

Review of “Multiphase processes in the EC-Earth Earth System model and their relevance to the atmospheric oxalate, sulfate, and iron cycles” by *Myriokefalitakis et al.* (2021)

The manuscript by *Myriokefalitakis et al.* (2021) describes the results of EC-Earth3, an Earth System Model, after the implementation of a detailed multiphase chemistry scheme. The focus of this study is on the cycling of oxalate (OXL), sulfate, and bioavailable/soluble iron (SFe) in the aerosol phase. The model is also tested with two sets of meteorological fields. Model results of the variables of interest are compared to available observations. The article is very dense; however, in general, is well written and describes complex issues clearly. The complex chemistry scheme implemented is well referenced and all assumptions were justified to the best of the author’s ability. While I feel the paper could benefit from some shortening and simplifying, the manuscript is scientifically sound and presents interesting results. I only have minor issues with this manuscript. After addressing the minor comments below I feel this paper is sufficient for publication in *Geoscientific Model Development* (GMD).

Scientific Comments

1. The authors mention that the metastable assumption in ISORROPIA II can lead to overprediction of aerosol acidity (i.e., lower pH values) compared to the stable aerosol assumption. Can the authors provide some estimate of how this might impact the results of the model simulations? For instance, is the atmospheric processing production of SFe noticeably larger due to the metastable assumption? It would be good to know how this uncertainty in aerosol acidity calculations might impact the multiphase chemistry incorporated in the model.
2. How many passive tracers had to be added to the model in order to simulate all the gas phase and aqueous species, and the multiphase chemical reactions, represented in Table S2? Did this significantly increase the computational expense of the model?
3. The standard deviation around the mean of the multi-year OXL (and some other species) is very small. Does this mean there is very little interannual variability (IAV) in the primary emissions (I see from Table 2 that the primary emissions are constant) and precursor species emission, and production/destruction processes? One would think that there would be IAV in emission source strength of precursor species, transport/deposition, and other meteorological conditions impacting OXL production/distribution.
4. The paper is very dense. The amount of quantitative values for species emission, production/destruction, deposition, and evaluation statistics of each species, and comparison to other recent studies, presented in the text of the article is a bit overwhelming. It makes reading and understanding the manuscript difficult. After much effort I feel that all the values seem reasonable; however, this took significant effort. This might impact the effectiveness of presenting the important results of this paper. I don’t think this is 100% necessary, but I would suggest that the authors think of ways to simplify the text of the paper.

5. How important is the comparison of the model results, in all sections of the manuscript, with both sets of meteorological fields? I almost think this part of the manuscript could be a supplemental section. This would reduce the density of the article's information in the main body of the paper.

Technical Comments

1. Line 563. considered in that latter study.
2. The use of “~”. The authors use the approximation symbol for nearly every value presented in the manuscript. It seems that the values are often pretty exact (e.g., line 576 for the atmospheric lifetime of 5.7 days) and likely do not need this symbol.
3. Line 583. “calculates that is ~ 3 % lower” needs rewording.
4. Line 603. ~~are~~ is produced.
5. Line 661. downwind of land areas.