Decrease of carbon tetrachloride (CCl_4) over 2004-2013 as inferred from global occultation measurement with ACE-FTS

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In this contribution, we use infrared solar occultation measurements performed by the ACE-FTS (Atmospheric Chemistry Experiment – Fourier Transform Spectrometer) instrument onboard the SCISAT-1 Canadian satellite (Bernath et al., 2005). Since its launch in August 2003, this spectrometer has been in continuous operation with no significant degradation of its performance, and global measurements are available from late February 2004 onwards, spanning now more than a decade.

ACE-FTS achieves a spectral resolution of 0.02 cm^{-1} and covers the 750-4400 cm^{-1} range, encompassing the strong unresolved and broad CCl_4 ν_3 band around 796 cm^{-1}, near a strong CO_2 Q-branch affected by line-mixing (Rinsland et al., 2012). Systematic analysis of sunset and sunrise occultation measurements in the 787.5 – 805.5 cm^{-1} window (version 3.5; Boone et al., 2013) provides mixing ratio profiles of CCl_4 in the 7 – 25 km altitude range, with mean vertical resolution of 2-3 km.

More than 24000 occultations have been included in the present study, covering the 85°N-85°S latitude range and updating the work of Allen et al. (2009). We determine a significant positive bias with respect to surface measurements by the AGAGE and NOAA networks, confirming the findings of Rinsland et al. (2012) when using ground-based column measurements at the Jungfraujoch station. However, when accounting for the systematic uncertainty affecting the CCl_4 line parameters and the impact of the CO_2 line-mixing, we show that it is possible to close the gap between the surface and remote-sensing measurements.

Focusing on the tropical observations near 9 and 17 km altitude, we characterize a significant yearly decrease for CCl_4 (at the 2-sigma level) of -1.3 ppt (or -1.35%) over the last decade, in agreement with results from Jungfraujoch (updated from Rinsland et al., 2012) and the *in situ* networks (WMO 2014). Finally, we analyze ACE-FTS global data in order to check for possible contrasted evolutions of CCl_4 in both hemispheres.

References

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