Review of Bell et al ACP 2021

This paper presents a new set of chamber experiments exploring the condensed-phase dark aging of NO3 + α-pinene SOA, which chemically speciated observations (EESI, FIGAERO-CIMS) illuminating the continued oxidation of the SOA. The analysis could be expanded to increase the impact of this paper, and I recommend additional analysis be included in the main body of the paper.

Major comments/suggestions:
1) The authors use the MCM model to determine the most likely RO2 bimolecular reaction partner, and then note that the RO2+RO2 product channel is apparently more dominant based on observed products. Based on this empirical observation, could you use the model to infer what the RO2+RO2 rate constant must be, for these C10 nitrate functionalized RO2’s? It seems to me this is an opportunity your data give you that should be exploited! Suggest to include an additional section on modeling RO2 fate.
2) I don’t understand the claim that peroxy nitrates are controlled by RO2 + NO3 reactions. I think they would be controlled by nitrato-RO2 + NO2 reactions, which makes one of your ideas about oxidant sources less sound.
3) You discuss fragmentation as yielding only CH2O as volatile fragments, but the loss of organonitrates that cannot be explained by hydrolysis to HNO3 suggests that there must also be some fragmentation to high-volatility organonitrates. This would be good to elaborate upon. Would these be detectable in any of your gas-phase measurements?
4) Could semivolatiles be repartitioning differentially to the chamber walls? Your discussion of wall losses seems to assume a consistent loss rate for all species, but these rates could be species-dependent. Could the apparent loss of O8 be due to greater wall repartitioning for that molecule than for higher-oxidized, heavier molecules? I suggest thinking about the speciated wall partitioning discussed in Krechmer et al. 2020 (https://pubs.acs.org/doi/abs/10.1021/acs.est.0c03381)
5) Sections 3.2 and 3.3 have the same title
6) Around line 155: can dimers also form in the EESI, in the reverse of the fragmentation you discuss?

Minor / technical points:
1) Line 11 “in or downwind of polluted”
2) Line 15: “in the absence of external stimuli” feels a bit vague to me -- what you really mean is aging in the dark, right? This phrase is also used later. Consider rewording? But this is a style choice, so just a suggestion to think about
3) Line 29: ‘Unlike isoprene, monoterpenes are emitted”
4) Line 30: “and NO3) play an important”
5) Line 50: “still not well understood”
6) Line 59: “determine the absolute scale” is unclear to me. Perhaps something like “determine the magnitude of the effect”?
7) Section 2.1: find a place here to mention that these experiments were run in batch mode; also suggest to spell out EESI and FIGAERO here too (even though they came also in the abstract), since you spell out other acronyms in this section

8) Line 67: which instrument is the thermo-denuder in front of?

9) Line 75: indicate the approximate concentration of the cresol contaminant?

10) Line 83: “cresol contamination constitute ~1-2%”

11) Line 86: “experiments 1 - 3, ~100 ppb”

12) Table 1 caption: I suggest to add some rate constant modeling info to this caption: “based on a fit to the VOC decay, assuming XXX as the NO3 + apin rate constant at XX C”

13) Eq 1 formatting: need a space between equation and label

14) Line 117: “sampling the aerosol at 1 L min^{-1}”

15) Line 122 “ion guides and were separated” -- in general, check for verb tense consistency: most are past tense, but some a present tense.

16) Line 127: “subtracting the background filter periods from the adjacent chamber sampling periods. A filtering”

17) Around line 132: this equation needs a number. There is some repeated text before and after “Avogardo’s number and a conversion factor…” - remove one

18) Line 135 “including particle size,”

19) Line 140: “and had low sensitivity”

20) Line 143: give formula for acetonitrile at its first instance, to help reader interpret later clusters you mention

21) Line 146; “ for all C20H32N2Ox molecules” -- if this is in fact what you mean?

22) Line 175: “Tofware”

23) Line 177: “For the first filter in Exp. 2, due to a software failure, the filter was stored wrapped in aluminum foil for ~7 H after … was done prior to desorption.”

24) Line 178: “ were similar to other experiments.”

25) Line 187: “consumed, because the injection of N2O5 was less”

26) Line 199: “ observations (Takeuchi and Ng, 2019), and with RO2+ RO2 reactions.”

27) Line 201: “dimers over monomers”

28) Around line 206: I don’t understand how peroxynitrates would be formed from RO2 + NO3 reactions. WOuld’n they be formed by nitrato-RO2 + NO2 reactions?

29) Lines 222-224: The first sentence of this section doesn’t make sense to me, sharpen / reword / make more specific?

30) Line 226; “larger SOA yields were observed under an RO2 + RO2 dominant”

31) …. Rest of section: make every instance consistently “RO2 + X” - currently some have the + sign and some have long dashes --

32) Line 236: “HO2 is not an important RO2 reaction partner since there”

33) Line 240:” peroxy linkage. This molecule is the dominant dimer”

34) Around line 257: See comment above about expanding interpretation of RO2 + RO2 rate constant based on your observations.

35) Line 261; “and the FIGAERO-CIMS, were used to”

36) Line 265 spelling FIGAERO
37) Line 282: units “ag s-1 h-1” don’t make sense to me. Per second and per hour? (on next line too)
38) Line 312: spurious comma at the end of the line
39) Line 331: reorder confusing sentence: “Approximately half of the total depletion observed arises from an increase in oxidation, with the remainder coming from evaporative losses.”
40) Line 343-345: this last line of the paragraph, about no specific loss of -ONO2 groups, is confusing to me.
41) Line 361: “initiated from the scission of O-O bonds in organic peroxides”
42) Next line: as mentioned above, I don’t see why PANs concentration would be drive by [NO3] (rather, I would expect a dependence on [NO2])
43) Around line 371-372: could the same N2O5 measurement be an artefact / wall background?
44) Line 373: “phase or organic peroxides could be”
45) Line 380: remove “making up the difference”
46) Line 384: “in the gas phase, dimers will”
47) Around line 387: doesn’t this say fragmentation isn’t just CH2O?
48) Line 389: ‘organic peroxides or from”
49) Line 390: “presented here, along with… ), demonstrate that”
50) Line 397: “Overall, particle-phase … regime, since .. is always an important sink of RO2”
51) Line 405: suggest to start new paragraph with this sentence and edit to:” The atmospheric consequence of these results is that we will typically over-predict…”
52) Figure 1: what are the numbers after the experiment numbers in panel B caption?
53) Figure 2 caption references to panels a and b are confusing, reorder text?
54) Figure 4 notation about #O is confusing. What about “#O(non-NO3) = #O(total) - 3*#N”?