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Triple oxygen isotope composition of tropospheric carbon dioxide and its temporal variation

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Conventional stable isotope analysis of carbon dioxide (^{18}O/^{16}O and ^{13}C/^{12}C) is an excellent tool to investigate the atmospheric carbon cycle. In recent years, it has been suggested that investigations on the triple oxygen isotope composition (^{17}O/^{16}O and ^{18}O/^{16}O) might complement traditional stable isotope analysis as this new tracer should reveal information on the terrestrial gross primary production (GPP) [1].

Here, we present high-precision triple oxygen isotope data of ambient air CO_2 sampled in Göttingen (NW Germany) over the course of 2 years. Triple oxygen isotope analysis on CO_2 was carried using a CO_2-CeO_2 equilibration technique [2]. All triple oxygen isotope data are reported as Δ^{17}O values relative to a CO_2-water equilibration line, i.e. Δ^{17}O = ln (δ^{17}O+1) - 0.522×ln (δ^{18}O+1) [3]. Accuracy and precision of Δ^{17}O (single analysis) was better than ±0.05‰ (1σ, SD). We compare our observational data to a revised triple oxygen isotope mass balance model of tropospheric CO_2 where we reconcile both ^{18}O/^{16}O and ^{17}O/^{16}O fractionation processes.

Carbon dioxide sampled in Göttingen has a long-term mean triple oxygen isotope composition with $\delta^{18}O_{VSMOW}$ = 41.6±0.9‰ (SD) and Δ^{17}O = -0.03±0.07‰ (SD). The δ^{18}O values follow the well-known seasonality with an amplitude of about 1.0‰ . The Δ^{17}O signal shows a temporal variation with a peak-to-peak range of about 0.25‰ that parallels the δ^{18}O cycle from summer 2010 to winter 2011, i.e. maximum Δ^{17}O values during summer. During June, July and August 2012, however, the Δ^{17}O values are consistently lower (Δ^{17}O = -0.07±0.05‰ (SD)) than the observational data from summer 2010 and 2011 (Δ^{17}O = +0.02±0.05‰ (SD)).

The revised global mass balance model predicts a Δ^{17}O value for tropospheric CO_2 of +0.06‰ . A Monte Carlo simulation gives an uncertainty estimate intrinsic to our model of ±0.05‰ (SD). The model confirms a sensitivity of Δ^{17}O of tropospheric CO_2 to variations in the terrestrial gross primary production. However, estimates for seasonal variations in regional gross fluxes from the biosphere cannot explain the observed temporal variation in Δ^{17}O of tropospheric CO_2. Variations in anthropogenic CO_2 emissions also cannot explain the Δ^{17}O trend. Thus, it may be that variations in the influx of stratospheric CO_2 cause the observed temporal variation in Δ^{17}O. However, we also suggest that future experimental studies should investigate in detail the Δ^{17}O signature of CO_2 from biological sources to clarify the mechanisms controlling the triple oxygen isotope composition of tropospheric CO_2.

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