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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Photochemical degradation of iron(III)-citrate/citric acid aerosol quantified with the combination of three complementary experimental techniques and a kinetic process model" *by* Jing Dou et al.

Anonymous Referee #2

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This manuscript (acp-2020-779) investigates iron(III) carboxylate photochemistry using three different experimental approaches. In an electrodynamic balance, the mass loss of a levitated droplet containing Fe(III)-citrate and citric acid upon irradiation was quantified. Using STXM/NEXAFS, changes to the oxidation state of deposited Fe-containing aerosol were measured. With a coated wall flow tube, the production of HO2Âů was monitored during photochemical reaction through a chemiluminescence measurement of NO. The Photochemical Reaction and Diffusion (PRAD) model was then applied to all three sets of experimental data to explore chemical processes occurring within a

single droplet. The results indicate that photochemical degradation and repartitioning of molecules to the gas phase could be very significant for Fe-containing aerosols in the atmosphere.

This manuscript combines interesting and well-conducted complementary experiments with a new modelling approach to develop a consistent interpretation of the complex chemical processes occurring within an individual droplet, representing an important advance. The manuscript is well-written, with clearly constructed and appropriate figures. This manuscript is within the scope of Atmospheric Chemistry and Physics and will be publishable once the comments below are addressed.

Comments:

1. PRAD model: The PRAD model appears to be a novel model to describe chemistry within a single droplet. The manuscript would be improved by providing some additional context identifying the distinct aspects of the PRAD model relative to existing models (e.g. KM-GAP, Kinetiscope) which appear to operate under similar principles. In addition, the authors indicate that the model is manually tuned. The authors should clarify how they minimized the possibility of tuning to a local minimum. Moreover, in discussing the number of parameters tuned and the fraction of parameters that are poorly known, the authors should also provide some indication about the sensitivity of the model to these parameters. This is discussed a bit on pages 21 and 22, but additional detail would strengthen the manuscript.

2. Page 12, line 247: In this line, the number of molecules is defined as Nn, whereas on the previous page (line 221), the number of moles is defined as Ni. Although the discussion for each revolves around different portions of the particle (n representing the outermost layer; i representing inner layers), the authors should ensure that terms are consistently used throughout the manuscript. Moreover, on page 12, line 270, n*Cit, nCit and nFeCit do not appear to be defined. Presumably these refer to molarity, in which case should not the appropriate term be c (as defined on page 9, line 205)?

3. Figure 6 caption: The colours listed in the caption do not match those in the figure.

4. Page 17, line 329 (“within about 6 hours”): This is not shown experimentally in Fig. 8. Experimental data at 60% RH only extend to $\sim$3 h. The model extends to 6 h, but this is not an observation.

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