We thank the reviewer for their thoughtful comments which have improved the manuscript. New manuscript text is italicized in the replies.

**Comment:** 1. Lack of observations and sensitivity to averaging are cited as sources of uncertainty in evaluating modeled soluble fraction. Is it possible that other drivers are important here? For example, does the presence or absence of other chemical species, or in-correct species distributions in the model, affect modeled iron solubility? In addition to evaluating sensitivity to averaging techniques, it makes sense to evaluate soluble fraction sensitivity to emissions of other soluble species.

**Response:** There are other factors beyond the analytical method under which iron solubility is calculated causing uncertainty. We feel we have covered many of the most important points in a detailed manner within L493-500 and the dedicated Future directions Section 5. The undertaking of many more detailed sensitivity simulations is beyond the scope of this model description paper; but this is an excellent suggestion for a follow-on paper within a more relevant journal (e.g., ACP). We wish to convey the Authors comments though and add the following text in opening paragraph to Section 5,

"In general, improving the modelled representation of secondary organic aerosol (including oxalate) and aerosol pH, particularly for remote regions, is an important task for aerosol modeling and one which have co-benefits for iron aerosol modelling. Comparisons of soluble fraction of other aerosol species with observations could also be used to guide model development."

**Comment:** 2. The explanation of results throughout the paper would benefit from the inclusion of additional quantitative information. While the figures are very comprehensive, highlighting more quantitative outcomes within the text would strengthen the paper.

**Response:** We have made the manuscript more qualitative by including more information from Tables and Figures within the text.

**Comment:** 3. Table 3: What is the fate of the remaining fraction of each mineral treated in the model?

**Response:** The remaining mineral fractions are advected as their respective mineral species. New Table 6, as suggested by R1, helps further highlight this for the reader by including the number of advected tracers,

"Table 6: Simulation time (in seconds per simulated year) for the CESM-MAM4 model. The CAM5 base model, with the addition of dust minerology, and with the addition of dust minerology and iron processing (i.e., MIMI v1.0) shown in black text. Cost of running the new higher resolution CAM6 model with dust minerology also shown for comparison in blue text. All CAM5 simulations executed on 10 nodes, with 36 cores per node, for two years (2006-2007) with consistent output fields."

		CAM5		CAM6
	MAM4	MAM4DU8	MAM4DU8FE6	MAM4DU8
	(Base model)	(dust minerology)	(MIMIv1.0)	(dust minerology)
Number advected aerosol species	24	45	63	46
Gridcell resolution (#lon x #lat)	144x96	144x96	144x96	288x192
Wall clock s a <sup>_1</sup> (simulation)	3954	5856	7836	20167
Core-hours	396	586	784	2017

and we also add the following text to the header of Table 3,

"Residual mineral dust mass is then advected as its respective tracer."

Note that this is covered in the main text already in L216-220 and so do not add any new text to the body of the manuscript.

**Comment:** 4. Table 4: Indicate in the header that these are fire emissions ratios.

**Response:** Added at end of header.

"Modelled fire emission ratio for Fe:BC then calculated from observed ratios."

Comment: 5. Line 344: Should this be statistically?

Response: Yes, thank you.

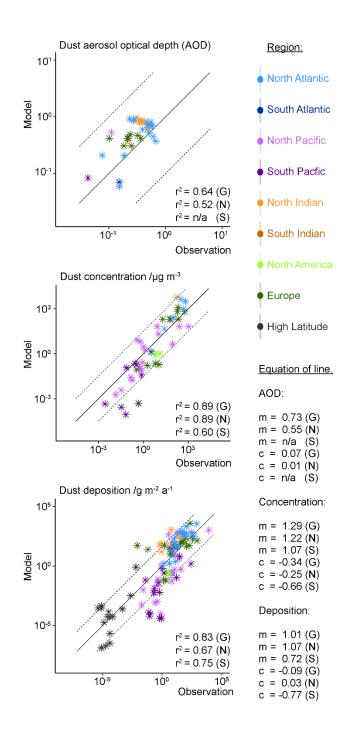
**Comment:** 6. Line 420, Figure 2: It would be more informative to include an additional table of slopes, intercepts, etc. for each region and for all regions combined.

**Response:** As Figure 2 has limited points per region (and so regional statistics would not be robust) we have included Hemispheric level details for Figure 2. However, we think this an excellent suggestion for a regional scale evaluation of total iron for Figure 4.

New text for Figure 2,

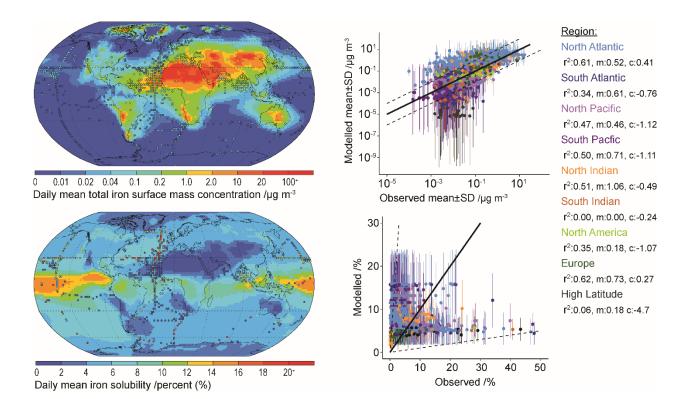
Globally, both dust concentrations (correlation:  $r^2 = 0.89$ ) and deposition (correlation:  $r^2 = 0.83$ ) are simulated well compared to observation within MIMI. A higher correlation of modelled dust concentrations with observations is calculated in the Northern Hemisphere (NH;  $r^2 = 0.89$ ) compared to the Southern Hemisphere (SH;  $r^2 = 0.67$ ), but with gradient of line of best fit is further from 1:1 (NH: 1.22 vs. SH: 1.07). Conversely, for dust deposition a lower correlation with observations is simulated in NH ( $r^2 = 0.75$ ) compared to the SH ( $r^2 = 0.60$ ) but with a gradient of the line of best fit closer to 1:1 (NH: 1.07 vs. SH: 0.72).

Updated Figure 2:



"Figure 2. Dust aerosol optical depth, surface concentrations and deposition in modal aerosol model and observations (Albani et al., 2014; Holben et al., 2000). Correlation ( $r^2$ ), gradient (m) and intercept (c) shown for global (G), Northern Hemisphere (N) and Southern Hemisphere (S) regions."

## Updated Figure 4:



**Figure 4.** Daily mean model total iron concentration and solubility from 2007 to 2011. Observations (circles) overlaid (at resolution of the model grid) as a mean from 1524 individual records in Mahowald et al. (2009) and in Myriokefalitakis et al. (2018). Also shown are scatter plots of the model mean and standard deviation compared to each available observation and identified by oceanic region. *Correlation (r<sup>2</sup>), gradient (m) and intercept (c) for total iron with observations shown for each region.* 

**Comment:** 7. Figure 4: Label the scatter plots as mean and standard deviation.

Response: Added (see above).

Comment: 8. Line 547: "...differences between method are not insignificant..."

Response: Altered as suggested.

Comment: 9. Lines 567-572: Repeated text.

Response: Removed repeated text.

**Comment:** 10. Lines 572-574: As written, this sentence could be interpreted as, the ratios of tails only exist in certain regions. Whether narrow or wide, many distributions will have tails. Perhaps rewriting the sentence to indicate that extreme ratios of tails are found in specific regions would eliminate ambiguity.

Response: Altered sentence as suggested, now reads as,

"The *extreme* ratio of the tails of soluble and total iron are only found *in specific* regions with the highest temporal variability [...] "

Comment: 11. Line 610: "...the iron it contains is~120% higher..."

**Response:** Altered as suggested.

**Comment:** 12. Lines 616-617: This designation of fire emissions as combustion emissions here is inconsistent with the emissions categories presented in the rest of the paper.

**Response:** We agree that keeping consistency throughout is important to keep the reader orientated. We have removed the reference to combustion and now refer to the sum of fires and anthropogenic combustion as pyrogenic iron both here and throughout the manuscript where confusion could occur.

**Comment:** 13. Line 690: The first instance of acronym should be spelled out.

**Response:** Line 40 contains first instance of acronym and is spelled out.

**Comment:** 14. Lines 692-694: Was the sensitivity to vertical resolution near the surface tested in this study? If not, please cite a reference here.

**Response:** It was not and to our knowledge no study in the literature has explicitly examined this for dust deposition (but have for PM10 and the vertical profile). Removed previous sentenced and replaced with new text,

"The dry deposition flux is sensitive to the aerosol properties, surface roughness and modelled turbulence. Although increasing the vertical resolution has been shown to increase surface  $PM_{10}$  concentration (Menut et al., 2013) and better simualte the dust vertical profile (Teixeira et al., 2016), it is not as yet clear if this would correspondingly increase the dry deposition flux."

Comment: 15. Line 709: The first instance of acronym should be spelled out.

**Response:** This is the only instance of said acronym in the main text and so remove it and just state as follows,

"Inter Tropical Convergence Zone"

**Comment:** 16. Section 5: This was by far the most well-written section of the paper. I found the writing of the majority of the other sections to be choppy and difficult to read.

**Reply:** We have included many improvements to the text from both Reviewers and made additional ones ourselves where we could see it would help improve the manuscript.

References:

Menut, L., Bessagnet, B., Colette, A. and Khvorostiyanov, D.: On the impact of the vertical resolution on chemistry-transport modelling, Atmos. Environ., 67, 370–384,

doi:10.1016/j.atmosenv.2012.11.026, 2013.

Teixeira, J. C., Carvalho, A. C., Tuccella, P., Curci, G. and Rocha, A.: WRF-chem sensitivity to vertical resolution during a saharan dust event, Phys. Chem. Earth, 94, 188–195, doi:10.1016/j.pce.2015.04.002, 2016.