

Review of Lin et al., “Rapid mass growth and enhanced light extinction of atmospheric aerosols during the heating season haze episodes in Beijing revealed by aerosol-chemistry-radiation-boundary layer interaction”.

This paper describes measurements of aerosol physical and chemical properties that were collected over a five month period in Beijing city. These data are combined with boundary layer height measurements, and calculated aerosol scattering efficiency to better understand the aerosol-radiation-boundary feedback relationship. The authors focus on a case study period of three days in February. The results illustrate that as boundary layer decreases and as pollution conditions develop, the contribution of nitrate containing particles increase dramatically. It is suggested that these larger ammonium nitrate particles contribute most towards the reduction in visibility during haze events and also slow the development of the mixing layer height. The low mixing layer height together with the increased concentration of particles is thought to favor the formation of secondary nitrate, sulfate and organic aerosols, and hence leads to marked increases in aerosol particle concentrations. This manuscript presents an interesting data set, although it feels that the authors are just skimming the surface of what could potentially be a very relevant study. Prior to publication, it is necessary to include additional details on instrument operation, and case study periods as outlined in the comments below.

General comments

There are many details of instrument operation, including that of the ACSM and also the LTOFMS that are omitted from the paper and supplementary material. The authors focus on measurements for the period from October 2018 to February 2019, but later on only shown data for a three day period in February. Please state this clearly in the abstract, introduction, and methods section if this is the only haze event encountered during this five month sampling period? If not how representative is this haze event compared to other events.

Introduction:

Please highlight better the added value of this work compared to previous studies (cited in the references) on the aerosol-radiation-boundary layer feedback.

Methods:

1. No information is provided on the inlet set up? How is the aerosol dried prior to sampling?
2. A ToF-ACSM fitted with a PM 2.5 inlet was used in this study. This is still a relatively new version of the instrument, and it merits a correct introduction. Please state if this instrument operating with a standard vaporizer or a capture vaporizer? The paper referenced here “Frohlich et al., “deployed a PM1 inlet and not a PM2.5 inlet. Please update the references.
3. What collection efficiency was applied to this data? Please show a plot of how the total mass measured by the ACSM compared with that of the TEOM (also PM2.5), how representative is the PM2.5 ACSM measurements of the total PM2.5.
4. Line 132: The authors state that they applied the correction for the m/z 44 artefact, without showing if this instrument was influenced by this artefact, please provide the artefact values calculated from this instrument from pure ammonium nitrate calibrations. More recent studies (Freney et al., AST 2019)

have additionally shown that this correction is often only necessary at highest NO_3 mass fractions ($> 40\%$). From Figures 2 through to 4, these contributions never appeared to be greater than 40%.

5. Please also state in the text the average values as well as the range for each species measured (for the period that concerns this study).
6. Additionally data from a LTOF-CIMS is provided. This is a complex instrument, and both the operation and the analysis of this data require a considerable amount of work. Please provide more details on the operation of this instrument and the subsequent analysis of the data. Unlike the ACSM used in this study, this instrument is usually operated with a PM1 inlet rather than PM2.5.
7. The authors need to provide comparisons of measurements between the ACSM and the LTOFCIMS. Please provide additional details for this instrument. Was it sampling alongside the ACSM for similar sampling periods?
8. For the OC/EC measurements, Can the authors also provide plots comparing the OM from the sunset with that of the ToF-ACSM and to the LTOFCIMS. How do the O/C plots compare with that of the LTOFCIMS and calculated from the ACSM with that measured by the OC of the Sunset instrument.

Results and Discussion

1. Figure 5 shows data collected during high and low pollution events. Are the differences between the high and low pollution periods significant for all measured species? The authors could perform a significance test (e.g. Wilcoxon rank-sum test).
2. Can the authors provide an estimation of how good these fits represent the data? Can these fits be used in the future to estimate the variability of the aerosol concentration over pollution/haze events?
3. In each plot there are data points (behind the box plots) that are different colors can the authors please provide an adequate legend for this figure.
4. Line 272: When comparing the NH_4 neutralization plots were there any periods where neutralization was not achieved that would suggest the presence of organic nitrate. The LTOFCIMS instrument is capable of providing a good assessment of the presence of organic nitrates.
5. Line 304: This is the first mention of the results of HOMS, can the authors also provide time series of these data together with those of the ACSM.
6. Line 315: It is mentioned that there is abundant ammonia but not mentioned if it is measured here. However in Fig. S4 there are plots of NH_3 as a function of MLH. How were these measurements obtained?
7. Line 320 and Figure 5. It appears in this figure that during a high pollution event the increase in SO_4 is more significant than nitrate.
8. Equally the HOMS appear to have a greater increase during polluted events compared with organics who have a little increase. Previously, it is mentioned that the OA decreases with low MLH. The authors should provide a detailed discussion of HOMS and OA. Also with a simple positive matrix factorization analysis of the ACSM data it would be possible to obtain additional information on the different “types” of organic aerosol measured. In Figure S4 we observe the OC increasing in a similar way to the HOMS. This could also be discussed.
9. Were there any gas phase measurements available to help in the interpretation of the formation of NO_3 ?

Supplementary material

There are 12 plots (3 lots of four labeled images a) through d)) in Figure S2. These plots are not sufficiently and incorrectly described in the figure caption, which refers to “different times” please indicate the times and labels (a) to (e). There is no ‘e).

Figure S3 starts at b) rather than a).

Can the authors improve the caption explanation of the figure. These emission sensitivities represent polluted /periods of high aerosol loadings. The figures show high values coming initially from the west and also from the south. In the figures there is little contribution from the north east sectors.

Figure S4: Only one panel is labelled with “f”). The others nothing. There are two representations of the sub 3 nm clusters (-dN/DlogDp and cm/1). Are both necessary? What is the difference between the two?

Figure S4: I do not believe that any reference is made to this plot in the main text of the manuscript, nor to any measurements of OH, HONO, NH3 [ppb]

Minor comments

Abstract: There is a repetition of information. Line 37 to 39 states that as the MLH decrease the fraction of nitrate aerosol and the total mass concentration increases. This is stated again in Line 41 where the ammonium nitrate and aerosol water increased during low MLH.

Table 1: Species instead of Specie

Line 180: Please include the reference to the previous publication here.

Line 231: I don’t believe that this acronym was correctly defined (NR-PM2.5)

Line 249: Can the authors rephrase this sentence: Assuming that particles of different sizes have the same chemical composition as PM2.5 (organics, NH₄NO₃, EC, (NH₄)₂SO₄, NH₄Cl), the light extinction of particles in the size range of 300-700 nm increased significantly from the relative clean period to the polluted period (namely from 12:00 to 16:00).

Line 255: Based on the available data, it might be better to say “that based on the observations it is likely that....

Line 305 to 309: This is a very long sentence, please try to rephrase.

Line 346: remove “rather” this is

Line 358: Remove brackets around AN.

Also add in ‘the calculated’ light extinction.

The authors mention the presence of a PSM but no measurements are shown. The SMPS data start at 6 nm

Figure 3: Since all other figures are based on 3 days of analysis please state in the figure caption the period that this data is collected over .In the main text it is suggested that this figure represents 6 months of data.

In the caption text change “All the date” to “All the data”