

Self broadening coefficients and improved line intensities for the ν_7 band of C_2H_4 near $10.5 \mu m$, and impact on ethylene retrievals from Jungfraujoch solar spectra

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Ethylene (ethene, C_2H_4) is a tropospheric pollutant on the Earth, also present as a by-product of methane photochemistry in the atmosphere of outer solar system bodies. Remote sensing of ethylene in the infrared range relies on the $10 \mu m$ region. This spectral range corresponds to the excitation of 7 modes of vibration of $^{12}C_2H_4$, 4 of which being infrared active (see Fig. 1 of [1]). The corresponding ν_{10} , ν_7 , ν_4 and ν_{12} bands are located near 826 , 949 , 1026 and 1442 cm^{-1} , respectively [1]. Among these, the ν_7 band is the strongest, indeed used for remote sensing measurements of ethylene.

Relying on high-resolution Fourier transform infrared (FTIR) spectra recorded in Brussels, the present work involved extensive measurements of individual line intensities and self broadening coefficients for the ν_7 band of $^{12}C_2H_4$. Compared to the corresponding information available in the latest edition of the HITRAN spectroscopic database (HITRAN 2012 [2]), the measured line intensities were found to be higher by about 10 % for high J lines in the P branch and lower by about 5 % for high J lines of the R branch, varying between these two limits roughly linearly with the line positions. Test calculations performed in this work indicated that these discrepancies could result from the relative values of the transition moments of the ν_{10} , ν_7 and ν_{12} bands used when the information provided in HITRAN was generated (the transition moment of the ν_4 band was set to zero). The measured self broadening coefficients exhibit a dependence on both J and K_a , which was modeled empirically. The spectroscopic information for ethylene available in HITRAN 2012 was modified to match the present observations. The impact of these modifications on retrievals of atmospheric ethylene was then evaluated via FTIR retrievals in the $949.0 - 952.0 \text{ cm}^{-1}$ microwindow, from a subset of ground-based high-resolution FTIR solar spectra recorded at the Jungfraujoch station. The new line intensities were found to lead to a reduction of the measured total columns of ethylene by $-4.1 \pm 0.1 \%$, compared to the use of HITRAN 2012.

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Références

- [1] F. Willaert, J. Demaison, L. Margulès, H. Mäder, H. Spahn, T. Giesen, A. Fayt, “The spectrum of ethylene from microwave to submillimeter-wave”, *Mol. Phys.* **104**, pp. 273-292 (2006).
- [2] L.S. Rothman *et al.*, “The HITRAN 2012 molecular spectroscopic database,” *J. Quant. Spectrosc. Radiat. Transfer* **130**, pp. 4-50 (2013).