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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source apportionment of PM_{10} in a North-Western Europe regional urban background site (Lens, France) using Positive Matrix Factorization and including primary biogenic emissions" *by* A. Waked et al.

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The authors of this manuscript would like to thank the editor and both referee's for their fruitful comments which accounted for the improvement of the quality of this work.

Below you will find our response to the questions of the anonymous referee 1.

Responses to the anonymous referee 1

Major comments

Question 1 " 1. In section 3.3.3 and 3.3.4 the authors comment that the direction and hotspots from the PSCF analysis for primary biogenic and biomass burning factors could be calculation artifacts. Can the authors provide more explanation for this argument? Since according to the weighting function low number of back trajectories associated with high concentrations is down weighted anyway and account for a lower fraction of the PSCF values in Fig 7. How can then the authors justify the PSCF analysis for other PMF factors? Please elaborate."

Response 1 The authors agree with the fact that the PSCF interpretation for the biomass burning and primary biogenic factors are challenging. Conceptually speaking, investigating potential mid-/long-distance transport can lead to ambiguous results for local emissions as the wind speed is notably not considered in this approach (which is now stated in the revised manuscript). Thus, calculation artifacts are then very likely to occur for these sources, as the weighing function is optimized for transported emissions. Nevertheless, the same patterns were found in a similar PSCF analysis performed on wood burning emissions in Paris, about 200km away from Lens, in another study (Bressi et al., submitted in ACPD). For other sources, such as the non-local and secondary sources, the PSCF model is considered as appropriate because backward trajectories plotted by the HYSPLIT model at levels above 500 m Above Ground Level and used in the PSCF calculation are not associated with significant artifacts for these types of sources.

Question 2 "2. Pg 25352, L 12-14: How can the authors claim that SOA is mostly linked to sulfate rich and nitrate-rich organics that are originating from anthropogenic sources? While the contribution of nitrate-rich factor to OC is small most of the year except for in spring, photochemical processing of biogenic VOC to SOA is known during summer, as is the formation of ammonium sulfate. So, can it be argued that the OC associated with sulfate rich factor is just photochemical processing in the atmosphere of VOC (from both biogenic and anthropogenic sources) and therefore a clear source of organics associate with sulfate-rich factor is not possible?"

Response 2 The authors would like to thank the reviewer for highlighting this point and for the suggestions given. The paragraph was corrected in the manuscript and became as follows:

“The different contributions of OC sources retrieved from the PMF analysis are presented in figure 9. On an annual basis, major contributors of OC are biomass burning (24 % of OC mass) and primary biogenic emissions (17 % of OC mass). These results underline the large contribution of modern carbon (as opposed to fossil fuel combustion – derived carbon) within organic aerosols, which is in good agreement with previous studies (Yttri et al., 2011; Favez et al., 2010; Caseiro et al., 2009; Puxbaum et al., 2007; Yttri et al., 2007). The sulfate-rich secondary factor appears to be the third contributors to total OC, illustrating similarities within the processes leading to the formation of ammonium sulfate and secondary organic aerosols (SOA) as previously observed (Lanz et al., 2007; Ulbrich et al., 2009). Indeed, a seasonal variation was observed for the sulfate-rich source with a maximum contribution during the summer season where photochemical reactions processes leading to SOA formation are increased due to intense solar radiation. However, associating a clear source of organics to the sulfate-rich factor is not possible and could be misleading due to the lack of sufficient geochemical information’s and to the fact that ammonium sulfate formation is suspected to increase during summer. Besides these three major OC contributors, it is noteworthy to mention that other sources (aged marine, traffic, oil combustion, and dust) are still making up together 41 % of the total OC load, notably highlighting the non-negligible emissions of primary organic matter from anthropogenic activities.

Question 3 “3. Pg 25352, L18-21: Can the authors rephrase this sentence? It is not clear what the author’s main point is Âż

Response 3 Here, authors mean that the lower contribution for the nitrate-rich factor to total OC during summer could be related in a part to sampling artifacts where a part of particulate nitrate is volatilized due to its thermal instability. However, and due to the fact that the chemical mass closure during summer was satisfactory and sam-

pling artifacts related to ammonium nitrate could not definitely explain the decrease of nitrate-rich contribution in summer, the sentence related to ammonium nitrate was now eliminated from the manuscript and the paragraph became now as presented for the answer to question 2.

Question 4 “4. Pg 25353, L 5: “These discrepancies are firstly due . . ..”. Which discrepancies are the authors referring to?”

Response 4 We are referring to discrepancies generally observed between AMS-PMF and filter-based-PMF studies, which are introduced within previous sentences. This paragraph will be merged with the previous one, so that it is clearer that they both belong to the same discussion.

Question 5 “5. Pg’s 25353-25354: The paragraph starting with “Indeed, biomass burning emissions..”. I understand that the authors are trying to convey that using a large number of biomass burning tracers could help in better resolving the biomass burning contribution and other sources during winter. What is not clear is the discussion of nitrate-rich factor, semi-volatile nature of OA and its contribution to OC. In fact, nitrate-rich factor has minimal contribution during fall and winter seasons. I suggest rewording this paragraph.”

Response 5 This paragraph also refers to discrepancies that could (hypothetically) be observed between AMS-PMF and filter-based-PMF studies. It is known that wood burning emits high amounts of Semi-Volatile OA. However, on the one hand, AMS-PMF analyses generally indicate good correlation between ammonium nitrate and Semi-Volatile OOA; while, on the other hand, we did not observed here any significant mixing of wood burning OA with the nitrate-rich factor (which cannot be neglected in fall and winter). It thus comes that semi-volatile wood burning OA are rather directly included in the wood burning factor in our study (while they artificially stand alone when using AMS-PMF analysis). This paragraph was corrected and became as stated for the question 2, the contribution for the nitrate-rich to OC and OA was now revised.

Question 6 “6. General comment: Why is nitrate higher in spring? and not in winter when ammonium nitrate formation is efficient and volatilization from the aerosol is low.”

Response 6 As stated in the manuscript, at the end of winter beginning of spring, as already vastly reported, the increase of nitrate concentrations could be related in a part to the semi-volatile character of ammonium nitrate and frequent occurrences of photochemical episodes during a period of the year which also corresponds to rather humid conditions, low temperatures and increased agricultural activities in this part of Europe (Dall'Osto et al., 2009). Accordingly, ammonium concentrations are also higher in spring and are mostly associated to nitrate with a coefficient of determination (R2 = 0.95) between nitrate and ammonium which is not surprising due to the fact that agricultural activities are increased during this season.

Minor comments

Question 1 “1. Were PM10 samples collected daily during this study period? First sentence of sections 2.1 and 2.2 implies that PM10 samples were collected daily. Or did the authors mean fitlers were collected every third day for 24 hrs?”

Response 1 Filters have been collected every day for 24 hrs, but every third filters only were analyzed.

Question 2 “2. Which meteorological fields were used for PSCF analysis?” Response 2 PSCF calculation is only based on counting back-trajectories into grid cells. However, as mentioned in section 2.4 of the discussion paper (line 9-12), wet deposition was roughly estimated with precipitation data, calculated along back-trajectories.

Question 3 “3. Pg 25333, L 8: Change “under Python” to “using Python”.

Response 3 “Under Python” was changed to “using Python”

Question 4 “4. Pg 25336, L 9: Change “were slowly increased” to “increased”.

Response 4 “were slowly increased” was changed to “increased”

Question 5 “5. Pg 25336, L 27: Delete “number of”

Response 5 “Number off” was deleted

Question 6 to 10 6. Pg 25337, L 16: Change “injected” to “used”. 7. Pg 25339, L 10: Change “dispatched” to “divided” or “separated". 8. Pg 25339, L 14-15: Please rephrase the last sentence” Finally, introducing..”. 9. Pg 25342, L 5: Change “largely decreasing” to “are lower”. 10. Pg 25343, L 20: Change “shares” to “fraction”.

Responses 6 to 10 Pg 25337 injected was changed to used Pg 25339 dispatched was changed to separated Pg 25339 the sentence “ Finally, introducing additional factor is lowering loosely the values of Q robust and Q true” was changed to “Thus, introducing additional factor to the PMF analysis is lowering inaccurately the values of Q robust and Q true” Pg 25342 “largely decreasing” was changed to “are lower” Pg 24343 “Shares” was changed to “fraction”

Question 11 “11. At several places in the text the authors cite different regions of Northern Europe (ex. Nord Pas de Calais etc.). It would be good to also include the general direction from the site as this would help readers who are not familiar with regions in this part of the Europe (world) Âż

Response 11 In the manuscript, the general directions from the site for these regions were added in the manuscript.

Question 12 “12. Pg 25349, L 23: The authors say that aged marine source shows strong seasonal variation but from Fig 4 it seems like this factor is lower only during spring and it pretty constant during the rest of the year. Its contribution to PM10 is slightly higher during summer but there is no strong seasonal variation as the authors suggest”

Response 12 The sentence was corrected in the manuscript.

“This source follows a strong seasonal variation with the highest contribution (23 %) in the summer season. This seasonal variation is also observed for the sulfate-rich

factor with a contribution increasing from 14% to 19% during summer. The comparable seasonal variation observed among the sulfate-rich and the aged marine aerosol factors along with an important share of sulfate (22% of the sulfate mass) suggests that these marine aerosols acquire an anthropogenic signature as a result of the combustion of. . .” Was corrected to : “The contribution of this source to total PM10 mass showed the highest values (23%) in the summer season. This variability was also observed for the sulfate-rich factor with a maximum contribution accounting for 19% during summer. The comparable variation observed among the sulfate-rich and the aged marine aerosol factors along with an important share of sulfate (22% of the sulfate mass) suggests that these marine aerosols acquire an anthropogenic signature as a result of the combustion of”

Question 13 “13. Pg 25355, L 6: global chemical composition of PM10 in Lens?”

Response 13 Global chemical composition was corrected to global chemical mass of PM10 in Lens

Question 14 “14. Fig 4: Adding PM10 mass concentration beside each pie chart might help the reader quickly estimate the contribution of each factor in terms of mass concentrations.”

Response 14 PM10 mass concentration was added to figure 4

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/13/C10190/2013/acpd-13-C10190-2013-supplement.pdf

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