Reply to the reviewer comments of the manuscript "Comparison of ambient aerosol extinction coefficient obtained from in-situ, MAX-DOAS and LIDAR measurements at Cabauw" by Zieger et al., 2010 (acp-2010-838)

February 14, 2011

Content:

Reviewer No. 1 (Anne Jefferson)

We thank Anne Jefferson for her constructive review comments, which helped us to improve the quality of our publication.

This paper describes results from a field campaign that compares in-situ and remote measurements of the aerosol extinction as well as a closure study between HTDMA *g*(RH) and humidified nephelometer *f*(RH) measurements. The extinction comparison identifies specific air mass source regions and aerosol types that account for differences in the remote and in-situ measurements.

Experimental Section:

Was the nafion tubing a bundle of many tubes or a single tube? What was the tube ID?

Reply: The Nafion dryer consists of a single Nafion tube (Perma Pure LLC), as described in the cited technical paper by Rahel Fierz-Schmidhauser et al. (2010b), the tube diameter is 1.7 cm.

Is it possible to apply the same O_4 correction factor to the MAX-DOAS retrievals so that this difference can be eliminated when comparing the measured extinctions to the surface in-situ measurements?

Reply: The O_4 correction factor (scaling factor) is a MAX-DOAS specific retrieval factor and there is no physical reason to apply this factor to the in-situ measurements. It accounts for an uncertainty in the absolute value of the existing O_4 cross sections Greenblatt et al. (1990); Hermans et al. (2002). The scaling factor here is based on studies by Wagner et al. (2009) and Clémer et al. (2010). They also have mentioned in their papers that the estimated scaling factors have an uncertainty as high as ±0.10, which is much larger than the difference between the scaling factors used by different groups in the present study.

4.1 Wet neph analysis

How much does the fitted value of *f*(RH) differ if RH values down to the lower limit of the dehydration branch are used? There are so few data points above 70% RH that doing the fits over a larger RH range will decrease the fit uncertainty. What is the variability in the scattering coefficient over the 3 hour time frame of the humidity scans? My concern is that the air mass and aerosol can change significantly over such a long time period. Is it possible to average the humidity scans over an hour or less?

Reply: Taking only values below 70% RH of the dehydration branch does not significantly improve the fitting result (these are anyway only few points). An entire humidogram was recorded every 3 hours with one hydration and one dehydration scan (each took 1.5 h). This time was chosen to ensure a RH equilibrium within the system during the scans. At the high RH of 85% the 1.5 h long hydration and dehydration scans were fitted separately for RH>70%. The scan from 70% up to around 90-95% and back again took approximately only 1 hour, so within this hour, we have to assume no air mass change. No significant difference was found for the two branches at this high RH, which additionally backed up our assumptions. One hour averaging time can be used, but the noise due to less data points is increased, while the general picture does not change (see Figure A below, where the 1-hour average has been added to Fig.1a as shown in the manuscript). Therefore, we would prefer leaving the Figures with a 3-hour averaging time frame.

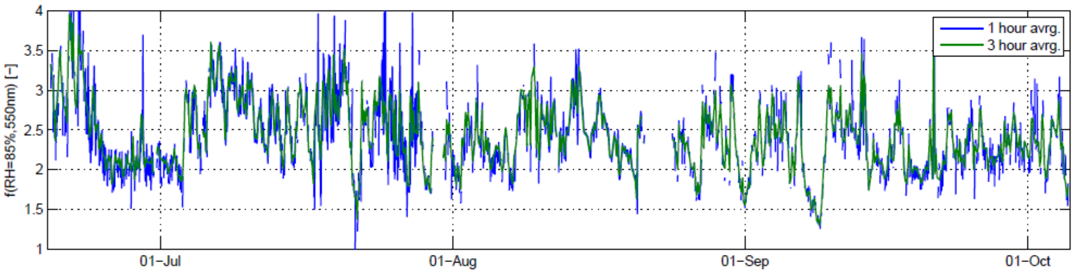

Figure A. Same as Fig 1a shown in the submitted manuscript, except that the result for a 1-hour averaging time has been added.

4.2 Factors influencing *f*(RH)

How does the 165 nm diameter size of the HTDMA compare to the median volume or surface area aerosol diameter during the measurement period?

Reply: The mean surface area diameter is not well correlated ($R^2<0.1$) with the *g*(RH) at 165 nm. This can already be seen in Fig. 3, where the mean diameter of the entire size distribution is only slightly (anti-) correlated with *g*(RH). The coarse mode volume fraction is e.g. a much better proxy for the sea salt contribution, as can be seen in Fig. 3 and 6.

Trish Quinn has a paper where she shows a nice correlation between fRH and the ratio of OC/(OC +SO4). (Geophys. Res. Lett., 32, L22809, doi:101029/2005GL024322) Can you compare fRH to this fraction or to the mass fraction of OC or total carbon rather than just BC? I don’t know if there was any aerosol chemistry measured or if there are past measurements from this region to which you can refer. BC may have a minor influence on fRH or its influence is coincidental with its co-emission with OC as OC will comprise a much larger fraction of the aerosol mass than BC. Keep in mind that BC usually comprises a larger portion of the aerosol mass in fine mode aerosol than in accumulation mode aerosol, so its influence will be more apparent for gRH than fRH.

Reply: Unfortunately, the aerosol chemical composition was not measured during our campaign at Cabauw. The BC volume fraction clearly anti correlates with the hygroscopic fraction (as can be seen in Fig. 3 and is discussed in the text).

4.3 Closure Study

In your calculation of g from Equation 2 what values did you use for a_w and κ? How did you determine κ?

Reply: For the water activity a_w the relative humidity can be used, if the Kelvin term is neglected, which is justified for large particles (as stated in the manuscript). The value κ is then calculated by inserting the RH of the H-TDMA measurement (in our case 90%) and the measured growth factor. For clarification, we have modified the sentence in Section 4.3: "Since the H-TDMA measured at a constant RH=90%, the value of *g*(RH) for different RH was calculated using Eq. (2), where instead of the water activity a_w the relative humidity RH is used."

Place the empirical equation for (*g*(RH=85%)= b_1+ . . . on a separate line with a number. The following sentence would be clearer if written "The result of the *f*(RH) calculation using Equation 9 for *g*(RH) compared to the measurements is presented in Fig. 5d." Otherwise it's unclear as to how *f*(RH) was calculated, whether from the measured *g*(RH) or from the empirical equation.

Reply: Yes, we agree and have modified this paragraph as suggested. Also reviewer #3 suggested to write *g*(RH) instead of just using *g*, so we changed it throughout the manuscript.

4.4.1 MAX-DOAS

The error bars for the in-situ data are for the dry neph measurements. Depending on the ambient RH, the single largest source of uncertainty is likely from the derived scattering enhancement factor or *f*(RH). An uncertainty of 1-2% in the measured RH can propagate to a very large uncertainty in *f*(RH) and the calculated ambient scattering coefficient. In the Experimental Section can you give information about the type of RH sensor used in the *f*(RH) measurement and how this sensor was calibrated? Can you give an approximate range to the uncertainty in the *g*(RH) and *f*(RH) measurements?

Reply: Yes, the exact RH inside the nephelometer is a crucial value for the determination of the exact *f*(RH). We use a combination of a Rotronic HygroClip (HC, accuracy ± 1.5% RH) and a dew point mirror (Edge Tech, Model 2000, Dewprime DF, accuracy ± 0.1°C) to determine the RH inside the nephelometer cell (see Fierz-Schmidhauser et al. (2010b) for more details). The HC inside the cell was calibrated with standard salts before and after the campaign. The dew point mirror was used to detect and correct for shifts in the HC calibration during the four months (the temperature is measured by the dew point mirror at the outlet of the nephelometer). From these calibrations, an uncertainty of 10% in the *f*(RH) measurements was estimated. This assumption is in addition supported by the good agreement between the growth factors retrieved via Mie theory compared to the H-TDMA measured values (for non-sea salt cases, see Fig. 6).

We have added in the WetNeph experimental section: "The RH inside the nephelometer cell is monitored by a HygroClip (Rotronic), which was calibrated before and after the campaign with standard salt solutions, and in addition by a dew point mirror (Edge Tech, Model 2000, Dewprime DF). More technical details can be found in Fierz-Schmidhauser et al. (2010b)."

Concerning the *g*(RH) uncertainties, we have modified the 2nd paragraph of Sect. 3.1.5 (Measurement of the hygroscopic growth factor):
"…The growth factors in this study were determined within ± 0.05, which is typical for a well-maintained TDMA system (Swietlicki et al., 2008).

In a complex location such as Cabauw, with several different aerosol sources, the particles are typically externally mixed. This is reflected in the hygroscopic growth factor spectrum by a widened distribution, or even by clearly separated growth modes, for a given particle size. The Piecewise Linear method of the TDMAinv Toolkit (Gysel et al., 2008) was used to retrieve the growth factor distributions. Although many different sources can contribute to the aerosol population, typically one of the sources dominated. Therefore, simply using the average growth factor for each distribution is sufficient to describe the temporal variation of the growth of the accumulation mode particles at 90% RH. In this work only the data at the largest dry size, 165 nm, was utilized as the larger particles contribute to the optical properties the most (Sundström et al. 2009)."

Reviewer No. 2 (anonymous)

We thank the 2nd anonymous reviewer for his or her useful and productive comments, which helped to improve the clarity and quality of the manuscript.

Overview
This paper utilizes humidified nephelometer measurements to (a) determine if there is a somewhat simple aerosol measurement that can be used to predict *f*(RH) – the scattering enhancement value due to hygroscopic growth and (b) compare in-situ extinction measurements adjusted to ambient conditions with remote sensing extinction measurements. The authors do a very nice job of pulling together information from many different instruments. The comparison with the MAX-DOAS and lidar imeasurements is especially nice as we need to be able understand how remote sensing instruments relate to the much longer term record of in-situ monitoring.

Science related comments
P29690
Lines 8-11 – the two sentence discussion of losses was vague – in what way were the attempts to characterize losses inconclusive? Be a little more detailed about the losses according to theory. I wouldn't be picky about this except that in the comparison with remote sensing instruments, losses are mentioned as a possible reason for disagreement but the reader doesn't get any feel for what the losses might be (i.e., what size of particles might be lost and the order of magnitude of the particle transmission efficiency is as a function of size – is it mostly coarse aerosol or tiny, optically inactive particles that are lost...or both?)

Reply: No direct measurements of the particle losses were made at the Cabauw tower yet (e.g. with two parallel operating size distribution instruments one at the inlet entrance and one at the basement). Nevertheless, we have made a basic estimate of the particle losses using the aerosol loss equations from Baron and Willeke (2001). The result for the Cabauw inlet (input values: 60 m long pipe with inner tube diameter of 45 mm, tilted by 0.5° from the vertical position, flow rate of 60 lpm) can be seen in Figure B (below) together with the differential scattering coefficient $d\sigma_{sp}/d\log D$ (derived from the mean number size distribution measured between 4-18 July 2009 and an assumed refractive index of 1.54+0.01i via Mie theory). In the optical active diameter range of 0.1 to 1 µm the losses are below ~10-20%. Below 0.1 µm losses through diffusion clearly reduce the collection efficiency, while the same is true for particles larger than ~1 - 2 µm where losses through sedimentation will occur. The diffusion losses occur for rather small particle which are less relevant for the extinction at our wavelengths, while the sedimentation losses occur at a size ranges where the aerosol number concentration is comparable small. The MAX-DOAS or LIDAR measurements were performed and compared for wavelengths between 355 and 477 nm where the influence of larger particles is much smaller compared to larger wavelengths like 700 nm.

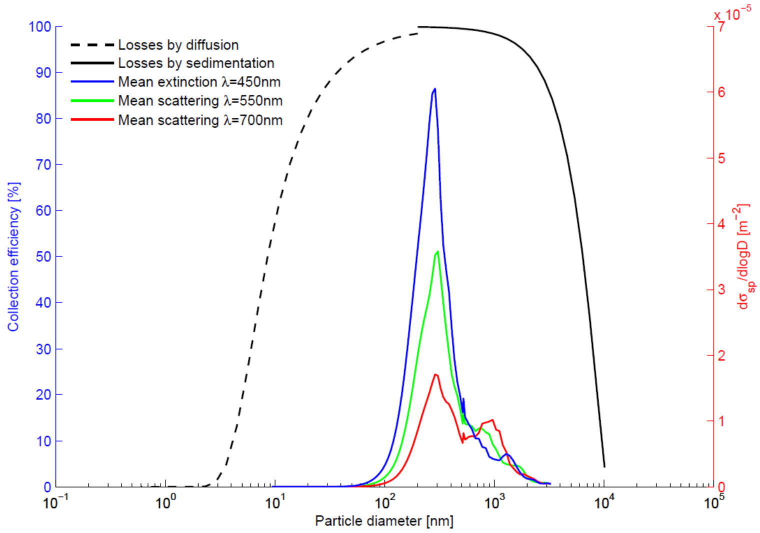

Figure B. Estimated losses for the inlet at Cabauw and the calculated differential scattering coefficient (using the mean size distribution measured between 4 July – 18 July 2009) for the three nephelometer wavelengths (assuming a refractive index of 1.54+0.01i).

In addition, the good agreement of the optical parameters calculated from the measured size distribution (APS and SMPS) points towards no significant losses within the different branches of the tubing system located in the basement of the tower, so the main spot for possible losses would be located before the branching, most probably in the Nafion dryer located at the inlet entrance. We would estimate the total losses to be lower than 10 – 20% to be measured in the scattering coefficient. For clarification, we have added the following in Sec. 3.1.1. (Inlet system): “Losses through diffusion (for smaller particles with D<0.1 µm) and sedimentation (for larger particles with D>2 µm) are expected to be below 10 - 20%. Since the main contribution to the extinction in the visible nephelometer wavelength will be in the size range between 0.1 and 1 µm, the effect of particle losses on the dry extinction coefficient is assumed to be smaller than 10 - 20%."

P29692
Out of curiousity – why not use the Collaud Coen 2010 correction for the MAAP? I think it's more different than merely changing the value of 'C' in the Weingartner correction. (i realize that for the purposes of comparing with ambient extinction it makes little difference).
Reply: Coen et al. (2010) discuss various aethalometer corrections and not a specific MAAP correction procedure. The correction value of C=4.09 was taken for the aethalometer and is specific for the Cabauw site. The Weingartner correction is a simplified procedure and absolutely suitable for our application. Only the spectral dependence (Ångström exponent) was taken from the aethalometer (if available) the absolute value for the absorption coefficient was taken from the MAAP which is assumed to be more precise.

P29701
Lines 1-27 – I could not follow the discussion in this paragraph and it needs to be rewritten for clarity. I think the point is that both chemistry and size distribution play a role in determining *f*(RH) but the role of each differs depending on the actual aerosol and perhaps chemistry is most important most of the time. One possible re-write could be making two paragraphs one describing the correlations (or lack thereof) with *f*(RH) and a second paragraph doing the same for *g*(RH). Some other things that might improve clarity would be to provide some scattering size distributions (i.e., dscat/dlogdp vs dp instead of dNdlogdp vs dp) for different types of aerosol – you should be able to do that using the measured size distributions and the mie code. I would imagine (perhaps incorrectly) that for the clean marine and slightly polluted marine most of the scattering at both low and high RH is for the larger particles. You could also present scatter plots of the different properties in figure 3 vs *f*(RH) colored by air mass type. It might at the very least be helpful to show a plot of *g*(RH) and *f*(RH) – either a scatter plot or a time series (maybe for the time period covered in figure 6), colored by some parameter such as airmass origin or coarse mode fraction. I'm not saying any of these plots would be required, I'm just trying to help figure out the best way to explain what you're trying to explain. I am particularly confused by the sentence: 'The rather low correlation to *f*(RH) and the significant correlation to *g*(RH) can be explained by the fact that a larger coarse mode fraction is an indicator for the presence of sea salt, which exhibits a higher hygroscopic growth while a larger BC fraction is an indicator for anthropogenic pollution with a reduced hygroscopic growth.'
Both *f*(RH) and *g*(RH) are indicators of hygroscopic growth of the aerosol and the *f*(RH) curves for different aerosol types in figure 2 suggest one might expect correlation between *f*(RH) and larger coarse mode fraction and/or BC fraction.
Reply: Yes, we have re-written that paragraph (also the sentences which were unclear to the reviewer). We have also moved the part discussing the trajectories to the previous Sect. 4.1. (WetNeph analysis). See revised manuscript.

The time frame with concurrent aerosol in-situ measurements was, compared to the entire campaign, quite short (4-18 July, same time period as used for the closure study). Showing the time series of all those measurements would be too confusing. We already show the main and most complete time series of the scattering enhancement plus the scattering and absorption coefficients to get an idea of the magnitude of the effect (in addition to Fig. 2 which shows the humidograms). Figure 3 is supposed to show in a summarized way the correlations of other intensive parameters to *f*(RH) and to demonstrate what parameters would be important to predict *f*(RH) (which will be discussed further in the closure study). Showing all different kinds of scatter plots might mislead the reader to find (and maybe use!) functional descriptions of certain relationships, which were not clearly seen. However, we have decided to add an additional figure in the review comments that covers the period 4-18 July and which hopefully further helps to communicate our message (see Figure C below).

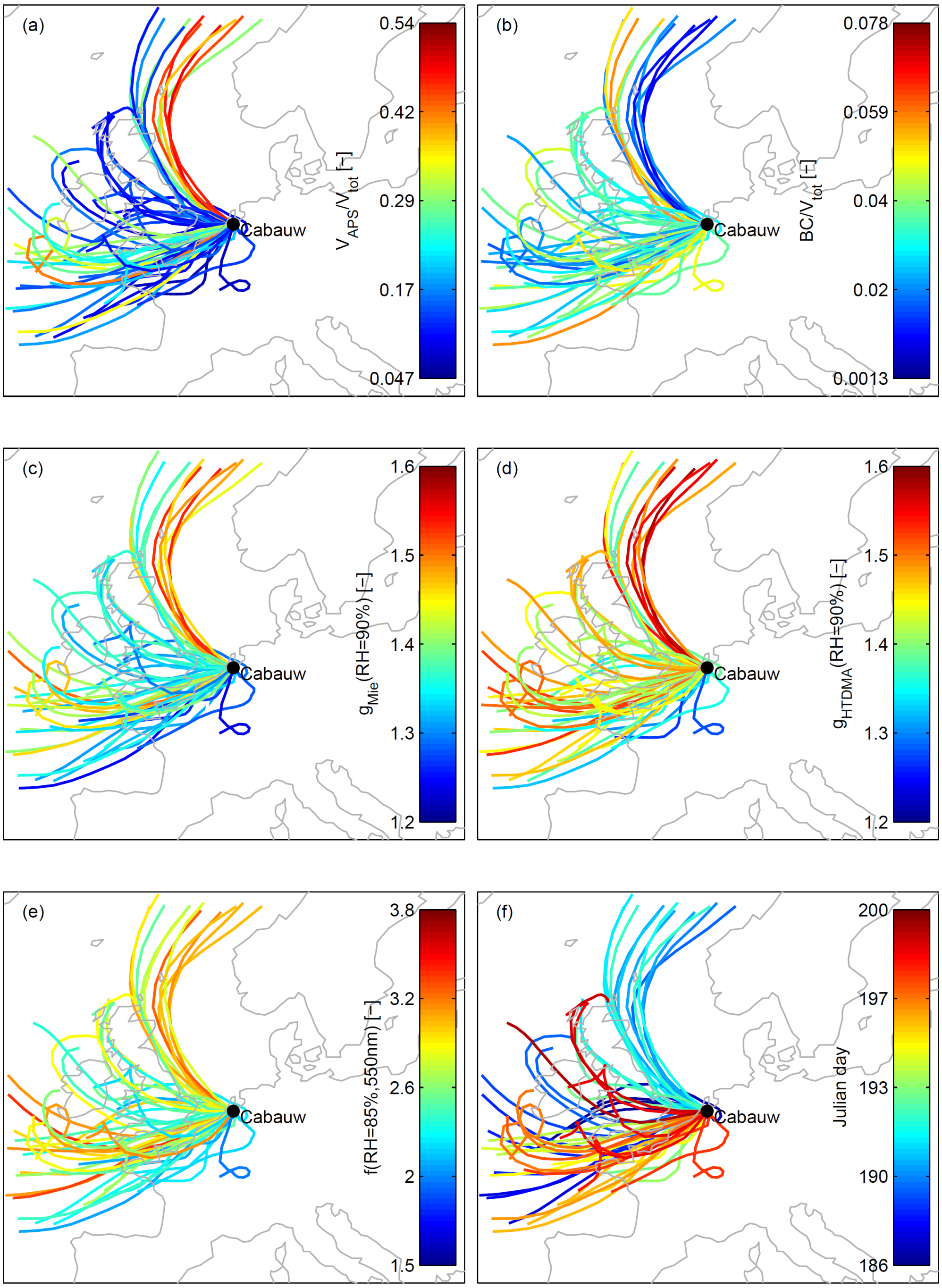

Figure C. 48-hours backward air trajectories (FLEXTRA) arriving at Cabauw during 4 – 18 July 2009 color coded with other in-situ measured properties. Panel (a): Volume coarse mode fraction; Panel (b): BC volume fraction; Panel (c): Hygroscopic growth factor retrieved from WetNeph, DryNeph, absorption and size distribution measurements via Mie theory; Panel (d): Hygroscopic growth factor measured by the H-TDMA (at 90% RH and at the dry size of 165 nm); Panel (e): Scattering enhancement factor *f*(RH=85%,550nm); Panel (f): Julian day.

P290702

Line 17 – ‘inversion of the dry scattering and absorption coefficients’ just for clarity: (a) does this mean you are adjusting refractive indices until calculated scattering matches measured scattering? (b) are you using a size-dependent refractive index or a constant refractive index for the whole size distribution?
Reply: Yes, this inversion means that the refractive index is changes until the measured scattering coefficient and single scattering albedo fits the calculated value. With this, we are only able to derive a mean value for the whole size distribution. For clarification, we have added the following sentence: “With this inversion only a mean refractive index (representative for the entire aerosol size distribution) can be derived.”

P29708
BIRA uses asymmetry and SSA from sunphotometer but comparison improves if use in-situ measurements. . .is this related to poor inversion results of sunphotometer data at low AOD? I believe AERONET only includes SSA and asymmetry values in their level 2 inversion products at AOD (blue) > 0.04
Reply: Yes, this could be a possible reason. In between missing AERONET measurements the SSA and asymmetry values were interpolated. We have added a sentence: “This however can be caused by the large uncertainty of the single scattering albedo and the asymmetry factor retrieved from AERONET at low AOD.”

Typos, wording suggestions
Please use g(RH) for growth factor. This makes it more consistent with the μ (RH) usage and also the letter g is used for other aerosol parameters such as asymmetry parameter. LIDAR is capitalized in title but not in text...
Reply: g(RH) and LIDAR is now being used consistently within the text.

P29685
Line 5 – add the word ‘also’: . . .are ALSO strongly dependent. . .
Reply: Changed accordingly.

Line 20 – second ‘needs’ should be ‘need’: . . .and size distribution NEED to. . .
Reply: Changed accordingly.

Line 25 – add ‘from MAX-DOAS’: . . .extinction coefficients FROM MAX-DOAS were.
Reply: Changed accordingly.

Line 27-28 – change sentence: Differences WERE SHOWN to be dependent on the MAX-DOAS retrieval algorithm applied.
Reply: Changed accordingly.

Line 28 – add the word ‘extinction’: . . .in-situ EXTINCTION data. . .
Reply: Changed accordingly.

P29686
Line 10-11 move the word ‘also’ on line 10 to before ‘strongly depend’ on line 11
Reply: Changed accordingly.

P29687
Line 24 – add the word ‘almost’: . . ..but ALMOST always. . . (since you provide the Morgan et al 2010 reference which does use humidified neph data)
Reply: Changed accordingly.

P29688
Line 12 – change ‘was’ to ‘were’: The data WERE compared. . .. (the word data is plural)
Reply: Changed accordingly.

Line 15 – add the word ‘also’: . . .measurements were ALSO compared. . .
Reply: Changed accordingly.

P29690
Line 2 – define MAAP and SMPS the first time they are used
Reply: Changed accordingly.

Line 20 – get rid of the word ‘and’: . . .humidifier followed by a dryer. . ..
Reply: Changed accordingly.

P29691
Line 2 – I don’t like the choice of the word ‘known’ but I’m not sure what is better – described?
Reply: Yes, we have modified the sentence: “Deliquescence is described as a sudden up-take of water of an initially dry and solid particle at the defined deliquescence relative humidity.”

Line 18 – the Anderson corrections also account for lamp non-idealities
Reply: We have added to that sentence: “… which also accounts for non-idealities of the light source in the nephelometer.”

P29692
Line 13 – rewrite: . . .larger than 0.8 most of the time. . ..
Reply: Changed accordingly.

P29693
Line 14 – rewrite: . . .which results in a reduction in size. . . .
Reply: Changed accordingly.

Line 25 – rewrite: . . .monodisperse particles are exposed to controlled. . ..
Reply: Changed accordingly.

P29697
Line 25 – replace ‘between’ with ‘over’: . . .varying LR’ OVER a range of values. . .
Reply: Changed accordingly.

Line 26 – replace 'verified' with 'determined'
Reply: Changed accordingly.

P29698
Line 5 – rewrite: The prediction of *f*(RH) without explicit wetneph measurements at Cabauw is also discussed in section 4.3.
Reply: Changed accordingly.

Line 26 – I'm guessing the 90th percentile value should be higher than 1.52?
Reply: Yes, an error has occurred here. The correct number should be 10^{th} perc=1.93 and 90^{th} perc=2.9), which has been corrected.

P29699
Line 1 – add the word 'dry' in front of absorption: . . .and DRY absorption. . .
Reply: Changed accordingly.

Lines 7-8 – rewrite: Distinct periods of lowered or elevated *f*(RH) values (see Fig 1a) were correlated with the origin of . . ..
Reply: Changed accordingly.

P29700
Line 6-8 – rewrite: The maritime slightly polluted cased (Fig 2d) reveals a similarly high magnitude of *f*(RH) as the clean maritime case (Fig 2b), but without deliquescence, while the maritime heavily polluted case is. . .. (I think you mean 'clean' instead of 'clear' in this sentence)
Reply: Yes. We have changed it accordingly.

Line 11 – Russell reference – two 'L's in Russell.
Reply: Reference corrected.

P29702
Line 21 – replace 'the one' with 'that': close to THAT of water
Reply: Changed accordingly.

P29703
Line 1 – 'low absorption' clarify – do you mean organics cause little to no absorption or that they have an absorbing component? If you mean the second should include a citation.
Reply: Yes, we want to state that the absorbing component of organics is small compared to BC. For clarification, we have modified that sentence: "… which is expected to lower the hygroscopic growth while having a minor influence on the refractive index (negligible imaginary part of the refractive index compared to BC, Nessler et al., 2005a)."

Line 7 – replace 'next to' with 'in conjunction with'

Reply: Changed accordingly.

P29706
Lines 13-20 – rearrange: put period after '. . .JAMSTEC are used'. Then move sentence 'BIRA and IUPHD retrievals to after . . .JAMSTEC are used. Then have new sentence: For the MPI data a mean aerosol extinction coefficient in the BL is estimated by retrieving the layer height and aerosol optical thickness.
Reply: Changed accordingly (Start of last sentence slightly changed to: "In the MPI retrieval a mean extinction …")

P29707
Line 6 - should be 12 pm ?
Reply: Yes, this has been corrected.

Line 13 – replace 'giving' with 'and gives': . . .being compared AND GIVES no information. . .
Reply: Changed accordingly.

Line 15 – replace ';displays' with 'display': Figures 8 and 9 DISPLAY the . . .
Reply: Changed accordingly.

P29711
Line 11 – replace 'main' with 'best': The BEST quantity to estimate. . .
Reply: Changed accordingly.

Line 13 – 'or derived from chemical composition measurements' I think you need to be careful here – I imagine you could get a useful g(RH) value from high time resolution, size-resolved chemistry measurements (e.g., AMS) but I'm guessing a g(RH) value derived from bulk filter measurements might not be as helpful (if one can even get g(RH) from bulk filter measurements!).
Reply: Yes, that's correct. And even an AMS will have problems to access the sea-salt contribution. Since we have not measured the chemical composition anyway, we've deleted that part of the sentence. New: "The best quantity to estimate f(RH) from other continuous in-situ measurements was found to be the hygroscopic growth factor measured e.g. by a H-TDMA."

Line 14 – replace 'provide favorable results' with 'correlate well with f(RH)'
Reply: Changed accordingly.

Reviewer No. 3 (anonymous)

We thank the 3rd reviewer for his or here helpful and productive comments, which significantly helped to improve the quality of the revised version of our manuscript.

Overall Quality of the Paper:

Measurements of the aerosol scattering enhancement factor *f*(RH) are presented for the Cabauw site for the first time and represent a sound contribution to additional scientific knowledge of *f*(RH), and is regarded as a main strength of the paper. This is complimented by quite a good closure study using a comparison between measured and calculated *f*(RH) values. The comparison of an in-situ extrapolated/interpolated aerosol extinction coefficient from a single point (60m altitude) with remote sensing data is less convincing and is considered to be a definite overall weakness of the paper. Some main weaknesses of the work stem from (a) a comparison of in-situ single point extinction coefficient with values inferred from MAX-DOAS aerosol extinction coefficient values – not yet validated in the open literature – and (b) a comparison with lidar signals requiring extrapolation from a height of about 750 m to close to ground. Referenced work is very good and the paper on the whole is clearly written. More detailed comments are given below.

Comments on results

(i) In-situ data

The work is centred on measurements and calculations to examine the effect of relative humidity on aerosol scattering at the Cabauw atmospheric site in the Netherlands. Aerosol optical and microphysical measurements were performed at the site over the period from the 19th June up to about 4th October, 2009. The aerosol measurements were taken at a 60m height on a 213m tower, which also housed meteorological sensors including that of temperature and dew point (relative humidity) at 6 heights between 10m and 200m. Results of the aerosol scattering enhancement factor *f*(RH) are presented for the Cabauw site for the first time. Closure in the form of a comparison between measured *f*(RH) and calculated *f*(RH) is quite good, which is helped by the fact that closure is made at a high relative humidity (85%), reducing dependencies on particle chemical composition (and therefore on particle refractive index). The assumption of a constant growth factor *g* (for particle diameter 165nm) with size, is likely to be incorrect, and is acknowledged by the authors. Indeed, the work of Swietlicki et al (2008) shows from HTDMA field data, that the growth factor g generally increases with particle size for rural, continental and marine aerosol – which will lead to an underestimation of calculated *f*(RH), as reflected by the closure study results in Figure 5(a). In the discussion of the effect of particle size and hygroscopicity on *f*(RH), a larger *f*(RH) is attributed to non-linearity in Mie-scattering, which could be elaborated upon in a more specific manner, since Mie non-linearity is a sensitive function of size (or size parameter) and refractive index. Indeed, Mie theory calculations show that the efficiency factor for scattering does increase, at visible wavelengths, with a decrease in imaginary index of refraction (as the particle becomes more hygroscopic).

Reply: We agree, maybe we haven’t enough emphasized this fact in the corresponding paragraph. For a fixed chemical composition (and fixed wavelength), *f*(RH) decreases with increasing particle diameter, as we have observed and modeled for Artic aerosol (see Fig. 9 in Zieger et al. (2010) where *f*(RH) has been calculated for different mean diameters and different NaCl to organic mass ratios). The influence of size and chemical composition on *f*(RH) is also the reason not to give an analytical equation for *f*(RH) as a function of *g*(RH) as asked by the other reviewer, although the correlation is comparable high. An additional Figure with model calculations would be beyond the scope of our manu-

script and has already been shown in other publications, but we have modified that sentence for more clarity:
“This may also point towards effects of non-linearity in the Mie-scattering, where both size and chemical composition are input parameters. If the chemical composition (hygroscopic growth and refractive index) is assumed to be constant for a given wavelength, f(RH) will decrease with increasing particle size. This can be compensated if the size changes concurrently with its hygroscopicity. A similar effect was e.g. observed and modeled for Arctic aerosol (see Fig. 9 in Zieger et al. 2010), where smaller but less hygroscopic particles had a similar magnitude of f(RH) compared to larger but more hygroscopic particles (in that case the coarse mode was also dominated by hygroscopic sea salt).”

Of course, the prediction of f(RH) remains quite a challenge in the absence of knowledge of aerosol chemical composition. In that regard, the assumed dry particle chemical composition is not given and should be specified. $(NH_4)_2SO_4$ was chosen for Arctic aerosol (Zieger et al, 2010), but is probably not appropriate for the Cabauw site.
Reply: Unfortunately no aerosol chemical composition measurements were available during the campaign. We have retrieved the dry refractive index using the measured dry absorption and scattering coefficients as well as the measured dry size distribution. Of course, one could argue that this procedure is going round in circles, but – as already mentioned – the closure was performed at high RH and in addition repeated with a fixed dry refractive index, which does not significantly change the agreement between the calculated and measured scattering coefficients since the measured number size distribution governs the magnitude of the calculated scattering coefficient. Using m=1.599+0.024i for 550 nm (used in Fierz-Schmidhauser et al. (2010a) for polluted air at Mace Head, Ireland) gives y=(1±0.014)x+(2.2e-07+2.4e-7)m^{-1} and R^2=0.94, similar to the one given in Fig.5a. This is then reflected in a slightly worse agreement between calculated and measured f(RH) (y=(0.64±0.029)x+(0.57+0.078), R^2=0.61) using the growth factor of the H-TDMA, caused by the calculated dry scattering coefficients which are in the denominator in the equation for f(RH).

We have clarified this at the end of the third paragraph in Sect. 4.3 (closure study):

“Keeping the dry refractive index at a fixed value does not significantly change the agreement within this closure study. Despite the fact that the number size distribution dominates the magnitude of the calculated dry scattering coefficient the variation of the dry refractive index still has an influence. Taking e.g. m=1.5291+0.024i at 550 nm (used in Fierz-Schmidhauser et al., 2010 for polluted air at Mace Head, Ireland) gives y=(1.0±0.014)x+(2.2e-07±2.4e-7)m^{-1} and R^2=0.94 for the comparison of the wet scattering coefficients to the calculated values (analog to Fig. 5a). For the comparison of the measured and calculated f(RH) using the growth factor of the H-TDMA (analog to Fig. 5b) gives a slightly lower agreement y=(0.64±0.029)x+(0.57±0.078) and R^2=0.61.

To further demonstrate the effect of the limited size range of the H-TDMA measurements for the closure study, the hygroscopic growth factor ...”

The paper would have benefited through examining more thoroughly the variation of *f*(RH) with air mass type, through a more detailed classification of air mass back trajectories, with respect to sector direction, distance from source, and possibly through extending the air mass travel time from 48hours. Since the Cabauw site is inland from the North Sea, varying in distance from about 60 to ~ 100km, depending on direction from the ocean, all so-called maritime air parcels are de facto 'modified maritime'. The authors should describe how 'maritime' air masses are differentiated from 'maritime slightly polluted'. The distinction between 'maritime slightly polluted' and 'maritime heavily polluted' should also be quantified.

Reply: The aerosol at Cabauw is highly variable in size and composition. We have tried to classify the air masses arriving Cabauw using the FLEXTRA trajectories to categorize them into groups to predict *f*(RH) for certain air mass types. We have also tested longer air mass travel times. Unfortunately, this attempt was not successful due to the high variability in the aerosol composition. For example, air masses directly coming from the oceans can be pure sea salt or be mixed with local anthropogenic pollution just before reaching Cabauw. A climatology that could also be used by users would need in our opinion a longer measurement period covering at least a full year. So we have decided to show only examples of averaged humidograms only selected individually applying certain criteria which is clearly mentioned in the text. Nevertheless, for clarification we will mention the selection criteria used in the revised manuscript. We have modified the following sentences in the second paragraph of Sect. 4.2. (Factors influencing *f*(RH) at Cabauw):

"A typical maritime case is presented in Fig. 2b (selection criteria used: direction of arriving air parcel between 45° < θ < 315°; *f*(RH=85%,550nm) > 3.5, average of 4 humidograms)."

"The maritime slightly polluted case (Fig. 2d; with 225° < θ < 315° and *f*(RH=85%,550nm) > 3, average of 31 humidograms) reveals a similarly high magnitude of *f*(RH) as the clean maritime case (Fig. 2b), but without deliquescence, while the maritime heavily polluted case is characterized by much lower values of *f*(RH) (see Fig. 2e; with 225° < θ < 315° and *f*(RH=85%,550nm) < 2, average of 25 humidograms)."

"Figures 2c and 2f show two examples of air masses having a continental origin (continental south: 135° < θ < 225° and *f*(RH=85%,550nm) < 2, average of 48 humidograms; continental east: 60° < θ < 135°, average of 75 humidograms)"

We have also modified the last sentence of this paragraph:

"A simple and generalized categorization using the air mass trajectories could not be established due to the high variability of size and composition and the short measurement period. For a better statistical analysis a longer time period of at least a year would be desirable."

The requirement of a second nephelometer to measure the aerosol scattering coefficient under dry conditions begs the question as to how the 'wet' nephelometer compared with the dry nephelometer under identical low relative humidity dry conditions? This is not addressed in the paper and should be.

Reply: Yes, of course, the comparison of the dry nephelometer and the WetNeph system measuring at dry conditions (humidifier turned off) has been done (in addition to the nephelometer span gas calibration before and after the campaign). We have forgotten to mention that in the manuscript and have added:

"Both nephelometers were calibrated (with particle-free air and CO_2) and compared directly (WetNeph without humidifier system). In addition, the scattering coefficients at dry conditions ($RH_{wetneph}$<40%) were compared for the entire campaign. From these measurements it was found that the WetNeph scattering coefficients at dry conditions were slightly higher than the ones of the DryNeph (450 nm: $\sigma_{wetneph} = 1.07*\sigma_{dryneph}+8.7*10^{-7}m^{-1}$, $R^2=0.99$; 550 nm: $\sigma_{wetneph} = 1.06*\sigma_{dryneph}+6.7*10^{-7}m^{-1}$, $R^2=0.97$; 700 nm: $\sigma_{wetneph} = 1.03*\sigma_{dryneph}+4.5*10^{-7}m^{-1}$, $R^2=0.94$), which was caused by differences in the absolute calibration of the nephelometer (WetNeph nephelometer measured directly higher scattering coefficients) and losses in the humidifier (~5%, see Fierz-Schmidhauser et al., 2010a). The WetNeph measurements were therefore corrected accordingly. "

The aerosol size distribution is measured using a combination of a SMPS (up to aerosol particle diameter of 520nm) and an APS which measures from a diameter of 500nm. Data from the 2 instruments is used to yield the complete size distribution, but how good was the overlap between the 2 instruments? There is no discussion of this in the paper, and certainly should receive attention.

Reply: We have found that the APS-SMPS agreement to be sufficiently good for our purposes. In the figure below, mean surface and volume size distributions are shown, which have been categorized according to their volume coarse mode fraction measured by SMPS and APS. The fine mode (d<500nm) dominates for most of the cases and the APS coarse mode fits well to the SMPS. Small differences were found for large volume coarse mode fractions (most probably sea salt), which might have been caused by non-sphericity and density effects in the APS size attribution. This is a commonly known problem of the APS sizing as can be found in the literature. For our purposes, we concluded the agreement between SMPS and APS to be sufficient and did not apply any further time (air mass) dependent corrections to the APS. The SMPS is definitely the more important instrument to measure optical relevant particles for our wavelengths (see differential scattering coefficient $d\sigma/d\log D$ in the Figure below). We have added in the instrumental section:

"The overlap between the SMPS and APS showed to be good for most of the cases. Small differences seen in the transition of the volume size distribution were caused by variations in density and shape influencing the APS sizing. However, they were found to be negligible for our purposes, since the scattering coefficient is dominated by contributions from the fine mode (D<500 nm, measured by the SMPS). The measured volume size distributions could be well fitted using a three modal lognormal size distribution equation."

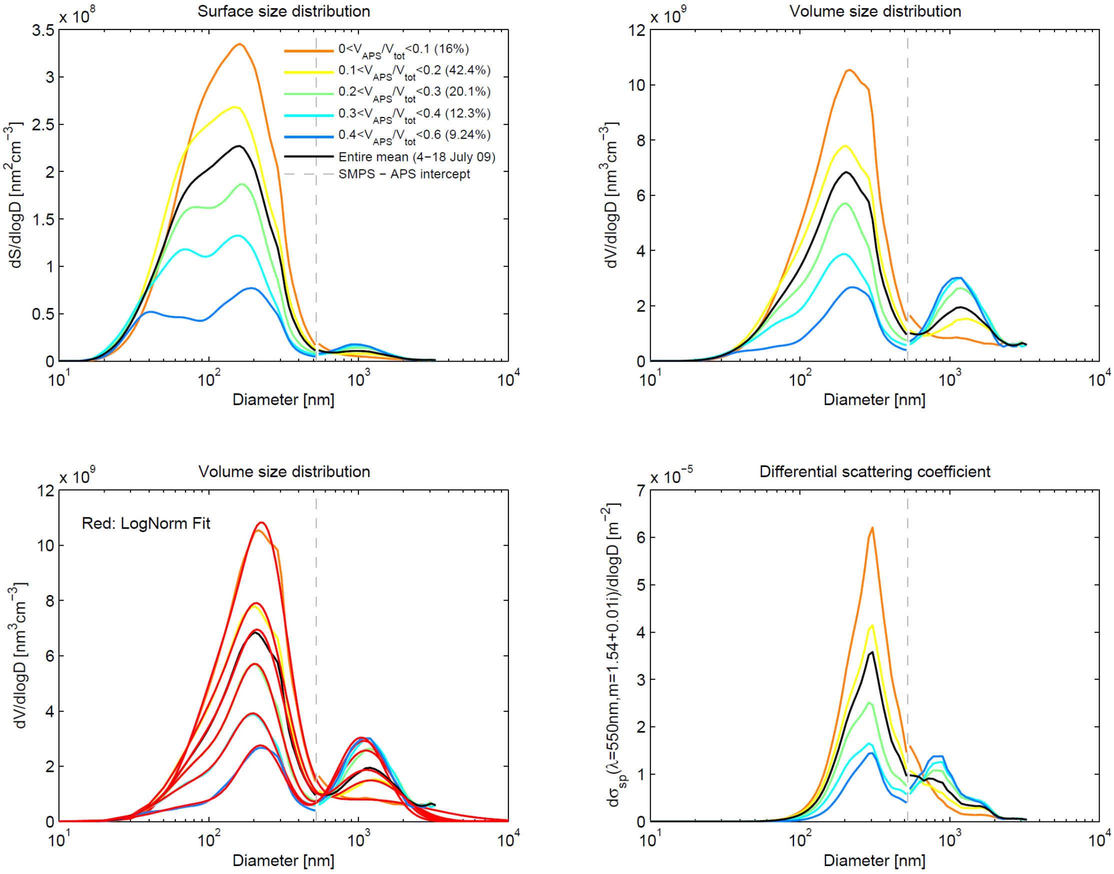

Figure D. Mean size distributions measured by the SMPS (<520nm) and APS (>520nm). The size distributions were averaged according to their coarse mode volume fraction (V_{APS}/V_{tot}) to show the influence of air mass types. The percentage of occurrence is given in the legend. The black curve represents the mean of the entire period (4-18 July 2009). Panel (a): Mean surface size distribution; Panel (b): Mean volume size distribution; Panel (c): Mean volume size distribution fitted with a 3 modal lognormal distribution; Panel (d): Differential scattering coefficient (for 550 nm and a refractive index of 1.54+0.01i).

Use is made of measured aerosol absorption coefficients at a range of wavelengths, using an Aethalometer Model AE-31, to derive an absorption Ångström exponent α_{ap}. The conversion of aerosol absorption coefficient as measured by the MAAP at the 'operational' wavelength of 637nm to a different wavelength is performed through the use of Eq. (6), i.e. using an absorption Ångström exponent α_{ap}, determined from a different instrument – an Aethalometer. However, there is no certainty that the 'instrumental' absorption Ångström exponent α_{ap}, obtained from the Aethalometer measurements at a range of wavelengths is necessarily the same as would be obtained by a MAAP, had it had a variable wavelength capability, in view of the different measurement principles and characteristics of the 2 instruments. This should be discussed and justified in the revised version of the paper.

The use of a constant value for Ångström exponent α_{ap} of 0.84 based on an initial relatively short period (~ 1/6 of the total measurement period) of the campaign, from ~ 19th June to July 6th, and assumed to be valid thereafter throughout the remaining period up to ~ 4th October, is certainly questionable, and needs both justification and discussion.

Reply: We think that the use of the measured Ångström exponent from the aethalometer and the assumption of a constant value for the absorption coefficient is justified in our case, because of the following reasons:

- The variation of the Ångström exponent within these 2.5 weeks was not very large (mean 0.84, 10th percentile: 0.71, 90th percentile: 0.98). Usually a value of 1 is used in the literature for urban aerosol (see e.g. Russell et al., 2010, ACP).
- The Ångström exponent is only needed to calculate the ambient extinction coefficient, where the scattering coefficient is clearly the dominating factor and the assumption of different constant Ångström parameters has only a negligible influence on the ambient extinction coefficient. This is demonstrated in Figure E below, where we have calculated the ambient extinction coefficient using other constant Ångström parameters for the absorption (1.0, 1.5, and 2.0). A value of around 1 would be a typical range for urban polluted aerosol (Russell et al. (2010)) and the comparison shows that the result would only change our ambient extinction coefficients by less than 1%. Also higher values of 1.5-2 have only a slight influence of 1-2%. A value of 2 would probably be a too high assumption at Cabauw, since it is a typical value for mineral dust Russell et al. (2010).

For clarification, we have added the following sentence in Sect. 4.4 (Comparison to remote sensing data):

"The assumption of a constant value of α_{abs} is justified in our case because of the low variation of the measured value (10th percentile: 0.71, 90th percentile: 0.98) and due to the negligible impact of α_{abs} on the ambient extinction coefficient where the scattering is the clearly dominant part (e.g. taking 1 or 1.5 as a fixed value for α_{abs} would increase the ambient extinction coefficient only by a factor of 1.002 or 1.01, respectively)."

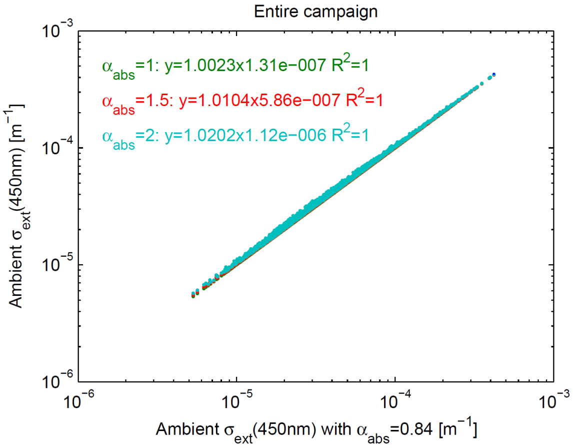

Figure E Ambient extinction coefficient recalculated taking fixed Ångström exponent for the absorption coefficient compared to the one used in the script. The comparison is shown for the entire campaign.

Calculated volume concentration of BC from BC mass concentration requires an assumed value for density of BC particles, which is not given in the paper.

Reply: Yes, we have used a density of 2.1 g/cm^3 (see http://www.inchem.org/documents/icsc/icsc/eics0471.htm) and have added this information in the manuscript. The value might be at the upper limit for possible BC densities, but it is suitable for our usage since we used a fix value to check for possible correlations of the BC volume fraction to other in-situ parameters and to parameterize the hygroscopic growth factor.

(ii) Comparison to remote sensing data
The paper describes the attempt to compare in-situ aerosol extinction coefficient (based on measured aerosol scattering coefficient + aerosol absorption – assumed not to vary with RH) at a single point (60m above ground), to aerosol extinction coefficient inferred from MAX-DOAS profiles for lowest altitude levels varying from 0-200m up to 20-5000m. Comparison of a single point measurement with profile measurements is at the best of times a seemingly precarious scientific exercise, since it requires a host of assumptions, so that attainment of apparent fair agreement using the 2 approaches may simply be just fortuitous. There is no evidence provided that the assumption of a well mixed aerosol layer is justified for each of the periods when in-situ versus MAX-DOAS intercomparisons were made.
I would have thought that available ceilometer data or lidar data would have provided evidence of aerosol layering? The MAX-DOAS technique has been shown to yield quite reliable retrieved profiles of gaseous components, such as NO_2 and O_4 (Roscoe et al, 2010). However, as far as the referee is aware, no published work has appeared to date on the intercomparison of the MAX-DOAS technique to reliably validate aerosol extinction coefficient profiles, with other profiling techniques such as lidar. Reference is made in the paper to a paper in preparation (Friess et al, 2010), but this has not yet appeared in print. In the absence of such validation work, doubt lingers in the referee's mind as to the reliability of MAX-DOAS inferred vertical profiles of aerosol extinction coefficient, presented in Figures 7 to 10.
Reply: We disagree with the reviewers' opinion that the MAX-DOAS in-situ comparison is just a “precarious scientific exercise”. We have shown that for certain conditions (low AOD and low PBL) an agreement exists between ambient in-situ and MAX-DOAS (see e.g. the golden day in Fig. 7) and found a good correlation between the different measurement techniques. This can not just be “fortuitous”. The MAX-DOAS retrieval is most sensitive to the lower height levels, where the LIDAR is blind due to the overlap problem, which in addition justifies such an approach as presented here. Nevertheless, the disagreement for most of the cases is an important finding for the MAX-DOAS community and will help to further quantify the capabilities (and limits) of this novel technique. It should also be mentioned that in a recent study by Li et al. (2010) a good agreement was found between aerosol extinction coefficients retrieved from MAX-DOAS and in-situ measurements.

The following reasons are brought forward to further justify our approach:

- The assumption of a well mixed layer between 0 - 200 m is an inevitable one, due to the configuration of the MAX-DOAS retrieval. The assumption of a constant aerosol type (=chemical composition without water content) is probably justified in this rather small layer during daytime measurements. An exception is the MPI group, which uses a simpler retrieval technique and therefore our assumption of a well mixed layer had to be extended to the actual retrieved layer height, which is of course more arguable compared to the one for the 0 - 200 m layer (this is clearly stated in the manuscript). At Cabauw, we are in the good position to have continuous relative humidity measurements within the 0 - 200 m layer, since the RH is one of the main factors influencing the extinction coefficient within this layer. Of course, for MPI the assumptions become more problematic since we had to use RH data from a weather model (COMSO). The good correlation to the four different MAX-DOAS retrievals - despite their differences - is a sign that the assumption of a well mixed layer can be made for this rather small level.

- The agreement of columnar densities is not a sign that the actual profile is correct (as we have stated in the paper). Roscoe et al. (2010) only compared slant column densities recorded during the CINDI campaign and do not show the agreement of the vertical profiles of NO_2 or O_4, they also do not show a validation by other independent measurement techniques. This has still to be shown, which is work in progress.
- The same is true for the vertical profiles of the aerosol extinction coefficient. Yes, there are only a few studies comparing the vertical extinction profiles with independent LIDAR measurements. Irie et al. (2008) and Irie et al. (2009) i.e. made comparisons between lower-tropospheric vertical profiles of the aerosol extinction coefficients retrieved from JAMSTEC MAX-DOAS and coincident LIDAR observations at Tsukuba, Japan. They found reasonable agreement for layers of 0-1 and 1-2 km to within 30% and 60%, respectively, for most cases. However, these very few studies also show the need for further independent validation studies like ours. Frieß et al. are currently analyzing the retrieved profile shapes of the different groups. To include these results and the discussion would go beyond the scope of our work and has to be discussed in a separate publication.
- For clarification, we have added in the manuscript the following sentence:

Added after the first paragraph of Sect. 4.4.1: “It should be pointed out that the comparison of the lowest MAX-DOAS extinction coefficient with in-situ measurements is of special interest since the MAX-DOAS retrieval has its highest sensitivity at the ground (Frieß et al. (2006)) while LIDAR measurements are usually challenged with the overlap problem at low altitudes. In a recent study (Li et al., 2010), good agreement was found between aerosol extinction coefficients retrieved from MAX-DOAS and surface in-situ measurements. MAX-DOAS aerosol extinction coefficient profiles have only been compared in very few studies. Irie et al. (2008) and Irie et al. (2009) made comparisons between lower-tropospheric vertical profiles retrieved from JAMSTEC MAX-DOAS and coincident LIDAR observations at Tsukuba, Japan. They found reasonable agreement for layers of 0-1 and 1-2 km to within 30% and 60%, respectively, for most cases. However, these very few studies also show the need for further independent validation studies like the one presented here.”

There are also some issues with the comparison of in-situ aerosol extinction coefficient, with that inferred from lidar. As pointed out by the authors, the lidar ratio (LR=aerosol extinction coefficient/aerosol backscatter coefficient) can be directly measured by the CAELI lidar above about 750m in altitude. Comment on how those lidar ratios compare

with previously measured values for similar type air masses would be informative. For example, Rosen et al, 1997 (in JGR) present measured LR values for near surface aerosols over the south western USA. Are the values of LR at the levels below 750m taken to be equal to the value of LR measured directly at the lowest possible level (at ~750m)? If so, this presumes that the layer between ~ 750m and ground is well mixed, but is there is no evidence presented for such an assumption, for each of the 22 profiles. What are the error bars associated with the inferred lidar aerosol extinction coefficient with height in Figure 11, which are desirable in order to better inter-compare with the in-situ values? Is there a reason as to why the mean ambient aerosol extinction coefficient in Figure 12, determined by the lidar, does not have accompanying error bars? It is rather odd that mean values for certain profiles do not seem to correspond with individual data points for some of the data shown. For example, three visible night-time square values (largest value of ~ 1.9 on y-axis), above about an in-situ value of 0.3 x 10^{-4} m^{-1} do not seem to have a mean value shown?, or am I simply misreading the points? Similarly, there are 4 vertical circular gray (daytime points) with largest value ~2.85 – is their mean value shown?
In summary, I do believe there is need to give more information on the data presented in Figure 12, probably in the form of a Table – number of points used to obtain each mean value – to be accompanied by a standard error and value of LR used for each of the 22 cases, corresponding to a specified (thickness) minimum level. On the face of it, the data below 1x 10^{-4} m^{-1} for σ_e (355nm) looks more like a scatter-plot, and it is difficult to see any good degree of agreement between the in-situ and lidar values in the range of σ_e values between 0 and 1x 10^{-4} m^{-1}.

> Reply: We agree that further information has to be added to the LIDAR/in-situ comparison and that Fig. 12 might have been a bit confusing to the reader in its original form, so we have substantially modified this Figure. However, we are still convinced that our approach is justified because of the following reasons:
>
> - Rosen et al. (1997) report LR from near-surface aerosols of an arid region in the southwestern U.S., which on the one hand is probably not comparable to the aerosol found at Cabauw, where more urban polluted and maritime aerosol is found and on the other hand is not measured by a LIDAR instead a combination of nephelometer and backscatter sonde is used. Nevertheless, they find values around 40 sr and compare their results to other studies where the LR e.g. measured in Japan ranged from 20 to 70 sr or in Tucson (U.S.) measurements showed LR around 20 (±9) sr. A good overview on measured LR is given by Müller et al. (2007) where observed LR from different aerosol types are being discussed. For marine aerosol typical values are 23 (±3) sr (at 532 nm, observed at the North Atlantic), and for urban haze in central Europe 58 (±12) sr (at 355 nm) are reported (both measurement refer to values in the PBL). The influence of RH on the LR for different aerosol types is nicely discussed and shown e.g. in Ackermann (1998). LR for continental aerosol might vary between 0 to 99 % RH in the range of roughly 40 – 70 LR, while for maritime aerosol the range is roughly between 15 and 25 sr (both at 355 nm). These ranges were also observed with the CAELI LIDAR (observed values LR ~ 8 – 60 sr). To demonstrate the effect of RH on the LR, we have added a second panel to Fig. 12 showing the LR measured in the individual layers between 700 and 1700 m versus the RH of the individual layer

taken from the COSMO weather model. One can generally observe for most of the cases an increase of LR with increasing RH in the same range as e.g. modeled in Ackermann (1998). Of course, it has to be assumed that the aerosol type is constant throughout this layer, which is certainly not given for all profiles. In addition, we have used these LR-RH-relationships to calculate (interpolate) the LR for the mean RH measured at the ground. These points were added in the scatter plot (red crosses). No extrapolation was performed due to the unknown functional relationship (so the number of compared profiles decreased from 22 to 15). As described in the paper, the LIDAR extinction coefficients were determined by averaging the measured backscatter coefficient below 200 m and multiplying this value with the measured LR of the upper layers. Instead of showing these individual points as in the original version, we have replaced these points by error bars giving the range of the retrieved extinction coefficient if the minima and maxima value of LR are taken to calculate the ground value. These error bars are therefore an indicator of the variability and uncertainty of our approach. Taking the measured LR from the lowest level is probably still a feasible way since it is probably still the most representative value. We have kept the linear (orthogonal) regression lines in Fig. 12 (now Panel b), but have added a third regression line for those points were the LR was interpolated to the value of the ground RH. The overall message has not changed but we have rewritten this section completely (see below).

- The averaging time of the LIDAR extinction profile was (1.85±0.5) hours (mean±std).
- The points that were missed by the reviewer were present in the plot (close to the 1:1 line, maybe a bit hidden behind the regression lines, but visible). In the new version this should be better visualized using only single points and error bars.
- We have found a minor error in the routines that calculated the average backscatter coefficient in the lowest layer (below 200 m), therefore the slopes and R^2 slightly changed (improved) compared to the previous version.

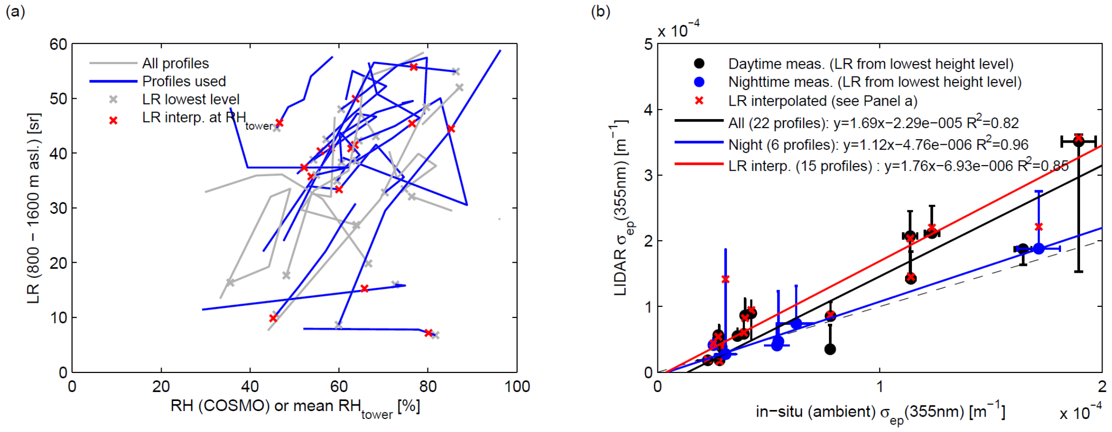

Figure F. Panel (a): The LIDAR ratio LR measured in 200-m altitude intervals between 700 and 1700 m versus the RH in the individual layer (taken from COSMO). Grey crosses denote the LR of the lowest layer, red crosses show the interpolated LR for the RH measured at the ground (mean ~60-200 m). The profiles used to retrieve the interpolated value are shown in blue. Grey are all profiles

(where no interpolation was possible or where no WetNeph measurements were available). Panel (b): Comparison of the extinction coefficient σ_{ep} at the ground retrieved from LIDAR vs. the in-situ values (mean for ~60-200 m). Circular points denote the mean value if the LR from the lowest level is taken (black daytime measurements; blue nighttime measurements). Red crosses show the mean value if the LR interpolated to the ground RH is taken (see Panel a). The error bars in the y-direction give the range of the retrieved σ_{ep} taking the measured minima and maxima LR of the upper layer. The x-error bars give the calculated error in the ambient in-situ measurement. The solid lines represent linear orthogonal regression (see legend). (This figure will replace Fig. 12 in the manuscript.)

Rewritten Section 4.4.2:
Due to the long averaging times, only 22 profiles (within the period 23 June--20 September, averaging time 1.85±0.5 hours (mean ± standard deviation) of the aerosol extinction coefficient measured by the CAELI LIDAR could be compared to the in-situ measurements. The aerosol extinction coefficient (at 355 nm) can be measured directly using the Raman channel above approximately 750 m. The backscatter signal, retrieved using the Raman method, starts at approximately 60 m and can be used to extrapolate the direct measurement of σ_{ep} if an appropriate LIDAR ratio LR (Eq. 7) is assumed. Instead of an educated guess, the measured LR of the upper layers between 700 and 1700 m was determined (mean values for 200 m thick levels) and multiplied with the backscatter signal.

An example day is presented in Fig. 11. The extinction is directly measured above ~750 m (black line). The LR of the upper layers increase with height from LR = 37 to LR = 48 (due to changing RH and/or aerosol type changes or lower signal to noise ratio). These values are used to calculate σ_{ep} by multiplying the backscatter signal with the LR. The in-situ values at dry (black square) and at ambient conditions at the RH measurement of the tower (color coded circles) are also shown. The large RH gradient results in a strong increase of σ_{ep} concurrently determined indirectly from both the in-situ aerosol measurements and the LIDAR measurements.

The LR values are within the range as e.g. modeled by Ackermann (1998) for marine (LR=~10-25 between RH=0-99%) or continental aerosol (LR=~40-70 between RH=0-99%) or as observed by Müller et al. (1997) for urban haze in central Europe (LR=58±12). As mentioned above, the LR depends besides the aerosol composition also strongly on the RH. To illustrate the effect of RH on the LR measured here, the LR of the individual layer versus the layer RH is shown in Fig. 12a. The RH-profiles were taken from a re-analyzed weather model (COSMO). One can observe that for most of the cases the LR increases with increasing RH, similar to the model results of Ackermann (1998) or the measurements of Salemnik et al. (1998). Of course, also the aerosol type might change with altitude which can not be excluded here.

The LR of the lowest possible height level was multiplied with the mean backscatter coefficient measured between (~60-200 m) to retrieve a mean extinction coefficient for the ground (see Eg. 7). In addition, the individual retrieved LR-RH-relationships (see Fig. 12a) were used to calculate (interpolate) the LR for the mean RH measured at the ground. With this method only 15 profiles could be compared since the no extrapolation was performed. The result is shown in Fig. 12b. The error bars denote the range of the retrieved extinction coefficient taking the maximum and minimum value of the measured LR to calculate σ_{ep} at the ground.

Orthogonal linear regressions (without weights) revealed that the LIDAR retrieved σ_{ep} were about ~1.7 -- 1.8 higher compared to the ambient in-situ values. There is no large difference if the LR interpolated to the ground RH (instead of the LR from the lowest layer) is being used, which indicates that the LR of the lowest level has been a good estimate for the LR at the ground (at least for most of the cases). Both sets of σ_{ep} are well correlated to the ambient in-situ values (R^2=0.82-0.96). Nighttime measurements showed to have a better agreement (slope 1.12, R^2=0.96) compared to daytime measurements, which might be due to lower noise in the LIDAR measurements during nighttime However, this improvement has to be treated with care since only 6 profiles were measured during nighttime.

Technical Corrections (in order of appearance in the article)
The number indicates the line number

Section 1

Page 5: 5 sizes more relevant Changed accordingly. 23 extinction coefficient Changed accordingly. 24 , but nearly always.. Changed to “but almost always“ (see reviewer #2)

Page 6: 5 MPI - should give the full name, followed by (MPI) Changed accordingly. 7 ambient aerosol extinction. Changed accordingly.. .15 in situ measurements of .. (should state what measurements) Changed accordingly “… in-situ measurements of the aerosol extinction coefficient …” 16 xtinction coefficient Changed accordingly. Section 2 22 ‘approx.’ should write the word out completely, and also elsewhere in the article Changed within the entire article. 22 33km north east of. . . 23 west south west of Utrecht Changes to north east and south west.

Page 7 ‘Section’ to replace ‘Sect.’ Section 3 “Sect.” is the official abbreviation within the text as wanted from ACP, so we kept it like it is. 18 lowest layer of the atmosphere from ground level up to ? We just changed it to “lowest ground layer.” and did not add the specific height since it was variable and will be explained in the corresponding chapter.

Page 8 2 and 3 Should give the full name of MAAP, followed by (MAAP), and also for SMPS and APS Changed accordingly.

Page 9 5 of inorganic salts Changed accordingly.

Page 11 8 counted by the CPC. Changed accordingly. 24 was set up to. . . Changed accordingly.

Page 14 24 Raman scattered wavelength (? nm) The Raman scattered wavelengths used here is 387 nm, shifted from 355 nm by nitrogen. The Raman shifted wavelength at 607 nm is also available, shifted form 532 nm, but this was not used in this study. For clarification, we have added “(here: 387 nm)”.

Page 15 9 60m? (on page 28, line 6, 70m is stated) . . .the numbers should be consistent with each other Yes, we changed it to 60 m (the lower limit).

Page 16 2 first sections – should give the Section numbers Changed accordingly. 11 was set up. Changed accordingly. .. After Equation (8) , where a is . . .and is. . ... We have added: “where *a* and γ are two independent curve fit parameters (*a* is the intersect at RH=0\% and γ parameterizes the magnitude of the scattering enhancement).” 20 at these high. Changed accordingly. . ..26 ‘ perc.’ should be written out fully Changed within the entire manuscript.

Page 17 2 (at 670nm?) or should it be (at 637nm?) – as per Section 3.1.3 line 24 Yes, it should be 637 nm, which was used (670 nm is the value given by the manufacturer). We have changed it within manuscript. 24 Should give the typical number of humidograms used to compute the average Yes, we have added that info to the selection criteria in the text (see comment above).

Page 18 2 ‘can not’ should be written as ‘cannot’ 11 (Ming and Russell) Changed accordingly.

Page 19 3,4 should state that *f*(RH) is correlated positively, even if somewhat weakly, with V_{APS}/V_{tot}, while *f*(RH) is correlated negatively with V_{APS}/V_{tot}, also somewhat weakly 4 correlation of ? to *f*(RH) Yes, we have changed that sentence to: “The BC volume fraction V_{BC}/V_{tot} (assuming a density of 2.1 gcm^{-3}) shows only a weak negative correlation, while the coarse mode volume fraction V_{APS}/V_{tot} shows only a weak negative correlation with *f*(RH).”

Page 23 10 does not change. . . Changed accordingly.

Page 24 1 , where h. . ... Changed accordingly.

Page 25 15 . . .entire data set, for the time periods given in Table 1. Changed accordingly.

Page 28 Section 4.4.2 17 concurrently determined indirectly from both the in-situ aerosol measurements and the lidar measurements. Changed accordingly.

References

Page 31 10 7113 – 7113? Changed to 7113-7131.

Page 32 27 . . .. -2319-2010, 2010a. . . . (delete ‘2010’) Corrected.

Page 34 8 droplets, Trans. Faraday. . ... Corrected.

Page 35 17 U., and Fiebig, M. . . .. . . 19 Roozendael, M. et al.? ('et al' should be deleted or else replaced by other authors' names) Corrected … "et al." removed.

Page 38 6 WMO/GAW reference should include the Report Number We have added "GAW Report No. 153"

Table 1
k More detail should be given in the text and in the Table caption of 'water soluble and soot particles' We have added in the footnote k of Table 1: "…with a number mixing ratio of 0.46 and 0.54, respectively."

Fig. 1 Line 2 ..Netherlands, over the period from mid June to the beginning of October, 2009. line 5 670 or 637nm? Changed accordingly. It should of course be 637 nm.

Fig. 2 Line 2 . . .mean aerosol scattering enhancement. . .. Changed accordingly.

Fig. 5 Why not use the same multiplication factor (either 10-4 or 10-5) on both axes of Fig. 5 (a)? Changed accordingly.

Fig.8 (b) x-axis label: 477nm or 450nm? It should be 450 nm. For clarification we have added a new footnote in Table 1. "σ_{ep} is retrieved at 450 nm due to specifications of the radiative transfer model and the employed OPAC database (Hess et al. 1998)."

Fig 8(c) 476nm or 477nm? We have added in Table 1 as a footnote: "The retrieved σ_{ep} is for 476 nm, which is the O_4 cross-section- weighted mean wavelength over the fitting window used."

Fig.8 caption , line 3: . . .interpolated in accordance with the appropriate wavelength; Changed accordingly.

Fig. 12 caption line 1: . . .extinction coefficient determined indirectly from lidar vs. inferred extinction coefficient obtained indirectly from in-situ aerosol optical and microphysical measurements (mean . . . Line 3 height level (should specify) We have changed that figure substantially and with this also it's caption (see comment above).

Further changes

- The FLEXTRA trajectories by NILU were in the original version calculated with slightly wrong coordinates for Cabauw (old: 51.3°N, correct: 51.97°N). This has been fixed and (as expected) the overall result has not changed. Acknowledgement to Ann Marie Fjaeraa added, who re-calculated the air mass trajectories.
- The fixed hygroscopic growth factor used in the closure study assuming a constant hygroscopic growth was $g(d_0,RH=90\%)=1.48$ (and not 1.4). This has been corrected in the figure labeling.
- Update of references and corrected minor typos.

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