Dear Editor,

Please find below our response to the last comments raised by Reviewer #2 about acp-2014-279 revised manuscript.

First of all, we sincerely thank Reviewer #2 for his interest in our work and for pointing out "the authors' effort on performing and documenting the extensive sensitivity tests, which is rarely seen in receptor model papers". In respect with this comment, the authors do not clearly understand the reason why the rating of the manuscript Scientific Quality moved from "Excellent" to "Good". Nevertheless, we have tried here again to take into consideration the new comments raised by Reviewer#2 with explanations that will, hopefully, convince the Editor and Reviewers that our manuscript meets *Atmospheric Chemistry and Physics* standards.

Reviewer #2: "However, the reviewer still has reservations on the "sequential" PMF analysis and does not believe it will ultimately perform better than conventional approaches with respect to PM source apportionment at this site. As pointed out by the other reviewer and also the authors, every step of PMF analysis, e.g., estimating weights, implementing constraints, as well as deciding number of factors and other parameters, require -work/assumptions. The two-step PMF analysis will likely make the solutions more guess subjective and dubious."

Authors:
First, the subjective part of each PMF calculation is user-related, and partly lies in the naming and interpretation of factor profiles and time-series. Previously published PMF results are then subjective from this point of view. Our first PMF approach (source apportionment of OA sources from ACSM data) is similar to the large number of those published in the literature; it is not more subjective and is successfully compared to previous OA source apportionment performed in the region of Paris and other megacities of the Northern Hemisphere (Zhang et al., 2007). Our second PMF uses mathematical investigations, which provide robust guidelines (e.g. residuals examination) to be as 'objective' as possible. These tests have already been addressed in the manuscript as highlighted by the Reviewer.
Second, PMF^2 is one among other alternative PMF approaches that are currently being submitted/published to *Atmospheric Chemistry and Physics* (Sun et al., 2012; Crippa et al., 2013; McGuire et al., 2014). These different PMF approaches do not address in more details than our paper the subjective and dubious pattern of their results.

For all these reasons, we strongly believe that the proposed methodology is not necessarily more subjective than similar approaches that have been and/or are currently being published.

Reviewer #2: "More works need to be done to understand how uncertainties multiply in the PMF×PMF process and provide convincing evidences that the "overall" uncertainty and/or bias are manageable. This may be achieved using synthetic data similar to what have been done for one-step PMF analysis (see the U.S.EPA PMF 5.0 manual for references). "

Authors: As mentioned previously, our first PMF results are, at most, as subjective as many others reported in the literature for OA source apportionment by AMS/ACSM. To our best knowledge, there are no tools currently available to better constrain this type of uncertainties (for instance, bootstrapping is not available in the current PMF tools used with ACSM/AMS data). Our second PMF results are certainly more robust given the sensitivity tests reported in the paper. As a result, there are no reasons to think that uncertainties are multiplied using PMF^2. Although such overall uncertainty of the PMF^2 remains challenging to assess, the authors believe that this should not prevent this first ever publication on real-time PM_1 source apportionment from ACSM to be published.
Moreover, the use of a synthetic database is challenging by virtue of the inherent complexity of building such data. It is essentially based on fixed factor profiles, and temporal contributions empirically managed, or determined through chemistry-transport models. We seriously doubt that any empirical approach would "validate" our results. Then, CTMs are surely a more robust methodology, but can suffer from significant uncertainties, especially regarding organic aerosols, as described in the region of Paris by Petetin et al. (2014).

Reviewer #2: "Whether or how the bootstrapping method (working on the original input data) can be applied to the two-step PMF is also worth examination."

Authors: As stated in the manuscript, bootstrapping is actually applied to the second step of our PMF^2 approach. Concerning the first step of our approach, it intentionally consists of a "traditional" AMS/ACSM PMF analysis, which is performed all over the world for about 10 years without any bootstrapping examination.

Reviewer #2: "PMF analysis including only total OA followed by multi-linear regression analyses, as suggested by this reviewer, will also "bridge OA fractions obtained using traditional AMS-PMF methodology". It can apportion HOA, p-BBOA, and OOA into sources likely as well as, if not better than, the PMF×PMF approach and mitigate the empiricism of HOA, p-BBOA, and OOA quantification in the PMF analysis."

Authors: Following reviewer's suggestion, a multilinear regression approach has been considered as follow: a PMF analysis with OM, inorganic ions and BC fractions led to a 4-factor solution; wood burning- and traffic-related factor, and ammonium nitrate- and sulphate-rich factor (see Fig. AR1 below). Although the stability of the runs over different seeds and bootstrap analysis strengthen the stability of the solution, OM is not fully reconstructed (slope=0.89 and r^2=0.90, Fig. AR2a), contrasting with the OM reconstruction from the PMF^2 approach (slope=0.99, r^2=0.91, Fig. AR2b). Thus, any further calculation from these results would suffer from additional uncertainties. Nevertheless, a multilinear regression was applied with OM concentrations from the obtained factors ($OM_{Fi}=f_{OM}$. $[F_i]$, where f_{OM} is the mass fraction of OM in F_i profile) to investigate the distribution of the OA fractions (HOA, pBBOA and OOA). It has been performed with the Analysis Toolkit macro of Microsoft Office Excel, where the origin ordinate has been fixed to zero. The resulting equations are listed below:

$HOA = 0.20*OM_{WB} + 0.53*OM_{TR} - 0.02*OM_{NO3\text{-}rich} - 0.11 *OM_{SO4\text{-}rich}$ $r^2=0.82$
$pBBOA = 0.46* OM_{WB} + 0.03* OM_{TR} - 0.05* OM_{NO3\text{-}rich} - 0.29* OM_{SO4\text{-}rich}$ $r^2=0.89$
$OOA = 0.44* OM_{WB} + 1.01* OM_{TR} + 1.05* OM_{NO3\text{-}rich} + 0.77* OM_{SO4\text{-}rich}$ $r^2=0.96$

These results similar general features of the PMF[2] approach: HOA is explained by both wood burning and traffic, BBOA is mostly explained by wood burning (but the significant negative coefficient for $OM_{SO4\text{-}rich}$ does not make much sense), and OOA seems to be distributed over the 4 factors. Although some of these results are kind of acceptable, several sources of uncertainties also need to be addressed. This methodology is based on 3 different steps (OA source apportionment, PM_1 source apportionment with OM, and multilinear regression). If consecutive analyses make results more dubious, due to uncertainties propagation, then this approach is conceptually more questionable. Also, multilinear regression does not allow positive constrains. Finally, it is recall here that the latter calculation is based on OM concentrations that are not fully reconstructed from the PMF analysis suggested by the Reviewer, while the first PMF analysis used in our approach allows a more accurate reconstruction of the total organic matter (See Fig.5b of the manuscript).

Reviewer #2: "Inter-comparison of various PMF results is indeed critical to this study if the goal of this paper is to demonstrate a "novel" method that can work better than existing methods and advance our understanding of source contributions. The reviewer does agree that this method will be a valuable addition to the current sets of methods providing weight-of-evidence for source apportionment at this and other monitoring sites."

Authors: Again, the authors do not claim that the "novel" methodology we are proposing is intrinsically better than other existing methods. As also agreed earlier, comparing the proposed methodology to other approaches is of interest. However, there is currently no standardized protocol for such a PM_1 source apportionment, so that an intercomparison exercise may not be useful to assess the validity of the presented results, and is *de facto* out of the scope of the paper.
Finally, the authors would like to underline that other alternative methodologies recently published within *Atmospheric Chemistry and Physics* (Sun et al., 2012; Crippa et al., 2013; McGuire et al., 2014) compared, as in our case, their results to a "traditional" OA source apportionment analysis, but have been accepted without any other kind of intercomparison.

On behalf of all the authors,
Best regards,
Jean-Eudes Petit

References

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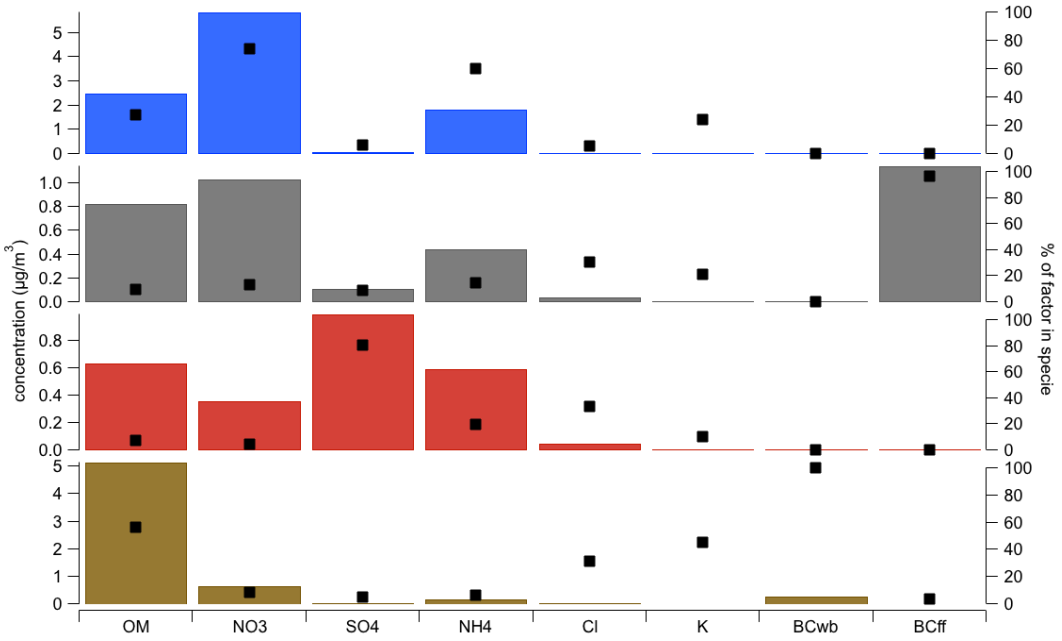

Figure AR1: Factor profiles of the 4-factor solution from a PMF analysis with OM, inorganics and BC fractions

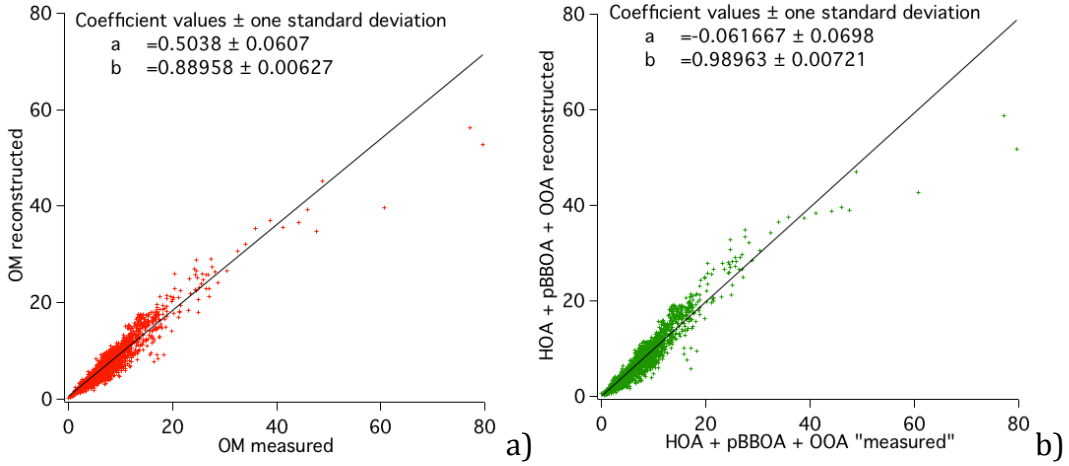

Figure AR2: comparison of measured OM versus reconstructed OM from a PMF analysis with OM, inorganics and BC fractions (a), and from the second PMF of the PMF^2 approach (b).