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Variable atmospheric iron solubility over the past 54,000 years and possible implications for atmosphere-ocean carbon cycling

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Deposition of aerosol Fe in the Southern Ocean during the last glacial period (LGP) has been linked to the coupled atmosphere-ocean carbon cycle via changes in the efficiency of the biological pump. Most Antarctic ice core studies that have explored this relationship either a) use the assumption that Fe concentrations measured at pH < 2 reflect biologically-relevant Fe (i.e., phytoplankton-accessible Fe is a constant proportion of acid-reactive Fe concentrations), or b) directly estimate biologically-relevant Fe using oceanographically defined, weak acid-leach approaches (e.g., pH = 4.5 –5.2). However, changes in modern Antarctic Fe solubility percentage (biologically relevant Fe/total Fe * 100; Fe%) appear to be coupled to atmospheric conditions (i.e., relative humidity, photoreduction) and source region minerology, indicating that the assumption of constant fractional proportions are misleading. Here we present the first record of biologically-relevant (buffer to pH 5 using ammonia acetic acid) and total (HF-HNO3 digestion) Fe concentrations and Fe% records spanning 50 –6 ka from the South Pole ice core (SPICEcore; SPC14). We use a combination of high- and low-resolution samples (866 high-resolution samples; ˜245 years per sample; and 41 low-resolution samples; ˜490 years per sample in the LGP, respectively), and compare the fractional Fe concentrations with corresponding particle concentration, size, and shape, as well as SPC14 δ18O and insolation variability. High resolution biologically-relevant Fe reaches a maximum concentration of 1.73 µg L-1 at ˜17.5 ka while maximum total Fe concentration is 81.8 µg L-1 at ˜25.9 ka. Both records are significantly related to particle concentration (biologically relevant, r = 0.7, p < 0.01; total, r = 0.85, p < 0.01). However, while both fractional concentrations increase in the LGP, Fe% values reach a minimum value of 0.53% at ˜25.6 ka and maximum value of 14.3% at 7.1 ka. Fe% has a significant negative correlation with particle concentration (r = -0.70, p < 0.01) and a weak relationship with particle size (r = 0.28, p = 0.05). Fe% has a positive correlation to SPC14 δ18O (r = 0.73, p < 0.01) and a negative log scale correlation with Fe total concentrations (r = -0.83, p < 0.01). SPC14 Fe% record has a negative correlation to insolation at 45°S and 90°S (r = -0.65, p < 0.01; r = -0.68, p < 0.01, respectively). We explore the idea that the proportion of biologically relevant Fe may be controlled by a combination of atmospheric water conditions (i.e., relative humidity) and activation of local dust source regions in Antarctica. Our findings suggest that direct measurements of biologically relevant Fe concentrations are needed to assess past relationships between aerosol deposition and potential ocean ecosystem impacts.

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