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 ($\Delta^{17}O = \delta^{17}O - 0.516 \times \delta^{18}O > 0$) is in the stratosphere, such isotopic signal provides a powerful tracer for studying biogeochemical cycles involving carbon dioxide. In addition to the exiting fluorination method that can achieve a precision as high as $\sim$0.1 per mil, we improved a previously proposed technique, CeO$_2$ + CO$_2$ equilibrium at high temperature, to a precision of $\sim$0.13 per mil and better. Such improvement allows us monitor the changes of biogeochemical CO$_2$ in detail. We successfully measure $\Delta^{17}O$ in near surface carbon dioxide. The temporal variation of $\Delta^{17}O$ 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 $\Delta^{17}O$, i.e., mass-dependent CO$_2$. Processes that affect $\Delta^{17}O$ in the troposphere are (1) chemistry in the atmosphere (mainly CO$_2$ + O($^{1}$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