

Second Round Review of Kang et al. ACPD 2017

This paper describes results from an OFR - AMS/SMPS study at an island in Korea. The ACPD version had a large number of problems. The authors have addressed some of the problems in this revision, but have unfortunately ignored others. I would estimate that $\frac{1}{2}$ of the major problems have been addressed in this revision. However new errors come to light with the added information. Before acceptance into a journal with high standards like ACP, the paper requires another round of major revision, followed by re-review.

Major Issues

- (1) L156: a direct comparison (time series and scatter plot) of the AMS vs SMPS concentrations measured in this study is still not shown, despite being explicitly requested. This should be done for both the ambient measurements, and for the enhancements measured in PAM. This is a critical indicator of data quality, and in my opinion the paper should not be published without it.
- (2) L156: the statement is made in the responses (p8) that for the conditions of this study “CE would stay constant near 0.5.” However the next paragraph says “The composition dependent CE resulted in much better agreement between the mass measured by AMS and SMPS.” This makes no sense. First we are told that CE did not change, but then we are told that when applying a variable CE, the comparisons improved a lot? This makes the request for the direct comparison of SMPS and AMS mass (previous comment) even more important.
- (3) L203-204: this statement is erroneous. SVOC have saturation concentrations (C^*) up to 1000 $\mu\text{g m}^{-3}$, and IVOCs are in the range 10,000 to $1\text{e}6$ $\mu\text{g m}^{-3}$. IVOCs will not partition significantly to the particle phase under these conditions, and would require temperatures of -40C or so to start partitioning. Similar much of the SVOCs will be in the gas-phase, under the conditions of this study. Thus these statements are incorrect and should be revised. The implications of this revision should be propagated through the manuscript conclusions.
- (4) L136: The difference between the outside and PAM temperature is not discussed clearly in the revised manuscript. The first two paragraphs on p35 of the responses document this issue, and should be included in the revised paper. The conclusion that evaporation (or condensation, as the authors rightly point out could be an issue for an air-conditioned trailer) is “insignificant” is not warranted. A more detailed discussion is needed.
- (5) L137-144: was the calibration of OHexp used here and shown in p23 of the responses done on the same, or a different PAM reactor than the one used in this study. If a different reactor and lights were used for this study, the uncertainty on the OHexp increases considerably, being of at least a factor of 3. Whether the reactor was the same or different should be stated in the paper. If it was a different reactor, the increased uncertainty on the resulting OHexp should be discussed. (As an aside, if the Figure on p23 has not been previously published in the peer-reviewed literature, it should be included in the Supp. Info.).
- (6) The authors have calculated the condensation losses in the PAM. However they inexplicably use a single value for all their measurements, when e.g. their Figure 2 shows extremely large variations on PM concentrations, which will lead to dramatic changes in this correction. Since the authors have time-resolved SMPS measurements, implementing a time-dependent correction is easy and should be done here.

(7) Supp. Info., P1: the statement “We calculated the predicted sulfate formation with measured SO₂ concentrations, assuming that all SO₂(g) was converted to sulfate(p) by reaction with OH inside the PAM reactor” indicates that this analysis was not performed correctly. At an OH_{exp} of 4.6 equivalent days, the fraction of SO₂ consumed should be $1 - \exp(-4.6/8.1) = 40\%$ of the SO₂, not 100% of the SO₂. This seems like a major error, which when corrected will result in a slope of ~3 in Figure S1 (before correcting the other error discussed below).

(8) Fig S1: it is erroneous to remove the values with negative SO₄ enhancement. Seeing some negative values is normal due to measurement noise under low SO₂ concentrations. But removing these negative values will bias the regression analysis. Thus needs to be redone with those measurements included.

(9) The figure shown on p20 shows that the mass distribution peaked at the upper limit of the SMPS measurement, which is close to the upper limit for the AMS measurement as well. This indicates a substantial limitation of the measurement size range. Some changes in particles before and after the reactor may actually be due to this effect, for example if particles grow out of the size of the SMPS / AMS, the enhancement in mass will not be counted. This effect needs to be described in the experimental section, and its effects assessed.

(10) L113: the use of a PM1.0 cyclone upstream of an AMS/SMPS is very unusual. Both AMS and SMPS have approximate PM1 size cuts already, so normally a PM2.5 cyclone is used. Otherwise the PM1 cut is imposed twice, and results in PM0.75 transmission, and the concentrations will be lower than those measured without a PM1 cyclone. This should be mentioned in the revised manuscript. Referring to the AMS measurements as “PM1” is incorrect under these conditions.

(11) The conclusion in the abstract that “... SO₂ is an unquestionably primary precursor of secondary aerosol in northeast Asia. In comparison, the contribution of organics to secondary aerosols is more variable during transport in the atmosphere” is still not justified. It is clear (from this study and many others) that SO₂ is an important secondary precursor of secondary aerosols in this region. But the present study has limitations, in particular the use of a single very high OH_{exp} and the use of inlets, that limit its ability to say much about the relative importance of SO₂ and organic precursors contributing to secondary aerosols in the real atmosphere. This needs to be clearly acknowledged, emphasizing the strong result (SO₂ is important) without stating a conclusion for which there is no evidence from this study (SO₂ is more important than organic precursors).

Other Important Comments

(12) Experimental methods: the diameter, material, length, flow rate, and flow regime (laminar or turbulent) of all the inlet tubes is still not given in the manuscript. This should include the tube between the cyclone and PAM input, and the tube(s) between the PAM output and the AMS & SMPS measurements.

(13) P45 of the responses states “the particles we observed were quite likely to be internally mixed. Thus, it wouldn't be inappropriate to use the constant density and we would like to leave it as a further study.” This mixes apples and oranges. Internal mixing is indeed expected at remote sites like this one. But the density of the internally mixed particles will change substantially with composition. Estimating that density is literature one line of code, and given the limited time required, it does not seem acceptable to this reviewer to say that it is outside the scope of the manuscript.

(14) Page 6: the reason why a single OHexp was used is still not stated in the revised paper. There is a long text in the responses, which seems to boil down to the fact that it was too difficult to do so experimentally at the time. This can just be stated in the paper, and serves to inform others that it is not that such variation was not considered important, but that it was not possible to implement it for this study.

(15) Supp. Info. p.1. The statement that attributes the high scatter in Fig. S1 to “mainly because the comparison was made for hourly averaged concentrations of AMS even though measurement was alternately made every 6 minutes between the ambient and PAM air” makes no sense. Averaging the AMS measurement to the same time resolution as the SO₂ measurement should *reduce* scatter, not increase it. It appears instead that the scatter is dominated by the low accuracy and precision of the available SO₂ measurement. The erroneous statement should be removed, and this section should be revised.

(16) The figure shown on page 20 of the responses, showing the losses of particles through PAM when the lights were off, must be added to the Supp. Info., as it is an important characterization of the reactor that is needed in order to compare with other studies. At present the figure is shown in the responses, but is not included in the revised manuscript or Supp. Info. Figures that are left only in the responses are very difficult to find, as most readers will only look at the final published paper. Thus it is the responsibility of the authors to document all the important details on the paper itself, and not only in the responses.

(17) Similarly the figure about the SO₂ losses needs to be added to the Supp. Info. In addition, SO₂ losses in tubing and the PAM reactor are known to strongly depend on RH. So at a minimum the RH for the tests shown in the figure needs to be stated.

(18) L133: The flow rate of N₂ through the sleeves needs to be given. The temperature increase may not be the same as in past studies, if a different N₂ flow rate was used.

(19) L142: no reference or description is given for the “chemistry model” (discussed as “oxidation chemistry model” in the text quoted in the responses, although the word “oxidation” is missing from the revised manuscript). The references at the end of the sentence appear to refer to OH reactivity. A description or reference for the model should be given (ideally after the word “model” in that sentence).

(20) Figure S1: it is unclear whether an appropriate regression method has been used for this figure. Since both the X and Y variables have errors, it is not appropriate to use standard regression, which assumes that only the Y variable has errors. A method such as orthogonal distance regression should be used, and this should be stated in the paper. By eye it seems to me that an ODR fit would be significantly steeper than the one shown. The uncertainties on the fitting parameters that the fitting function should provide should also be given. (But overall this is not a bad comparison, given how poor the available SO₂ data were).

(21) The experimental setup shown on p36 of the responses should be added to the Supp. Info. This type of information is invaluable for future readers of the paper.

(22) L410: the Underwood et al. reference reports results for mineral dust. There are chemical pathways available for dust that are not active for the sulfate-organic-nitrate aerosols of the submicron mode, that are studied here. I recommend that this text and reference are removed to avoid creating confusion.

Minor items and Typos

L94: possible typo on Lamb

New Figure 6 has a typo on “organics” on the right side.