Iron chemical speciation in atmospherically processed industrial aerosols

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Abstract

How human activities impact the transport of bioavailable iron to the oceans is a fundamental question addressed in atmospheric and oceanographic studies. Fe becomes more soluble during atmospheric transport and when associated with anthropogenic aerosols. However, Fe speciation in these aerosols is poorly known. The Fe solubility strongly depending on its oxidation state, this study focuses on the impact of cloud processes on the Fe chemical speciation for industrial aerosols.
The studied sample was collected on the industrial filter located in the chimney stack of a metallurgy plant (cooling area of the sintering unit). This sample was fully characterized by X-Ray diffraction, laser diffraction particle sizing and scanning electron microscopy (SEM-EDX). Weathering processes are simulated using 300±1 mg of particles into 1L of a leaching solution (80μM oxalic acid solution) mimicking cloud droplets. This represents 16.2±0.1mg of total Fe. The leaching experiment is performed for 90 minutes under simulated solar irradiation. Fe solid state chemical speciation is investigated by M´ossbauer spectroscopy before and after the leaching experiment.

In unprocessed particles, the most intense quadrupole doublet (47.7% of the total resonance pattern) of the 57Fe M´ossbauer spectrum corresponds to high-spin Fe3+ species in oxide environment. α-Fe2O3 rather well crystallized, with a slight distortion of the Fe local environment, in rhombohedral α-Fe2O3 lattice and ill-crystallized and impure magnetite Fe3O4, are the two other Fe species evidenced by M´ossbauer spectroscopy. In the magnetite octahedral sites, Fe sites are partially occupied by metallic cations of similar size (Mn2+ or Ca2+).
After simulated cloud weathering (about 0.2% of solubilized Fe), only small changes are evidenced in the α-Fe2O3 and Fe3O4 57Fe M´ossbauer spectrum hyperfine interaction parameters. This suggests that, if cloud processes can significantly affect the iron solubility, speciation in metallurgy aerosols remains unaffected by medium or long range transport atmospheric processes.

Keywords: aerosols, metallurgy, iron speciation, cloud chemistry, simulation, M´ossbauer spectroscopy

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