

Atmos. Chem. Phys. Discuss., referee comment RC1
<https://doi.org/10.5194/acp-2022-838-RC1>, 2023
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Anonymous review of "Secondary Aerosol Formation in Incense Burning Particles by Ozonolysis and Photochemical Oxidation" for Atmos. Chem. Phys.

Anonymous Referee #1

Referee comment on "Secondary Aerosol Formation in Incense Burning Particles by Ozonolysis and Photochemical Oxidation via Single Particle Mixing State Analysis" by Zhancong Liang et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-838-RC1>, 2023

General Comments

Liang et al. (manuscript) describes the experimental formation of SOA and nitrate through chemical aging induced in an oxidation flow reactor (OFR). The authors injected combustion air from incense burning into the OFR at high RH, which were rapidly aged by controlling UV light and O₃ to mimic UV-aged, O₃-aged, and OH-aged scenarios. The authors used a single-particle AMS (SPAMS) to obtain the chemical composition of the particles and the Gothenburg PAM OFR (Go:PAM) as the reaction vessel.

The authors use the adaptive resonance theory method (ART-2a) algorithm to perform cluster/categorization analysis with the mass spectra (Zhao et al., 2008). The authors conclude that the OH-aged case generates more secondary nitrate than the O₃-aged case based on the higher relative peak area (RPA). The enhanced secondary nitrate formation is attributed to higher uptake of nitrogen-containing species.

The manuscript overall lacks quantitative information, and I am confused about the OFR configuration. The authors provide some [NO_x] information in Figure S10 and in the text, but a NO_x instrument is not shown in the OFR set up in Figure S1. Moreover, the flow rates entering and exiting the OFR in Figure S1 already match, so adding a NO_x monitor would cause a flow imbalance.

Moreover, the methodology and instrument details are lacking for replication, and additional explanations are needed to connect the chemistry in these OFR conditions to those of the atmosphere. For instance, the manuscript is missing the Go:PAM temperature and experiment residence times. The generated particle number concentrations from the

WCPC, the particle mass collected on the filters for IC analysis, and the mass of incense used are not available. I do not have a clear picture of how much aerosol entered and exited the OFR.

The experiments and discussions fall within the scope of *Atmos. Chem. Phys.*, and the content is topical to the atmospheric chemistry community. However, the manuscript is currently underprepared for publication, and there are technical issues that need resolution. Given the importance of understanding how chemical aging impacts aerosol evolution, I request the authors perform major revisions and resubmit the manuscript.

Specific Comments

1. The SPAMS calibration is not outlined, and details on the ART-2a solution is inadequate. Details on the SPAMS operation would help assess the data quality. The Aerodyne soot particle AMS requires laser alignments for consistent measurements (Avery et al., 2020); does the SPAMS in this manuscript need a similar calibration? Particle transmission through the aerodynamic lens is size-dependent (Huang et al., 2013). How would size-dependent particle detection influence the data interpretation?

The absolute peak area (APA), relative peak area RPA, and number fraction (NF) are frequently invoked in the data interpretation. The authors use APA and RPA as analogues to concentration (or fraction of total aerosol). However, I suspect that depends on how efficiently different species are ionized by the pulsed Nd:YAG laser, and I would like to know if adjustments have been made to the RPA based on the ionization efficiency. What is the ionization efficiency (IE) of species mentioned in the manuscript, and is IE consistent across species?

As for the ART-2a solution, Zhao et al. (2008) and Huang et al. (2013) note that there is no general rule for the vigilance factor, and that a comparative approach (like re-grouping or comparing with other clustering algorithms) may be needed. I also note there is no PAH category, despite particulate PAH found in previous incense combustion studies (Ji et al., 2010) and a PAH contribution being found in a similar mass spectrometer with ART-2a (Passig et al., 2022). Can the authors provide more detail and justification for their ART-2a solution?

2. OFR characterization and operation details are missing. Offline OHexp calibrations may be inaccurate when OH reactive species suppressed OH. Basically, OH suppression is when the external OH reactivity (extOHR) entering the OFR is high enough to titrate the OH, which results in OH-aging being suppressed. In such scenarios, offline OHexp calibrations become unreliable, possibly by orders of magnitude

(see Section 3.1.4 of Peng and Jimenez, 2020). Peng and Jimenez (2020) also notes that OFR254 is susceptible to OH suppression at low O₃ injections.

Operational information of the OFR would be valuable for replication and should be mentioned in the supplementary. Watne (2018) describes the Go:PAM as being made of quartz; have there been efforts to constrain electrostatic particle wall loss (Cao et al, 2020)? How would gas wall loss (Palm et al., 2016) affect the results reported, or is gas wall loss negligible? What cleaning procedure was taken to minimize carryover effects between experiments?

3. Kinetic modeling may be needed for interpretation.

The authors' argument on secondary nitrate formation, either heterogeneously or in the gas phase is limited by the lack of quantification HNO₃, HONO, NO_x, NO_y etc. The difference in condensed nitrate between the O₃ and OH-aged cases may be due to differences in HNO₃/HONO/NO_x uptake as the authors allege. A kinetic calculation showing that the formation of these species under the difference OFR conditions are comparable would be more demonstrative.

Moreover, gas-phase organic nitrate formation, either through VOC+NO₃ or RO₂+NO (Ziemann and Atkinson, 2012) and condensation should be considered. Kinetic modeling may be needed to connect the experimental aging conditions in the Go:PAM to those of the atmosphere (Peng and Jimenez, 2020).

Line 30: I recall incense burning is found in other cultures and am unsure if the practice is "especially" common in Asian and African religious rituals. I suggest either providing a reference for that point or removing "especially" in this sentence.

Line 35: The incense burning references cited here mention that there is variation in the particle emission factor (EF) across incense varieties. How does the particle EF in these experiments compare with those previous works? Were the combustion conditions comparable to those previous works?

Line 62: There is no information on the incense sticks used, like the manufacturer or composition, and Liang et al. (2022) used several as shown in their Figure S21. What type of incense was used here? How much incense was burned? This information could be valuable for replication studies.

Line 64: The methods reference (Liang et al., 2022) states there were four UVA lamps, while here the authors say they used "two UVC light tubes." Please confirm that the Go-PAM set up had changed for this manuscript and specify that in the text. Moreover, what lamps were used? Rowe et al. (2020) found that 185 and 254 nm photon fluxes would vary across manufacturers, which may be re

Line 65: I am confused on how many experiments this manuscript is describing. I see in Figure 2 that there were 7 involving aging and 1 fresh; were some of these the "control" experiments? From this sentence I expect at least 2 types of controls, with either a charcoal absorber or HEPA filter. Were the control cases then also aged?

Line 66: How did the authors obtain these removal efficiencies?

Line 67: Compressed air or zero air? If the air is coming from a compressor, were efforts made to scrub the air of contaminants?

Line 74: The methods reference (Liang et al., 2022) does not mention using a diffusion dryer. At ~0.1 LPM, what was the residence time in the dryer and is there an estimate of particle loss in the dryer? Was the dryer effective in removing H₂O?

Line 76: What were the estimated number of particles collected?

Line 83: Please provide additional details on the OH_{exp} calibration with SO₂, in particular the concentrations of SO₂ used and timescales to reach equilibrium. An estimate of extOHR during the experiments should be compared with that of the SO₂ calibrations.

Line 101: See specific comment 1.

Line 113: Explaining the abbreviations would improve readability. For instance, OC and ONEC do not appear prior in the text?

Line 147-149: How does the charcoal absorber remove NO_x without removing VOC? How would the removal of VOC affect the interpretation here?

Line 156: How would the loss of SVOC/LVOC in the HEPA filter (Shilling, 1997) affect the conclusion of the control experiment?

Line 182: How did the authors arrive at the "~90 % reduction of [NO_x]?" Was this a separate test? If so, please add a quick summary of how that test was performed.

Line 183: Was a NO_x monitor available? If so, please provide the monitor's location in Figure S1 and specify in the text. Also, please explain how the flow rates entering and exiting the Go:PAM would be reconciled.

Line 191-193: Do OH and O₃ oxidation form similar functional groups? Are those functionalities evenly represented in these general markers? The RPA increase of SOA markers in OH over O₃-aging may be skewed if these markers overrepresent one oxidation case over the other.

Line 214: Do larger and smaller particles have similar surface properties, at least with regards to nitrate uptake?

Line 222: I suspect SOA formation is not "potential," but rather inevitable under the aforementioned OFR conditions, so I suggest removing "potential" from the section heading. Also, are oxalate and malonate universal and proportional indicators of SOA? That is, do different SOA precursors form these indicators evenly under different oxidants (O₃/OH)? I am concerned that there are specific chemical conditions where these species are enhanced without a proportional enhancement of SOA, which may skew the NF.

Line 229: Is NF of oxalate and malonate proportional with SOA concentrations? As it stands, "30 and 9 folds" increases of these tracers sounds like SOA increased by that much.

Technical Comments

Line 47: Awkward grammar in "For instance ... nitrate."

Line 325: Please check the citation styles; they are inconsistent.

Lines 409/412: Same reference cited twice?

Figure 2: The data appears almost randomly scattered in the lower panels, which may be due to points heavily overlapping with each other. The authors may want to replace the box and whiskers/scatterplot with a violin plot for easier visualization.

Figure S1: Please display where the charcoal absorber, HEPA filter, and NOx instrument would have been placed.

References

- Avery et al. (2020): <https://doi.org/10.1080/02786826.2020.1844132>
Cao et al. (2020): <https://doi.org/10.1016/j.atmosenv.2019.117086>
Huang et al. (2013): <https://doi.org/10.1016/j.atmosenv.2012.09.044>
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