

Comments on “Field characterization of the PM_{2.5} 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 ACSM equipped with newly developed PM_{2.5} 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 PM_{2.5} mass), BAM etc. Apparently, this is a sufficient and valuable dataset to investigate the performance of PM_{2.5}-ACSM. Good linear correlations have been shown between the non-refractory species detected in PM_{2.5}-ACSM with other measurements, suggesting a full detection of PM_{2.5} 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 PM_{2.5}-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 PM_{2.5}-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 PM_{2.5}-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 PM_{2.5} are varied with different inorganic and organic aerosol species. What are the PM₁ vs PM_{2.5} ratios of OOA and POA?

(2) Line 298-300: Since the authors have the TDMPS, how is the volume comparison between PM₁, PM_{1-2.5} and PM_{2.5} 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 PM_{2.5} 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_3 was also reported in this paper. Has the author compare the modeled NH_3 with measurement NH_3 to validate their results?

(4) Line 432-437, The NO_2 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_4NO_3 particles, the authors can derive their own CO_2/NO_3 ratio following Pieber et al. (2016).

Line 353-355: The authors should estimate their own fCO_2 interferences based on the calibration data. The fCO_2 production from other crustal nitrate can be roughly estimated based on the relationship between CO_2/NO_3 from pure NH_4NO_3 particles and CO_2/NO_3 from pure NaNO_3 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_3 . The author cannot assume all Na^+ exist in forms of NaNO_3 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^+ , 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_3 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 PM_{2.5}, then the ratio between OM from PM₁-ACSM vs OC from PM_{2.5} 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 PM_{2.5}-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 NH₄/NH₃+NH₄ 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/m³) 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.

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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.

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