Review of *Schlag et al.*

Schlag et al. report aerosol measurements taken at Cabauw, Netherlands that span a period of 1 year from July 2012 to June 2013. The PM_1 aerosol composition at the site was measured using an aerosol chemical speciation monitor (ACSM) as well as a multi-angle absorption photometer (MAAP). In addition, a monitor for aerosol and gases (MARGA) was used to characterize $PM_{2.5}$. A number of gas phase measurements (e.g. CO, CO_2, NO_X) are used in the analysis of the aerosol measurements as well.

It is found that during the measurement period there were 12 exceedances of the WHO's daily mean limit for $PM_{2.5}$ (25 µg m^{-3}). In addition, the average campaign PM_1 concentration of 9.50 µg m^{-3} was near the WHO annual mean limit of 10 µg m^{-3}. A large majority of the PM_1 mass is due to secondary formation of aerosols including NH_4NO_3 and $(NH_4)_2SO_4$ as well as secondary organic aerosol (SOA). During two pollution events, the secondary fraction of PM_1 reached 90%. Apportionment of OA mass is also carried out using PMF analysis driven by multilinear engine 2 (ME-2) and source finder (SoFi). Four components of OA are identified: hydrocarbon organic aerosol (HOA), biomass burning organic aerosol (BBOA), oxygenated organic aerosol (OOA), and humic-like substances (HULIS) aerosol.

The scientific quality and presentation of the manuscript are good. The manuscript will need copy-editing because there are numerous grammatical errors, although the text is still clear. There is agreement between the various instruments utilised at the site (ACSM vs SMPS vs MARGA), and the PMF analysis follows established procedures. I do have questions about the determination of organic nitrates and about the correlations between the OA components and tracers, and these questions are given below. On the other hand, the scientific significance of the manuscript is not clear. Aerosol mass spectrometry (AMS) including ACSM measurements are fairly standard now, and AMS measurements were previously carried out at this site, as indicated in the manuscript. I acknowledge that the long duration of the measurements in this manuscript is somewhat novel, but it is not clear how having simply more measurements provides new significant insight into atmospheric chemistry. This concern will need to be addressed before final publication, in addition to the comments provided below.

General Comments:

Organic nitrates: The difference in the concentration of nitrate measured by the ACSM and MARGA is not a direct means of identifying and quantifying the presence of organic nitrates. Is there other evidence from the ACSM or other instruments to support the conclusion that these compounds are present and account for 9% of the total ACSM NO_3? If not, then this conclusion is rather weakly supported by the data.

Correlation of PMF time series with tracers: The correlations of the PMF factors with the tracer time series seems very low for HOA, BBOA and HULIS, and the correlation coefficients (R^2) are all below 0.5 for all the tracers used in this study. For example, the HULIS factor, which is presumably secondary in nature, exhibits the highest correlation with BC rather than one of the primary combustion factors (i.e. HOA and BBOA). This lack of correlation indicates that PMF is not identifying OA components that can be unambiguously associated with specific sources. The lack of correlation should be examined in more detail as it calls into question the validity of the PMF results.

Specific Comments:

P35121, L20: What is the difference between intensively and extensively managed grassland?

P35122, L2: Remote sensing is a very general term. The manuscript should specify exactly which remote sensing measurements are made at the tower site.

Page 35123, L27: In contrast with what is implied in the text, I believe that the Middlebrook et al. (2012) CE correction algorithm also accounts for high ammonium nitrate.

Page 35124, L5 – L8: Why was the detection limit not calculated during the measurement period or afterwards using data from the measurement period? Data from periods when the instrument was sampling behind a particle filter could be used for this analysis.

Page 35125, L23 – 29: The correction for the wall loses in the 60 m sampling line is a critical point for the manuscript. However, the description of how the losses were calculated or estimated is not sufficient. Only a reference to a personal communication is provided. Given that this correction can impact the aerosol measurements substantially – by 33% as indicated by the authors – the manuscript must contain a detailed explanation of how the aerosol losses in the sampling line are determined.

Page 35126, L13 – L16: During the measurements with the MARGA system, a polyethylene tube was used as the sampling line. Could the use of a non-conducting material for the line lead to substantial electrostatic deposition of the particles to the walls of the tube?

Page 35129, L7: Why is SO_2 included in the MARGA $PM_{2.5}$ mass? In the atmosphere, this compound is found in the gas phase.

Page 35129, L9: It appears that there is an error on this line and the concentration for the annual mean limit should be 10 $\mu g\ m^{-3}$ rather than 25 $\mu g\ m^{-3}$ as is currently written.

Page 35130, L6 – L9: This sentence is confusing. First, it is written that the pollution events are highlighted in green, but (to my eye) the highlighting is grey. Second, there are four periods that are highlighted in Figure 2, but only three periods are listed in the text. The dates for the missing period should be given as well.

Page 35132, L21: From Figure S10, it appears that the intercept is negative rather than positive (i.e. –0.80 versus 0.80). The sign is important as there is likely a small amount of dust and perhaps sea salt which is measured by the SMPS and not the ACSM that is responsible for the non-zero intercept value.

Page 35133, L18 – 20: Why couldn't the fragmentation table be adjusted? Adjustment of the fragmentation table is fairly standard during analysis of AMS data. If it is believed that the fragmentation table may be responsible for the underestimation of SO_4, then the authors should explore making possible adjustments of the values in the table.

Figure 1: Using zeros to fill in the missing values for black carbon for periods when measurements are not available is not a standard approach (to my knowledge). It would be preferable if the existing BC data was simply just averaged. It is already very clear from the manuscript that data is missing in certain periods, so a reader can take this into account when evaluating the data. Alternatively, the pie chart could be modified to include only the ACSM data for when the MAAP was functional.

Figure 4: I assume the authors mean to say "grey shaded areas" rather than "green shaded areas".

<u>Supporting Information:</u>

Page 2, L3: I think this equation should be $MF_{NO3} \geq 0.78$.

Figure S1: The figure legend text is very small and hard to read. The font size should be increased to improve readability.

Figure S2: I assume the authors mean to say "gray shaded areas" rather than "green shaded areas".

Figure S6: Similar to the previous comments, the shaded areas look grey to me rather than green. Perhaps there is a difference in how the colors are displayed by the author's monitor and my monitor? There are subsequent figures were this comment applies as well, but I will not repeat it to avoid being overly repetitive.

Figure S13: It would improve the presentation of the results if all the bar graphs were plotted with the same format. The size of the bars is not consistent.