

Kang et al. present size- and chemically-resolved measurements of ambient aerosols from a Baengnyeong Island receptor site in South Korea. The measurements alternate sampling through a conventional inlet line and a PAM oxidation flow reactor to subject the ambient air to 5 days of simulated photochemical aging by OH radical. In general, passing ambient air through the reactor resulted in increased sulfate concentrations from SO₂ oxidation, and decreased organic and nitrate concentrations due to fragmentation/evaporation. The oxygen-to-carbon ratio of the organic aerosols increased from 0.6-0.8 to 1.0-1.2 depending on whether the air mass originated from China or the Korean Peninsula. Overall, I thought that the manuscript demonstrates a useful application of the flow reactor paired with particle instrumentation to examine effects of oxidative aging on the physical and chemical properties of ambient aerosols. There were a few sections that might benefit from further clarification or discussion. I would support publication in Atmospheric Chemistry and Physics after consideration of my comments below.

1. The authors never explain why the PAM reactor was only operated at a single OH exposure of 7×10^{11} molec cm⁻³ sec (5 days at [OH] = 1.5×10^6 cm⁻³). This condition should favor fragmentation reactions, resulting in the net decrease in organic concentrations that was observed. Why wasn't the reactor also operated at lower OH exposures that might favor functionalization reactions, as has been done in other field studies that used the PAM reactor? (see, for example: Tkacik et al., 2014; Ortega et al., 2016; Palm et al., 2016). Presumably, this would have given more information about the precursors contributing to SOA formation. Because air masses had already been processed for ~30-60 hrs by the time they reach Baengnyeong Island (Fig. 8), I can see why it wouldn't be necessary to survey as wide a range of conditions as the studies referenced above. But it isn't clear to me why more reactor operating conditions were not studied (or, if they were studied, why the results weren't presented).
2. What was the chloride concentration that was measured with the AMS?
3. **P6, L143-L146:** The authors state: "The difference in mass concentrations between the PAM aerosols and the ambient aerosols was in the range of ~3–7 μg m⁻³, indicating that photo-oxidation resulted either in the loss of pre-existing particles or in the formation of secondary aerosols." This statement seems to contradict the data presented in Table 1. For both the "organics-dominated" and "sulfate-dominated" episodes, the mean increase in mass from ambient to PAM-oxidized aerosols was +2.3 μg m⁻³ (14.0 to 16.3 and 24.3 to 26.6). Additionally, the net change in sulfate + nitrate+ ammonium + organics mass was -1.1 μg m⁻³ (8.0 to 6.9 and 11.2 to 10.1 μg m⁻³). So, how do the authors calculate that mass concentration increased by 3-7 μg m⁻³ following oxidation in the PAM reactor?
4. **P8, L206:** What is "non-linear oscillatory nucleation"?

5. **P10, L235-L236:** The authors state: “Thus, chemical transformation from low O:C to high O:C is more likely to explain the organic mass loss.” What is the change in temperature inside the reactor (with lamps on) relative to ambient temperature? How much of the decrease in organics can be explained by simple temperature-induced evaporation?
6. **P14, L343-L347:** The authors state: “The nitrate loss is rather explicit in the PAM reactor because of efficient conversion of SO₂ to sulfate, causing the aerosols to become acidic and causing particulate nitrate (HNO₃(p)) to evaporate.” How much of the decrease in nitrate can be explained by simple temperature-induced evaporation from the lamps being turned on in the reactor?
7. **Table 1 and elsewhere:** For the “organics-dominated” episode, the total mass of ambient sulfate + nitrate + ammonium + organics was 8.0 μg m⁻³, whereas the mass calculated from the SMPS size distributions was 14.0 μg m⁻³ (assuming density of 1.2 μg m⁻³). Then, for the “sulfate-dominated” episode, the total mass of ambient sulfate + nitrate + ammonium + organics was 11.2 μg m⁻³ compared to the mass of 24.3 μg m⁻³ calculated from SMPS. EC was typically ~1 μg m⁻³ (Fig. 4). Thus, in both cases the aerosol mass measured by AMS was ~50% of the mass calculated from SMPS. What collection efficiency was applied to the AMS data? Is it possible that a CE of 0.5 is approximately valid but was not applied? If not, a composition-dependent CE should be calculated and applied to the AMS mass concentrations that are presented. See, for example, Middlebrook et al., 2012.
8. **Table 1 and elsewhere:** The authors assume a density of 1.2 g cm⁻³. I think it would be better to calculate an effective density directly from the AMS and SMPS size distributions that would be more applicable to the data. See, for example, DeCarlo et al., 2004.
9. **Figure 3:** It is difficult to quantitatively interpret this figure. Instead of presenting this figure, please consider adding rows to Table 1: Mass (10-50 nm), Mass (50-200 nm), Mass (200- 400 nm) for all “ambient” and “PAM” columns during the organic and sulfate episodes.
10. **Figure 4:** The authors mention PM₁₀ mass measurements (P6, L131) but as far as I can tell do not present the results – please add time series here and discuss briefly in the text.
11. **Figure 5:** Why/how is there organic signal at m/z = 32?
12. **Figure 6:** The authors might consider smoothing some of the size distributions, especially for ammonium and Organics/Org43/Org44 during the sulfate episode. Also, is there sufficient signal-to-noise to resolve chloride size distributions?

References

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