Review: Importance of SOA formation of a-pinene, limonene and m-cresol comparing day- and night-time radical chemistry. Mutzel et al., acp-2019-1066.

This paper describes an experimental study looking at the SOA formation potential of these three important species with OH radicals in the presence of NO and from nitrate radical chemistry. It also investigates the effect of changing the RH in the chamber on the SOA yield and on the yield of individual SOA tracer species. The experiments and the premise of this paper are very interesting and could provide valuable information for the community. However, I felt that some of the analysis was rather limited and hypothesis were presented without evidence, even though the types of analysis used, could have provided this. In addition, some of the language was hard to follow and could benefit from being tidied up. I think if the major points below are addressed the paper would be suitable for ACP.

Major comments

Page 8, line 260-268. I do not agree with the reasoning that the inorganic nitrate is not higher under humid conditions. At <5 % RH, the ratio of DM:NO3- is 29.9:3.1 = 9.65. In contrast, at 75 % RH, the ratio is 16:3.5 = 4.65. A much larger fraction of the aerosol produced is nitrate at higher RH. Also, if any organic nitrates that are present in the SOA do hydrolyse, then they could appear as NO_3^- in the IC analysis, since this uses a water extraction step.

Page 10, line 340-345: What were the results of the blank tests? The a-pinene results are indeed very unexpected. Can you estimate the proportion of RO_2 reacting with HO_2 versus NO in these experiments? What is the temporal evolution of NO in the chamber, how quickly is it converted to NO_2? Also, how much ozone is formed? Could you be seeing ozonolysis leading to SOA peroxides? Your values seem so much higher than previous studies. Could other species be giving a response? Needs more information in the experimental about the analysis method, not just giving a previous paper. For limonene, surely you could use the MS data to determine if high molecular weight peroxide dimers were present?

Page 11, section on Biogenic SOA markers: There is no comparison of the a-pinene SOA tracer data to previous studies. Are the levels observed in alignment with previous studies (especially when considering such high peroxide content)? There needs to be a better discussion here about the amount of liquid water content present at the different RH levels. Is there really less pinonic acid formed at 75 % RH or is there just more water in the SOA, leading to a more dilute aerosol? Line 406: Some of the dimers can be observed in negative ionisation ESI. Could you qualitatively determine whether this pathway increases at higher RH using the LC-MS data?

Page 12, section on Anthropogenic SOA markers: I don't really understand why you are comparing the composition to an "ASOA marker" mixture, rather than using existing knowledge about cresol chemistry to use appropriate standards of oxidation products. Also, there is discussion here about biomass burning but you have called this an anthropogenic tracer. The Hoffmann paper (2007) calls this a method for biomass burning. How many of these compounds would you expect to form from m-cresol oxidation? Are there species you would have expected to see but don't? While the 3MNC shows a positive trend, I don't really feel that this data adds very much to our understanding of cresol SOA.

Page 13, line 465: Have there been any studies of the OH yield from NO3+a-pinene? The formation route for pinonic acid is not obvious. Do you have any idea where it is coming

from in the NO3 reaction? Is it similar to the norpinonic and a possible hydrolysis product? Or could it be from residual ozone or OH?
Page 13, line 478: I felt the last few lines made a very important comment that was not really discussed in the main paper. To me it seems very odd that so little SOA is formed from this route as such a large amount of m-cresol is reacted. In your table 1, there is a literature reference suggesting a 35-49 % yield, but I think perhaps this should be for OH chemistry? A recent paper on o-cresol +NO_3 indicated a SOA yield of 7-17%. There needs to be much more discussion about the reasons for this in the main paper on in the section on page 7.

Sathiyamurthi Ramasamy, Tomoki Nakayama, Takashi Imamura, Yu Morino, Yoshizumi Kajii, Kei Sato, Investigation of dark condition nitrate radical- and ozone-initiated aging of toluene secondary organic aerosol: Importance of nitrate radical reactions with phenolic products, Atmospheric Environment, 2019, https://doi.org/10.1016/j.atmosenv.2019.117049

Minor comments
Abstract: why only give a-pinene peroxide levels here? Also, “pinonic acid as the most important particle phase constituent” What do you mean by “important”? Do you mean highest proportion based on the small subset of products measured?
Page 2, line 57 – is the uncertainty on the limonene + NO_3 rate coefficient really a factor or almost 100? Seems very high.
Page 2 -the introduction is very focussed on NO_3 chemistry and very little on OH. There is clearly much more literature information available for OH and I would like to see some of this incorporated here.
Page 2, line 69: Why is cresol used as the anthropogenic tracer? No explanation given for this choice, since it can also be released from biomass burning.
Page 3, line 96: “equals this a dilution” – consider rewording this sentence.
Page 3, line 104: Can you explain how the pre-reactor was set up to avoid ozone entering the chamber. How much ozone did you detect in the chamber during NO_3 experiments?
Page 4, line 114: 1 g cm^{-3} seems a rather low value to use for density of SOA.
Page 4, line 134: should this say micro L not milli L?
Page 5, section 2.3.4: The contents of the anthropogenic SOA mix need to be included here. The reader does not want to have to go and look this up. It is important to know whether you would expect to see any other tracers in the mix in the cresol experiment or not. If not, why not just use a 3MNC standard?
Page 5, line 164: Reconsider this sentence. Im not sure how it can be “significantly or at least slightly higher”?
Page 6, line 190: This sentence doesn’t make sense. I think you are trying to say the yield from NO3 is higher than from OH. But this sentence seems to say the opposite. Only one a-pinene + OH reference is given in Table 1. I am sure there are many more than that. Page 6,
Page 6, line 201: I don’t follow this sentence. Do you mean SOA is being formed from the oxidation of first generation products rather than the products themselves?
Page 6, line 204: Please give structures for the non-standard named compounds like Myrtenal.
Page 6, line 216 – please give typical limonene ozonolysis SOA yields.

Page 7, line 222 – is there no literature data for SOA from cresol for comparison? The start of this section is rather imprecise. Using cresol as a proxy for "anthropogenic" VOCs and comparing whether they form more SOA than biogenics is not accurate. Hildebrandt uses the parent hydrocarbons and so cannot simply be applied to the cresol.

Page 7, line 240 – there is no mention here about the impact of RH on aerosol phase and hence the uptake of SVOC.

Page 7, line 250: You have not mentioned limonene + OH, which seems to have a different pattern to the others, with the highest yield at 50 % RH?

Page 8, line 277: can you really say this based on a SAR derived rate constant from the MCM?

Page 8, line 280: Were these two sets of reactions done in this study? What is the "one study that investigated this reaction channel"?

Page 8, line 297 – I don't understand what "the evolution in dependency on consumption" means.

Page 9, line 305: This whole section is really confusing, with long sentences with too many parts. There needs to be some more discussion about the data in the figure. Why are some values over 100 %? The proportion of OM also is much lower in limonene + NO_3, why is this? I felt there was a lot of information in the figure, but very little discussion of what it means and the levels of uncertainty.

Page 9, line 321: What is the sensitivity of this analysis as a result of changing the molar mass? There is increasing evidence of auto-oxidation in some systems, which can occur in the presence of NO, which could shift the peroxide mass distribution.

Page 10, line 355 to 360: can you make it clear that you are talking about HO2 concentrations in the GAS phase.

Page 11, line 415. I don't follow this sentence. To me the RH dependence looks very different – nowhere near as dramatic at 75 % RH.