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Atmos. Chem. Phys., 6, 2453-2464, 2006

www.atmos-chem-phys.net/6/2453/2006/

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## Kinetics and mechanism of heterogeneous oxidation of sulfur dioxide by ozone on surface of calcium carbonate

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**Abstract.** Sulfate particles play a key role in the air quality and the global climate, but the heterogeneous formation mechanism of sulfates on surfaces of atmospheric particles is not well established. Carbonates, which act as a reactive component in mineral dust due to their special chemical properties, may contribute significantly to the sulfate formation by heterogeneous processes. This paper presents a study on the oxidation of SO<sub>2</sub> by O<sub>3</sub> on CaCO<sub>3</sub> particles. Using Diffuse Reflectance Infrared Fourier Transform Spectroscopy (DRIFTS), the formation of sulfite and sulfate on the surface was identified, and the roles of O<sub>3</sub> and water in oxidation processes were determined. The results showed that in the presence of O<sub>3</sub>, SO<sub>2</sub> can be oxidized to sulfate on the surface of CaCO<sub>3</sub> particles. The reaction is first order in SO<sub>2</sub> and zero order in O<sub>3</sub>. The reactive uptake coefficient for SO<sub>2</sub> [(0.6–9.8) × 10<sup>14</sup> molecule cm<sup>-3</sup>] oxidation by O<sub>3</sub> [(1.2–12) × 10<sup>14</sup> molecule cm<sup>-3</sup>] was determined to be (1.4 ± 0.3) × 10<sup>-7</sup> using the BET area as the reactive area and (7.7 ± 1.6) × 10<sup>-4</sup> using the geometric area. A two-stage mechanism that involves adsorption of SO<sub>2</sub> followed by O<sub>3</sub> oxidation is proposed and the adsorption of SO<sub>2</sub> on the CaCO<sub>3</sub> surface is the rate-determining step. The proposed mechanism can well explain the experiment results. The atmospheric implications were explored based on a box model calculation. It was found that the heterogeneous reaction might be an important pathway for sulfate formation in the atmosphere.

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Citation: Li, L., Chen, Z. M., Zhang, Y. H., Zhu, T., Li, J. L., and Ding, J.: Kinetics and mechanism of heterogeneous oxidation of sulfur dioxide by ozone on surface of calcium carbonate, Atmos. Chem. Phys., 6, 2453-2464, 2006. ▣ [Bibtex](#) ▣ [EndNote](#) ▣ [Reference Manager](#)

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