



Determination of triple oxygen isotope composition of tropospheric CO₂

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The isotopic composition of carbon dioxide originating at the surface is modified in the stratosphere. Given that the only source of oxygen anomaly ($\Delta^{17}\text{O} = \delta^{17}\text{O} - 0.516 \times \delta^{18}\text{O} > 0$) is in the stratosphere, such isotopic signal provides a powerful tracer for studying biogeochemical cycles involving carbon dioxide. In addition to the existing fluorination method that can achieve a precision as high as ~ 0.1 per mil, we improved a previously proposed technique, $\text{CeO}_2 + \text{CO}_2$ equilibrium at high temperature, to a precision of ~ 0.13 per mil and better. Such improvement allows us monitor the changes of biogeochemical CO_2 in detail. We successfully measure $\Delta^{17}\text{O}$ in near surface carbon dioxide. The temporal variation of $\Delta^{17}\text{O}$ is apparent. In addition, a 3-D numerical model that includes the isotopic fractionation in the atmosphere has been developed. In this model we assume the biological processes produce neutral $\Delta^{17}\text{O}$, i.e., mass-dependent CO_2 . Processes that affect $\Delta^{17}\text{O}$ in the troposphere are (1) chemistry in the atmosphere (mainly $\text{CO}_2 + \text{O}^{1\text{D}}$ isotope exchange), (2) large circulation that brings the isotopically heavy CO_2 to near the surface, and (3) biological processes that “reset” the isotopic composition. Implications for constraining the sources and sinks of CO_2 using its isotope signature are discussed

References

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