

Response to reviewer comment (Rev. 1):

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 useful and reasonable comments to improve the manuscript. We followed the advices of the reviewer to change the structure of the introduction adding a discussion on primary/secondary particle processes and reducing the paragraph regarding health effects of particulate matter. As recommended, we changed our strategy regarding the analysis of particle volume patterns and decided to omit this part completely, preserving it for an additional paper.

In the following we give answers to the comments by reviewer 1:

General Comments:

1)

Rev.1: The introduction section which is in my opinion misleading concerning the motivation of the concept and misleading about the usability of the outputs. The key note and how you structure your article is missing. The nature of secondary particles and related processes such as nucleation and condensation which play a key role in this article isn't addressed at all. The benefits and further usage of this approach must be illustrated in a better way, they must become clearer. In contrast, a lot of information is within the introduction given, which is not addressed later in the article. A major fraction of the introduction is dedicated to health related aerosol characteristics (composition & size) and health related articles. However, later in the article no reference is made to all this information. I would recommend that authors either amend 3.1 and conclusions or shorten these parts of the introduction.

Answer: We revised and restructured the entire introduction section. Health aspects were reduced so that misunderstandings about the motivation of the paper are prevented. Instead the discussion on primary/secondary particles and related processes was extended including references. We hope that the intention and usability of the study results is now outlined more clearly.

We omitted a sentence on the structure of this article, since in our point of view it seems not necessary in a research paper.

2)

Rev.1: The second one are the results presented in Figures 4c and 4d, but also Figures 4e and 4f. One might expect cyclic behavior at the time of the day 0:00 and 24:00. Please clarify or give reasoning why these patterns are different at 0:00 and 24:00!

Answer: We included a paragraph in section 2.3.2 to give a short explanation for the behavior of the NMF patterns. Please refer also to the response we gave earlier on ACPD. Additionally, we added time series of the NMF weights to the figure with NMF patterns (now Fig. 5) to underline the fact that NMF factors and NMF weights have to be considered together.

3)

Rev.1: The presentation of results in 3.2 Particle volumes NMF is neither fish nor fowl. I looked at the information provided in the supplement and I think these patterns may be interpreted / analysed as well. However there is possible the same ambiguity as already stated in 2). On the other hand the article is already far too long, so I would recommend either to omit 3.2 and related results or to write a supplementing companion article.

Answer: We followed the recommendation to revise the particle volume section and decided to remove this section from the manuscript, because otherwise it would be too long. Instead it is intended to address NMF on particle volume data in another paper, where the interesting outcomes will be explained and interpreted as well. Thank you for this helpful advice.

Specific Comments:

Rev. 1: P13017 I1: Changing car fleet... Why? Please give reasoning (later in text) or delete. NB: Higher emission standards may improve a changing car fleet).

Answer: We included discussion on the development of traffic volume and emissions in Beijing in the introduction.

The introduction is in some parts too comprehensive and lacks clarity. In particular the main objectives and the motivation and use of this study remain vague or even misleading. On health effect related research many words are spent. However later in article there is no real connection with health relevance to PM composition, or PM size ranges or particle burden.

See above, general comments 1).

Authors state initially that "In order to derive source-dependent measures for reduction of particle concentrations it is necessary to gain insight into the exact composition of the particle burden. " In order to have efficient measures a good understanding of sources, their health relevance and subsequent transport & transformation of pollutants is important. However, the method presented does neither provide exact composition nor any air quality standard related information. Air quality standards are defined in mass; however PM2.5 or PM10 do not provide information about constituents. In that context it would very interesting how the particle volume presented in Fig. 6c relates roughly to PM10 measurements (levels would be interesting, there are many standard air

quality monitoring stations in Beijing). I would rather suggest motivating this work with “process understanding” i.e. getting further insight into the complex processes in Beijing air, which is important with regard to the interpretation of measurement results, aerosol characteristics and eventually subsequently health assessment. Hence the introduction should become far more specific towards process understanding or should provide a better overview about what is important and where you will discuss important issue in your article.

and

P 13018 l12-l16: Here authors may motivate their work. In the article no information is given in which way these results may be used in the context of “measures”, see also above.

Further: The aim of this study is not to present an exact and complete composition of Beijing’s particle pollution. We rather aimed at initiating a new method that differs from previous approaches and helps to find a classification of patterns and their association to sources for daily data sets. Secondly, we intended to extract the main patterns and sources for the Beijing data set, interpreting the results by means of meteorological and physical conditions as well as known patterns from the literature. We revised the paragraphs motivating our work to clarify these goals.

P13019 l9 – l24 lacks structure and contains some misplaced or not related statements (e.g. l9-l10 or l13 are meaningless). The attempt to provide an abstract about particle origin in number and mass or volume, and composition, primary, secondary, anthropogenic and natural origin needs a clear structure. I would suggest distinguishing between main number sources and main volume/mass sources and what is known about these particle classifications or concepts in Beijing. Finally: Consequently, mode of action and hazardousness of primary particles... are predetermined by their origin. Concerning number, nucleation is an important process; however the health relevance is yet unclear, please check with your co-authors, they are experts in this field. Concerning mass/volume condensation is as well an important process in particular for the fine particle range. However the role of secondary particles and related processes and maybe their health relevance is not described. However later in the article they play a prominent role.

We rewrote the paragraph and extended discussion on possible primary/secondary particle processes in order to provide a basis for further discussion on the resulting patterns.

P13019 l19-l21 ... *influencing air pollutant concentrations* ... appears incomplete in which way, which impact, e.g. low wind speeds?

Deleted and replaced by “The basin-like topography of Beijing’s urban area reinforces temperature inversions in the lower troposphere, occurring frequently in the winter months during low wind weather situations. As a result, emitted particles are trapped in the basin and accumulate over time.”

P13019 l24 - P13020 l1. Unclear, message? The statement l26-l28 appears as a trivial statement. This needs certainly specification, please revise. I doubt that industrial emissions are similar within the “whole area”. They may be distributed quite uniformly in certain districts. The only concept to assess spatial variations which may work is to have representative stations close to different main sources but also different “background” monitoring stations, supplemented by a smart modelling approach. The spatial variation (inclusive temporal) may be assessed by a combined approach by monitoring and modeling. However all models have their limitations or deficiencies and rely on the availability of proper input data such as meteo, topographical data (terrain, land use, buildings), emissions (spatially and temporally resolved) ... Please specify or maybe omit this information.

Omitted, since it is not crucial for the study.

P13020 I5 please specify “receptor model” (for what? & in which way?) – or is it a statistical concept to be used in source receptor modeling applications, probably based on: A non-negative factor model with optimal utilization of error estimates of data values, based on P. Paatero, U. Tapper (1994).? Consequently I6: The concept is used for characterizing aerosol sources by calculating dominant positive factors on the basis of ...

and

P13020 I8 – I15: May need specification. What is finally the effect on analysing PSD data or particle composition?

The paragraph is changed for better clarity.

P13020 I16 and subsequent lines: the use of NMF is not very well motivated. Bring forward the information provided in 2.3 NMF P13023 I23-I27 and move the specific information towards 2.3 NMF.

After describing the traditional PMF approach we give a short explanation of NMF, leaving details for the methods section.

P13022 I6-10: nice listing about the characteristics. However, the important information about prevailing wind systems and wind speed is missing!

We added brief information on the wind systems and wind speed/direction during the study period, underlined by a new figure (now Fig. 2).

P13023 I23-I27: bring forward this information - see last comment at introduction.

We improved the paragraph in the introduction, but preserved detailed information for NMF for section 2.3.

P13029: Table 1: I wonder why there is no internal combustion engine / district heating related pattern indicating a clear soot mode in the size range of 40 nm – 150 nm. The optimum number of 5 may points to limits of this approach. However, this might be related also to a different car fleet compared to Europe , rapid changes in the fleet during 2004 – 2008 and possibly as well on traffic bans for HDV vehicles?

and

P13029: I16-I17 please rewrite It provides a background pattern ... sounds strange, I guess the “urban background” is meant.

The number of NMF factors was chosen as 5, because in this case the best physical interpretability of the patterns was obtained. For more factors, the patterns become too local and an assignment to sources is more difficult. Unfortunately, some processes are mixed together in the r=5 case. We extended our interpretation of the patterns and their related sources. Heating was assigned to factor 1, which is interpreted as urban background containing emissions from fuel combustion and construction sites. Soot is contained in pattern 1 and pattern 2 (related to traffic).

P13030 NMF-N2 interpretation / analysis Fig 4c: why is the maximum in the late afternoon until midnight?

We addressed this issue in the revised section on NMF-2, supported by a study of Kai et al. (2007). The maximum of the particle concentrations is achieved in the late afternoon due to rush hour emissions, meteorological conditions and secondary particle processes.

P13030 I25-30: this is highly unclear. The role of daily NMF weights and resulting effects must be explainer earlier. Please clarify or provide appropriate references within your article.

Additional information regarding the interpretation of both NMF factors and corresponding weights is added in the section 2.3.2.

P13031: line 14 – I21: Highly confusing, imprecise and part wise too specific. More specifically: Afterwards, with increasing solar radiation fostering what? I doubt that particles grow up to that extent by coagulation, I guess a major process is as well or mainly condensation! The Beijing vehicle fleet from 2004 – 2008 provided certainly an enormous amount of pre-cursor gases such as NMVOC, SO₂, NO_x, H₂O ... then there are certainly district heating units located in the vicinity of the monitoring station providing NMVOC, & SO₂. Processes important for NMF-N4 and NMF-N5 are highly complex and there might be several paths and interactions. Moreover, there are numerous studies about the interaction of atmospheric processes, particle properties such as surface area and pre-cursor gases. Therefore it is important to discuss very briefly the main processes leading to secondary formation (including SOA formation) already within the introduction or here in this section. I think the basic concepts should be mentioned providing references. It will be not possible to go give too much detail about the concepts. You may find some valuable information like an overview of processes and a study of different aerosol dynamical processes in interaction with atmospheric processes e.g. in U. Uhrner et al., Particle formation at a continental background site: comparison of model results with observations, Atmos. Chem. Phys., 3, 347-359, 2003. There in section 3, two basic general mechanism are given. F. Stratmann, et al. New-particle formation events in a continental boundary layer: first results from the SATURN experiment; Atmos. Chem. Phys., 3, 1445-1459, 2003, studied in detail the impact of atmospheric mixing process on new particle formation and there are several studies from Nilsson, E. D. which are related to new particle formation and atmospheric processes.

See above remarks on structure of the paper and overview of primary/secondary particle processes. We changed the paragraphs for NMF4 and NMF-5 and concerned these issues in the introduction (as recommended). Secondary particle processes were discussed briefly and additional references were provided. The influence of traffic was analyzed in the interpretation of factor 2.

3.2 Particle volume NMF-V

See general comment. There might be other processes relevant but not mentioned here. P13032 I23-I25 ... several NMF-V factors obviously present patterns associated with this phenomenon at different times of the day. This is a confusing statement with an unclear message! Do you want to point to possible deficiencies of your methodology? Or is the problem that dust storms cannot provide a distinct pattern since they occur occasionally?

The section is deleted from the manuscript.

P13034 I27-I28, P13035 I1-I12: see above suggestions to improve introduction needs better structure, please revise. Sulphuric acid and water are certainly key species triggering nucleation, however there might be different mechanisms e.g., organics might provide as well formations paths. Cold air breaks in winter related to the Siberian anticyclone may be as well important (Nilsson, E. D.). Concerning growth: SOA formation is certainly a major process to explain particle growth in summer!

We followed the advices and changed the introduction section as well as the interpretation of the NMF factors. The section for categorization of the NMF patterns was included in the direct interpretations of the factors (3.1).

P13035 I13-I14 confusing sentence please revise. The inferred impact of industrial emissions and subsequent transport from industries located southward of the monitoring site ...

Sentence was removed.

P13037, l11-l16: where are the results illustrated to support these findings?

We deleted these remarks from the conclusions section and changed the according remarks in the section for NMF initialization 2.3.1. We did not include further NMF results, because this would blow up the paper. We decided not to provide additional information in the supplementary material, because NMF results (found by the two other initialization strategies) were extremely close to the results of NNDSVD and would thus not provide additional information. NNDSVD fastened the NMF algorithm convergence and is generally more reasonable. A figure on the algorithm convergence would not provide any new information and blow up the paper.

Minor Comments

Changes according to all of the minor comments have been applied in the revised manuscript except for these cases, where the corresponding phrases were removed or replaced.