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Atmospheric ammonia over China: emission estimates and impacts on air quality

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Ammonia (NH3) in the atmosphere is an important precursor of inorganic aerosols, and its deposition through wet and dry processes can cause adverse effects on ecosystems. The ammonia emissions over China are particularly large due to intensive agricultural activities, yet our current estimates of Chinese ammonia emissions and associated consequences on air quality are subject to large errors. Here we use the GEOS-Chem chemical transport model and its adjoint model to better quantify this issue. The TES satellite observations of ammonia concentrations and surface measurements of wet deposition fluxes are assimilated into the model to constrain the ammonia emissions over China. Optimized emissions show a strong seasonal variability with emissions in summer a factor of 3 higher than winter. We improve the bottom-up estimate of Chinese ammonia emissions from fertilizer use by using more practical feritilizer application rates for different crop types, which explains most of the discrepancies between our top-down estimates and prior emission estimates. We further use the GEOS-Chem adjoint at 0.25x0.3125 degree resolution to examine the sources contributing to the PM2.5 air pollution over North China. We show that wintertime PM2.5 over Beijing is largely contributed by residential and industrial sources, and ammonia emissions from agriculture activities. PM2.5 concentrations over North China are particularly sensitive to NH3 emissions in cold seasons due to strong nitrate formation. By converting shorted-lived nitric acid to aerosol nitrate, NH3 significantly promotes the regional transport influences of PM2.5 sources.