

Interactive comment on “Secondary organic aerosol formation from in situ OH, O₃, and NO₃ oxidation of ambient forest air in an oxidation flow reactor” by Brett B. Palm et al.

Anonymous Referee #2

Received and published: 21 March 2017

This work describes the first field observations of in-situ OH, O₃, and NO₃ exposures to ambient air using an oxidative flow reactor. This is highly important work in the field of atmospheric chemistry today, with extensive field and lab studies being performed to better understand the chemical mechanisms and potential to form (or fragment) secondary organic aerosol. Observations here are conducted in a forested environment with biogenic precursor gases (monoterpene dominant) and highlight the dominance of OH oxidation chemistry, but show potential for O₃ and NO₃ reactions with C=C bond VOC species at night. Several studies have been performed using a similar method since the 2011 BEACON-RoMBAS study described here, making the analysis and results of this study very relevant for upcoming manuscripts for this research team and

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others. In-situ NO₃ chemistry and modeling is especially novel.

Specific comments to be addressed:

Pg. 4, Line 22: Discussing MT's here, but haven't defined how these are measured, if cumulative MT's by PTR, or summed by GC/MS.

Pg. 5, Line 5: Please provide average concentration increases for "moderate increases" of NO_x, CO, and anthro VOCs. Also, what anthro VOCs?

Pg. 5, Line 20: Are periods with very high local winds excluded from the analysis?

Pg. 6: The thorough explanation of NO₃ exposure estimates here and in supplemental material is appreciated. It seems worth considering how representative one equivalent day of NO₃ aging would be of atmospheric conditions. Given the typical diel pattern of NO₃, and relatively low concentrations, would it ever be expected that a whole day's worth of oxidation could occur prior to further oxidation from OH?

Pg.8, Line 22: Can further argument be provided for the assumption in this modeled correction (of no fragmentation for O₃ or NO₃ reaction LVOC products)? I'm wondering to what extent does the assumption drive conclusions? Figure 5 suggests lower OA concentrations at 2-3 days NO₃ eq. aging compared to 1 day eq. aging.

Pg. 9, line 1: The acronym for sesquiterpene (SQT) has not yet been defined.

Pg. 10, line 15: The negative values in Figure 2d for the fraction of monoterpenes reacted, along with the instances of OFR output MT concentrations that exceed ambient levels shown in Figure S7, should be mentioned. Can this be attributed to instrument uncertainty, or are there other factors at play that give these apparent MT generation events?

Pg. 10, line 24: Change "didn't" to "did not".

Pg. 12, Line 6: please provide average daytime MT+SQT concentration and average nighttime MT+SQT concentration here.

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Pg. 12, line 19: In Figure 6, there is an uptick in OA enhancement with the highest level of O₃ oxidation for the nighttime air. However, in Figure S7 it appears that the MTs are largely depleted prior to reaching this extent of aging. Would this suggest that something beyond the measured monoterpenes is contributing to SOA formation from O₃ oxidation at these highest levels of aging?

Pg. 13, Line 11: abstract says factor of 3.4. Here it states factor of 4.4. Are these numbers referring to the same discrepancy?

Pg. 18, line 24: Change “formed from primary VOCs” to “formed from reaction with primary VOCs”.

Pg. 20, line 13: Please explain further where 620 g mol⁻¹ is coming from.

Figure 5: This method of binning seems to limit comparison of low and high monoterpene conditions at the same levels of oxidation. Particularly for NO₃, why are there not average values for the high monoterpene case at high levels of NO₃ eq. age?

Supplemental Information

Fig S3: Should reiterate in figure caption that these fractional fates are modeled, not measured. Additionally, it seems that the fraction of LVOCs condensing on the aerosol will decrease slightly at higher NO₃ exposures. Would this be due to a greater frequency of fragmentation reactions occurring as opposed to functionalization?

Fig S6: why higher NO₃ exposures on the limited data points on Aug9-10?

Fig S8: Which quantile averages are being shown by the black trace?

Table S2: revisit for formatting

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