

GENERAL COMMENTS:

First of all I have to say that I do not have professional experience with methods like principal components analysis or more specifically non-negative matrix factorization. I hope the other reviewer is an expert in this field and can evaluate pages 13024 – 13029. I have communicated this to the Editors in my reply to the invitation.

This study covers the full range of complex aerosol dynamics including complex atmospheric processes and authors endeavour to interpret their results in detail. Sometimes these efforts remain vague or too general or may provide even too many details. An interesting concept and interesting results are presented in this study.

In order to approve submission there are from my point three main issues / concerns which need clarification:

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.
2. The second one are the results presented in Figures 4c and 4d, but also Figures 4e and 4f. One might expect cyclic behaviour 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!
3. 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.

In order readers can grasp your results easier, I would recommend that you provide in Figures 3, 4, 5 and 6, information about the "physical" meaning of the NMFs. At least please refer to Table 1.

The English needs to be improved.

Given the length and details of this article there are many bits and bobs, see below.

SPECIFIC COMMENTS:

Abstract

P13017 l1: *Changing car fleet...* Why? Please give reasoning (later in text) or delete. NB: Higher emission standards may improve a changing car fleet).

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. 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.

P 13018 I12-I16: 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.

P13019 I8 *Reviews are given in ...* -which reviews, about what? Please state precisely.

P13019 I9 – I24 lacks structure and contains some misplaced or not related statements (e.g. I9-I10 or I13 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.

P13019 I19-I21 *... influencing air pollutant concentrations ...* appears incomplete in which way, which impact, e.g. low wind speeds?

P13019 I24 - P13020 I1. Unclear, message? The statement I26-I28 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 modelling. 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.

P13020 I5 please specify “*receptor model*” (for what? & in which way?) – or is it a statistical concept to be used in source receptor modelling 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 ...

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

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.

2.2 Weather and climate of Beijing

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

2.3 NMF

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

3. Results and discussion

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?

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

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

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.

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.

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?

3.4 Categorization of NMF patterns

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!

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

4. Conclusions

P13037, I11-I16: where are the results illustrated to support these findings?

MINOR COMMENTS

Abstract minor

L5 Source group-related → source related

Full diameter range [...] → in the diameter range

Introduction

P 13018 I22: ... *associated to* ... → ... associated with ...?

P 13018 I23: ... *chemical compositions hazardousness for human health may differ.* ?

P 13019 I3 ... *penetrate more deeply into the lungs.* I would suggest stating this more precisely, in particular to motivate the role of UFP concerning health relevance in contrast to PM2.5 in a better way. The following sentence is too specific. I am not a health expert, but to my knowledge both PM2.5 & UFPs enter the alveoli, (the deposition efficiency of UFPs is a bit higher, according to Heyder). However the UFPs enter the next “barrier” they assess the tissues, hence able to enter the blood circulation.

P13019 I21-I23: Do dust storm contribute to particle pollution or maybe better the particle burden? It might be interesting to provide information about the PM levels encountered associated with dust storms. The storm may dilute harmful substances.

P 13019 I6-I7 ... these particles may contribute to the observed health effects largely (). – Which health effects? & what means “largely”?

P13019 I19-I21 ... *influencing air pollutant concentrations* ... in which way?

P13020 I9 please specify sub factor **profiles** and ...

P13020 I20 ... *traditional* ... what is traditional? References missing?

P13020 I29 *as a pattern recognition tool to the field of* - isn't it: ... **in** the field of ...(NB: I am not a native English speaker)

2. Material and methods

P13021 I22-25: transformation process is described not very clear. Only specialists who know the instrument characteristics can understand what is going on.

P13022 I12: *After all the restrictions in total* ... → (Finally,) in total ... ?

2.2 Weather and climate of Beijing

P13023 l1-5 change to: Temperature inversions (with related low mixing heights) may occur. Emitted particles are practically trapped beneath the inversion layer and accumulate over time resulting in high particle number and mass concentrations.

3. Results and discussion

P13029: l16 *Permanent emissions* → Continuous emissions

P13030: l20 ... *turns upside down* ... At early noon this process is reversed?

P13033 l5: By means of ...?

4. Conclusions

P13036 l19 With the aid of ... ?

P13038 l1-l5 Please revise! Sentence is too long and not clear. For the data sampled during period of the Olympic ... it is intended to use / apply ...NMF factors to assess ... These measures were introduced ...