**Atmospheric
Chemistry
and Physics
Discussions**

Interactive
Comment

Interactive comment on "Comparison of ambient aerosol extinction coefficients obtained from in-situ, MAX-DOAS and LIDAR measurements at Cabauw" *by* P. Zieger et al.
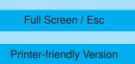
Anonymous Referee #3

Received and published: 18 January 2011

acp_2010-838_Referee_Comments

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 pa-

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

per. 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 750m 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 hygro-scopic). 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. (NH4)2 SO4 was chosen for Arctic aerosol (Zieger et al, 2010), but is probably not appropriate for the Cabauw site. 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 tra-jectories, 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 $\sim$ 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. 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. 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. 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 αap. 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 αap, determined from a different instrument – an Aethalometer. However, there is no certainty that the 'instrumental' absorption Ångström exponent αap, obtained from the Aethalometer measurements at a range of wavelengths is nec-

essarily 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 αap of 0.84 based on an initial relatively short period ($\sim$ 1/6 of the total measurement period) of the campaign, from $\sim$ 19th June to July 6th, and assumed to be valid thereafter throughout the remaining period up to $\sim$ 4th October, is certainly questionable, and needs both justification and discussion. 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.

(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 coeffienct 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 NO2 and O4 (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 (Frieβ 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. There are also some issues with the com-

parison 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 $\sim$ 750m)? If so, this presumes that the layer between $\sim$ 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 $\sim$ 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 mis-reading the points? Similarly, there are 4 vertical circular gray (daytime points) with largest value$\sim$ 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 σe (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 σe values between 0 and 1x 10-4 m-1.

Technical Corrections (in order of appearance in the article)

The number indicates the line number

Section 1 Page 5: 5 sizes more relevant 23 extinction coefficient 24 , but nearly always.. Page 6: 5 MPI - should give the full name, followed by (MPI) 7 ambient aerosol extinction. . .15 in situ measurements of .. (should state what measurements) 16 extinction coefficient Section 2 22 'approx.' should write the word out completely, and also elsewhere in the article 22 33km north east of. . . 23 west south west of Utrecht Page 7 'Section' to replace 'Sect.' Section 3 18 lowest layer of the atmosphere from ground level up to ? Page 8 2 and 3 Should give the full name of MAAP, followed by (MAAP), and also for SMPS and APS Page 9 5 of inorganic salts Page 11 8 counted by the CPC. 24 was set up to. . . Page 14 24 Raman scattered wavelength (? nm) Page 15 9 60m? (on page 28, line 6, 70m is stated) . . .the numbers should be consistent with each other Page 16 2 first sections – should give the Section numbers 11 was set up. . .. After Equation (8) , where a is . . .and γ is. . . .. 20 at these high. . . .26 ' perc.' should be written out fully Page 17 2 (at 670nm?) or should it be (at 637nm?) – as per Section 3.1.3 line 24 24 Should give the typical number of humidograms used to compute the average Page 18 2 'can not' should be written as 'cannot' 11 (Ming and Russell) Page 19 3,4 should state that f(RH) is correlated positively, even if somewhat weakly, with VAPS/Vtot, while f(RH) is correlated negatively with VAPS/Vtot, also somewhat weakly 4 correlation of ? to f(RH) Page 23 10 does not change. . . Page 24 1 , where h. . . .. Page 25 15 . . .entire data set, for the time periods given in Table 1. Page 28 Section 4.4.2 17 concurrently determined indirectly from both the in-situ aerosol measurements and the lidar measurements.

References Page 31 10 7113 – 7113? Page 32 27 . . .. -2319-2010, 2010a. . . . (delete '2010') Page 34 8 droplets, Trans. Faraday. . . ... Page 35 17 U., and Fiebig, M. . . . ... 19 Roozendael, M. et al.? ('et al' should be deleted or else replaced by other authors' names) Page 38 6 WMO/GAW reference should include the Report Number

Table 1

k More detail should be given in the text and in the Table caption of 'water soluble and soot particles'

Fig. 1 Line 2 ..Netherlands, over the period from mid June to the beginning of October, 2009. line 5 670 or 637nm? Fig. 2 Line 2 . . .mean aerosol scattering enhancement. . .. Fig. 5 Why not use the same multiplication factor (either 10-4 or 10-5) on both axes of Fig. 5 (a)? Fig.8 (b) x-axis label: 477nm or 450nm? Fig 8(c) 476nm or 477nm? Fig. 8 caption , line 3: . . .interpolated in accordance with the appropriate wavelength; 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)

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 29683, 2010.