

2.169 Atmospheric fate of anthropogenic aerosols: Iron mobility, nitrite formation, and water uptake from fly ash particles.

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Abstract:

Combustion particles, such as fly ash (FA) aerosol from coal-fired power plants, may represent a source of bioavailable iron in the open ocean, with significant implications in global biogeochemical cycles. While FA's iron content and speciation depends closely on source region and combustion process, few studies have been made to compare the atmospheric processing and iron leaching from FA from multiple sources. We present here an investigation of iron dissolution in acidic aqueous solutions for fully characterized FA samples from three different sources: US Midwestern (USFA), European (EUFA) and Indian fly ash (INFA). To better understand their atmospheric fate, water uptake on these fly ash particles was also investigated.

Under the acidic atmospheric conditions, iron was found to mobilize primarily due to proton-promoted mechanisms. Suspensions of FA in HCl resulted in larger fractions of iron leached compared to suspensions on HNO₃. In general, dissolution showed a variability between source regions with a relative iron leach in the order USFA > INFA > EUFA. A similar order was also observed in the initial rate of iron leach, suggesting that source region is a determining factor in iron leaching from fly ash. While poorly combusted samples leached a larger fraction of bioavailable Fe(II) in HCl, in the presence of HNO₃ only Fe(III) was observed, as all leached Fe(II) was rapidly oxidized, reducing nitrates (NO₃⁻) into nitrites (NO₂⁻). Because of composition variability with source region, water uptake was studied at room temperature as a function of relative humidity (RH) on fly ash. Overall, water adsorbs on the fly ash particle surface in both an ordered (ice-like) and a disordered (liquid-like) structure. A discussion is presented on water monolayer coverage on each sample as well as a comparison between surface sites of fly ash samples the corresponding enthalpies of adsorption.