our data analysis procedure.
But we also want to emphasize here that our methodology has to be seen from a remote-sensing perspective. In contrast to well designable in-situ experiments our focus lies an ambient measurements and the selection of appropriate cases from a much larger amount of measurements in order to derive optical hygroscopic effects on a statistical bases. The general differences between the two research paradigms shall not be debated in this paper. We try to ease the language accordingly.
Also the in-depth comparison of in-situ data with remote sensing data could only be performed for 2009 in this study, while most of the remote sensing data analyzed here are from 2009-2012.

Our response to the comments is given below.

1 General comments
Although I have no doubts on the high technical quality of the performed measurements,
I have strong doubts on the validity of the main statements and its underlying assumptions. As already mentioned at the quick-review stage of this manuscript, the main concern is that for the retrieval of the enhancement factor different atmospheric processes that cause a change in particle light extinction are mixed into one parameter. The authors have selected days with a pronounced RH cycle and used an empirical parametrization to fit their data. As nicely shown in their case studies (Fig. 5 and 10) a decrease in RH was often coincident with a decrease in particle light extinction, which of course can be partially due to hygroscopic growth. However, the diurnal cycle also causes a change in PBL height and thus a dilution of the aerosol concentration which directly influences the measured light extinction. In addition, local sources must largely have affected the measurements, i.e. a major highway runs below the line of sight of the instrument, which must have had an influence on the measurements, e.g. with diurnal variations during rush hours. Despite these major issues no satisfactory discussion can be found in the manuscript.

We will include these points in our discussion. You are absolutely right, since our method works at ambient measurement conditions there are of course a lot of factors that might influence the aerosol load during the day. For this reason we carefully selected 143 days from our four-year long measurement period were we think our method is applicable.
For cases were in-situ data are available at TROPOS we can show that the “dry” (i.e. at 30-40% RH) volume concentration did not change significantly during the single evaluation periods. While we observe extinction enhancement on the order of a factor of 2 to 5 (at 95% RH) the dry volume changes only on the order of 30% during the time period of such a case. Especially for our statistical dataset of extinction enhancement by relative humidity the uncertainty caused by PBL dilution is still small and not larger than for example the uncertainties caused by precision of RH measurements or of natural inhomogeneities along our light path.
Moreover, the argument that a PBL growing (let’s say from a nocturnal depth of ~200-300 m to a full extent of ~1.5-2 km in the afternoon) will dilute the aerosol in the same way is almost never so straightforward. As seen from lidar measurements, it regularly occurs that the residual aerosol layer from the prior day is present above the PBL. In this way the new PBL is mixed with the residual layer resulting in a much smaller dilution effect as expected from the PBL growth alone.
The influence of local sources (highway) may have affected the measurements, but not as stated in a largely way. If you estimate a local source on the scale of 100 m with even 10 times higher aerosol extinction than average, then after our entire measurement path of 6 km the overall extinction is only increased by 15%. And as stated before the hygroscopic enhancement we are interested in is on the order of 200-500%.

But you are right, the argumentation was missing in the manuscript. We include your remark in our discussion:
At Section 3.3:
“*The calculation of the extinction enhancement factors relies on the assumption that the initial air mass, and more specifically the dry aerosol extinction, is constant throughout the measurement while the relative humidity changes. For the calculation we excluded all days with precipitation to exclude wet depositional loss or days with a distinct change of the air-mass origin during a measurement. We also had to exclude all measurements with visibilities less than the optical path length and days with no significant aerosol load (clean days, bext < 0.05 km-1).*

Secondary aerosol production, advection of aerosol from local sources to the site or an air mass with lower concentration, temperature-driven partitioning of ammonium nitrate (e.g. Morgan et al., 2010) and of semi-volatile material (e.g. Donahue et al., 2006), and boundary-layer dilution can force this assumption to fail. Accompanying in-situ measurements of the extinction coefficient by the dried aerosol would be one choice to provide a dry reference. However, in Leipzig such data were not available for the long-term period investigated here. Therefore, in a first step we have analyzed the backward trajectories to ensure a constant air-mass origin during the measurement. Secondly, the time periods we used to quantify the dominant optical-enhancement process where usually no longer than four hours. For the effect of boundary-layer dilution it was often found from lidar measurements that the residual layer from the prior day is still present in the morning above the nocturnal inversion layer. The turbulent PBL growth process then mixes the residual layer downwards while the surface aerosol is mixed upwards. Hence, statistically the net dilution effect is smaller than expected from PBL growth alone so that in a lower extreme considering a negligible nocturnal aerosol production at the surface and no deposition of aerosol from the residual layer the dilution effect could even be nonexistent. On average, the possible uncertainties given by the reasons above are still small (on the order of 20-30% throughout a measurement) compared to extinction enhancement be relative humidity (on the order of 200-300%). With the given preconditions we were able to select 143 days out of our 4 year data set in order to derive the extinction enhancement factor on a statistical basis. The main results of the analysis are summarized in Fig. 6."

During days with a high variability in air temperature, the diurnal cycle of aerosol (optical) properties will also be influenced by partitioning processes e.g. of ammonium nitrate (see e.g. Morgan et al., 2010) or semi-volatile organics (see e.g. Donahue et al., 2006). These factors and the course of the PBL will clearly influence the curvature of the recorded and selected humidograms (and thus the apparent hygroscopicity parameter as discussed here).

Of course, such processes can occur. We tried to argue on this in the paragraph included above. Still, we believe the by far dominant process in our recorded humidograms is the hygroscopic growth. On average the variability of the dry aerosol extinction is much smaller.

To retrieve reliable enhancement factors, the authors have to relate and normalize their extinction measurements to a second independent measurement (e.g. using the particle size distribution or the in-situ particle light extinction coefficient) or at least introduce an appropriate dilution factor.

You are absolutely right. Normalization would be the ideal approach. A good method would be probably be using an aerosol inlet with a dryer together with a CAPS instrument. Unfortunately such instrumentation was not available at TROPOS during the course of our study. Measurements of particle size distributions would be another approach, but then uncertainties of unknown refractive indices also would bring errors to our "baseline". However, there is always the possibility that the volatile particle mass, as well as the coarse particles, can vanish drying process. Considering these limitations our approach was to carefully select days with unchanged air masses, which resulted in the remaining 143 cases.
In our outlook we include: *"In this study, we had to rely on a persistent dry particle extinction coefficient while the ambient humidity changed. Various measures were taken to ensure this precondition. For future studies however, we intend to use co-located in-situ measurements to not only ensure but to directly measure a dry baseline. In this way, more valid cases of hygroscopic extinction enhancement could be obtained from a measurement campaign."*

In its current state, the given parametrization is highly questionable and thus not very useful to the reader and scientific community. The authors have to thoroughly revise their method (and manuscript) to prove the validity of their method, which is unfortunately not given at the current state. This includes a substantial improvement of their method, result and discussion section. It is for the reason that I suggest major revision of the manuscript.

In the point of a highly questionable parameterization we disagree. We tried to explain the method and the limitations more thoroughly but our results of e.g. Fig. 9 show a very good performing parameterization. Also, from Fig. 6 we find a mean extinction enhancement factor of 2.41 (at 85% RH and 550 nm, with a range from 1.5 - 4.4) which is consistent with previous findings for the Leipzig region (e.g. of the scattering enhancement by Zieger 2014). Of course, a more solid foundation of a "dry baseline" is favorable and will for sure be used in upcoming studies.

2 Specific comments

There are further major concerns (besides the major one described above) which should be carefully regarded. The following comments are given in arbitrary order.

Page 12585, Line 22-25: There is actually a high number of publications on that topic (incl. very recent ones). Therefore, the authors should conduct a more thorough review and discussion of the literature; at the very least more recent results from Melpitz should be discussed given its vicinity to the sample station used during this campaign (Zieger et al., 2014). Zieger et al. (2014) measured the scattering enhancement directly during the winter months of 2009, which overlaps with the measurements discussed here. Their enhancement factors were significantly higher (median of 2.78 at 85% and 550 nm) than the values presented here. These observations, although made using different techniques, can be directly compared since the RH-related scattering enhancement is larger than the absorption enhancement, and the scattering coefficient exceeds largely the absorption coefficient. Therefore I strongly encourage the authors to have a second look at this work and discuss potential agreement/differences.

Yes, we are aware of the study by Zieger et al. Melpitz is the rural background measurement site of TROPOS. But especially close to the surface there can be significant differences of the aerosol between Melpitz and the city of Leipzig. So differences can be expected. However, we do not find the mentioned disagreement of the two methods. In Fig. 6 of our study (mean enhancement factor) we find the extinction enhancement to be 2.41 at 85% RH which is close to the previous finding of 2.78. Indeed, here it seems astonishing, how well (on average) the in situ and SAEMS techniques agree in terms of the enhancement factor. We will include the discussion in our text**.**

• Page 12587, Line 2: Unfortunately, the influence of sea spray can not be regarded as low at this site (Spindler et al., 2010) and it also affects the optical properties (incl. hysteresis effects, see Zieger et al., 2014). This sentence is also in contradiction with the authors statement made below (page 12593, line 16-18:
”The largest value of 3.5 was observed for northerly air flows with the comparably largest influence of marine particles (at comparably low levels of pollution advection from the Baltic Sea and Scandinavia).”). Please revise and discuss appropriately.

You are right. We will include the following sentence in our discussion:
P12587L2:
Replace “The influence of marine particles can be regarded as low.”
By:
Although the cases with north and northwesterly flows reaching Leipzig from marine regions are very common continental and local aerosol sources still dominate the particle fraction in Leipzig. However, the occurrence of marine particles at the site cannot be excluded in general (see: Spindler 2010, Zieger 2014).”
Hysteresis on the other hand we did not observe with our method, as it is very unlikely to observe and recrystallize pure marine particles at ambient RH which occur in Leipzig. Such a hysteresis effect would probably always be masked by a mixture of particle species, by turbulent processes, and by much longer time spans during which the aerosol is conditioned.

• Eq. (3) and Eq. (4): It is not clear, if Eq. (3) is only used for the low RH range (RH< 70%), while Eq. (4) is used for the range above 70% RH throughout their analysis (see statement on page 12588, line 14). If so, then it has to be clearly described and discussed. In this case, many of the statements in the result section (incl. the given parametrization and the figures) need to be revised to avoid this ambiguity (would be the parameter for the low and intermediate RH range and the c-values representative for the high RH range).

Hänel introduced the second equation (Eq.4, c1,c2) in order to describe the curvature more exactly, because the slope could be adjusted. For the RH range below 70% he used Eq.3. For RH>70% Eq.4 (c1, c2) was used. Eq.5 is then the mandatory requirement to match both equations at 70% RH.
We followed the reviewer’s suggestion from below to only rely on the gamma equation (Eq. 3) in our updated manuscript. After thoroughly considerations we found it not useful to fit the humidograms with three unknowns (b(f=0), c1, c2) because in contrast to in-situ measurements the dry extinction (RH<30%) is not always available in our ambient data. Then, b(f=0) and c1 cannot be independently determined. Also a combined fit (Eq.3 for RH<70% and Eq.4 for RH>70%) did not give satisfactory results as artificial discontinuities of the slope can arise at 70% RH.

• The factor of 0.3 in Eq. (5) is probably referring to the dry reference RH of 30%. However, all the following results are later given at 0 %. Does this assumption have an effect? In addition, if entering the coefficients given below, the results would be = 0.4413 and not 0.4364. Please clarify.

The factor 0.3 does not refer to the dry reference of 30%. This factor is a mandatory requirement in Haenels formula in order to define the transition at 70% RH between the two formulas. Haenel defined the gamma formula up to 70% RH and the c1-c2 formula for humidities above 70%. However, we will only use the c1-c2 formula to plot and compare to Haenels original parameterization in Fig 6.

• Sect. 3.3: How exactly were these 143 days out of the four years of data selected? What criteria were applied and how many days were neglected?

Reviewer 2 had a very similar comment. The selection was carefully performed as stated in the beginning of our response letter. Only these days with a significant change of R.H. within a certain time and no hints on air mass changes were selected for our study. Of course, many data had to be discarded in this way as it is often the case for ambient observations. We now state the selection criteria above and in the text.

The fitting procedure is described (and maybe performed?) in a careless way. As it reads now, both Equations 3 and 4 were fitted separately to the recorded data, however, if the mean values of c1 and c2 are then inserted into Eq. 5 a value of = 0.2732 is retrieved, while the second fit gave = 0.464 as stated in Fig. 6, which is almost a factor of two different. Why?

We cannot see how the reviewer get the value of 0.732. We have used the following Eq. with our values: 0.8329 – ln(0.6417)/ln(0.3) = 0.464.
Regardless of this discussion, we removed c1 and c2 from the manuscript, as suggested by the reviewer.

Ignoring for a moment all the major limitation mentioned here: Would it not be totally sufficient to just use Eq. 3 (with and the intercept as the only free parameters)? The separation as derived by Haenel (1984) probably can't be made since the measurements were not performed in an controlled environment. Haenel (1984) is an empirical parametrization and many other parameterizations were used later on within the literature. Therefore I would suggest to only use one parametrization (i.e. Eq. 3).

You are right. Although Eq. 4 has one degree of freedom more to describe the curvature better than Eq. 3, we decided to provide only the gamma values as suggested. We agree that this parameter is much easier to handle and often used in the literature. The matter was resolved as described above.

• Sect. 3.4: How many days backwards in time were the trajectories being used? What criteria led to exactly eight clusters? Has the residence time within the PBL (as the main source region of aerosols) being considered for the analysis?

We calculated four-day backward trajectories for all cases. At first we obtained 12 typical clusters (also including slow and fast transport regimes towards Leipzig) but we combined several of the clusters for which no significant differences were found for mean extinction and mean gamma values. Only for cluster 6 (west, slow, more anthropogenic pollution) and cluster 7 (west, fast, more probable Atlantic air masses) we still discriminate because of the different mean ambient extinction. Thus, the residence time in the PBL was not specifically analyzed but was originally reflected in the trajectory velocity.
We will include:
"3.4.... based on 4-day HYSPLIT backward trajectories…."
"The cluster analysis revealed eight significant ~~different~~ air flow regimes for which different optical properties were obtained."

• Page 12592, Line 2 and Fig. 7: Please add to the figure (for the ambient values) the mean RH for each sector.

We had a look at our data and found no significant RH differences. We will now add to the text:
The ambient RH for our measurements was found to be 65% on average for each cluster with a standard deviation of 15% RH within each cluster. The mean differences between the clusters RH were found to be low (max. 5%) with the maximum of 70% RH for cluster 3 and the minimum of 63% RH for cluster 4. Figure 7 presents ..."

• Page 12592, Line 5: Were the mean values of c1 and c2 from the entire campaign or for each cluster separately being taken? Would it give the same results if the mean cluster values of (Fig. 9) was used?

Yes, as it was written in the text ("Figure 7b shows the cluster mean extinction values after normalization to 0 % relative humidity by using cluster mean values for c1 and c2.") the parameterizations used here were taken from each cluster separately. Now that we changed only to the gamma parameterization, it can be stated that the clustered gammas from Fig. 9 (now included in Fig 7, see later comment) where used for each of the data points summarized in Fig 7a to derive Fig 7b by the individual relative humidity.
We now write: "*Figure 7b shows the cluster mean extinction values after normalization of the individual data points to 0 % relative humidity by using the derived cluster mean values for gamma (Fig. 7c) and the respective RH of each data point.*"
Also we changed Figure 7 caption: *".., (b) same as (a), except prior to averaging all individual cases were normalized for dry conditions (f=0 %) by use of the derived cluster mean parameter gamma (c), and (d) same as (b) but separately for …"*

• Page 12593 and Fig. 8: It is surprising that the pattern of the enhancement factor changes when going from 80% to 95% RH. This is somehow counterintuitive given the monotonic increasing function shown in Fig. 6. Are these the same datasets or why for example is suddenly sector 6 at 95% RH below sector 7 and 5, while it showed larger values than the two at 80%? If I use the –values given in Fig. 9, the result for the enhancement factors are much different than presented in Fig. 8. I could imagine that the authors used the c-values to derive Fig. 8, however these values should be consistent with the results using the - values. This probably relates to the fact that the coefficients given in Fig. 6 are not consistent or that the coefficients are valid for different RH-ranges. Please clarify.

Yes you are right, the explanation of the Figure 8 was probably misleading or incomplete. For Figure 8 we only selected cases where the ambient RH indeed changed from 40 to 80% and from 40 to 95% within a short period of time. No parameterization was involved. Of course these cases can be from different observational days. The figures intention is to show that up to 80% RH the extinction enhancement is rather similar on average while for higher RH significant differences are found for the sectors. Logically this figure should probably be located before the parameterization analysis, but then the clusters were not introduced. So we decided to place it after Fig 7.
However, we will include the number of observed cases in each bar to make more clearly, that only observed data are used in this figure.
Also we change: *"Figure 8 provides an overview of the mean particle enhancement factor (and corresponding SD) for the different airflow clusters. The shown mean values and SD of the ratio of particle extinction at 80 or 95 % relative humidity to the one at 40 % relative humidity were calculated from the available individual days with strong humidity variability for each of the eight air flow regimes separately. Nine (south cluster) to 29 days (northwest cluster) were available for the cluster-related investigations."*
To: *"Figure 8 provides an overview of the mean particle enhancement factor (and corresponding SD) for the different airflow clusters. The shown mean values and SD of the ratio of particle extinction at 80 or 95 % relative humidity to the one at 40 % relative humidity were directly calculated from the available individual days with strong humidity variability (either from 40 to 80% or from 40 to 95% RH, respectively) for each of the eight air flow regimes separately. Because of the larger required RH span in ambient conditions Fig 8a includes additional observational cases with respect to Fig 8b."*

Fig 8 caption from: "*Particle extinction enhancement factor (four-year mean value and SD), for (a) 80-to-40% RH enhancement, and (b) 95-to-40 % enhancement for the eight air mass transport regimes."*
To: *"Particle extinction enhancement factors (80-to-40 % and 95-to-40 % RH enhancement) observed from days with occurring humidity variations between at least 40 and 80 % RH (a) and only from days with variations between at least 40 and 95 % RH (b) separated for the eight air mass transport regimes. Four-year mean values and SD are given.*"

• Page 12591, Line 24: Are the 18 000 single observations identical with the 143 selected days? What is the time step for each observation?

The 143 selected days are only a small part of the 18000 single observations of our dataset. We take one measurement every 25-30 minutes depending on the automatically adjustment process (Skupin et al 2014). The 143 days however are only the days we could use to obtain a good parameterization. The 18000 observations stem from all available measurements of >800 days.

• Were any seasonal trends or monthly patterns in the enhancement factors observed? This discussion would be interesting, since partitioning effects, differences in aerosol emissions and the PBL development probably caused a clear seasonal variation.

This is an interesting suggestion. However, our data amount is strongly limited by the requirement of a humidity cycle while the dry aerosol extinction remains constant. The majority of the 143 cases are in spring/summer/autumn and we have only a few cases in the winter season. In winter there was often a high relative humidity and no significant RH cycle. Thus we were not able to analyze a trustworthy seasonal trend of the enhancement parameters.
A seasonal cycle in the extinction coefficient and the Angström-Exponent was observed (probably also determined by different mean ambient humidities) but this topic was not intended to be part of this publication.

• Page Page 12593, Line 1-3: This statement is too speculative. Are the authors really sure that a regulation introduced in 2011 already shows an effect the year later? What exactly has been regulated?

On 1 March 2011 the so called Environmental Green Zone was implemented in Leipzig in order to meet the EU regulations on particulate matter. Vehicles which didn't meet the requirements were banned from the city. As a result the effects were immediate, especially on soot emissions.
From: "In contrast, in 2011 new traffic emission reducing regularizations were brought into operation and may have caused the overall low particle extinction values observed in 2012."
To: "In contrast, on 1 March 2011 the Environmental Green Zone restriction were brought into operation in Leipzig to meet the European Union's regulation on particulate matter to ban vehicles which didn't meet certain requirements from the city. This implementation may have caused the overall low particle extinction values observed in 2012."

• Sect. 3.5 (Comparison to in-situ measurements): As a kind of validation, the authors present a 5-day comparison (as a timeline) to Mie calculations using the measured particle size distribution. However, this is described in inadequate detail and more needs to be added here.
– With what kind of instrument has the particle number distribution been measured?
What was its upper size limit?

The in-situ data at TROPOS were taken with TDMPS and APS. The combination of both instruments covers particles with diameters of 3 nm to 10 µm.
After "*A so-called PM 10 inlet is used so that very coarse particles with diameters larger than about 10 µm are not measured.*" we add to the text: "*The particle size distributions were measured with a tandem differential-mobility particles sizer (TDMPS, 3 – 800 nm in diameter) and with an aerodynamic particle sizer (APS, 0.8 – 10 µm in diameter). The in-situ data we used for this study are 1-hour averages.*"

– How has the coarse mode (above 1 micron) in the Mie calculations been treated?

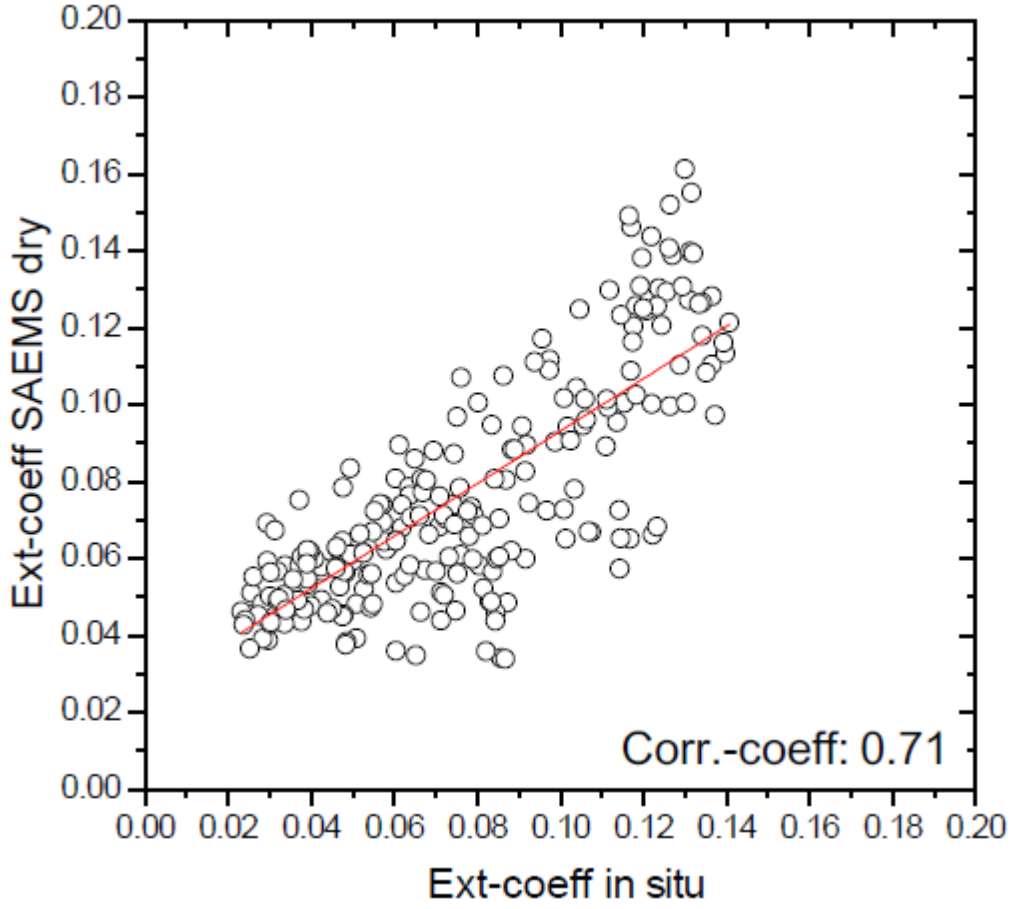
Yes, we are aware that the coarse mode particles can have different refractive indices. But a sophisticated Mie calculation was not possible for us and is also not in the major focus of our manuscript, so we applied a Mie code with a constant refractive index for all particle sizes. For a more elaborated way a continuous chemical analysis would be needed in addition in order to fix the refractive index. What is not shown is that we tested a range of reasonable refractive indices and found that the typical value of (1.53+0.01i) was sufficient for our cause. Also a mixing rule for the refractive index of aerosol and water based on the hygroscopic growth described by the Kappa-Köhler theory was tested. But again, the uncertainties of dry in-situ data of the refractive index are small compared to the hygroscopic enhancement process which we focus on.

– It is not clear how long exactly the in-situ measurements were performed.
Please clarify.

The continuously archived in-situ data we had available were 1-h averages. See above.

– Please show a scatter plot of the entire comparison of the (dry) extinction coefficient and discuss the degree of agreement (incl. regression line and statistical parameter). A nice looking example as a time series is not enough to judge on the overall performance.

The scatter plot below of the selected period (former Fig. 10) shows the correlation (Pearson correlation coefficient 0.71) of dry extinction coefficients based on in situ and SAEMS data. The discrepancies have several causes. At first we calculate the extinction coefficient with an average refractive index without any knowledge of the chemical composition. In situ and ambient measurements are based on different average times.



– Why were the results not directly compared to the long-term records of (dry) optical properties measured in situ at the Melpitz site, which is close to Leipzig and run by the same institute (as I believe)? This comparison would definitely help to strengthen the message of this work and partially clear out the strong doubts brought up within this review.

You are right, Melpitz is a long-term site of TROPOS, about 50 km northeast of Leipzig. But it is a continental background station and hence significantly cleaner than the city of Leipzig. Therefore it would also be questionable to compare our measurements from TROPOS to those of Melpitz. Of course, a SAEMS instrument in Melpitz would be favorable. For the future it is planned to install a mobile version there, indeed.

• Sect. 5.3 (Comparison to AERONET):
– It is not clear why the differences in wavelengths has not been corrected for. Please use the Angstrom law (Eq. 1) and transfer the AERONET measurements to 550 nm. Secondly, the reviewer wants to see a scatter plot with a linear regression line and statistical values. Figure 11 is to the reviewer's opinion not sufficient to show a direct comparison.

Yes, the Angström transfer of the AERONET values to 550 nm was also mentioned by the other reviewer. We changed this in the manuscript and explained the calculation briefly.

Text change from: "*In the case of the AERONET observations, the extinction distribution curve shows PBL mean extinction values (vertical column mean values). All measured 500 nm aerosol particle optical thickness (AOT) values were divided by the respective PBL height, obtained from numerical weather prediction data (GDAS:..".…*

To:*"In the case of the AERONET observations, the extinction distribution curve shows PBL mean extinction values (vertical column mean values). First, we converted the measured 500-nm particle extinction with the Angström-Exponent (500-870nm) to 550 nm wavelength by Eq. 1. Then, all calculated 550nm aerosol particle optical thickness (AOT) values were divided by the respective PBL height, obtained from numerical weather prediction data (GDAS: ..."*

...

"As can be seen in Fig. 11, a rather good agreement between the SAEMS (ambient) and the AERONET observations is found. A systematic overestimation of the PBL mean extinction value must be kept in consideration in the interpretation of the AERONET observations, because, on average, 20\,{\%} of the AOT is caused by particles in the free troposphere (Mattis2004). ~~*We did also not correct for a wavelength dependence of particle extinction. On average, 550 nm extinction coefficients are about 10–15 % lower than the values at 500 nm."*~~

For Figure 11 we cannot derive a meaningful scatter plot as suggested because the values taken for SAEMS, in-situ, and AERONET are not matched in time. AERONET only measures during daytime and only under cloud-free conditions and with unspecific time resolution. In-situ data are taken from all days but only from 13-17 UTC (daytime like AERONET, only for mixed PBL). SAEMS data from all measurement days (no rain, no system downtime, 13-17 UTC). So it is expected that the statistical results are similar, but individual comparison (scatter plot) is not possible for these data.

– What is the average ratio (mean and SD) of the AERONET and SAEMS ambient extinction coefficient exactly? Can it be fully explained by particles above the PBL?

We will add the values in the text: "*The mean extinction coefficients and SD for 550 nm is 0.12±0.09 km^{-1} (AERONET) and 0.11±0.0.06 km^{-1} (SAEMS)* ."(Graphs are shown in former Fig.11). We did recalculate the extinction coefficients for AERONET data with Eq.1 to get the 550-nm value.

– Why was the PBL height retrieved from the lidar not being used or (in another way) how does the PBL height given by the model compare to the continuous lidar measurements?

For our long-term dataset continuous lidar data are not available in 2009 because the autonomous lidars were used in campaigns elsewhere. Baars et al. (ACP, 2008) compared lidar derived PBL with model data. It was found that at least for the fully developed PBL (13-17UTC) the model data are in good agreement with reality. In a statistical sense the model slightly underestimated the maximum PBL height, but not more than 20%.

• Sect. 3.6: The spectral dependency of the extinction coefficient shown in Fig. 13 clearly indicate a decreasing Angstrom exponent with increasing RH, however the discussion of the Angstrom exponent using two pairs (390/440nm and 390/881 nm) is not very convincing.

We hope that the discussion becomes clearer with the additional text from below with the argument of: One explanation would be that the coarse mode doesn't grow and keeps the 881 nm extinction constant (Fig 2). At the same time the small particles do grow which increases the 380 and 440 nm extinction.

How did the in-situ/AERONET comparison look for the 390 and 881 nm? The 390 and 440nm are influenced by the absorption of NO_2, which might be relevant when considering that the instrument measured across a large highway.

We didn't calculate the dry in situ Angström exponent. There are assumptions to made to compare the dry particle size fraction to spectral ambient extinction measurements. In case of the AERONET Angström exponent we did some case studies and found a larger Ängström exponent for these AERONET measurements.
We didn't correct our spectral measurements for NO2 for the broadband absorption spectrum in the 250-650 nm spectral region with the maximum absorption cross section at 414 nm. The large highway covers only approx. 1% of the measurement path and the overall influence is considered as low (< 2%) for 390 and 440 nm.

How was this corrected for? Could there also be a calibration or technical (temperature dependent?) issue of the red channel? The results as presented now are largely based on small variations of the 881nm measurement where the relative uncertainties are higher. I would suggest to use a numerical fit to determine the Angstrom exponent and repeat/improve the discussion.

You are right. The red channel had some difficulties with strong water vapour absorption bands. They might influence the spectral measurement. For this reason we looked at every measurement and selected the spectral data carefully. In future studies we will use a spectrometer with a 20-times higher resolution (than the spectrometer in this study , 10 nm) to get better information about the particle extinction coefficient in the red channel.

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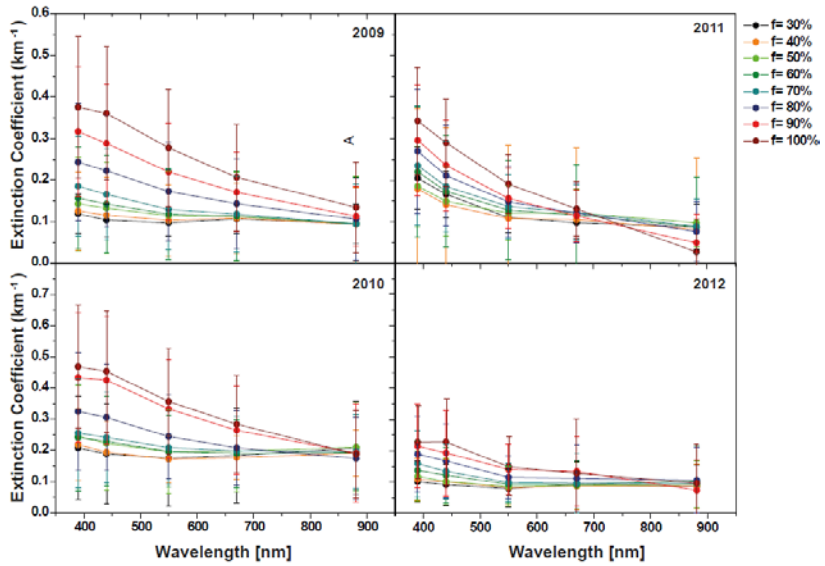
• Page 12596, Line 25: Why were the following two years not included?

The period of 2009 and 2010 was the focus of the PHD work. In these two years SAEMS was constantly monitored and observed for good performance. While the 550nm measurements are very robust, especially the measurement of spectral data required more attention and manual data evaluation. Hence, the quality of this 2-year dataset is higher than for the 2011 and 2012 data. Therefore we show only 2009 and 2010 here in this final paragraph. A quick analysis with automatic data analysis for 2011 and 2012 showed very similar results (see graph below) but these spectral data are not quality checked and therefore not shown in the manuscript.



• Page 12597, Line 5: I would soften the discussion on non-hygroscopic coarse particles here, since hygroscopic sea spray particle can't be fully excluded (see comment above).

We changed the discussion in the following way:
"The impact of fine-mode particles on the extinction coefficient at 881 nm is low. In contrast, the coarse particles (road dust and others) obviously do not grow significantly by water-uptake so that the extinction coefficient at 881 nm, dominated by large particles, remains low for all ambient humidity conditions. Consequently, the overall 390–881 nm Ångström exponent increases with relative humidity."
"Furthermore, the impact of fine-mode particles on the extinction coefficient at 881 nm is low. At this wavelength the extinction coefficient is primarily determined by larger particles. Although coarse-mode sea-spray particles cannot be fully ignored in Leipzig, most of the time the coarse mode consists of road and soil dust particles which do not grow significantly by water-uptake. As a consequence, the extinction coefficient at 881 nm might remain constant for all ambient humidity conditions (c.f. Fig. 2) while the 390-nm extinction coefficient increases by fine-mode particle hygroscopic growth. Consequently, the overall 390–881 nm Ångström exponent might also increase with relative humidity."

• Page 12586, Line 21: It is here (and further down) often referred to the PhD thesis of the main author ("The full set of analysis results can be found in Skupin (2014)."), also for critical points ("More case studies and more details to the parameterization efforts can be found in Skupin (2014)", line 18, page 12591). Unfortunately, this thesis is written in German and thus not accessible to the majority of the scientific community. Please list all needed information to the reader in the revised manuscript and state in the reference list that this is a thesis written in German.

Yes, the thesis is written in German and can be found at the TROPOS website (http://lidar.tropos.de/publikationen/dissertation_skupin.pdf). We will indicate this in our reference list.
With respect to the parameterization routine we will add to the end of section 2:
Since for remote sensing in ambient conditions it is not possible in general to observe a dry extinction coefficient b(f=0). Without such a known value a fit with Eq. 4 becomes problematic because b(f=0) and c1 would be dependent on each other. Therefore for further statistical investigations we relied on Eq. 3."

• Page 12585, line 18: The wording "manipulation" is not appropriate here since it implies a certain willful intention and the authors should bear in mind that all measurement techniques in aerosol science have certain drawbacks. I suggest to replace it with "introduction of certain potential measurement artifacts".

Manipulation was never meant to state that there was a willful intention. It simply has the meaning of handling, management or treatment. Maybe it is a misunderstanding as in German language Manipulation" indeed has a very negative touch. We will change:
"However, it is not a simple task to accurately determine the volume extinction coefficient for a given aerosol scenario without any manipulation of the aerosol system. Such a manipulation cannot be avoided when aerosols are sampled and analyzed by means of in situ measurement techniques. Only remote sensing methods are able to avoid the disturbance of the aerosol conditions to be measured."
To:
"However, it is not a simple task to accurately determine the volume extinction coefficient for a given aerosol scenario without any alteration of the aerosol system. The introduction of certain potential measurement artifacts can occur when aerosols are sampled and analyzed by means of in situ measurement techniques. In contrast, remote-sensing methods are able to completely avoid the disturbance of the aerosol conditions to be measured but as a drawback these methods always rely on ambient conditions and careful case selection."

• In general, the authors should tone down their language. Statements like "manipulation" (see comment above), "corroborates the usefulness" (page 12595, line 5), "corroborates the high quality and reliability of our long-term observations" (page 12591, line 17), "year-by-year differences are also obvious" (page 12592, line 27), "nicely shows" (page 12595, line 8), "obviously do not grow" (page 12592, line 6) are often empty and subjective statements without any statistical backbone. Real numbers would be more useful.

Thank you for pointing this out. We will modify the passages.

• There are a lot of figures in the current manuscript and there is some space for improvement.
– Figure 3: This figure could be merged with Fig. 10, where one example RH time line is already shown.
It is unfortunately not possible to combine Fig 10 and 3 as the additional RH sensors were not available before 2010. Within our study we had to use different sensors from time to time. Fig. 3 is supposed to show typical RH variations at our site and that the use of different sensors can help analyzing the homogeneity along our measurement path.

– Figure 4: This figure can be omitted. Not much is learned here and the numbers can be given in the text.

The figure shows the log-normal distribution of all extinction values measured for this 4-year campaign and the distribution after dehumidification by our parameterization. Since this is one major statistical finding we want to keep this figure. Later on, the comparison to other methods (former Fig 11) only includes the year 2009 where we had the comparison data available.

– Figure 5: Please add the confidence intervals to the fit parameters in panel c and f. As mentioned above, I believe it is fully sufficient to just use the -parametrization as a one-parameter fit for the humidograms. Otherwise the authors should justify why the c_1 and c_2 are needed.

Yes, as suggested, we only used the gamma parameterization and now added the confidence intervals for gamma.

– Figure 6: Please reduce the fit-coefficients (SD) to their significant digits.

Sure, we reduced the number of digits to two. Note, the SD represents the variations between the 143 fits, not the uncertainty. Therefore we keep 0.46 as an average now, as 0.3/sqrt(143) = 0.02.

Why is Eq. 5 not fulfilled here (see comment above)? #

Since we now only use the gamma fit, the comment is not applicable.

– Figure 7 and 9 could be merged into one since they are related as a result of trajectory analysis. The bar plots (panel c of Fig. 7) could be omitted, not much is learned here and a sentence in the text would be sufficient.

We combined both figures into a 4-panel graph. However, we did not want to skip the dehumidified extinction values in their annual statistic (bar graphs). They show on the one hand that the mean dry extinction values in different years can vary by a factor of two. On the other hand, the inter-annual consistency of more polluted vs. less polluted transport regimes is clearly seen.
The text passages were sorted accordingly.

• For RH ! 100% the enhancement factor goes towards infinity (Eq. 3 and 4), but@ the authors show in Figure 13 the extinction coefficient for it (red curve). Please clarify.

You are right, for 100% RH the parameterization reaches infinity. In reality, droplet activation takes place, fog forms, and the extinction grows to a much larger, but finite value. However droplet activation is not treated in this study, such cases were excluded. We focus on hygroscopic growth. Also because of measurement limitation by the path length our maximum observable extinction coefficient is approx. 1 km^{-1}. And finally, we used different humidity classes (Δf=10%) for this plot. To clarify this we will change the legend of Fig 13 to:
"f=<30%, f=30-39.9%, …, f=80-89.9%, f= 90-99.0%"

3 Technical corrections
• Please add the respective wavelengths to all the figures where needed.

Yes, we added the wavelength in the Figure captions where needed. Most of the time it was 550 nm.

• Figure 9: Please replace "extinction-enhancement-describing parameter" by "hygroscopic exponent" to be consistent with the text.

Done.

• Table 1: Please replace 'Norge', 'Suisse' and 'anthropogen' by its correct English words. Also add the wavelengths to the literature values.

Thanks! Of course we changed these words to *Norway, Switzerland* and *anthropogenic*

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Four-year long-path monitoring of ambient aerosol extinction at a central European urban site: dependence on relative humidity

A. Skupin, A. Ansmann, R. Engelmann, P. Seifert, and T. Müller

Leibniz Institute for Tropospheric Research, Permoserstraße 15, 04318 Leipzig, Germany

Correspondence to: A. Skupin (skupin@tropos.de)

Abstract. The ambient aerosol particle extinction coefficient is measured with the Spectral Aerosol Extinction Monitoring System (SÆMS) along a 2.84 km horizontal path at 30–50 m height above ground in the urban environment of Leipzig (51.3° N, 12.4° E), Germany, since 2009. The dependence of the particle extinction coefficient (wavelength range from 300–1000 nm) on relative humidity up to almost 100 % was investigated. The main results are presented. For the wavelength of 550 nm, the mean extinction enhancement factor was found to be 1.75 ± 0.4 for an increase of relative humidity from 40 to 80 %. The respective four-year mean extinction enhancement factor is 2.8 ± 0.6 for a relative-humidty increase from 40 to 95 %. A parameterization of the dependency of the urban particle extinction coefficient on relative humidity is presented. A mean hygroscopic exponent of 0.46 for the 2009–2012 period was determined. Based on a backward trajectory cluster analysis, the dependence of several aerosol optical properties for eight air flow regimes was investigated. Large differences were not found indicating that local pollution sources widely control the aerosol conditions over the urban site. The comparison of the SÆMS extinction coefficient statistics with respective statistics from ambient AERONET sun photometer observations yield good agreement. Also, time series of the particle extinction coefficient computed from in-situ-measured dry particle size distributions and humidity-corrected SÆMS extinction values (for 40 % relative humidity) were found in good overall consistency, which verifies the applicability of the developed humidity parameterization scheme. The analysis of the spectral dependence of particle extinction (Ångström exponent) revealed an increase of the 390–881 nm Ångström exponent from, on average, 0.3 (at 30 % relative humidity) to 1.3 (at 95 % relative humidity) for the four-year period.

1 Introduction

The importance of atmospheric aerosols in the global climate system due to scattering and absorption of radiation and the influence on the formation of clouds is well known (Charlson and Heintzenberg, 1995; Heintzenberg and Charlson, 2009). However, a realistic consideration of atmospheric aerosols in climate models and the quantification of aerosol-related climate effects is a rather crucial task, not only because of the high horizontal, vertical, and temporal variability of aerosol concentrations, but also as a result of the highly variable microphysical and chemical properties of the aerosols originating from many and rather different anthropogenic and natural sources. Furthermore, as a function of particle chemical composition, particle age, and state of aerosol mixture, aerosols can show a very different hygroscopic behavior (i.e., water uptake with increasing relative humidity), which further complicates the impact of aerosol particles on the Earth's radiation budget. There is a clear need for more field observations of ambient aerosol optical properties as a function of relative humidity from low (< 40 %) to very high values (> 95 %) to better describe aerosols in climate models as well as to better separate of aerosols and clouds in satellite remote sensing products. However, it is not a simple task to accurately determine the volume extinction coefficient for a given aerosol scenario without any affect on the aerosol system. Such an affect can not be avoided when aerosols are sampled and analyzed by means of in situ measurement techniques. In contrast, remote-sensing methods are able to completely avoid the disturbance of the aerosol conditions to be measured but as a drawback these methods always rely on ambient conditions and careful case selection.

Only a few publications are available for particle growth in high-humidity environments with relative humidities up to almost 100 %, before cloud droplet activation begins (Arnulf et al., 1957; Goes, 1963; Elterman, 1964; Goes, 1964; Ba-

dayev et al., 1975; Stratmann et al., 2010; Liu et al., 2011; Chen et al., 2014; Zieger et al., 2014). These efforts were partly based on controlled laboratory studies. Motivated by the need for more aerosol field observations with emphasis on undisturbed, but complex aerosol mixtures at ambient humidity conditions, we designed and setup the Spectral Aerosol Extinction Monitoring System (SÆMS) (Skupin et al., 2014) which allows us to continuously monitor the wavelength spectrum of the particle extinction coefficient at a height of 30–50 m above ground between two towers which are 2.84 km apart from each other. The measurements cover all seasons of the year. Simultaneously, relative humidity and temperature are recorded at both towers at the height level of the aerosol extinction measurement path. The most interesting days for our study are those with a strong change in relative humidity, e.g., from nearly 100 % in the early morning to 30–40 % later on during the day and correspondingly strong changes in the particle extinction coefficient.

In our first article, we described the Spectral Aerosol Extinction Monitoring System (SÆMS) in detail (Skupin et al., 2014), discussed the quality and uncertainties of the observations, and presented case studies to show the potential of the newly designed remote sensing facility. In this article, we summarize the main findings of our long-term observations which cover the four-year period from January 2009 to December 2012. Besides the study of the dependence of particle extinction on relative humidity, we provide a general overview of the four-year statistics of particle extinction coefficients. We further compare the statistics with simultaneously performed Aerosol Robotic Network (AERONET) photometer observations and the optical properties derived from in situ measurements of the dry particle size distribution close to the SÆMS instrument. A similar study was presented by Müller et al. (2006) based on a short-term data set measured at the Leibniz Institute for Tropospheric Research (TROPOS) in March 2000. Here we expand the study and compare the entire year-2009 observations. The full set of analysis results can be found in Skupin (2014).

2 Instrumentation and data analysis methods

The long-term SÆMS aerosol measurements are performed in a suburban environment about 3 km northeast of the city center of Leipzig (51.3° N, 12.4° E, 120 m a.s.l.) in the eastern part of Germany since the beginning of 2009 (Skupin et al., 2014). Aerosol conditions are dominated by anthropogenic pollution (gas, oil, benzin, and coal burning, biomass-burning smoke, road dust) and natural continental aerosols (soil dust). Although the cases with north and northwesterly flows reaching Leipzig from marine regions are very common continental and local aerosol sources still dominate the particle fraction in Leipzig. However, the occurrence of marine particles at the site cannot be excluded in general (see: Spindler et al. (2010), Zieger et al. (2014)). SÆMS is installed in the roof laboratory of the main TROPOS building with a dome on top, and free view in all direction. The system is fully automated and allows us to measure the particle extinction spectrum from the from 300 to 1000 nm. SÆMS is part of the Leipzig Aerosol and Cloud Remote Observations System (LACROS) (Wandinger et al., 2012; Bühl et al., 2013), which includes European Aerosol Research Lidar Network (EARLINET) lidars, a Cloudnet station consisting of a ceilometer, cloud radar, and microwave radiometer (Illingworth et al., 2007), and the AERONET sun/sky photometer (Holben et al., 1998).

The measurement principle is illustrated in Fig. 1. The radiation beam of a broad-band 450 W Xe-arc-high-pressure lamp is alternatively pointed to retroreflectors mounted at two towers at heights of 30 and 50 m above ground. The steering unit for light transmission and the receiving and detection units of SÆMS are mounted in the roof laboratory of TROPOS. The towers are 300 and 3140 m northeast of the TROPOS building. As explained in detail by Skupin et al. (2014) and Skupin (2014) the measurements allow us to determine the volume extinction coefficient $b_{\mathrm{p,e}}$ of particles along the horizontal path of 2840 m between the two towers. Figure 2 shows all extinction measurements for the 2009–2012 period for three different wavelengths as a function of relative humidity. The relative humidity (RH) as well as the air temperature (T) are simultaneously measured close to the retroreflectors at the towers as well as on the roof of the TROPOS building. Figure 3 shows an example of a week-long time series of relative humidity, measured at the different sites. We use the total set of meteorological data (measured at all three locations) to check the homogeneity of the air mass along the SÆMS beam.

In this article, we concentrate on the influence of relative humidity on the optical properties, and briefly introduce several quantities used in this context. Following the notation of Skupin et al. (2014), the Ångström exponent (Ångstrom, 1964), which describes the spectral dependence of the extinction coefficient, is defined as

$$\alpha(\lambda_1,\lambda_2) = -\frac{\ln[b_{\mathrm{p,e}}(\lambda_1)/b_{\mathrm{p,e}}(\lambda_2)]}{\ln(\lambda_1/\lambda_2)} \tag{1}$$

with the particle extinction coefficient $b_{\mathrm{p,e}}(\lambda_N)$ for wavelength λ_N.

The particle extinction coefficient $b_{\mathrm{p,e}}(\lambda)$ increases with relative humidity. We consider this by introducing the humidity parameter f_1 with, e.g., $f_1 = 0.8$ for 80 % relative humidity. The so-called extinction enhancement factor $b^*_{f_1,f_0}(\lambda)$ is defined as:

$$b^*_{f_1,f_0}(\lambda) = \frac{b_{\mathrm{p,e}}(\lambda,f_1)}{b_{\mathrm{p,e}}(\lambda,f_0)} \tag{2}$$

which describes the increase of the particle extinction coefficient at $f_1 > f_0$ with respect to the dry-particle extinction coefficient at, e.g., $f_0 = 0.4$. Following Hänel (1984) with

focus on anthropogenic pollution (mixture of urban haze and rural background aerosol), we can describe the dependence of particle extinction on ambient relative humidity conditions by means of:

$$b_{\mathrm{p,e}}(\lambda, f_1) = b_{\mathrm{p,e}}(\lambda, f_0 = 0)(1 - f_1)^{-\gamma}\,. \quad (3)$$

Hänel (1984) introduced an extension for the high humidity range ($0.7 < f_1 < 0.99$) as follows:

$$b_{\mathrm{p,e}}(\lambda, f_1) = b_{\mathrm{p,e}}(\lambda, f_0 = 0)c_1(\lambda)(1 - f_1)^{(-c_2)}\,. \quad (4)$$

The empirical parameters c_1 and c_2 are related to γ according to

$$\gamma = \left(c_2 - \frac{\ln c_1}{\ln 0.3}\right). \quad (5)$$

For urban aerosols, $c_1 = 0.7008$ and $c_2 = 0.7317$ for 550 nm so that $\gamma = 0.4364$ after Hänel (1984). Since for remote sensing in ambient conditions it is not possible in general to observe a dry extinction coefficient $b_{\mathrm{p,e}}$(f=0). Without such a known value a fit with Eq. (4) becomes problematic because $b_{\mathrm{p,e}}$(f=0) and c_1 would be dependent on each other. Therefore for further statistical investigations we relied on Eq. (3).

3 Results

3.1 Overview

Figure 4 provides an overview of the particle extinction conditions at Leipzig. Shown is the frequency distribution of measured 550 nm ambient extinction coefficients (top panel) and, for comparison, the extinction frequency distribution after normalization of all values to 0 % relative humidity (bottom panel) by using Eq. (3) and appropriate input parameter γ discussed below. The 2009–2012 mean values and standard deviations (SD) are $210 \pm 170\,\mathrm{M\,m^{-1}}$ for ambient conditions and $110 \pm 80\,\mathrm{M\,m^{-1}}$ for dry aerosol conditions. Thus the particle water content is responsible for roughly 50 % of particle extinction in the lowermost part of the troposphere at this urban site. Mattis et al. (2004) analyzed the Leipzig EARLINET Raman lidar observations conducted from 2000–2003, and found a mean extinction coefficient for 532 nm wavelength and ambient humidity conditions of $94 \pm 50\,\mathrm{M\,m^{-1}}$ in the upper part of the planetary boundary layer (PBL, above 1000 m height). The surface extinction values found in this study are a factor of two larger than the ones found from EARLINET. Most likely, the EARLINET lidar statistic is biased by drier cloud free days while the SÆMS data are taken at all ambient conditions. Also the present statistic is based on all measurement cases including cases with near-surface capped inversions and not only based on well-mixed conditions. So it is reasonable that the surface mean extinction values shown here are larger than the EARLINET data.

3.2 Case studies

Days with a strong decrease in relative humidity during the morning hours or a strong increase in the evening served as the basis for our specific investigation of the influence of water uptake by particles on their optical properties. We sampled 143 days during the four-year period with a pronounced diurnal cycle in terms of relative humidity. For the parameterization we only used cases with a diurnal cycle of the relative humidity from max 75% in minimum to min 80% in maximum with $> 20\%$ difference from minimun to maximum without any changes in air-mass origin or precipitation during the measurement. Figure 5 presents two examples. Besides the influence of the relative humidity, changing air flow direction (long-range transport) and the daily evolution of the PBL can have a sensitive impact on the surface-near particle extinction coefficient. The backward trajectories (HYbrid Single-Particle Lagrangian Integrated Trajectory Model, HYSPLIT, http://www.arl.noaa.gov/HYSPLIT.php) (**?**Draxler and Hess, 1998; Draxler, 1999) indicate almost constant long-range aerosol transport conditions during the shown measurement periods. The 96-h back trajectories from 20 August (Fig. 5a) all originated at 3500 m height and indicate an almost identical descend linearly in height until their arrival in Leipzig with a maximum height separation between the individual trajectories of < 500 m (trajectory heights not shown in the plot). The three back trajectories from 27 August revealed that the air masses remained at a constant height of 500–1000 m for 96 h. The particle optical depth at 500 nm as observed with the AERONET photometer was around 0.1 ± 0.04 over the whole day until 16:00 UTC on 20 August, and thus confirmed the almost constant aerosol conditions during time period shown in Fig. 5b.

According to the lidar observation on 20 August 2009, the PBL development (growth of the PBL height with time) was found to influence the aerosol extinction properties close to the surface not before about 11:30 UTC. As a general result of the 2009–2012 lidar observations we found that the diurnal PBL evolution only affects the surface-near aerosol concentration to a significant amount when the growing PBL grasps into the clean free troposphere so that any further increase in PBL depth reduces the aerosol concentration in the entire PBL by downward mixing of clean free tropospheric air. As long as the convectively active PBL is developing into the polluted residual layer ontop of the growing, but shallow PBL, the impact of the PBL development on the measured surface-near extinction coefficient was usually found to be low. The steady decrease of the extinction coefficient from 11:30 to 15:00 UTC on 20 August 2009 in Fig. 5b is the result of the growing PBL and corresponding downward mixing of clean air from the free troposphere. The PBL depth increased from 1300 to 1900 m (30 % increase). This is directly reflected in the decrease of the extinction coefficient from values around 0.2 to values around 0.14, while the relative humidity decreased from 53 to 48 % only.

On 27 August 2009, cloudy weather prevailed. The trajectories in Fig. 5d show a constant air flow from southwest.The lidar detected a deep, aged aerosol layer (residual layer) up to 2 km height in the morning. The depth of this stable stratified layer increased only slightly up to 2.5 km height until the evening probably driven by shallow PBL convection as detected from ceilometer measurements near Leipzig. The AERONET photometer recorded an optical depth of 0.2 +/- 0.05 for 500 nm throughout the day, indicating a polluted, aged European air mass. Thus the average PBL extinction coefficient remained constant within a relative uncertainty of 20% throughout the day which indicates that the precondition of a constant aerosol load (i.e., a constant dry aerosol extinction) is valid. Finally, only the near-surface extinction decreased significantly with decreasing relative humidity which itself was caused by near-surface temperature increase after sunrise. The humidity was close to 100 % in the early morning around 03:30 UTC and decreased to almost 35 % in the afternoon around 13:30 UTC. The correlation between the simultaneously measured relative humidity and particle extinction coefficient for the two different days is shown in Fig. 5c and f. Curve fitting (assuming a relative-humidity dependence according to Eq. 3 reveals the value for γ as given in Fig. 5c and f. For the pronounced relative-humidity dependence on 27 August 2009, the parameter is quite similar to the one for urban haze after Hänel (1984). For 27 August 2009, we obtain for the exponent $\gamma = 0.50$ after Eq. (3). Hänel (1984) found $\gamma = 0.44$.

3.3 Extinction enhancement factor

The calculation of the extinction enhancement factors relies on the assumption that the initial air mass, and more specifically the dry aerosol extinction, is constant throughout the measurement while the relative humidity changes. For the calculation we excluded all days with precipitation to exclude wet depositional loss or days with a distinct change of the air-mass origin during a measurement. We also had to exclude all measurements with visibilities less than the optical path length and days with no significant aerosol load (clean days, $b_{\mathrm{p,e}} < 0.05\,\mathrm{km}^{-1}$). Secondary aerosol production, advection of aerosol from local sources to the site or an air mass with lower concentration, temperature-driven partitioning of ammonium nitrate (e.g. Morgan et al., 2010) and of semivolatile material (e.g. Donahue et al., 2006), and boundary-layer dilution can force this assumption to fail. Accompanying in-situ measurements of the extinction coefficient by the dried aerosol would be one choice to provide a dry reference. However, in Leipzig such data were not available for the long-term period investigated here. Therefore, in a first step we have analyzed the backward trajectories to ensure a constant air-mass origin during the measurement. Secondly, the time periods we used to quantify the dominant optical-enhancement process where usually no longer than four hours. For the effect of boundary-layer dilution it was often found from lidar measurements that the residual layer from the prior day is still present in the morning above the nocturnal inversion layer. The turbulent PBL growth process then mixes the residual layer downwards while the surface aerosol is mixed upwards. Hence, statistically the net dilution effect is smaller than expected from PBL growth alone so that in a lower extreme considering a negligible nocturnal aerosol production at the surface and no deposition of aerosol from the residual layer the dilution effect could even be nonexistent. On average, the possible uncertainties given by the reasons above are still small (on the order of 20-30% throughout a measurement) compared to extinction enhancement be relative humidity (on the order of 200-300%). With the given preconditions we were able to select 143 days out of our 4 year data set in order to derive the extinction enhancement factor on a statistical basis. The main results of the analysis are summarized in Fig. 6. For each of the 143 days, the optimum curve after Eq. (3) and the corresponding value for γ were determined. From these data set, the mean value $\overline{\gamma}$, and the corresponding SD $\delta\gamma$ as presented in Fig. 6 were calculated. The curve for the mean enhancement factor (blue curve in Fig. 6) is obtained with Eq. (3) and the mean value $\overline{\gamma}$. The upper and lower boundaries of the gray-shaded area in Fig. 6 are obtained by using $\overline{\gamma} + \delta\gamma$ (upper boundary) and $\overline{\gamma} - \delta\gamma$ (lower boundary) in Eq. (3). The close agreement of the blue curve with the green curve for urban haze after Hänel (1984) in Fig. 6 indicates the high quality and reliability of our long-term observations. More case studies and more details to the parameterization efforts can be found in Skupin (2014). In Fig. 6 of our study (mean enhancement factor) we find the extinction enhancement to be 2.41 at 85% RH which is very close to the previous finding of 2.78 in Melpitz, the rural background measurement site of TROPOS (Zieger et al., 2014).

3.4 Extinction coefficient and enhancement factor for different air flow conditions

In order to investigate to what extend regional and long-range transport of aerosols influenced our measurements we performed an extended cluster analysis based on 4-day HYSPLIT backward trajectories for all selected observations. We considered 18 000 individual SÆMS observations performed in the years 2009–2012 in this study. The cluster analysis revealed eight significant air flow regimes for which different optical properties were obtained.The ambient RH for our measurements was found to be 65% on average for each cluster with a standard deviation of 15% RH within each cluster. The mean differences between the clusters RH were found to be low (max. 5%) with the maximum of 70% RH for cluster 3 and the minimum of 63% RH for cluster 4. Figure 7 presents an overview of the surface-near particle extinction conditions over Leipzig for different airflow directions. In Fig. 7a, mean values and SD of the particle extinction coefficient for ambient conditions are given. Note the two westwind clusters (for

strong westerly winds and for slow air mass transport from the west). Figure 7b shows the cluster mean extinction values after normalization of the individual data points to 0 % relative humidity by using the derived cluster mean values for γ (Fig. 7c) and the respective RH of each data point. Fig. 7c presents the cluster-mean γ values which were calculated by Eq. (3) for individual days. Higher In Fig. 7c γ values reaching almost 0.6 and indicating more hygroscopic particles were found for the north and east clusters, whereas the lowest values around 0.4 were observed when the air was advected from the west or northeast. γ is closely correlated with the 80-to-40 % extinction growth factor and takes values of around 0.4, 0.5, and 0.6 for growth factors around 1.55, 1.7, and 1.85, respectively. In Fig. 7d, the dry particle extinction coefficients are given for the individual years from 2009–2012.

The main findings can be summarized as follows: after removing of the humidity effect on light extinction, the extinction coefficients are generally a factor of 2 lower then for ambient conditions, disregarding specific airflow conditions. The largest extinction coefficients with a mean value of $0.23\,\mathrm{km}^{-1}$ ($0.11\,\mathrm{km}^{-1}$ for dry particles) were observed when the air masses were advected from easterly directions, i.e., from the eastern parts of Leipzig (with the highway A14), from the most eastern parts of Germany, Poland, Ukraine, and polluted southeastern European regions. The lowest extinction coefficients (about a factor of 2 lower then the east-cluster values) were observed during situations with fast westerly air mass transport. Pronounced contributions to particle extinction by the Leipzig city center (clusters 5–7 in Fig. 7c) were not found. On average, the surface-near extinction coefficients are about $0.17\,\mathrm{km}^{-1}$ ($0.08\,\mathrm{km}^{-1}$ for dry particles) with an only weak dependence on the airflow conditions. Particle extinction conditions at our SÆMS measurement site were seemingly widely controlled by local and regional aerosol sources and, only to a second order, by long-range aerosol advection.

The year-by-year statistics of dry particle extinction coefficients in Fig. 7d support this impression. Air masses advected from the east show the highest extinction values in each of the four years and the variations of the individual cluster-mean extinction values around the overall mean are in the 10–20 % range (except for the east cluster). However, year-by-year differences are also obvious. The comparably large 2010 extinction values are caused by strong construction activties in the eastern parts of the Leipzig greater area. Highway construction works covered the whole year to extend the four-lane highway A14 to a six-lane road. In contrast, on 1 March 2011 the Environmental Green Zone restriction were brought into operation in Leipzig to meet the European Union's regulation on particulate matter to ban vehicles which didn't meet certain requirements from the city. This implementation may have caused the overall low particle extinction values observed in 2012. There is almost no difference in the precipitation amount for the years 2011 and 2012 which could explain a potentially stronger wash out effect in 2012 and frequent cleaning of the streets (and reduced road dust effects). Figure 8 provides an overview of the mean particle enhancement factor (and corresponding SD) for the different airflow clusters. The shown mean values and SD of the ratio of particle extinction at 80 or 95% relative humidity to the one at 40% relative humidity were directly calculated from the available individual days with strong humidity variability (either from 40 to 80% or from 40 to 95% RH, respectively) for each of the eight air flow regimes separately. Because of the larger required RH span in ambient conditions Fig 8a includes additional observational cases with respect to Fig 8b As can be seen in Fig. 8, large differences between the clusters were not found. The 80-to-40 % extinction growth factor was 1.75 ± 0.4, on average with variations between the clusters mean values of the order of 0.1. Stronger differences between the clusters were found for the 95-to-40 % extinction growth factors. The largest value of 3.5 was observed for northerly air flows with the comparably largest influence of marine particles (at comparably low levels of pollution advection from the Baltic Sea and Scandinavia). The lowest growth factor of 2.3 was found for the south-wind cluster with a high amount of anthropogenic less hygroscopic pollution particles. On average, the 95-to-40 % extinction growth factors was 2.8 ± 0.6.

Table 1 provides literature values of the extinction growth factors for comparison. Values between 1.1 and 3.3 have been published for the 530–550 nm wavelength range. For biomass burning aerosol or background (rural) particles extinction growth factors as low as 1.0–1.2 were found. For polluted continental areas the growth factors accumulate from 1.6–2.0, and for marine particles values above 3.0 are observed. Our observations fit well into the larger frame of observed growth factors and adds new values for the high humidity range (95-to-40 % growth factors).

3.5 Extinction coefficient statistics: comparison of SÆMS-, AERONET-, and in situ observations

In Fig. 9, we compare our SÆMS measurements for a time period of five days in September 2009 with particle extinction coefficients at 550 nm derived from ground-based in situ measurements of the dry particle size distribution (Birmili et al., 2009). Such a comparison was already successfully performed for a ten-day period in March 2000 (Müller et al., 2006), with a similar apparatus as SÆMS but by using a very short optical path in the vicinity of the in situ measurement stations. A successful comparison between in situ aerosol observations on the roof of the TROPOS building and the SÆMS observations along the 2.8 km path was also shown in Fig. 8 in Skupin et al. (2014) for 3 May 2009.

The in situ extinction coefficients are computed from the measured size distributions of dried particles, i.e., for particle size distribution measured at relative humidities around 30 %. A so-called PM_{10} inlet is used so that very coarse par-

ticles with diameters larger than about 10 µm are not measured. The particle size distributions were measured with a tandem differential-mobility particles sizer (TDMPS, 3–800 nm in diameter) and with an aerodynamic particle sizer (APS, 0.8–10 µm in diameter). The in-situ data we used for this study are 1-hour averages.The particle extinction coefficient was calculated by means of a Mie scattering code based on Bohren and Huffman (1983) as described in Skupin (2014). The real part of the refractive index was set to a constant value of 1.53 (typical value for urban haze). Absorption by particles was considered by assuming an imaginary part of 0.01i.

As can be seen, a good overall agreement between the in situ and SÆMS dry extinction time series (black and red curves) is obtained. The 2009 mean (±SD) and median dry particle extinction coefficients are $0.061 \pm 0.055\,\mathrm{km}^{-1}$ and $0.046\,\mathrm{km}^{-1}$ (in situ), respectively, and $0.073 \pm 0.036\,\mathrm{km}^{-1}$ and $0.065\,\mathrm{km}^{-1}$ (SÆMS, dry), respectively. The humidity-corrected SÆMS extinction coefficients in Fig. 9 are calculated from the ambient SÆMS extinction values by using the extinction enhancement parameterization shown in Fig. 6. The good agreement between the black and red curve indicates the usefulness of the developed parameterization. The correlation coefficient is found to be 0.71.

The strong impact of relative humidity on particle extinction (SÆMS, ambient) is illustrated in Fig. 9. During the gray-shaded time periods from 13:00–17:00 UTC, when the PBL is well mixed at sunny days (days 267–269 in Fig. 9), the relative humidity and particle extinction take their daily minimum. During the afternoon hours, the PBL has the largest vertical extent which contributes to the observed low extinction values around 15:00 UTC. The systematically lower in situ extinction coefficients on these sunny days compared to the SÆMS (dry) values may be partly caused by the used constant refractive index which is probably not appropriate for all aerosol conditions throughout the day, especially not when aged particles (after long-range transport) are mixed down from higher altitudes and partly substitute the less aged urban haze close to the ground. The humidity correction may be also not valid at all for the aerosol conditions found during the convectively active period. Furthermore, we compare point measurements with long path measurements 300–3140 m apart from the PM_{10} inlet. TROPOS is part of an area with complex urban building structure, whereas the optical path of SÆMS crosses areas with much less buildings, even areas without any building, and is parallel to several large motorways and crosses the A14 highway.

In Fig. 10, extinction distributions derived from 2009 AERONET sun photometer and SÆMS (ambient) measurements are compared. The shown distribution curves are optimum fits to the respective frequency-of-occurrence distributions of measured and derived extinction coefficients. As before, we considered only data measured in the afternoon from 13:00 to 17:00 UTC, when the probability is highest that the PBL is well mixed. In the case of the AERONET observations, the extinction distribution curve shows PBL mean extinction values (vertical column mean values). First, we converted the measured 500-nm particle extinction with the Ångström exponent (500–870 nm) to 550 nm wavelength by Eq. 1. Then, all calculated 550-nm aerosol particle optical thickness (AOT) values were divided by the respective PBL height, obtained from numerical weather prediction data (GDAS: global assimilation system, http://www.arl.noaa.gov/gdas.php) (Kanamitsu, 1989), before the calculation of the frequency-of-occurrence distribution. At well-mixed conditions the PBL mean particle extinction coefficient is closest to the extinction value measured with SÆMS during the day.

As can be seen in Fig. 10, a rather good agreement between the SÆMS (ambient) and the AERONET observations is found. The mean extinction coefficients and SD for 550 nm is $0.12 \pm 0.09\,\mathrm{km}^{-1}$ (AERONET) and $0.11 \pm 0.06\,\mathrm{km}^{-1}$ (SÆMS). A systematic overestimation of the PBL mean extinction value must be kept in consideration in the interpretation of the AERONET observations, because, on average, 20 % of the AOT is caused by particles in the free troposphere (Mattis et al., 2004).

For comparison, also the distribution of dry extinction coefficients as obtained from the SÆMS observations after humidity correction and the extinction distribution calculated from the in-situ-measured dry particle size distributions are shown for the specific 13:00–17:00 UTC time period. The possible reasons for the found deviations between the two dry extinction frequency-of-occurrence distributions were discussed above.

3.6 Extinction wavelength dependence as a function of relative humidity

Finally, we briefly summarize the influence of a relative-humidity increase on the spectral slope of the particle extinction coefficient for the wavelength range from 390 to 881 nm. Figure 11 shows a steady increase of the Ångström exponent (see Eq. 1) with increasing relative humidity for the entire spectrum from 390–881 nm and a decrease for the short wavelength range (390–440 nm). The figure is based on all measurements in 2009 and 2010. The reason for the increase of the 390–881 nm Ångström exponent and the decrease of the 390–440 nm Ångström exponent is shown in Fig. 12. A strong increase of the 390 nm particle extinction coefficient was observed with increasing relative humidity, an even stronger increase was observed at 440 nm, whereas no or even a decreasing trend of the extinction strength with increasing relative humidity at 881 nm. A strong water-uptake effect for fine-mode particles with radius < 100 nm can explain the strong increase of the extinction coefficient at the shorter wavelengths as our Mie scattering calculations indicate. Furthermore, the impact of fine-mode particles on the extinction coefficient at 881 nm is low. At this wavelength the extinction coefficient is primarily determined by larger

particles. Although coarse-mode sea-spray particles cannot be fully ignored in Leipzig, most of the time the coarse mode consists of road and soil dust particles which do not grow significantly by water-uptake. As a consequence, the extinction coefficient at 881 nm might remain constant for all ambient humidity conditions (c.f. Fig. 2) while the 390-nm extinction coefficient increases by fine-mode particle hygroscopic growth. Consequently, the overall 390–881 nm Ångström exponent might also increase with relative humidity. Significantly different Ångström exponents for the eight air-flow classes were not observed pointing again to the dominating influence of local and regional pollution on the aerosol conditions at our field site. It is finally worthwhile to mention that the mean value and SD for the 440–881 nm Ångström exponent for the years of 2009 and 2010 is 1.55 ± 0.42 in the case of the AERONET column measurements. In contrast the 390–881 nm SÆMS Ångström exponents show a mean value of $0.91 \pm 0.68.0$ (Skupin, 2014) for the 2009–2010 period, a clear indication of the strong impact of coarse particles on the SÆMS observations.

4 Conclusions

For the first time, a long-term study of the surface-near particle extinction coefficient at undisturbed aerosol and humidity conditions at a central European urban site has been presented. The dependence of particle extinction on relative humidity could be studied from 20 to almost 100 % relative humidity. For the wavelength of 550 nm, the mean extinction enhancement factor was found to be 1.75 ± 0.4 (for a humidity increase from 40 to 80 %) and 2.8 ± 0.6 for a relative humidity increase from 40 to 95 %. A parameterization of the humidity dependence of the particle extinction coefficient was derived. A mean hygroscopic exponent γ of 0.46 for the 2009–2012 period was retrieved. Based on an extended backward trajectory cluster analysis, a weak dependence of the particle optical properties (AOT, extinction enhancement factor, Ångström exponent) from the air flow condition has been observed. Locally produced aerosol particles widely controlled the measured ambient aerosol optical properties.

In this study, we had to rely on a persistent dry particle extinction coefficient while the ambient humidity changed. Various measures were taken to ensure this precondition. For future studies however, we intend to use co-located in-situ measurements to not only ensure but to directly measure a dry baseline. In this way, more valid cases of hygroscopic extinction enhancement could be obtained from a measurement campaign. As an outlook, a mobile SÆMS (based on a simplified setup with, e.g., three diode lasers as radiation sources operating around 400, 550, and 850 nm) would be desirable to study basic ambient aerosol conditions at very different places (rural areas, background stations, marine environments, regions influenced by desert dust). However, to investigate the dependence of particle extinction on relative humidity, strong ambient humidity changes must occur, which may not be observable on islands or desert sites.

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Table 1. Overview of published particle extinction enhancement factors based on extinction values measured at different values of relative humidity RH (%).

Region	Aerosol type	RH (wet/dry)	(λ)	Enhancement factor	Reference
Brazil	biomass burning	80/30	(550 nm)	1.01–1.51	Kotchenruther and Hobbs (1998)
USA	urban/industrial	80/30	(550 nm)	1.81–2.3	Kotchenruther et al. (1999)
Portugal	anthropogenic	82/27	(550 nm)	1.46	Carrico et al. (2000)
India	biomass burning or dust	85/40	(550 nm)	1.58	Sheridan et al. (2002)
Africa	biomass burning	80/30	(550 nm)	1.42–2.07	Magi and Hobbs (2003)
Korea	dust	85/20	(550 nm)	2.00	Kim et al. (2006)
Switzerland	rural	85/20	(550 nm)	1.21–3.3	Fierz-Schmidhauser et al. (2010)
Norway	marine	85/20	(550 nm)	3.24	Zieger et al. (2010)
Italy	rural	90/0	(550 nm)	2.1	Adam et al. (2012)
United States	polluted continental, marine-pollution mixtures	80/30	(530 nm)	1.6	Im et al. (2001)
China	urban	80/40	(550 nm)	1.9	Liu et al. (2013)
China	polluted continental	90/40	(550 nm)	1.93	Chen et al. (2014)
Germany	polluted continental	85/10	(550 nm)	1.2–3.6	Zieger et al. (2014)
Germany	urban	80/40	(550 nm)	1.86	Müller et al. (2006)
Germany	urban	80/0	(550 nm)	2.12	Müller et al. (2006)
Germany	urban	80/40	(550 nm)	1.37–1.99	this work
Germany	urban	95/40	(550 nm)	2.35–3.49	this work

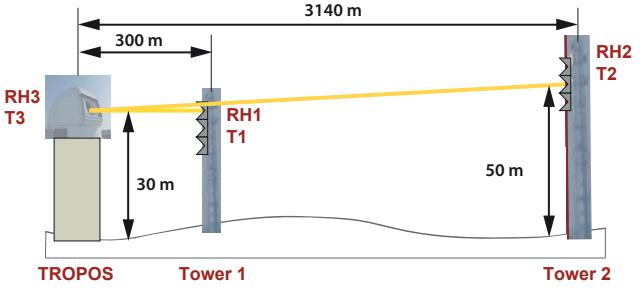

Figure 1. Sketch of the SÆMS measurement configuration. A light beam is transmitted at TROPOS and direct to a retroreflector array mounted at Tower 1 for several minutes. Afterwards the beam is moved to the second retroreflector array at Tower 2 for several minutes, followed by the next round in which the beam is again directed to Tower 1, and so on. Particle extinction is derived from the Tower 1 and Tower 2 long-path transmission observations, and thus is related to an almost horizontal path of 2840 m at a height of 30–50 m above ground. The aerosol particle extinction measurements are set into context with meteorological observations of temperature (T) and relative humidity (RH) which are measured at the roof of TROPOS (T3, RH3) and close to the retroreflectors at Tower 1 (RH1, T1) and Tower 2 (RH2, T2).

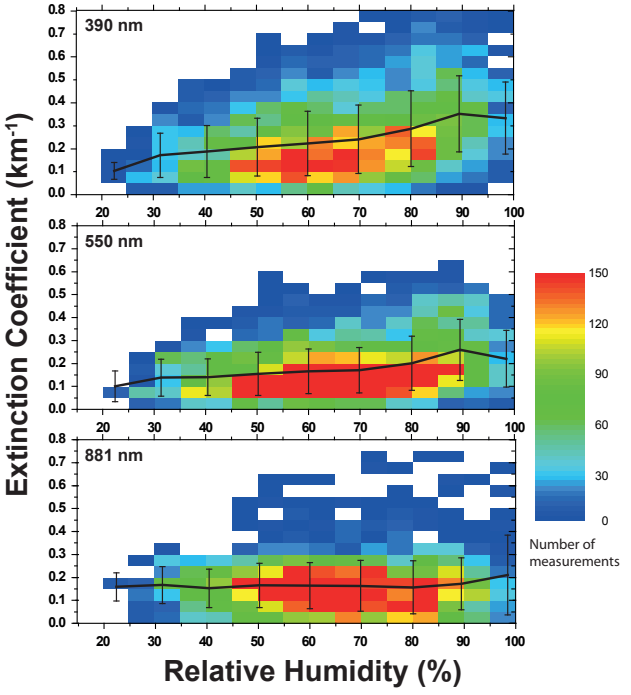

Figure 2. Measured particle extinction coefficients for the wavelengths of 390 nm (top), 550 nm (center), and 881 nm (bottom) as a function of relative humidity. The color scale indicates how frequently a given extinction coefficient was measured during the 2009–2012 period. Mean values (bold lines) of extinction coefficients and corresponding SD (vertical bars) are shown for 10 % humidity intervals.

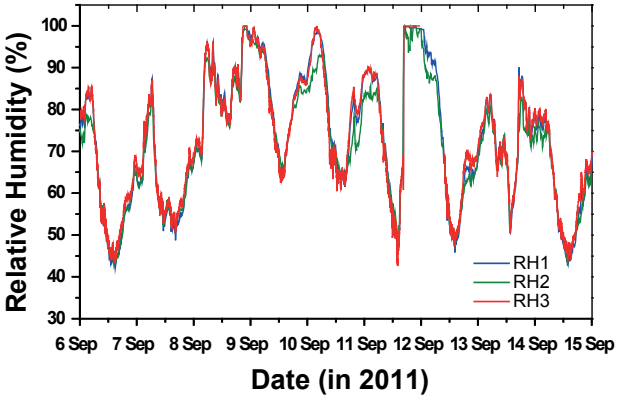

Figure 3. Example of the three-point relative humidity observation (over 9 days) with humidity sensors on top of the TROPOS building and at the two towers (see Fig. 1).

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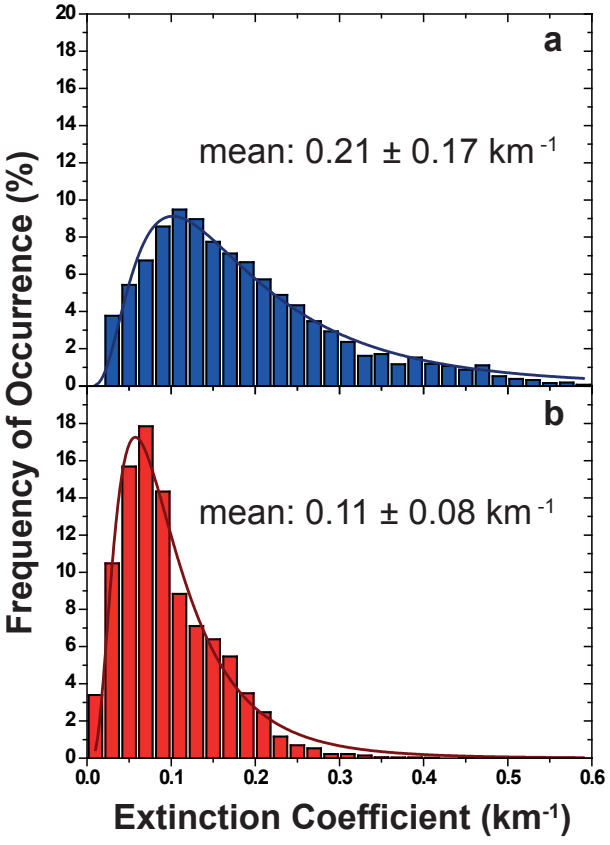

Figure 4. **(a)** Frequency distribution of ambient 550 nm particle extinction coefficient observed with SÆMS at Leipzig from 2009–2012, **(b)** same distribution after correction of the particle water uptake effect, i.e., after normalization of all values to 0 % relative humidity by means of Eq. (3) with the parameter for urban aerosol derived from the four-year SÆMS study. 2009–2012 mean value and respective SD are given as numbers.

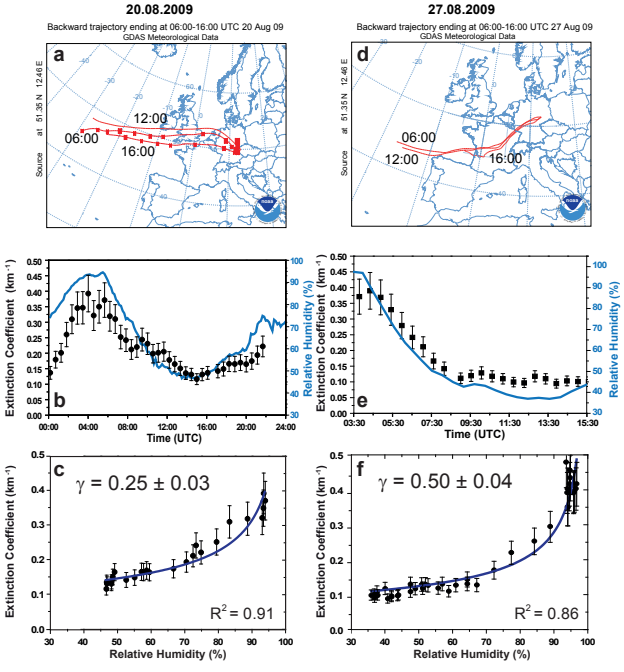

Figure 5. SÆMS observations on (left) 20 August 2009 and (right) 27 August 2009. Almost constant horizontal transport of polluted air from westerly to southwesterly directions is indicated by 4-day HYSPLIT backward trajectories (**a**, **d**, arrival height of 500 m). The temporal variation of the 550 nm particle extinction coefficient for 550 nm with relative humidity is shown in **(b)** for 20 August 2009 and in **(e)** for 27 August 2009, and the corresponding relationship between ambient extinction coefficient and relative humidity is presented in (**c** and **f**). The curves fitted to the data points in (**c** and **f**) are obtained with Eq. (3). The coefficient of determination R^2 for each fit is given as number.

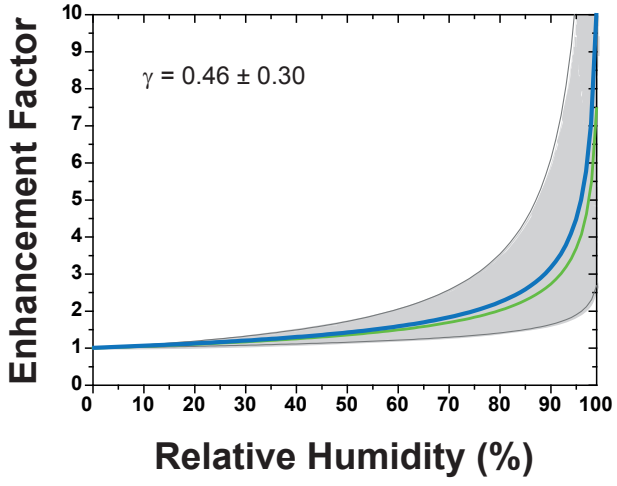

Figure 6. Mean value of the enhancement factor for the 550 nm particle extinction coefficient (blue line, obtained with Eq. (3) for the mean value $\overline{\gamma}$). The upper and lower boundaries of the gray-shaded area are obtained by using $\overline{\gamma}+\delta\gamma$ (upper boundary) and $\overline{\gamma}-\delta\gamma$ (lower boundary) in Eq. (3). The given mean values and SD of the parameter γ result from the evaluation of 143 observational cases collected in the years 2009–2012. The green curve is shown for comparison and represents urban haze conditions after Hänel (1984).

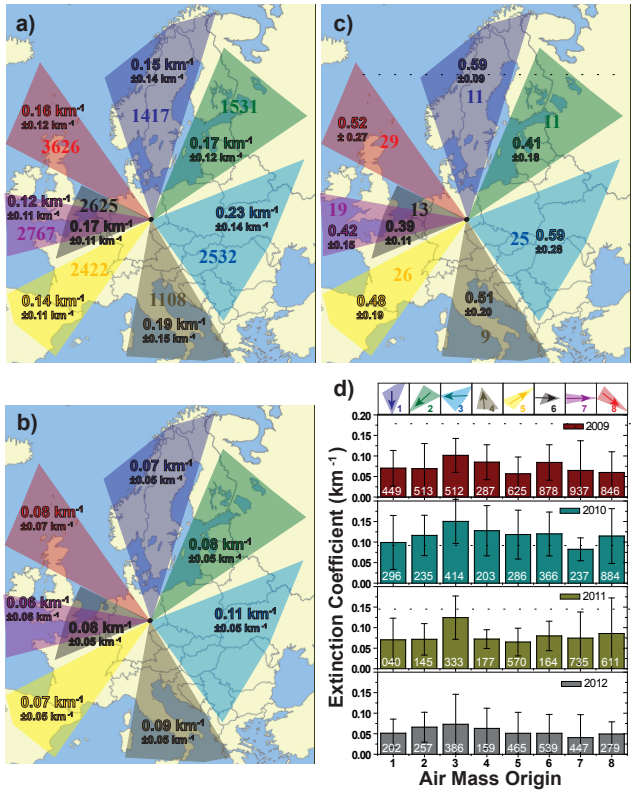

Figure 7. **(a)** Extinction coefficient for 550 nm (mean value, SD, number of measurements) for eight defined air mass transport regimes based on SÆMS observations from 2009–2012, **(b)** same as **(a)**, except prior to averaging all individual cases were normalized for dry conditions (RH = 0 %) by use of the derived cluster mean parameter $\gamma(c)$, **(c)** $Hygroscopic exponent \gamma$ for 550 nm (mean value and SD, computed with Eq. 3) for the eight air mass transport regimes derived from SÆMS observations from 2009–2012. Numbers of available cases per cluster are given in addition, and **(d)** same as **(b)**, but separately for for dry conditions (RH = 0 %) for each year of the period from 2009–2012.

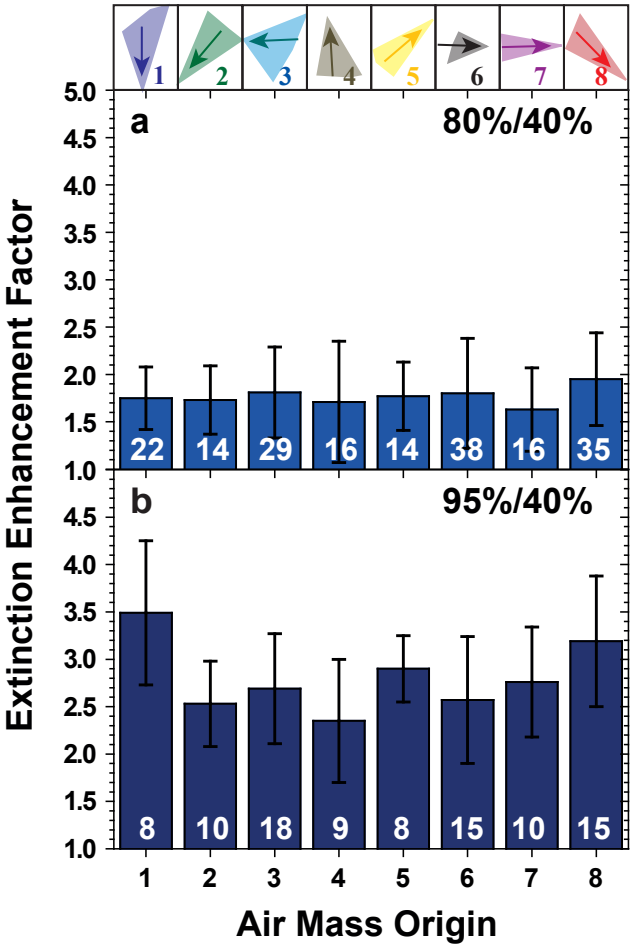

Figure 8. Particle extinction enhancement factors for 550 nm (80-to-40 % and 95-to-40 % RH enhancement) observed from days with occurring humidity variations between at least 40 and 80% RH (a) and only from days with variations between at least 40 and 95 % RH (b) separated for the eight air mass transport regimes. Four-year mean values and SD are given.

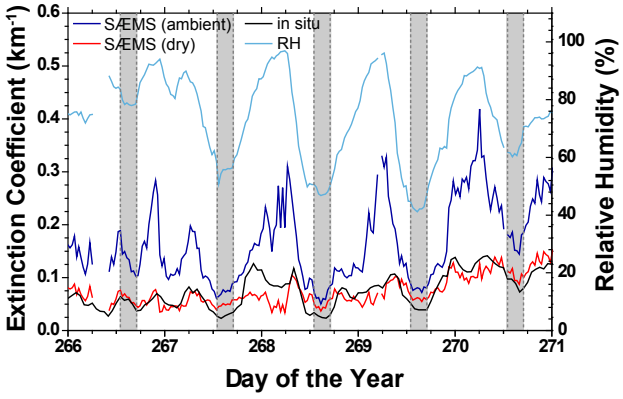

Figure 9. Comparison of 550 nm extinction coefficients for 550 nm measured with SÆMS (ambient, dark blue) and computed from dry particle size distributions (black) measured in situ at the roof of the TROPOS building from 24–29 September 2009. The humidity-corrected SÆMS (dry, 0 % relative humidity) extinction time series is shown as red curve. Relative humidity is given in addition as light blue line. Gray shaded areas indicate the 13:00–17:00 UTC periods during which the PBL is assumed to be well mixed, PBL depth takes its maximum, and relative humidity and particle extinction take their minmum during sunny days (days 267, 268, 269).

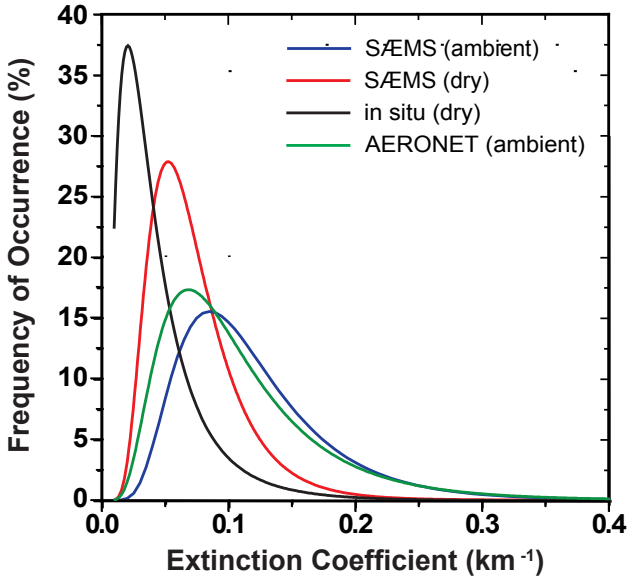

Figure 10. Frequency of occurence of 550 nm particle extinction coefficient measured with SÆMS (ambient) at TROPOS, Leipzig, between 13:00–17:00 UTC of each day in the year of 2009 (blue line). For comparison, the respective distribution for the PBL-mean extinction coefficient (ambient, green) is shown. These values are derived from AERONET sun photometer observations of the 500 nm particle optical depth divided by the PBL depth, which was estimated from GDAS model data. The red SÆMS (dry) curve shows the distribution of humidity-corrected SÆMS 550 nm particle extinction values (for 0 % relative humidity). The black distribution (in situ, dry) shows the 550 nm extinction values calculated from in situ observations of the dry particle size distribution at the roof of the TROPOS building exclusively for the time period from 13:00–17:00 UTC.

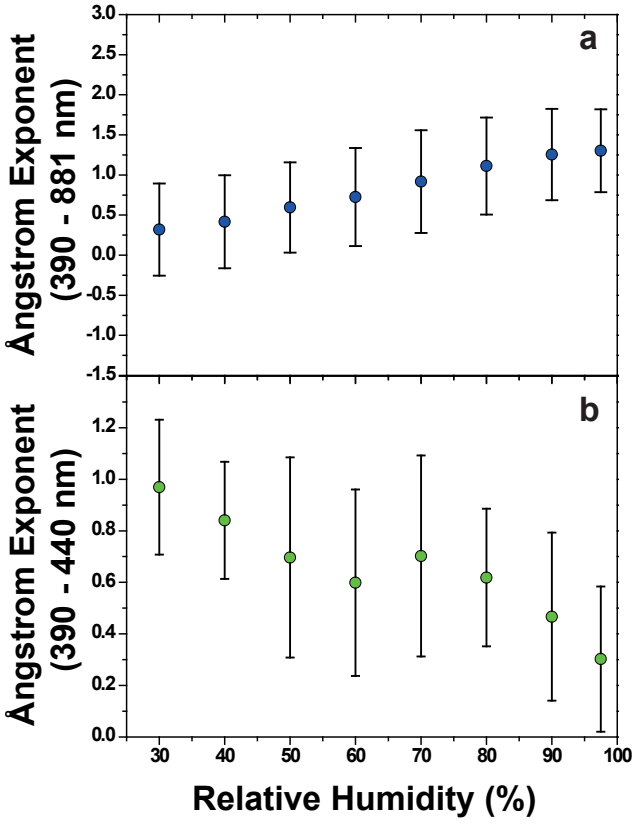

Figure 11. 2009–2010 mean Ångström exponents and SD for the 380–881 nm (top) and 390–440 nm (bottom) wavelength range for eight relative humidity classes.

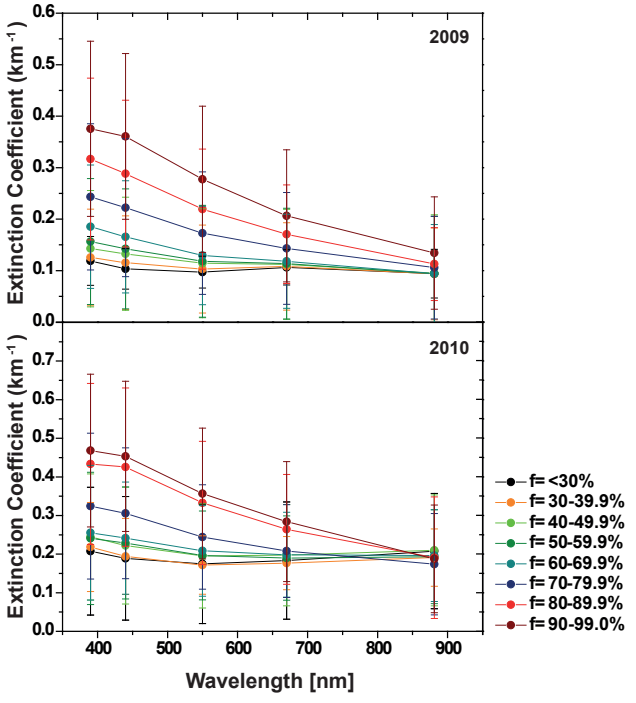

Figure 12. (Top) 2009 and (bottom) 2010 mean extinction coefficient spectrum for eight relative humidity classes (indicated by different colors). Vertical bars indicate the SD for each of the shown five extinction coefficients (for five wavelengths) for a given humidity interval.