Geophysical Research Abstracts
Vol. 20, EGU2018-8001-1, 2018
EGU General Assembly 2018

Attempting to retrieve peroxyacetyl nitrate from ground-based infrared solar spectra

Emmanuel Mahieu (1), Emily V. Fischer (2), Zitely A. Tzompa-Sosa (2), Bruno Franco (3), Maxime Prignon (1), Christian Servais (1), James W. Hannigan (4), Ivan Ortega (4), Mathias Palm (5), Justus Notholt (5), and Geoffrey C. Toon (6)

(1) University of Liège, Institute of Astrophysics and Geophysics, LIEGE, Belgium (emmanuel.mahieu@uliege.be), (2) Department of Atmospheric Science, Colorado State University, Fort Collins, CO, USA, (3) Quantum Chemistry and Photophysics, Université Libre de Bruxelles, Belgium, (4) National Center for Atmospheric Research, Boulder, Colorado, USA, (5) Institute of Environmental Physics, University of Bremen, Bremen, Germany, (6) Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA, USA

Given its significant lifetime in the upper troposphere, $CH_3C(O)O_2NO_2$ (known as peroxyacetyl nitrate, PAN) acts as a reservoir for the nitrogen oxide radicals (NO_x = NO + NO_2), allowing the long-range transport of pollution, far from the regions of production (Fischer et al., 2014). Its thermal decomposition in remote areas leads to the efficient formation and redistribution of tropospheric ozone, justifying its global monitoring.

This compound is the subject of many investigations, and efforts are ongoing in the satellite community in order to produce reliable remote-sensing products. This is the case, e.g., for ACE-FTS (Atmospheric Chemistry Experiment-Fourier Transform Spectrometer; Tereszchuk et al., 2013), MIPAS (Michelson Interferometer for Passive Atmospheric Sounding; Pope et al., 2016) and TES (Tropospheric Emission Spectrometer; Zhu et al., 2015). In situ surface and airborne measurements are available for the validation of such global satellite measurements, but to our knowledge, PAN has not been retrieved yet from ground-based Fourier Transform InfraRed (FTIR) solar spectra.

There are two candidate spectral features for the retrieval of PAN from ground-based FTIR spectra, one centered on 1162 cm^{-1}, and a second one near 795 cm^{-1}. For both, the broad unstructured absorptions of PAN require to select wide spectral window, of several tens of wavenumbers, for its retrieval. As a result, numerous strong interfering lines have to be accounted for, by H_2O, CO_2, O_3, N_2O. The inversions are further complicated by other interfering broad features associated to halogenated source gases (e.g., CCl_4, HFC-23, HCFC-22, CFC-113). Last but not least, strong line-mixing affects a CO_2 Q-branch around 791 cm^{-1}, leading to systematic and significant residuals (Rinsland et al., 2012).

In this contribution, we present first attempts to retrieve PAN, using FTIR spectra recorded at the Jungfraujoch station (Swiss Alps, 3580 m a.s.l.). The implementation of the line-mixing representation in the SFIT-4 algorithm (v0.9.4.4) allows fitting, and comparing, both windows. For the heavy species, pseudo-linelists were used. These line-by-line parameters were determined from the fit to cross-section laboratory spectra typically recorded at several atmospheric pressures and temperatures (http://mark4sun.jpl.nasa.gov/pseudo.html). Finally, our investigations are supported by a full-chemistry simulation by the 3D Chemistry Transport GEOS-Chem Model, performed with a horizontal resolution of 2° x 2.5°.

Acknowledgments

This research received support from the F.R.S.–FNRS (Brussels), under Grants J.0093.15 and J.0147.18.

References

Fischer, E. V., et al.: Atmospheric peroxyacetyl nitrate (PAN): a global budget and source attribution, ACP, 14, 2679–2698, doi:10.5194/acp-14-2679-2014, 2014.

Pope, R. J. et al.: Intercomparison and evaluation of satellite peroxyacetyl nitrate observations in the upper troposphere–lower stratosphere, ACP, 16, doi:10.5194/acp-16-13541-2016, 2016.

Rinsland, C. P. et al.: Decrease of the carbon tetrachloride (CCl_4) loading above Jungfraujoch, based on high resolution infrared solar spectra recorded between 1999 and 2011, JQSRT, 113, doi:10.1016/j.jqsrt.2012.02.016, 2012.

Tereszchuk, K. A., et al.: Observations of peroxyacetyl nitrate (PAN) in the upper troposphere by the Atmospheric Chemistry Experiment-Fourier Transform Spectrometer (ACE-FTS), ACP, 13, doi:10.5194/acp-13-5601-2013, 2013.

Zhu, L., et al.: TES observations of the interannual variability of PAN over Northern Eurasia and the relationship to springtime fires, GRL, 42, doi:10.1002/2015GL065328, 2015.