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Sulfur and Oxygen Isotopic Constraints on Atmospheric Sulfur Cycles in East Antarctica

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Sulfate (SO_4^{2-}) preserved in Antarctic ice cores is of major interests for understanding the linkage between marine biogenic activity and climate, since atmospheric SO_4^{2-} in Antarctica originates mainly from organic sulfur compounds produced by phytoplankton in the surrounding oceans and then SO_4^{2-} influences climate through the formation of aerosols and clouds scattering solar radiation. It has been shown that there is no significant fluctuation of SO_4^{2-} flux in ice cores through the last eight glacial cycles, which is considered to indicate that there was no significant change in marine biogenic activity in response to climate change. To this context, stable sulfur ($\delta^{34}S$) and triple oxygen isotopic compositions ($\Delta^{17}O$) of SO_4^{2-} are expected to provide different perspectives since they reflect variations in sulfur sources and chemistry in the atmosphere which cannot be unraveled by concentration observation only. This thesis aimed at examination of applicability of these isotopic signatures and development of understanding on sources and formation processes of SO_4^{2-} in the present Antarctic atmosphere, by analyzing seasonal variations of these signatures at inland and coastal sites of East Antarctica.

The analysis of $\delta^{34}S$ of SO_4^{2-} examined its homogeneity between inland and coastal sites throughout the year, which suggests there is no significant isotopic fractionation during transport towards inland. This result ensures that $\delta^{34}S$ can be a strong signature for the source apportionment between marine biogenic and non-marine SO_4^{2-}. This finding, in conjunction with the SO_4^{2-} records in Antarctic ice cores, will open a perspective implying the increased relative importance of non-marine SO_4^{2-} during glacial periods. Furthermore, significant increase in non-marine SO_4^{2-} in early summer was found for the first time, which was hidden in the usual increase of marine biogenic SO_4^{2-} from spring to summer. Identification of sources of this non-marine SO_4^{2-} will be a key step for elucidation of linkage between Antarctic atmosphere and environmental changes at lower latitudes.

The analysis of $\Delta^{17}O$ of SO_4^{2-} suggested that specific chemistry in Antarctica such as snow photochemistry of reactive nitrogen has considerable influence on SO_4^{2-} formation processes in addition to general variations induced by sunlight-driven changes in oxidants (i.e., relative abundance of OH and H_2O_2 relative to O_3). Furthermore, the significant gap of $\Delta^{17}O(SO_4^{2-})$ signatures between atmosphere and snow was found, which indicates the existence of unknown processes at atmosphere-snow interface. These findings will contribute to precise interpretation of $\Delta^{17}O(SO_4^{2-})$ records in Antarctic ice cores and thus the reconstruction of the past atmospheric chemistry.