Response to reviewer comment (Rev. 3):

The revised paper contains changes according to comments of all of the three reviewers. The abstract was revised according to the remarks of the three reviewers. As recommended, single paragraphs were shifted to new positions for better understanding. The entire introduction was restructured. In section 2 the discussion on weather conditions in Beijing was extended, specifications for stopping conditions of the NMF algorithm were included, the role of NMF factors and weights was clarified and the subsection for relevance of the NMF factors was improved. In Section 3 interpretation of the resulting pattern was improved and extended regarding weather conditions and NMF weights. The subsections in chapter 3 were combined to form at least two subsections in the revised paper.

Figures and Tables: We changed the content of the figures as follows. Figure 1 was replaced by the temporal mean, which is considered useful for comparison with the NMF factors and their patterns. Since dust storms do not play a role anymore, when shifting the NMF volume application to a separate study, the former graphs of 2 exemplary days with and without dust storm influence are removed. Figure 2 is new, comprising information about wind direction and speed in the study period. The former figure 2 becomes now figure 3. The former figure 3 becomes figure 4 (recomputed because of the use of mean instead of median values). The former figure 4 becomes figure 5, where time series of the NMF factors have been added for all 5 factors. The new figure 6 is the upper part of the former version. Since volume NMF results are omitted, the lower part does not play a role. Figure captions have been adapted and the content of Table 1 was changed according to the changes in the manuscript.

The authors thank the reviewer for its helpful comments to improve the manuscript.

In the following we give answers to the comments by reviewer 3:

Rev. 3:
However, the authors emphasize the methodology (i.e., NMF) over a more in depth analysis of the data.

This is our intention, since we want to introduce this new methodology and explain its benefits by the observed results. In other words, there are two main goals: (1) introduce the method and (2) interpret the results for the study in Beijing during 2004-2008.

It is not especially useful to "discover" well-documented associations between number and volume size distributions and sources, as determined by actual source tests, several of which are cited in the paper.

Since the method is a new way to obtain size-dependent patterns of PM, we have to support our results by findings of recent studies with traditional approaches to verify our results. Otherwise, the interpretation of the patterns was based on the patterns, their weights and meteorological variables.

This reviewer believes that different data processing methodologies could have been more helpful for interpreting this high temporal resolution data set which covers number and volume size distributions over a 4 year period at only one monitoring site. Is it possible that sources in a given sampling site have changed from year to year within these 4 years? The weekly and seasonal variations in PM

sources are identifiable, but unless there were unusual circumstances, I don't see how source in general can change drastically from year to year in a 4 year period. Basically other than episodic events like wildfires, etc., the yearly variations in PM sources at a given site must be negligible. Thus analysis of the data from 1 year of this study could yield similar results.

We do not share the view of the reviewer. PSD data have been available for the 5-year period 2004-2008 comprising a lot of missings. Therefore we intended to make use of all available data sets to obtain a large data base. Why should we reduce the data base? Moreover, if there are unusual weather conditions in a certain year (or a season) the data of this year are not representative for a typical particle burden during the whole year. Using the period 2004-2008 provides a mean of the conditions in these years, which is more representative.

On a separate note, authors claim in the abstract (P. 12017, L. 8) that the number concentrations have been measured in high time- and space-resolution; while the data is obtained at a single site, so why do they claim that this is also a high space resolution data set?

Thank you for this advice. This was a misleading statement. The term "space" should refer to the particle size (data were available in the size range 3nm to 10µm). We corrected this irritating phrase.

NMF would have been most useful if there were more than one sampling site in a one year period of time. Ideally these sampling sites must have been chosen to represent different areas of the city with different PM sources (i.e. one site affect by local traffic, one relatively out of city to reflect background PM, one in agricultural areas to reflect increasing effect of windblown dust, etc.).

This was not the basis for our study. If data had been available for different monitoring sites and in the same time- and diameter-resolution, we would have used it in an appropriate study.

The data could have been interpreted without using NMF, and by comparing the diurnal variations of particle number and volume size distributions in different seasons, and on days with unusual events.

Of course, but our aim was to get another access to the topic of source identification and therefore we applied NMF on daily PSD data. It is not a simple case study, but an introduction in "using NMF for the purpose of source apportionment", that necessarily includes an interpretation of the results.

Using NMF, the authors' interpretation of the factors is purely speculative. The absence of meteorology data is one of the major caveats of this study.

Interpretation of the obtained patterns is difficult. We used NMF factors and NMF weights for this interpretation. Of course we also made use of meteorological data (presented in section 2.2) and included relations between patterns and meteorological variables in the interpretation process.
In the revised paper we improved this discussion including correlations between temporal NMF weights and meteorological parameters. We added a figure for wind conditions during the study period.

The authors talk about variations in traffic density and changing car fleet, without using this information in interpretation of NMF results. If such data was available, it would have been significantly useful for identifying vehicular sources, based on diurnal variations in traffic density (and changing car fleet as the authors mention in the abstract) and comparing it to the diurnal variations in number and volume size distributions.

Thanks for this advice. We included information on traffic in the introduction and the discussion sections.

The discussions are too general, and more emphasize is on methodology than the quantitative results. The authors should at least present some specific PM concentration averages, and highlight some of seasonal variations in number and volume concentrations. For example, more discussion of the highest PM concentrations (for each size fraction) would be useful. The dates on which these occurred, wind speeds/directions, fractions of size fractioned concentrations to total PM concentrations, etc. would provide far more insight into the causes of these high levels that is currently available from NMF.

This is not the intention of the paper. We intended to introduce a new method to the field of PSD data analysis and source apportionment and explain the results in an application to the Beijing data set. PM concentration averages and variations could be addressed in another study.

Moreover using NMF, has resulted in 'mixed factors', also pointed out by the other reviewer, where the first factor, NMF-N1, is affected by three different sources. If the authors want to use this method, it is advisable to do the analyses for each season separately. This way these mixed factors may appear differently in each season, enabling authors to gain more insight in the possible sources of their observations.

Thank you for this advice. We also applied the method to summer and winter seasons separately, but in the results the first factor is still a mixed factor of some sources. There only slight differences to the patterns of the whole year. We analyzed seasonal variations in terms of the mean NMF weights per season.

Most of the discussion focused on the methodology can be moved to supplemental material in order to include more comprehensive discussion on the single events, presenting quantitative data and discuss in more detail the seasonal variations is number and volume size distributions.

We want to introduce and describe the method in application to PSD data pointing out some results. We do not focus on particular PM features.