

Dear editorial board of ACP,

We thank both reviewers for the interesting comments we have received on our article. Major issues which arose and have been addressed concern the backfilling of AERONET level 2.0 data with 1.5 data (reviewer 1) and the treatment of coarse mode aerosol and the validation/comparison procedure (reviewer 2). We have included the backfilled L2.0 AERONET data in our analysis. We have clarified the way the coarse mode is described and extended the justification for the neglect of sea salt and dust in this study. A few plots and a table with statistics have been added to the revised manuscript to improve the comparison with surface observational data. Furthermore, we have made a few small changes to our model in order to improve the consistency with the AERONET retrievals. We have implemented the T-Matrix software described in Dubovik et al. (2006) which includes the AERONET (non-) spherical fraction in our optimization. We have calculated columnar relative humidity (RH) by averaging the water vapor pressure and temperature from instead of relative humidity directly. This did not lead to significant changes in our results. Individual reviewer comments will be addressed separately in the following sections.

## 1 Comments on review report of referee 1.

### 1.1 Major issues.

#### 1.1.1 L2.0 backfilling.

We have added an analysis based on the backfilled L2.0 data to our original analysis and added to our manuscript in section 2.1.2: *“We use the AERONET level 2.0 inversions and applied a backfilling of missing SSA and RI with level 1.5 data. The lack of level 2.0 SSA and RI is predominantly caused by the strict condition of high (>0.4) AOT. L2.0 does not contain retrievals from high solar zenith angles (which is based on only a small range of scattering angles since the range of angles scanned during an AERONET almucantar is twice the solar zenith angle). Therefore, level 1.5 data (Dubovik and King, 2000; cloud screened but not quality assured, Smirnov et al., 2000) are considered when level 2.0 data are not available.”*

Generally, we find that most of the discrepancies (bad fits) between our model and AERONET relate to the L1.5 data. While L2.0 data improves the overall consistency with the surface observations, not all discrepancies are removed, and also several data points are removed from which reasonable results are obtained. We have added in our manuscript (Section 3): *“Most of the discrepancies relate to L1.5 data and not to the backfilled L2.0 data, which may reflect the difference in accuracy.”*

#### 1.1.2 OPAC BC values.

The reported OPAC BC values for density and for refractive index (RI) are 1.0 and 1.75 -0.44i respectively. The very low density and low imaginary refractive index for BC from OPAC are suspect (Bond and Bergstrom, 2006). We will leave them in the table because they are still widely used, but add the following warning as a footnote to Table 3: *“Bond and Bergstrom state that a density of 1.0 has never been observed.”* and to Table 4: *“Bond and*

*Bergstrom argue that the low IRI (0.44) value for BC (from the OPAC database) should be discarded.”*

### 1.1.3 Page 9: Accuracy of AERONET retrievals.

Dubovik et al. (2000) present their errors for low AOT (<0.2) and high AOT (>0.2), see e.g. Table 4 in their article. We rewrote this statement into (section 2.1.2 in our revised manuscript): “*The uncertainties ( $E_y$ ) used in our model can be found in Table 2. They are derived from Dubovik et al. (2000) and pertain to the general AERONET performance. The uncertainties in AERONET SSA and RI are larger for low AOT (<0.2) than for high AOT.*”

### 1.1.4 Page 10: Angstrom exponents.

Indeed this is true, actually AERONET shows a significant coarse mode volume fraction for  $AE > 1$ . We have adapted section 2.2.2 in our revised manuscript:

*“The Ångström exponent generally exceeds 1 ( $1.48 \pm 0.26$  and  $1.51 \pm 0.24$  for level 1.5 and 2.0 respectively), indicating that the fine mode fraction dominates the aerosol optical properties.”*

### 1.1.5 Page 17: correlation of RRI with radiosonde RH is not necessarily linear.

Yes, the relation between RRI and RH is not linear. The linear correlation coefficient is used to indicate that there is a large spread between RRI and RH, and even though an exponential fit might be slightly better, it is likely not statistically significant. Figure 1 (also added to our manuscript as figure 4) shows the relation between the boundary layer average RH and the RRI. Here, also the very low RRI values at very low RH can be seen.

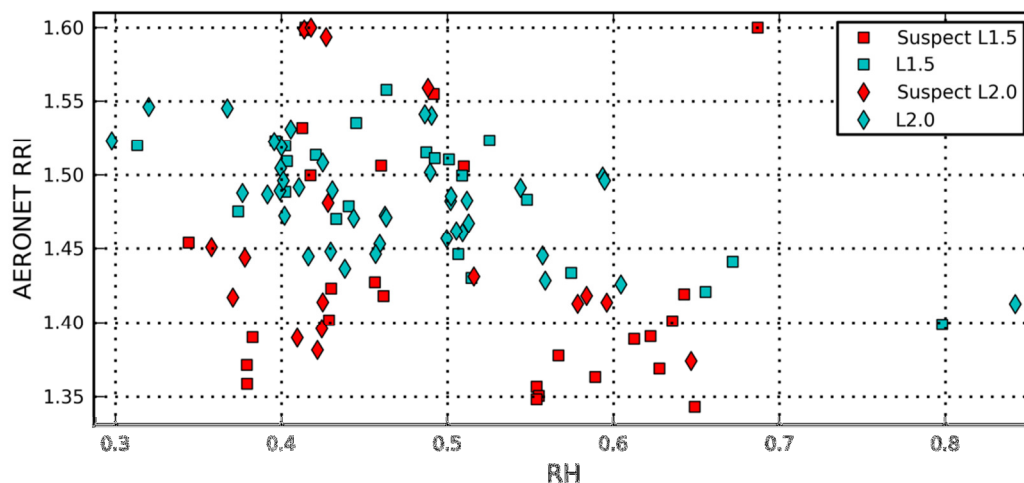


Figure 1. Boundary layer effective relative humidity (RH) versus AERONET real refractive index (RRI).

## 1.2 Minor issues.

### 1.2.1 Does the model ever choose substantial populations of particles larger than 15 $\mu\text{m}$ ?

No, extensive testing with these parameters and comparing the resulting distributions with the AERONET distributions has shown that our model never chooses substantial populations of particles larger than 15  $\mu\text{m}$  during our analysis period.

### 1.2.2 Model uses refractive index for each mode, AERONET only one refractive index for both modes.

For each mode a single RI is calculated separately. The median absolute difference of RRI between modes is very small (0.01) in our study, but it can occasionally be significant.

### 1.2.3 First paragraph of Section 2.2.2 needs to be moved above 2.2.1

We agree, this has been done in the revised manuscript.

### 1.2.4 Page 3: citations.

Citations have been added to the manuscript in section 1: “(e.g MODIS (King et al., 1992); MISR (Diner et al., 1991); POLDER/PARASOL (Deschamps et al., 1994; Tanré et al., 2011))”

### 1.2.5 Page 9: Uncertainty in AERONET volume distribution.

This is from Dubovik et al. (2000), the reference has been added. Dubovik et al. state that the uncertainty in the bins between (and including) 0.1 and 7  $\mu\text{m}$  is 15 %, increasing to 100 % at the distribution edges. We have set the uncertainties of the three outlying bins below 0.1 and above 7  $\mu\text{m}$  to 30 %, 60 % and 100 % respectively. The uncertainties obtained by the sensitivity study of Dubovik et al. (2000) are based on randomly oriented spheroids (see Dubovik et al., 2002). AERONET does not provide error estimates for each separate retrieval. The uncertainty of the bins is related to the optical contribution of each bin, and thus also dependent on the specific distribution. There is very little optical information in the outlying bins, hence the large uncertainty.

We have adapted section 2.1.2 in the manuscript: “*The uncertainties ( $E_y$ ) used in our model can be found in Table 2. They are derived from Dubovik et al. (2000) and pertain to the general AERONET performance.*”; and “*Dubovik et al. (2000) state that the uncertainty in the AERONET volume distribution is 15 % between 0.1 and 7  $\mu\text{m}$ , increasing up to 100 % at the distribution edges. We have set the uncertainties of the three outlying bins below 0.1 and above 7  $\mu\text{m}$  to 30 %, 60 % and 100 % respectively.*”

### 1.2.6 Page 13: “surface” observations.

Indeed, surface observations is what we meant and has been added.

### 1.2.7 Page 17: large variation of AERONET RRI from poor retrievals at low SZA.

Although most of the discrepancies are in the L1.5 data, the use of L2.0 data does not (fully) resolve the issue of these large (chaotic) daily variations in AERONET inversions of real refractive index.

## References

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## 2. Comments on review report of referee 2.

### 2.1 Response to general comments.

Reviewer 2 requests a more thorough validation of our model results, involving scatterplots and statistics of all the model and observational data and has issues with the neglect of sea salt in the aerosol. The following section addresses the general comments of the reviewer.

#### 2.1.1 The comparison with observational data is unsatisfactory.

We added a comparison in our revised manuscript for the (AERONET versus optimized) total aerosol volume (Fig. 5 in the revised manuscript), the monthly average volume size distribution (Fig. 6 in the revised manuscript), and a discussion of the parameters which showed significant or interesting deviations. Two examples of scatterplots of the volume in an individual size bin have been presented here (Fig. 2) for the reviewers. We also added a plot (Fig. 10 in the revised manuscript) of daily averaged aerosol concentrations including uncertainty estimates, and a table with statistics (Table 7 in the revised manuscript). We have refrained from showing all scatterplots because this would be excessive.

We have used the AERONET (almucantar) L1.5 and L2.0 inversion products (“retrievals”), for which uncertainty estimates have been described in Dubovik et al. (2000). These are AOT, SSA and real and imaginary refractive index at 440, 676, 870 and 1020 nm, and the volume distribution in 22 size bins. Spherical fraction is also part of this retrieval, however, no uncertainty estimates are available. We have compared all these parameters (fits) to our model, but did not include all plots and statistics in our article (for AERONET alone this would yield 40 parameters, each for 3 subsets of the data). These retrievals contain the full set of information we need for our optimization. We did not use other AERONET parameters, because they are derived products from the AERONET retrievals mentioned above and are thus redundant.

We have not shown a comparison with other campaign data (e.g. other surface optical measurements than AERONET) because this data has not yet undergone quality assurance and has not yet been published. For the H-TDMA specifically, it is not known what reference RH is used for the 90 % RH growth factors (GF), and we have noticed from our model that the results are very sensitive to this. The GF are only available for aerosol diameters  $D_p \leq 165$  nm, which is smaller than most optically active particles. A comparison of the AERONET size distribution with the surface measurements at Cabauw has not been done because during IMPACT 2008 unfortunately there were problems with the SMPS (data only available between May 14 and 31 because the SMPS 3034 CPC cooler was broken) and with the CPC 3762 (from May 3 onwards).

#### 2.1.2 Comments on coarse mode and neglect of sea salt.

It is a misunderstanding that the model is restricted to coarse mode free aerosols. In the absence of significant amounts of sea salt and mineral dust, a coarse mode can still consist of mixtures of inorganic salts, organic matter and (inclusions of) black carbon, which are

included in our model in this study. We have left sea salt out of our retrieval because the synoptic set up (dominated by high pressure and easterly winds) makes a significant amount of sea salt in the aerosol unlikely during our analysis period. The measured concentration of  $\text{Cl}^-$  at the surface is negligible ( $\ll 1 \mu\text{g m}^{-3}$ ). There are no signs of significant elevated aerosol layers on lidar images (only lidar backscatter images are available from the EUCAARI IOP Cabauw database at <http://www.knmi.nl/eucaari/>), at the times of the AERONET inversions (including those with very low RRI), except at the few AERONET inversions which were influenced by dust events as discussed in our article (e.g. p15203.27-30). Also Morgan et al. (2010) report very low aerosol concentrations above approximately 2 km altitude between May 1-14.

The neglect of sea salt is not the reason why our model cannot reproduce the AERONET RRI at the observed RH. Our model cannot reproduce the low AERONET RRI values, even if we assume that the (dry) aerosol consists purely of sea salt (or any other strongly hygroscopic compound). The variability of RRI during the day seems larger than expected from changes in composition or RH (e.g. May 9, 10 in figure 2 in our manuscript). Besides, very low RRI exists at low columnar RH, e.g. all AERONET RRI  $< 1.4$  occurs when the columnar RH  $< 65\%$  (Fig. 1). Highwood et al. (2012) report a similar range of  $f(\text{RH})$  from airborne nephelometer measurements than calculated by our model. If sea salt aerosol was present in elevated layers, it would have a high RRI (1.55), because RH above the boundary layer was generally very low. Finally, most of the very low RRI retrievals relate to AERONET level 1.5 data, confirming that they are lower quality AERONET retrievals.

We have excluded the second half of the IMPACT campaign (only 15 % of all data in May 2008), because AERONET retrievals are sparse in this period. On the last few days some surface and radiosonde observations are not available. Besides this, the weather was quite unsettled and variable at that time, so that our assumption of a 2 km well mixed boundary layer is certainly not valid.

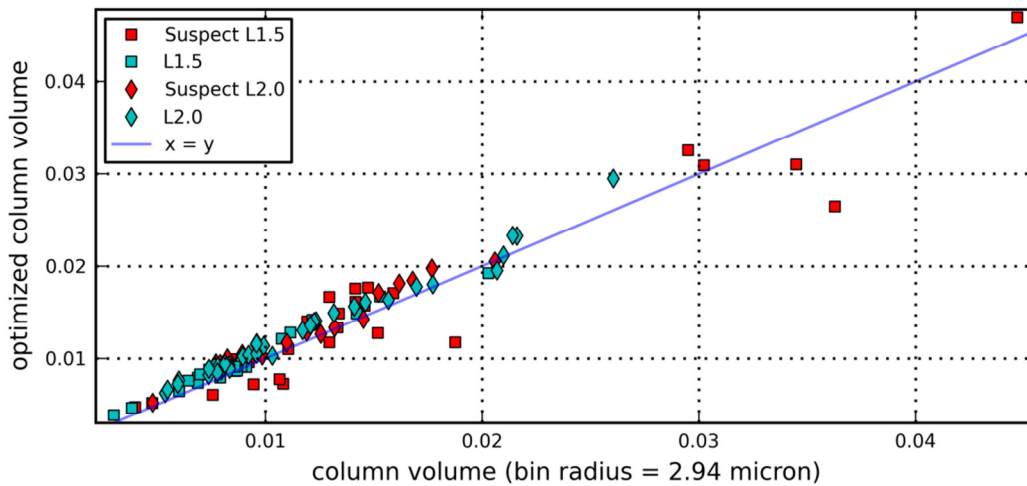
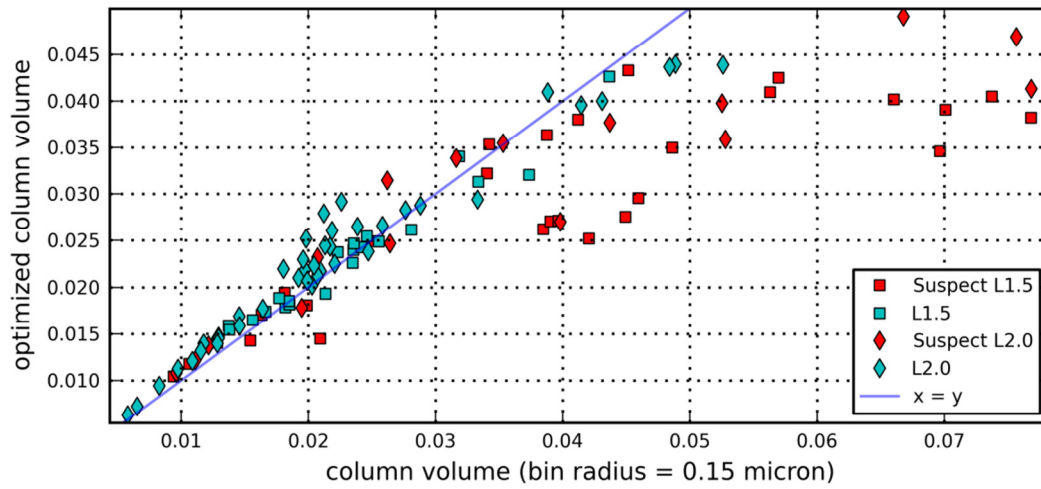


Figure 2. Scatter plots of the AERONET volume distribution (L1.5: squares, L2.0: diamonds) versus modeled (optimized) volume distribution in a single size bin for a) a bin radius of 0.15 micron, and b) a bin radius of 2.94 micron, corresponding approximately to the average median volume radius of the fine and coarse mode respectively.

## 2.2 Detailed response to the individual comments

Below we will address the specific comments of reviewer 2. Each response will refer to the page number and line number in the original manuscript in the following format (page.line: response).

15192.20: We have rephrased this as: “*The retrieved water volume fraction is highly variable and strongly dependent on composition. During this campaign we find that it is >50 % at approximately 80 % RH when the aerosol composition is dominated by hygroscopic inorganic salts, and <0.1 below 40 % RH, especially when the composition is dominated by less hygroscopic compounds such as organic matter.*”

15193.17: Aerosol optical properties such as refractive index and single scattering albedo are available for example from the PARASOL satellite (e.g. Hasekamp et al., 2011). We have added references to our manuscript (see 1.2.4).

15193.18: We have adapted this section in the revised manuscript.

15193.21: Our study can be useful to understand discrepancies between modeled and satellite observations of aerosol (optical) properties, which can be caused by the description of the aerosol processes or RH in the model (e.g. Bian et al., 2009; Zhang et al., 2012).

15193.27: the aerosol humidification factor  $f(\text{RH}) = k(\text{RH})/k(40\%)$ , where  $k$  is the aerosol scattering coefficient. We have adapted the sentence to “*(the ratio of scattering coefficient of aerosol at ambient RH and at dry (40 % RH) conditions)*”, in order to be more specific.

15194.14: Several potential issues are addressed here by the reviewer. We have changed the word “validated” to “compared”. The modeled size distribution is directly fitted to the AERONET distribution. Problems with the SMPS and CPC prevented a comparison with the full size distribution, as described here in section 2.1.1 (and in section 2.2 of our revised manuscript). We have added a scatter plot of the total aerosol volume in the model compared to the AERONET volume (Fig. 5 in our revised manuscript). We have also added some information on the modeled number concentrations (section 3 in our revised manuscript), and compared them to some available flight measurements. The optimized aerosol number concentration is in the same order of magnitude as the observations (Hamburger et al., 2011).

15195.20: Yes, black carbon inclusions may be present in coarse mode aerosols and significantly influence absorption (e.g. Berner et al., 1996).

15196: We have added growth factors to the text where relevant. Growth factors for each individual compound can be directly calculated from the polynomials presented in the studies we have referred to in our article.



15196.2: They are included in the model but have been left out of the retrieval during May 2008. It has been discussed in section 2.1.2 why we left these compounds out. We have adapted the methodology so that it includes a section (2.2.1 in our revised manuscript) containing the assumptions we have made specifically for Cabauw during IMPACT.

15196.12: We have left this argument out.

15197.4: We use AOT at 4 wavelengths, which includes the information present in the angstrom exponent. For the discussion about fine and coarse AOT and other AERONET derived compounds, see section 2.1.1.

15197.6: We changed “for several wavelengths” to “at four wavelengths (440, 675, 870 and 1020 nm)”.

15197.20, Table 3: The model is free to vary the geometric standard deviations of the fine and coarse mode. We have added to Table 1: “*Geometric standard deviation of each mode can be varied by the model.*”.

15198.3: There is not enough AERONET data influenced by dust and sea salt during May 2008 to evaluate the performance of our model. This will be done in a future study. More arguments on why we left out this period are presented in section 2.1.2 above.

15198.19: see 15197.4.

15199.1: This section has been revised (see comment 1.1.3 to reviewer 1). The uncertainties as a function of  $AOT_{440nm}$  can be found in Table 2, which are derived from the sensitivity study of Dubovik et al. (2000); as described in the manuscript.

15199.4: Yes, in Table 2 it says 0.15V (approximation see text). We have added to the revised manuscript (section 2.1.2):  
“*Dubovik et al. (2000) state that the uncertainty in the AERONET volume distribution is 15 % between 0.1 and 7  $\mu m$ , increasing up to 100 % at the distribution edges. We have set the uncertainties of the three outlying bins below 0.1 and above 7  $\mu m$  to 30 %, 60 % and 100 % respectively.*” See also comment 1.2.5 on reviewer 1.

15200.11: Yes, reviewer 1 also comments on this (see 1.1.4). Note that here we specifically focus on the period between May 6 and May 12. On May 2, May 4 and the afternoon of May 12 the angstrom exponent is relatively low (near or below 1). In our manuscript (section 3 of our (revised) manuscript) it is discussed that the AERONET retrievals at May 4 are likely influenced by traces of dust which traversed the AERONET region, and that the retrieval for these points may not be valid. At May 2 (2 L1.5 measurements) and the afternoon of May 12 (1 L1.5 measurement) the retrievals may also have been influenced by traces of dust (likely on May 2), but they turned out to be “suspect” in our retrievals.

15200.21:  $R^2 \sim 0.14$  (0.36 for non-suspect data), this has been added in the manuscript. I also expected a strong relationship between the AERONET RRI and the radiosonde RH. However, there is a very strong correlation between modeled water volume fraction and RH but not between RH and AERONET RRI. This leads to the large variation of composition in our model which we discussed in our article.

15200 Sect. 2.2.2: This has been discussed in section 2.1.1 of this review response.

15201.11: We have added to the revised manuscript (section 2.2.3): “*Measured AMS PM1 mass concentrations for inorganic and organic ambient aerosol species are reproducibly accurate to approximately  $\pm 25$  % (Canagaratna et al., 2007).*” and changed the order which the MAAP BC measurements and the AMS measurements appear in the text.

15201.17: The AMS data is equivalent to  $PM_1$  as described in the article (Canagaratna et al., 2007; Mensah et al., 2012). Even though the inlet might be effectively  $PM_5$  or  $PM_{10}$  we doubt that this will influence the AMS data (being  $PM_1$ ). The MARGA data were collected from a different inlet at 4 m altitude at ambient RH and temperature. We are using quality assured (published) data corrected by the measurement groups, which implies that losses in the instrument have been accounted for.

15202.4: This does not seem to be the case (see section 2.1.2).

15202.13(a): Suspect points do not generally have low angstrom exponent (Fig. 3).

15202.13(b): This has been done, also by request of reviewer 1.

15202:16, Table 5: We have added to the description of Table 5: “*Squared Pearson correlation coefficients*”. For a discussion of why we did not describe the other (derived) AERONET parameters in our article, the reader is referred to section 2.1.1 of this review response.

15202.19: We have added a plot of daily averaged aerosol composition with uncertainty estimates (Fig. 10 in our revised manuscript) and a table with statistics (Table 7 in our revised manuscript). We have made a rough estimate of the uncertainties in our model results and observations. Largest uncertainties stem from the uncertainty in AERONET RRI and the representativeness of column average model data versus surface observations (e.g. inhomogeneous distribution of aerosol in the layer and its depth), see section 3 in our revised manuscript for more details. We decided not to focus on a golden day separately, as the whole period was characterized by similar meteorological conditions.

15202.28: The boundary layer is relatively well mixed, especially between May 6–12 and in the afternoon (e.g. see Fig. 4). No elevated layers can be found on lidar images (available on [www.knmi.nl/eucaari](http://www.knmi.nl/eucaari)) above this layer, except sometimes some slight signal from 10-11 km altitude (dust or cirrus). Even during the early morning hours, most of the aerosols are still in

a layer of approximately 2 km, but on some days some layering can be seen (e.g. May 6, 7). The lidar however is not able to pick up the near surface layer, which can be very shallow, and thus have a relatively small contribution to the columnar optical properties, but a large influence on the surface measurements. The approximation of a 2 km deep well-mixed boundary layer for this study is a crude, but reasonable, assumption.

15203.10: The mixing by itself will not do this, but the partitioning effects of temperature on semi-volatile compounds will, as indeed discussed in the next sentence in our manuscript. In a strict sense, the PBL is not perfectly mixed, but here it is generally a reasonable assumption (in light of the uncertainties in our model and in the AERONET inversions). The surface will be much more influenced by small scale local effects of emissions, aerosol processes, temperature and humidity than the column average.

15203.15: No, this may not be visible in the surface measurements because of the combined effects of mixing (meteorological processes reducing concentrations near the surface) and partitioning of semi-volatiles by temperature, photochemical production and emissions. Our model does not represent specific aerosol processes, but aims to interpret the aerosol optical data which reflects the results of these processes. The increase in modeled (optimized) dry aerosol concentration during the afternoon is caused by a (near) two-fold increase in AERONET AOT (e.g. May 8, 9), which occurs without a significant change in AERONET refractive index.

15205.4: We have adapted this in our revised manuscript (section 3, Fig. 12). The figure shows that inorganic and organic fractions depend strongly on RRI, with decreasing inorganic and water volume fractions and increasing organic fractions for increasing RRI. In general the aerosol composition appears to be predominantly defined by RRI, other factors, specifically RH, may also exert significant influence (e.g. at RRI = 1.41).

15205.13: This is correct, we have added “*single scattering albedo*,” here in the revised manuscript.

15205.24: We agree that “validated” should not have been used here. We have changed this in our manuscript to “compared to”. We have commented on the completeness of the comparison in previous sections, e.g. in section 2.1.1 and at 15194.14 of this response.

15206.26: Yes, we agree. We have changed “*relatively hydrophobic*” to “*weakly hygroscopic*” here in our revised manuscript.

15207.16: This has been done.

15207, 3<sup>rd</sup> point: Yes, we agree. Reviewer 1 also suggested this, so the data has been added to the tables and graphs and the results are discussed in the text.

Precise conclusion.

We have added a short and precise conclusion to section 4 of our revised manuscript. *“The accuracy of the optimized aerosol chemical composition strongly depends on the accuracy of AERONET inversions of RI, the description of aerosol RI as a complex mixture of components and the optical model of scattering and absorption. Another uncertainty is associated with the depth and mixing of the boundary layer. As a first estimate, based on the model results and sensitivity tests, uncertainties in computed volume fractions are approximately 50 % for inorganic salts, black carbon and water, and 100 % for organic matter. We expect that the uncertainty caused by the discrepancy between column average and surface concentrations is approximately 25 % for this study specifically. Combined, this corresponds to uncertainties of approximately 50 %, 100 %, 50 %, 30 % and 65 %, for inorganic salts, OC, BC, dry mass and water in the total column aerosol mass.”*

Table 1: They are not defined in terms of the particle diameter, except by the fits to the AERONET volume distribution. The fit with the largest effective radius or volume median radius is the coarse mode.

Table 5: We have separated the data into L1.5, backfilled L2.0 and non-suspect data, and presented the results separately. We have selected a relevant subset of the available parameters and discussed the discrepancies between modeled and AERONET data.

Fig. 1: No, the columnar temperature has not been included in the optimization.

Fig. 2: We have added to the manuscript in section 2.2.1: *“We assume that the aerosol is homogeneously distributed in a layer of 2 km depth at the surface. The “effective” RH in this layer is derived from radiosonde measurements by means of averaging water vapor pressure and temperature in this layer.”*

Fig. 3: Yes, we have done this and many other tests, but we did not find any other explanation on why the model fails at these points. We have also tried many other RRI values for each compound within the range found in the literature (see discussion in section 4 in the revised manuscript), but these RRI values are less likely and do not resolve the issue of simulating very low RRI at relatively low RH.

Fig. 4 – Fig. 6: We have refrained from putting these figures together for reasons of clarity, because each figure already shows a lot of data. We have included daily average aerosol composition, including a table with statistics, see 15202.19.

Fig. 7: A plot of the growth factors versus RH is presented here (Fig. 5). The range of numbers we find (when leaving out the “suspect” data) corresponds roughly to the flight nephelometer measurements presented in Highwood et al. (2012) (e.g. between 1.01 and 1.08 when RH is 40 % and 1.3-1.4 when RH 80 %). We are not aware however of any H-TDMA curves measured at Cabauw.

Title: We have changed the title to: “*Estimation of aerosol water and chemical composition from AERONET sun photometer measurements at Cabauw, the Netherlands*”

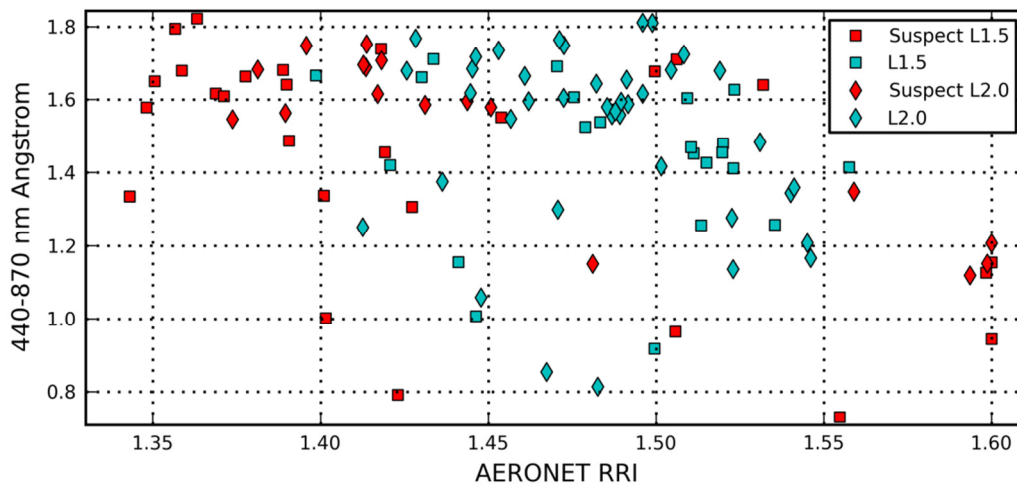


Figure 3. The AERONET RRI versus the (440–870 nm AERONET) Angstrom exponent, with the “suspect” data in red.

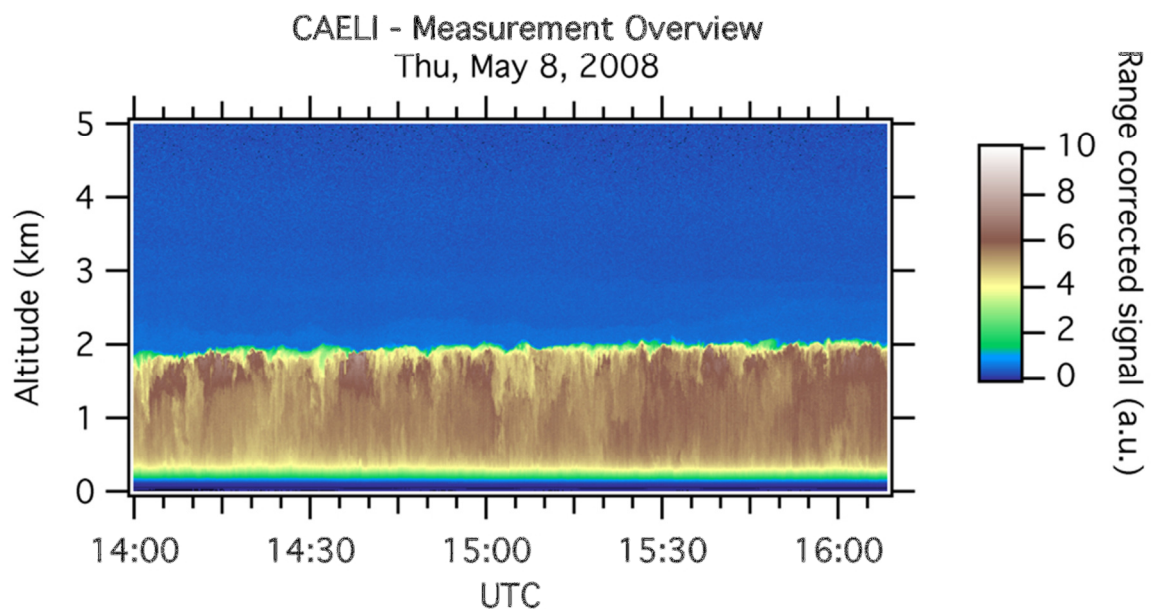


Figure 4. An example of the “well mixed” boundary layer at Cabauw, during the relatively quiet period between May 6 and 12. Sample image taken from [www.knmi.nl/eucaari/](http://www.knmi.nl/eucaari/), original data is not (yet) available.

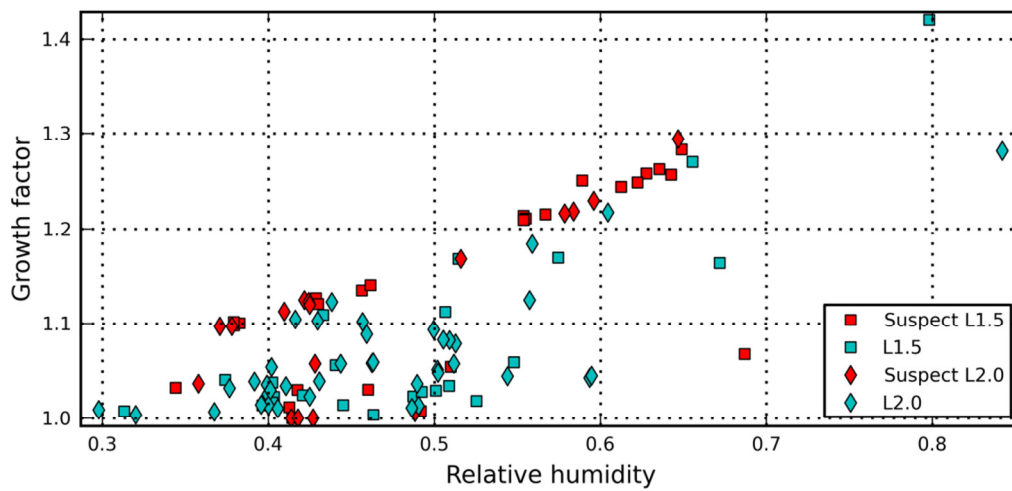


Figure 5. The columnar relative humidity versus the aerosol growth factor. Results based on AERONET L1.5 (squares) and L2.0 (diamonds) have been presented red when “suspect”.

## References

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