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Instrumental and methodological limitations of CO_2 isotopologue eddy covariance measurements

Patrick Sturm (1), Werner Eugster (1), Alexander Knohl (1,2)

(1) Institute of Agricultural Sciences, ETH Zürich, Zürich, Switzerland (pasturm@ethz.ch), (2) Chair of Bioclimatology, Georg-August University Göttingen, Göttingen, Germany

Stable isotope measurements of the biosphere-atmosphere CO_2 exchange can be used to constrain carbon balance estimates. In particular, the terrestrial ecosystem isotope discrimination is an important quantity to determine the magnitude of the terrestrial carbon sink on different spatial and temporal scales. Eddy covariance (EC) measurements of stable isotopes in CO_2 could provide such a direct measure of the ecosystem isotope discrimination and thus indicate the ecosystem fingerprint on the atmosphere's isotopic budget. EC measurements require high-precision and very fast instruments, which became available only recently with new developments in laser absorption spectroscopy.

We present the first EC flux measurements of stable CO_2 isotopologues over a forest canopy. The field experiment was conducted at the Lägeren research site in Switzerland from 22 May to 17 June 2008. We used a quantum cascade laser absorption spectrometer (QCLAS, Aerodyne Research Inc.) for the simultaneous measurement of $^{16}O^{12}C^{16}O$, $^{16}O^{13}C^{16}O$ and $^{18}O^{12}C^{16}O$ isotopologues with a sampling rate of 10 Hz. The 1σ precision estimated from Allan variance plots is about 0.60 ‰ at 0.1 s and about 0.09 ‰ at 30 min averaging time for both $\delta^{13}C$ and $\delta^{18}O$ in CO_2. Hourly calibrations are performed with two calibration gases of known isotopologue concentrations and by dynamically diluting a third calibration gas with CO_2-free air in order to account for the concentration dependence of the isotope ratios.

The isotopic CO_2 fluxes show as expected a strong diurnal cycle and the total CO_2 flux measured with the QCLAS matches well the half-hourly fluxes measured with an LI-7500 open-path infrared gas analyser (QCLAS vs. LI-7500 slope: 0.987, r^2 = 0.98). The ecosystem isotope discrimination calculated from the ratio of the isotopologue fluxes does not show any diurnal variation (in contrast to results from branch bag flux measurements at the same site). However, the noise in the half-hourly EC flux ratios is large and we explore what is limiting the precision of the flux ratios. On the one hand we use data simulations to estimate the error contribution from the instrument. On the other hand different EC data processing procedures are examined to quantify the uncertainty inherently associated with turbulent flux measurements and calculations. We show that the uncertainty from the EC data processing is comparable or larger than the error from instrument precision. Therefore, a direct determination of ecosystem discrimination on hourly or diurnal time scales with the EC technique remains a challenge.