

Comments on “Field characterization of the PM<sub>2.5</sub> Aerosol Chemical Speciation Monitor: insights into the composition, sources and processes of fine particles in Eastern China”

This Paper described the first comparison results of ACMS equipped with newly developed PM2.5 lens +capture vaporizer with other multiple on-line instruments, including a traditional PM1 ACSM (with standard vaporizer), TDMPS (for particle size distribution), On-line EC/OC, MARGA (for inorganic species), TOEM (total PM2.5 mass), BAM etc. Apparently, this is a sufficient and valuable dataset to investigate the performance of PM2.5-ACSM. Good linear correlations have been shown between the non-refractory species detected in PM2.5-ACSM with other measurements, suggesting a full detection of PM2.5 masses in this type of ACSM. Then the authors discussed secondary inorganic aerosol formation, POA and SOA, the aqueous/photochemical reactions, two case studies and the geography origins of those aerosols. Overall, I recommend this manuscript to be published in ACP. However, a major revision is suggested here based on the reason addressed below.

The authors tried to combine multiple topics into one paper, which is very distracting. I do not know the topic of this paper is to evaluate the PM2.5-ACSM or to investigate the aerosol formation. The analysis in the Section 3.3-3.5 is quite shallow. Exclusive similar results and analysis on aerosol composition and sources have been published in China before (Li et al., 2017 and references therefor in). I did not see any new finding in the analysis reported in this paper. I suggest the authors cut and combine these parts and focus on more interesting points. For the comparison part, the authors should pay more attention to the details for validating their results, since this is the main selling point based on the abstract.

**Major comments:**

(1) Line 248-267: I found the comparison ratios of PM1-ACSM vs PM2.5-ACSM for different species are quite different in Fig. 2. i.e. OA: 0.5~; sulfate ~0.35; nitrate ~0.72; Ammonium ~0.46; Chloride: ~1. Any comment on this? To explain this difference is quite important for understanding the PM2.5-ACSM instrument. Does this indicate the aerosols are externally mixed? However, when I looked at the time series of mass concentration in Fig.3, nitrate correlate quite well with sulfate and ammonium, implying the aerosols might be internally mixed.

In line 397-398, the authors also stated the SOA and SIA are internally mixed. If the SOA and SIA are internally mixed, then why the ratios of PM1 vs PM2.5 are varied with different inorganic and organic aerosol species. What are the PM1 vs PM2.5 ratios of OOA and POA?

(2) Line 298-300: Since the authors have the TDMPS, how is the volume comparison between PM1, PM1-2.5 and PM2.5 based on the TDMPS data..

More importantly, This dataset, as another independent dataset, can help to confirm the comparison results between ACSM with BAM and TOEM. The authors should calculate the PM1 and PM2.5 masses based on volume conc. calculated based on TDMPS measurement and

density calculated from ACSM, which can help the quantification of total PM1-ACMS and PM2.5 ACSM in Fig. 1 and Fig. 2.

(3) The authors should quantify if the PM2.5-ACMS really detect the PM2.5 masses. How much mass was lost at smaller size ranges in the PM2.5-ACSM. Xu et al. (2016) showed a larger mass loss below 200 nm of aerodynamic particle size. The author can calculate the lost masses based on TDMPS size distribution measurement.

(4) Line 212-216: Which calculation mode of ISORROPIA-II did the authors use for this calculation? Is there any gas-phase measurement to constrain the input or evaluate the output of the model? Gas phase NH<sub>3</sub> was also reported in this paper. Has the author compare the modeled NH<sub>3</sub> with measurement NH<sub>3</sub> to validate their results?

(4) Line 432-437, The NO<sub>2</sub> accelerate the sulfate formation is based on the fact that the aerosol is neutral. What is the pH of aerosol in this study? The authors had run the ISORROPIA-II, thus the pH should be easily calculated (Guo et al. 2017).

(5) Line 283-284: Since the authors have calibrated the instrument with NH<sub>4</sub>NO<sub>3</sub> particles, the authors can derive their own CO<sub>2</sub>/NO<sub>3</sub> ratio following Pieber et al. (2016).

Line 353-355: The authors should estimate their own fCO<sub>2</sub> interferences based on the calibration data. The fCO<sub>2</sub> production from other crustal nitrate can be roughly estimated based on the relationship between CO<sub>2</sub>/NO<sub>3</sub> from pure NH<sub>4</sub>NO<sub>3</sub> particles and CO<sub>2</sub>/NO<sub>3</sub> from pure NaNO<sub>3</sub> or other particles reported in Pieber et al. (2016).

(6) Line 260-264: This is a paradox: if all the Na came from NaCl, then it will not exist as NaNO<sub>3</sub>. The author cannot assume all Na<sup>+</sup> exist in forms of NaNO<sub>3</sub> in the aerosols then exist as NaCl at the same time. Meanwhile, (1) the author can assume a maximum Cl mass balanced from Na, Ca, K<sup>+</sup>, Mg. To see if this calculated maximum Cl can explain the difference of Cl between AMS and Marga. (2) the author could correlate the time series of NO<sub>3</sub> difference between AMS and MARGA with that of Cl. In such a way, the authors could check if these differences come from the same source.

### **Other comments:**

Line 54: Please define “high time-resolution”

Line 78-80: A paper published recently suggested the aerosol pH in Beijing is less than 5, typically close to 4, even under the highest levels of ammonia. This level of acidity suppresses potential multi-phase sulfur oxidation pathways recently suggested to explain missing sulfate sources in the region (Liu et al. 2017). The author should also consider the possibility of this point.

Line 88-89: Any evidence for this? Is there any other potential reason that could lead to this difference of source apportionment?

Line 160: Change “response factor” to be “ionization efficiency (IE)” or “sensitivity”

Line 243-245: To better address the comparison result, the comparison uncertainty, propagated from the measurement uncertainty of each instrument should be fully addressed, which can give a better understanding of how good of the comparison results and also will be useful references for other users.

Line 271-278: What the size cut of on-line EC/OC instrument. If it is PM2.5, then the ratio between OM from PM1-ACSM vs OC from PM2.5 EC/OC is meaningless, which should not be considered at all.

Line 276-278: The authors did not show any evidence to support this statement.

Line 344: What is the f60 and f73 from the PM2.5-ACSM-CV compared with these from the PM1-ACSM-SV.

Line 345: Specify the m/z 55/57 ratio value here.

Line 386-389: Have the authors considered new particle formation process could be a potential reason for the higher NH<sub>4</sub>/NH<sub>3</sub>+NH<sub>4</sub> ratios at smaller size ranges.

Line 380: Delete “e”

Line 393-395: What is the “different roles”? Please specify.

Line 398-399: What kind of SOA enhancement? I only saw an enhanced SOA at lower LWC concentration (<50 ug/m<sup>3</sup>) in Fig 8a.

Line 419-423: The authors stated cooking emission included in the POA factor and POA showed clearly noon and night peaks for cooking emissions. Thus, the POA/CO ratios should not be dominated by urban traffic emissions. In contrast, I think the POA vs CO regression ratio reported here should be larger than POA vs CO ratio from urban traffic emissions, based on Hayes et al. 2013.

What is the regression method for the POA/CO calculation. Could the authors show the scatter plots. Orthogonal distance regression should be used. Is the intercept fitted to be zero or not?

## References:

Liu, M., Song, Y., Zhou, T., Xu, Z., Yan, C., Zheng, M., Wu, Z., Hu, M., Wu, Y., and Zhu, T.: Fine particle pH during severe haze episodes in northern China, Geophys Res Lett, n/a-n/a, 10.1002/2017gl073210, 2017.

Hayes, P. L., Ortega, A. M., Cubison, M. J., Froyd, K. D., Zhao, Y., Cliff, S. S., Hu, W. W., Toohey, D. W., Flynn, J. H., Lefer, B. L., Grossberg, N., Alvarez, S., Rappenglück, B., Taylor, J. W., Allan, J. D., Holloway, J. S., Gilman, J. B., Kuster, W. C., de Gouw, J. A., Massoli, P., Zhang, X., Liu, J., Weber, R. J., Corrigan, A. L., Russell, L. M., Isaacman, G., Worton, D. R., Kreisberg, N. M., Goldstein, A. H., Thalman, R., Waxman, E. M., Volkamer, R., Lin, Y. H.,

Surratt, J. D., Kleindienst, T. E., Offenberg, J. H., Dusanter, S., Griffith, S., Stevens, P. S., Brioude, J., Angevine, W. M., and Jimenez, J. L.: Organic aerosol composition and sources in Pasadena, California, during the 2010 CalNex campaign, *Journal of Geophysical Research: Atmospheres*, 118, 9233-9257, 10.1002/jgrd.50530, 2013.

Guo, H., Liu, J., Froyd, K. D., Roberts, J. M., Veres, P. R., Hayes, P. L., Jimenez, J. L., Nenes, A., and Weber, R. J.: Fine particle pH and gas–particle phase partitioning of inorganic species in Pasadena, California, during the 2010 CalNex campaign, *Atmos. Chem. Phys.*, 17, 5703-5719, 10.5194/acp-17-5703-2017, 2017.

Li, Y. J., Sun, Y., Zhang, Q., Li, X., Li, M., Zhou, Z., and Chan, C. K.: Real-time chemical characterization of atmospheric particulate matter in China: A review, *Atmos Environ*, 158, 270-304, <https://doi.org/10.1016/j.atmosenv.2017.02.027>, 2017.