Authors response to editorial boards of Kang et al., ACPD, 2017

Co-Editor Decision: Publish subject to minor revisions (review by editor) (06 Mar 2018) by Sergey A. Nizkorodov

Comments to the Author:

Dear authors. I am sorry about the delay in reviewing your paper. As you have seen the reviewers have had a larger number of suggestions, and it takes time to review and re-review papers, which require major revisions. However, we are not converging to the "Minor Revision" stage. Please address the remaining minor comments and the paper will be accepted.

[Response]

The point-by-point responses for comments are given below.

Your paper title "Photochemical aging of organic and inorganic ambient aerosol from the Potential Aerosol Mass (PAM) reactor experiment in East Asia" is too general in my opinion. I would choose a title that better reflects the primary finding of the paper, as opposed to the technical method used to achieve this result.

[Response]
As you suggested, we changed the title to "Photochemical aging of aerosol particles in different air masses arriving at Baengnyeong Island, Korea".

Last paragraph of the introduction section can be improved. This is usually where you explain the motivations for the study and summarize the main results. Good questions to discuss here: What is the hypothesis? Why was this particular location chosen? What makes it better than other possible locations for testing your research hypothesis? What are the main findings summarized in one sentence?

[Response]

We modified the last paragraph of the introduction section as suggested.

In this study, we deployed a PAM reactor at an island site in the Yellow Sea to investigate the photochemical aging of ambient aerosols in sulfated-dominated northeast Asia. The size, mass, and chemical characteristics of ambient and PAM-processed aerosols were compared for different air masses transported from nearby continents. Their aging characteristics were examined in terms of secondary aerosol formation and evolution of pre-existing aerosol particles. The discussion mainly focused on the formation of sulfate and further oxidized organics and the loss of less oxidized organics upon photochemical oxidation in PAM reactor.

L37 and elsewhere: m/z is generally written in italic in ACP and other journals
L63: delete "(SA)" since you are not using this abbreviation anywhere in the paper
L43: replace "NO3" by "inorganic nitrate"

L179 and L292: there is no need to define "CS" twice and not use it in the paper – please remove this abbreviation

L187: replace "COO" by "CO2"

[Response]

We corrected all parts following the comments.

Figure 2: symbol for micro did not print correctly in the Y-axis labels. The size and X-axis labeling of panels c and d are not the same as size of panels a and b. Are these figures meant to be on the same packed stacked on top of each other?

[Response]

We will provide the original figure files to avoid the error during conversion. Figure a and b are for mass concentration, c for volume concentration, and d for number concentration. They are all time-series plots for the same period. We will change the X-axis label of Figure d.

Figures 3,4: same issue with the symbol for micro not printing. Perhaps it is only in my Adobe Acrobat program.

[Response]

It is probably due to conversion problem. To make sure that the symbol "μ" correctly printed in the figure, we will submit all figures separately in original format at the final stage of submission.

Figures 5, 6: very low image quality – need to be fixed for the final publication.

[Response]

It seems like Adobe Acrobat automatically lowers the image quality. We will submit high quality figures as original files at final stage.

L286: replace "formation of nuclei-mode particles" with "nucleation of particles"
L284 and L293: I would not use "nuclei-mode particles", a more common term if "nucleation mode particles". Perhaps you can say "freshly nucleated" on L284.

[Response]

The term of "nuclei-mode" was replaced with "nucleation mode" in the revised

manuscript.

The title 4.1 "Formation of nuclei-mode particles" was replaced with "Nucleation of particles", too.

In Line 287, "the formation of nuclei-mode particles" was modified to "freshly nucleated particles".

Page 14: I think it is not accurate to use terms "organic m/z 43" and "organic m/z 44". I would explain at the beginning that m/z correlates with less oxidized organics and m/z correlates with carboxylic acids and just use "m/z 43" and m/z 44" from then on.

[Response]

In the revised manuscript [Line 188-191], the following sentences were added to explain the term of m/z 43 and m/z 44.

The term m/z 43 and m/z 44 from the mass spectra correlate with less oxidized OAs and further oxidized organics such as carboxylic acids, respectively. Therefore, the discussion on single mass of *m/z* 43 and *m/z* 44 represent the entire organic classes.

Page 14: I also fine the use of terminology such as "m/z 43-like compounds" and "m/z 44-like compounds" in accurate. Please avoid such lose terms.

[Response]

In the revised manuscript, "m/z 43-like compounds" and "m/z 44-like compounds" were reworded to "less oxidized organics" and "further oxidized organics", respectively.

L425: It is not clear to me what microN refers to in the formula

[Response]

The unit "μN " represents the normality of 10^{-6} equivalent per liter. Thus, it was replaced with "μeq/L" in the revised manuscript.

Conclusion section: it is not clear that the conclusion section is needed since you have an atmospheric implication section before it. I think they should be merged together.

[Response]

Conclusion section summarizes the significant results obtained from this study. However, the main findings are all stated in Atmospheric implication section. Therefore, Conclusion section was removed as you suggested.

COPY OF REVIEWER COMMENTS:

This 2nd revision of the Kang et al. manuscript is improved vs. the previous one. I still disagree about a number of analyses and details (or e.g. leaving out PMF), but this is the authors' paper and not mine, so we'll leave it at that. I think it should be acceptable into ACP after the authors address a few remaining items:

- Abstract L45: referring to the volatility of the organics is not justified. This should just refer to the more oxidized organics. Oxidation was measured, volatility was not. And the two are not always correlated. Early studies found and reported a correlation, but more studies have accumulated since then that have shown that oxidation and volatility are often not correlated. This is also an issue around L272 and L347 and perhaps elsewhere.

For this reason the terms LO and MO OOA (less and more oxidized OOA) are must more commonly used now instead of SV and LV-OOA. I recommend that the authors adopt this terminology.

[Response]

We adopted the terms of "less oxidized OAs" and "more oxidized OAs" and modified the manuscript and references accordingly.

Line 304 in revised manuscript: The paragraph starting with "Mohr et al. (2012) observed that … " was reworded as follows.

A previous study showed that the less oxidized organic aerosols were found more often than more oxidized OAs in particles smaller than 200 nm (Sun et al., 2012). In addition, the mass concentration of *m/z* 43 was higher in less oxidized OAs than more oxidized OAs (Sun et al., 2012). In the present study, the contribution of *m/z* 43 to total organics was greater in the organics-dominated episode than in the sulfate-dominated episode, and the loss of organics in PAM reactor was also greater in the organics-dominated episode. These results suggest that the less oxidized OAs were more in the organics-dominated episode than in the sulfate-dominated episode.

Line326 in revised manuscript: "SV-OOAs and LV-OOAs (Jimenez et al., 2009)" was changed to "less oxidized OAs and more oxidized OAs (Zhu et al., 2018)".

References added:

Sun, Y.L., Zhang, Q., Schwab, J.J., Yang, T., Ng, N.L., Demerjian, K.L.: Factor analysis of combined organic and inorganic aerosol mass spectra from high resolution aerosol mass spectrometer measurements, Atmos. Chem. Phys., 12, 8537-8551, 2012.

Zhu, Q., Huang, X.-F., Cao, L.-M., Wei, L.-T., Zhang, B., He, L.-Y., Elser, M.,

Canonaco, R., Slowik, J.G., Bozzetti, C., El-Haddad, I., Prévôt.: Improved source apportionment of organic aerosols in complex urban air pollution using the multilinear engine (ME-2), Atmos. Meas. Tech., 11, 1049-1060, 2018.

- The use of solenoid valves for switching, which get quite hot in operation, could have caused some evaporation of the aerosol, which should at least be mentioned in the methods section.

[Response]

We added the statement to the manuscript and supplements as follows.

Line 133 in revised manuscript: In addition, the three-way switching valve might cause the evaporation of ambient and PAM aerosols when it was getting hot during operation.

Line 17 in Supplements: The 3-way switching valve might cause the evaporation of ambient and PAM aerosols when it was getting hot during operation."

- Responses p4-5 (and relevant paper text and figures): the scatter plots of AMS vs SMPS do not look consistent with BC or soil at all. BC is typically a much smaller fraction of total PM1 at remote sites (more like 1-2%). Soil is highly variable and typically uncorrelated with secondary species, and would not explain the nearly constant slopes observed in many periods. Likely differences in particle transmission or remaining calibration uncertainties are the real reasons. The accuracy of both the AMS and the SMPS (for mass or volume) are ~35%, so perfect agreement is not expected. I suggest revising this text accordingly, as otherwise confusion can result since the reasons given are not plausible for experienced practitioners. Actually acknowledging the real limitations and uncertainties of the measurements inspires more confidence than blaming effects that are implausible causes of what's observed.

[Response]

Surely, there is uncertainties involved in AMS and SMPS measurement, which is added to the manuscript. As you mentioned, the contribution of EC to mass is small and soil contribution is highly variable in other remote regions. However, the maximum EC concentration of ~1.5 $\mu g/m^3$ is pretty high particularly in summer season, which is mostly associated in submicron particles (PM1.0). In previous study conducted in Jeju, the average mass fraction of EC to PM1.0 was 10 % (Lim et al., 2012). Although soil contribution is substantial in supermicron particles during cold season, its influence is consistent through the year even in submicron particles (e.g., Shang et al., 2018). Other than natural dust, construction dust is substantial source of PM2.5 and it accounts for ~10 % of PM2.5 mass in Beijing (haze study conducted in 2017 by Peiking University).

Line 266 of the revised manuscript: The relevant part was modified as follows.

The disagreement is largely associated with AMS and SMPS measurement uncertainties. However, the role of elemental carbon or soil particles that are abundant in the study region (Lee et al., 2007; Lim et al., 2012) may not be ruled out because there are captured by SMPS but not by AMS.

In supplements, the discussion on Figure S7 was also modified as follows.

Figure 2(c) in the manuscript is a time series of AMS and SMPS particle volume concentration and Figure S7 is a scatter plot of AMS and SMPS particle volume concentration. For ambient aerosol particles, the AMS and SMPS particle volume concentrations agree to within measurement uncertainties, but for the PAM aerosol particles, the SMPS volume concentration was greater than AMS volume concentration by a factor of 1.6. While this difference can be explained by the measurement uncertainties of the two instruments, it is also possible that elemental carbon and soil particles are being detected by the SMPS but not by the AMS. In ambient aerosol particles data for the organics-dominated episode, the AMS volume concentration was slightly greater than or similar to that of the SMPS, but in sulfate dominated episode, AMS volume concentration was smaller than that of the SMPS.

X. Shang, M. Lee, J. Han, E. Kang, S.-W. Kim, Ö. Gustafsson, L. Chang, Identification and chemical characteristics of distinctive Chinese outflow plumes associated with enhanced submicron aerosols at the Gosan Climate Observatory, Aerosol and Air Quality Research, 18, 330-342, 2018.

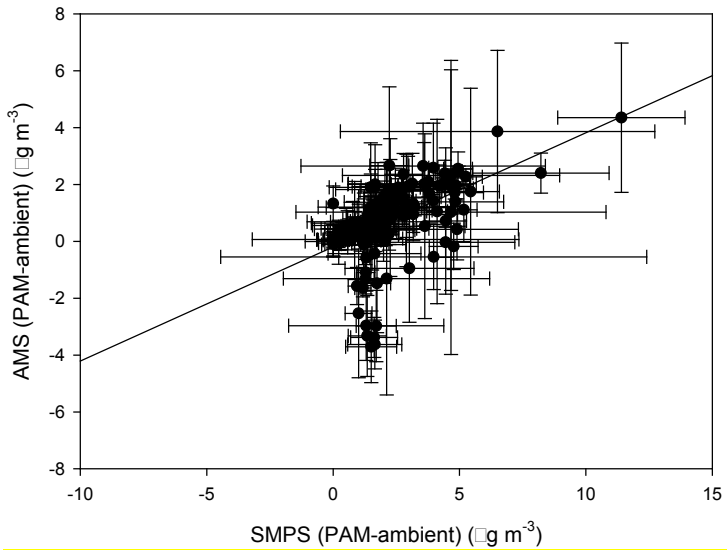
- An important plot, which I had requested before, but it is still not shown, is a scatter plot of the added mass in the SMPS (SMPS_after_PAM – SMPS_ambient) vs the added mass in the AMS (AMS_after_PAM – AMS_ambient) using contiguous points in time. That plot is important for confidence on the quantification of the reactor enhancements.

[Response]

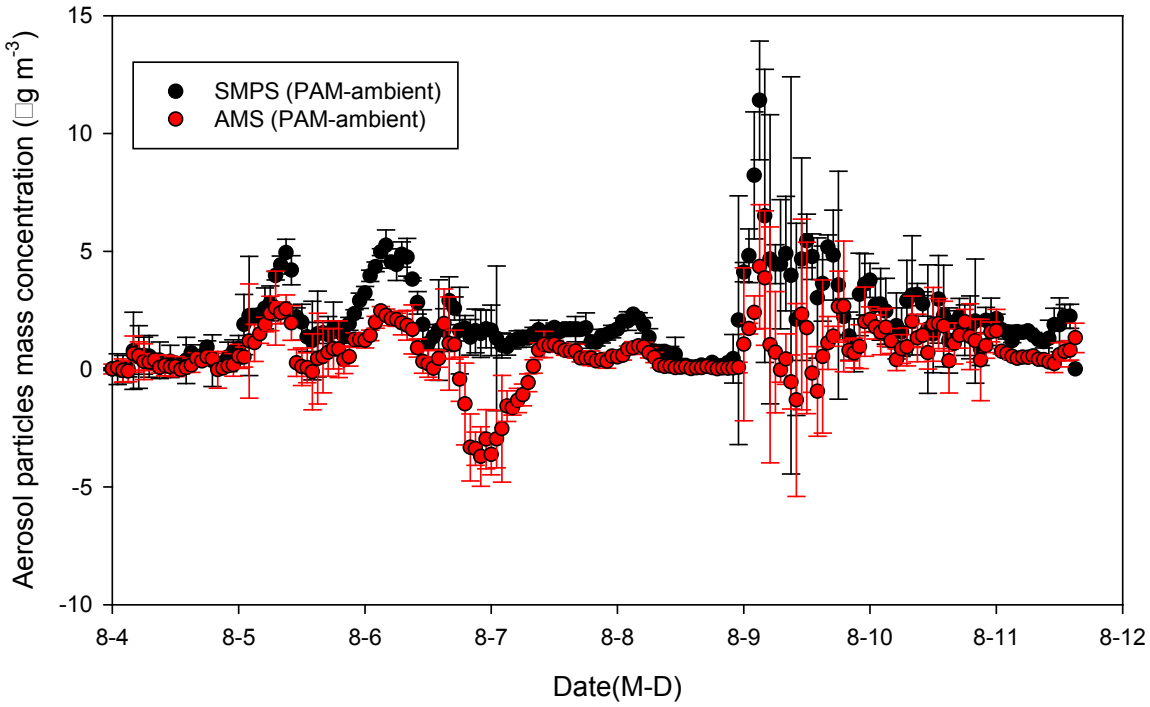
The plots are given below.

Compared are the difference in mass concentrations between the PAM aerosol and the ambient aerosol measured by AMS and SMPS. In general, the SMPS concentrations were equal to or slightly greater than those of AMS. However, when the ambient mass concentrations were high in the organics-dominated and the sulfate-dominated episode, the difference was visibly large. It might be related to your questions addressed above. the reason is not clear right now but it does not affect the conclusions.

(a)



(b)



[The difference in mass concentrations between the PAM aerosol and ambient aerosol

measured by SMPS and AMS. Data are averages for 6 minutes with 2σ confidence intervals. (a) The SMPS measurements were correlated with the AMS measurements. (b) Time series plots of the two measurements

- Responses p18: some text is quoted here (which I can't find on the revised manuscript): "L144, added the sentence. "Contrary to other studies (Ortega et al., 2016; Palm et al., 2016), we designed the experiment as simple as possible with a single OH exposure considering the dynamic change of meteorological conditions in the study region, a remote island."

This does not make sense. Time variations are slowest at remote sites such as the one in the current paper, and they are faster in forests and much faster in urban areas, due to much faster changes of emission sources and smaller spatial scales coupled with advection. So that should be revised (or just not included in the paper, which seems to be the case at present).

[Response]

Please see the line 150 in the revised manuscript.

In this study, the OH exposure of the PAM reactor was dependent on the humidity of the ambient air but not modulated by UV lamps. The exposure was originally set to 3-4 days for spring time to get near-maximum mass concentration. However, the experiment was delayed by logistic problem and the OH exposure was closer to 4.6 days due to high humidity in summer.

- Supp L 124: chlorine should be chloride

[Response]

It was changed.

- The text in many of the figures (axis labels etc.) are often very small, and will be hard to read in the final ACP version. I strongly suggest making them larger.

[Response]

We will enlarge the labels of figures at the final submission stage.