

## Modeling Study on Spatial and Temporal Variations of Aerosols and their optical properties around Beijing

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Regional aerosol model calculations were made using the WRF-CMAQ and WRF-chem models to study spatial and temporal variations of aerosol mass concentrations of individual components and their optical properties around Beijing, China. Model calculations were compared with in-situ intensive measurements (CAREBEIJING-2006 campaign), which had been conducted in and around the Peking University in the summer 2006, and satellite measurements (MODIS).

During one-month intensive observation period, enhancements in aerosol concentrations were observed 5 times around Beijing following by clean air events in association with a passage of the cold front. Model calculations generally reproduced well these temporal variations of aerosol concentrations as well as, their precursor gases and meteorological parameters. Model calculations also reproduced the temporal variations of aerosol optical parameters at the ground surface, vertical profiles of aerosol extinction coefficient, and column aerosol optical depth (AOD) observed around Beijing during the CAREBEIJING-2006 campaign. In addition, spatial and temporal variations of AOD obtained by the MODIS satellite sensors were reproduced well by model calculations.

Analyses on these validated model calculations show that spatial and temporal variations of primary (emitted directly into the atmosphere as a particulate form, such as EC) and secondary (formed in the atmosphere through the oxidation of gaseous precursor compounds, such as sulfate) aerosols were resulted by different mechanisms. Spatial variations of primary species reflect those of emissions, and their distinct diurnal variations with a maximum between mid-night and early morning are likely due to lower planetary boundary layer height during the night. Further analyses show that concentrations of primary species in Beijing are controlled by local emissions within 100 km around and preceding 24 hours. On the other hand, an accumulation of secondary aerosols produced in air from their precursor gases under the synoptic-scale meteorological conditions, resulted in movements of high aerosol loading air with a regional scale of more than a thousand kilometer. These regional scale behaviors caused observed temporal variations of secondary aerosol species at Beijing with a timescale of 5 to 7 days, and they are found to be controlled by emissions as far as 500 km within preceding 3 days.

Spatial and temporal variations of aerosol optical parameters are found to be interpreted by the combination of those of primary and secondary aerosol mass concentrations. The spatial and temporal variations of the aerosol extinction and scattering coefficients are found to generally be controlled by secondary aerosols, such as sulfate, that show marked regional enhancements/reductions in association with synoptic-scale meteorological conditions. In contrast, the spatial and temporal variation of the aerosol absorption coefficient is mostly controlled by EC, which is primary aerosol showing a distinct diurnal variation and determined by local-scale emissions. Observed decreases of single scattering albedo following a passage of the cold front are therefore, interpreted by the differences in spatial and temporal variations of primary and secondary aerosol concentrations.

Mass fraction of secondary aerosols increases with altitude within the planetary boundary layer (PBL) owing to secondary aerosol formation at the upper levels, leading to an increase in AOD by 40% around Beijing. Aerosol water mass also increases with altitude owing to higher relative humidity, leading a further AOD increase by 110%. An importance of these two factors on column aerosol radiation effects is clearly demonstrated in this study.