

Response to reviewers. EGUSPHERE-2022-1414.

Modeling dust mineralogical composition: sensitivity to soil mineralogy atlases and their expected climate impacts.

We gratefully acknowledge the comments and suggestions from Dr. Akinori Ito and Anonymous Reviewer #2, who have contributed to improve the quality of this manuscript. Please, find below the detailed response to each of their comments and a summary of the changes included in the manuscript.

RC#1

General comments

Model predictions of dust mineralogical composition and its effect on climate are highly uncertain. The authors implemented two mineralogical data sets to a global atmospheric chemistry model. They evaluated the model results against observations of dust mineralogy. They conducted sensitivity experiments to assess the impact of the modeled mineralogy on the dust single scattering albedo and iron deposition. Overall, the paper is well written and will help us to advance modeling dust mineralogical composition. I have some comments to improve the paper.

Specific comments

I.99: Please check and correct the reference for BFT.

The reference has been corrected and it is now included as:

Kok, J. F.: A scaling theory for the size distribution of emitted dust aerosols suggests climate models underestimate the size of the global dust cycle, *Proceedings of the National Academy of Sciences of the United States of America*, 108, 1016–1021, <https://doi.org/10.1073/pnas.1014798108>, 2011

I.117: Please include more comprehensive references for different models which used C1999 or J2014NN to estimate the dust DRE and the atmospheric soluble iron deposition.

The introduction section has been enriched with the references listed below. Additionally, the reference lists have been, in some cases, introduced by “e.g.,” to account for the fact that they constitute a sample of the current publications in each area.

As studies considering the radiative effect of mineralogy we have included: **Ito et al. (2018)** which considers J2014NN to derive the impact of dust radiative effect and assess its relevance compared to other aerosols.

As studies that assess the impact of dust mineralogy as a source of iron in the atmosphere, we have included: **Wang et al. (2015)** and **Ito and Shi (2016)**, both use J2014NN. We have also included an additional reference to the relevance of dust for biogeochemical cycles: **Kanakidou et al. (2018)**.

We have also slightly modified the sentence on line 121: “*and even fewer (Li et al., 2021) make use of different soil maps in a common modelling framework*”, to specify that we refer to single models using the two soil maps, rather than model intercomparison exercises.

Ito, A., & Shi, Z. (2016). Delivery of anthropogenic bioavailable iron from mineral dust and combustion aerosols to the ocean. *Atmospheric Chemistry and Physics*, 16(1), 85–99. <https://doi.org/10.5194/acp-16-85-2016>

Ito, A., Lin, G., & Penner, J. E. (2018). Radiative forcing by light-absorbing aerosols of pyrogenetic iron oxides. *Scientific Reports*, 8(1), 1–11. <https://doi.org/10.1038/s41598-018-25756-3>

Kanakidou, M., Myriokefalitakis, S., & Tsigaridis, K. (2018). Aerosols in atmospheric chemistry and biogeochemical cycles of nutrients. *Environmental Research Letters*, 13(6). <https://doi.org/10.1088/1748-9326/aabddb>

Wang, R., Balkanski, Y., Boucher, O., Bopp, L., Chappell, A., Ciais, P., Hauglustaine, D., Peñuelas, J., and Tao, S.: Sources, transport and deposition of iron in the global atmosphere, *Atmos. Chem. Phys.*, 15, 6247–6270, <https://doi.org/10.5194/acp-15-6247-2015>, 2015.

I.130: Why don't you show the dust DRE (Klose et al., 2021) and compare it with previous studies which used C1999 or J2014NN?

The MONARCH model version presented here accounts for individual tracers for the different minerals present in dust, however it does not include their impact on radiation online. Such work is currently being developed and will be published in a follow-up paper. In this study, we present an offline calculation of the dust single scattering albedo (Figure 12). There, we assume that all minerals have the same optical properties, equivalent to those of homogeneous dust (panel A), and compare it with the result obtained from the mineralogy of C1999 and J2014 (panels B and C). Using a reference for the homogeneous dust that spans for the same period and has the same aerosols' load than our speciated dust allows us to assess the specific impact of mineralogy on the optical properties. The results presented in Klose et al. (2021) correspond to the year 2012 and different dust configurations. On the other hand, in Section 6 we compare our off-line calculated optical properties with the results of previous studies using Claquin et al. (1999) and Journet et al. (2014).

I.164: Why did you disable the gas-phase chemistry and the other dynamic aerosols? Please clarify this effect on the dust model prediction and its evaluation (see also comment on I.214).

The main goal of this work is to assess the ability of our model to reproduce dust mineralogy as compared to observations, and its sensitivity to the soil mineralogy information, we decided to use climatological values for aerosols different from dust and to deactivate the gas-phase chemistry, defining a set of equivalent experiments in which we only vary the emitted dust composition. Each of our simulations span for 5 years and including the mineralogy adds respectively: 72 (C1999), 112 (J2014) and 120 (J2014NN) new tracers to the model.

We have now included a sentence in the manuscript to justify this choice (I166-169), which reads as follows:

“All the experiments are configured to represent only the atmospheric dynamics and the speciated dust cycle, disabling the gas-phase chemistry and the other dynamic aerosols. The dust interaction with radiation, along with that of other aerosols, is solved by using prescribed aerosol climatologies. This configuration allows us to reproduce the main features of the minerals life cycle, while it optimizes the computational time needed to conduct the experiments.”

Although there are ongoing developments to assess the impact of dust mineralogy on heterogeneous chemistry and the thermodynamic equilibrium of inorganic species within MONARCH, in this study dust is assumed to be an externally mixed aerosol and without specific interactions with the gas-phase chemistry. Therefore, differences in the dust cycle with or without the gas-phase chemistry and the other aerosols are expected to be small. Those would be exclusively attributed to the impact of resolving online atmospheric chemistry and aerosols in the meteorological drivers of dust emission and deposition (e.g., wind speed, precipitation or soil humidity) as compared to the use of climatological averages, but not to specific effects of the mineralogy.

I.186: Why did you select G01-UST? Which option did you select for the soil moisture? Presumably, you selected the default option, why did you select this? The use of this equation in G01-UST means that the threshold friction velocity and thus dust emission is hardly sensitive to the soil wetness. Please justify this.

This work aims at assessing the airborne dust mineralogy modeled by MONARCH and the sensitivity of the results to the specific soil mineralogy map. Testing the sensitivity of our results to different dust emission schemes and configurations is beyond the scope of the paper.

Klose et al. (2021) assesses four different configurations for the representation of dust emission currently available in MONARCH. The configuration G01-UST was selected for the following reasons: (1) it is an empirically-based approach the representation of dust sources is to a certain extent observationally constrained, (2) overall it produces similar results to the other schemes in Klose et al. (2021) as compared to MODIS and AERONET observations and (3) it shares many of the characteristics of the dust emission configuration for the operational forecasts routinely produced at BSC as part of the WMO Barcelona Dust Regional Center (<https://dust.aemet.es/about-us/monarch>).

The reviewer is right in that the soil moisture correction for the threshold friction velocity (Belly, 1964; Ginoux et al. 2001) is not very sensitive to soil moisture. We think that this lack of sensitivity towards soil wetness does not have a significant impact on the ability of the model to reproduce the dust annual cycle (see Fig. 7 in Klose et al., 2021: MONARCH G01-UST shows a monthly correlation of 0.91 against MODIS retrieved coarse Dust Optical Depth, and of 0.88 for the Frequency of Occurrence of DOD greater than 0.2). In addition, the geographical distribution of the modeled surface dust concentration and deposition fields is highly correlated with climatological observations (Figure 4) and the shown biases are well within the range of current uncertainties shared by different models (e.g., Huneeus et al., 2011; Albani et al., 2014; Checa-Garcia et al., 2021).

I.200: How did you consider phosphorus?

We have not considered phosphorus (P). Our simulations include different minerals, rather than nutrients. Unfortunately, there is a lack information on the abundance of relevant minerals in soils for an offline diagnostic on the phosphorus content, e.g., apatite. Past works have included a representation of phosphorus associated with dust via the explicit definition of their content in the

soils (e.g., Okin et al. 2004; Zhang et al., 2015) and Nickovic et al. (2012) reports the P content in different FAO soil types. However, we have focused on this work exclusively in the minerals reported by Claquin et al. (1999) and Journet et al. (2014).

Okin, G. S., Mahowald, N., Chadwick, O. A., & Artaxo, P. (2004). Impact of desert dust on the biogeochemistry of phosphorus in terrestrial ecosystems. *Global Biogeochemical Cycles*, 18(2). <https://doi.org/10.1029/2003GB002145>

Y. Zhang, N. Mahowald, R. Scanza, E. Journet, K. Desboeufs, S. Albani, J. Kok, G. Zhuang, Y. Chen, D. D. Cohen, A. Paytan, M. D. Patey, E. P. Achterberg, J P. Engelbrecht, and K. W. Fomba, Modeling the global emission, transport and deposition of trace elements associated with mineral dust, *Biogeosciences*, 2015, <http://www.biogeosciences.net/12/5771/2015/>

I.205: Please elucidate the form of iron oxides and specify the iron content for each mineral.

Iron oxides in the soil mineralogy maps we use are assumed to be hematite and goethite. We use lines 84-90 from the introduction to explain that although Claquin et al. (1999) only identifies hematite in their mean mineralogy table, their work and that of Nickovic et al. (2012) often assumes that hematite is a proxy for both hematite and goethite. In Journet et al. (2014) both minerals are identified separately.

The iron content is estimated from the iron content per mineral reported in Journet et al. (2014), except for the hematite and goethite, where we consider the values reported in Journet et al. (2008). This was explained in the Figure 13 caption in the original version of the manuscript, we have since included it and appropriately referenced it in **Table 1**.

I.212: How did you consider the effect of the mineralogy on the in-cloud scavenging (see also comment on I.214)?

At present, minerals have the same treatment in in-cloud scavenging, i.e., they are assumed to share the same solubility.

I.214: How did you consider the effect of the coatings on the removal processes?

The current model implementation does not consider any internal mixture between minerals and other species, therefore coatings are not accounted for on the removal processes.

This, together with the comment on I212, is now clarified in the manuscript (I219-221), as follows:

“[...]At present, minerals have the same treatment in in-cloud scavenging, i.e., they are assumed to share the same solubility, and they do not chemically interact with other gas-phase or aerosol species in the model. [...]”

I.320: How did you use the Noah model and aerosol climatological data? Please rephrase this sentence.

The paragraph read:

“The atmospheric meteorological variables are initialized from the ERA-Interim reanalysis (Berrisford et al., 2009; Dee et al., 2011) every 24 hours, to keep the modelled circulation close to

observations and hence allow for a more precise comparison of the dust cycle to observations. Soil temperature and moisture, as well as aerosol fields are recycled everyday.”

The last part has been now re-written to (I324-328):

“The atmospheric meteorological variables are initialized from the ERA-Interim reanalysis (Berrisford et al., 2009; Dee et al., 2011) every 24 hours, to keep the modeled circulation close to observations and hence allow for a more precise comparison of the dust cycle to observations. However, the soil temperature and moisture, as well as the mineral dust fields, are those prognostically estimated by MONARCH (i.e., every day the model uses as initial state for these variables their modeled value at 24.00 h of the day before).“

I.358: How did you consider the seasonality of the contribution of iron from dust?

We applied the filter to minimize the anthropogenic influence on the observations at the annual scale. Our evaluation results show little sensitivity to its application. We attribute this insensitivity to the fact that most of the observations in Perlwitz et al. (2015) are located close to dust sources, where dust contribution is high.

We agree with the reviewer that the filtering method could be refined accounting for the seasonality of dust, which would affect our classification of “anthropogenically” influenced observations. However, given the characteristics of the observational data, this refinement would not have a significant effect on the results. We have slightly rephrased the methods section to clarify that the filter is applied on an annual basis, the new sentence reads as follows (I366-367):

“We assume that mineralogy is dominated by dust in those areas where the annual mean contribution of iron from dust to the total iron concentration is of 75 % or larger (see Section 2 in the Supplementary Materials).”

I.404: Please show the refractive index in a supplementary table to compare with the measurements for hematite and goethite.

The refractive indexes for the different mineral components (host minerals, pure iron oxides and mixtures of iron oxides with other minerals) and for the compositionally homogeneous mineral dust used in this study have been added to section 3 of the Supplement. This information has been referenced in the main text accordingly.

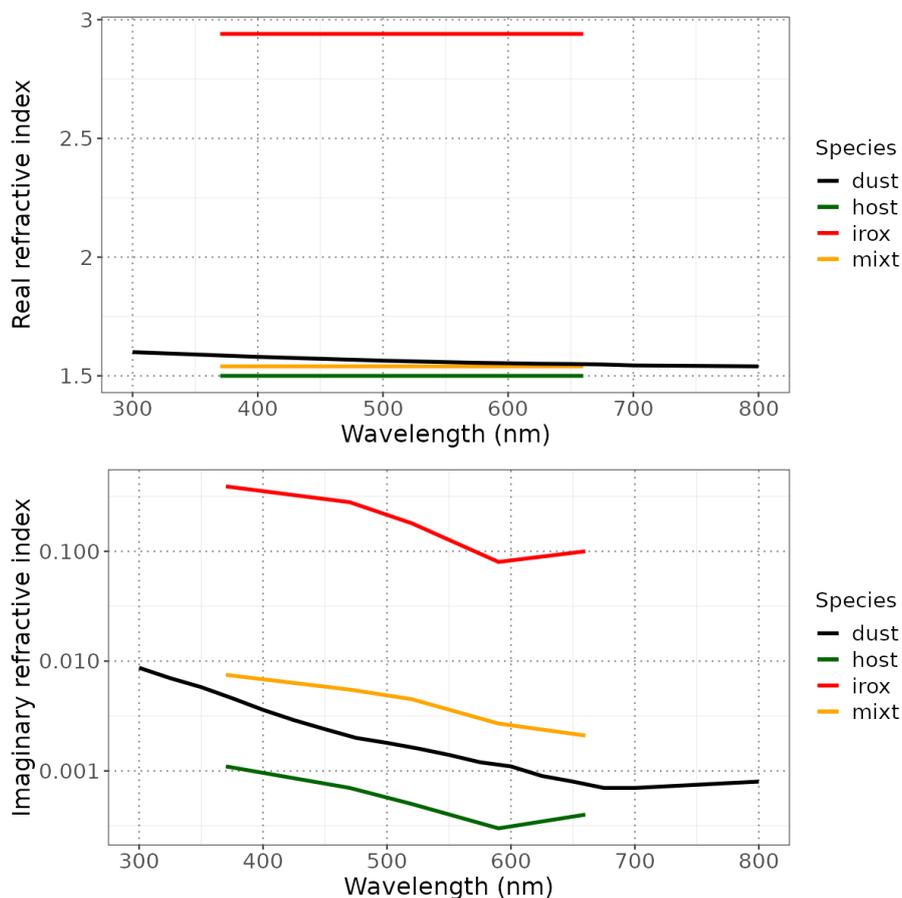


Figure S2. Real and imaginary part of the refractive index used in this work for compositionally homogeneous dust, pure iron oxides, host minerals and mixtures of iron oxides and host minerals (see main text for details).

I.431: Please specify the differences in the evaluation of the dust cycle between homogeneous dust and sum of the minerals.

The differences in surface concentration and mass deposition fluxes between the homogeneous dust and the sum of the minerals included in the simulations are negligible. Our current implementation accounts for the effect of the density and size distribution of the different minerals in the transport and deposition processes. However, these alone do not substantially change the dust cycle (when we sum the minerals mass as compared to the definition of a compositionally homogeneous dust tracer). We have included a comment in the main manuscript text to explain this fact (I456-459):

“Our current implementation accounts for the effect of the density and size distribution of the different minerals in the transport and deposition processes, however, these alone do not substantially change the dust cycle (when we sum the minerals mass as compared to the compositionally homogeneous dust tracer).”

I.490: The total iron content at emission should be compared with other models which used C1999 or J2014NN. Please indicate iron content for accretion at emission.

The total iron content at emission is reported in section 6 of the submitted manuscript. There, we had included a comparison between this iron content and the relatively common 3.5w% value used by other models. We have complemented that section with additional references to compare our estimates:

“On the other hand, dust is known to be a major source of iron to the atmosphere, which is relevant for ocean biogeochemistry upon deposition. The choice of a specific soil map has a large impact on the total iron estimates (Figure 12a and b). The C1999 experiment emits 70.23 Tg/yr of iron on average for 2006-2010, which globally represents a 2.0 w% of the total dust emitted mass, while J2014 derives in 97.64 Tg/yr, a 2.8 w%. In J2014NN, as we account for the fraction of dust with an unknown composition, to which we do not attribute any iron content, thus the total iron emitted adds up to 89.5 Tg/yr (i.e., 2.6 w% of dust). All of them fall below the 3.5 w% iron-to-dust ratio commonly assumed in modelling studies that do not resolve dust mineralogy (e.g., Luo et al., 2008; Mahowald et al., 2009; Ito and Xu, 2014). Mineralogy resolving models produce higher iron-to-dust ratios when they represent smaller size ranges than MONARCH (e.g., Scanza et al. 2018, three of the models in Myriokefalitakis et al. 2018; Hamilton et al. 2019; Bergas-Massó et al. 2023 or Wang et al. 2015 emit dust up to 10 μm in diameter with 3.2 to 4.6w% of iron). Models sharing similar size ranges (e.g., the IMPACT model in Myriokefalitakis et al. 2018, traces dust particles up to 20 μm in diameter) result in similar mass ratios (i.e., 2.65w%). This enrichment in small size ranges is attributed to the high abundance of phyllosilicates with large structural iron content (see Table 1). Finally, the use of J2014 (or J2014NN) yields high iron at emission for comparable size ranges due to the contribution of iron-rich minerals (i.e., chlorite and vermiculite) which are not represented in C1999, and also to the higher illite and smectite mass fractions in J2014.”

I.609: Please specify the differences from Fig. 10 (b), which showed lower hematite levels in J2014 than those in C1999 over Izaña (Tenerife, Spain).

Thanks to the reviewer comment, a small bug was found in the evaluation script used for the J2014 case, which used modelled goethite instead of hematite for the comparison. We corrected this bug and the resulting hematite evaluation is presented in Figure R1 below. The discussion and the figures of the main manuscript have been modified accordingly (I631-633):

“For these spots, located or influenced by African sources, both C1999 and J2014 underestimate the hematite content, however C1999 shows a slightly lower error and higher correlation than J2014 (Figure 11a and c).”

Unfortunately, this bug has also an effect on the other J2014 evaluation plots. The differences with the submitted version are minimal and they do not induce any change in our discussion. The relevant panels for J2014 of Figures 7 and 9 have been updated in the corrected manuscript.

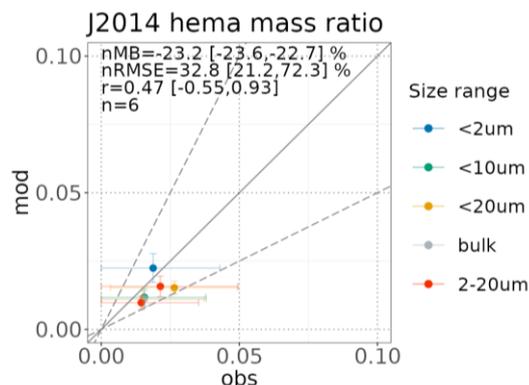


Figure R1. Evaluation of the J2014 hematite modelled mass fraction against observations.

I.613: Why don't you show the comparison of iron content? The comparison of iron concentration is also recommended.

Following the reviewers' suggestion, we have produced an evaluation of the total iron concentration against the observations compiled in Myriokefalitakis et al., 2018. The new section "4.4. Total iron from dust and evaluation" explains the methodology followed to estimate offline the total iron content and the evaluation procedure. A new panel has been added to Figure 2 to locate the total iron observations. Section 6 includes now a description of the evaluation of the iron fields for the C1999 and J2014 experiments, as well as for a homogeneous dust reference using a 3.5w% of iron, which is now shown in Figure 13 (I682-688).

"The comparison of our diagnosed total iron with surface concentration observations suggests that C1999 underestimates the elemental iron present in dust. As a result of the higher iron-to-dust ratios, this bias is slightly reduced when we consider J2014NN (see section 7 in the Supplement) and gets closer to zero with J2014 on average. On the contrary, applying a constant 3.5w% of iron in dust would lead to an overestimation of the observed total iron surface concentration (Fig. 13). The spatio-temporal variability of the modelled iron is primarily driven by the ability of the model to derive the dust fields, for that reason, the correlation of the diagnosed iron surface concentration is close in all the experiments and in the case of the homogeneous dust."

The evaluation of J2014NN (see Figure R2) shows a slightly larger underestimation of the total iron content than J2014, but lower than C1999. This is explained due to the abundance of phyllosilicates rich in iron, as compared to C1999, and to the presence of a "other" class mineral, to which we don't assign any iron content. As this two characteristics are already mentioned in the manuscript, and to maintain consistency with the rest of the evaluations, we have included the results of J2014NN in section 7 of the Supplement.

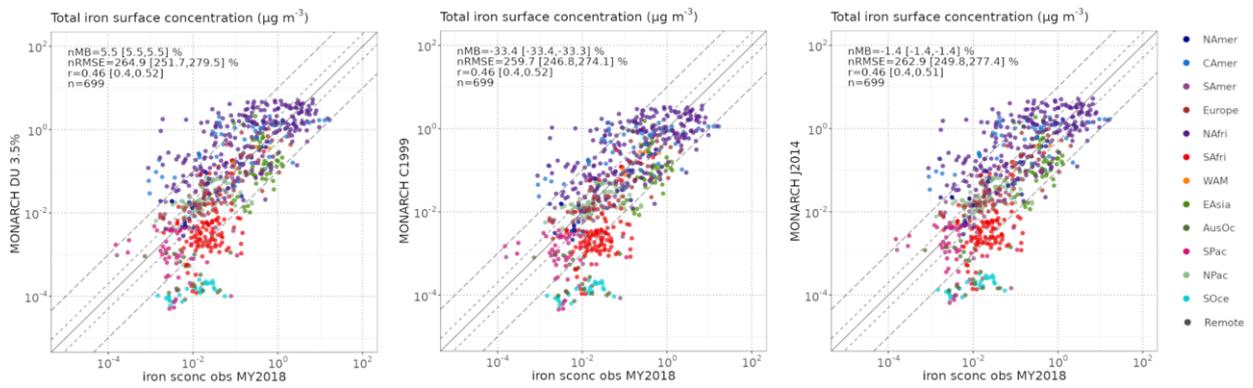


Figure 13. Diagnostic of the total iron surface concentration ($\mu\text{g m}^{-3}$) associated to the MONARCH modelled (a) dust with an average 3.5w% of iron content, (b) C1999 and (c) J2014 dust mineralogy compared the observations compiled in Myriokefalitakis et al. (2018) identified as dust-dominated (see Fig. 1c and section 4.4). Where n is the number of measurements, nRMSE, the normalized Mean Root Square Error, nMB, the normalized Mean Bias, and r , the correlation, over all points. The ranges for the nMB, nRMSE and r correspond to the 95% confidence level (see Section 1 of the Supplement for details). Regions are depicted in Figure 1c. Ashed lines represent differences of 2 times and one order of magnitude.

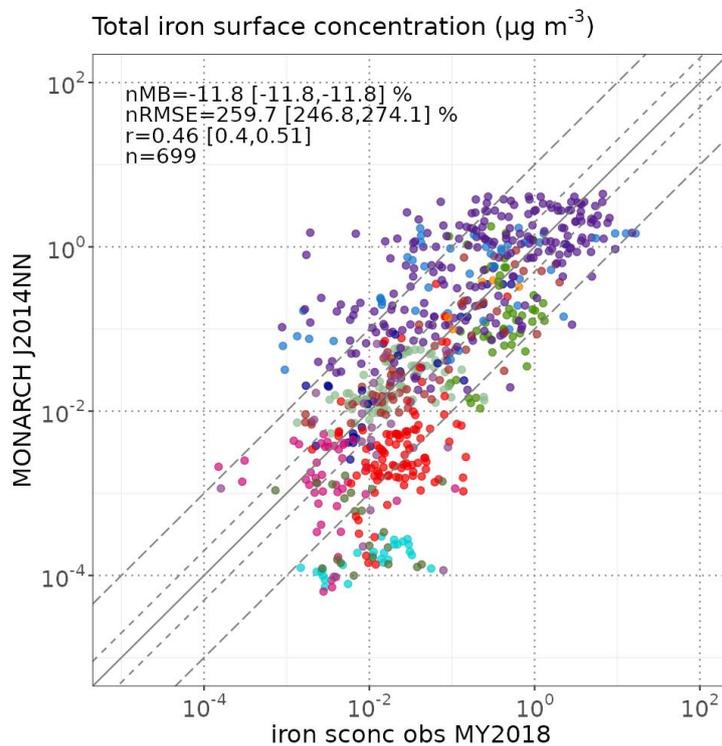


Figure R2. Evaluation of the total iron surface concentration estimated from the J2014NN mineralogy against the observations in Myriokefalitakis et al. (2018).

I.652: The total iron content at deposition should be compared with previous studies which used C1999 or J2014NN. Please indicate iron content for accretion at deposition. Please also show J2014NN.

We have now included in the manuscript an estimate of the total iron deposition budgets and compared them with other modelling studies (I689-697):

“In line with the emission, globally the amount of total iron deposited in J2014 (and J2014NN) is larger than in C1999, with 95.0 (87.0) Tg/yr (Figure 14a and b, and section 7 of the Supplement). In all experiments, dry deposition is the predominant removal process (representing 57% and 58% of the total iron deposition in C1999 and J2014, respectively). These estimates fall within those reported in previous modelling studies (71.5 +/- 43 Tg/yr) applying both C1999 and J2014NN (Myriokefalitakis et al., 2018), which also highlights the relevance of dry deposition for dust-originated iron removal. Regionally, the largest differences in total iron deposition between C1999 and J2014 occur over the Atlantic and Indian oceans, while over the Southern Ocean (and in general the Southern Hemisphere basins), both soil mineralogy maps yield similarly low total iron deposition fluxes. Our model tends to underestimate the total deposited dust mass in this region (see Fig. 4 and Table S02 in the Supplement), which has a direct impact on the estimated total iron deposition fluxes.”

The total iron deposition from J2014NN has been added to Section 7 of the supplementary materials. In all our experiments the largest mass of iron oxides is transported in accreted form. Free iron oxides constitute a 3w% (in J2014 and J2014NN) to 4w% (in C1999) of the total deposited iron from iron oxides. Independently of the mixing state we assume for the transport of iron oxides, both accreted and free iron oxides are considered as part of the crystalline FeOx used for our diagnostic of the solubility of iron from dust (Figure 14). To avoid confusion to the reader, we leave the information about the distribution in accreted and free forms out of the manuscript.

I.708: Please discuss the reasons of the excessive allocation of quartz to the coarser sizes.

Unfortunately, the soil maps available only distinguish between two soil size classes (clay up to 2 μm in diameter) and silt (from 2 to 63 μm in diameter). Quartz is more abundant the coarser the particle. Therefore assuming the same fraction of quartz in soil sizes from 2 to 63 μm in diameter will likely provide an unrealistically high fraction at diameters close to 2 μm , and the opposite in the largest diameter sizes. With the application of the Brittle Fragmentation Theory for dust emission to derive the emitted minerals' size distribution, we correct for the overestimation of phyllosilicates in fine aerosol fractions and, indirectly, we partially improve the overestimation of quartz in the silt sizes (see Figure 1 in the main article). However, our results still show an excessive allocation of quartz for those aerosol sizes greater than 2 μm .

This has been further clarified in the article conclusions (I745-749), as follows:

“Even with this re-aggregation, in all our experiments, the model overestimates the observed quartz mass fractions at coarse sizes, and underestimates those in the finer ones (below 2 μm). This bias is attributed to the poor representation of the minerals' size distribution in the soils, which neglects the increased abundance of quartz with size. The SMAs provide a constant quartz proportion for soil grains from 2 up to 63 μm in diameter, which is likely too large for the smaller silt particles. As a consequence of the overestimation of the relative fraction of quartz, the model underestimates other typically coarse minerals over the same size ranges, e.g., feldspars.”

RC#2

In this study, multiple soil mineralogy maps were input to a global atmospheric chemistry model, and calculations were performed to reproduce atmospheric mineral dust for each soil composition and validated using available observational data. In most conventional climate models, mineral dust composition is assumed to be globally uniform and its optical properties are independent of its origin. Therefore, although this study is limited to the validation of single scattering albedo, it is expected to be carried over to the next step of researches, which are to improve the accuracy of the specific assessment of climate change due to mineral dust. Overall, the manuscript is well written, but I recommend the following revisions to ensure accurate information to the reader.

L107: “Despite these uncertainties, there are evidences that the spatio-temporal variability in modelled dust absorption when considering the soils’ varying composition gets closer to that observed, independently of the soil information used (Obiso et al., in prep.a, a).” It is not clear on the meaning of this sentence. In the immediately preceding sentence, it is stated that differences in iron oxide content between regions are important, but does this mean that radiative absorption by mineral dust is independent of soil conditions? If that was the case, then it would be reasonable to assume that the optical properties of mineral dust are uniform across the globe, and the significance of this study would be lost. The authors should rewrite the sentence so as not to mislead the reader.

The sentence has been rephrased (I107-109), to avoid confusion, it now reads:

“Despite these uncertainties, accounting for the soil's varying composition improves the spatio-temporal variability in modelled dust absorption as compared to observations (Obiso et al., 2023).”

L111: “... proving that these estimates have a positive impact on the modelled ice nucleating particles as compared to observations,” It is not clear what "positive impact" means. Please explain in detail how the ice nucleating particles are affected.

The sentence has been clarified as follows (I110-113):

“Some models already incorporate explicitly the number concentration of K-feldspar and quartz in the atmosphere (Atkinson et al., 2013; Vergara-Temprado et al., 2017; Chatziparaschos et al., 2023) to derive the concentration of ice nucleating particles (INPs). Their evaluation against observations suggests improvements in the geographical distribution of the INPs (Atkinson et al., 2012), as well as their relevance at the global scale (Vergara-Temprado et al., 2017; Chatziparaschos et al., 2017).”

L194: Does the MONARCH solve for time evolution using not only the mass mixing ratio of each bin size, but also that of each mineral composition as independent prognostic variables? Please note this clearly as it is a technically important point.

Yes, MONARCH accounts for the 9, 14 or 15 minerals (including free iron oxides and accreted iron oxides) in 8 bin sizes as independent variables (meaning, to represent mineralogy with C1999 involves adding 72 tracers, J2014, 112, and J2014NN, 120). Our model has been configured so that the user can select specific minerals to be represented, and therefore optimize the computational burden for certain applications. This has been further clarified in the manuscript, following the reviewer’s comments (I201-202):

“[...]The representation of the minerals’ size distribution in MONARCH follows that of the compositionally homogeneous dust, i.e., each mineral will be represented by 8 tracers with diameters ranging from 0.2 to 20 μm .[...]”

L404: Information on the optical data of the mineral dust used in this study is important to the validity of this study. Please illustrate the wavelength distribution of the complex refractive index for each mineral composition and iron oxide.

This has been integrated together with the response to reviewer#1 comment. The refractive indexes used in this study to compute the SSA have been plotted and added to a Table, both are now shown in section 3 of the Supplement and appropriately referenced in the main text.