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Atmospheric dissolved iron from coal combustion particles

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It is known that mineral dust is the largest source of aerosol iron (Fe) to the offshore global ocean, but acidic processing of coal fly ash (CFA) may result in a disproportionally higher contribution of dissolved Fe to the surface ocean. In this study, we determined the Fe speciation and dissolution kinetics of CFA from Aberthaw (United Kingdom), Krakow (Poland), and Shandong (China) in acidic aqueous solutions which simulate atmospheric acidic processing. The CFA bulk samples were re-suspended in a custom-made chamber to separate the PM_{10} fraction. The Fe speciation in the PM_{10} fractions was determined using sequential extraction methods. In the PM_{10} fractions, 8%-21.5% of the total Fe was as hematite and goethite (dithionite extracted Fe), 2%-6.5 % as amorphous Fe (ascorbate extracted Fe), while magnetite (oxalate extracted Fe) varied from 3%-22%. The remaining 50%-87 % of Fe was associated with aluminosilicates. At high concentrations of ammonium sulphate ($(NH_4)_2SO_4$) and low pH (2-3) conditions, which are often found in wet aerosols, the Fe solubility of CFA increased up to 7 times. The oxalate effect on the Fe dissolution rates at pH 2 varied considerably, from no impact for Shandong ash to doubled dissolution for Krakow ash. However, high concentrations of $(NH_4)_2SO_4$ suppressed this enhancement in Fe solubility. The modelled dissolution kinetics suggest that magnetite may also dissolve rapidly under acidic conditions, as the dissolution of highly reactive Fe alone could not explain the high Fe solubility at low pH observed in CFA. Overall, Fe in CFA dissolved up to 7 times faster than in Saharan dust samples at pH 2. These laboratory measurements were used to develop a new scheme for the proton- and oxalate- promoted Fe dissolution of CFA. The new scheme was then implemented into the global atmospheric chemical transport model IMPACT. The revised model showed a better agreement with observations of surface concentration of dissolved Fe in aerosol particles over the Bay of Bengal, due to the rapid Fe release at the initial stage at highly acidic conditions.