



Evolution of methanol (CH₃OH) above the Jungfraujoch station (46.5°N): variability, seasonal modulation and long-term trend

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Methanol (CH₃OH) is the second most abundant organic compound in the Earth's atmosphere with concentrations close to a few ppbv, after methane, despite a short lifetime of a few days (Jacob *et al.*, 2005). Natural sources of CH₃OH include plant growth, oceans, decomposition of plant matter, oxidation of methane and other VOCs, ... while anthropogenic sources are from vehicles, industry, ... biomass burning completes the emission budget. The main sink is the oxidation by hydroxyl radical, leading to the formation of carbon monoxide (CO) and formaldehyde (H₂CO).

The retrieval of methanol is very challenging due to the presence of strong absorption of ozone and its isotopologues in addition to water vapour and carbon dioxide in the region of the selected strong nu8 band of CH₃OH. First retrievals from satellite observations using the Atmospheric Chemistry Experiment infrared Fourier Transform Spectrometer (ACE-FTS) on board the SCISAT satellite have been performed by Dufour *et al.* (2007 and references therein) using a spectral interval going from 995.5 to 1008.3 cm⁻¹. In 2009, first retrievals from a ground-based FTS, using spectra recorded at Kitt Peak (31.9°N) and a window going from 992 to 999 cm⁻¹ have been reported by Rinsland *et al.* (2009), followed by Vigouroux *et al.* (2012 and references therein) who used yet another spectral interval going from 1029 to 1037 cm⁻¹.

From those former retrieval strategies and also considering the Mahieu *et al.* (2012) contribution, we redefined our spectral intervals to maximize the information content. Indeed, our first window, starting from 992 to 1008.3 cm⁻¹, is issued from the merge of Rinsland *et al.* and Dufour *et al.* windows while our second, going from 1029 to 1037 cm⁻¹, is the one used by Vigouroux *et al.* With this new combination of windows, we were able to enlarge the range of zenith angles providing robust results while maintaining good correlation between our two windows; this also resulted in an improvement of the fitting residuals and of the information content.

We used the 2008 HITRAN compilation (Rothman *et al.*, 2009) for spectroscopic parameters. However, systematic residuals still remain in the 1033 cm⁻¹ region which are attributed to unsatisfactory line parameters for methanol. New cross sections recorded at the Molecular Spectroscopy Facility of the Rutherford Appleton Laboratory (Harrison *et al.* 2012) and calibrated in intensity by using the reference spectra from the Pacific Northwest National Laboratory (PNNL) IR database will be tested as soon as converted into pseudolines.

In this work, we will present the first long-term time series of methanol total columns, resulting from the implementation of our new retrieval strategy. All retrievals have been performed with the SFIT2 algorithm (v 3.91) (Rinsland *et al.*, 1998) using a series of about 7 000 spectra recorded between 1995 and 2012, with zenith angles between 60 and 85°. These solar absorption observations have been recorded with a high-resolution FTIR Bruker 120HR instrument, at the high altitude station of the Jungfraujoch (46.5°N, 8°E, 3580 m asl), within the framework of the Network for the Detection of Atmospheric Composition Change (NDACC, visit <http://www.ndacc.org>).

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