

Supplementary information

Table S1. Set of differential equations used in GEOS-Chem/Fe<sub>d</sub>

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|---|---|
| $\frac{d\text{Fe(II)}}{dt}$               | $= j_1\text{Fe(OH)}^{2+} + j_2[\text{Fe(C}_2\text{O}_4)_2^-] + j_3[\text{Fe(C}_2\text{O}_4)_3^{3-}] + k_7[\text{Fe(OH)}^{2+}][\text{O}_2^-]$<br>$+ k_8[\text{Fe(OH)}^{2+}][\text{HO}_2] + k_{17}[\text{Fe(C}_2\text{O}_4)_n^{3-2n}][\text{O}_2^-]$<br>$+ k_{18}[\text{Fe(C}_2\text{O}_4)_n^{3-2n}][\text{HO}_2] - k_1[\text{Fe}^{2+}][\text{H}_2\text{O}_2] - k_2[\text{FeC}_2\text{O}_4][\text{H}_2\text{O}_2]$<br>$- k_3[\text{Fe}^{2+}][\text{O}_2^-][\text{H}^+]^2 - k_4[\text{Fe}^{2+}][\text{HO}_2][\text{H}^+] - k_5[\text{Fe}^{2+}][\text{NO}_3]$<br>$- k_6[\text{Fe}^{2+}][\text{NO}_2][\text{H}^+] - k_9[\text{Fe}^{2+}][\text{O}_3]$ |
| $\frac{d\text{Fe(III)}}{dt}$              | $= -\frac{d\text{Fe(II)}}{dt}$  |
| $\frac{d[\text{FeO}^{2+}]}{dt}$           | $= k_9[\text{Fe}^{2+}][\text{O}_3] - k_{10}[\text{FeO}^{2+}][\text{H}_2\text{O}] - k_{11}[\text{FeO}^{2+}][\text{OH}^-][\text{H}^+] - k_{12}[\text{FeO}^{2+}][\text{H}_2\text{O}_2]$<br>$- k_{13}[\text{FeO}^{2+}][\text{HO}_2]$  |
| $\frac{d[\text{C}_2\text{O}_4^{2-}]}{dt}$ | $= j_2[\text{Fe(C}_2\text{O}_4)_2^-] + j_3[\text{Fe(C}_2\text{O}_4)_3^{3-}] - k_{19}[\text{C}_2\text{O}_4^{2-}][\text{OH}^-] - k_{20}[\text{C}_2\text{O}_4^{2-}][\text{NO}_3]$<br>$- k_{21}[\text{C}_2\text{O}_4^{2-}][\text{O}_2]$   |
| $\frac{d[\text{H}_2\text{O}_2]}{dt}$      | $= k_3[\text{Fe}^{2+}][\text{O}_2^-][\text{H}^+]^2 + k_4[\text{Fe}^{2+}][\text{HO}_2][\text{H}^+] + k_{11}[\text{FeO}^{2+}][\text{OH}^-][\text{H}^+] + k_{14}[\text{HO}_2][\text{HO}_2]$<br>$+ k_{15}[\text{HO}_2][\text{O}_2][\text{H}^+] - k_1[\text{Fe}^{2+}][\text{H}_2\text{O}_2] - k_2[\text{FeC}_2\text{O}_4][\text{H}_2\text{O}_2]$<br>$- k_{12}[\text{FeO}^{2+}][\text{H}_2\text{O}_2]$  |
| $\frac{d[\text{HO}_2]}{dt}$               | $= k_{12}[\text{FeO}^{2+}][\text{H}_2\text{O}_2] - k_4[\text{Fe}^{2+}][\text{HO}_2][\text{H}^+] - k_8[\text{Fe(OH)}^{2+}][\text{HO}_2] - k_{13}[\text{FeO}^{2+}][\text{HO}_2]$<br>$- k_{14}[\text{HO}_2][\text{HO}_2] - k_{15}[\text{HO}_2][\text{O}_2][\text{H}^+] - k_{18}[\text{Fe(C}_2\text{O}_4)_n^{3-2n}][\text{HO}_2]$   |
| $\frac{d[\text{OH}^-]}{dt}$               | $= j_1\text{Fe(OH)}^{2+} + k_1[\text{Fe}^{2+}][\text{H}_2\text{O}_2] + k_2[\text{FeC}_2\text{O}_4][\text{H}_2\text{O}_2] + k_{10}[\text{FeO}^{2+}][\text{H}_2\text{O}]$<br>$- k_{11}[\text{FeO}^{2+}][\text{OH}^-][\text{H}^+] - k_{19}[\text{C}_2\text{O}_4^{2-}][\text{OH}^-]$  |
| $\frac{d[\text{O}_2^-]}{dt}$              | $= k_{16}[\text{CO}_2^-][\text{O}_2] + k_{21}[\text{C}_2\text{O}_4^{2-}][\text{O}_2] - k_3[\text{Fe}^{2+}][\text{O}_2^-][\text{H}^+]^2 - k_7[\text{Fe(OH)}^{2+}][\text{O}_2^-]$<br>$- k_{17}[\text{Fe(C}_2\text{O}_4)_n^{3-2n}][\text{O}_2^-]$  |
| $\frac{d[\text{C}_2\text{O}_4^-]}{dt}$    | $= j_2[\text{Fe(C}_2\text{O}_4)_2^-] + j_3[\text{Fe(C}_2\text{O}_4)_3^{3-}] + k_{19}[\text{C}_2\text{O}_4^{2-}][\text{OH}^-] + k_{20}[\text{C}_2\text{O}_4^{2-}][\text{NO}_3]$<br>$+ k_{21}[\text{C}_2\text{O}_4^{2-}][\text{O}_2]$   |

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Table S2. Fe-species absorption cross-sections used in GEOS-Chem/Fe<sub>d</sub>

| Absorption cross-section (cm <sup>2</sup> molecule <sup>-1</sup> ) |                         |  |   |
|--|-------------------------|--|---|
| Wavelength (nm)*   | Fe(OH) <sup>2+</sup>    | Fe(C <sub>2</sub> O <sub>4</sub> ) <sub>2</sub> <sup>-</sup> | Fe(C <sub>2</sub> O <sub>4</sub> ) <sub>3</sub> <sup>3-</sup> |
| 294  | 8.8 × 10 <sup>-18</sup> | 1.7 × 10 <sup>-17</sup>                                      | 1.7 × 10 <sup>-17</sup>                                       |
| 303  | 8.4 × 10 <sup>-18</sup> | 1.5 × 10 <sup>-17</sup>                                      | 1.5 × 10 <sup>-17</sup>                                       |
| 310  | 7.6 × 10 <sup>-18</sup> | 1.1 × 10 <sup>-17</sup>                                      | 1.1 × 10 <sup>-17</sup>                                       |
| 316  | 6.5 × 10 <sup>-18</sup> | 8.9 × 10 <sup>-18</sup>                                      | 8.9 × 10 <sup>-18</sup>                                       |
| 333  | 4.8 × 10 <sup>-18</sup> | 6.7 × 10 <sup>-18</sup>                                      | 6.7 × 10 <sup>-18</sup>                                       |
| 380  | 1.1 × 10 <sup>-18</sup> | 2.5 × 10 <sup>-18</sup>                                      | 2.5 × 10 <sup>-18</sup>                                       |
| 574  | 3.8 × 10 <sup>-21</sup> | 3.8 × 10 <sup>-21</sup>                                      | 3.8 × 10 <sup>-21</sup>                                       |

\*Effective wavelengths used in Fast-J.

Absorption cross-section ( $\sigma$ ) calculated by:  $\sigma = \epsilon * 3.82 \times 10^{-21}$ .

$\epsilon$  is the molar extinction coefficient (M<sup>-1</sup> cm<sup>-1</sup>) derived from Zuo and Holgne (1992).

Table S3. Comparison of model-predicted Fe<sub>d</sub> (goethite simulations) values to ambient data

| Fe(III) (ng m <sup>-3</sup> )      |      |       |      |                  |
|------------------------------------|------|-------|------|------------------|
| <i>Measurement Campaign</i>        | R    | Bias  | RMSE | NMB <sup>a</sup> |
| MP01 (Atlantic Ocean) <sup>b</sup> | 0.75 | -0.14 | 0.03 | -31.46           |
| MP02 (Pacific Ocean) <sup>c</sup>  | 0.59 | -0.19 | 0.06 | -41.07           |
| MP03 (Atlantic Ocean) <sup>b</sup> | 0.76 | 0.96  | 1.09 | 86.72            |
| MP05 (Pacific Ocean) <sup>c</sup>  | 0.53 | -0.10 | 0.01 | -79.34           |
| Fe(II) (ng m <sup>-3</sup> )       |      |       |      |                  |
| <i>Measurement Campaign</i>        | R    | Bias  | RMSE | NMB <sup>a</sup> |
| MP01 (Atlantic Ocean) <sup>b</sup> | 0.44 | -0.19 | 0.37 | -33.89           |
| MP02 (Pacific Ocean) <sup>c</sup>  | 0.63 | -0.36 | 0.83 | -31.75           |
| MP03 (Atlantic Ocean) <sup>b</sup> | 0.74 | 0.17  | 0.31 | 43.76            |
| MP05 (Pacific Ocean) <sup>c</sup>  | 0.70 | -0.16 | 0.02 | -57.95           |

<sup>a</sup> NMB is in percent.

<sup>b</sup> Chen and Siefert (2004).

<sup>c</sup> Chen (2004).

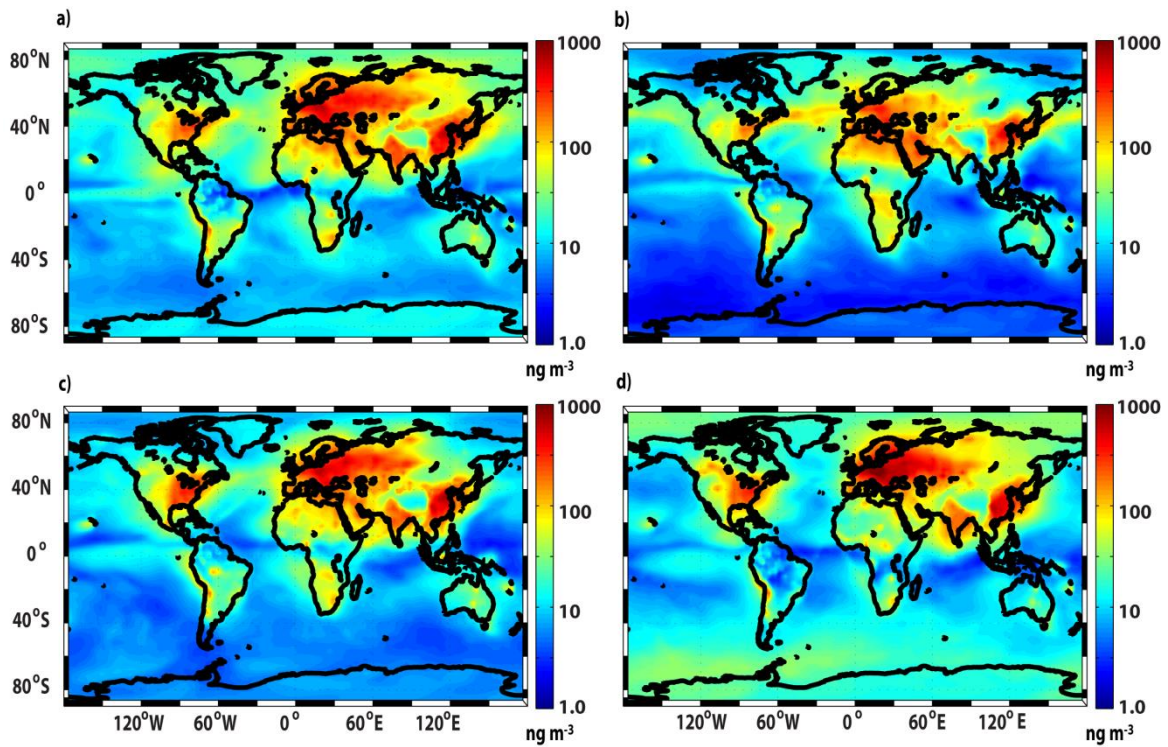
Table S4. Comparison of model-predicted Fe<sub>d</sub> values (illite simulations) to ambient data

| Fe(III) (ng m <sup>-3</sup> )      |      |       |      |                  |
|------------------------------------|------|-------|------|------------------|
| <i>Measurement Campaign</i>        | R    | Bias  | RMSE | NMB <sup>a</sup> |
| MP01 (Atlantic Ocean) <sup>b</sup> | 0.64 | 0.27  | 0.24 | 47.87            |
| MP02 (Pacific Ocean) <sup>c</sup>  | 0.60 | 0.04  | 0.02 | 9.33             |
| MP03 (Atlantic Ocean) <sup>b</sup> | 0.87 | 2.12  | 3.74 | 191.93           |
| MP05 (Pacific Ocean) <sup>c</sup>  | 0.69 | -0.02 | 0.01 | -19.27           |
| Fe(II) (ng m <sup>-3</sup> )       |      |       |      |                  |
| <i>Measurement Campaign</i>        | R    | Bias  | RMSE | NMB <sup>a</sup> |
| MP01 (Atlantic Ocean) <sup>b</sup> | 0.43 | 0.05  | 0.32 | 8.19             |
| MP02 (Pacific Ocean) <sup>c</sup>  | 0.58 | 0.24  | 0.91 | 22.14            |
| MP03 (Atlantic Ocean) <sup>b</sup> | 0.74 | 0.51  | 0.27 | 136.35           |
| MP05 (Pacific Ocean) <sup>c</sup>  | 0.81 | -0.05 | 0.02 | -21.08           |

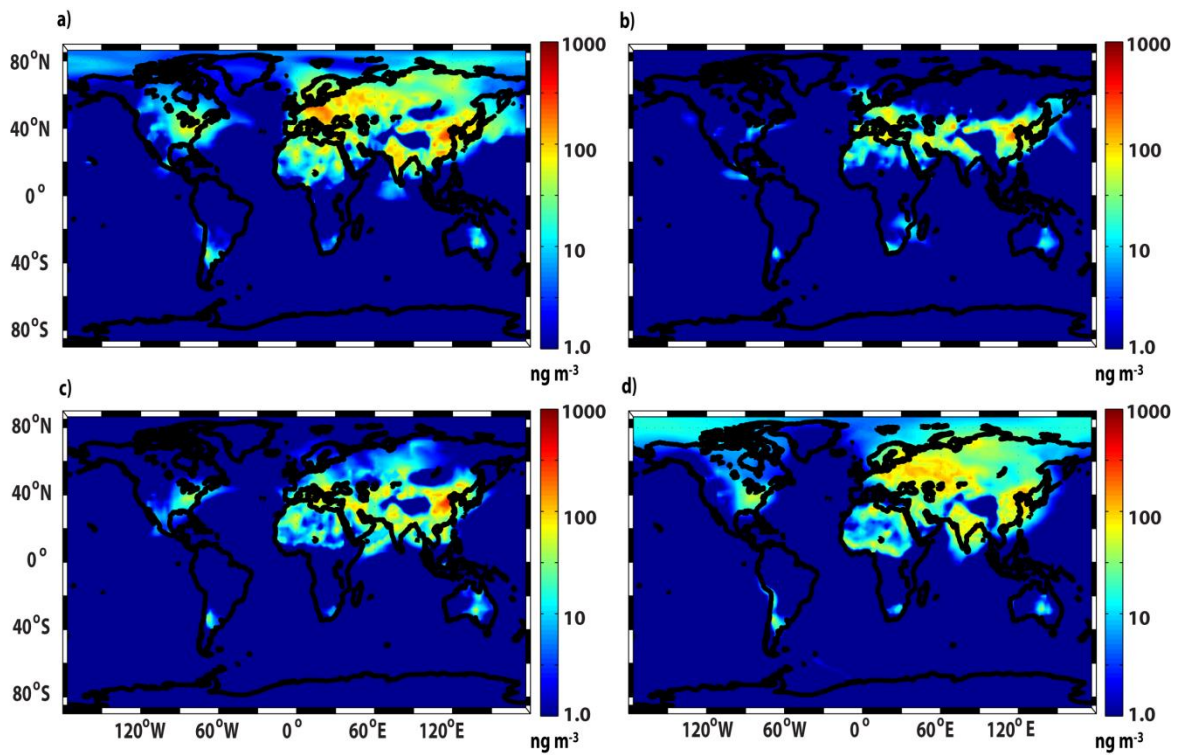
<sup>a</sup> NMB is in percent.

<sup>b</sup> Chen and Siefert (2004).

<sup>c</sup> Chen (2004).



**Figure S1.** GEOS-Chem-predicted seasonally-averaged surface level total oxalate concentration ( $\text{ng m}^{-3}$ ) for a) March-May (MAM), b) June-August (JJA), c) September-November (SON), and d) December-February (DJF).



**Figure S2.** GEOS-Chem-predicted seasonally-averaged surface level dust-oxalate concentration ( $\text{ng m}^{-3}$ ) for a) March-May (MAM), b) June-August (JJA), c) September-November (SON), and d) December-February (DJF).