

5.034 Spatio-temporal variations of ^{17}O excess of sulfate over Antarctica: reconciling observations and modeling.

Early Career Scientist

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Abstract:

^{17}O excess of sulfate ($\Delta^{17}\text{O}(\text{SO}_4^{2-})$) in aerosol samples reflects relative contribution of different oxidation pathways towards sulfate formation, which can be a diagnostic tool to constrain atmospheric chemistry models. In addition, it has been proposed that the application of $\Delta^{17}\text{O}(\text{SO}_4^{2-})$ to Antarctic ice cores potentially provides past information on oxidant chemistry. Extension of its applicability requires precise understanding of the spatio-temporal variability and the controlling factors of the signature over the present Antarctic atmosphere. In this study, we perform seasonal observations of $\Delta^{17}\text{O}(\text{SO}_4^{2-})$ for aerosol samples collected at both coastal (Dumont d'Urville, 66°40'S, 140°01'E) and inland Antarctica (Concordia, 75°06'S, 123°33'E). At Dumont d'Urville, $\Delta^{17}\text{O}(\text{SO}_4^{2-})$ exhibits a clear seasonal pattern characterized by summer minimum (December) and winter maximum (June), with the range of 1.0 to 3.4‰. This trend is generally caused by sunlight-driven changes in the relative contribution of S(IV) oxidation by O_3 versus other oxidants (e.g., OH radicals and H_2O_2). On the other hand, at Concordia, $\Delta^{17}\text{O}(\text{SO}_4^{2-})$ reaches minimum in late summer (February) and maximum in spring (October). This regional difference implies the existence of S(IV) oxidation processes that results in high $\Delta^{17}\text{O}$

$\delta^{34}\text{S}(\text{SO}_4^{2-})$ values during spring to early summer in the inland site relative to the coastal site. To discuss the possible processes, we evaluate the relative importance of different S(IV) oxidation pathways (i.e. gas phase oxidation by OH radicals, in cloud oxidation by H_2O_2 , O_3 , hypohalous acids ($\text{HOX} = \text{HOCl}$, HOBr), and metal-catalyzed O_2) using the GEOS-Chem 3-D global chemical transport model and simulate the $\Delta^{17}\text{O}(\text{SO}_4^{2-})$ over Antarctica.