Source apportionment of highly time resolved trace elements during a firework episode from a rural freeway site in Switzerland

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Response to Reviewer #3

We kindly thank the reviewer#3 for taking our manuscript into consideration and we value the comments raised to improve the manuscript. A point-by-point answer (in regular typeset) to the reviewers' remarks (in italic typeset) follows. Changes to the manuscript are indicated in blue font.
In the following page and line references refer to the manuscript version reviewed by anonymous Reviewer#3.

The paper "Source apportionment of highly time resolved trace elements during a firework episode from a rural freeway site in Switzerland" by Rai et al. deals with Positive Matrix Factorization analysis of a dataset of 1-h resolved trace elements. Despite the authors declare the existence of much more information on high time resolved scale (mass concentration by TEOM, equivalent black carbon by MAAP, ACSM data), they carried out the analysis on elements only. This limits strongly the information provided by the study (e.g. they can apportion only the mass related to elements, that they estimated to be about 20% of PM10 mass). Thus, the results cannot be representative
of the total contribution of the sources to the measured PM. Furthermore, lots of constraints were implemented to reach the final solution. In some cases, both source profiles and temporal trends were constrained. So many constraints to the model make questionable the validity of the results, also considering that these constraints are not adequately supported by a methodological description of the way they were obtained, as explained in more detail below. Whether the paper is well structured and well written in most parts (even if some obscure descriptions remain), the scientific aspect is not fully convincing.

Major concerns:
Page 5, line 15: excluding mass data (mentioned at p.4 l.2) should be carefully discussed as it strongly reduces the interest of the results, preventing an absolute quantification of the factor contributions to the measured PM. Furthermore, maybe the authors decided to analyse separately ACSM data, but at least equivalent BC information could be of help in source resolution.

This issue was also raised by Reviewer #2 (major comments section) and our response is presented in both places for clarity. The exclusion of elements was done based on online (Xact 625) and offline (ICP analysis) elemental quality comparison as discussed in Furger et al. (2017). We modified the text on Pg 5 line 20:

Elements (% of data points below MDL, r^2 value) which had more than 50 % of data points below MDL and low r^2 (0.5) between Xact and offline data were not included in the PMF input, such as: V (98 %, 0.57), Co (100 %, 0.05), Ni (32 %, 0.22), As (96 %, 0.5), Se (62 %, 0.3), Cd (87 %, 0.18), Sn (15 %, 0.27), Sb (6%, 0.42), Hg (13 %, 0.12) and Pt (98 %, not measured on the filters). The element Bi (93% data points below MDL) was an exception to include in the PMF input due to an excellent correlation between Xact and offline data (r^2 = 0.98) during fireworks peaks. The detailed description of the Xact data quality is given in the previous study (Furger et al., 2017).

Concerning ACSM and BC data, the same issue was also raised by Reviewer#1 and Reviewer#2. We repeat here the same. In our opinion, the combination of PM_1 data for Q-ACSM, $PM_{2.5}$ for MAAP and PM_{10} data for Xact may introduce artefacts in the ME-2 analysis. We have used NO_x, MAAP and Q-ACSM data sets in the manuscript to compare some of the ME-2 factor time series and diurnal patterns. We are not planning to publish a separate paper on the other data. A major focus was the exploration of the use of the Xact for source apportionment of inorganic elements / metals in Europe where the concentrations are considerably lower than in polluted areas in Asia.

Furger, M., Minguillón, M. C., Yadav, V., Slowik, J. G., Hüglin, C., Fröhlich, R., Petterson, K., Baltensperger, U., and Prévôt, A. S. H.: Elemental composition of ambient aerosols measured with high temporal resolution using an online XRF spectrometer, Atmos. Meas.Tech., 10, 2061–2076, https://doi.org/10.5194/amt-10-2061-2017, 2017.

Page 6, lines 11-12. "The unconstrained PMF solutions yielded mixed factor solutions. Therefore, it was essential to constrain specific factor profiles and the time series in the PMF analysis to avoid mixing (see details in the Supplement, section S1, Fig. S1)". This is the weakest aspect of all. The way in which the factor profiles/time series are constrained is the key point for all the rest of the analysis. Constraints determination strongly affects the final results (see described differences between the preliminary analysis and the constrained one) and merits detailed description and attention. Opposite, its description was moved to the Supplemental Material, and what is reported there is a not sufficient to determine the robustness of the approach. The following information should have been provided: 1) How many factors were present in the final analyses of the fireworks and non-fireworks periods? 2) Were the sources other than fireworks and sea-salt comparable between the datasets? (In terms of profile and tracer species?) 3) Were 2 firework-related factors identified in the analysis of the firework-days subset? 4) What about the residual of S in the unconstrained 9 factor solution? 5) The need to constrain both profile and temporal trend of the sulphate source is very suspicious (the whole "secondary sulfate" factor was constrained in the final solution).

We agree with the reviewer#3. We have rewritten the section S1 below and we moved this section into the main text. However, we would like to respond point by point to the reviewer's five questions.

The input data set was divided into two parts: fireworks days (FD; 31 July–4 August) and non-fireworks days (NFD; all days except 31 July–4 August). To obtain a specific fireworks profile, we further selected only fireworks hours (FH; 31 July 21:00–1 August 07:00 LT) as PMF input data. The PMF analysis was performed on the NFD, FD, FH and the complete datasets separately for three to ten factors, with each of these solutions investigated with ten seeds (each seed represents a different pseudorandom initialization).

1) Seven factors were resolved in the NFD PMF analysis while a five-factor solution was identified in the FH PMF analysis.
2) The unconstrained NFD PMF analysis resolved factors such as sea salt, secondary sulfate, traffic-related, road dust, background dust, industrial and a K-rich factor. The FD and FH constrained PMF analyses resolved a five-factor solution with secondary sulfate, sea salt, fireworks, background dust and a K-rich factor with fireworks related elements. The traffic-related and road dust factors were resolved in FD and FH PMF analyses in seven and eight-factor solution, which were comparable to NFD PMF analysis. The fireworks factor contribution was going down in seven and eight-factor solution than the five-factor solution. The industrial factor in the FD and FH PMF analysis was resolved in eight-factor solution where it was mainly characterised by high contribution to Pb (67 %) and Zn (30 %) whereas in NFD PMF analysis the relative contribution to Pb and Zn were 90 % and 87 %, respectively.
3) The FD and FH constrained PMF analyses identified two fireworks-related factors, which were comparable to the final two fireworks-related factors profiles.
4) The scaled residual of S in the unconstrained nine-factor solution was between -0.46 and 0.85.
5) We tested several approaches for the secondary sulfate constraint: a) constraining only the profile; b) constraining only the time series to preserve secondary sulfate temporal trend during fireworks; c) constraining the entire time series along with factor profile; d) constraining the factor profile together with a segment of the time series (i.e., only during fireworks days). From all the above four tests, unmixed secondary sulfate was possible only in case of c and d, and we prefer the approach in d as it provides maximum freedom to the algorithm. We suggest that the need to constrain both the profile and part of the time series is driven by the very high concentration and variation in composition during the fireworks period. Because the signal-to-noise is very high, imperfections in the model description exert a strong influence on Q, and the model therefore tries to compensate by assigning fireworks mass to other factors. The double constraint tactic in d avoids this problem, while minimizing overall constraints on the solution.

These points, as well as the requested general information regarding the PMF analysis, are added to the manuscript beginning at Page 6 line 9:

In a first step, we examined a range of solutions with three to ten factors at ten seeds (number of PMF repeats) from unconstrained runs. The unconstrained PMF solution resulted in mixed factors, such as sea salt mixed with fireworks, even for higher numbers of factors (Fig. S1). This is likely because of the very high concentration and variation in composition of fireworks emissions during the fireworks period. Because the signal-to-noise is very high, imperfections in the model description exert a strong influence on *Q*, and the model therefore tries to compensate by assigning fireworks mass to other factors. This was particularly evident for the sea salt and secondary sulfate factors, where constraints on factor profiles and/or time series were necessary to obtain clean separation. Here we discuss the method for achieving this separation.

The input data set was divided into two parts: fireworks days (FD; 31 July–4 August) and non-fireworks days (NFD; all days except 31 July–4 August). To obtain a specific fireworks profile, we further selected only fireworks hours (FH; 31 July 21:00–1 August 07:00 LT) as input data. The PMF analysis was performed on the NFD, FD, FH and the complete datasets separately for three to ten factors, with each of these solutions investigated with ten seeds (each seed represents a different pseudorandom initialization).

The unconstrained NFD PMF analysis resolved seven factors at all ten seeds such as sea salt, secondary sulfate, traffic-related, road dust, background dust, industrial and a K-rich factor. The sea salt factor profile shows excellent correlation (r^2= 0.99) between the Cl and the identified sea salt factor time series. Solutions with less than seven factors showed significant scaled residuals for elements and time series, while solutions with more than seven factors revealed a split of the traffic-related, industrial and background dust factors. The NFD analysis therefore provides a sea salt profile that can be used as a constraint in the complete dataset.

The unconstrained PMF analysis of the complete dataset identified a secondary sulfate factor (most of the S is apportioned in this factor, with 91 % of the factor mass) in the nine-factor solution (Fig. S1). The identified secondary sulfate factor time series correlated very well with ACSM sulfate (r^2 = 0.91) (Fig. S1) at one seed out of ten seeds while r^2 was ≤ 0.88 for the remaining nine seeds. The scaled residual (over time series) of S in this solution was within the range of -0.46 to 0.85. Although these r^2 are quite similar, the solution characteristics are notably different. For the r^2 = 0.91 solution the secondary sulfate factor did not respond significantly to the fireworks event, while the other factors time series, such as the sea salt and a mixed traffic plus K-rich factors, were enhanced during the fireworks peaks. For the other nine seeds, visible contamination (i.e. concentration spikes) during the fireworks plumes were observed, suggesting mathematical mixing. S is one of the major components of fireworks emissions, the composition of which is highly variable. Because of their high sensitivity (and thus high signal-to-uncertainty ratio), imperfections in the model description of the fireworks composition yields high-signal residuals which strongly influence *Q*. The model responds by apportioning fireworks residuals to the other factors during the fireworks days. A similar issue also occurred for the sea salt factor due to the significant amount of Cl in the fireworks factor profile. Therefore, such events are often excluded from traditional PMF analyses (i.e., time periods removed from the input matrix), to avoid modelling errors due to the pulling of a solution by outliers. Here we take a different approach, exploiting the rotational control available in ME-2 to isolate environmentally reasonable, unmixed solutions.

We then performed the constrained PMF analysis on the FD and FH datasets. Here we constrained the secondary sulfate factor profile (*a*-value 0.1) and the time series (*a*-value 0.01) using the results of the 9-factor unconstrained PMF analysis of the NFD dataset. We tested several approaches for the secondary sulfate constraint: a) constraining factor profile only; b) constraining the factor time series during fireworks days only; c) constraining both the factor profile and the entire factor time series; d) constraining the factor profile and the factor time series during fireworks days only. Of the above methods, only c and d yielded secondary sulfate factors without visible mixing from the fireworks period. The approach d was used for PMF analysis as it provides maximum freedom to the algorithm.

In the FD and FH PMF analyses, the sea salt factor profile (*a*-value 0.1) was also constrained from the NFD unconstrained seven-factor PMF analysis. To resolve the unmixed sea salt factor time series from the fireworks, the background Cl concentration was calculated for the fireworks data points (K > 220 ng m^{-3}) only. A Cl concentration < 30 ng m^{-3} was considered as a background Cl concentration, and the fireworks data points were replaced with the linear interpolation between the background Cl concentrations adjacent to the fireworks peaks. In this way 42 % of the data points were interpolated during the fireworks days. The calculated background Cl during the FD was constrained (with *a* -value 0.01) in the sea salt factor time series. After applying all the above four constraints, the FH PMF analysis identified the fireworks factor profile on the basis of the K / S elemental concentration ratio (~2.76) in black powder (Dutcher et al., 1999), and on the concentration peak of 42 µg m^{-3}, which is close to the total elemental concentration peak of 48.4 µg m^{-3} on 1 August 23:00 LT in the factor time series. The FD PMF analysis also identified fireworks factor but the highest peak was 30 µg m^{-3} on 1 August 23:00 LT in the factor time series and the K / S elemental concentration ratio was 2.55 in the factor profile. Therefore, the fireworks factor profile from FH PMF analysis was considered for the final complete dataset PMF analysis. The FD and FH PMF analyses resolved a five-factor solution with secondary sulfate, sea salt, fireworks, background dust and a K-rich factor with fireworks related elements. In the K-rich factor, the K / S ratio was slightly higher (3.56) than the black powder ratio. The common factors resolved by FD, FH and NFD PMF were comparable in terms of factor time series and factor profile.

In the final complete dataset PMF analysis, the factor profiles of fireworks, secondary sulfate and sea salt were constrained (*a*-value 0.1) while the time series of secondary sulfate and the calculated background Cl concentration interpolation were constrained during the fireworks period only (*a*-value 0.01). The solution that best represented the input data was an eight-factor solution, consisting of factors interpreted as sea salt, secondary sulfate, traffic-related, industrial, two dust-related and fireworks-related (two factors).

Residual analysis (Q-contribution over time series) of the PMF runs showed significant structure in the residuals (Q maximum value was 15 during fireworks period as shown in Fig. S3) for solutions having up to seven factors. Increasing the number of factors to eight gave evidence of structure removal, with mostly random errors remaining, by another fireworks-II factor which was explained by K, S, Ba, Ti, Cu, Bi, while a further increase led to a new mixed factor of traffic-related and background dust which however had a noisy diurnal pattern (Fig. S3). All the variables were approximately unimodal scaled residuals between ±3 (Paatero and Hopke, 2003) (Fig. S5).

Pag 6, line 13: "obvious structure": completely obscure what the authors mean.

We meant significant structure in residuals. We modified the text on Page 6 line 13 as follows:

Residual analysis (Q-contribution over the time series) of the PMF runs showed significant structure in the residuals ((Q maximum value was 15 during fireworks period as shown in Fig. S3) for solutions having up to seven factors.

Pag 6, line 28: "both the sea salt and secondary sulfate factor time series were also constrained with a-value 0.01". Further constraint implemented. Please note that a small a-value was used. Is it consistent with uncertainty estimates for the non-firework sub-set and for the sulfate factor identified in the preliminary 9-factor solution? Please also note that the constraints come from two different analyses.

We understand the concern of the reviewer#3. The selected *a*-values are not intended to reflect the uncertainties y in the factor time series of secondary sulfate and sea salt during the fireworks period; we now assess these using a different method, as discussed below. Rather, the selected values are empirically chosen to provide a clean separation of these factors from the fireworks emissions. We performed a sensitivity analysis on the small *a*-value from 0 to 0.1 over partial (fireworks period only) time series of secondary sulfate and sea salt. The structure during the fireworks period in the sea salt and the secondary sulfate factor time series was mixing with the fireworks factor time series for *a*-values > 0.01.

We have now added the following text on page 6 line 28:

The small *a*-value (0.01) for the sea salt and the secondary sulfate factor time series (for the fireworks period only) were estimated based on sensitivity analyses on the *a*-value from 0 to 0.1 with an increment of 0.01. The time series of both factors were showing fireworks peaks during the fireworks period for *a*-values greater than 0.01.

In the original manuscript, section 3.3 presented an analysis of the uncertainty of the time series across the entire campaign. However, this analysis did not account for the tight constraints applied to the sea salt and secondary sulfate factors during the fireworks period, and as a result these uncertainties were underestimated. We have now added the following text (page 7 line 12), which describes the uncertainty estimates of these factors during the fireworks period, and updated Fig. 3a accordingly.

During the non-fireworks period, uncertainties in the source apportionment results are assessed by a bootstrap analysis as described above. However, this approach cannot be used to assess uncertainties in the sea salt and secondary sulfate factors during the fireworks period, as during this period these factor time series are constrained with an artificially low *a*-value selected to optimize deconvolution. For these two factors, uncertainties during the fireworks period are determined by our ability to accurately predict the factor time series. The secondary sulfate and sea salt factors cases are discussed separately below.

Secondary sulfate concentrations during the fireworks period were estimated from the linear fit of the secondary sulfate factor to ACSM sulfate during the entire non-fireworks period. Uncertainties of ±5 % were calculated as the standard deviation of the actual secondary sulfate concentrations to the predicted values and included for the fireworks period only in Fig. 3a.

As described above, the sea salt factor time series during the four-day fireworks period was investigated to determine measurements that were affected or not affected by fireworks, where the measurements determined to be affected were replaced with a linear interpolation between the nearest good points. To determine the uncertainties of this approach, we applied this calculation to random segments of the non-fireworks data. Specifically, the four day-long sequence of affected/non-affected time points determined during the fireworks period was applied to a randomly chosen segment of data, and the standard deviation of measurement data to the estimated values calculated by interpolation was determined. This analysis was repeated for 38 randomly selected locations through the non-fireworks data, and a mean standard deviation of ±42 % was determined. This value is used as the uncertainty of the sea salt factor time series (during the fireworks period only) in Fig. 3a.

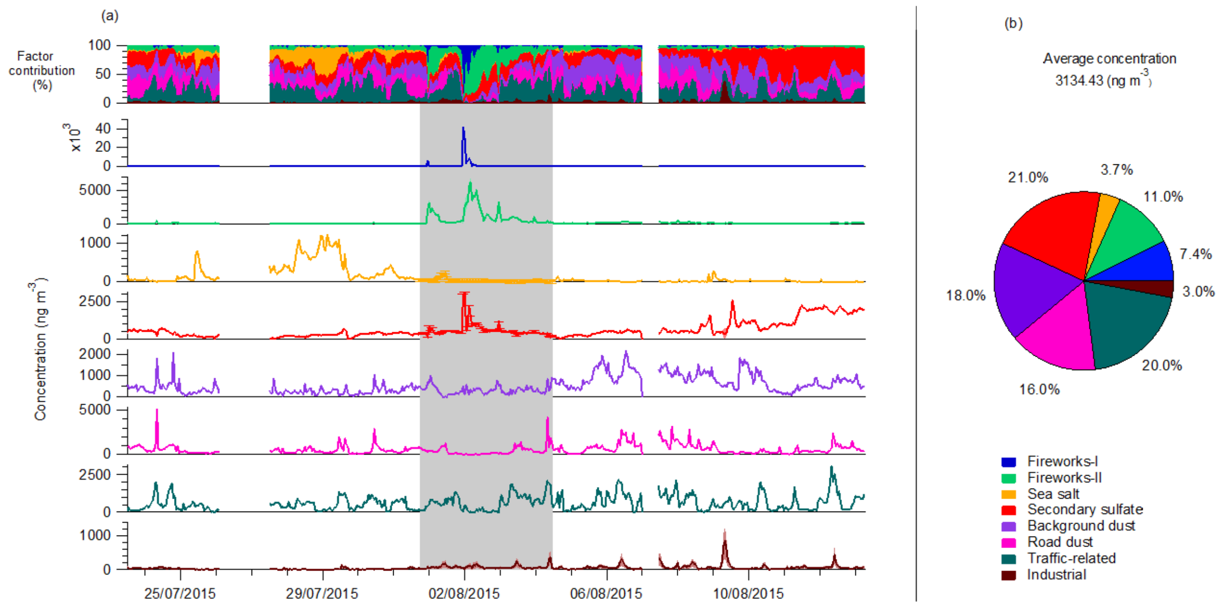

Figure 3: (a) Time series of the PM_{10} elemental sources and relative contributions of the different sources over time; shaded areas indicate the uncertainties (interquartile) of selected bootstrap runs; grey background color represent the fireworks period; estimated uncertainty of the secondary sulfate (± 5%) and the sea salt factors (± 42%) during fireworks period are added as error bars; (b) Mean relative contributions of PM_{10} elemental sources.

Page 7, line 19: obvious. It was checked in the preliminary analysis at 9-factors and then constrained both as far profile and temporal patterns are concerned in the final solution.

This factor (secondary sulfate) was identified in the unconstrained PMF solution at a specific seed where it was mostly dominated by S and had a high correlation with ACSM sulfate. However, the high and variable S concentrations during the fireworks period led to mixing between the fireworks and the secondary sulfate factors during this period. Therefore we constrained part of the secondary sulfate time series (only during fireworks days) to avoid mixing along with factor profile.

Page 8, "Fireworks" paragraph. Considering that the fireworks profile was constrained, it sounds very strange that two fireworks sources were identified in the final analysis. As previously required, the existence of two fireworks sources should be supported by their identification at least in the unconstrained analysis of the fireworks period. If not, the strength of the imposed constrain should be verified to get if it artificially generates the presence of two factors.

Based on the suggested changes due to the previous comments, we believe that we have responded to this concern as well. We have already discussed the residuals structure in Fig. S3 from 4 to 10 factors solution. In the final solution, the secondary sulfate, sea salt and fireworks-I factor profiles were constrained along with the factor time series (for the fireworks period only) of sea salt and secondary sulfate. Because of significant scaled residuals in the 7-factor solution, we found that fireworks related elements such as K, Ba, Ti, Cu, Bi had significant residuals during fireworks days (scaled residual over time: ±8) which is consistent with Fig. S8. Both analyses (residual and Fig. S8) indicate that the fireworks composition was highly variable and not well explained by a single factor.

We identified a K-rich factor in both the fireworks PMF and in the non-fireworks PMF analyses which were dominated by these elements along with other elements (Si, S, Fe, Mn, Zn, Pb). Although the second fireworks factor profile yielded a slightly higher K / S ratio (3.56) than the black powder ratio (2.76), we decided to keep it based on the improved residual structure and the factor time series which captured the fireworks variability.

Page 9, lines 1-4 and Figure 4. Completely obscure. I interpret "normalized" as "divided by", but in this case how can the negative values be justifiable? Furthermore, what is "composition"? Average contribution of the factor to each element?

The similar issue was also raised by Reviewer #1, and our response is presented in both places for clarity. The PMF results (Fig. 2) describe potentially complex and time-dependent sources as a single factor (or linear combination of factors). This is a fundamental limitation of the model, and may not accurately describe the behaviour or a complex source (e.g., residuals may be significant). In contrast, Fig. 4 provides an estimate of overall fireworks composition and temporal variability, complementing the PMF results (which are shown for reference). One can then see both the explanatory power and limitations of the SA model for this source.
We moved Fig. 4 to the supplement as Fig. S8 and removed ± SD from the figure as suggested by reviewer#1.
We added in the supplement on page 8 line 4:
Fig. S8 provides an estimate of the overall fireworks composition and temporal variability, complementing the PMF results (which are shown for reference). The figure is constructed in two stages. First, the time series of fireworks contributions to each element is estimated by subtracting the non-fireworks factors (NFF) from the original measurements. Then, the estimated fireworks contribution for each element is normalized by the total fireworks element contribution, and the displayed statistics are calculated. This is represented mathematically below, and the expression has been added to the main text. Note that the variation in fireworks profiles implied by this figure supports the representation of fireworks by 2 fireworks factors.

$$normalized\ concentration_{ij} = \frac{x_{ij} - (g_{ik}f_{kj})_{k=NFF}}{\Sigma_j\left(x_{ij} - (g_{ik}f_{kj})_{k=NFF}\right)}$$

(S1)
where X represents the input data matrix for PMF, while G and F represent the factor time series and factor profiles driven by six non-fireworks factors (NFF). Here *i* and *j* denote time series and variables, respectively.
NFF and fireworks measurements have some uncertainties, and therefore for elements and/or time periods in which the concentration of fireworks is low, positive and negative values fluctuating around zero are expected. Here the negative values occur mostly for Si (for which fireworks are a minor fraction of the total signal), while the other elements were mostly captured very well. We used the factor composition for the factor profiles (fingerprints). We have already added the mathematical expressions for the factor composition as well as for the factor relative contribution on Page 7, lines 26-27.

Page 11, lines 22-23: "We established that data sets including extreme events such as fireworks can be apportioned by ME-2 without disturbing the model solutions". Untrue. The imposed constraints completely modified the output of the unconstrained analysis.

We have discussed the unconstrained nine-factor solution which was basically mixed with the fireworks in the factor time series during fireworks period. To avoid mixing, we used the ME-2 method by restricting the fireworks structure in the S and Cl driven factors time series.

We have modified the text as follows:

We show that the rotational control available in ME-2 provides a means for treating extreme events such as fireworks within a PMF analysis.

Minor comments

Page 2, Line 12: A reference to: "Fe in brake lining can reach up to 60 % by weight" is needed.

Done. (Chan and Stachowiak, 2004; Schauer et al., 2006)

Chan, D. and Stachowiak, G. W.: Review of automotive brake friction materials, Proc Inst. Mech. Eng. Part D: J Automob. Eng., 218, 953–966, https://doi.org/10.1243/0954407041856773, 2004.

Schauer, J. J., Lough, G. C., Shafer, M. M, Christensen, W. C., Arndt, M. F., DeMinter, J. T., and Park, J. S.: Characterization of emissions of metals emitted from motor vehicles, Research report (Health Effects Institute), 133, 1–76; discussion 77, 2006.

Page 2, line 27: add "among others" after "Wang et al., 2018". Indeed, the list is far from being complete (see e.g.: Li et al. 2017 http://dx.doi.org/10.1016/j.jenvman.2017.02.059; Zhou et al., 2018 https://doi.org/10.5194/acp-18-2049-2018 among the most recent). If the authors intend providing a full list, a much more detailed research has to be done.

Done.

Page 3, lines 10-12. "The later, being essential in particular when separating extreme events such as fireworks which are most often excluded from the PMF input matrix (Ducret-Stich et al., 2013; Norris et al., 2014) to avoid distortion in the PMF solution due to unusually high emissions". This sentence is questionable. As reported by Paatero et al., (doi:10.5194/amt-7-781-2014) wrong decision on the outlier status of data can introduce serious modelling errors. Please also note that opposite to what stated by the authors, examples in the literature tried to exploit fireworks tracers to quantify the source contribution, with different approaches (e.g. Scerri et al., 2018 https://doi.org/10.1016/j.chemosphere.2018.07.104, Ji et al., 2018 https://doi.org/10.1016/j.scitotenv.2018.01.304, Vecchi et al., 2008 http://dx.doi.org/10.1016/j.atmosenv.2007.10.047)

We modified the text in the manuscript as follows:

The rotational control available in ME-2 provides a means for treating extreme events such as fireworks within a PMF analysis. Such events are often excluded from the PMF input matrix to avoid modelling errors due to the pulling of a solution by outliers (Ducret-Stich et al., 2013; Norris et al., 2014; Paatero et al., 2014).

Regarding the papers mentioned by the reviewer, it was shown that fireworks are not necessarily outliers as considered in the PMF analysis. However, these articles have discussed different methods to apportion fireworks contribution in the PMF analysis e.g. Scerri et al. (2018) has down weighted some of the elements (not specific to fireworks-related elements) as "weak" variables without discussing the influence of fireworks on the other non-fireworks factors. The down weighting approach is not clear in this paper due to lack of criteria defining down weighting random elements. In contrast Vecchi et al. (2018) reported the up weighting of fireworks tracer by a factor of 2 to highlight its role during fireworks. At the same time, they down weighted some of the variables by the factor of 2 to 4 with trial and error method until the model resolved fireworks sources. This approach may have worked to resolve fireworks contribution but no discussion has been provided for other non-fireworks sources. They discussed the other sources in a separate paper (Bernardoni et al., 2011) with a slightly different PMF input set on the same data by excluding fireworks-related tracers. Ji et al. (2018) quantified fireworks factor on the basis of signal to noise ratio instead of up/down weighing elements specific to fireworks which yielded the mixed factors with fireworks (in terms of factor profile and time series) in their PMF analysis. This mixing issue can be resolved by using ME-2 approach as adopted in our study.

Bernardoni, V., Vecchi, R., Valli, G., Piazzalunga, A., and Fermo, P.: PM_{10} source apportionment in Milan (Italy) using time-resolved data, Sci. Total Environ., 409, 4788–4795, https://doi.org/10.1016/j.scitotenv.2011.07.048, 2011.

Ji, D., Cui, Y., Li, L., He, J., Wang, L., Zhang, H., Wang, W., Zhou, L., Maenhaut, W., Wen, T., and Wang, Y.: Characterization and source identification of fine particulate matter in urban Beijing during the 2015 Spring Festival, Sci. Total Environ., 430–440, 628–629, https://doi.org/10.1016/j.scitotenv.2018.01.304, 2018.

Scerri, M. M., Kandler, k., Weinbruch, S., Yubero, E., Galindo, N., Prati, P., Caponi, L., and Massabò, D.: Estimation of the contributions of the sources driving $PM_{2.5}$ levels in a Central Mediterranean coastal town, Chemosphere, 211 465–481, https://doi.org/10.1016/j.chemosphere.2018.07.104, 2018.

Vecchi, R., Bernardoni, V., Cricchio, D., D'Alessandro, A., Fermo, P., Lucarelli, F., Nava, S., Piazzalunga, A., and Valli, G.: The impact of fireworks on airborne particles, Atmos. Environ., 42, 1121–1132, https://doi.org/10.1016/j.atmosenv.2007.10.047, 2008.

Page 7, line 28: "absolute mass". It should be recalled that it refers only to the mass related to elements, as no PM mass was inserted in the analysis.

We have rewritten it as follows:
Fig. 3a shows the time series of the factors contributions in ng m^{-3} (bottom panels) and relative contributions (top panel) of the retrieved PM_{10} factors.

Page 9, line 25-26: "The two dust factors together explain 95 % of Ca, with no other factor explaining more than 93 %". Obscure. If already explained at 95%, how can other factors explain Ca for more than 93%?

We apologize for the misunderstanding. For clarity, we have rewritten it as:
The two dust factors together explain 95 % of Ca, while the remaining factors explain only 5% of Ca.

Page 9, lines 29-30: "In general, Ca is commonly associated with mineral dust, construction activities, vehicular emissions and iron/steel plants". References are needed.

Done.

(Lee and Pacyna, 1999; Vega et al., 2001; Bukowiecki et al., 2010; Crilley et al., 2016; Maenhaut, 2017)

Lee, D. S. and Pacyna, J. M.: An industrial emissions inventory of calcium for Europe, Atmos. Environ. 33, 1687–1697, https://doi.org/10.1016/S1352-2310(98)00286-6, 1999.

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