We thank the reviewer for the careful reading of the manuscript and the insightful comments. Please find below our point-by-point replies:

Scientific Comments

SC1. **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.**

- We thank the reviewer for this comment. In retrospect, this comment should have been rephrased differently: "*The metastable assumption produces pH values that are different from the stable assumption. Work to date, such as Bougiatioti et al. (2016), Guo et al. (2018; 2015) and others identified in the review of Pye et al. (2020), has shown that the metastable solution tends to provide semi-volatile partitioning of pH-sensitive species (like NH_3/NH_4 and HNO_3/NO_3) and aerosol liquid water content that is closer to observations – at least for when the RH is above 40%. For this reason, we assume that the most plausible estimates of acidity are to be obtained with the metastable assumption – and we base our simulations on that.*"
  Nevertheless, we have further investigated the sensitivity of dissolved Fe to the phase state assumption, by running the model (ERA-Interim) for one year with ISORROPIA-II in the forward mode but following the stable assumption.

SFe Deposition, Annual
STABLE - METASTABLE
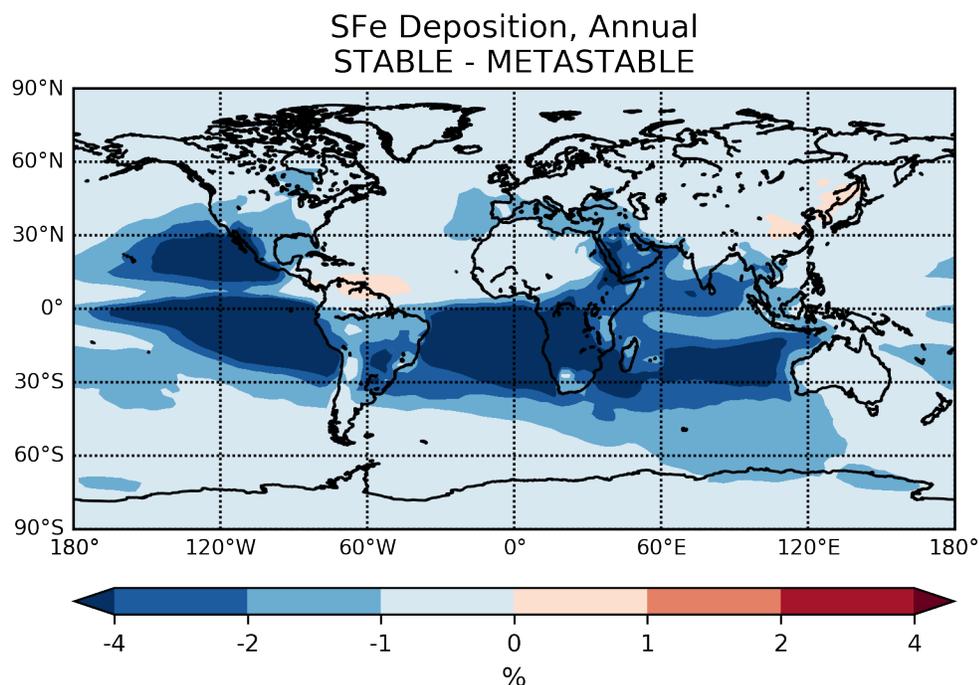
Our results indicate that the total Fe dissolution is lower by ~20 % on average due to the total cumulative effects of the different aerosol pH and liquid water content predicted. Moreover, the expected location of soluble Fe production is also affected by the different aerosol phase state assumptions (i.e., metastable vs. stable), which in turn affects the spatial distribution of soluble iron deposition (SFe). The stable aerosol

assumption leads generally to lower solubilities in deposition over source regions, such as for dust-dominated (~ -1%) as for combustion-dominated (~ -3%). However, the larger impact is calculated over the remote ocean (to up to ~ -4%), where high iron solubilities are combined with low iron-containing aerosol concentrations. We note, however, that dust outflow and wildfire-dominated regions still receive most of the soluble iron, in contrast to the Southern Ocean that receives much lesser soluble iron.

SC2.   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?

- The aqueous phase driver alone accounts for 68 tracers that can either be partitioned, dissolved (in the case of minerals), and/or oxidized in the aqueous phase of the atmosphere. However, upon the calculation of the aqueous phase chemistry, the model does not trace all the different forms of the species participating in the aqueous-phase chemistry scheme, meaning that the different aqueous-phase forms are not prognostic variables (tracers) in the model, as we also stated in the manuscript. Instead, we trace the final concentrations that can be transported and deposited. Thus, for this work we account only for tracers needed to properly describe the mineral-Fe solubilization processes; specifically, oxalate and glyoxylic acid, as well as the mineral dust-Fe according to three dissolution classes; namely fast, intermediate, and slow Fe pools (overall, 10 species additionally).

  The aqueous-phase chemistry calculations, nevertheless, do increase the computational time of the model. To make it more clear to the reader as the reviewer proposed, we added a new subsection (i.e., Sect. 2.6) focusing specifically on the model's performance: "*The coupling of the aqueous-phase chemistry scheme along with the description of the atmospheric iron cycle for this work increases the model runtime. EC-Earth3-Iron uses 109 transported and 33 non-transported tracers, which are significantly larger numbers than in the EC-Earth3-AerChem configuration (i.e., 69 transported and 21 non-transported tracers). Note, however, that EC-Earth3-Iron used for this work employs the MOGUNTIA gas-phase chemistry scheme configuration, in contrast to the mCB05 configuration used in EC-Earth3-AerChem, which is overall found to be computationally more expensive by ~27% alone (Myriokefalitakis et al., 2020). In the Marenostrum4 supercomputer architecture (2x Intel Xeon Platinum 8160 24C at 2.1 GHz), the EC-Earth3-AerChem configuration (van Noije et al., 2021) simulates 1.85 years per day simulation time (SYPD) with 187 CPUs, while to reach to a comparable performance (i.e., 1.41 SYPD) with the EC-Earth3-Iron configurations, 432 CPUs are required, respectively. This means overall, that the EC-Earth3-Iron corresponds to 7353 computation hours per year (CHPY), which is roughly 3 times larger than the standard EC-Earth3-AerChem.*"

SC3.   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.

- The EC-Earth3-Iron version used for this study is built on EC-Earth3-AerChem (van Noije et al., 2021). Thus, we used the same emission set-up for anthropogenic, biomass burning, as well as for biogenic and other natural emissions as in van Noije et al. (2021).

However, in EC-Earth3-AerChem, the biogenic emissions are prescribed using monthly estimates from the MEGAN-MACC data set (Sindelarova et al., 2014) for the year 2000, as produced by the Model of Emissions of Gases and Aerosols from Nature (MEGAN) version 2.1 under the Monitoring Atmospheric Composition and Climate (MACC) project. Accounting that OXL is mainly produced from precursors of biogenic origin (i.e., mainly isoprene), we do not thus expect a strong IAV on the calculation of its production, i.e., in EC-Earth the OXL production is 12.61 ± 0.06 Tg yr^{-1}. On the other hand, the primary emissions of OXL, although very uncertain as we note in the manuscript, are very low based on the published estimates and are not expected to have a substantial impact on OXL atmospheric concentrations. Note that in Table 2 we provide the average of the simulated period (see also caption), but in the text, we also provide the IAV, i.e., 0.373 ± 0.005 Tg yr^{-1}; see also Sect. 3.1.1. Nevertheless, we here present simulations only for a short period (2000-2014) of present-day, thus no important IAV is expected.

SC4. **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.**

- This manuscript aims to include the most important information about the calculations of the precursors of oxalate and how this can impact Fe-containing minerals dissolution. Such a layout can help the reader to have in one place all the critical information concerning the chemistry calculations, along with a comparison to other state-of-the-art studies. This layout is, thus, necessary to describe the complex chemical formation pathways of OXL and its precursors, as in other studies available in the literature (e.g., Lin et al., 2012; Liu et al., 2012; Myriokefalitakis et al., 2011), as well as to provide enough references to guide the reader for further reading.

SC5. **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.**

- Our goal here is to couple a chemistry scheme that can satisfactorily simulate the aqueous-phase production of oxalate to properly simulate the mineral Fe dissolution processes in the atmosphere. However, using only the calculated (online) meteorology of EC-Earth, we cannot come to a safe conclusion based on present-day observations concerning the actual strength of oxalate production in the aqueous phase of the atmosphere, due to biases in the meteorological conditions that unavoidably exist in an ESM. On the other hand, the ERA-Interim setup allows constraining the model with the observations-based assimilated meteorological data and is therefore used for the budget analysis and comparison with other modeling estimates from the literature. All in all, the comparison with ERA-Interim is used to explore uncertainties regarding the aqueous-phase chemistry scheme in EC-Earth and thus its impact on Fe-containing aerosol solubilization processes. For this, we keep in the discussion the results of both sets of meteorological fields.

Technical Comments

TC1. **Line 563. considered <u>in</u> that latter study.**
- Corrected.

TC2. **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.**
- Corrected.

TC3. **Line 583. "calculates that is ~ 3 % lower" needs rewording.**
- Rephrased as "*while in EC-Earth is calculated 3 % lower.*"

TC4. **Line 603. ~~are~~ <u>is</u> produced.**
- Corrected.

TC5. **Line 661. downwind <u>of</u> land areas.**
- Corrected.

References

Guo, H., Xu, L., Bougiatioti, A., Cerully, K. M., Capps, S. L., Hite, J. R., Carlton, A. G., Lee, S.-H., Bergin, M. H., Ng, N. L., Nenes, A. and Weber, R. J.: Fine-particle water and pH in the southeastern United States, Atmos. Chem. Phys., 15(9), 5211–5228, doi:10.5194/acp-15-5211-2015, 2015.

Lin, G., Penner, J. E., Sillman, S., Taraborrelli, D. and Lelieveld, J.: Global modeling of SOA formation from dicarbonyls, epoxides, organic nitrates and peroxides, Atmos. Chem. Phys., 12(10), 4743–4774, doi:10.5194/acp-12-4743-2012, 2012.

Liu, J., Horowitz, L. W., Fan, S., Carlton, A. G. and Levy, H.: Global in-cloud production of secondary organic aerosols: Implementation of a detailed chemical mechanism in the GFDL atmospheric model AM3, J. Geophys. Res. Atmos., 117(D15), 1–15, doi:10.1029/2012JD017838, 2012.

Myriokefalitakis, S., Tsigaridis, K., Mihalopoulos, N., Sciare, J., Nenes, A., Kawamura, K., Segers, A. and Kanakidou, M.: In-cloud oxalate formation in the global troposphere: a 3-D modeling study, Atmos. Chem. Phys., 11(12), 5761–5782, doi:10.5194/acp-11-5761-2011, 2011.

Myriokefalitakis, S., Daskalakis, N., Gkouvousis, A., Hilboll, A., van Noije, T., Williams, J. E., Le Sager, P., Huijnen, V., Houweling, S., Bergman, T., Nüß, J. R., Vrekoussis, M., Kanakidou, M. and Krol, M. C.: Description and evaluation of a detailed gas-phase chemistry scheme in the TM5-MP global chemistry transport model (r112), Geosci. Model Dev., 13(11), 5507–5548, doi:10.5194/gmd-13-5507-2020, 2020.

van Noije, T., Bergman, T., Le Sager, P., O'Donnell, D., Makkonen, R., Gonçalves-Ageitos, M., Döscher, R., Fladrich, U., von Hardenberg, J., Keskinen, J.-P., Korhonen, H., Laakso, A., Myriokefalitakis, S., Ollinaho, P., Pérez García-Pando, C., Reerink, T., Schrödner, R., Wyser, K. and Yang, S.: EC-Earth3-AerChem: a global climate model with interactive aerosols and atmospheric chemistry participating in CMIP6, Geosci. Model Dev., 14(9), 5637–5668, doi:10.5194/gmd-14-5637-2021, 2021.

Sindelarova, K., Granier, C., Bouarar, I., Guenther, A., Tilmes, S., Stavrakou, T., Müller, J.-F., Kuhn, U., Stefani, P. and Knorr, W.: Global data set of biogenic VOC emissions calculated by the MEGAN model over the last 30 years, Atmos. Chem. Phys., 14(17), 9317–9341, doi:10.5194/acp-14-9317-2014, 2014.

Guo, H., Xu, L., Bougiatioti, A., Cerully, K. M., Capps, S. L., Hite, J. R., Carlton, A. G., Lee, S.-H., Bergin, M. H., Ng, N. L., Nenes, A. and Weber, R. J.: Fine-particle water and pH in the southeastern United States, Atmos. Chem. Phys., 15(9), 5211–5228, doi:10.5194/acp-15-5211-2015, 2015.

Lin, G., Penner, J. E., Sillman, S., Taraborrelli, D. and Lelieveld, J.: Global modeling of SOA formation from dicarbonyls, epoxides, organic nitrates and peroxides, Atmos. Chem. Phys., 12(10), 4743–4774, doi:10.5194/acp-12-4743-2012, 2012.

Liu, J., Horowitz, L. W., Fan, S., Carlton, A. G. and Levy, H.: Global in-cloud production of secondary organic aerosols: Implementation of a detailed chemical mechanism in the GFDL atmospheric model AM3, J. Geophys. Res. Atmos., 117(D15), 1–15, doi:10.1029/2012JD017838, 2012.

Myriokefalitakis, S., Tsigaridis, K., Mihalopoulos, N., Sciare, J., Nenes, A., Kawamura, K., Segers, A. and Kanakidou, M.: In-cloud oxalate formation in the global troposphere: a 3-D modeling study, Atmos. Chem. Phys., 11(12), 5761–5782, doi:10.5194/acp-11-5761-2011, 2011.

Myriokefalitakis, S., Daskalakis, N., Gkouvousis, A., Hilboll, A., van Noije, T., Williams, J. E., Le Sager, P., Huijnen, V., Houweling, S., Bergman, T., Nüß, J. R., Vrekoussis, M., Kanakidou, M. and Krol, M. C.: Description and evaluation of a detailed gas-phase chemistry scheme in the TM5-MP global chemistry transport model (r112), Geosci. Model Dev., 13(11), 5507–5548, doi:10.5194/gmd-13-5507-2020, 2020.

van Noije, T., Bergman, T., Le Sager, P., O'Donnell, D., Makkonen, R., Gonçalves-Ageitos, M.,

Döscher, R., Fladrich, U., von Hardenberg, J., Keskinen, J.-P., Korhonen, H., Laakso, A., Myriokefalitakis, S., Ollinaho, P., Pérez García-Pando, C., Reerink, T., Schrödner, R., Wyser, K. and Yang, S.: EC-Earth3-AerChem: a global climate model with interactive aerosols and atmospheric chemistry participating in CMIP6, Geosci. Model Dev., 14(9), 5637–5668, doi:10.5194/gmd-14-5637-2021, 2021.

Sindelarova, K., Granier, C., Bouarar, I., Guenther, A., Tilmes, S., Stavrakou, T., Müller, J.-F., Kuhn, U., Stefani, P. and Knorr, W.: Global data set of biogenic VOC emissions calculated by the MEGAN model over the last 30 years, Atmos. Chem. Phys., 14(17), 9317–9341, doi:10.5194/acp-14-9317-2014, 2014.