Aerosol source apportionment from 1-year-measurements at the CESAR tower at Cabauw, NL

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Responses to anonymous referee #2

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We thank the reviewer for the careful review of our manuscript; the comments and suggestions are greatly appreciated. All the comments have been addressed. In the following please find our responses to the comments one by one and the corresponding changes made to the manuscript. The original comments are shown in italics.

General Comments:

1) Order of sections: Please exchange 3.1. and 3.2 (data quality assurance should be before the results). You might even consider moving section 3.2 to chapter 2. Also in the PMF results section, the applied contraints should be described first, and then the results should be discussed. See also my comments below. With respect to the discussion on organic nitrate, I am very sceptical that the data support the conclusions. The uncertanties of both instruments are too large for an estimation of organic nitrate from the difference between ACSM and MARGA.

Response:

The authors agree with the reviewer that the cross validation chapter should be placed before the presentation of the results themselves. In the revised manuscript chapter 3.1 and 3.2 were exchanged.

We also accept the suggestion from the reviewer to move the description for the applied a values to the beginning of chapter 3.3 in the revised manuscript. Referring to comments #12 and #14 from the reviewer, we added a sentence describing that the choice for the a-values resulted from a series of sensitivity tests:

“Figure 4 summarizes the time series of PMF factor classes (two POA factors: HOA and BBOA; and two SOA factors: OOA and a humic-like substances (HULIS) related factor, see below for descriptions) found for all seasons, except for BBOA in Summer 2012. A corresponding graph dividing these PMF results into the five periods according to Fig. 2 is shown in Fig. S13. The POA profiles were constrained within ME-2 using the HOA and BBOA mass spectra found by ME-2 operated PMF analyses by Crippa et al. (2014) at the CESAR tower in Cabauw in March 2009. For the HOA profile, a constant a-value of 0.1 was found to be most suitable for every season. If observed, the BBOA a-value was set to either

0.2 (Autumn 2012) or 0.3 (Winter and Spring 2013). The a-values based on different sensitivity tests for each season as described by Canonaco et al. (2013).”

Organic nitrate related aspects are answered in the specific comments below.

<u>Specific Comments:</u>

2) Abstract: Please mention the height above ground of the ACSM. Until the end of section 2.2 the reader thinks that the ACSM was located on top of the tower.

Response:

The first sentence of the abstract was changed in the revised manuscript to:

“Intensive measurements of submicron aerosol particles and their chemical composition were performed with an Aerosol Chemical Speciation Monitor (ACSM) at the Cabauw Experimental Site for Atmospheric Research (CESAR) in Cabauw, NL, sampling at 5 m height above ground.”

Section 2.2: Instrument description (second paragraph of this section) should be placed before the statements on the agreement between different ACSMs and the ACTRIS comparisons.

Response:

In the revised manuscript we accepted the suggestion of the reviewer and placed the instrument description before the statements on the agreement between different ACSMs and the ACTRIS comparisons.

3) Page 35124, line 5-8: Detection limits? It is not clear to me why they were not determined? As far as I understand, a blank filter measurement is done on a routine time schedule by an ACSM, and then the detection limit can be calculated as three times the average noise level.

Response:

As responded to a similar comment from referee #1, there were no measurements done with this ACSM during the presented campaign, where a separate particle filter was introduced in the sample line, in addition to the filter including in the gas-phase background filter cycle as described by Ng et al. (2011). Furthermore, the ACSM Local software version used in this study could not show data acquired during the filter cycle measurements (e.g. closed mass spectra/time series), which would be needed for the determination of the detection limits. The software could only show the differential mass spectra/time series.

4) Page 35124, line 12-15: Particle losses: I recommend using the PLC by von der Weiden et al (AMT, 2009) to estimate the losses in such long a sampling line (all vertical or horizontal?) www.atmos-meas-tech.net/2/479/2009/ http://www.mpchmainz. mpg.de/drewnick/PLC/

Response:

As responded to a similar comment from referee #1, we used now the particle density deriving from the chemical composition not as the campaign average but time resolved for each data point in the revised manuscript. In addition we introduced SMPS data which is now size dependently corrected as published by Henzing (2011). The description of the sampling losses of the 60 m inlet for eBC given in the manuscript derived from a series of measurements at the Cabauw tower performed in a previous campaign. Unfortunately these results are not published yet. To clarify how these losses were determined in the revised manuscript and to account for a comment from referee #2, the loss description was changed as follows:

“SMPS data was corrected size dependently for (diffusional) losses in the inlet system and SMPS system itself according to (Henzing, 2011) who compared theoretical findings with measured losses that are obtained by measuring simultaneously before and after the various parts of the inlet system at the CESAR tower. In addition, particles of different compositions were measured in 2013 simultaneously at the pipe entrance at 60 m height and in the basement (J. S. Henzing, personal communication). For more than 8000 simultaneous observations, the results showed that aerosol measurements through this 60 m sampling line underestimate PM_{10}-eBC by approximately 33% with an uncertainty of 7%. Therefore, eBC obtained from the MAAP are divided by a factor of 0.66 to account for these losses. For the inorganic species penetrations through this inlet line were reported to be 62-73% for nitrate, 55-64% for sulfate, and 54-56% for ammonium. However these results were not used for corrections in this work”

Please note that these losses influenced only data acquired by the MAAP and SMPS. Since the contribution of eBC is rather low (average: 5%) a potential overall error for total aerosol masses is low and would not significantly alter one quintessence of the paper, namely total mass concentrations above the air quality limits. We think theoretical particle loss calculations would not add additional information in the context of this paper.

As a consequence of the newly evaluated SMPS data the correlation values between ACSM+MAAP data with SMPS data changed as seen in Fig. S3 and S4 in the revised supplement. Nevertheless, the overall qualitative and quantitative agreement is still given except that the ACSM+MAAP data is now overestimating the total PM_1 mass by 16%, excluding the eBC data the ACSM overestimates total mass by 12%. As seen in Fig. S3 the difference between both systems is significantly higher during the pollution events 16 to 27 January 2013 and 5 to 8 May 2013. Since the quantitative agreement with the MARGA is much higher at these times the discrepancy to the SMPS is likely due to the fact that the losses within the 60 m inlet could not be corrected for individual species as mentioned above. Therefore the following paragraph was added at the end of the cross validation chapter in the revised manuscript:

"Major discrepancies to the SMPS especially during some of the pollution events like 16 to 27 January 2013 and 5 to 8 May 2013 (see below) can be explained by the correction of losses through the 60 m inlet line which was done size dependently and did not account for losses of individual species as mentioned in chapter 2.3. As the quantitative agreements of individual inorganic species as well as of total inorganics between the ACSM and the MARGA during these periods are much higher, the mass loadings determined from these instruments are more reliable than the SMPS data."

5) Page 35124, lines 24 ff: MAAP: Please mention that a MAAP reports equivalent black carbon (eqBC), see Petzold et al., Atmos. Chem. Phys., 13, 8365-8379, doi:10.5194/acp-13-8365-2013, 2013

Response:

We thank the reviewer for the supportive remark and pointing out the reference. In the revised manuscript, the MAAP description was written in more detail as follows:

"The MAAP instrument has been introduced by Petzold and Schönlinner (2004) and Petzold et al. (2005). It is designed for the determination of the black carbon (BC), which is a product of incomplete combustion. There is in the scientific community a general consensus over what black carbon is in terms of properties (Bond et al., 2013) The MAAP measures the strong visible light absorption property of BC by simultaneous measurements of the radiation penetrating through and scattered back from a particle-loaded fiber filter. According to Petzold et al. (2013), optical BC determined by MAAP is to be referred to as equivalent black carbon (eBC). One property of BC is that it is highly refractory with a vaporization temperature near 4000K (Schwarz et al., 2006), thus BC is not vaporized at 600°C and cannot be measured by the ACSM. The MAAP achieves a time resolution of 5 minutes with an uncertainty of 12% (Petzold and Schönlinner, 2004)."

In addition, we used the term equivalent black carbon or its abbreviation (eBC) in the revised manuscript and supplement including tables and figures, whenever its measured values are given, since eBC represents the actual quantitative the MAAP is acquiring.

6) Page 35125, lines 23-29: For the 60 m inlet the sampling losses were calculated. So why not for the ACSM inlet?

Response:

As pointed out in the response to the reviewer's comment #4, the losses in the 60 m sampling line were measured on site in a previous campaign. These measurements could reasonably not be done for the ACSM inlet within the scope of the presented study.

7) Page 35127, line 15: 38Cl should read H37Cl, I assume? There is no 38Cl isotope

Response:

Indeed, the interference on m/z 38 is due to the $H^{37}Cl$ ion. The respective sentence was changed in the revised manuscript to:

"This was done because the signal at these masses showed high interferences with the chloride related ions ^{37}Cl and $H^{37}Cl$."

8) Page 35128, line 9-11: "Even when only considering the ACSM + MAAP PM1 concentration, where a campaign average of 9.5 $\mu g\ m^{-3}$ was determined, the WHO PM2.5 limit was approximated." I suggest rephrasing as: Even the PM1 concentration inferred from ACSM + MAAP data (campaign average 9.5 $\mu g\ m^{-3}$) approached the WHO PM2.5 limit.

Response:

We accepted the suggestion of the reviewer and changed this part accordingly in the revised manuscript.

9) Page 35131 line 29 – page 35132, line 8: You calculate organic nitrate from the difference of ACSM and MARGA. I think that bears quite some uncertainty considering the large uncertainties of the instruments. At least you should provide an error estimation. What about looking at ion ratios, e.g., 30/46? This is known to differ between inorganic and organic nitrates? Can you plot 30/46 vs. the difference between MARGA-nitrate and ACSM-nitrate? Later on, on page 35133, lines 6-7, you state that the uncertainties are 30% for ACSM-nitrate and 10% for MARGA-nitrate. Thus, how can you infer 9% organic NO3?

Response:

As responded to the similar comment from referee #1, there are strong evidences for the existence of particulate organic nitrates during this campaign despite the uncertainties of both instruments. We agree with the reviewer that these explanations are contradictory to those given in the cross-validation chapter on the comparison of ACSM and MARGA total nitrates. The latter part (page 35133, lines 4-12) was changed in the revised manuscript to:

"The quantitative difference to the AMS-organics is also very low, and the discrepancies in case of ammonium and nitrate are within the stated ± 30% accuracy of the AMS and ACSM (Ng et al., 2011) and the ± 10% for the MARGA–NO_3, respectively (Makkonen et al., 2012). Similar variations were also found by Crenn et al. (2015) as well as Budisulistiorini et al. (2014). The latter reported of a comparison between two collocated ACSM's (± 27%, R^2 = 0.21 for Chl, R^2 > 0.8 for the other species) and between these ACSM's and a continuous Tapered Element Oscillating Microbalance (TEOM, $PM_{2.5}$) instrument. Note that the major ions used for nitrate detection in AMS and ACSM (NO^+ and NO_2^+) are produced from both inorganic and organic nitrate (e.g. Farmer et al. (2010)). The higher ACSM nitrate compared to MARGA nitrate can therefore also be explained by the presence of organic nitrates. In case of nitrate the ASCM and MARGA comparison cannot be seen as independent"

As the reviewer pointed out correctly, the ratio of m/z46 (NO_2^+) to m/z30 (NO^+) can be used as a parameter to determine to organic and inorganic nitrate fractions. To do that, the 46/30 ratios of purely inorganic and pure organic nitrates need to be known. The value of purely inorganic nitrate for individual instruments can be taken from ammonium nitrate measurements during IE calibrations. For AMS instruments this ratio was commonly seen to be around 0.4 (Fry et al., 2011; Fry et al., 2009; Mensah, 2011). The corresponding ratio of purely organic nitrates (R_{OrgNO3}) was found to be approximately 0.1 in chamber experiments, measuring SOA from the reactions of BVOC's with NO_3 radicals with an HR-ToF-AMS (Fry et al., 2011; Fry et al., 2009; Mensah, 2011). Analogous experiments were not yet published for ACSM instruments. During the presented campaign in Cabauw, a 46/30 ratio of around 0.2 was observed during the response factor calibrations of the ACSM. Crenn et al. (2015) reported a similar value (0.23) for this particular ACSM and that this ratio is strongly varying between the investigated ACSM's (0.20-0.58). That implies that it is highly uncertain that the 46/30 ratio of 0.1 for organic nitrates should be used for ACSM data, at least for the campaign presented here. Furthermore, as stated in the response to a comment from referee #1, the fragmentation table of the ACSM Local software used here could not be user defined. Thus possible interferences on m/z30 and m/z46 between nitrate and organic fragments could not be corrected. Those interferences would in turn increase the uncertainty of the NO_2^+/NO^+ ratio determination from unit mass resolution data.

10) Page 35132, line 10-12: What value was assumed for the density of organics?

Response:

As mentioned on page 35125, line 12, a density of 1.4 g cm^{-3} for organics was assumed.

11) Page 35133, line 12 ff: There seems to be a problem with sulfate here. The ACSM appears to underestimate SO4 by a factor of two compared to the HR-ToF-AMS. In the paper by Crenn et al (Atmos. Meas. Tech., 8, 5063–5087, 2015, www.atmos-meastech.net/8/5063/2015/ doi:10.5194/amt-8-5063-2015) it looked much better. How were the RIEs for the ACSM and for the HR-ToF-ASM determined? How often for the ACSM? Did all sulfate calibrations suffer from high background signals?

Respond:

The RIE of sulfate was calculated using a common procedure for AMS and ACSM instruments (e.g. Budisulistiorini et al. (2014) and Elser et al. (2015)). Monodisperse ammonium sulfate particles were measured right after the measurement of monodisperse ammonium nitrate. Using the RIE of ammonium deriving from the ammonium nitrate data, the RIE of sulfate was determined by an ion balance of data derived by the ammonium sulfate measurements. For the ACSM, this was done every 1-2 months (AMS: once per week) with relatively stable results (RIE_{SO4} = 0.81 ± 0.10). Close after the sampling of ammonium sulfate (up to 120 µg m^{-3}), a largely increased background of sulfate was observed during all calibrations, leading to the conclusions mentioned in the manuscript.

As mentioned in Crenn et al. (2015), the accuracy of AMS- and ACSM-sulfate measurements is still under debate. In their work, RIE calibrations were done for all instruments, including the particular ACSM used here. For the inter-comparison of the ACSM data sets with the collocated HR-ToF-AMS, Crenn et al. (2015) took the default RIE_{SO4} of 1.2 for all ACSM's, although the calibration results were partly much lower with strongly varying values of RIE_{SO4} between 0.48 and 0.97, while for the AMS the experimental RIE_{SO4} was applied (Fröhlich et al., 2015). Unfortunately, Crenn et al. (2015) did not publish comparison results with the AMS for individual ACSM's. Overall we think that in the case of sulfate it is difficult to generalize from Crenn et al. (2015) on AMS-ACSM comparisons for the presented work.

The reader should also take into account that the overlap period of the AMS and ACSM comprises only one week (294 common data points) in the beginning of the ACSM campaign during a time with low mass concentrations, while the comparison with the MARGA PM_1 data could be done over the whole ACSM data set (1943 common data points), leading to more representative correlation results. There, a better quantitative and qualitative agreement in case of sulfate was found comparing to the ACSM-AMS cross-validation.

12) Section 3.3 Factor analysis:

You prefer the 4-piece separate analysis over the one-year analysis, but I don't see an objective reason for this. To my opinion this discrepancy reflects the uncertainty of the factor analysis. I appreciate that you openly show and discuss the differences, but the conclusion that the 1-year analysis "overestimates" something is not backed up by any data, or am I wrong? If you argue that BBOA is overestimated because of the high degree of freedom (a = 0.3), why didn't you restrict it more (a = 0.1) as the HOA? You should state the reasons for choosing a certain a value at the beginning for the section. If the 1-year analysis would be run with a=0.1, would it be better? In general I would describe first the constraints, then the factors, then the alternative 1-year solution, and then conclude that latter cannot be correct (no m/z 60 in summer).

Response:

As described in the response to comment #1 of the reviewer, we added an explanation that the a-values were determined by a standardized procedure described in detail by Canonaco et al. (2013) and Crippa et al. (2014), in addition to the description of the source apportionment as applied in this work in chapter 2.4 (specifically page 35128, lines 1-13). In contrast to unconstrained PMF commonly published for AMS data, a higher amount of exploration runs including sensitivity tests are necessary to choose the most suitable a-values for constrained factors and overall the most suitable solution for a data set within a ME-2 analysis. It is not feasible to report all these runs with individual explanations on choices of solutions.

We agree with the reviewer that the conclusion of a possible overestimation of BBOA in summer due to the high degree of freedom cannot be sufficiently proven. Further PMF analyses with a constrained BBOA and a-values lower than 3 gave the same results with only very minor differences regarding profiles and time series. As suggested by the reviewer, the description of the 1-year PMF was moved to the end chapter 3.3 in the revised manuscript and changed as follows:

"The source apportionment as described here used a data set which was subdivided into the four seasons prior to PMF analysis. A single PMF analysis of the whole data set with constrained HOA and BBOA profiles lead to solutions with a highly overestimated BBOA factor in the summer, compared to the results when the seasons were explored individually (see Fig. S17 and S18 in the supplement). Furthermore, the contributions of individual factors change significantly in some periods, especially for the OOA factor during pollution events. This is mainly driven by the different OOA-*f44* and -*f43* values. This behavior is independent from applied a-values for BBOA and may result from the uncertainty of this statistical tool. Since there was no evidence of BBOA seen in the separate analysis of the summer period (e.g. low fraction of m/z 60 in the organic mass spectrum and no correlation of the constrained BBOA factor with POA tracers, no matter which a-value was used), the solution derived from the single PMF analysis was reasonably rejected. "

13) Page 35136 line 16: Typo: it's -> its

Response:

This part was corrected in the revised manuscript.

14) Page 35137 line 10: Explain humic-like substances (HULIS) on first occurrence

Response:

To clarify the HULIS term, the introductory sentence on page 35134, line 18, was changed in the revised manuscript to:

"Figure 4 summarizes the time series of PMF factor classes (two POA factors: HOA and BBOA; and two SOA factors: OOA and a humic-like substances (HULIS) related factor, see below for descriptions) found for all seasons, except for BBOA in Summer 2012."

15) Conclusions: This section is too short and too weak and should be expanded.

Response:

To meet this suggestion by the reviewer we restructured the conclusions section and moved some concluding statements from previous sections to this part. The conclusions now emphasize the important finding that NH_3 emission control can significantly decrease PM loading at this rural site. Furthermore the high SOA fraction in the aerosol challenges mitigation strategies in particular when large background fractions are observed as is the case with HULIS here. To account for comments from other reviewers as well the revised section now reads:

This work provides chemical composition data of atmospheric aerosols acquired during one year at the CESAR tower in Cabauw, the Netherlands, which is a representative rural site for North Western Europe. The concentration of submicron particles from combined ACSM and MAAP data showed 12 exceedances from the WHO $PM_{2.5}$ daily mean limit. The respective campaign average of 9.5 $\mu g\, m^{-3}$ approached the WHO $PM_{2.5}$ annual mean limit. Taking MARGA $PM_{2.5}$ into account the number and proportions of these exceedances are even higher, emphasizing the importance of these high ACSM PM_1 results shown here which represent lower limits. As carbonaceous compounds are estimated to be five times more toxic than inorganic particles (Lelieveld et al., 2015) MARGA data alone would not give sufficiently possible implications regarding adverse health effects. While few people live in the direct vicinity of the measurement site, the high aerosol concentration measured at the site can be considered to represent the regional background. This regional background is adding to local aerosol contributions in high populated urban sites (Pandis et al., 2013), namely the 4 largest cities of the Netherlands which have a distance of 40 km or less from the CESAR tower.

Particulate mass loadings found at this rural site are dominated by secondary aerosol formation through atmospheric gas phase chemistry and particle phase aging. It is shown that particulate ammonium nitrate is the major aerosol component (39% on average) and represents the more hygroscopic aerosol fraction. Since the human respiratory system is characterized by high humidity more hygroscopic aerosols have a higher deposition tendency within the human lung than less water soluble particle compounds (Asgharian, 2004; Broday and Georgopoulos, 2001). With regard to adverse health effects this is very crucial because Asgharian (2004) also found that especially hygroscopic submicron particles can deposit in the entire lung. The high ammonium nitrate fraction also implies that inorganic SA reduction in Cabauw can be most efficiently achieved through the reduction of gaseous ammonia emissions in the area.

As indicated by model results for the South Western United States from Zhang and Wu (2013), the reduction of NH_3 emissions, in conjunction with already implemented SO_2 and NO_x emission reductions, can further reduce $PM_{2.5}$ than reducing SO_2 and NO_x emissions alone, particularly for regions with high emissions of NH_3 from agricultural sources. The latter is clearly the case for Cabauw with its high number of animal husbandry and the use of nitrogen containing fertilizers around the CESAR tower. Banzhaf et al. (2013) derived similar conclusions for PM_{10} using different emission scenarios within domains covering Germany and Europe. Applying a 3d chemical transport model over Europe Megaritis et al. (2014) found that a reduction of NH_3 emissions by 50% would have a much higher effect on reducing $PM_{2.5}$ than decreasing NO_x emissions by 50%. The latter scenario would even result in negative side effects such as higher tropospheric ozone concentrations (especially in summertime 4% over Western Europe and up to 40% in major urban areas) and higher amounts of particulate sulfate and OA by 8% and 12%, respectively, in winter.

The local mitigation of organic aerosol mass (29% contribution on average) is more challenging, as secondary organic aerosols are highly abundant at the Cabauw site (74% and 22% of OA and total PM_1 on average, respectively). The presented data set shows a large and ubiquitous HULIS fraction (37%) which based on diurnal patterns and a lack of correlation with wind direction can be considered as long-range background aerosol formed from atmospheric aging processes. In turn, primary organic aerosols emitted mainly from traffic and biomass burning (12% and 13% of OA on average) have only minor importance. For a more detailed identification of the SOA sources compound specific measurements of OA as well as routine VOC monitoring are needed.

Finally, the presented data set and interpretations provide an important contribution to the EU-FP7-ACTRIS project which supported building of new knowledge as well as policy issues on climate change, air quality, and long-range transport of pollutants on a European scale."

Figures:

16) Figure S2: Wind speed is hard to see. Please make extra graph.

Response:

As suggested by the reviewer, the wind speed is now shown in a separate graph in Fig. S2 in the revised supplement material

17) Figure S11: Mention in figure caption that MARGA data are PM1.

Response:

In the revised supplement material the figure caption was changed to:

"Figure S11: Correlation graphs of Chl, NH_4, SO_4, NO_3, and total inorganic mass concentrations from ACSM and MARGA PM_1 data"

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