

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

This manuscript describes the development of a detailed multiphase chemistry scheme within EC-Earth with the worthy aim to realistically represent the atmospheric iron cycle. This can improve understanding of marine biogeochemistry perturbations, and thus carbon and nitrogen cycles, under past and future climate scenarios. The model contains iron from both natural and anthropogenic sources and contains schemes for the dissolution of insoluble iron to a soluble form which account kinetically for the solution's acidity, oxalic acid, and irradiation. The chemistry required is discussed in great detail and results from two sets of simulation are compared to observations of oxalate, sulphate, and iron aerosol. The analysis is very good, and results are interesting given that representing the full atmospheric aerosol iron cycle within a global Earth System Model is still a new development. In my opinion this manuscript is very well written and in particular the description of the chemistry was very well made. I feel this manuscript is thus suitable for publication in *Geoscientific Model Development* after addressing a series of, mainly minor, comments.

1. Please double check the level of significance throughout the manuscript. There are some instances of where a higher precision is given than likely should be. Also please be consistent with the level of precision within a given paragraph, best to not let it vary without giving the justification.
2. When reporting the average for shipborne observations (and comparing to the model) it is recommend using the median and not the mean (or give both). This is how the observational papers report these values (e.g., any Baker et al. paper) and a full reasoning is given in (Hamilton et al., 2019) outlining why for such sparse (spatially and temporally) datasets the mean can often be misleading. Using the median may also result in a better model: obs correlation. Please add medians and alter discussion where needed.
3. Maybe useful to point to Table S2 earlier within the introduction to help guide the reader through the many reaction mechanisms discussed.
4. Methods: Please describe the aerosol model further. Some points to include for example would be mixing assumptions (internal vs. external), which modes the iron aerosol goes into (and if ageing of aerosol information is needed please add), how the aerosol number concentrations are calculated and the associated new constants used for iron aerosol, the vertical distribution of biomass burning emissions (if any), and how dry and wet deposition are handled (briefly). There may be more beyond this list too.
5. If Fe fractions are used to generate combustion iron emissions, how are the coarse sized emissions of carbonaceous aerosol estimated? CMIP6 inventories have assumed all BC and OC is in the accumulation mode.
6. Please add the required additional model simulation time (e.g., core hours) for the new tracers and chemistry. It may be easiest to give both the base model EC-Earth3 run time and the new EC-Earth3-Iron runtimes.
7. Using a hematite fraction of 66% iron (Table S1) is higher than other studies (e.g., (Journet et al., 2008) gives 57.5%). What is the reasoning for using this value?
8. Have any of the modelled dust optical properties been modified by accounting for dust mineralogy? is the new iron aerosol from combustion sources interacting with the radiation scheme? Please briefly what are the potential impacts/feedbacks on relevant variables for

the chemistry (temperature, humidity, etc.) when comparing online (EC-Earth) vs. offline (ERA-Interim) simulations of adding (or not adding) these couplings.

9. What is the dust emission flux (Tg/a)? How does that compare to recent estimates (e.g., (Kok et al., 2021; Wu et al., 2020))? And were dust emissions tuned in any way (e.g., (Ridley et al., 2016) recommendations to attain a global mean dust AOD of 0.03)?
10. Does dust mineralogy alter the AOD estimates in the model?
11. Is dust aerosol internally mixed with sea spray aerosol? How does this impact dust lifetimes?
12. Can the Authors describe why is the lifetime of dust iron much larger than combustion iron? I would have guessed the other way as dust is larger and thus more prone to being lost from the atmosphere by dry deposition. This is therefore maybe interesting.
13. The discussion section compares results with Myriokefalitakis et al., 2018 and Ito et al. 2021. However, neither of these studies contain results from an Earth System Model using a modal aerosol scheme containing iron aerosol. It therefore would be insightful to include a comparison to the more similar model aerosol framework from (Hamilton et al., 2020, GRL) or (Hamilton et al., 2019). Furthermore, (Hamilton et al., 2020) covers the same study years as presented here, which the other studies do not.

L70-75: What of other anthropogenic fuel sources beyond oil? How do these compare?

L140: Can the authors elaborate more on why a continental source is important?

L185: Is DMS in EC-Earth truly prognostic as it uses a climatological monthly mean ocean surface conc. from Lana et al. (2011)? The gas transfer velocity is then parameterized following Wanninkhof (2014).

L194: Please define anthropogenic (sources are those from Table S1?) and biomass burning (anthropogenic or also wildfires?). Was a metal smelting source accounted for? At L219 it states, "Fe is also emitted in the model from anthropogenic combustion and biomass burning sources following Ito et al. (2018) and Hajima et al. (2019)", Ito et al. do consider metal smelting, but I can find no reference here to this particular source.

L213: What is the mean PSD (accumulation: coarse dust) ratio?

L215: Is the 0.1% placed in "fast" at point of emission? And can the Authors describe a little more their reasoning for using 0.1% solubility for all iron bearing minerals regardless of mineralogy.

L219: I cannot quite link up how this methodology follows Ito et al. (2018) and Hajima et al. (2019). Maybe it is best to describe what was done here to estimate combustion iron emissions in more detail. For example, Ito et al. uses supermicron and submicron PM values while Hajima et al. states that MIROC uses a 0.4gFe gBC<sup>-1</sup> ratio. But I cannot find how either of these link to the different values given in Table S1, and if values in Table S1 are iron fraction w.r.t. to total aerosol or to carbonaceous aerosol only. Some more description of how these were derived in the main text and the Table header would thus be beneficial. Also, within Table S1 where are values for biomass burning 0.63 (accumulation) and 2.30 (coarse) from – also Ito et al. 2019? (maybe add refs to Table S1?). For the biomass burning how do these values compare to the multi-biome ratio estimates given in (Hamilton et al., 2019, 2022)?

L225: For the study period this is the GFED4s fire emission dataset, maybe also reference as such?

L226: While an iron solubility of 79% for oil has been recorded, it is a value at the high end of the literature (8-85%; e.g., (Rathod et al., 2020)). Furthermore, MIROC uses 79% for the final solubility of oil sourced iron at the point of deposition. As 79% is used at point of emission here this iron will only increase after atmospheric processing. I think it would be useful therefore to describe what possible implications this has for the current study, future model development, and comparison to observations shown.

L228: I found it a little confusing to read biomass burning alongside anthropogenic in the same sentence here. This could be read to imply that only fires from human activity are accounted for in the model. Maybe best to separate these sentences and explain why this assumption holds for biomass burning (e.g., assumed no change in vegetation type over the study period?).

L598: Maybe duplicate this information about no ocean sources to the methods, alongside where it is discussed there are no gasoline engines sources (L237), to be complete in the methods.

Figure 4: Would be nice to also have the statistics on the figure

Figure 6 (g+h) Would be nice to also have the statistics on the figure

Figure 10: The use of contouring in the shading of the map creates a somewhat false impression of the values given it interpolates between cells and there are some high gradients and lone cells. I feel that it would be better to colour each grid cell individually. Also how were the observations averaged; all observations in a given cell collected and the average taken I am assuming, but maybe this can be more explicitly described in the caption. Can the standard error also be included for the ERA line in the bottom panels?

Figures (general): Please alter line colours where red and green are used to make figures more accessible for colour blind readers.

Table S1: Please add refs for values.

Figure S5: Is it possible to add a scatter plot for iron solubility.

### **Technical comments:**

L59: uptake of *atmospheric* CO<sub>2</sub>

L132: Seems quite a precise approximate value

L211: PSD is only used twice and in the same paragraph; likely does not warrant an acronym.

L765: 'outstandingly' is hyperbole, please give the values and allow the reader to come to that conclusion if merited.

### **References:**

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