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SO_2 photo-oxidation on mineral dust: The missing link to explain $\delta^{33}S$ anomalies in urban sulfate aerosols

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Sulfate aerosols exert a net cooling effect on the earth-atmosphere system, yet their radiative forcing remains associated with largest of uncertainties in the assessment of climate change. One of the contributing factors is the poor knowledge of the sulfate formation pathways. The presently known major pathways of atmospheric SO_2 conversion to sulfate are understood to be mostly following the mass-dependent fractionation model (i.e., $\delta^{33}S \sim 0$). However, urban sulfate aerosols collected worldwide exhibit significant non-zero $\delta^{33}S$ compositions, resulting of sulfur mass-independent fractionation (S-MIF) processes. The origin(s) of these S-MIF anomalies remain unclear. Here, we conducted dual-isotope ($\delta^{33}S$, $\delta^{34}S$) probing of sulfate aerosols from summertime megacity Delhi in South Asia. Our results show a shift towards concomitantly high $\delta^{33}S$ and low $\delta^{34}S$ values with proceeding of the summer season. This is attributed to the influx of mineral dust showing also significant correlations with sulfate loadings and $\delta^{33}S$ signatures. We postulate that the SO_2photo-oxidation on mineral dust pathway generates an S-MIF anomaly in sulfate aerosols. As such, this also provides a plausible explanation for the span of positive $\delta^{33}S$ values previously observed in several urban locations globally. Taken together, the findings in this study improve our understanding of S-isotope dynamics in urban regions wherein non-anthropogenic influence (i.e., the role of mineral dust) is found to be an important factor in the formation of aerosol sulfate contributing to air pollution.