# Response to review

In my last comment there were two questions to be explained: the linearity of the system and the usage of standard deviation in the analysis of climate variability.

For the first question the authors included an additional experiment with the endmember effect set to 0 and used it to confirm the linearity. I agree that the system seems to be linear and the differences between two calculation ways are negligible. However, I expected an explanation why a system with processes which obviously interact work approximately in a linear way. I suppose that not the model setup causes the linearity but the nature of the Fe isotope system. A good example for this approximation is the system of carbon isotopes (e.g. Hayes 1982). Therefore, I suggest the authors to make it clear if and which model setup leads to the linearity or for which system such an approximation can be made.

Hayes, J.M., 1982. Fractionation et al.: An introduction to isotopic measurement and terminology. Spectra, 8(4), 3–8.

We are glad that the reviewer agrees that we have demonstrated that the system is linear. The references to the Hayes summary was not clear, but seems to ask whether, as for carbon, we can sum describe the isotope system as a linear sum of different components or whether we have imposed this linearity through our model parameterisations.

Ultimately, the set up of the model is responsible for this linearity in the modelled iron isotope system and we have observed this for all different experimental set-ups tested so far. In responding to this, we note that fractionation factors are constant (i.e., independent of Fe concentration) and only applied to Fe transferred between two Fe pools for each time step, i.e., there is no equilibration between Fe isotope pools. Similarly, source endmembers are constant (in time) and applied to the input of Fe independent of flux strength or other factors. To exemplify this approach, a simplified version of how light ($^{54}Fe_{diss}$) and heavy ($^{56}Fe_{diss}$) tracers of dissolved Fe are modelled as shown below (eq. 1-4) was added to the supplement. Note that Fe supply by other external sources are modelled similarly to dust supply, and other non-fractionating internal transformation processes are modelled similarly to remineralisation. Fractionation during Fe complexation is modelled similarly as uptake fractionation whereby the inverse of its fractionation factor is applied to the scavenged Fe flux, as scavenging only removes uncomplexed Fe. Other tracers (phytoplankton Fe, Fe particles) are modelled similarly as dissolved Fe, although without any external supply.

$$\frac{\partial^{56}Fe_{diss}}{\partial t} = \overset{\text{External Fe supply}}{\boxed{J_{dust,Fe} * \frac{R_{dust,Fe}}{R_{dust,Fe}+1}}} \overset{\text{Int. trans. without fractionation}}{\boxed{+J_{remin,Fe} * \frac{R_{Fe,part}}{R_{Fe,part}+1}}} \overset{\text{Int. transf. with fractionation}}{\boxed{-J_{up,Fe} * \frac{R_{Fe,diss} * \alpha_{up}}{R_{Fe,diss} * \alpha_{up}+1}}} \qquad (1)$$

$$\frac{\partial^{54}Fe_{diss}}{\partial t} = \boxed{J_{dust,Fe} * \frac{1}{R_{dust,Fe}+1}} \boxed{+J_{remin,Fe} * \frac{1}{R_{Fe,part}+1}} \boxed{-J_{up,Fe} * \frac{1}{R_{Fe,diss} * \alpha_{up}+1}} \qquad (2)$$

$$R_{dust,Fe} = \frac{^{56}Fe_{IRMM-014}}{^{54}Fe_{IRMM-014}} * \left(\frac{\delta^{56}Fe_{dust}}{1000}+1\right) \qquad (3)$$

$$R_{Fe,part} = \frac{^{56}Fe_{part}}{^{54}Fe_{part}} \qquad (4)$$

$$R_{Fe,diss} = \frac{^{56}Fe_{diss}}{^{54}Fe_{diss}} \qquad (5)$$

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|---|---|
| $J_{dust,Fe}$ | dissolved Fe supply rate from dust |
| $R_{dust,Fe}$ | Isotope ratio of supplied dust Fe |
| $\delta^{56}Fe_{dust}$ | Endmember signature of dust Fe supply |
| $J_{remin,Fe}$ | dissolved Fe supply by remineralisation of Fe particles |
| $R_{Fe,part}$ | Isotope ratio of Fe particles, see eq. 5 |
| $^{56}Fe_{part}$ | Heavy particulate Fe concentration |
| $^{54}Fe_{part}$ | Light particulate Fe concentration |
| $J_{up,Fe}$ | dissolved Fe uptake by phytoplankton |
| $R_{Fe,diss}$ | Isotope ratio dissolved Fe, see eq. 4 |
| $\alpha_{up}$ | fractionation factor for phytoplankton Fe uptake |
| $^{56}Fe_{diss}$ | Heavy particulate Fe concentration |
| $^{54}Fe_{diss}$ | Light particulate Fe concentration |

Note that the linearity of the iron isotope system may be challenged by developments in the modelling of iron isotopes in the future and new work (based on either refined modelling or new observations) may depart from these assumptions. Unfortunately, a discussion of this linearity, and where or when it may not apply, is beyond the scope of this study, which instead focusses on interannual variability in the cycling of iron, iron limitation and iron isotopes. But to respond to the reviewers comment, we have added the full system of equations and associated text to the supplement.

Unfortunately the second question can not be answered with the linearity. By combining independent variables (the central limit theorem), standard deviations of variables can not be added together but variances. Therefore, Eq. 4 is wrong. Here I suggest to use the root of the sum of variances of three distributions instead of the sum of SDs, and redo the analysis of climate variability.

We agree with the reviewer that the calculation of the “contribution” of $\delta^{56}Fe_{EM}$, $\delta^{56}Fe_{UF}$, and $\delta^{56}Fe_{CF}$ to $\delta^{56}Fe_{diss}$ as outlined in equation 4 is not ideal. However, we note that using the root of the sum of the three variances would also lead to misleading values, since there is a degree of covariance between the three components, as their temporal response to climate variability can be aligned (or opposing). Thus, we decided to use the ratio between

the variance of each component to the variance of $\delta^{56}Fe_{diss}$ as an indicator of their relative importance in contributing to $\delta^{56}Fe_{diss}$ variability, and also show the covariance between $\delta^{56}Fe_{EM}$ and $\delta^{56}Fe_{UF}$, as they are generally more variable than $\delta^{56}Fe_{CF}$. Note that we did not replace standard deviation with variance for plots that illustrate the variability in $\delta^{56}Fe_{diss}$, dFe concentration and primary productivity (Fig. 1a, Fig. 2a,b, Fig. S2) as it allows for easier comparison between the variability of a parameter and its average, or between the degree of variability for two model experiments.
We updated Section 2.3 as follows:

Line 137 of the revised manuscript:

“To compare the interannual variability of each of the three components to $\delta^{56}Fe_{diss}$ variability, we calculated their “relative variability”, i.e., the ratio between their VAR and that of $\delta^{56}Fe_{diss}$, where subscript i=UF, CF or EM (Eq. 4).

$$\delta^{56}\mathrm{Fe_i}\ \text{relative variability} = \frac{\mathrm{VAR}(\delta^{56}\mathrm{Fe_i})}{\mathrm{VAR}(\delta^{56}\mathrm{Fe_{diss}})} \qquad (4)$$

Note that the sum of the three ratios are lower (higher) than 1 in areas with positive (negative) covariance between two or three of the components, i.e., where their response to climate variability are reinforcing (opposing) each other.”

We also updated the sections 3.1 and 4 of the manuscript accordingly (see tracked version of the manuscript), namely by adding the relative variability of each component and the covariance between $\delta^{56}Fe_{EM}$ and $\delta^{56}Fe_{UF}$ to Figures 1, 2, S5, and S11. Note that this new approach left Fig. S3 and Fig. S6 (which showed standard deviations of drivers) obsolete, so that they were removed. We also updated Fig. S4 (now Fig. S3) to show both examples of negative and positive covariance between parameters. Note that the major results and conclusions drawn are not changed.