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Application of synchrotron radiation for measurement of iron red-ox speciation in atmospherically processed aerosols

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Abstract. In this study, ambient atmospheric particulate matter samples were collected using a size-resolved impactor sampler from three urban sites. The purpose of this study is to gain a better understanding of transformations of aerosol-bound iron as it is processed in the atmosphere. Thus, the aerosol samples were artificially aged to represent long-term transport (10 to 40 days) or short-term transport (1 to 10 days) and were measured for iron at several time points. At each time point, iron was measured in each size fraction using three different techniques; 1) inductively coupled plasma-mass spectrometry (ICPMS) for total iron, 2) x-ray absorbance near edge structure (XANES) spectroscopy for the measurement of total Fe(II) and Fe(III), and 3) a wet-chemical method to measure soluble Fe(II) and Fe(III). Prior to aging, the XANES spectroscopy results show that a majority (>60% for each size fraction) of the total iron in the PM is in the form of Fe(III). Fe(III) was shown to be a significant fraction of the soluble iron (sometimes >50%), but the relative significance of Fe(III) was found to vary depending on the site. Overall, the total soluble iron depended on the sampling site, but values ranged from less than 1% up to about 20% of the total iron. Over the course of the 40 day aging period, we found moderate changes in the relative Fe(II)/Fe(III) content. A slight increase was noted in the coarse (>2.5 μm) fraction and a slight decrease in the 0.25 to 0.5 μm fraction. The soluble fraction generally showed (excepting one day) a decrease of soluble Fe(II) prior to 10 days of aging, followed by a relatively constant concentration. In the short-term transport condition, we found that the sub-micron fraction of soluble Fe(II) spikes at 1 to 3 days of aging, then decreases to near the initial value at around 6 to 10 days. Very little change in soluble Fe(II) was observed in the super-micron fraction.

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