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Determination of isotopic fractionation δ^{13}C of methane from ground-based FTIR observations performed at the Jungfraujoch

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Atmospheric methane (CH_4) is a strong greenhouse gas that has important chemical impacts on both the troposphere and the stratosphere. In the troposphere, oxidation of methane is a major regulator of OH and is a source of formaldehyde, carbon monoxide and hydrogen. In the stratosphere, CH_4 plays a central role (i), due to its contribution to the stratospheric water vapor budget, and (ii), as a sink for chlorine atoms which reduces the rate of stratospheric ozone depletion.

Because the different sources of methane (natural and anthropogenic like wetlands, rice paddies, termites, natural gas escape, biomass burning, etc) have distinct $^{13}C/^{12}C$ ratios (usually reported in "delta" notation $\delta^{13}C$), measurements of atmospheric $^{13}CH_4$ content, in addition to those of the main isotopologue ($^{12}CH_4$), can be used to investigate individual source strengths as well as their spatial and temporal distributions. Characterization of the isotopic fractionation of methane is therefore important, for example, to help models constrain estimates of the global methane budget. However, experimental data for the $^{13}C/^{12}C$ isotope ratio are sparse. The currently accepted average value of $\delta^{13}C$ in atmospheric methane is about -47‰ (Platt et al., 2004).

The first goal of this work is to develop and to characterize (in terms of information content and error budget) an original retrieval approach to derive $^{13}CH_4$ columns from ground-based Fourier transform infrared (FTIR) spectra recorded at the International Scientific Station of the Jungfraujoch (ISSJ; 46.5°N, 8.0°E, 3580m a.s.l., Swiss Alps). The retrieval strategy is based on a Tikhonov L1 approach which has been originally developed for $^{12}CH_4$ by Sussmann et al. (2008) [see also contributions by Sussmann et al. to this conference (EGU2009-7869)]. In order to validate our $^{13}CH_4$ products, comparisons with satellite ACE-FTS (Atmospheric Chemistry Experiment - Fourier Transform Spectrometer) measurements are performed. Then, atmospheric $\delta^{13}C$ ratios derived from the FTIR measurements will be compared to values published in the literature and critically discussed.

References:

Platt, U., W. Allan and D. Lowe, Hemispheric average Cl atom concentration from $^{13}C/^{12}C$ ratios in atmospheric methane, Atmos. Chem. Phys., 4, 2393-2399, 2004.

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