Comment on acp-2021-483

Anonymous Referee #1

Referee comment on "The number fraction of iron-containing particles affects OH, HO$_2$ and H$_2$O$_2$ budgets in the atmospheric aqueous phase" by Amina Khaled et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-483-RC1, 2021

This is an interesting paper, as the first one to address how iron distribution across aerosols and droplets may affect multiphase chemistry and reactive oxygen species (ROS). The author has done a good job on examining the budget of ROS in different scenarios. The results are interesting and useful to the community. I have several major comments:

- Aqueous diffusion. Given the short lifetime of ROS in aqueous phase, one would expect a large difference between surface concentration and bulk concentration of ROS for individual aerosols or droplets. This can be addressed by introducing a diffuso-reactive parameter (see Mao et al. 2013 for example). I am wondering how this may affect the results. I would expect that for most cases, OH and HO$_2$ transferred from gas-phase will be consumed in the surface layer of aerosols or droplets, which is different from the assumption of homogeneous mixing in this work for individiual aerosols or droplets.

- While the authors show detailed concentrations of ROS in different scenarios, I am wondering if the authors can also provide gas-phase concentrations of OH, HO$_2$ and H$_2$O$_2$ for each simulation?

- Mass accommodation coefficient and reactive uptake coefficient. The authors show in Table S4 the mass accommodation coefficient (alpha) of OH to be 0.05 and HO$_2$ to 0.01. The mass accommodation coefficient is considered to be the upper limit for reactive uptake coefficient. It is surprising that in Figure 8 the derived reactive uptake coefficient for HO2 is higher than the mass accommodation coefficient of HO$_2$ (0.01).

- Gas-phase diffusion. The equation (E.7) should include a gas-phase diffusion term. Note that for cloud droplets, the reactive uptake process is limited by gas-phase diffusion, not by reactive uptake. Some good explanation on this can be found from Jacob (2000) paper ("Heterogeneous chemistry and tropospheric ozone"). Therefore, the quantification of reactive uptake coefficient (gamma) for cloud droplets could be of minor importance. More importantly, I am wondering if the authors could discuss the importance of gas-phase diffusion throughout this work. Potentially, some of the

difference between aerosols and cloud droplets can be attributed to the difference in gas-phase diffusion.

Technical comments:

- Line 69 "…perform box model simulations a box model with…", remove "a box model"
- Line 90 Equation 1-3 needs a reference. How are they derived?
- Line 108 why 400 seconds?
- Line 125 "Fig. 1" should be "Fig. 2".
- Table 1 Iron-containing aerosol particles should be "30", not "2".
- Line 212-214, this should go to Line 108. "This time corresponds approximately to the lifetime of a single cloud droplet but underestimates the time an aerosol particle might be exposed to a given relative humidity. We have chosen this relatively short time, as in our box model setup, the initialized gases are not replenished over time as no emissions are considered."
- Line 387: It seems that Figure 6 should go after Figure 8.