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Short communication

Exploration of heterogeneous chemistry in model atmospheric particles using extended X-ray absorption fine structure analysis

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Abstract

As models of the composition and heterogeneous chemical reactions of the troposphere undergo refinement, novel application of state-of-the-art analytical techniques will be necessary to propound realistic characterizations of mineral dust chemistry. In this study, strontium carbonate particles treated with gaseous nitric acid and nitrogen dioxide were examined with X-ray absorption fine structure analysis (EXAFS). The X-ray spectra of carbonate and nitrate standards were fitted to ab initio calculations, which were used to determine the structure and consistency of strontium nitrate formed on strontium carbonate. By examining differences in mean square radial displacement and lattice spacing values obtained for bulk $Sr(NO_3)_2$ as compared to $Sr(NO_3)_2$ formed on $SrCO_3$, EXAFS proves effective as a tool for investigating the local structure and composition of heterogeneous aerosol particles. The implications of findings on reacted strontium carbonate for atmospheric models of calcium carbonate aerosol are discussed.

Keywords: Atmospheric particles; Strontium carbonate; Strontium nitrate; EXAFS; Mineral dust

1. Introduction

Tropospheric mineral aerosols affect global atmospheric composition and thermal equilibrium. Efforts to understand these impacts, however, are complicated by the chemical diversity of mineral dust. Field measurements and laboratory studies have identified carbonate minerals as an important component because of their robust reactivity with gaseous nitric acid and the facile water uptake of the nitrate products. For example, calcite's conversion to hygroscopic calcium nitrate in the presence of gaseous HNO_3 can be expressed by reaction (R1):

$$CaCO_3(s) + 2HNO_3(g) \rightarrow Ca(NO_3)_2(s) + H_2O(ads) + CO_2(g) \quad (R1)$$

Including the uptake of HNO_3 on mineral dust via such reactions has improved the predictions of tropospheric chemical models (Tang et al., 2004). This reaction also has significant implications for the radiative balance as highlighted by a recent study detailing the increased activity of $Ca(NO_3)_2$ particles as cloud condensing nuclei (Gibson et al., 2006).

To clarify the tropospheric interaction of carbonates and gaseous nitric acid, numerous studies

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