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Stable sulfur isotope study on the tropospheric cycle of carbonyl sulfide

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Carbonyl sulfide (OCS) can be a main sulfur source of background stratospheric sulfate aerosols (SSAs) and tracer of growth primary production (GPP). However, there are large uncertainty in global OCS budget and it makes the flux from OCS to SSAs and the GPP estimation uncertain. Here I investigated the tropospheric OCS cycle using recently developed method for sulfur isotope ratios ($\delta^{34}\text{S}$ value) of OCS. The determination of isotopic fractionation constants ($^{34}\epsilon$) for reactions and ambient measurements of $\delta^{34}\text{S}$ values were performed to quantify OCS budget.

To determine $^{34}\epsilon$ values, laboratory incubation experiments using OCS-degrading bacterial degradation of OCS were performed. The determined $^{34}\epsilon$ values for OCS degradation by bacteria using Rayleigh plot range from -2‰ to -4‰ . Furthermore, the $^{34}\epsilon$ value for OCS degradation by plant uptake was discussed.

To collect OCS samples from air, a portable sampling system was developed for collecting approx. 10 nmol of OCS from ambient air coupled with a purification system by referring to the halocarbon collection method. Salient system features are accommodation of samples up to 500 L of air at 5 L min^{-1} and the OCS is concentrated in portability of adsorption tubes approx. 0.0014 L volume.

I observed OCS concentration and $\delta^{34}\text{S}$ value from south to north, Japan. The result indicated anthropogenic OCS emission in explaining the spatial variation of atmospheric OCS concentration near the Asian region, suggesting that the results show that human activity greatly contributes to atmospheric OCS based on observation. Additionally, identification of missing sources, estimation of GPP, and OCS contribution to SSAs were estimated.

This thesis shows that the $\delta^{34}\text{S}$ value is a useful tool for understanding the tropospheric OCS cycle and mechanisms. In the future, it will be possible to clarify the dynamics of the troposphere OCS and estimate the OCS missing source by observing the $\delta^{34}\text{S}$ values. In addition, I propose that it is important to constrain the 3-D chemical transport model.