Recent increase of ethane detected in the remote atmosphere of the Northern Hemisphere

B. Franco¹, W. Bader¹, B. Bovy¹, E. Mahieu¹, E. V. Fischer², K. Strong³, S. Conway³, J. W. Hannigan⁴, E. Nussbaumer⁴, P. F. Bernath⁵,⁶,⁷, C. D. Boone⁷ & K. A. Walker³,⁷
¹University of Liège (bruno.franco@ulg.ac.be), ²Colorado State University, ³University of Toronto, ⁴NCAR Boulder, ⁵Old Dominion University, ⁶University of York, ⁷University of Waterloo

Abstract (1/3)

Ethane (C₂H₆) has a large impact on tropospheric composition and air quality because of its involvement in the global chemistry responsible for generating and destroying tropospheric ozone. The abundance of C₂H₆ influences the atmospheric content of carbon monoxide and impacts the lifetime of methane. It is an important source of PAN, a thermally unstable reservoir for NOₓ radicals.

On a global scale, the main sources of C₂H₆ are leakage from the production, transport of natural gas loss, biofuel consumption and biomass burning, mainly located in the Northern Hemisphere. Due to its relatively long lifetime of approximately two months, C₂H₆ is a sensitive indicator of tropospheric pollution and transport.

C₂H₆ from FTIR at Jungfraujoch: Recent burden increase

C₂H₆ in the Southern Hemisphere: No recent upturn

Cause of the C₂H₆ renewal: Shale gas exploitation?

Comparison with GEOS-Chem: Underestimation in the inventories
Using an optimized retrieval strategy (Franco et al., 2015, *JQSRT*), we present here a 20-year long-term time series of C$_2$H$_6$ column abundance retrieved from ground-based Fourier Transform InfraRed (FTIR) solar spectra recorded from 1994 onwards at the high-altitude station of Jungfraujoch (Swiss Alps, 3580m), part of the Network for the Detection of Atmospheric Composition Change (NDACC, see [http://www.ndacc.org](http://www.ndacc.org)).

After a regular 1994-2008 decrease of the C$_2$H$_6$ amounts, which is very consistent with prior major studies (e.g., Aydin et al., 2011, *Nature*; Simpson et al., 2012, *Nature*) and our understanding of global C$_2$H$_6$ emissions, trend analysis using a bootstrap resampling tool reveals a C$_2$H$_6$ upturn and a statistically-significant sharp burden increase from 2009 onwards.
Recent increase of ethane detected in the remote atmosphere of the Northern Hemisphere

B. Franco¹, W. Bader¹, B. Bovy¹, E. Mahieu¹, E. V. Fischer², K. Strong³, S. Conway³, J. W. Hannigan⁴, E. Nussbaumer⁴, P. F. Bernath⁵,⁶,⁷, C. D. Boone⁷ & K. A. Walker³,⁷

¹University of Liège (bruno.franco@ulg.ac.be), ²Colorado State University, ³University of Toronto, ⁴NCAR Boulder, ⁵Old Dominion University, ⁶University of York, ⁷University of Waterloo

Abstract (3/3)

We hypothesize that the \( \text{C}_2\text{H}_6 \) increase could affect the whole Northern Hemisphere and may be related to the recent massive growth in the exploitation of shale gas and tight oil reservoirs. This hypothesis is supported by measurements derived from solar occultation observations performed from space by the ACE-FTS instrument and at other NDACC sites. The recent rates of changes characterizing these data sets are consistent in magnitude and sign with the one derived at Jungfraujoch. In contrast, the atmospheric \( \text{C}_2\text{H}_6 \) burden in the Southern Hemisphere shows a monotonic decrease over the last two decades.

An ongoing work focuses on combining an analysis of \( \text{C}_2\text{H}_6 \) measurements from ground-based FTIR and ACE-FTS observations with dedicated GEOS-Chem simulations with updated inventories to identify the cause of the \( \text{C}_2\text{H}_6 \) renewal.
Recent increase of ethane detected in the remote atmosphere of the Northern Hemisphere

B. Franco¹, W. Bader¹, B. Bovy¹, E. Mahieu¹, E. V. Fischer², K. Strong³, S. Conway³, J. W. Hannigan⁴, E. Nussbaumer⁴, P. F. Bernath⁵,⁶,⁷, C. D. Boone⁷ & K. A. Walker³,⁷

¹University of Liège (bruno.franco@ulg.ac.be), ²Colorado State University, ³University of Toronto, ⁴NCAR Boulder, ⁵Old Dominion University, ⁶University of York, ⁷University of Waterloo

Regular decrease of atmospheric C₂H₆ burden consistent with the global decline of fugitive emissions from fossil fuel sources from the mid-1980s (Aydin et al., 2011, Nature; Simpson et al., 2012, Nature)

See Franco et al., 2015, JQSRT for more information on the FTIR retrievals
Recent increase of ethane detected in the remote atmosphere of the Northern Hemisphere

But since 2009, the atmospheric $\text{C}_2\text{H}_6$ burden is increasing at a rate of $5\%\text{ yr}^{-1}$
Recent increase of ethane detected in the remote atmosphere of the Northern Hemisphere

B. Franco¹, W. Bader¹, B. Bovy¹, E. Mahieu¹, E. V. Fischer², K. Strong³, S. Conway³, J. W. Hannigan⁴, E. Nussbaumer⁴, P. F. Bernath⁵,⁶,⁷, C. D. Boone⁷ & K. A. Walker³,⁷
¹University of Liège (bruno.franco@ulg.ac.be), ²Colorado State University, ³University of Toronto, ⁴NCAR Boulder, ⁵Old Dominion University, ⁶University of York, ⁷University of Waterloo

• Two independent partial columns may be deduced from the FTIR retrievals (DOFS ≈ 2.1)
• Vertically-homogeneous increase of C₂H₆ throughout the troposphere and lower stratosphere

Rates of change relative to 2009.0
- Total column
  4.85 ± 0.85 % yr⁻¹
- Lower troposphere (3.6-8 km)
  4.23 ± 1.06 % yr⁻¹
- Upper trop. – lower strat. (8-21 km)
  6.05 ± 1.09 % yr⁻¹
Recent increase of ethane detected in the remote atmosphere of the Northern Hemisphere

B. Franco¹, W. Bader¹, B. Bovy¹, E. Mahieu¹, E. V. Fischer², K. Strong³, S. Conway³, J. W. Hannigan⁴, E. Nussbaumer⁴, P. F. Bernath⁵,⁶,⁷, C. D. Boone⁷ & K. A. Walker³,⁷

¹University of Liège (bruno.franco@ulg.ac.be), ²Colorado State University, ³University of Toronto, ⁴NCAR Boulder, ⁵Old Dominion University, ⁶University of York, ⁷University of Waterloo

Cause of the C₂H₆ renewal since 2009?

- Sharp fluctuations of OH concentration in the atmosphere?
  - The global OH levels have not exhibited large interannual variability since the end of the 20th century (Montzka et al., 2011, *Science*)
  - Neither CO nor other species that have oxidation by OH as their major removal pathway such as HCN and C₂H₂, do not present an upturn in their retrieved columns

=> hence this hypothesis should be rejected
Recent increase of ethane detected in the remote atmosphere of the Northern Hemisphere

B. Franco¹, W. Bader¹, B. Bovy¹, E. Mahieu¹, E. V. Fischer², K. Strong³, S. Conway³, J. W. Hannigan⁴, E. Nussbaumer⁴, P. F. Bernath⁵,⁶,⁷, C. D. Boone⁷ & K. A. Walker³,⁷

¹University of Liège (bruno.franco@ulg.ac.be), ²Colorado State University, ³University of Toronto, ⁴NCAR Boulder, ⁵Old Dominion University, ⁶University of York, ⁷University of Waterloo

Cause of the C₂H₆ renewal since 2009?

- **Product of enhanced fugitive emissions?**
  - May be linked to the recent massive growth in the exploitation of shale gas and tight oil reservoirs, especially in North America
  - Increases of hydrocarbons related to oil and gas industries are detected over North American regions where the drilling productivity began to grow rapidly after 2009
  - Could represent a change in C₂H₆ throughout the Northern Hemisphere
  - FTIR and ACE-FTS measurements over North America support this hypothesis
Recent increase of ethane detected in the remote atmosphere of the Northern Hemisphere

B. Franco\textsuperscript{1}, W. Bader\textsuperscript{1}, B. Bovy\textsuperscript{1}, E. Mahieu\textsuperscript{1}, E. V. Fischer\textsuperscript{2}, K. Strong\textsuperscript{3}, S. Conway\textsuperscript{3}, J. W. Hannigan\textsuperscript{4}, E. Nussbaumer\textsuperscript{4}, P. F. Bernath\textsuperscript{5,6,7}, C. D. Boone\textsuperscript{7} & K. A. Walker\textsuperscript{3,7}

\textsuperscript{1}University of Liège (bruno.franco@ulg.ac.be), \textsuperscript{2}Colorado State University, \textsuperscript{3}University of Toronto, \textsuperscript{4}NCAR Boulder, \textsuperscript{5}Old Dominion University, \textsuperscript{6}University of York, \textsuperscript{7}University of Waterloo

FTIR observations of $\text{C}_2\text{H}_6$ over North America

Note: the same a priori and variability profiles as Jungfraujoch are used for all ground-based FTIR retrievals.
Recent increase of ethane detected in the remote atmosphere of the Northern Hemisphere

B. Franco\textsuperscript{1}, W. Bader\textsuperscript{1}, B. Bovy\textsuperscript{1}, E. Mahieu\textsuperscript{1}, E. V. Fischer\textsuperscript{2}, K. Strong\textsuperscript{3}, S. Conway\textsuperscript{3}, J. W. Hannigan\textsuperscript{4}, E. Nussbaumer\textsuperscript{4}, P. F. Bernath\textsuperscript{5,6,7}, C. D. Boone\textsuperscript{7} & K. A. Walker\textsuperscript{3,7}

\textsuperscript{1}University of Liège (bruno.franco@ulg.ac.be), \textsuperscript{2}Colorado State University, \textsuperscript{3}University of Toronto, \textsuperscript{4}NCAR Boulder, \textsuperscript{5}Old Dominion University, \textsuperscript{6}University of York, \textsuperscript{7}University of Waterloo

FTIR observations of $\text{C}_2\text{H}_6$ over North America

Note: the same a priori and variability profiles as Jungfraujoch are used for all ground-based FTIR retrievals
Recent increase of ethane detected in the remote atmosphere of the Northern Hemisphere

B. Franco¹, W. Bader¹, B. Bovy¹, E. Mahieu¹, E. V. Fischer², K. Strong³, S. Conway³, J. W. Hannigan⁴, E. Nussbaumer⁴, P. F. Bernath⁵,⁶,⁷, C. D. Boone⁷ & K. A. Walker³,⁷

¹University of Liège (bruno.franco@ulg.ac.be), ²Colorado State University, ³University of Toronto, ⁴NCAR Boulder, ⁵Old Dominion University, ⁶University of York, ⁷University of Waterloo

FTIR observations of C₂H₆ over North America

C₂H₆ at Eureka (Canada, 610 m, 80.1° N, 86.4° W)

- FTIR daily mean with 1σ StDev
- Trend component + seasonal modulation

Rate of change relative to 2009.0
3.49 ± 0.67 % yr⁻¹

Note: the same a priori and variability profiles as Jungfraujoch are used for all ground-based FTIR retrievals
Recent increase of ethane detected in the remote atmosphere of the Northern Hemisphere

B. Franco$^1$, W. Bader$^1$, B. Bovy$^1$, E. Mahieu$^1$, E. V. Fischer$^2$, K. Strong$^3$, S. Conway$^3$, J. W. Hannigan$^4$, E. Nussbaumer$^4$, P. F. Bernath$^{5,6,7}$, C. D. Boone$^7$ & K. A. Walker$^{3,7}$

$^1$University of Liège (bruno.franco@ulg.ac.be), $^2$Colorado State University, $^3$University of Toronto, $^4$NCAR Boulder, $^5$Old Dominion University, $^6$University of York, $^7$University of Waterloo

Note: the same a priori and variability profiles as Jungfraujoch are used for all ground-based FTIR retrievals.

FTIR observations of C$_2$H$_6$ over North America

Rate of change relative to 2010.0
3.38 ± 3.49 % yr$^{-1}$
Recent increase of ethane detected in the remote atmosphere of the Northern Hemisphere

B. Franco¹, W. Bader¹, B. Bovy¹, E. Mahieu¹, E. V. Fischer², K. Strong³, S. Conway³, J. W. Hannigan⁴, E. Nussbaumer⁴, P. F. Bernath⁵,⁶,⁷, C. D. Boone⁷ & K. A. Walker³,⁷

¹University of Liège (bruno.franco@ulg.ac.be), ²Colorado State University, ³University of Toronto, ⁴NCAR Boulder, ⁵Old Dominion University, ⁶University of York, ⁷University of Waterloo

Satellite measurements of C₂H₆ over North America

C₂H₆ from ACE-FTS (15 - 88° N, 50 - 175° W)

Rate of change relative 2004.0
-1.75 ± 1.30 % yr⁻¹

Rate of change relative to 2009.0
9.40 ± 3.21 % yr⁻¹

EGU General Assembly 2015, April 13th, Vienna, Austria
Recent increase of ethane detected in the remote atmosphere of the Northern Hemisphere

B. Franco\textsuperscript{1}, W. Bader\textsuperscript{1}, B. Bovy\textsuperscript{1}, E. Mahieu\textsuperscript{1}, E. V. Fischer\textsuperscript{2}, K. Strong\textsuperscript{3}, S. Conway\textsuperscript{3}, J. W. Hannigan\textsuperscript{4}, E. Nussbaumer\textsuperscript{4}, P. F. Bernath\textsuperscript{5,6,7}, C. D. Boone\textsuperscript{7} & K. A. Walker\textsuperscript{3,7}

\textsuperscript{1}University of Liège (bruno.franco@ulg.ac.be), \textsuperscript{2}Colorado State University, \textsuperscript{3}University of Toronto, \textsuperscript{4}NCAR Boulder, \textsuperscript{5}Old Dominion University, \textsuperscript{6}University of York, \textsuperscript{7}University of Waterloo

- FTIR and ACE-FTS observations in the Southern Hemisphere do not reveal any recent increase
- Latitudinal exchange of C\textsubscript{2}H\textsubscript{6} between both hemispheres is weak
- The observed renewal of C\textsubscript{2}H\textsubscript{6} seems to be currently \textbf{limited to the Northern Hemisphere}

\begin{figure}
\centering
\includegraphics[width=\textwidth]{ethane_plot.png}
\caption{C\textsubscript{2}H\textsubscript{6} at Lauder (New Zealand, 370 m, 45.0° S, 169.7° E)}
\end{figure}

- Rate of change relative to 1993.0
  \(-1.49 \pm 0.17 \text{ % yr}^{-1}\)
- Rate of change relative to 2009.0
  \(0.34 \pm 0.87 \text{ % yr}^{-1}\)
Recent increase of ethane detected in the remote atmosphere of the Northern Hemisphere

B. Franco¹, W. Bader¹, B. Bovy¹, E. Mahieu¹, E. V. Fischer², K. Strong³, S. Conway³, J. W. Hannigan⁴, E. Nussbaumer⁴, P. F. Bernath⁵,⁶,⁷, C. D. Boone⁷ & K. A. Walker³,⁷

¹University of Liège (bruno.franco@ulg.ac.be), ²Colorado State University, ³University of Toronto, ⁴NCAR Boulder, ⁵Old Dominion University, ⁶University of York, ⁷University of Waterloo

- FTIR and ACE-FTS observations in the Southern Hemisphere do not reveal any recent increase
- Latitudinal exchange of C₄H₆ between both hemispheres is weak
- The observed renewal of C₄H₆ seems to be currently limited to the Northern Hemisphere

C₄H₆ from ACE-FTS (10.0 - 40.0° S)

Rate of change relative to 2004.0
-1.62 ± 1.08 % yr⁻¹
Recent increase of ethane detected in the remote atmosphere of the Northern Hemisphere

B. Franco¹, W. Bader¹, B. Bovy¹, E. Mahieu¹, E. V. Fischer², K. Strong³, S. Conway³, J. W. Hannigan⁴, E. Nussbaumer⁴, P. F. Bernath⁵,⁶,⁷, C. D. Boone⁷ & K. A. Walker³,⁷

¹University of Liège (bruno.franco@ulg.ac.be), ²Colorado State University, ³University of Toronto, ⁴NCAR Boulder, ⁵Old Dominion University, ⁶University of York, ⁷University of Waterloo

C₂H₆ derived from FTIR retrievals and GEOS-Chem simulations

- GEOS-Chem v9-02 standard full-chemistry simulation
- 2005-2013 results displayed on a 1-year time base
- The vertical resolution and specific sensitivity of the FTIR retrievals are accounted for

Underestimation of C₂H₆ emissions in the current inventories implemented by GEOS-Chem

Incorporating updated inventories to reproduce the observed changes is part of an ongoing work
Recent increase of ethane detected in the remote atmosphere of the Northern Hemisphere

B. Franco¹, W. Bader¹, B. Bovy¹, E. Mahieu¹, E. V. Fischer², K. Strong³, S. Conway³, J. W. Hannigan⁴, E. Nussbaumer⁴, P. F. Bernath⁵,⁶,⁷, C. D. Boone⁷ & K. A. Walker³,⁷

¹University of Liège (bruno.franco@ulg.ac.be), ²Colorado State University, ³University of Toronto, ⁴NCAR Boulder, ⁵Old Dominion University, ⁶University of York, ⁷University of Waterloo

Acknowledgments:

The University of Liège contribution to the present work has mainly been supported by the AGACC-II project of the Science for Sustainable Development (SSD) program of the Belgian Science Policy Office (BELSPO, Brussels). Additional support was provided by MeteoSwiss (Global Atmospheric Watch), the Fédération Wallonie–Bruxelles and the F.R.S. – FNRS. We thank the International Foundation High Altitude Research Stations Jungfraujoch and Gornergrat (HFSJG, Bern). E. Mahieu is Research Associate with F.R.S. – FNRS. We are grateful to the many colleagues who have contributed to FTIR data acquisition at the Jungfraujoch station. The Atmospheric Chemistry Experiment (ACE), also known as SCISAT, is a Canadian-led mission mainly supported by the Canadian Space Agency and the Natural Sciences and Engineering Research Council of Canada. We thank D. Smale from NIWA Lauder for producing and making public the ethane time series of Lauder in the framework of NDACC. We warmly thank Jeremy Harrison and colleagues for their high quality lab measurements of C₂H₆.