

Response to Reviewer 2:

General Comments

This article describes a series of experiments used to explore the aging of incense particles in the presence of different oxidants by applying a flow reactor and a single particle mass spectrometer. The major conclusions are that nitrate formation on/in incense burning particles is enhanced in the presence of OH radicals relative to dark ozone oxidation and that oxalate and malonate formation is also enhanced in the presence of OH. While the experiments are interesting the main text and conclusions need more clarity around the actual implications of these findings for (presumably both indoor and outdoor) air quality and exposure.

Author's Response: Thank you for the constructive comments. We have revised the manuscript accordingly and added clarifications to the main text and conclusions around the implications of the findings.

Major comments

Section 3.2 is hard to follow and a clearer explanation and validation of the use of RPA and APA to draw conclusions is needed. A relatively low laser fluence was used in this work and partial ablation/matrix effects can be problematic for particles with secondary coatings (see Zelenyuk group research), which could lead to misclassification of particles based on ion intensities. The appearance of the nitrate-rich particle types after aging is quite conclusive though. Relative ionization/detection efficiencies for the different particle types are not discussed however, which would have a large impact on the relative particle type abundances shown in Figure 1. Some pure secondary organic particles (which may be externally mixed with primary particles in the reactor) for example are just not ionized effectively at all by SPMS. Some discussion of the drawbacks of SPMS are needed in the text.

Author's Response: A control experiment doubling the laser fluence from 0.6 to 1.2 mJ showed minor differences in the classification of the aged particles and the RPA of total nitrate peaks (Table S1). This suggests that the partial ablation due to the formation of nitrate coating is not significant. It is possible that there were some pure secondary organic particles still not ionized at 1.2 mJ, which resulted in an underestimation of secondary organic aerosol formation. We have added this information to the manuscript.

Table S1. The classification of particles and RPA of total nitrate peaks of aged particles at different laser fluences.

Experimental Conditions	Laser fluence (mJ)	OC-ON (%)	OC-N (%)	K-ON (%)	K-ONN (%)	K-N (%)	K-ONEC (%)	K-Cl (%)	RPA _{Total} nitrate
O ₃ -dark-aged (800 ppb)	0.6	(7.6±1.0)	(21.9±1.7)	(29.0±0.7)	(11.2±0.9)	(2.5±0.5)	(22.8±1.8)	(3.9±0.4)	(0.40±0.15)
	1.2	(7.2±1.4)	(22.6±4.0)	(28.5±4.3)	(10.6±0.2)	(3.3±1.3)	(22.8±2.5)	(4.0±0.1)	(0.37±0.09)
O ₃ -UV-aged (800 ppb)	0.6	(0.0±0.0)	(36.7±7.2)	(0.0±0.0)	(26.7±2.1)	(35.5±4.2)	(0.0±0.0)	(0.0±0.0)	(0.66±0.03)
	1.2	(0.0±0.0)	(35.6±2.3)	(0.0±1.0)	(28.6±1.9)	(33.6±4.7)	(0.2±0.2)	(0.0±0.0)	(0.67±0.07)

Line 229-232: A control experiment doubling the laser fluence from 0.6 to 1.2 mJ showed minor differences in the classification of the aged particles and the RPA of total nitrate peaks (Table S1). This suggested that the partial ablation due to the formation of nitrate coating is not significant. It is possible that there were some pure secondary organic particles still not ionized at 1.2 mJ, which resulted in an underestimation of secondary organic aerosol formation.

Where is the NO_x added? Or is it just the NO_x from the combustion process. How were the NO_x concentrations monitored or validated to be similar between experiments?

Author's Response: The NO_x was from the combustion process. We have added that the location of NO_x monitor to the schematic figure (Figure S1).

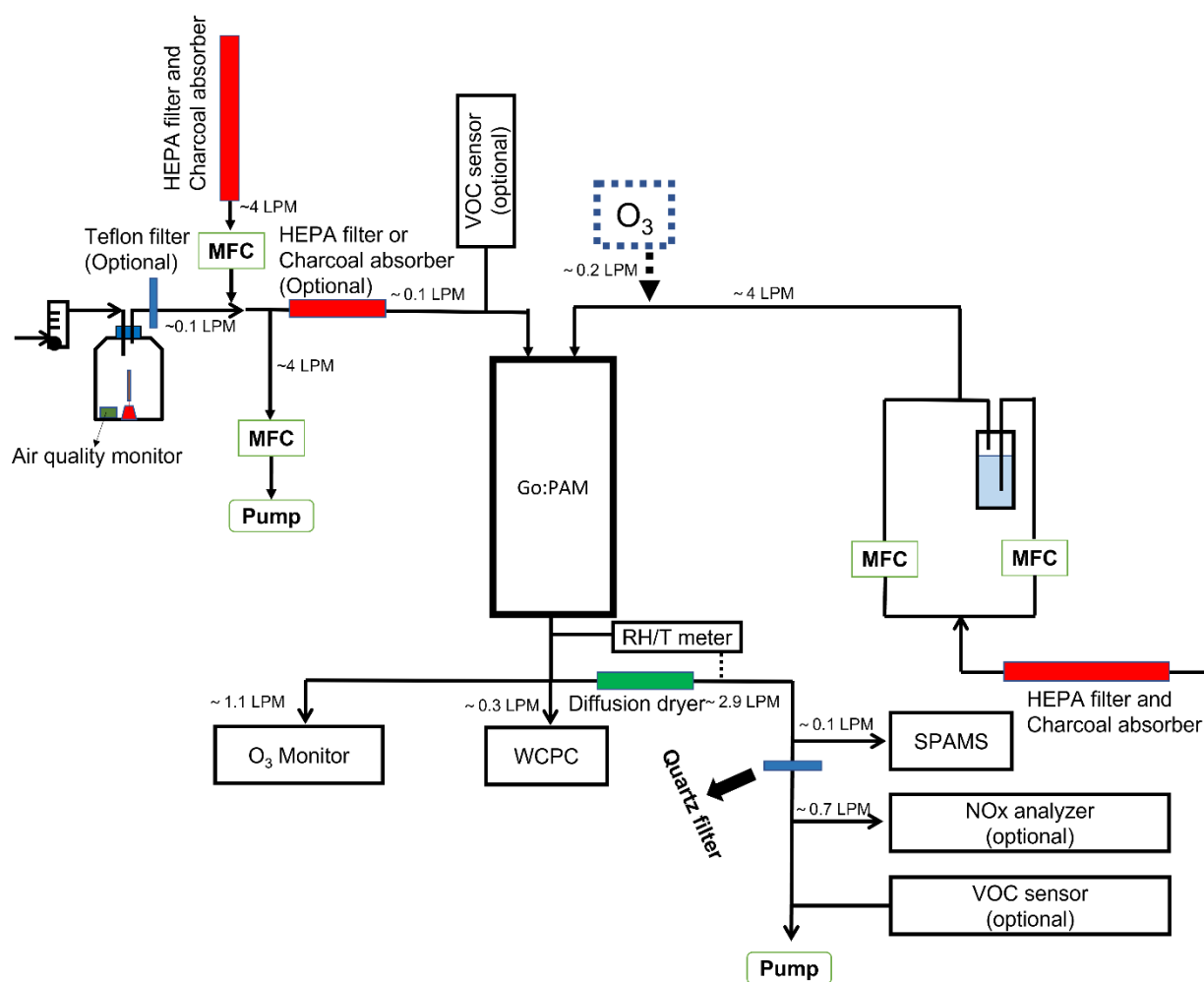


Figure S1. The schematic of the experimental set-up. The NO_x analyzer and the VOC sensor were used only in the experiment for determining the gas removal efficiency and NO_x decay under OH exposure.

Figure S1 is missing the scrubber mentioned in the text.

Author's Response: We have added the scrubber location to Figure S1.

Why are most of the figures in the supplementary information? There are only 3 main text figures and a lot of reference to the supplementary figures.

Author's Response: The main text figures are combined ones, converting the key messages of the manuscript. The supplementary figures play minor roles. The key results are shown in the manuscript.

The manuscript is hard to follow in places, including the conclusions. A clear central message needs to be stated in the conclusions, with implications for air quality or public health.

Author's Response: We have added a central message and implications.

Line 292-295: In this work, we report the single-particle mixing state characteristics of incense burning particles upon ozonolysis and photochemical oxidation. Formation of secondary aerosol including nitrate and organics was found. This indicates that besides the significant primary emission of particles, additional particulate pollutants could be formed upon atmospheric aging, further worsening the air quality in both outdoor and indoor environments.

No correlation (mentioned in the caption) is shown in Figure S4. Same for S6

Author's Response: We have corrected the captions.

Figure S7. APA of ON as a function of APA of total nitrate in fresh and O₃-aged particles.

Figure S9. RPA of total nitrate as a function of RPA of formate.

Why was no support analysis done (especially ion chromatography for the oxalate/malonate yields) to rule out potential matrix-effect artifacts. Quantification (even semi-quantification) exclusively using SPMS is associated with high uncertainty.

Author's Response: We have added TOC analysis to the water-extract of collected particles. While the TOC ratio at different O₃+UV shows an overall trend similar to the NF ratios of oxalate and malonate, which indicates the formation of SOA, the TOC ratio was 1.2-7.1 folds higher than the NF ratios of oxalate and malonate (Figure S15). However, this could be due to the formation of many other species as well as the matrix effects. We hesitate to speculate too much here. SOA formation during aging of incense burning plume should be further assessed by other quantitative online instruments in future works. We have added this clarification to the manuscript:

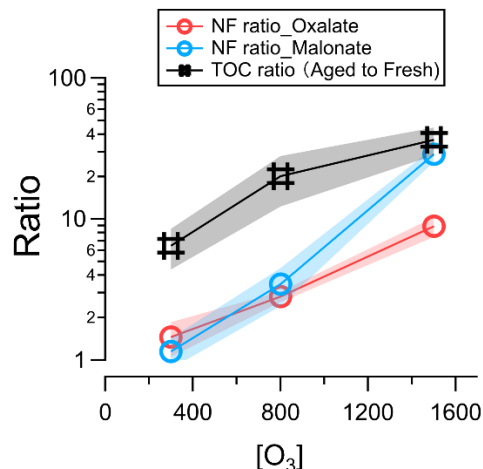


Figure S15. The NF ratios (aged to fresh) of oxalate and malonate, and TOC ratios as a function of [O₃] in O₃+UV aging experiments.

Line 264-268: While the TOC ratio at different O₃+UV shows an overall trend similar to the NF ratios of oxalate and malonate, which indicates the formation of SOA, the TOC ratio was 1.2-7.1 folds higher than the NF ratios of oxalate and malonate (Figure S15). However, this could be due to the formation of many other species as well as the matrix effects. SOA formation during aging of incense burning plume should be further assessed by other quantitative online instruments in future work.

Specific comments

Abstract- remove (photo-) because dark oxidation is also investigated

Author's Response: The (photo-) has been removed.

Page 2 line 58, missing 'tracers'

Author's Response: We have removed the empty row line 58.

"We studied the aging of the particles under 'UV', 'O₃ and dark', and 'O₃ and UV' in the PAM. Since UV at 254 nm is expected to photolyze O₃ to form OH radicals in the presence of water vapor, we named these aged particles UV-aged, O₃-aged, and OH-aged, respectively." Why not: 'UV-aged', 'O₃-dark-aged' 'O₃-UV-aged' for clarity

Author's Response: We have replaced all 'O₃-aged' and 'OH-aged' with 'O₃-dark-aged' and 'O₃-UV-aged'.

Where are the nitrate peaks in Figure 1b?

Author's Response: Figure 1b showed the 'organic difference spectra' with EC and inorganic removed. We have further clarified the contents of the figure.

Line 152-157: To compare the changes in the organic signals, we first excluded all inorganics and EC peaks (Table S2). Control experiments atomizing KNO₃ solution (as K⁺ is the main inorganic cation found in incense burning particles) showed the RPA ratio of -16[O] to nitrate peaks is (6 ± 1.7) % due to fragmentation. Sulfate shows negligible fragmentation under our experimental conditions (Liang et al., 2022). Thus, we subtracted the RPA of -16[O] by 6% RPA of nitrate. Then, we recalculated the relative peak area (RPA) of the organic peaks only, defined as "organic spectra". Figure 1b shows the differences in the organic spectra of the aged and fresh particles.

"The instrument 98 was routinely calibrated with polystyrene latex spheres of 0.2-2.5 μm diameter (Nanosphere Size Standards, Duke Scientific 99 Corp., Palo Alto)" It should be specified that this only calibrates the sizing accuracy, not chemical species. "and more than 98% of the particles were analyzed" should be "were classified"

Author's Response: We have added clarification and revised the text:

Line 110-112: The size accuracy of the instrument was routinely calibrated with polystyrene latex spheres of 0.2-2.5 μm diameter (Nanosphere Size Standards, Duke Scientific Corp., Palo Alto).

Line 114: more than 98% of the particles were classified.

"K-" particles contain a dominant +39 peak and a small +41 peak attributed to isotopic 116 potassium (Bi et al., 2011). On the other hand, the "OC-" particles are rich in typical organic fragments such as +27[C₂H₃] (Cheng et al., 2017). According to the negative spectra, "-ON" particles have dominant ON signals. "-ONEC" particles have elemental carbon (EC) peaks of -12n[C_n -], with intensities comparable to typical ON peaks (Zhou et al., 2020). "-Cl" particles have prominent Cl⁻ (m/z=-35, -37(isotopic)) and KCl₂ - (m/z=-109, -111(isotopic)) peaks (Bi et al., 2011)." These are not the correct references for the original identification of these peaks by single particle mass spectrometry. See Prather group research.

Author's Response: We have replaced the reference with those for original identification.

Line 135-139: "K-" particles contain a dominant +39 peak and a small +41 peak attributed to isotopic potassium (Silva et al., 1999). On the other hand, the "OC-" particles are rich in typical organic fragments such as +27[C₂H₃] (Silva et al., 2000). According to the negative spectra, "-ON" particles have dominant ON signals. "-ONEC" particles have elemental carbon (EC) peaks of -12n[C_n⁻], with intensities comparable to typical ON peaks (Whiteaker et al., 2002). "-Cl" particles have prominent Cl⁻ (m/z=-35, -37(isotopic)) and KCl₂⁻ (m/z=-109, -111(isotopic)) peaks (Guazzotti et al., 2001; Dall'osto et al., 2004)."

“except for the rise of -62[NO₃⁻] and -46[NO₂⁻], which indicates the formation of nitrate and probably nitrite” Note that -46 is observed from nitrate and can't be used to confirm nitrite

Author's Response: We agreed that -62[NO₃⁻] can be fragmented into -46[NO₂⁻], but we can not exclude the existence of nitrite. Therefore, we used 'probably' for conservation.

“However, the RPA shows an opposite trend, which can be interpreted as lower nitrate concentration in larger particles. Larger particles have larger surfaces but smaller surface-to-volume ratios, which lead to a larger absolute amount of nitrate formed but a lower relative concentration of particulate nitrate (Figure 2c, e). Under O₃+UV, it is also possible that comparable HNO₃ was generated under excess [OH] and contributed to the similar total nitrate RPA since the [NO_x] reductions under different OH exposure are similarly high (Figure S10). The insensitivity of nitrate formation to O₃ and OH exposure can be potentially explained by the diffusion limitation of interfacial uptake due to the poor hygroscopicity of fresh incense burning particles (Li and Hopke, 1993; Zaveri et al., 2018; Slade and Knopf, 2014; 220 Liang et al., 2022a)”

Caution should be taken interpreting the RPAs due to the potential for partial ablation and matrix effects.

Author's Response: Control experiments using different laser fluence indicate that partial ablation is insignificant. We have further clarified that the RPA is not equivalent to concentration due to the matrix effect.

Line 119-122: The relative peak area (RPA), defined as the peak area of a specific peak divided by the total positive or negative mass spectral peak area, can reflect the relative abundance of particulate components (Liang et al., 2022). Note that RPA is not equivalent to concentration due to the matrix effect. Furthermore, control experiments using different laser fluences indicate that partial ablation was insignificant (Table S1).

“Figure 3a shows the NF ratio (aged particles to fresh particles) of oxalate and malonate. We used the NF ratio rather than the APA or RPA, to avoid large uncertainties in organic abundance due to the much weaker peaks of organics in the spectra.” For the number fraction is there a query using a minimum APA or RPA for oxalate or malonate to be classified as a count? Furthermore, the point made here does raise the issue with using APAs/RPAs at all considering the potential for matrix effects.

Author's Response: The APA threshold for oxalate or malonate was 15 arbitrary unit (a.u.) with a baseline of zero unit, the same as that used in ambient measurements (Zhu et al., 2020). We have clarified this and mentioned the potential matrix effects.

Line 256-259: We used the NF ratio rather than the APA or RPA to avoid large uncertainties in organic abundance due to the much weaker peaks of organics in the spectra, which was potentially

due to the matrix effects. The APA threshold for oxalate or malonate was 15 arbitrary unit (a.u.) with a baseline of zero unit, the same as that adopted in an ambient study (Zhu et al., 2020).

References

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