Interactive comment on “Insights into a historic severe haze weather in Shanghai: synoptic situation, boundary layer and pollutants” by C. Leng et al.

Anonymous Referee #1

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The manuscript by Leng et al. presents a case study for a regional haze event in China. The authors analyzed the mass concentration, optical properties, CCN concentration, and chemical species of the aerosol particles monitored during the event. Weather charts, PBL heights, back trajectories, and satellite-based AOD data were used to complement the analysis. Although the dataset can be useful for elucidating the severe air quality issue in China megacities, I feel some analyses are not well developed. The main conclusions and the novelty are not clear to me. On English context, the manuscript needs a lot of edits to improve the grammar and readability. The manuscript should be improved both on the technical and presentation quality before it can be accepted. Detailed comments are listed below.
Major concerns:

1. The paper compared several controlling factors for their correlations with visibility. These factors include RH, PM, and particle number concentration at different sizes. However, these correlations were not based on the physical relationship. Directly computing the correlation coefficients for visibility (km) may not be appropriate, because the visibility values are generally low during hazy days and the correlation coefficient would be largely determined by the high visibility data. The authors could consider using the inverse of visibility as a proxy of extinction coefficient (at ambient RH) to do such analysis. Alternatively, the Mie model with particle size distribution and hygroscopicity as input can calculate visibility/extinction coefficient. This approach has been previously employed in several field campaigns in China. It considers the physical relationship between visibility and other factors thus more sensible than the regression used in the present study.

2. Calculation of kappa.

The authors used the measured inorganic ions to calculate the hygroscopicity. The contribution of water-soluble organic carbon (WSOC) was entirely omitted in this analysis. WSOC can contribute 5%-40% percent of the water content for urban aerosols, depending on the particle size and composition. The value of kappa was therefore underestimated. In addition, the authors simply added sulfate, nitrate, and ammonium ions and assigned a kappa value of 0.6 (ammonium sulfate) to this group. This treatment was somewhat oversimplified. In some conditions, the bisulfate or sulfuric acid can present when the particles are acidic. The contribution of H+ to hygroscopicity would be omitted in this simplified treatment. Although this may not be the case for current study (because the concentration of nitrate is high, as an indication of excess ammonium), the method used in current manuscript can be very misleading for readers. The data should be analyzed in a more rigorous way, e.g., by considering ion balances or using a thermodynamic model.
In addition, the kappa value calculated in this work should be considered as an effective value for the particle population of PM2.5. It is distinct from the common definition for kappa, which is based on HTDMA or CCN measurements for monodisperse particles. This should be at least discussed in the manuscript.

3. CCN data. For about half of the CCN data shown in Fig. 6, the CCN number concentration for 0.2% SS is higher than that for 0.4% SS. What does that mean? Show some calibration data as supplementary information if possible.

4. Criteria of haze. In section 3.1.1, the authors state that "It has been widely accepted that the key criterion for discerning a haze event is to identify an apparent decrease of atmospheric visibility less than 10 km, and ambient relative humidity (RH) below 80% lasting for several hours (Fu et al., 2008; Du et al., 2011). When 80% < RH < 90%, the event is referred to as a complex of haze-fog co-occurring or transition, and it is also classified into hazy episode in the present study (Leng et al., 2014a)."

These definitions are vague and not widely accepted. Fog consists mainly of activated liquid droplets. Supersaturation is needed for the formation of fog (not 80%<RH<90%). It is the same as cloud from the point of view of microphysics. Haze particles, in contrast, are still below of the activation point in the Kohler curve, even the RH is higher than 90%. A classification guideline for fog and haze using RH and visibility might be acceptable as an operational definition. However, I would not recommend it in any scientific literature.

Minor comments:

5. Abstract: the abstract should be revised. Please use short and clear sentences to improve the readability.

6. The language, in particular terminology, should be carefully checked throughout the whole manuscript to ensure that it is precise. Here are a few examples:

P32564, L6, "aerosol particles can aggregate and result in air pollution..." "aggregate"
in aerosol science usually refers to the formation of clusters. A more precise word can be "accumulate" in this context.

P32564, L7, "diffusion conditions" -> "dispersion conditions"?
P32567, L13, "DMA transferring function" -> "DMA transfer function"?
P32568, L23, "active carbon" -> "activated carbon"?
"hazy episode" -> "haze episode"?
"haze weather" -> "haze" or "haze event"?
"vertical diffusion" -> "convection"?

7. Section 2.2.
Specify the principle of MAGRA. Are the gases measured by MAGRA also analyzed?
Was the size distribution measured at dry condition?
P32567, L1 "for the instructions of sampling" -> "for the protocols of sampling"
P32567, L13 "LPS was calculated ..." How LPS was calculated? by Mie model? Or it is just a calibration using the PSL particles?
P32568, L7 "without obscuration due to relatively lower aerosol loading and well mixed atmosphere". What does this sentence mean? Why lower aerosol loading can obscure the atmosphere?
P32569, L3. Specify the operation principle of PM monitors. Does the RH affect the measurements?

8. Section 3.1.1.
I would suggest to combine fig. 1 and fig. 2 and label the panels as a, b, and c. It is difficult to align low visibility and high RH in separate figures.
9. Section 3.1.3
P32571 It seems that the record-breaking haze event was regional rather than a local event, because the PM concentrations in other cities in the same region were also high. Consider moving the discussions for AOD maps closer to this paragraph and make this point clearer to readers. When did the PM2.5 measurements start in Shanghai? The 600 ug/m3 was a record since when?

10. Section 3.1.4
An implicit assumption of the absorption coefficient calculation is that an averaged mixing state of black carbon was used. This should be discussed.

Specify the wavelength of data reported. (532 nm?)

Also include the single-scattering albedo (SSA) in the analysis?

11. Section 3.1.6
P32575, L14 "suggesting that atmospheric oxidation of NO2 and SO2 contributed significantly to the formation of nitrate and sulfate". This sentence is confusing. Shouldn't nitrate and sulfate be entirely contributed by NO2 and SO2, respectively? Did the authors mean "suggesting that atmospheric oxidation of NO2 and SO2 contributed significantly to the formation of particulate matter"?

The authors used nitrate/sulfate ratio to discuss contribution of motor vehicles. Nitrate concentration should also depend on the acidity. Abundance of ammonia can play a key role. As suggested in my previous comment, the inorganic species data should be analyzed in a more sensible way by analyzing ion balance or using a thermodynamic model.

12. Section 3.2.1
P32576 L10 "clod"→"cold"?
Include citations for the original data source of weather charts in Fig. 8 and 9. (KMA?)

13. Section 3.2.6 Cite original literature for classification of aerosol modes.

Would the mass concentration calculated by integration of size distribution consistent with measured PM2.5 and PM10? Consider the difference between aerodynamic diameter and mobility diameter in the calculation. This kind of closure calculation can be useful for validating data quality.

"...no significant correlation was derived between atmospheric visibility and aerosol size of 10–600nm and 1.4–10 μm " What does this sentence mean? Were the number concentration used in the regressions? Please specify.

14. Figure 3 and Figure 7.

Please use discernible colors other than red and pink.

15. Figure 11.

Use a more discernible color scale.

16. Figure 13.

Avoid the bright yellow color in the figure.

R2 = 19 -> R2=0.19 for PM2.5?

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