Geophysical Research Abstracts
Vol. 12, EGU2010-9367, 2010
EGU General Assembly 2010

Real-time measurement of δ^{13}C and δ^{18}O in CO_2 by QCLAS - from the soil to the free troposphere

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Isotope ratios of CO_2 are highly valuable to investigate CO_2 sources, sinks and transport at local and global scales. However, relevant studies often require extensive and long term measurements under field conditions, which may not be feasible with standard isotope ratio mass spectrometers (IRMS). We have developed and validated an entirely cryogen-free quantum cascade laser absorption spectrometer (QCLAS) using a room temperature 4.3 μm QC laser. The QCLAS is based on the differential absorption technique and suited for long-term, unattended field applications, delivering both δ^{13}C and δ^{18}O in CO_2 with a precision of 0.05 permil. The spectrometer was validated against IRMS in laboratory and field studies, and has been employed in a range of studies, including forest soil respiration, grassland sub canopy respiration and long-term measurements at a remote location.

In a beech forest of the Swiss Jura, we applied the QCLAS to investigate ^{13}C and ^{18}O dynamics of CO_2 in a closed chamber system placed on soils with a ^{13}C labelled litter layer. A recirculation sampling scheme permitted the real-time monitoring of the CO_2 isotope ratios in the chamber headspace on a timescale of seconds. We performed 90 measurement cycles, resulting each in Keeling plots of 1200 data points. The high time-resolution data revealed very distinct and at first sight surprising patterns for δ^{13}C and δ^{18}O.

High frequency (ca. 5 Hz) measurements were performed during summer at an intensively managed grassland site in central Switzerland with the aim to quantify atmosphere-biosphere carbon exchange for this type of ecosystem. The effects of a grass cut during the measurement period could be detected and resulted in a sub-canopy source conditional flux classification, for which the isotope composition of the CO_2 could be confirmed to be of a respiration source.

Currently, the instrument is operating at the high alpine research station Jungfraujoch (3580 m a.s.l.), and delivers for the first time continuous measurements of CO_2 isotopologues of the free troposphere. Several pollution events during January and February 2009 were identified by the strong patterns for δ^{13}C and δ^{18}O which may also be analyzed using the Keeling intercept method. In combination with Lagrangian backward trajectory models and other trace compounds, these data can be employed for the characterization of CO_2 source regions.

In this presentation we will highlight the characteristics, strength and limitations of our current QC laser spectrometer based on laboratory data and the above case studies.