Multiple sulfur isotopes of sulfate in Beijing $PM_{2.5}$

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In recent years, air pollution has become a serious problem in China. Mass-dependent (S-MDF) and mass-independent fractionation of sulfur isotopes (S-MIF) of sulfate in $PM_{2.5}$ samples collected during four seasons in Beijing is used to better constraining potential sources and formation processes of sulfate aerosol. The mean $\delta^{34}S$ value (S-MDF) of sulfate in $PM_{2.5}$ is close to that of coals used in North China, suggesting a considerable contribution of sulfate from coal combustion to the atmospheric sulfate pool. The $\Delta^{33}S$ value (S-MIF) of sulfate in $PM_{2.5}$ shows a pronounced seasonality with positive values in spring, summer and autumn (mean=0.227±0.110‰, n=30) and negative values in winter (mean=-0.211±0.188‰, n=18). Sulfur isotope anomalies ($\Delta^{33}S$ up to 0.480 ‰) in spring, summer and autumn are interpreted to result from SO_2 photolysis with self-shielding. The negative $\Delta^{33}S$ signature ($-0.300‰<\Delta^{33}S<0‰$) in winter may be related to incomplete combustion of coal in residential stoves during the heating season. However, negative $\Delta^{33}S$ anomalies ($-0.664‰<\Delta^{33}S<-0.300‰$) in winter and positive $\Delta^{33}S$ anomalies ($0.300‰<\Delta^{33}S<0.480‰$) in spring, summer and autumn suggest sulfur isotopic equilibrium on an annual time frame (mean $\Delta^{33}S$ value=0.075±0.253‰, n=46), which may provide an implication for the absence of S-MIF in sediments of the younger geologic record.

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