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 efficiencyð measured as a molar ratio of SO_4^{2-} to total SO_x ($SO_x = SO_2 + SO_4^{2-}$), referred as S-ratioð have been performed during a major long range plume transport to northeast India (Shillong: 25.67ęN, 91.91ęE, 1064 m ASL) in March 2009. Anomalously low S-ratios (median, 0.03) were observed during the episodeð associated with a cyclonic circulationð and the $SO_4^{2ï}$ and SO_2 exhibited unusual features in the órelative phaseô of their peaks. During initial days, when SO_2 levels were dictated by the long range influx, the $SO_4^{2ï}$ and SO_2 variabilities were in anti-phaseð 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 ódepleted OH levelôð 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 ósuppressed oxidation conditionsôð with characteristic low S-ratios and poor SO_2 ï $SO_4^{2ï}$ phase agreements. The modelled OH decreased steadily from the initial days, and OH normalized to SO_2ð referred as $OH_{specific}$ð was consistently low during the ósuppressed S-ratio periodô. Further, the geographical distribution of modelled OH showed a pronounced minimum over the region surrounding (20ęN, 95ęE) spanning parts of northeast India and the adjacent regions to the southeast of itð prevalent throughout the year, though the magnitude and the area of influence have a seasonality to itð with significant implications for reducing the oxidizing power of the regional atmosphere. A second set of measurements during January 2010ð when prominent long range transports were absentð 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 ótransient fluctuationsô in the oxidation efficiency, down to a regional scale.

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