Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2019-450-RC2, 2019

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Rapid transition in winter aerosol composition in Beijing from 2014 to 2017: response to clean air actions" *by* H. Li et al.

Anonymous Referee #2

Received and published: 15 July 2019

This study combines in-situ measurements of PM1 composition in Beijing during the winters of 2014 and 2017 and the simulation results from a regional chemical transport model to investigate the impacts that the clean air actions in China have had on aerosol chemistry. The relative contributions of anthropogenic emissions, meteorological conditions, and regional transport to the changes in aerosol composition in Beijing are also investigated. This is an interesting work and provides a timely and relevant analysis of a current problem – how aerosol pollution in Beijing responded to the implementation of the Air Pollution Prevent and Control Policy. Overall, the manuscript is well written and fits well within ACP's aims and scope. However, the current version may require substantial revision before publication can be considered. A major shortcoming in this manuscript is that the Methods section is very much lacking of important

technical details, for example on aerosol source contribution analysis and CMT model performance, thereby raises doubts about the credibility of the results. Moreover, there are some inconsistencies in the results and discussions which could cause concerns about the quality of the data, representativeness of the findings, or validity of the conclusions. Additionally, the figure legends and captions are often too brief to make the figures understandable.

Detailed comments:

Line 44-46, how much did air pollutants reduce between 2013 – 2017 in the Beijing area?

Section 2.2. is cursory and provides very little information on the organic aerosol source apportionment analysis. Details must be provided on how the PMF/ME-2 analysis of the ACSM data was performed, what data treatments were implemented, and how the solution conditions (eg number of factors, a value, fpeak) were selected and evaluated.

In Section 2.3. more information should be provided on the performance of the CMT model at simulating PM2.5 composition and how the modeling results compare to observations for 2014 and 2017 winters separately.

Section 2.5 lists three assumptions about aerosol properties in the ISORROPIA modeling. Several references are cited and claimed to support these assumptions for this study. But upon reading the references more closely, they don't seem so as the references either talked about aerosols from different locations or under different meteorological conditions, or simply did not provide direct evidence on aerosol physical states. In fact, given the wintry weather (very low RH and T) and intense local emissions in Beijing, it is hard to believe that internal mixing and single aqueous phase were the prevailing aerosol conditions relevant to this work.

Line 168 – 171, there was no mentioning of NH3 and HNO3 measurements in 2.1., but

it is mentioned here that the values of NH3+NH4 and NO3+HNO3 were input into the model. Where did the NH3 and HNO3 data come from?

Line 110, missing numbers after "0." What "a" values were used?

Line 167 -168, what does "the transition in aerosol composition" mean in this sentence?

Line 186 – 188, the Xu et al. study was also conducted in Beijing in the winters of similar years, but the nitrate to sulfate ratios reported there were much lower than in this study. Normal measurement uncertainty could not explain such large discrepancies (more than a factor of two in difference). Was it due to measurement artifacts or does it suggest some issues with the representativeness of the measurement data? What's the implication for the validity of the conclusions presented in this paper?

Line 205 – 206, organics were higher than sulfate and nitrate and in both 2014 and 2017, so calling Beijing aerosol pollution being "sulfate-driven" or "nitrate-driven" does not seem logical.

Line 259, for coal usage reduction in addition to quoting the absolute amount, it would be also interesting in knowing the relative amount of reduction.

Figure 2, according to the diurnal profiles , HOA concentration in 2014 was 2-3 times lower than 2017. If HOA is representative of emissions from transportation, is this level of decrease consistent with the decrease in emissions according to emission inventory? Moreover, the decrease of BC concentration from 2014 to 2017 was between 30-40% but the reduction of total combustion POA (sum of CCOA, BBOA and HOA) was close to 70%. This would suggest some very large, probably unrealistic, changes in the combustion emission factors.

Line 289 – 292, the comparisons of PM1 concentrations between different air trajectory classes do not logically lead to a conclusion about how much Beijing aerosol was influenced by polluted air masses transported from surrounding areas. Beijing has local pollution sources which could cause high PM events as well. In fact, in the paragraph

immediately beneath, the authors reported that CMT simulation indicates that regional transport contributes to only 30 -40% of PM2.5 in Beijing.

Section 3.2.4, 2nd paragraph, what are the rationales for using SO4/BC and NO3/BC ratios in the analysis? Sulfate is a secondary species with formation time scales usually much longer than the emission time scales of BC. So the physical meaning of SO4/BC ratio is unclear.

Section 3.2.4, 2nd paragraph, nitate/(nitrate+NOx) is not a proper index for the oxidation ratio of nitrogen. Discussions related to NOR should be either removed or revised.

For the discussions of the relationship between SOR and RH, it is important to point out that RH was measured locally at the sampling site but sulfate was mostly formed on a regional scale, ie, in air masses upwind of Beijing. Is it valid to assume that in-situ RH measurement data are representative of the RH conditions in the air masses where sulfate was formed?

Line 330 -331, does the higher SOR in 2017 than 2014 necessarily demonstrate "a higher sulfate production rate in 2017"? A relatively larger contribution from background air masses could also lead to higher SOR.

Line 544- 545, this citation is incomplete

Figures in the supplementary materials are fuzzy, need to use better resolution.

Figure S6d, explain how to read the figure and the meanings of N-E, W-N, E-S, S-W?

Figure S7, what do the color bars stand for?

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