SO$_2$ Oxidation Efficiency Patterns during an Episode of Plume Transport over Northeast India: Implications to an OH Minimum

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Abstract: Systematic monitoring of the fluctuations in atmospheric SO$_2$ oxidation efficiency measured as a molar ratio of SO$_4^{2-}$ to total SO$_x$ (SO$_x$ = SO$_2$ +SO$_4^{2-}$), referred as S-ratio, have been performed during a major long range plume transport to northeast India (Shillong: 25.67°N, 91.91°E, 1064 m ASL) in March 2009. Anomalously low S-ratios (median, 0.03) were observed during the episode associated with a cyclonic circulation and the SO$_4^{2-}$ and SO$_2$ exhibited unusual features in the relative phase of their peaks. During initial days, when SO$_2$ levels were dictated by the long range influx, the SO$_4^{2-}$ and SO$_2$ variabilities were in anti-phase for the differing mobility/loss mechanisms. When SO$_2$ levels were governed by the boundary layer diurnality in the latter days, the anti-phase is explained by a depleted OH level major portion being consumed in the initial period by the elevated SO$_2$ and other pollutants. Simulations with a global 3D chemical transport model, GEOS-Chem (v8-03-01), also indicated suppressed oxidation conditions with characteristic low S-ratios and poor SO$_2$ - SO$_4^{2-}$ phase agreements. The modelled OH decreased steadily from the initial days, and OH normalized to SO$_2$ referred as OH-specific was consistently low during the suppressed S-ratio period. Further, the geographical distribution of modelled OH showed a pronounced minimum over the region surrounding (20°N, 95°E) spanning parts of northeast India and the adjacent regions to the southeast of it prevalent throughout the year, though the magnitude and the area of influence have a seasonality to it with significant implications for reducing the oxidizing power of the regional atmosphere. A second set of measurements during January 2010 when prominent long range transports were absent exhibited no anomalies, and the S-ratios were well within the acceptable limits (median, 0.32). This work highlights the GEOS-Chem model skill in simulating/detecting the transient fluctuations in the oxidation efficiency, down to a regional scale.

Keywords: Sulphur Dioxide, Sulphate, Atmospheric Oxidation, GEOS-Chem, OH Radical, Plume Transport