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Interactive comment on “Seasonal and diurnal variations of particulate nitrate and organic matter in the Central European atmospheric aerosol” by L. Poulain et al.

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We would like to thank the referee for the constructive comments and suggestions made to improve the manuscript. Our responses to each comment are given below.

This manuscript investigates the seasonal and diurnal variations of organics and nitrate from 3 campaigns in central Europe during 3 different seasons, summer, autumn, and winter. While it is of no doubt that a lot of data have been collected, the data included in this manuscript seem inadequate to support many of the conclusions the authors intended to make. The discussions of the organics seem to be focused only on two m/z s, m/z 43 and m/z 44. I understand that these two m/z s are important markers for

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oxygenated OA (OOA), however, the authors did not perform any factor analysis in this work and so it did not provide any context why they only focused on these two m/z s. It appears that most of the discussions are based on the assumption that secondary organic aerosols dominate the total OA. However, no data are provided in the manuscript to support this. Instead of focusing the work on these two m/z s, when discussing the “aerosol composition”, they should at least show some mass spectra and also discuss other m/z s, such as m/z 55 and 57. The section regarding the partitioning of nitrates is highly speculative owing to the lack of many gas-phase measurements. Since the authors cannot provide the data to support their hypothesis, the arguments in that section are weak and require more work.

Response: We concede that a detailed description of the organic aerosol composition prior to the discussion on the seasonality OA oxidation state was missing. Our approach was mainly based on the high resolution analysis of the main organic fragments, especially the elemental analysis (OM/OC and O/C ratios). Therefore, we added a subsection describing the organic mass spectra of each season and a discussion on the seasonal and diurnal changes of the composition of the main organic fragments. Details can be found in our answer to the comment 12 (below). Additionally, the description of the nitrate diurnal profiles was improved by adding a discussion on the different factors influencing the ammonium nitrate concentration. Finally, to better estimate the contribution of nighttime chemistry, a simple model, based on reactions R3 and R4 and considering a complete heterogeneous dissociation of N_2O_5 in particulate nitrate, was run. This model was used to estimate the maximum of nitrate that could be expected by the nighttime chemistry. The results were then compared to the measured nitrate concentration. Details can be found in our answer to the comment 13 as well as in our answers to the comments of referees #1 and #2.

Specific comments:

1. Abstract, first sentence. The sentence is unclear. Are the authors referring to the uptake of PAHs and n-alkanes onto aerosol surface? As far as I understand, under

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most cases, these compounds do not exist in particle phase, on the other hand, the oxidation of such compounds can form products of low volatility that then partition into the aerosol phase (SOA formation).

Response: PAH and n-alkanes are semi volatile organic compounds that partition between gas and particle phases. The gas-to-particle phase partitioning of PAH and n-alkanes has been investigated in a number of theoretical and field studies (e.g. Radonic et al., 2011; Wang et al., 2011; Cincinelli et al., 2007; Sofowote et al., 2010 and references therein).

2. Abstract, line 15. What “discrepancy” are the authors referring to here?

Response: The abstract was changed according to the modifications made in the nitrate analysis; the corresponding sentence was removed.

3. Page 11613, lines 18 and 19. “depending” should be “dependent”.

Response: corrected

4. Page 11614, line 3. “depending” should be “dependent”.

Response: corrected

5. Page 11615, line 22. Delete “to”.

Response: corrected

6. Page 11616, lines 9 and 10. Awkward sentence. The aerodynamic lens does not “remove” the gas phase. It simply increases the particle concentration relative to the gas-phase species.

Response: We agree with this remark. However, the sentence was removed according to the specific comments #7 (below).

7. Section 2.2 in general. The authors can simplify the instrument description and operations of the HR-AMS and cite DeCarlo et al., 2006.

Response: As suggested by the referee, the AMS description was simplified.

8. Page 11616, line 17. “gas phase” should be “gas-phase species”.

Response: We agree with this remark. However, the sentence was removed according to the specific comments #7.

9. Page 11618, line 25. “considers” should be “taken into consideration”.

Response: corrected

10. Page 11618, line 28. “intake” – do the authors mean “inlet”?

Response: Absolutely. The text was corrected.

11. Page 11620. I think it would be better if the authors present the data in Table 2 as pie charts.

Response: Due to the large number of figures, we did not plot the content of Table 2 as pie charts.

12. Section 4.2. The title of this section is “Changes in organic particulate composition.”, however, the authors pretty much only discussed two m/z s in all of the AMS data, i.e., m/z s 43 and 44. After reading this section I did not get a very good idea regarding the organic particulate composition, other than the variations of these two m/z s and the OM/OC ratio. With respect to the amount of information one can obtain from HR-AMS data, the results presented in this section seem extremely inadequate.

Response: We totally agree with the referee. The section was renamed “Changes in organic aerosol” and was divided in two subsections, “Comparison of the organic aerosol mass spectra” and “changes on the organic oxidations states”.

a. The authors did not even show a single mass spectrum in this manuscript. How does the organic mass spectrum look like? Do they vary in different seasons? Such information should be included (The authors should at least present an average mass

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spectrum for each season).

Response: Details on the organic mass spectra over the campaigns were added in a new subsection 4.2.1. We now present a description of the organic mass spectra based on the presence of different ion categories ($Cx+$, $CxHy+$, $CxHyO+$ and $CxHyOz+$) as well as on the element composition of C, H, O atoms. We added the following text passage: “The averaged high resolution organic mass spectrum (HRMS) of each campaign is presented in Fig. 2. According to their composition, the organic fragments were divided in different ion categories ($Cx+$, $CxHy+$, $CxHyO+$ and $CxHyOz+$). The contribution of each category to total organic signal as well as the corresponding elemental composition of C, H and O atoms is also presented in Fig. 2. The contribution of $CxHy+$ was rather constant during the three campaigns and represented almost 50% of OA. The oxygenated categories, $CxHyO+$ and $CxHyOz+$, represented the second half of OA and a small change of the respective contribution of $CxHyO+$ and $CxHyOz+$ has been observed between summer and the two other seasons. However, the average elemental composition did not show pronounced seasonal variation.”

b. The discussions in this section appear to be built on the assumption that secondary organic aerosols dominate the total OA observed in these campaigns. Yet, the authors did not provide any data to support this. Do the authors have some ideas on the contributions of primary organic aerosols (POA) in these campaigns? One way to get some ideas on the relative importance of POA is to look at m/z 57. How does the times series / diurnal cycles of m/z 57 look like?

Response: The fragment m/z 57, which is commonly considered to be a tracer of primary organic sources, was made of two fragments, C_4H_9+ and C_3H_5O+ . While C_3H_5O+ represented 68% during summer, the contribution of the non-oxygenated fragment (C_4H_9+) increased in autumn (53%) and winter (57%). The diurnal profiles of m/z 57 for each season reveal a small increase of C_4H_9+ early in the morning during summer and autumn. This possibly indicates an increase of primary organic aerosol at this time. A similar conclusion can be drawn for m/z 55. Additionally, the

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contribution of m/z 60 to the total organics signal was close to 0.3% which has been considered to be representative for a background level value of SOA dominated ambient organic aerosol (Cubison et al., 2011). Therefore, we considered that during the different measurement campaigns, the organic aerosol measured at Melpitz was basically dominated by oxygenated organic aerosol whereas the contribution of primary organic aerosol was rather small.

The following text was added: “The fragment m/z 57 which is commonly considered as tracer for primary organic emissions of fossil fuel combustion (Zhang et al., 2005b; Canagaratna et al., 2004; Aiken et al., 2008) had a low contribution to total HRMS. The m/z 57 was made of two fragments, $C_3H_5O^+$ and $C_4H_9^+$. The contribution of each fragment to the total m/z 57 signal changed during the three campaigns (Fig. SI 2). While in summer, m/z 57 was principally made of $C_3H_5O^+$ (68% of total m/z 57 signal), the contribution of $C_4H_9^+$ increased in autumn (53%) and winter (57%). Comparing the diurnal profiles of these two fragments during the different campaigns, a small increase of $C_4H_9^+$ early in the morning during summer and autumn can be observed (Fig. SI 2). This might indicate a possible increase of primary organic aerosol at this time. It has been shown for a rural station in Canada that the ratios of $C_3H_5O^+$ to $C_4H_9^+$ increased with the photochemical aging (Liggio et al., 2010). Therefore, these authors suggest that the $C_3H_5O^+$ might be associated with SOA and/or oxidized POA. Following this, the largest contribution of the $C_3H_5O^+$ during summer might be attributed to a larger photochemistry aging OA, while the high correlation between $C_3H_5O^+$ and $C_4H_9^+$, observed during each campaign ($r^2 = 0.60$, 0.65 and 0.65 in summer, autumn and winter, respectively), might suggest a similar source for these two ions. Another important organic fragment is the m/z 55 which like m/z 57 is often associated to primary organic emission (Alfarra et al., 2004; Canagaratna et al., 2004). The fragment m/z 55 was made of $C_3H_3O^+$ and $C_4H_7^+$. Similarly to m/z 57, m/z 55 was basically made of the oxygenated fragment, $C_3H_3O^+$. However, a slight increase of the non oxygenated fragment could again be observed in autumn and winter (Fig. SI 2). In addition, the diurnal variation of the different seasons was very similar to m/z

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57. The composition of the main organic fragments (m/z 43, 55 and 57) indicated that in Melpitz, organic aerosol was dominated by oxygenated organic aerosol, with a low contribution of primary organic aerosol during the different measurement campaigns. Because of a larger concentration of $C_4H_9^+$ and a clear peak of $C_4H_7^+$ and $C_4H_9^+$ early in the morning during the autumn campaign, a small contribution of POA might be expected during this period. Finally, the presence of biomass burning organic aerosol is generally linked to the m/z 60 (Alfarra et al., 2007; Schneider et al., 2006). During the different seasons, m/z 60 had a small contribution to total OA (around 0.3%). Such low contribution of m/z 60 to total OA was considered as representative for a background level value of SOA dominated ambient organic aerosol (Cubison et al., 2011). Based on the m/z 60 contribution, it is concluded that there is only a minor influence of biomass burning aerosol on total OA, even during winter. However, like for POA, a limited contribution of biomass burning may be expected during the autumn campaign due to the small increase of m/z 60 contribution to OA.”

c. The authors wrote that the changes in organic oxidation levels are mainly examined through m/z 44 and 43 (mostly $C_2H_3O^+$). The authors seem to have framed their discussions based on that m/z 43 is mainly $C_2H_3O^+$, but no data were included to support this statement. Is the statement “ m/z 43 is mostly $C_2H_3O^+$ ” based on the HR analysis of their data? What do the authors mean by “mostly” (What % of m/z 43 is $C_2H_3O^+$ actually)? m/z 43 can be $C_3H_7^+$ or $C_2H_3O^+$. How does the diurnal cycle of these two fragments (instead of the “total m/z 43”) look like? Do the relative contributions of these two fragments change at different times of the day (e.g., in the afternoon hours when the photochemical activity is stronger, I would imagine the m/z 43 would be more dominated by $C_2H_3O^+$)?

Response: Our assumption was based on the fact that the fragment $C_2H_3O^+$ represented 92%, 84% and 80% of the total m/z 43 signal in summer, autumn and winter, respectively. We added the following description of the m/z 43 composition and the diurnal profiles: “The second most important peak was m/z 43. However, the contri-

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bution of m/z 43 to total HRMS decreased from summer to winter. The fragment m/z 43 was basically made of two fragments, $C_2H_3O^+$ and $C_3H_7^+$ (Fig SI 2). The fragment $C_2H_3O^+$ was the most important fragment of m/z 43 during the three campaigns. However, the contribution of $C_2H_3O^+$ was slightly higher in summer (92%), than in autumn (84%) and winter (80%) due to a slightly higher contribution of $C_3H_7^+$ in these seasons.”

d. m/z 44 can be used as an indication of the oxidation level of the aerosols, however, the data here at HR-AMS data, so instead of relying on m/z 44 the authors should present the O:C ratios calculated from elemental analysis of their data.

Response: As requested, the O/C ratio for each season was added. In addition, we also discussed the seasonality of the OA oxidation state using the newly developed Carbon Oxidation State (OSc) (Kroll et al., 2011). The following discussion was added: “Kroll et al. (2011) developed another approach to follow and compare changes of OA oxidation state. The authors determined the carbon oxidation state (OSc) from the O/C and H/C ratios, as follows: $OSc = 2 O/C - H/C$

The OSc diurnal profiles for each campaign are shown in Fig. 6. The resulting OSc values are in the range of previously reported values for remote/biogenic aerosol (-0.9 to -0.2) and in the lower range of the reported values for aged OA (-0.6 to +0.6) (Kroll et al., 2011). Summer and autumn periods presented similar OSc diurnal profiles. In line with the discussion above, this corresponds to an increase of the oxidation state during day time and can be attributed to aging processes. The lowest values of OSc and the absence of well pronounce diurnal variations in winter confirm the presence of less processed OA and only a small amount of aging during day. “

e. Page 11622, the sentence “Moreover, it is known that $f_{44} > f_{43}$ characterized a highly oxygenated and low volatile OA, while $f_{44} < f_{43}$ characterized a less oxygenated and semi-volatile OA (Ng et al., 2010)”. I think the authors are over-generalizing the results from Ng et al. (2010). Ng et al. (2010) put all the PMF OOA components from multiple

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sites into the f44 vs. f43 space and found that all components fall into a triangular region, with the LV-OOA components concentrating on the top of the triangle and the SV-OOA components concentrating on the bottom of the triangle. In this case, the authors did not perform any factor analysis on their data. First of all, they have to at least show that the m/z 43 is indeed dominated by the C₂H₃O⁺ fragment. Secondly, the statement “f44<f43 characterized a less oxygenated and semi-volatile OA” is not always true, for instance, in Ng et al. (2010), the f43 of SV-OOA components range from ~7% to 18%, there are actually SV-OOA components with f44>f43. Overall, I think the authors over-emphasized on f44 and f43 and ignored many other information they could have obtained from the analysis of their HR-AMS data.

Response: we concede that our sentence over-generalized the conclusion made by Ng et al. (2010). Therefore, we modified our sentences to “f44 > f43 generally characterized highly oxygenated and low volatile OA, while f44 < f43 generally characterized less oxygenated and semi-volatile OA.” Moreover, as previously mentioned (see our answer to comment b, above), OA at Melpitz was considered to be mainly made of oxygenated OA. Therefore, a direct comparison between m/z44 and m/z 43 could be performed.

f. Page 11624, line 10. Is this true for all m/zs? Otherwise the authors cannot say “organic compounds”. What about m/z 55, 57 etc?

Response: Here, we referred to the diurnal profile of the total organic signal and compared it to the profile observed during summer. The term “organic compounds” was replaced by “total OA”.

g. Page 11624, line 28. The authors stated that the local sources of OA in winter time are limited. Does this include POA? If so, it is not necessarily true that the POA contribution is lower in winter? What about biomass burning OA (BBOA)? Are there any BBOA in winter time?

Response: Biomass burning aerosol was estimated regarding the contribution of m/z

60 to the total OA (Cubison et al., 2011). Even in the winter campaign, the low f60 values (0.03%) suggest a limited contribution of biomass burning aerosol. As mentioned in our response to comment b, the following sentences were added: “Finally, the presence of biomass burning organic aerosol is generally linked to the m/z 60 (Alfarra et al., 2007; Schneider et al., 2006). During the different seasons, m/z 60 had a small contribution to total OA (around 0.3%). Such low contribution of m/z 60 to total OA was considered as representative for a background level value of SOA dominated ambient organic aerosol (Cubison et al., 2011). Based on the m/z 60 contribution, it is concluded that there is only a minor influence of biomass burning aerosol on total OA, even during winter. However, like for POA, a limited contribution of biomass burning may be expected during the autumn campaign due to the small increase of m/z 60 contribution to OA.

13. Section 4.3. This section is highly speculative, mainly owing to the lack of many important gas-phase measurements, such as NH₃, HNO₃, HONO etc required for their hypothesis.

Response: This section was entirely rewritten. We invite the referee #3 to read our comments and answers to the referees #1 and #2 for a complete description of our modifications. Nevertheless, we responded to the specific points mentioned by Referee #3.

a. The authors discussed the partitioning of nitrate as if this was a pure ammonium nitrate system. In reality, as the authors have shown, the aerosols consisted of organics, sulfate, chloride, and soot. These other components are present in non-negligible amounts. How does the presence of these components (esp. organics and sulfate) affect the discussions and conclusions in this section?

Response: We effectively considered a pure ammonium nitrate system and admit that the influence of the aerosol composition has to be considered. As discussed in section 4.1, aerosol can be considered to be fully neutralized during the three seasons. There-

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fore, we assumed that there is enough ammonia in the gas phase to fully neutralize nitrate and sulfate.

However, we added the following discussion to go describe the influence of the aerosol chemical composition in more detail: "The formation of ammonium nitrate also depends on the aerosol chemical composition as well as on the available ammonia in the gas phase. Indeed, ammonia is preferentially scavenged by sulfuric acid. Only when sulfate is neutralized, the excess of ammonia leads to the formation of ammonium nitrate (Seinfeld and Pandis, 2006). For all sampling periods, the ratio of total ammonium (TA = $\text{NH}_3 + \text{NH}_4^+$) to total sulfate (TS = $\text{SO}_2 + \text{SO}_4^{2-}$) was calculated. Most of the time, the ratio was larger than 2 corresponding to an excess of ammonia. At this level, the formation of ammonium nitrate is therefore possible. Moreover, the particulate ammonium to sulfate ratio was also estimated and the resulting averaged ratio of each campaign was larger than 2 which is interpreted as corresponding to ammonium sulfate formation (inserts in Fig. 8). The ammonium to sulfate ratio is in agreement with the neutralization state of the particles previously discussed (see section 4.1). However, during a few afternoons in summer, the ammonium to sulfate ratio was slightly below 2 (down to 1.6). This may indicate a possible small amount of ammonium bisulfate. These periods also coincided with important new particle formation events. Figure 8 shows the relationship between K_p^* and the ammonium to sulfate ratio. The highest K_p^* values happened during a low ammonium to sulfate ratio and a lower nitrate concentration. The relationship between nitrate formation and the ammonium to sulfate ratio was very well pronounced during the summer campaign, a period of strong photochemistry and sulfuric acid formation. This suggests a possible enhancement of nitrate depletion during day by larger ammonia consumption to sulfuric acid neutralization during summer. In Europe, the formation of ammonium nitrate is considered to be limited by the formation of HNO_3 (Sartelet et al., 2007). Unfortunately, no measurements of HNO_3 were performed here. However, we can note that HNO_3 does not react with NH_3 only; it can also directly interact with the inorganic fraction of the aerosol, e.g. calcium carbonate, magnesium carbonate, and sodium chloride, leading to the formation of calcium

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nitrate, magnesium nitrate, and sodium nitrate, respectively. As previously mentioned in section 4.1, contribution of Na^+ , Ca^{2+} , K^+ and Mg^{2+} was negligible due to their low concentrations compare to NH_4^+ , SO_4^{2-} and NO_3^- . Moreover, the presence of these cations on the coarse mode will limit the formation of ammonium nitrate in the fine mode. The comparison of the daily PM_{10} and $\text{PM}_{2.5}$ inorganic composition during autumn and winter is presented in Table 3. Larger differences between fine and coarse modes are observed for sodium, chloride and nitrate. This is in agreement with the western air masses influence, reported during the autumn and winter campaigns, which included a certain maritime influence. Nevertheless, as Melpitz is located far away from the sea coast (minimum distance of approximately 400 km); the sampling place is influenced by aged marine air masses rather than fresh ones. However, aging degree and for example conversion rate of NaCl to NaNO_3 depends on the history of the air mass over the continent (crossing or not polluted area, RH ...). Hence, in some specific situations (e.g. marine air mass with little processing), coarse mode may contribute to trap HNO_3 and therefore limit the ammonium nitrate formation. With only a daily time resolution, however, it is difficult to estimate the real contribution of the coarse mode on the nitrate equilibrium”.

b. The authors should include RH and T in Fig. 4 c.

Response: RH and T were added for all diurnal cycles.

Figure 5 is difficult to follow – I think scatter plots will serve the purpose better.

Response: In order to illustrate the dependency of nitrate concentration on both its equilibrium constant and the ammonium to sulfate ratio, the scatter plot of K_p^* vs. the ammonium to sulfate ratio colored by nitrate concentration was included in the figures.

d. The argument regarding the surface chemistry of HONO and HNO_3 is speculative owing to the lack of HONO and HNO_3 measurements. If the morning peak of NO is indeed produced by photolysis of HONO , shouldn't the authors expect OH also to have a peak around the same time NO peaks? (This is not seen from Fig. 6).

Response: We admit that the suggested HONO photodissociation early in the morning was rather speculative, especially due to the lack of any gas measurements. Based on the referee's comments, we decided to remove Fig. 6 and the corresponding discussion.

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Please also note the supplement to this comment:

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Interactive comment on *Atmos. Chem. Phys. Discuss.*, 11, 11611, 2011.

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