Recent ethane increase above North America: comparison between FTIR measurements and model simulations

B. Franco[1], W. Bader[1], E. Mahieu[1], B. Bovy[1], E. V. Fischer[2], Z. A. Tzompa-Sosa[2], K. Strong[3], S. Conway[3], J. W. Hannigan[4], E. Nussbaumer[4], K. Sudo[5], P. F. Bernath[6,7,8], C. D. Boone[8] & K. A. Walker[3,8]

[1]University of Liège (bruno.franco@ulg.ac.be), [2]Colorado State University, [3]University of Toronto, [4]NCAR Boulder, [5]Nagoya University, [6]Old Dominion University, [7]University of York, [8]University of Waterloo

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Outline

1. Ethane retrieval strategy
2. Recent ethane increase
3. Comparison with GEOS-Chem
4. Ongoing work

1. Ethane retrieval strategy

Strategy originally developed by E. Mahieu (project UFTIR) and then adopted by the NDACC network

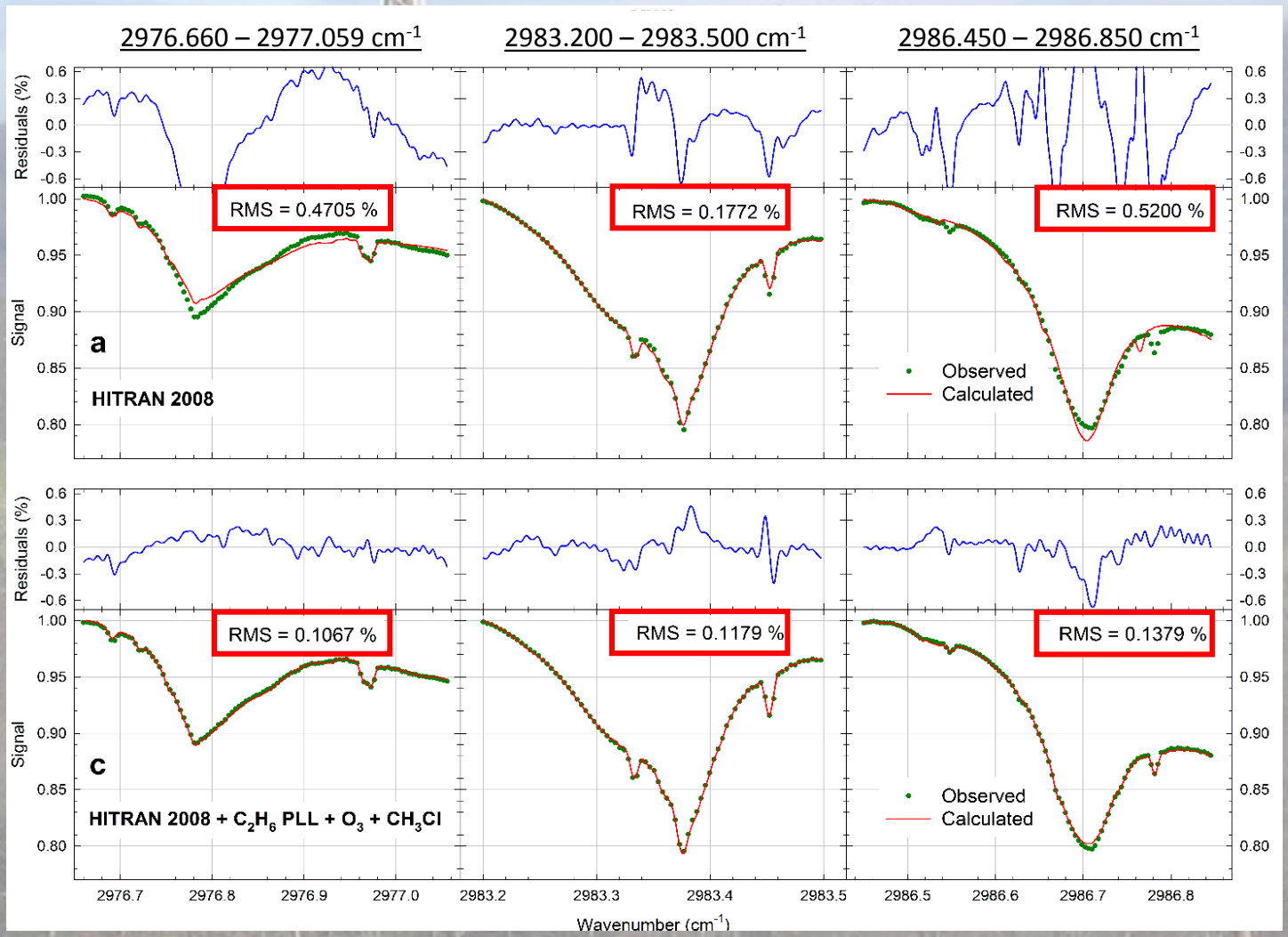
<u>Spectroscopic parameters</u>

HITRAN 2008

C_2H_6 pseudolines (G. C. Toon) based on J. J. Harrison's lab measurements

Update for three O_3 lines (P. Chelin)

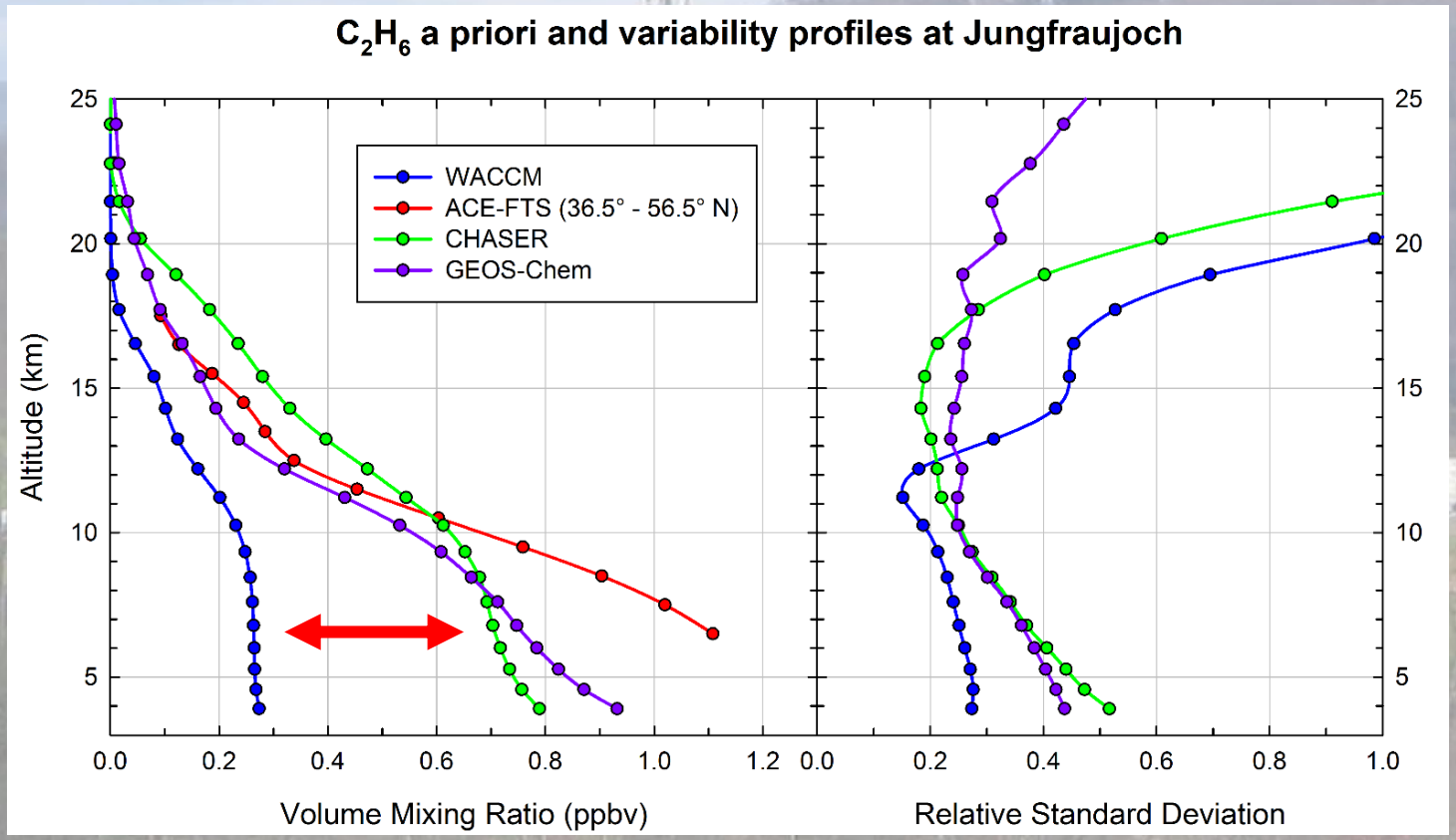
Update for CH_3Cl line positions and intensities (C. Bray)



NB: Changes in HITRAN 2012 temperature and pressure-dependency parameters of the H_2O feature at 2983.316 cm^{-1} increase the residuals

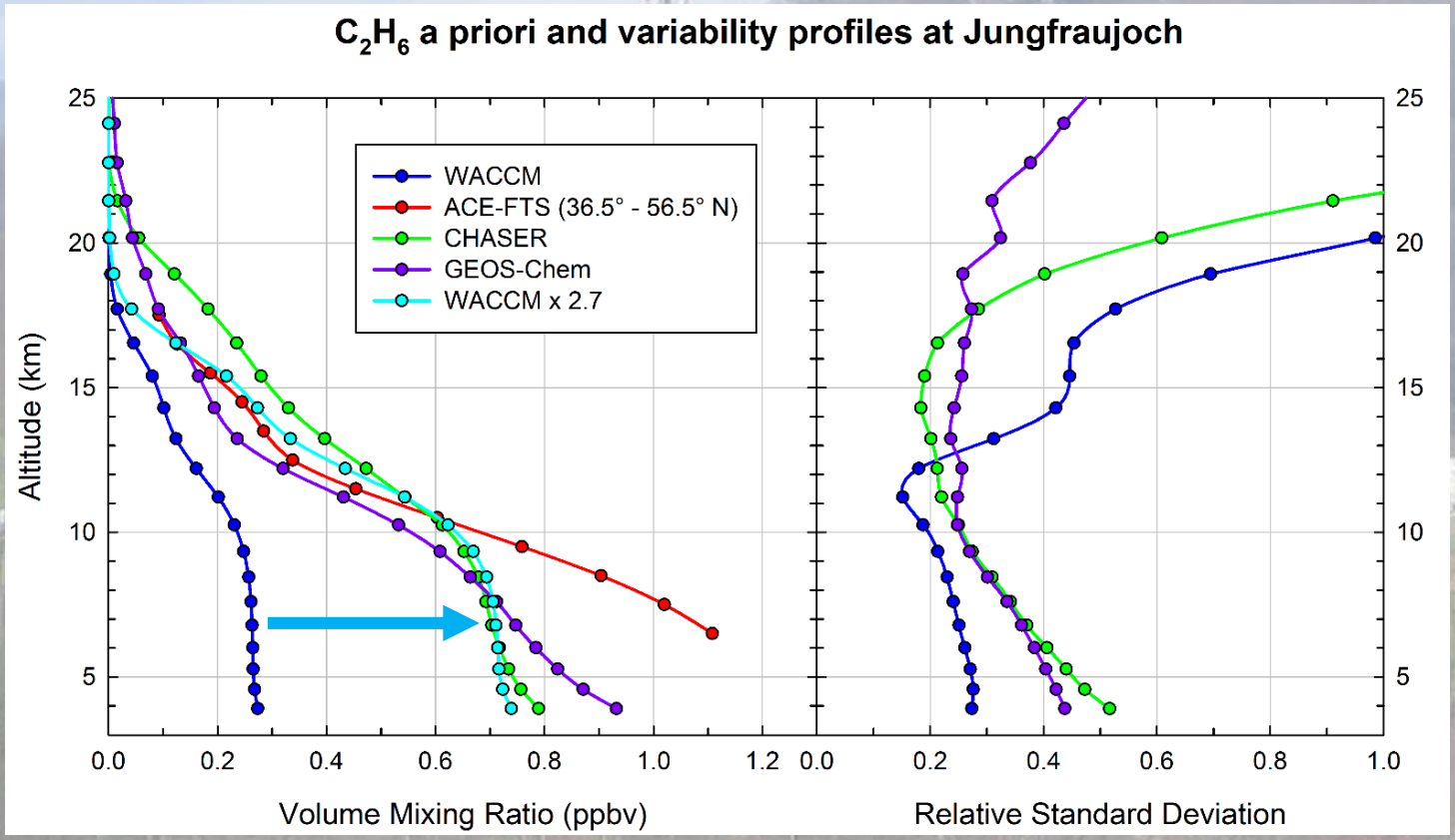
1. Ethane retrieval strategy

=> A priori and variability profiles derived from a 2007-2009 **CHASER** simulation (K. Sudo)



=> Large discrepancies between **WACCM**, and **ACE-FTS**, **CHASER** and **GEOS-Chem** profiles

1. Ethane retrieval strategy

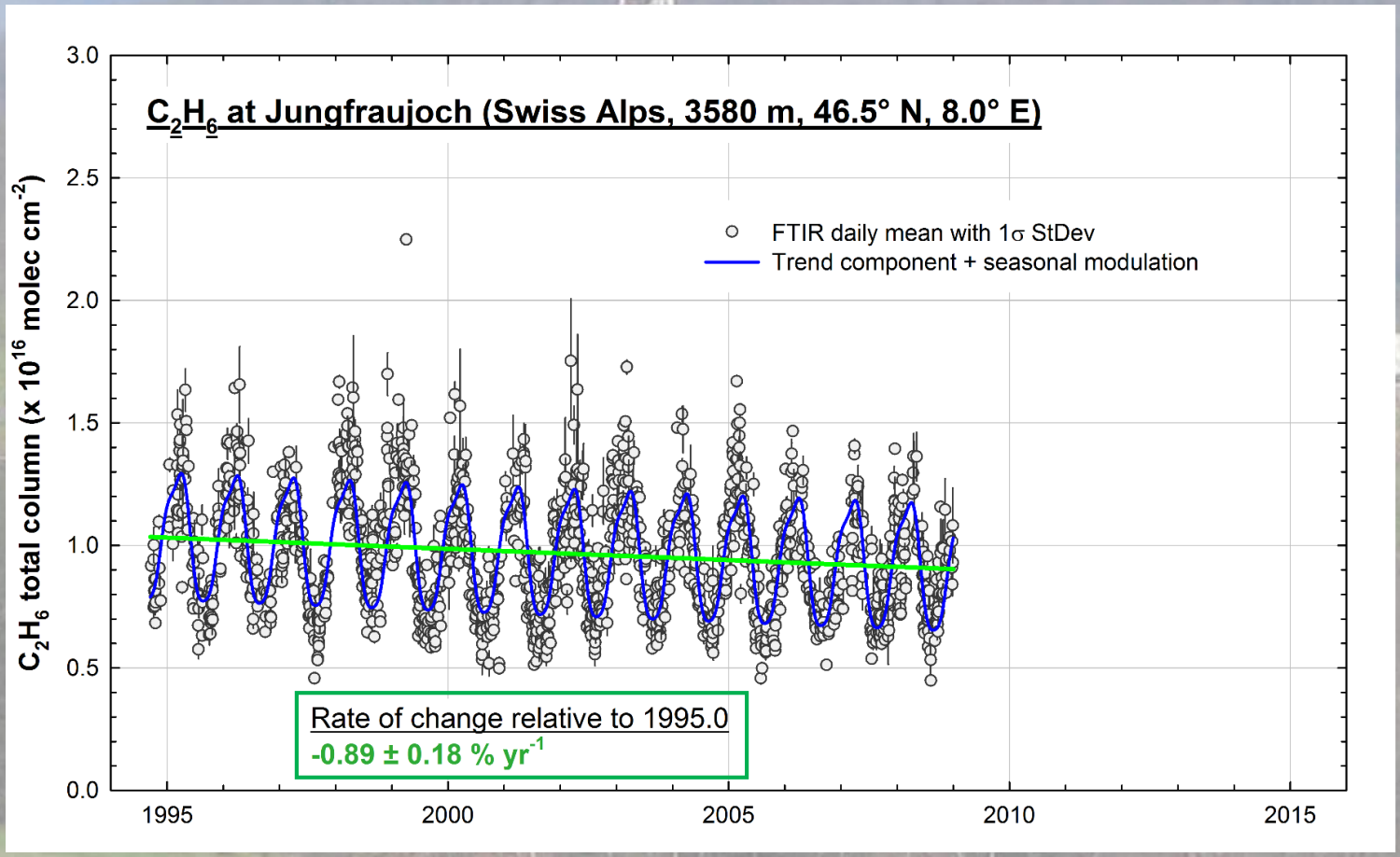


⇒ **WACCM** can be scaled by a constant factor to match the **ACE-FTS**, **CHASER** and **GEOS-Chem** profiles

⇒ No significant impact on the retrieved C_2H_6 columns, residuals of fitting and DOFS

2. Recent ethane increase

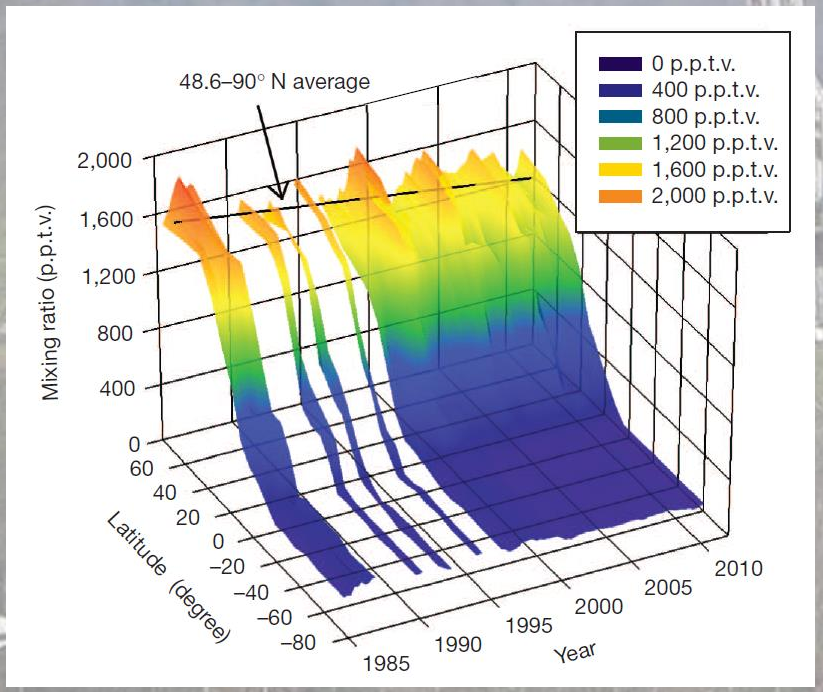
Trend analysis: bootstrap resampling tool, combining a linear function and a 3rd order Fourier series accounting for the intra-annual variability (Gardiner et al., 2008)



2. Recent ethane increase

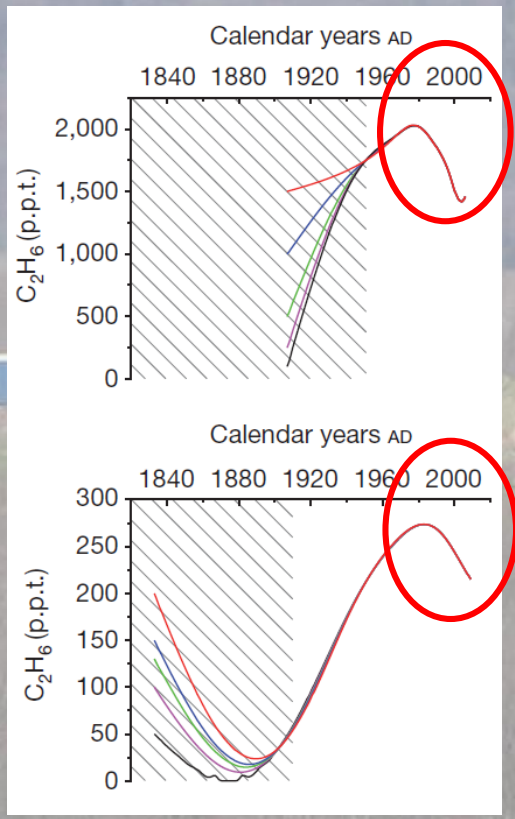
Regular decrease of atmospheric C_2H_6 burden consistent with the global decline of fugitive emissions from fossil fuel sources from the mid-1980s

-> from air sampling measurements



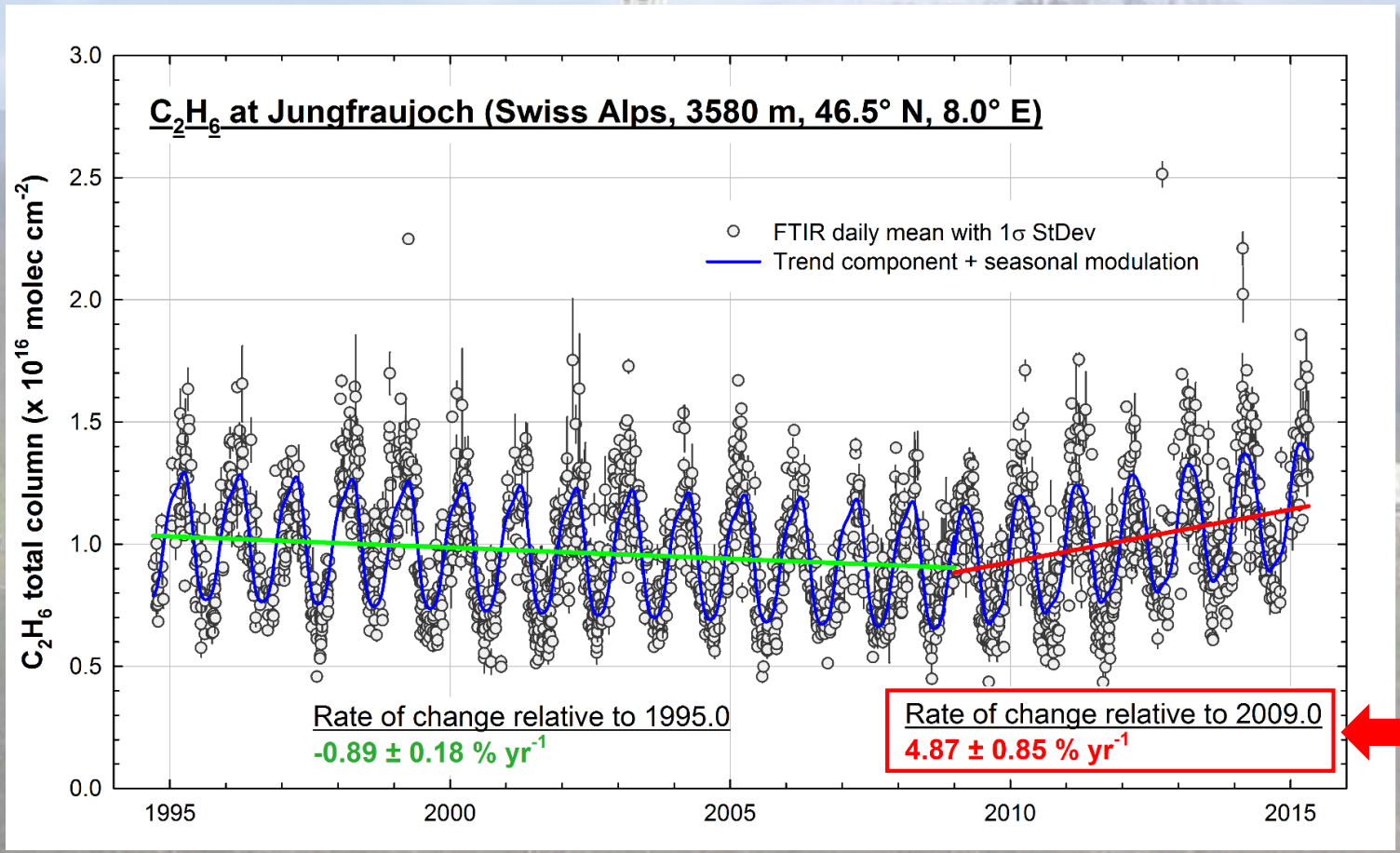
Simpson et al., 2012, *Nature*

-> from firn air measurements



Aydin et al., 2011, *Nature*

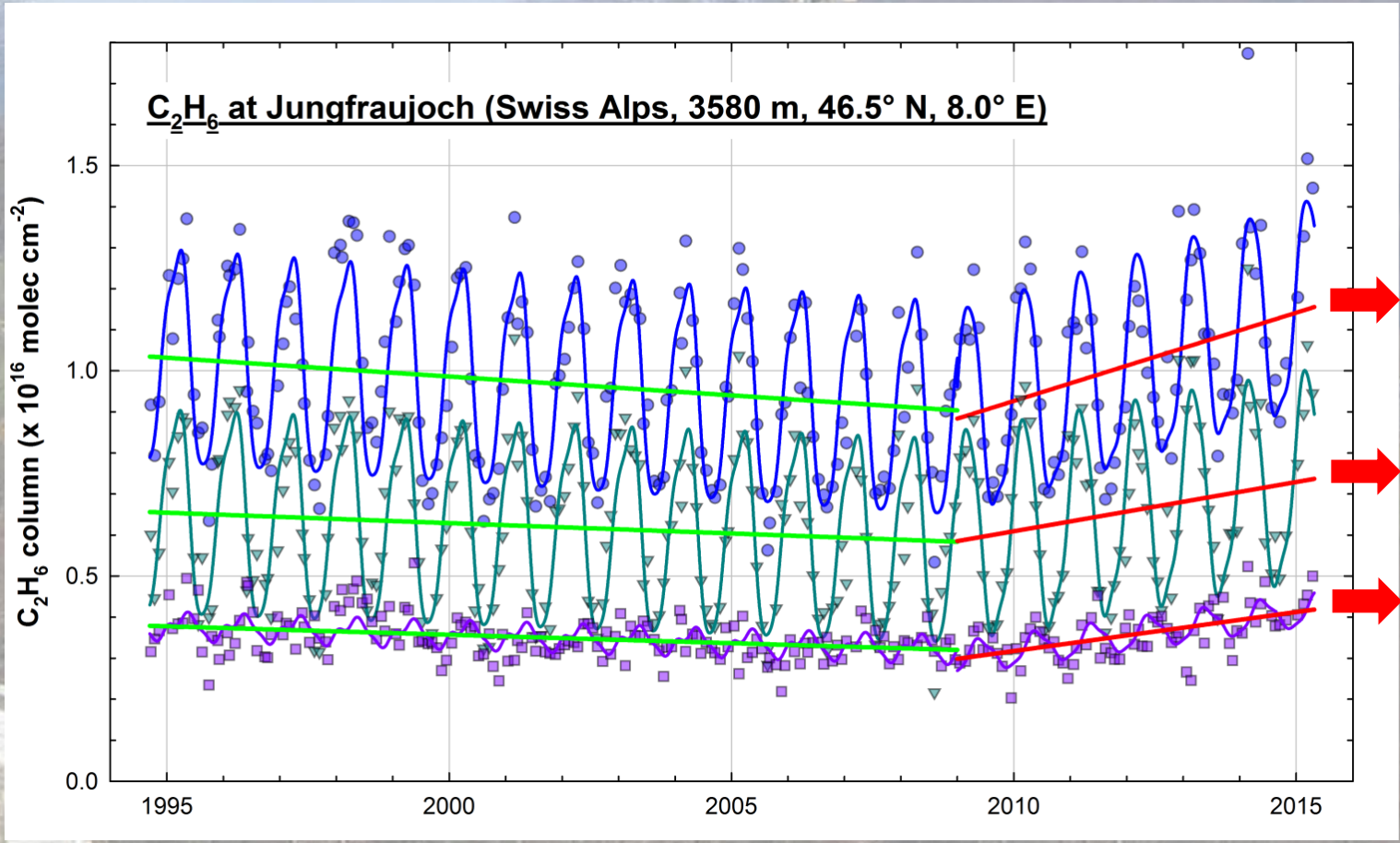
2. Recent ethane increase



Franco et al., 2015, *JQSRT*

2. Recent ethane increase

- Two independent partial columns may be deduced from the FTIR retrievals (DOFS ≈ 2.0)
- **Vertically-homogeneous** increase of C_2H_6 throughout the troposphere and lower stratosphere



Total columns:
=> +4.9 ± 0.8 % yr^{-1}

Lower troposphere (< 8 km)
=> +4.1 ± 0.9 % yr^{-1}

UTLS (8 - 20 km)
=> +6.4 ± 1.0 % yr^{-1}

2. Recent ethane increase

Cause of the C_2H_6 rise from 2009 onwards?

- **Sharp fluctuations of OH concentration in the atmosphere?**

  ✓ The gobal OH levels have not exhibited large interannual variability since the end of the 20th century

  => Montzka et al., 2011, Small Interannual Variability of Global Atmospheric Hydroxyl, *Science* ***331****, 67*

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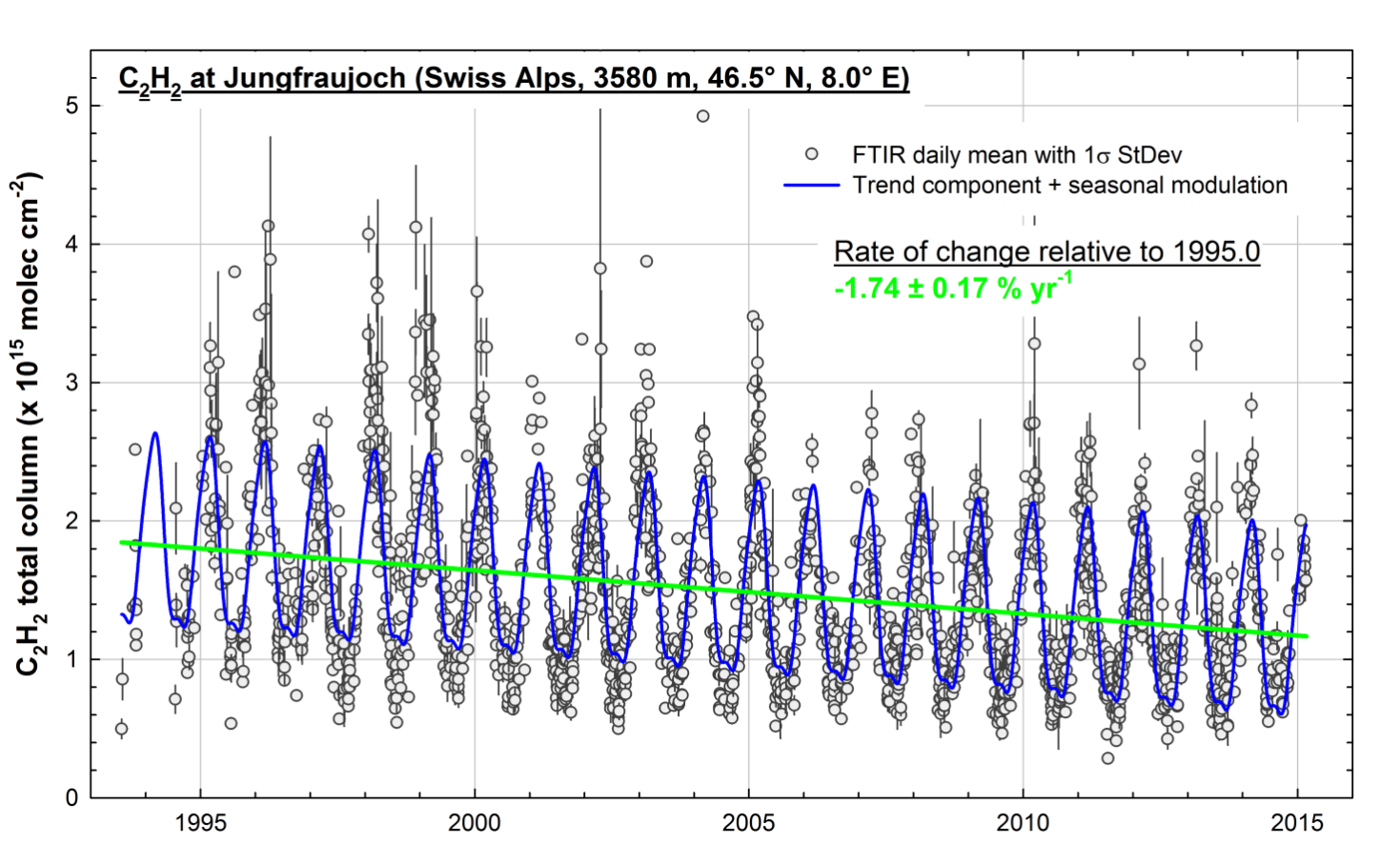
  ✓ Neither CO nor other species that have oxidation by OH as their major removal pathway such as HCN and C_2H_2, present an upturn in their retrieved columns

2. Recent ethane increase



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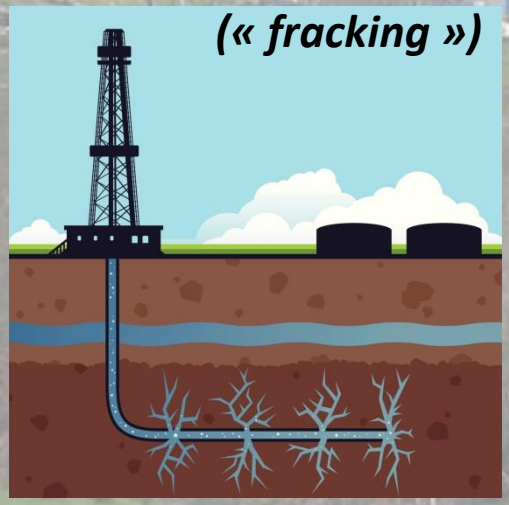
- **Product of enhanced fugitive emissions?**

  ✓ May be linked to the recent massive growth in the **exploitation of shale gas and tight oil reservoirs**, especially in North America

Hydraulic fracturing
(« fracking »)



www.huffingtonpost.com

Texas Barnett Shale gas drilling rig near Alvarado, Texas. Courtesy of David R. Tribble

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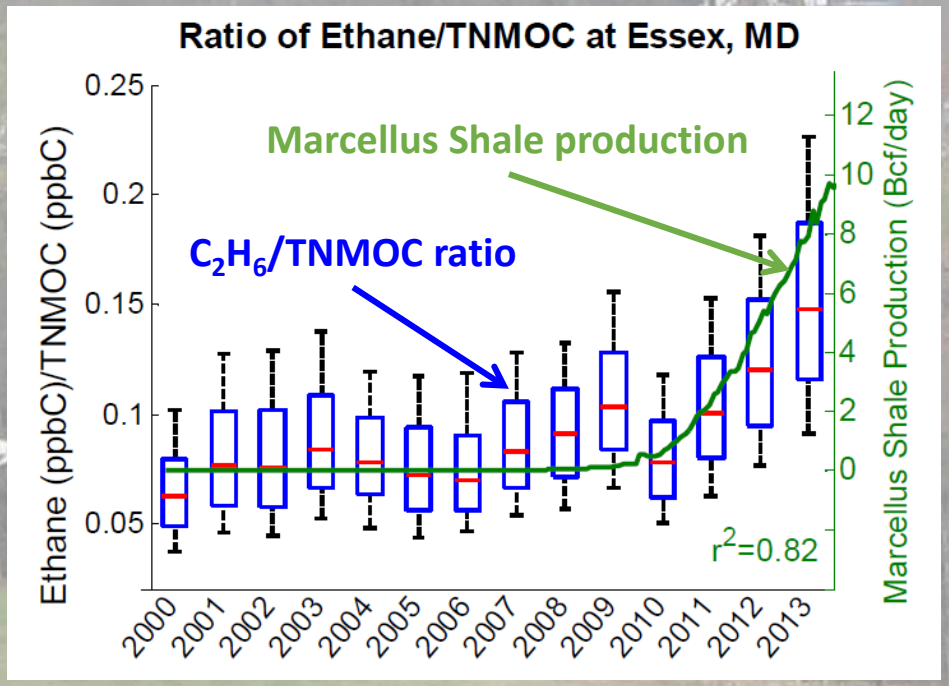
  ✓ Increases of hydrocarbons related to oil and gas industries are detected over North American regions where the drilling productivity began to grow rapidly after 2009



PAMS measurements at Essex, MD, located downwind from the giant Marcellus Shale play (WV, PA and NY)

Vinciguerra et al., 2015, Regional air quality impacts of hydraulic fracturing and shale natural gas activity: Evidence from VOC observations, *Atmospheric Environment 110, 144-150.*

2. Recent ethane increase

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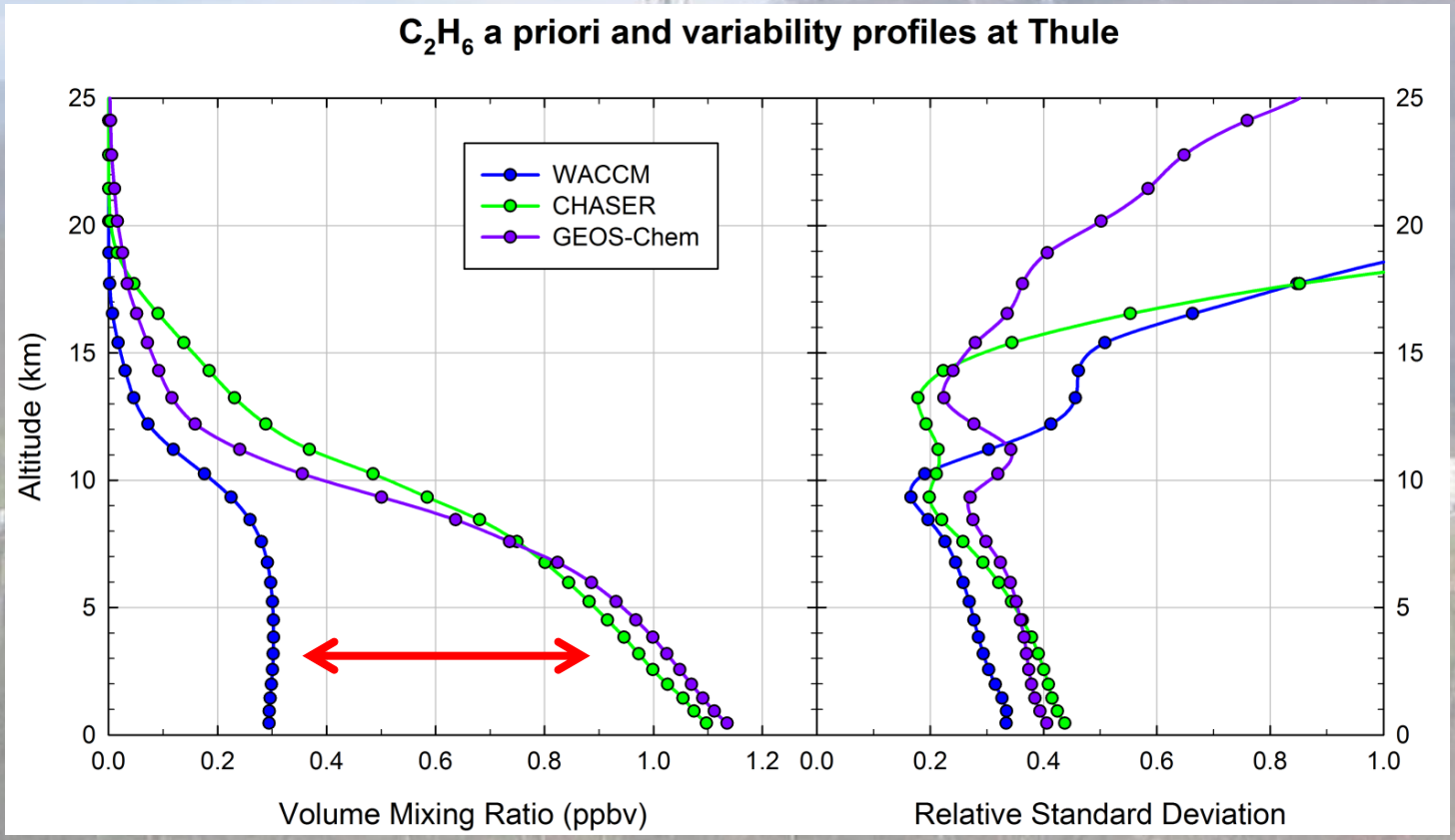
  - ✓ Increases of hydrocarbons related to oil and gas industries are detected over North American regions where the drilling productivity began to grow rapidly after 2009

  - ✓ Could represent a change in C_2H_6 throughout the Northern Hemisphere

  - ✓ **FTIR and ACE-FTS** measurements over North America support this hypothesis

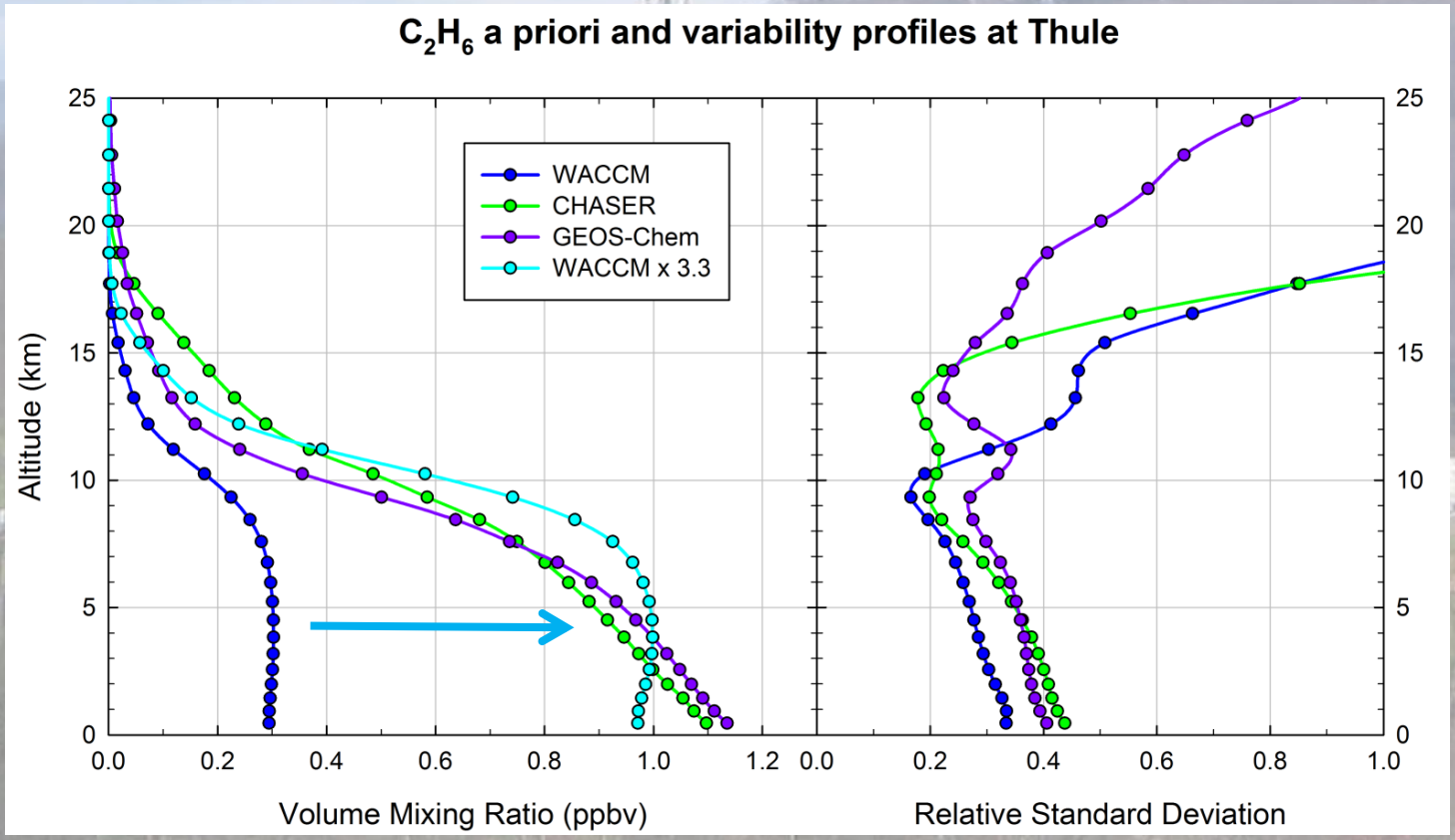
Toronto, Boulder, Thule, Eureka…

2. Recent ethane increase



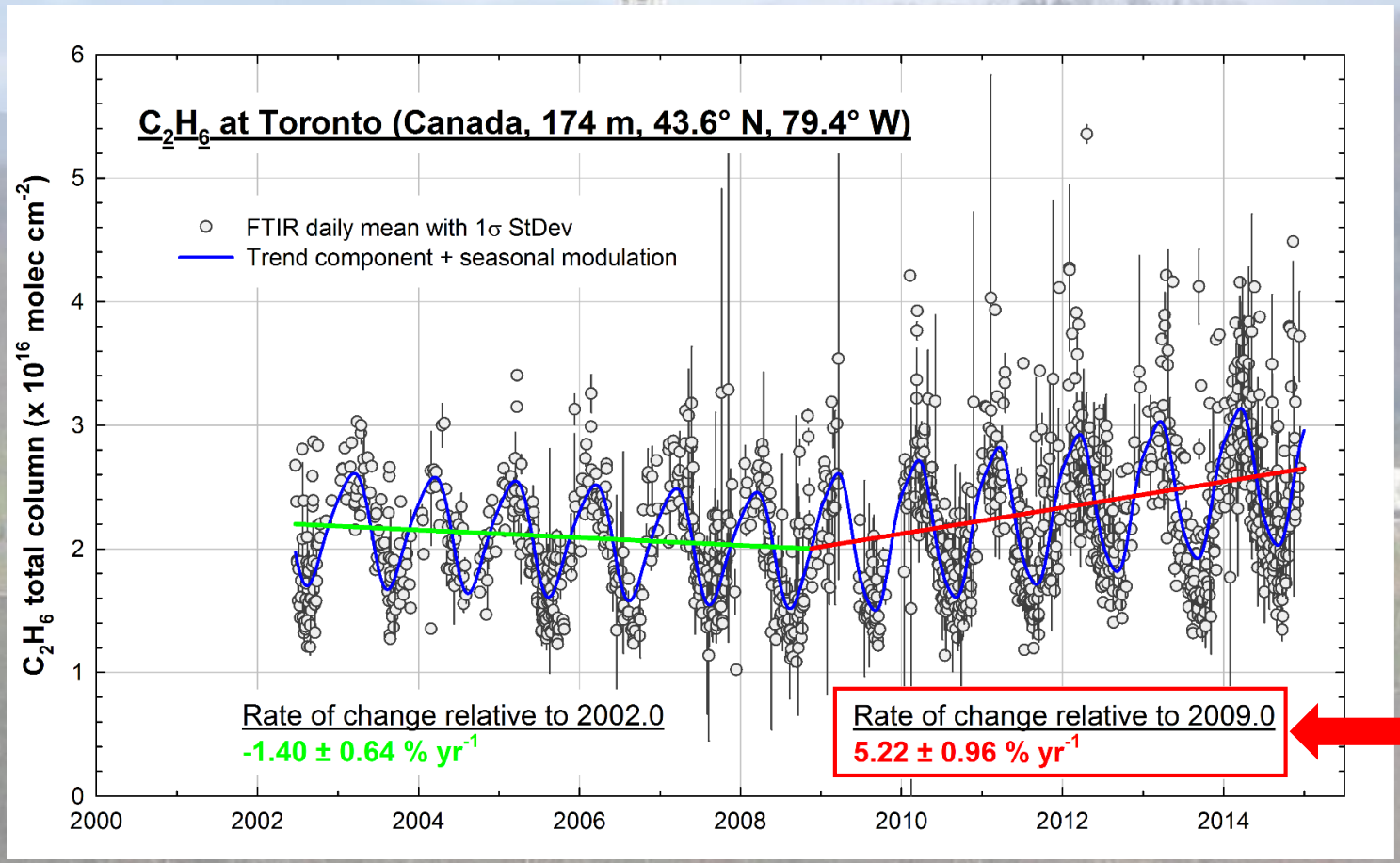
=> Large discrepancies between **WACCM**, and **CHASER** and **GEOS-Chem** profiles

2. Recent ethane increase



=> **WACCM** is scaled by a constant factor to match the **CHASER** and **GEOS-Chem** profiles

2. Recent ethane increase



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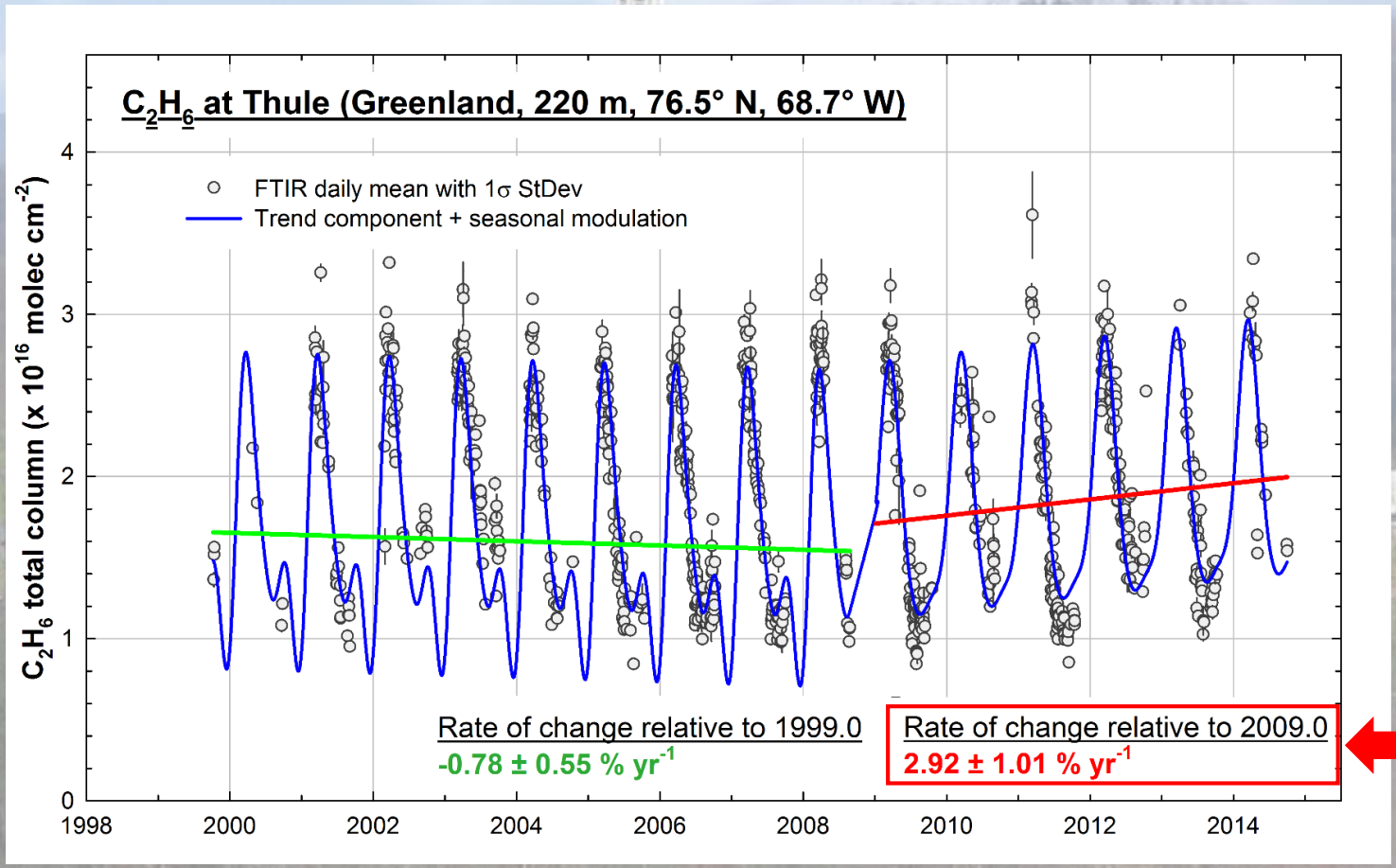

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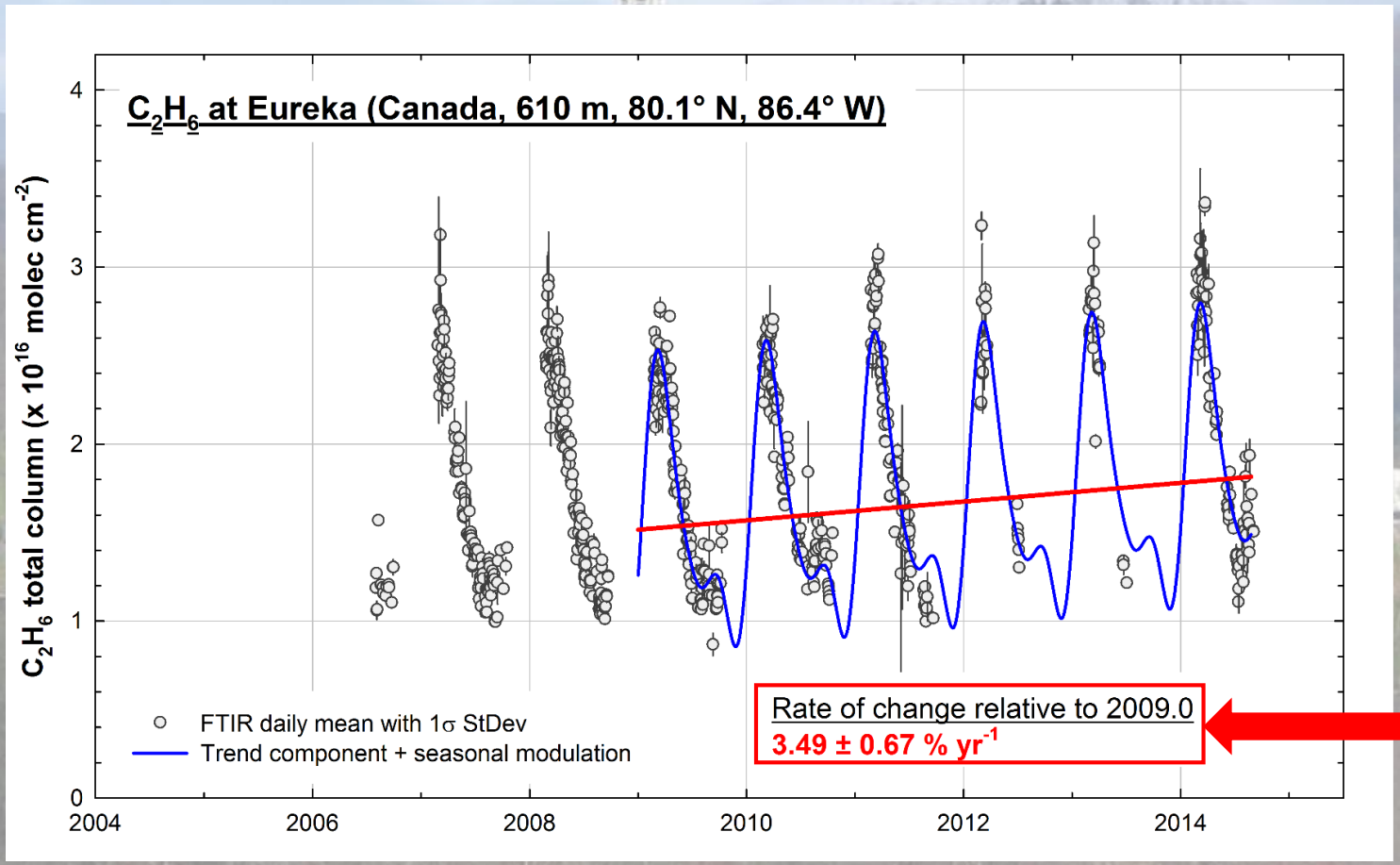
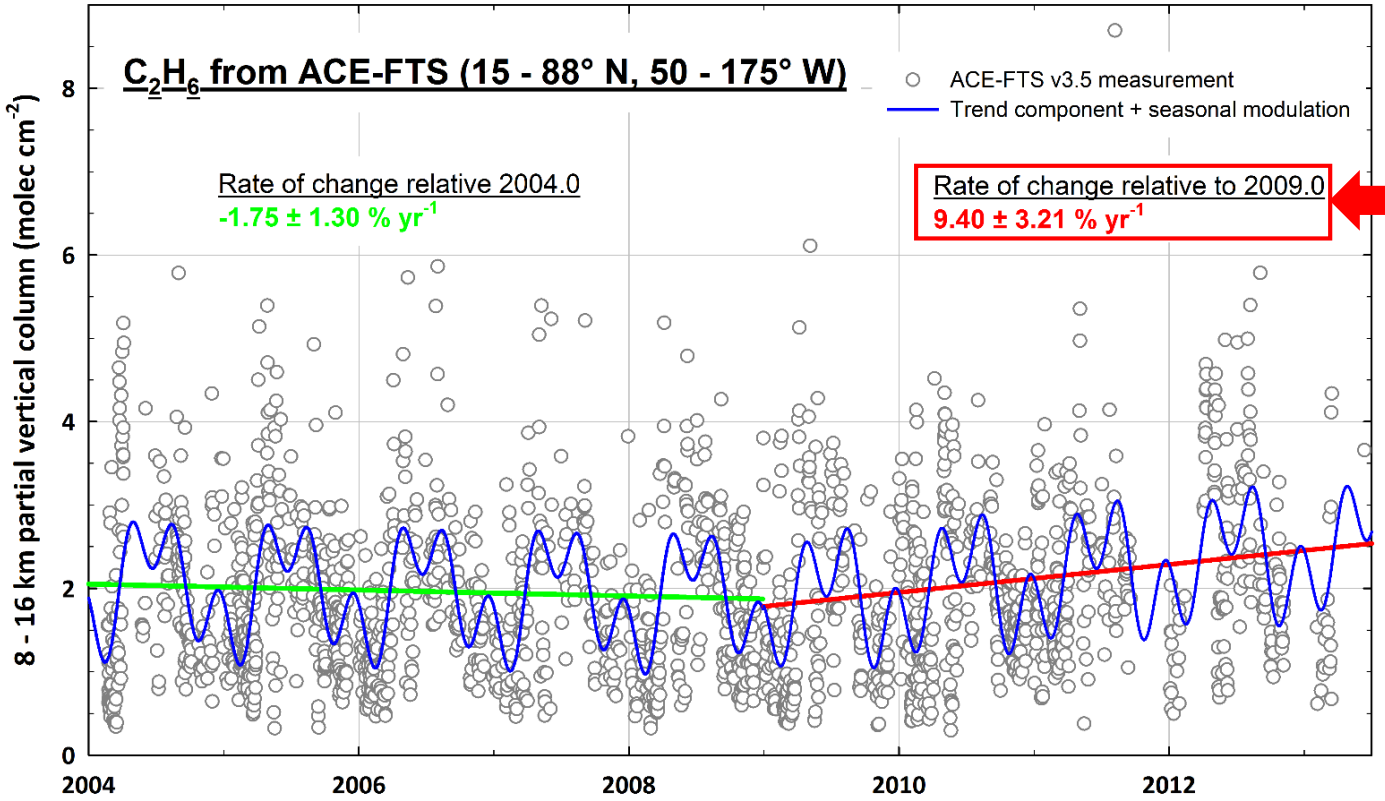


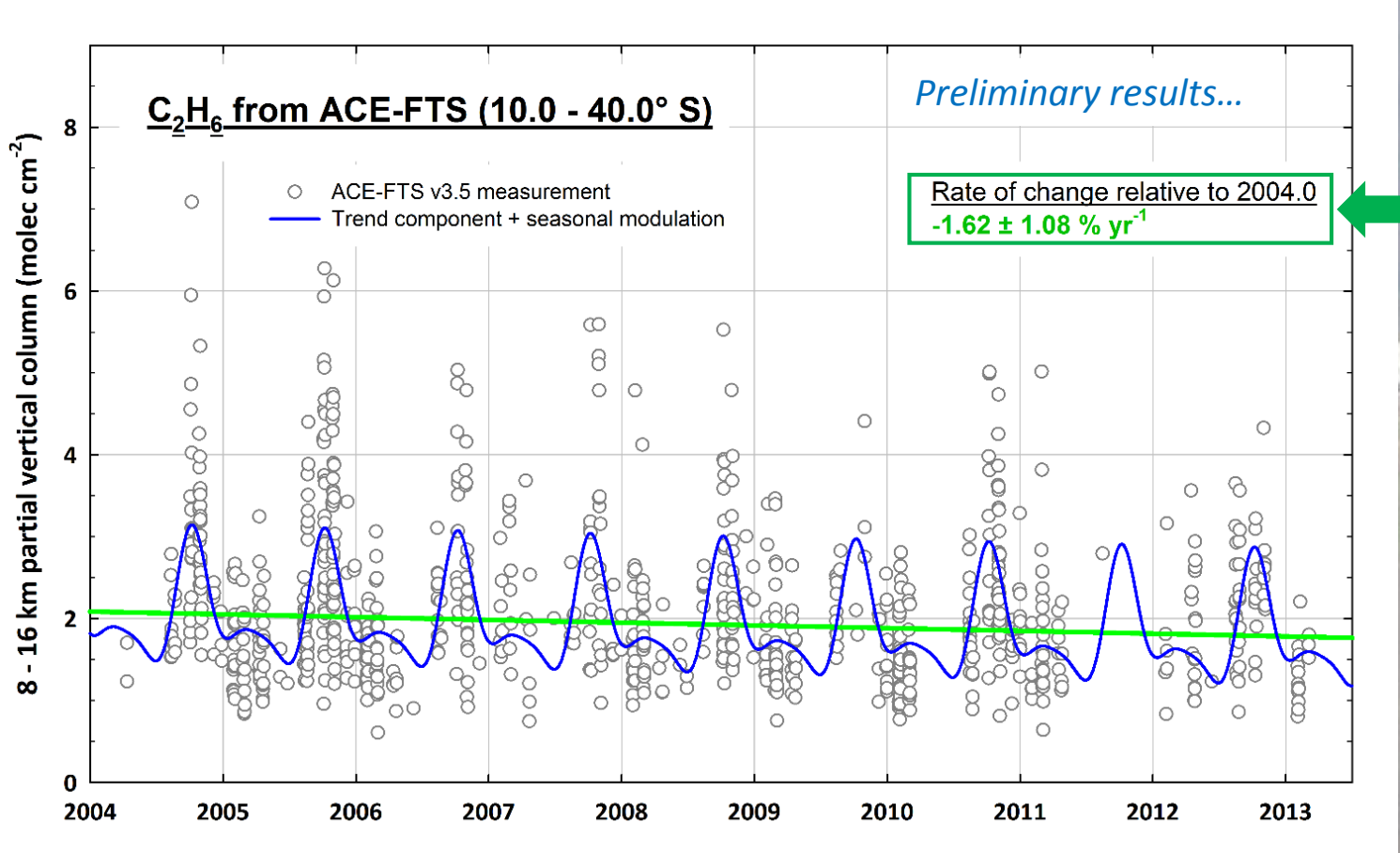
2. Recent ethane increase

Preliminary results…



2. Recent ethane increase

- FTIR and ACE-FTS observations in the Southern Hemisphere do not reveal any recent increase
- Weak latitudinal exchange of C_2H_6 between both hemispheres
- Observed increase of C_2H_6 currently **limited to the Northern Hemisphere**



3. Comparison with GEOS-Chem

Comparison between **FTIR** measurements and **GEOS-Chem** simulations

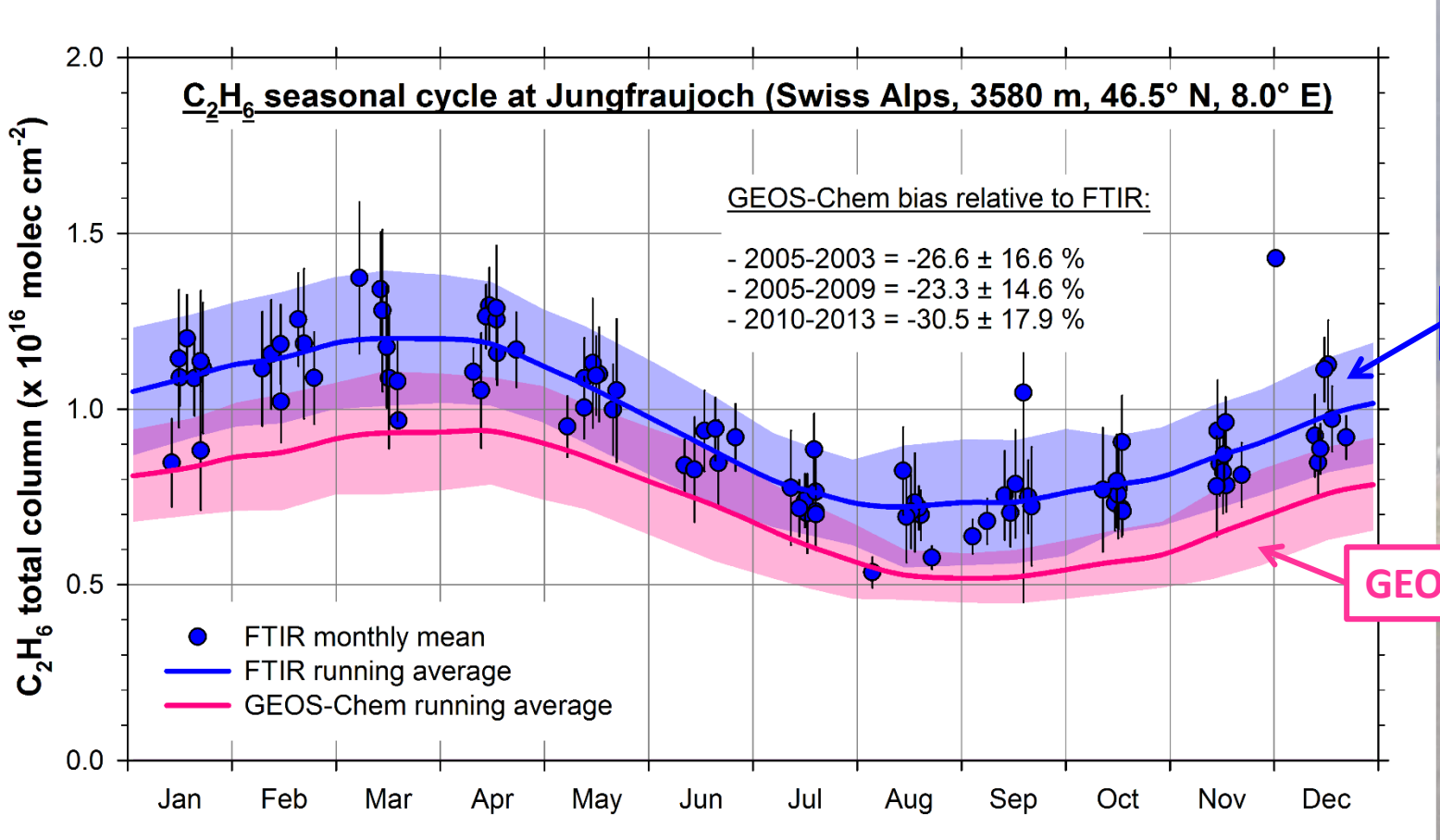
- **GEOS-Chem CTM**:
  => v9-01-03 standard full-chemistry simulation
  => 2° x 2.5° horizontal resolution
  => driven by the GEOS-5 meteorological fields
  => simulations over the mid-2005 – mid-2013 time period

- The vertical resolution and specific sensitivity of the FTIR retrievals are accounted for:
  => VMR profiles interpolated onto the FTIR vertical layer scheme
  => then smoothed by applying the FTIR AVKs

- The comparisons are made for the 2005 – 2013 observational days only

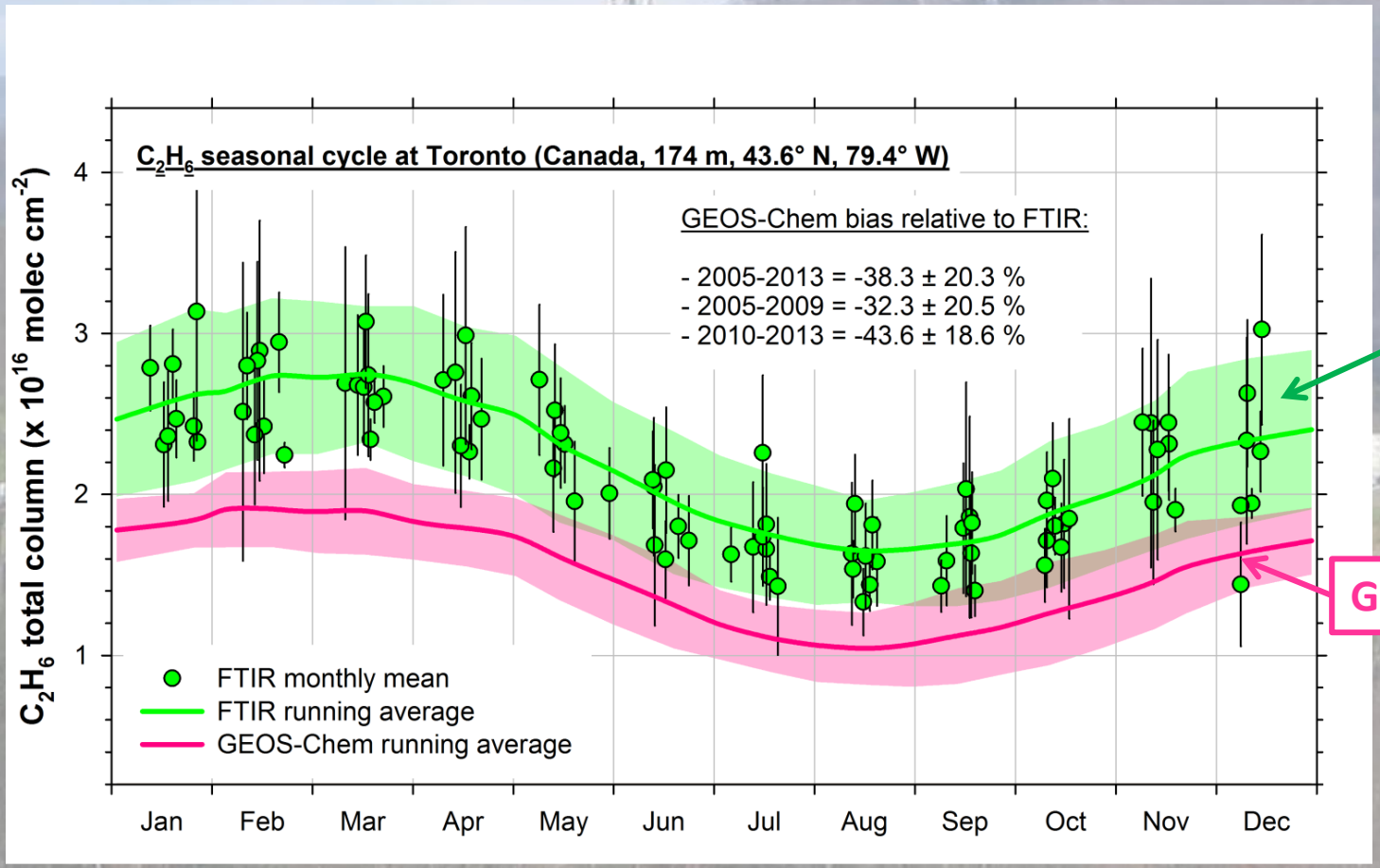
3. Comparison with GEOS-Chem

- C_2H_6 total columns (2005-2013) displayed on a 1-year time base
- Comparison made on the basis of the daily means



=> Good agreement (R = 0.77)… but systematic negative bias relative to FTIR

3. Comparison with GEOS-Chem



=> Good agreement (R = 0.73)… but systematic negative bias relative to FTIR

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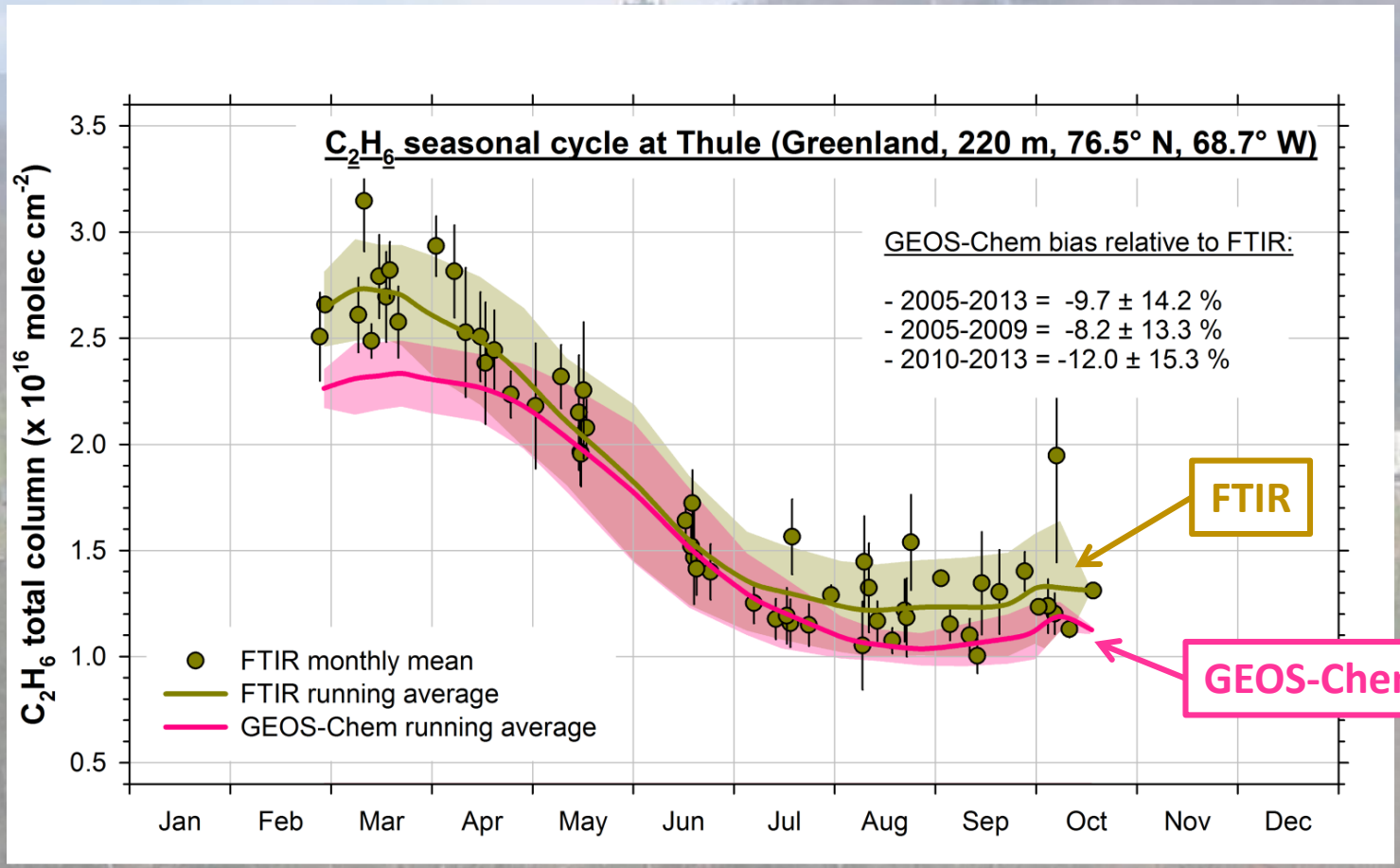
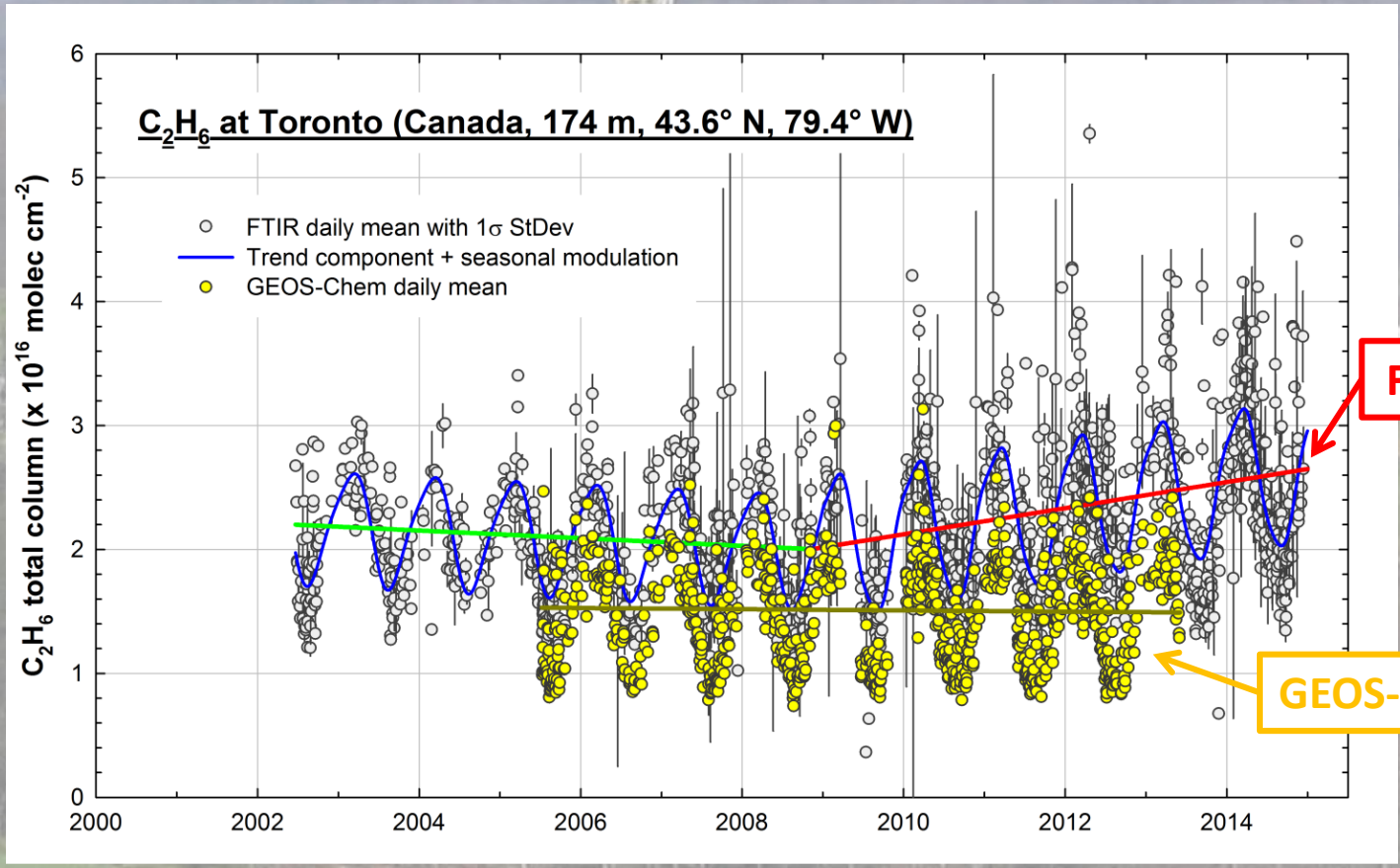


=> Good agreement (R = 0.93)… but systematic negative bias relative to FTIR

3. Comparison with GEOS-Chem

- Underestimation of C_2H_6 emissions in the current inventories implemented by GEOS-Chem
- The recent C_2H_6 increase is not captured by the model



4. Ongoing work

Incorporating updated inventories in GEOS-Chem

- to reconcile FTIR and GEOS-Chem results
- to reproduce the observed C_2H_6 increase

Current C_2H_6 emission inventories:

- Anthropogenic and pyrogenic sources from Xiao et al., 2008 (*JGR*)
  => offline simulation based on surface and aircraft observations
- Biomass burning sources from the GFED3 database (monthly means)

Updated C_2H_6 emission inventories: (E. V. Fischer and Z. A. Tzompa-Sosa, CSU)

- By applying C_2H_6/CH_4 emission ratios to satellite-derived CH_4 emissions (Turner et al., 2015, *ACPD*)
  => CH_4 emissions inferred from GOSAT and subsequently evaluated by surface and aircraft data

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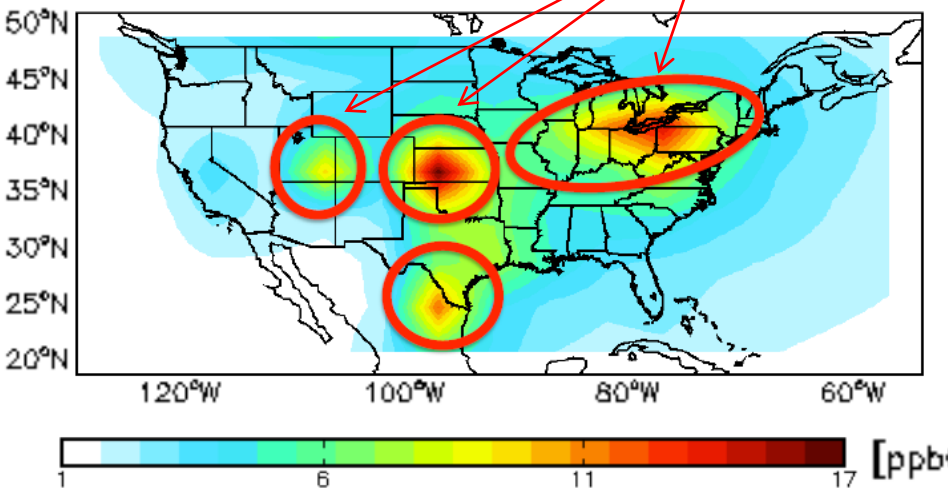
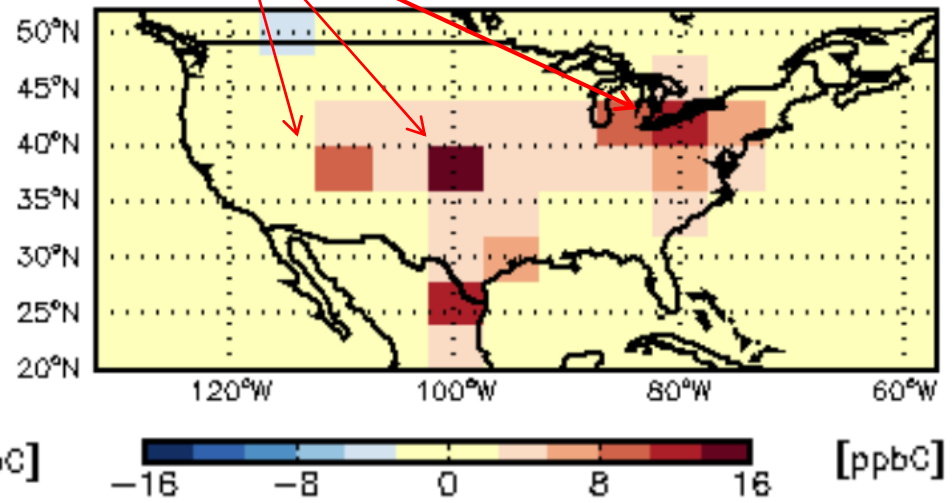
Preliminary results…

Major oil and gas basins

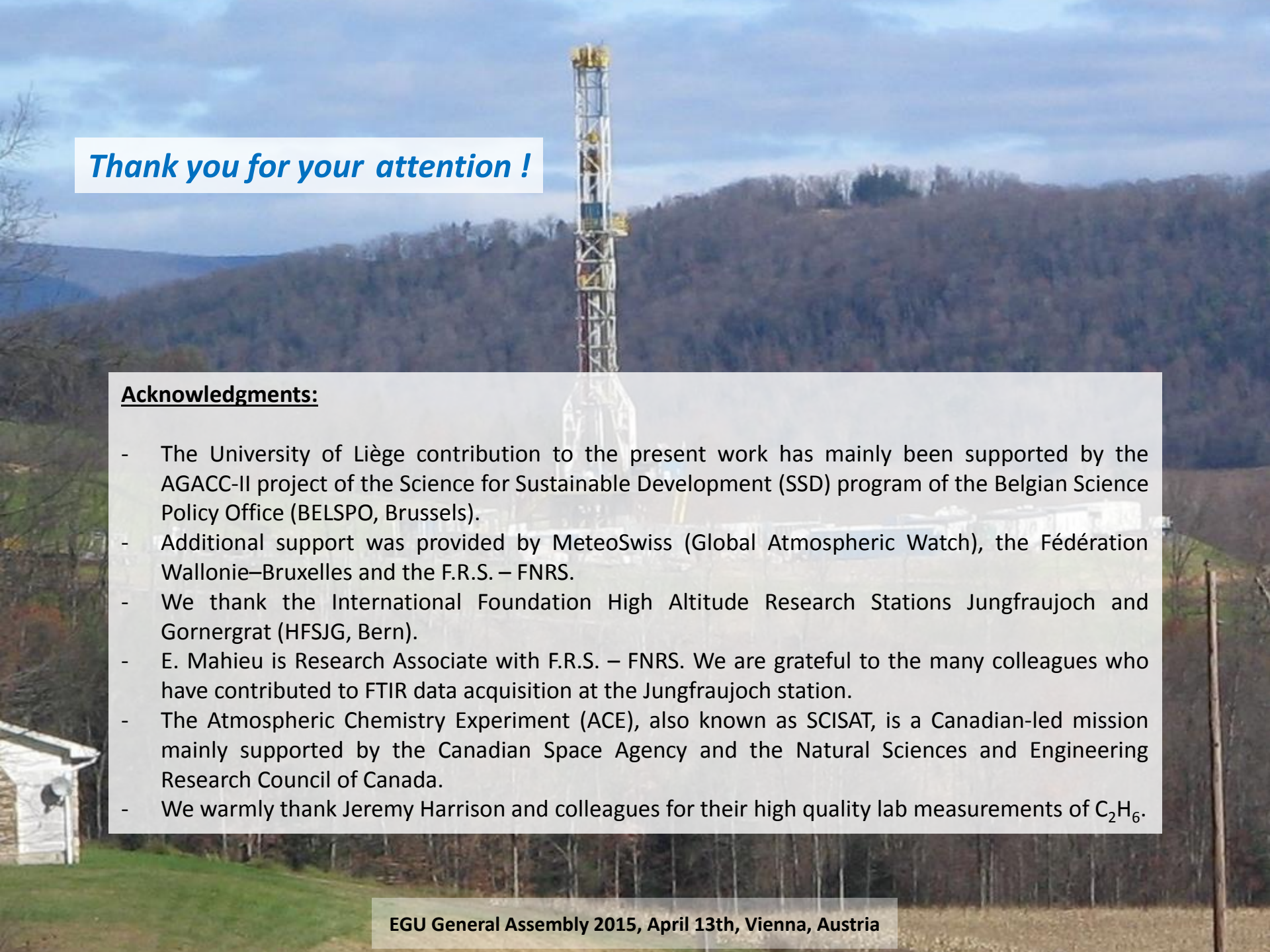
Courtesy of Z. A. Tzompa-Sosa (CSU)



GEOS-Chem C_2H_6 surface concentration for December 2010



December 2010 absolute difference in surface C_2H_6 (ppbC) for a simulation using emission ratios to CH_4 vs. Xiao et al. (2008) + GFED3 emission inventories



Thank you for your attention !

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- 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 warmly thank Jeremy Harrison and colleagues for their high quality lab measurements of C_2H_6.