

Title: Atmospheric dissolved iron deposition to the global oceans: Effects of oxalate-promoted Fe dissolution, photochemical redox cycling, and dust mineralogy

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Abstract: Mineral dust deposition is suggested to be a significant atmospheric supply pathway of bioavailable iron (Fe) to Fe-depleted surface oceans. In this study, mineral dust and dissolved Fe (Fe_d) deposition rates are predicted for March 2009 to February 2010 using the 3-D chemical transport model GEOS-Chem implemented with a comprehensive dust-Fe dissolution scheme. The model simulates Fe_d production during the atmospheric transport of mineral dust taking into account inorganic and organic (oxalate)-promoted Fe dissolution processes, photochemical redox cycling between ferric (Fe(III)) and ferrous (Fe(II)) forms of Fe, dissolution of three different Fe-containing minerals (hematite, goethite, and aluminosilicates), and detailed mineralogy of wind-blown dust from the major desert regions. Our calculations suggest that during the yearlong simulation $\sim 0.26 \text{ Tg}$ ($1 \text{ Tg} = 10^{12} \text{ g}$) of Fe_d was deposited to global oceanic regions. Compared to simulations only taking into account proton-promoted Fe dissolution, the addition of oxalate to the dust-Fe mobilization scheme increased total annual model-predicted Fe_d deposition to global oceanic regions by $\sim 75\%$. The implementation of Fe(II)/Fe(III) photochemical redox cycling in the model allows for the distinction between different oxidation states of deposited Fe_d . Our calculations suggest that during the daytime, large fractions of Fe_d deposited to the global oceans is likely to be in Fe(II) form, while nocturnal fluxes of Fe_d are largely in Fe(III) form. Model simulations also show that atmospheric fluxes of Fe_d can be strongly influenced by the mineralogy of Fe-containing compounds. This study shows that Fe_d deposition to the oceans is controlled by total dust-Fe mass concentrations, mineralogy, the surface area of dust particles, atmospheric chemical composition, cloud processing, and meteorological parameters and exhibits complex and spatiotemporally variable patterns. Our study suggests that the explicit model representation of individual processes leading to Fe_d production within mineral dust are needed to improve the understanding of the atmospheric Fe cycle, and quantify the effect of dust-Fe on ocean biological productivity, carbon cycle, and climate.

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