

# Detection of elevated tropospheric hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) mixing ratios in atmospheric chemistry experiment (ACE) subtropical infrared solar occultation spectra

C.P. Rinsland<sup>a,\*</sup>, P.F. Coheur<sup>b,1</sup>, H. Herbin<sup>b</sup>, C. Clerbaux<sup>c</sup>, C. Boone<sup>d</sup>,  
P. Bernath<sup>d,e</sup>, L.S. Chiou<sup>f</sup>

<sup>a</sup>NASA Langley Research Center, Science Directorate, Mail Stop 401A, Hampton, VA 23681 2199, USA

<sup>b</sup>Spectroscopie de l'Atmosphère, Chimie Quantique et Photophysique CP 160/09, Université Libre de Bruxelles, 50 Avenue F.D. Roosevelt, B 1050 Brussels, Belgium

<sup>c</sup>Service d'Aéronomie/Institut Pierre-Simon Laplace, Université Pierre et Marie Curie-Paris 6, France

<sup>d</sup>Department of Chemistry, University of Waterloo, Waterloo, Ont., Canada N2L 3G1

<sup>e</sup>Department of Chemistry, University of York, Heslington, York YO10 5DD, UK

<sup>f</sup>Science Systems and Applications, Inc., Hampton, VA 23666 USA

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## Abstract

We report measurements of hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) profiles from infrared solar occultation spectra recorded at  $0.02\text{ cm}^{-1}$  resolution by the atmospheric chemistry experiment (ACE) during 2004 and 2005. Mixing ratios as high as 1.7 ppbv (1 ppbv =  $1 \times 10^{-9}$  per unit volume) were measured in the subtropical troposphere. Back trajectories, fire count statistics, and simultaneous measurements of other species from the same occultation provide evidence that the elevated  $\text{H}_2\text{O}_2$  mixing ratios originated from a young biomass-burning plume. The ACE time series show only a few cases with elevated  $\text{H}_2\text{O}_2$  mixing ratios likely because of the short lifetime of  $\text{H}_2\text{O}_2$  and the limited sampling during biomass-burning time periods.

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## 1. Introduction

A variety of techniques have been used to measure hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) [1], an atmospheric oxidant almost solely formed from the self-reaction of  $\text{HO}_2$  radicals and thus can serve as sensitive indicator for  $\text{HO}_x$  ( $\text{OH} + \text{HO}_2$ ) chemistry [2]. The  $\text{H}_2\text{O}_2$  molecule is a  $\text{HO}_x$  reservoir with a relatively short lifetime in the

\*Corresponding author. Tel.: +1 7578642699; fax: +1 7578648197.

E-mail addresses: c.p.rinsland@larc.nasa.gov (C.P. Rinsland), pfcheur@ulb.ac.be (P.F. Coheur), hherbin@ulb.ac.be (H. Herbin), cathy.clerbaux@aero.jussieu.fr (C. Clerbaux), cboone@acebox.uwaterloo.ca (C. Boone), bernath@uwaterloo.ca, pfb500@york.ac.uk (P. Bernath), l.s.chiou@larc.nasa.gov (L.S. Chiou).

<sup>1</sup>P.F. Coheur is Research Associate with the F.N.R.S.

troposphere ( $\sim 1$  d) [2]. Emissions from surface sources can be transported to the upper troposphere by deep tropical convection [3]. Complex interactions in the troposphere affect the production of ozone ( $O_3$ ) and atmospheric  $HO_x$  and  $NO_x$  ( $NO + NO_2$ ) levels, though non-methane hydrocarbons cannot photolyze and participate in  $HO_x$  formation [2]. Volatile organic compounds (VOCs) include non-methane hydrocarbons (NMHC) and oxygenated NMHCs, are generally short-lived, although species such as ethane ( $C_2H_6$ ) and acetone ( $C_3H_6O$ ) are longer-lived and impact tropospheric chemistry on hemispheric scales [4].

Methods for measuring  $H_2O_2$  include *in situ* sampling from an aircraft during the transport and atmospheric chemistry near the Equator-Atlantic (TRACE-A) experiment [5], far infrared stratospheric emission measurements from a balloon-borne platform ( $34^\circ N$ ,  $104^\circ W$ ) in September 1989 [6], and space-based limb emission measurements with a high spectral resolution Fourier transform spectrometer [7]. However, weak absorption by  $H_2O_2$  in the mid-infrared combined with its short lifetime and the complex region where it absorbs make it a challenging molecule to detect and quantify with remote sensing techniques.

Atmospheric  $H_2O_2$  is formed primarily by self-reaction



although production can be suppressed at high  $NO_x$  in the troposphere where the peroxy radical ( $HO_2$ ) reacts with  $NO$ , i.e.



Loss of  $H_2O_2$  is primarily through



In the troposphere gas-phase reactions are augmented by the loss of dissolved  $H_2O_2$  mainly through reaction with dissolved  $SO_2$  and rainout [2,7–9]. Photolytic loss is predicted to result in diurnal and seasonal variations in  $H_2O_2$  [7].

$H_2O_2$  is an important sink for  $OH$  and an important oxidant of sulfur compounds in the aqueous phase and can be deposited both wet and dry as a result of its high solubility [2]. However, photochemical pathways may lead to substantial recycling to reform  $H_2O_2$  [2]. Despite the importance, to our knowledge only zonal mean space-based profiles of  $H_2O_2$  have been reported for the stratosphere and troposphere [7]. Mean measured profiles from that work did not show elevated tropospheric  $H_2O_2$ , but a three-dimensional model run for the same time period (September 2003) predicted an upper tropospheric peak of  $>0.33$  ppbv ( $10^{-9}$  per unit volume) in the tropics [7].

The purpose of this investigation is to present  $H_2O_2$  tropospheric retrievals obtained during 2004 and 2005 by the atmospheric chemistry experiment (ACE). We report several cases with elevated upper tropospheric  $H_2O_2$  mixing ratios. We show back trajectories and fire count statistics for the measurement with the highest  $H_2O_2$  mixing ratios. Those results support the hypothesis that the emissions originated from deep convective transport from a tropical biomass fire source regions to the upper troposphere [3]. Tropical fires are a well-documented source of elevated trace gases emissions and notably  $H_2O_2$ , as confirmed from measurements inside biomass-burning plume during aircraft flights near the equator as well as from laboratory biomass-burning emission experiments [5].

## 2. Measurements

The ACE payload, also known as SCISAT-1, was successfully launched on 12 August 2003 into a  $74^\circ$  inclined orbit by a NASA-supplied Pegasus XL at 650 km altitude [10]. Routine ACE FTS science measurements began in February 2004. This small Canadian-designed and built satellite contains three instruments with a shared field of view, and has the primary goal of recording high-resolution atmospheric spectra taking advantage of the high precision of the solar occultation technique. The infrared instrument is a Fourier transform spectrometer (FTS) that records solar spectra below altitudes of 150 km at a spectral resolution of  $0.02 \text{ cm}^{-1}$  (maximum optical path difference of  $\pm 25 \text{ cm}$ ) with a  $1.25 \text{ mrad}$  ( $10^{-3} \text{ rad}$ ) diameter

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