

To Dr André Prévôt

Head of Gasphase Chemistry Group at the Paul Sherrer Institute (PSI) Paul Scherrer Institut Labor für Atmosphärenchemie CH-5232 Villigen PSI Switzerland

December 11th, 2013

Dear Editor,

We have revised the manuscript entitled " Source apportionment of PM_{10} in a North Western Europe regional background site (Lens, France) using positive matrix factorization and including primary biogenic emissions " (ACP-2013-454), taking into account the editor and the referee's comments.

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 2.

Sincerely yours,

Antoine WAKED.

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 54, rue Molière, Domaine Universitaire, BP96, 38402 Saint-Martin d'Hères cedex, France Tél. +33 4 76 82 42 00 Fax +33 4 76 82 42 01

 E-mail : direction@lgge.obs.ujf-grenoble.fr
 Web : http://www-lgge.obs.ujf-grenoble.fr









Laboratoire de Glaciologie et Géophysique de l'Environnement

Responses to the anonymous referee 2

General comments

Question 1

"No mention is made about whether the authors considered rotational ambiguities in the solution sets. If any tests were performed, it would be very appropriate to document them in the supplementary material. I am also concerned that not enough attention has been paid to documenting the reasons for choosing a 9 factor solution. The only reason reported in the supplementary material appears to be because at 10 factors starts to produce less favorable diagnostics from the bootstrapping analysis. It would be more informative if more detail on the rejected solution set could be given, such as the authors' opinions on whether the profiles were physically meaningful, or the results of any other tests (e.g. seed variation) performed on the data."

Response 1

In fact, rotational ambiguities were indeed considered in the solution sets and the results were robust and satisfactory for the 9-factor solution; these results from rotational ambiguities were added to the supplementary material. It is now clarified in the manuscript and supplementary material that the reasons for choosing a 9-factor solution are not related only to the diagnostics from the bootstrapping analysis. As it was stated in the manuscript, tests were performed up to 14 factors, and adding factors was lowering the values of Q robust and Q true loosely. In addition, with 10 factors, an identified additional profile was not physically meaningful and it was impossible to link this factor to a common source.

Question 2

"I think the emphasis placed on a 'discrepancy' between this and PMF results from the AMS is largely unwarranted for the pure and simple reason that two different size fractions are being studied here. Even if the results of the analytical methods were directly comparable (which they are not), I personally would not expect a factorization of PM1 (which is dominated by combustion and secondary aerosols) to yield the same results as PM10 (which includes dust, sea salt and biological particles) anyway. That is not to say that the discussion comparing the techniques isn't warranted, but I would not interpret them as being in disagreement."

Response 2

We agree that the studied size fraction is an important parameter to be taken into account when trying to compare AMS-PMF analyses to filter-based-PMF studies. This is already stated Page 25353 (L.5), and is certainly true for sources leading to some fraction of coarse particles (like crustal sources) However, as an example, contributions of wood burning aerosols (with PM mostly in the submicron fraction) obtained from AMS-PMF analyses are generally lower when compared to filter-based methods, an indication that the different sampling size range is probably not the sole reason of these differences.

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Specific/technical comments:

Question 1

"Page 25331, line 17: Why were these species excluded and not dramatically 'downweighted' (i.e. uncertainties artificially increased)?"

Response 1

At first, we tried to downweight these species. However, increasing uncertainties has its limits to the point where the results are not meaningful anymore and do not add any constrain to the model. For this purpose, and due to the fact that downweighting these species did not solve the problem, we were obliged to exclude them.

Question 2

"Page 25336: I would suggest the authors do not use uppercase sigma to denote 'total' in line with freeform text (e.g. 'sugars'), as combining the two makes reading difficult. Simply writing e.g. 'total sugars' or using an acronym would make it clearer."

Response 2

In the manuscript "total polyols" replaced "SPolyols"

Question 3

"Page 25336: mg/g or mg g-1 are not legitimate units. The fractions should be expressed as a decimal or a percentage, qualified as 'by mass' if necessary".

<u>Response 3</u> A percentage was added to mg/g values as recommended by the reviewer

Question 4 "Page 35339, line 17: Correct 'specie' to 'species'".

<u>Response 4</u> Specie was replaced by species

Question 5

"Page 25342: Both on- and offline measurements of PM10 are known to be affected by losses of semivolatile ammonium nitrate, especially the methods that require the substrate to be heated to remove water vapor. While the authors refer to this later on, the authors should comment on whether they think that this is an issue for these measurements here."

Response 5

We agree with the reviewer that a part of the decrease of ammonium nitrate might be due to sampling artifacts. However, for quantification of ammonium nitrate using ion chromatography, there is no heating of the substrate to remove water vapor. Moreover, good chemical mass balance is achieved in summer (when compared to Beta gauge instrument equipped with a "smart heater"

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sampling line, the so-called RST system), implying small losses of semi-volatile species, including ammonium nitrate. Indeed, for the artifacts related to PM_{10} measurements, we believe that the uncertainty is not important due to the fact that the coefficient of determination R^2 between measured PM_{10} and reconstructed PM_{10} is 0.97 and 0.90 in the summer season. This issue was more clarified in the manuscript as follows :

"lower summer contributions of ammonium nitrate might partly be related to meteorological conditions and to the semi-volatile character of this compound as discussed above. Indeed, during summer, a part of ammonium nitrate is suspected to volatilize due to higher surface air temperature. This sampling artifact could also slightly affect the contribution of this factor during summer. Besides, artifacts related PM_{10} measurements should not be very significant due to the fact that the coefficient of determination R^2 between measured PM_{10} and reconstructed PM_{10} is 0.97 and 0.90 during summer."

Question 6

"Page 25350, line 2: The turn of phrase 'as a matter of fact' is not appropriate in this context, because the PSCF does not point to an irrefutable detail."

Response 6

The term "as a matter a fact" was replaced by "furthermore" in the manuscript.

Question 7

"Page 25350, line 5: Change 'highest ships density' to 'highest density of ships"

Response 7

'highest ships density' was changed to 'highest density of ships".

54, rue Molière, Domaine Universitaire, BP96, 38402 Saint-Martin d'Hères cedex, France Tél. +33 4 76 82 42 00 Fax +33 4 76 82 42 01 E-mail : <u>direction@lgge.obs.ujf-grenoble.fr</u> Web : <u>http://www-lgge.obs.ujf-grenoble.fr</u>







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