



Secondary inorganic aerosol formation and its shortwave direct radiative forcing in China

Xin Huang
(xinhuang@nju.edu.cn)

Secondary inorganic aerosol (SIA), including sulfate, nitrate and ammonium, is an important part of fine particle. SIA plays a significant role in shortwave radiation transfer. Numerical simulation is usually used to study SIA formation and its climate effect. In this work, we used the Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) to study SIA formation and its direct radiative forcing (DRF) over China.

SO₂ oxidation pathways related to mineral aerosol, including transition metal-catalyzed oxidation in aqueous phase and heterogeneous reactions, play an important role in sulfate production, but they are not well treated in current atmospheric models. In this work, we improved the WRF-Chem model by simulating the enhancement role of mineral aerosol in sulfate production. Firstly, we estimated mineral cations based on local measurements in order to well represent aqueous phase acidity. Secondly, we scaled the transition metal concentration to the mineral aerosol levels according to the existing observations and improved transition metal-catalyzed oxidation calculation. Lastly, heterogeneous reactions of acid gases on the surface of mineral aerosol were included in this simulation. Accuracy in the prediction of sulfate by the model was significantly improved and we concluded that mineral aerosol can facilitate SO₂ oxidation and subsequent sulfate formation. It was demonstrated that, over China, mineral aerosol was responsible for 21.8% of annual mean sulfate concentration. The enhanced aqueous oxidation was more significant compared to the heterogeneous reactions. In winter, mineral aerosol was responsible for 39.6% of sulfate production. In summer, gaseous oxidation and aqueous oxidation of SO₂ by hydrogen peroxide and ozone were the dominant pathways of sulfate formation. Mineral aerosol only contributed 11.9% to the total sulfate production. The increase in annual mean sulfate concentration due to mineral aerosol could reach up to over 6 μg/m³ in northern China, middle and lower reaches of the Yangtze River and the Sichuan Basin.

We integrated the updated ammonia emission inventory and improved model to simulate SIA formation and its DRF over China in 2006. At the top of the atmosphere (TOA), the annual mean DRF of SIA was -2.5 W/m², in which sulfate, nitrate and ammonium contributed -1.5 W/m², -0.4 W/m² and -0.6 W/m² respectively. At the surface, the annual mean DRF of sulfate, nitrate and ammonium were -2.1 W/m², -0.5 W/m² and -0.8 W/m² respectively. In the atmosphere, the annual mean DRF of sulfate, nitrate and ammonium were +0.6 W/m², +0.1 W/m² and +0.2 W/m² respectively. The DRF of SIA at TOA displayed a distinct seasonal variation. The national mean DRF reached its maximum of -3.4 W/m² in summer. There were several reasons for the summer maximum. Firstly, enhanced atmospheric oxidizing capacity and increased ammonia emission in summer accelerated the formation of sulfate and ammonium. Secondly, high humidity favored the hygroscopic growth of SIA in summer. Lastly, shortwave radiation flux at the TOA peaked in summer. On the contrary, the hygroscopic growth of SIA was limited in winter. Simultaneously, the shortwave radiation flux decreased to the minimum in winter. Consequently, the national mean DRF reached its minimum of -1.7 W/m² in winter. SIA could play an important part in climate change by offsetting BC's radiative heating effect in China.