

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.

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(a) Overview The authors used fine particle data collected in Beijing during the winters of 2014 and 2017 to understand changes in the chemical composition of the PM over a period when particle mass decreased significantly. The results here are consistent with past data, showing a large decrease in PM mass, but also revealing that this is largely because of decreases in sulfate, organics, and unidentified components, as a result of decreasing local and regional emissions. The authors find that meteorology also contributed to lower PM concentrations in 2017 but that this was a smaller factor.

This is an interesting paper with a number of complementary components. On the other hand, there are a number of issues that need to be addressed, as described

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below. The thermodynamic modeling seems problematic and needs more clarification and information.

(b) Major Points 1a. Figure 2 and lines 190 – 191: The text states that “All SIA species showed similar diel trends in the two winters. . .”. For some species this is true, but for many of the species, the diel variability is much lower in 2017 than in 2014, suggesting that while more local sources dominated in 2014, regional sources are more important in 2017. Take CCOA as an example: in 2014, the concentration varied by a factor of 3, while in 2017 the variation was probably only a factor of 1.5 or less. Can the change in diel variability be used as an indicator of local vs. regional pollution?

1b. Of course boundary layer height is a major influence on concentrations as well. I suggest you add a plot of CO as a tracer for BL height. Does plotting the ratio of PM component/CO help disentangle chemistry and BL height? These might be useful supplemental plots.

2. Figure 8 and its corresponding text. (i) Why normalize panels (a) and (b) to BC concentrations? Since both the numerator and denominator are changing between 2014 and 2017, the ratio seems less effective as a normalizing factor. What do the plots look like if sulfate and nitrate are not normalized by BC? (ii) The data are very noisy and so the authors have chosen to plot the median values (as the solid circles). But what does it look like if the mean and standard deviation are plotted instead? Are there any significant differences between RH values for a given year? If the authors want to stay with median, they should at least show some interquartile ranges or other measures of variability about the median. (iii) Line 323: “. . .the starting point of SOR growth was clearly delayed in 2017. . .”. Given the spread in the data, this is speculative since the median has no associated uncertainty or variability. Can this difference be tested statistically? (iv) Lines 330 – 335. The authors explain that the higher sulfate / (sulfate + SO₂) ratio (i.e., SOR) in 2017 at low RH results from less of an oxidant limitation. But isn't an alternative explanation that a higher fraction of the sulfur pollution in Beijing is from regional transport, which would have a higher SOR since it is more aged? (v) The nitrate / (ni-

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trate + NO₂) ratio (i.e., NOR) discussion in lines 339 – 344 seems to be missing a few points. First, NOR is a poor measure for the extent of NO_x oxidation since it considers only particulate nitrate and not gas-phase HNO₃. Under acidic particle conditions (as seems typical in 2014), most N(V) will be gas-phase HNO₃, a result of sulfate acidity driving out the particle nitrate. But this is not accounted for if NOR is calculated only with particulate nitrate, as is done here. Running the e-AIM thermodynamic model on the average 2014 data shows that the gas-phase HNO₃ concentration is over an order of magnitude higher than particulate nitrate. Thus N(V) / NO_x + N(V), with both phases considered for N(V), would be a better measure of NO_x oxidation than NOR. ISORROPIA could be run to examine HNO₃(g) in the two years. (vi) As a consideration for future work, it would be enormously helpful to have gas-phase measurements of NH₃ and/or HNO₃ to constrain the modeled aerosol pH.

3. Figure 9 and its corresponding text. (i) This figure indicates that the aerosol pH in 2014 was approximately 5, but this is inconsistent with the average composition indicated in Table 1. Using the averages for 2014 in Table 1, assuming protons make up the missing positive charge (which I assume was also done in ISORROPIA) and using a relative humidity of 60% in the e-AIM model results in a pH of -0.04 assuming a proton activity coefficient of 1; the pH is -0.7 if the e-AIM activity coefficient (4.22) is used. These results were done allowing solids to form, but it shouldn't change significantly if solids were not allowed to form. While the pH undoubtedly varies between samples, it is hard to believe the typical pH is near 5 given the ion imbalance in the data. (ii) Is the range of pH values given in line 361 (pH 5.0 to 6.2) from their work? Why is it for 2013 – 2017, rather than just the two winters of this study? It's not clear that the authors did much with the ISORROPIA data: why not report ALW and pH for every sample? This would be very useful information. (ii) The 2017 average data in Table 1 has a net positive charge, requiring a nonsensical negative concentration for protons that is of similar magnitude (though smaller) than the proton concentration from the 2014 data. This prevents use of a thermodynamic model and suggests that there are other charged species, likely organics, which are affecting the ion balance and the pH

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calculation. This should be discussed. Were the authors able to calculate ALW and pH values for the 2017 data? (iii) Under the acidic conditions I calculated for 2014, the value of NO₃⁻ / (HNO₃ + NO₃⁻) is very small, in contrast to the values described in the manuscript. (v) Similarly, I calculate a NH₄⁺/NH_x ratio near unity in 2014, in contrast to the small values reported in the text.

(c) Minor Points 1. Throughout the manuscript, it would be much better (and more consistent with the other inorganic nomenclature in the paper) to use "Cl⁻" as the abbreviation for chloride rather than "Cl".

2. lines 183-184: This sentence is unclear. Clearer wording would be "The decreases in particulate nitrate and ammonium during this period were 1.3 and 1.5 μg m⁻³, respectively."

3. Figure 1. The second line of the figure caption is unclear. These are not "rates", but rather decreases in concentrations, yes? Also, three significant figures in the decreases seems one too many.

4. line 188: It is odd that the Xu nitrate/sulfate ratios are approximately half of the values in the current work. Why this discrepancy?

5. lines 203-204: Sulfate is a minor fraction of haze in 2014 as well as 2017, so this statement isn't very useful. It would be better to indicate the sulfate contributions to each of the pollution classifications in 2017.

6. lines 204–205: This description of nitrate contribution is for 2017?

7. line 208: State the 2014 and 2017 OA concentrations along with the overall decline.

8. line 209: "The contribution from HOA was 2.6 μg m⁻³...". This is not as clear as it should be: better to state something like "HOA decreased by 2.6 μg m⁻³...".

9. lines 240 – 241: It is not clear what cases A, B, C, and D refer to. Are these panels of Fig. S7?

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10. lines 258-259: This is unclear. Should “Until 2017...” be “By 2017...”?

11. line 298: The 57.9% is not a “rate” and likely has one too many significant figure.

(d) Recommendation I recommend that the paper be accepted after major revisions to address the points above.

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