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Comment on acp-2020-1214

Anonymous Referee #2

Referee comment on "Kinetics and impacting factors of HO_2 uptake onto submicron atmospheric aerosols during the 2019 Air QUAlity Study (AQUAS) in Yokohama, Japan " by Jun Zhou et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-1214-RC1, 2021

This is a good paper. HO2 uptake to aerosol can be important as a HOx loss process, especially if NO mixing ratios are low and aerosol surface area is high. There have been a number of lab studies of the uptake coefficient of HO2 to specific aerosol types but to validate these uptake coefficients experiments have to be done in the field with ambient aerosol. However, this requires the use of an online HO2 detection technique (e.g. chemical conversion LIF), an online HO2 source, and aerosol surface characterization techniques, i.e. the experiment is not straightforward to complete.

There are very few online measurements of HO2 uptake using ambient aerosol. This paper presents a new generation of the approach, involving the use of an aerosol concentrator. The concentrator enhances the aerosol surface area and so too the HO2 loss rate, making the experimental results more robust. As well, the authors had a large number of simultaneous aerosol composition measurements, both online and filter based. Importantly, they measured metals by ICP-MS. So, all in all, this is a very challenging but well executed study.

The major results are that the uptake coefficient measured in a field campaign in Yokohama is large, roughly 0.2 to 0.3 on average. This is consistent with lab studies if transition metals are involved. In support, a correlation analysis shows positive correlation between the measured kinetics and metals such as copper.

I have only minor comments, and I recommend that the paper is published.

- The paper would benefit from a cleaner description of the role of the mass accommodation coefficient. Most of the lab studies have found values of between 0.2 to 0.5 to be lower limits to the true mass accommodation coefficient, and so I think it is unlikely that alpha controls the uptake. This is the reason that in Figure 4 the model

with alpha = 0.2 is predicting too low values. Overall, I found the paper was a bit unclear about how to describe the potential impact/role of alpha.

- It would be valuable to point out in the main manuscript to those unfamiliar with the VACES system that it operates by condensational growth followed by inertial separation and then drying. Thus, the particles are not in exactly the same state as they were in the ambient atmosphere. In particular, I wonder whether transition metal ions that were not soluble under ambient conditions were solubilized in the VACES? As well, the morphology of the particles (e.g. phase separated or not) may not be the same after exiting the VACES.
- What sizes of particles are concentrated by the VACES? Was there a PM2.5 or PM1 impactor or cyclone on the inlet line? Was there any surface area that the SMPS did NOT measure? Were the temperature and RH of the SMPS systems exactly the same as the T/RH in the flow reactor, i.e. were the aerosol surface areas the same between these two regions? What are the uncertainties in the surface area enrichment factors?
- For my version of the paper, the quality of Figure 1 was a bit fuzzy.
- Line 254 – The topic of gas phase diffusion limitations was discussed here for micrometer sized particles. I am confused because earlier in the paper, it was stated that gas phase diffusion limitations were negligible (a few percent at most). As well, the SMPS systems used did not scan up to one micrometer.
- Line 349 – I don't understand the relationship of the activity coefficient of copper to the topic being discussed.
- The role of BC as a site for HO2 reactivity is tenuous. I agree there is a weak correlation with BC, but I expect the BC will be coated with other aerosol components, and so I don't see the need to invoke reactivity with solely the BC component. Perhaps BC is just a surrogate for some other component of the aerosol, such as a metal?
- Line 72 – I don't understand the comment about electron transfer from O2- to HO2.
- Line 214 – What is the meaning of this "alpha" term?
- The English in the paper could be improved at places.