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First retrievals of methyl chloride from ground-based high-resolution FTIR solar observations

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Methyl chloride (CH_3Cl) is one of the most abundant chlorine-bearing gas in the Earth's troposphere and a significant contributor to the organic chlorine budget. Measurements by in situ networks indicate a mean volume mixing ratio of 550 pptv, with a significant seasonal cycle of about 80 pptv, peak to peak. This species also exhibits inter-annual variability, but no long-term trend. Major sources are from tropical and sub-tropical plants and dead leaves, the oceans and biomass burning. Some industrial processes and waste incineration further add to the emissions. Oxidation by the hydroxyl radical is by far the largest CH_3Cl sink, followed by soil uptake. Although balanced, its atmospheric budget is still affected by large uncertainties and contributions from unidentified sources and sinks cannot be ruled out. Methyl chloride has an atmospheric lifetime of 1 year, a global warming potential of 13 (100-yr horizon) and an ozone depleting potential of 0.02.

The retrieval of methyl chloride from ground-based infrared (IR) spectra is very challenging. Indeed, numerous interferences by strong water vapor and methane lines complicate the detection of small CH_3Cl absorptions, close to 1%, near 3 microns. In addition, and although weak, ethane features contribute to the difficulty, in particular since a significant number of ethane branches were absent until very recently from official HITRAN compilations. Therefore, the scientific literature does not report thus far about any investigations of CH_3Cl from ground-based remote sensing observations.

In this contribution, we will present first CH_3Cl total column retrievals, using the SFIT-2 algorithm (v3.94) and high-resolution Fourier Transform Infrared (FTIR) solar absorption observations recorded with a 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).

In our retrievals, we use new ethane absorption cross sections recorded at the Molecular Spectroscopy Facility of the Rutherford Appleton Laboratory (Harrison et al., 2010). They were calibrated in intensity by using reference low-resolution spectra from the Pacific Northwest National Laboratory (PNNL) IR database. These new cross sections were recently released as a HITRAN update (see http://www.hitran.com). Pseudoline parameters fitted to these ethane spectra have been combined with HITRAN 2004 line parameters (including all the 2006 updates) for all other species encompassed in the selected microwindows, including our target CH_3Cl.

We will evaluate the improvement brought by the new ethane line parameters on the fitting residuals, and characterize the quality, the precision and the reliability of the retrieved product. If successful, a long-term CH_3Cl total column time series will be produced using the Jungfraujoch observational database, and we will perform preliminary investigations of the seasonal and inter-annual variations of methyl chloride total columns at northern mid-latitudes.

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