The question of the methane increase through the retrieval of its isotopologues CH_3D and $^{13}CH_4$ from FTIR ground-based observations

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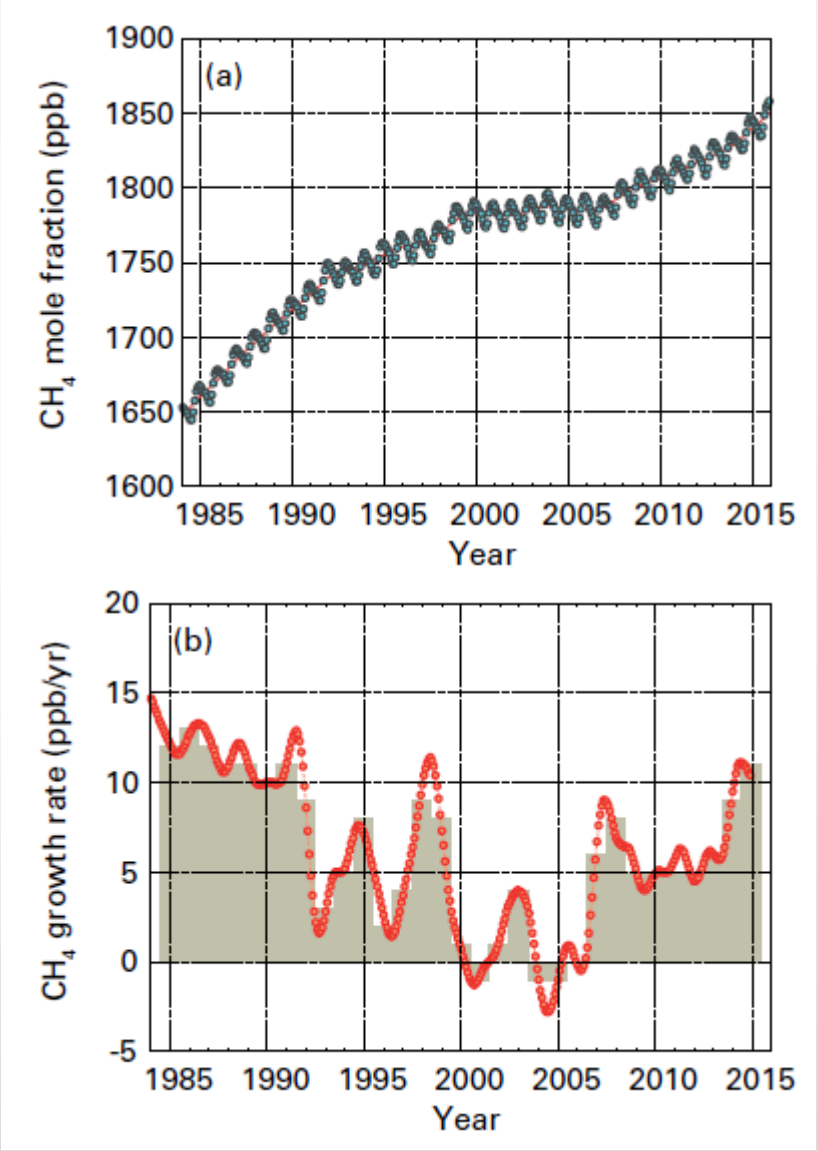
Introduction and motivation

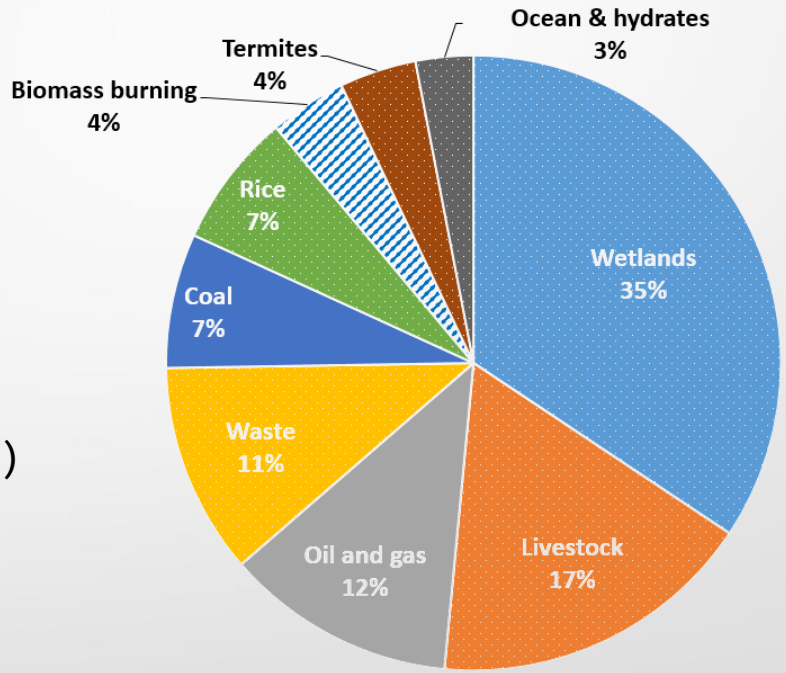
- Atmospheric methane CH_4
  - Lifetime of 8-10 years
  - 2nd most important anthropogenic GHG
  - 1/5th of radiative forcing since 1750
  - New high of 1845 ± 2 ppb
  - + 256 % since pre-industrial times
  - + ~0.3 %/year since mid-2000s
  - Changes remain unexplained
  - Rigby et al., 2008; Dlugokencky et al., 2009; Bloom et al., 2010; van der Werf et al., 2010; Frankenberg et al., 2011; Montzka et al., 2011; Spahni et al., 2011; Sussmann et al., 2012; Hausmann et al., 2016; Helmig et al., 2016; Schaefer et al., 2016; Bader et al., 2017; Tuner et al., 2017; Rigby et al., 2017



WMO, Greenhouse gas bulletin N°12, 2016

How useful are FTIR isotopologues measurements ?

- 2 main isotopologues : CH_3D, $^{13}CH_4$
  - CH_3D : 6.158×10^{-4} rel. abundance
  - $^{13}CH_4$: 1.11×10^{-2} rel. abundance
- Specific emission ratio
  - Process type
    - Biogenic (dotted area)
    - Pyrogenic (plain area)
    - Thermogenic (hatched area)
- Kinetic Isotope Effect
  - Removal pathway



- CH_3D and $^{13}CH_4$ are good tracers of the methane budget and bring complementary information on the CH_4 isotopic signature

How useful are FTIR isotopologues measurements ?

- Satellite : Only ACE-FTS has a methane isotopologue product (Buzan et al., 2016)
- In situ : No CH_3D long term time series exist so far

  < detection limit for in situ measurements
- Aircraft : NASA ER-2 (Rice et al., 2003)

- Revised isotopic signature for sources (Schwitzke et al., 2016)
- Total columns of CH_3D and $^{13}CH_4$ will bring useful information on the methane increase discussion.

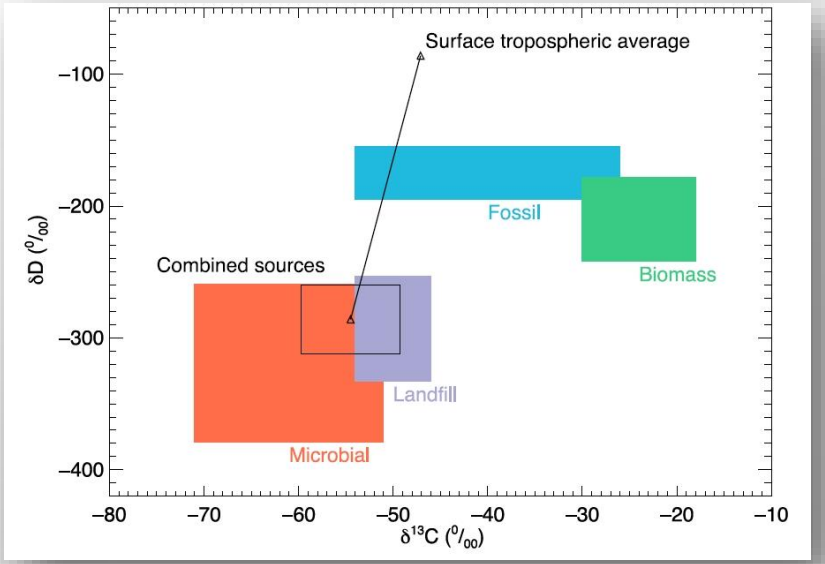


Fig. 2 from Rigby et al., 2012

Project Overview

2 years

Project timeline

Year 1

Remote sensing

CH_3D & $^{13}CH_4$
Eureka *(BONUS : PARIS instrument)*
Toronto
Lauder (few tests)
Jungfraujoch (need to implement SFIT-4)
+ BIRA sites (timeline to be discussed)

Year 2

GEOS-Chem Isotopic simulation

- ✓ Tagged simulation
- ✓ Emission inventories
- ✓ Emission ratios

Methane increase ?

Observations & Simulation
- ✓ CH_4
- ✓ CH_3D
- ✓ $^{13}CH_4$

Retrieval strategy for CH_3D

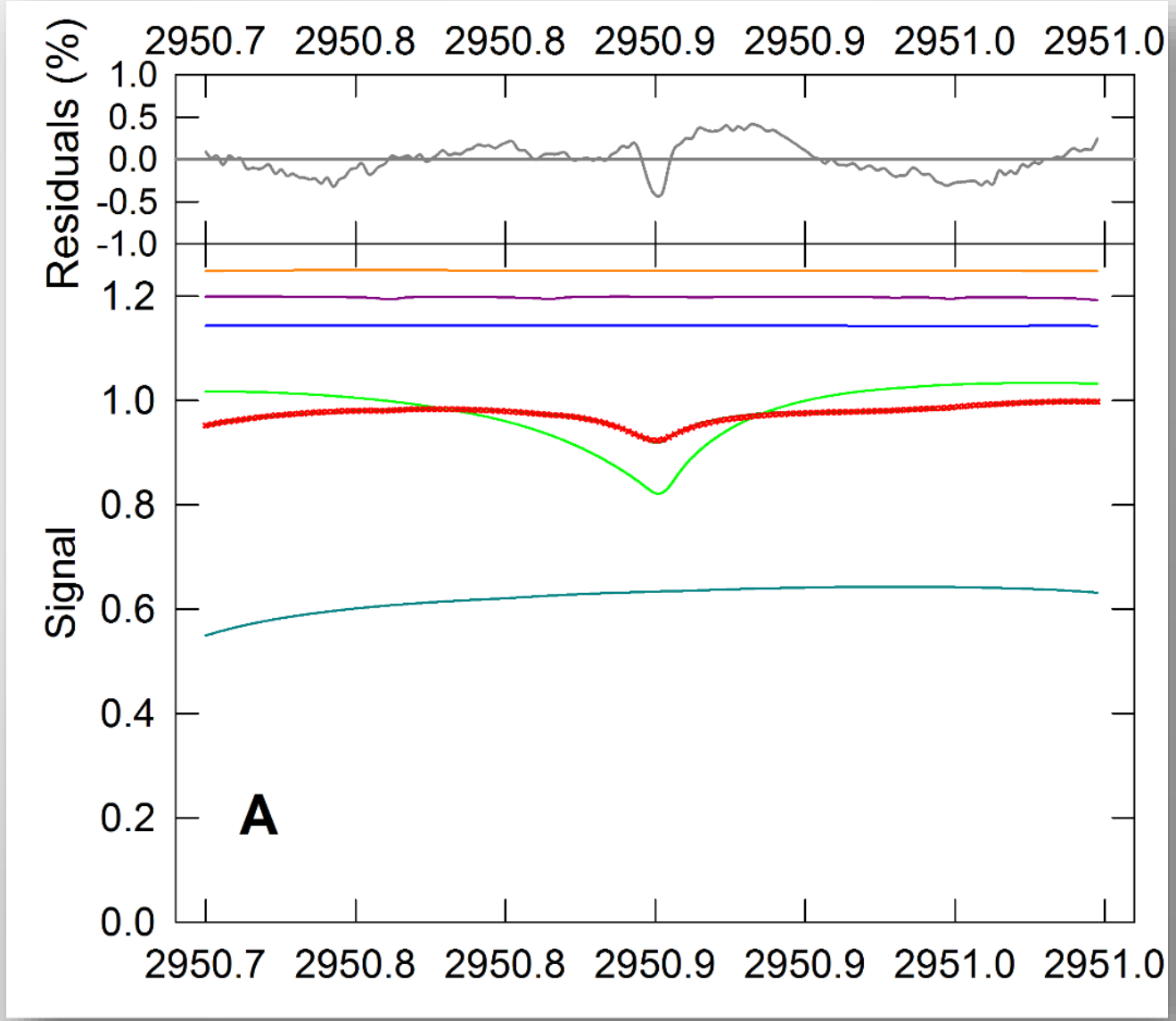
Tests over a whole year of observations

CH_3D Retrieval Strategy

Computed

Target gas
CH_3D

Interfering lines
CH_4
H_2O
O_3
Solar lines



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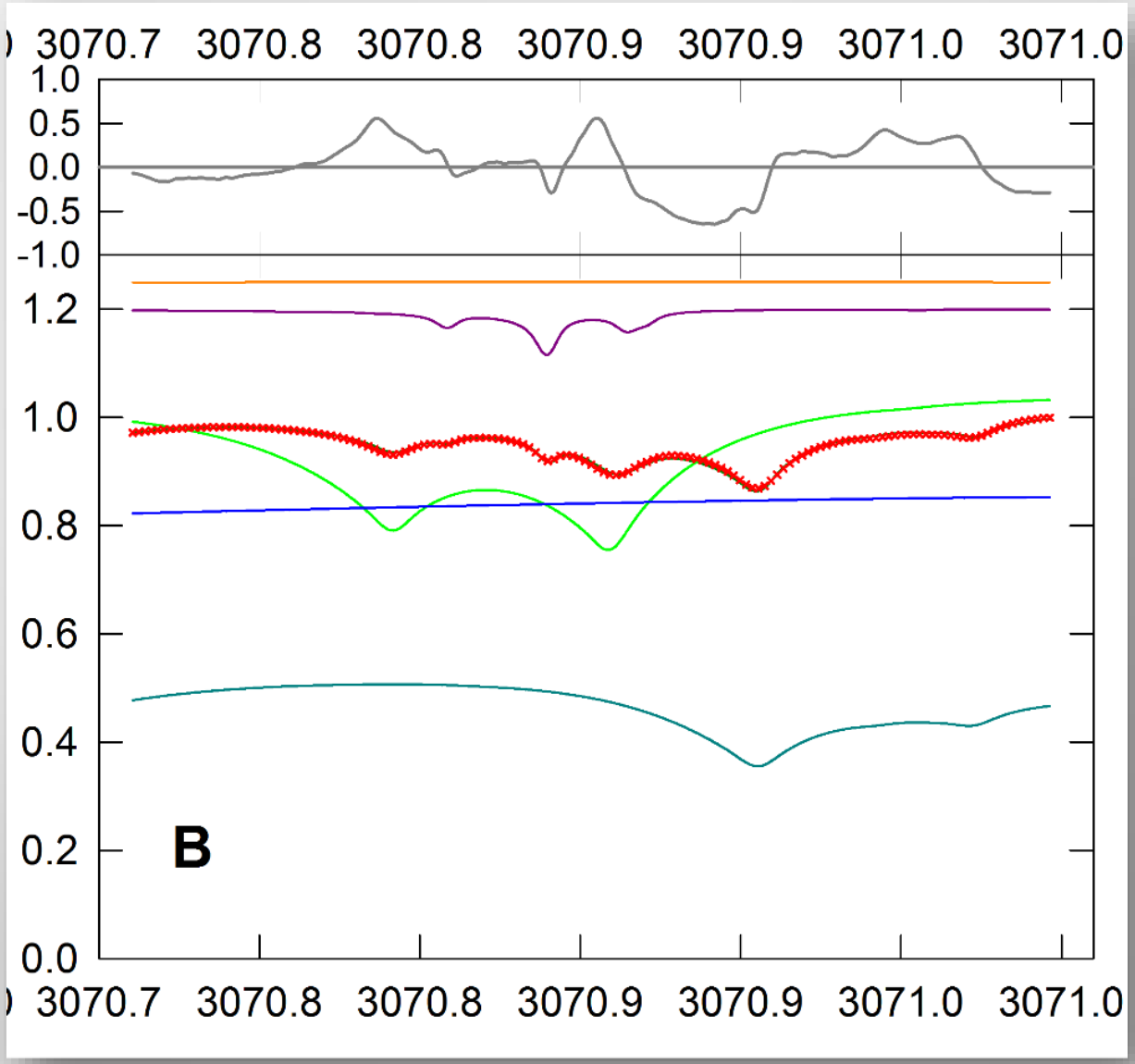

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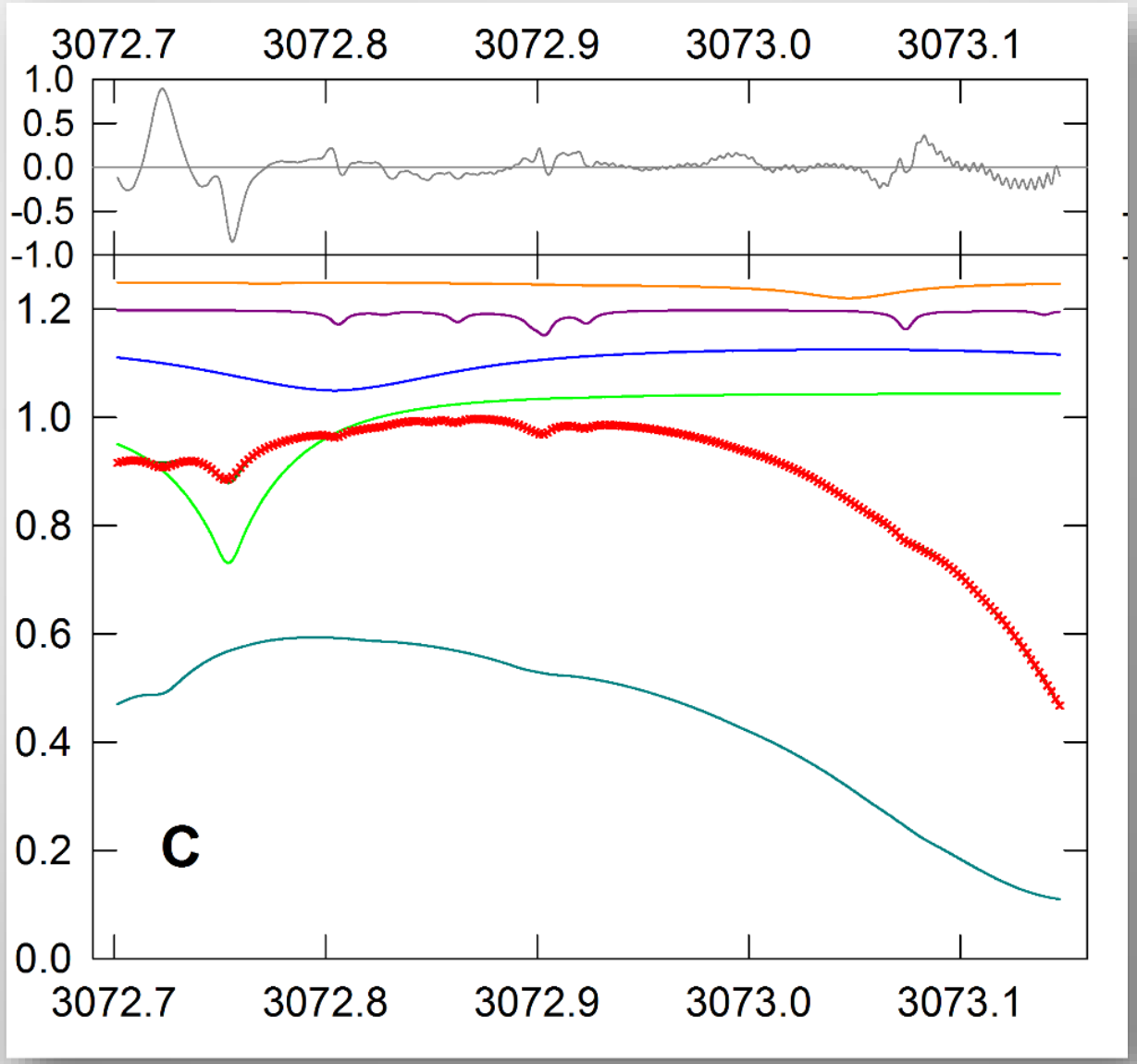

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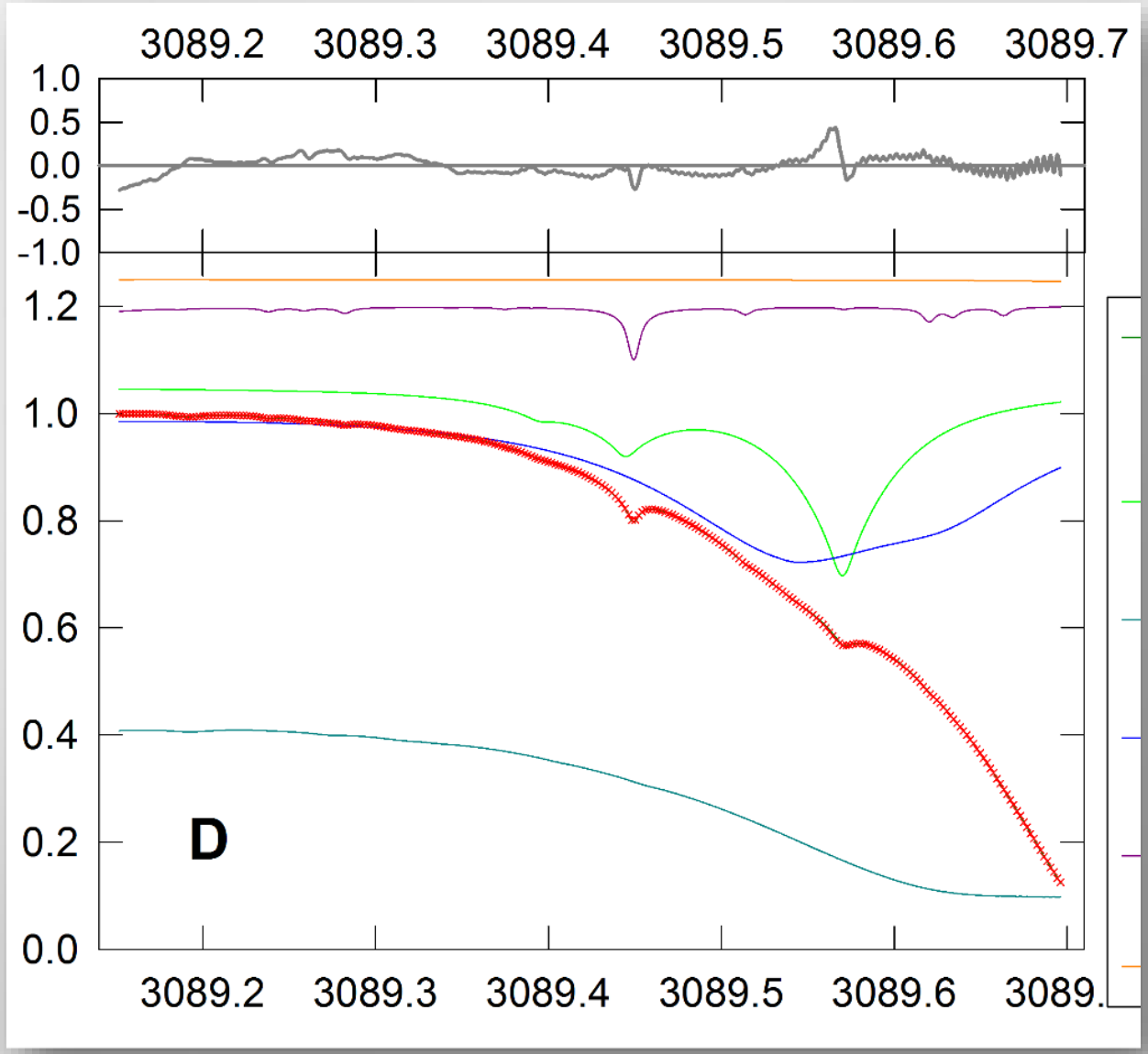
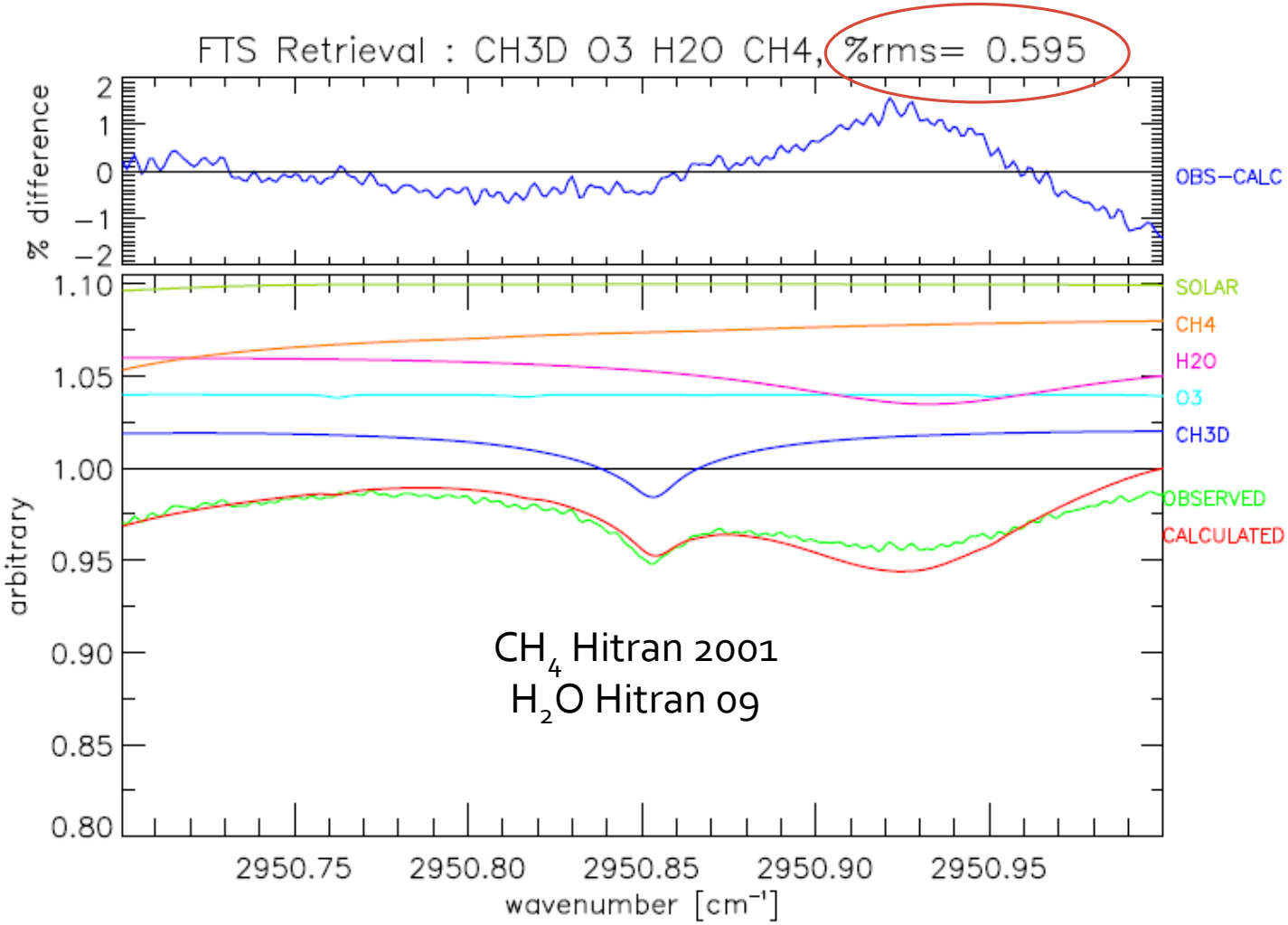


CH_3D Retrieval Strategy

- A priori from a WACCM simulation by Eric Buzan (Buzan et al., 2016)
  - Global simulation including both CH_3D and $^{13}CH_4$ from modified oxidation rate constants and boundary conditions from Röckmann et al., 2011
- No climatology available to build proper covariance matrices
  - Tikhonov regularization (similar to CH_4 strategy)
- Current spectroscopic linelists
  - CH_4 : Geoff Toon's atm linelist
  - H_2O : DLR's new linelist (thanks to Matthias Palm)
  - Still need to quantify the improvement from those linelists

Spectroscopy : example of Lauder (A)



Thank you Dan Smale !

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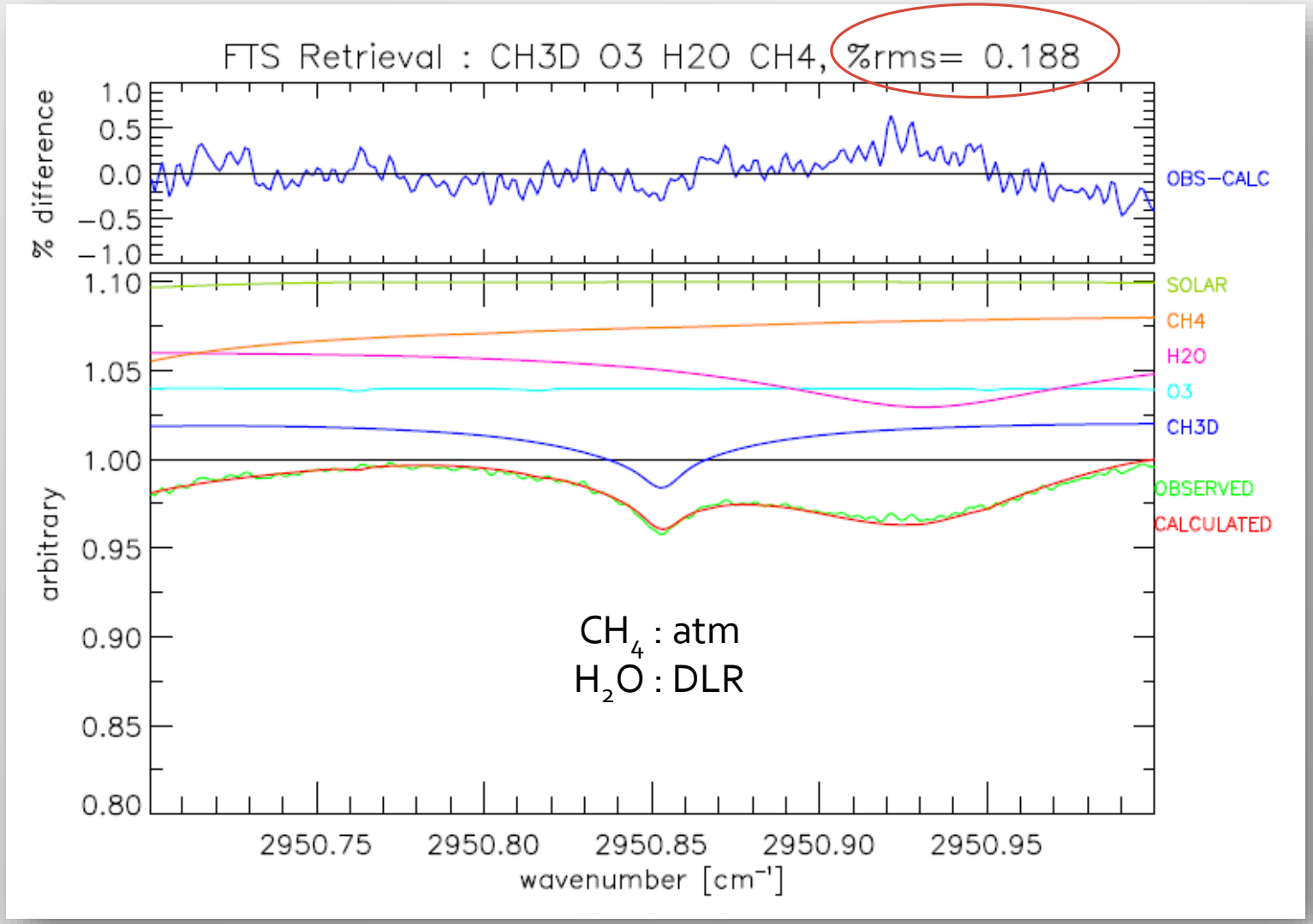

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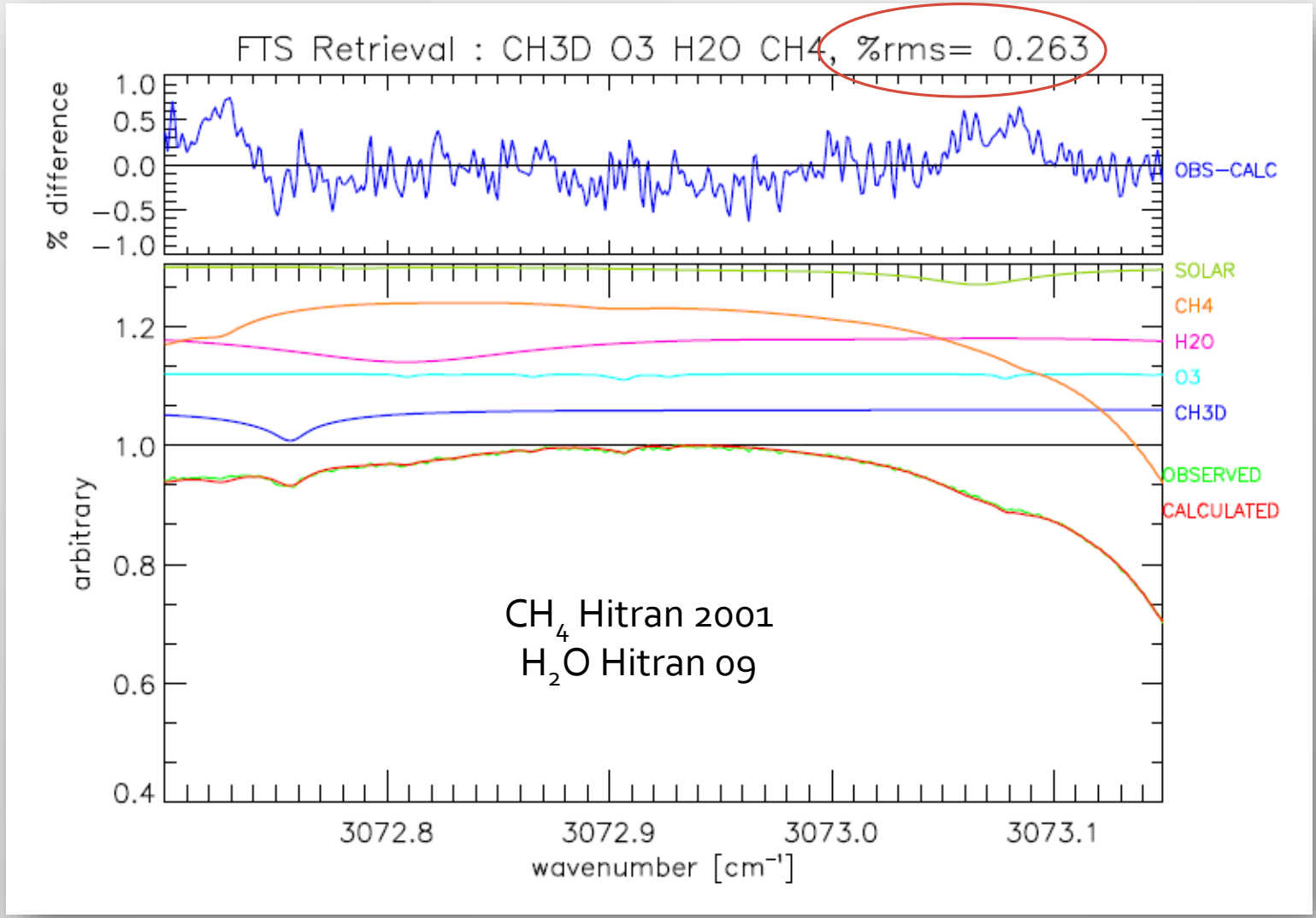
Spectroscopy : example of Lauder (C)



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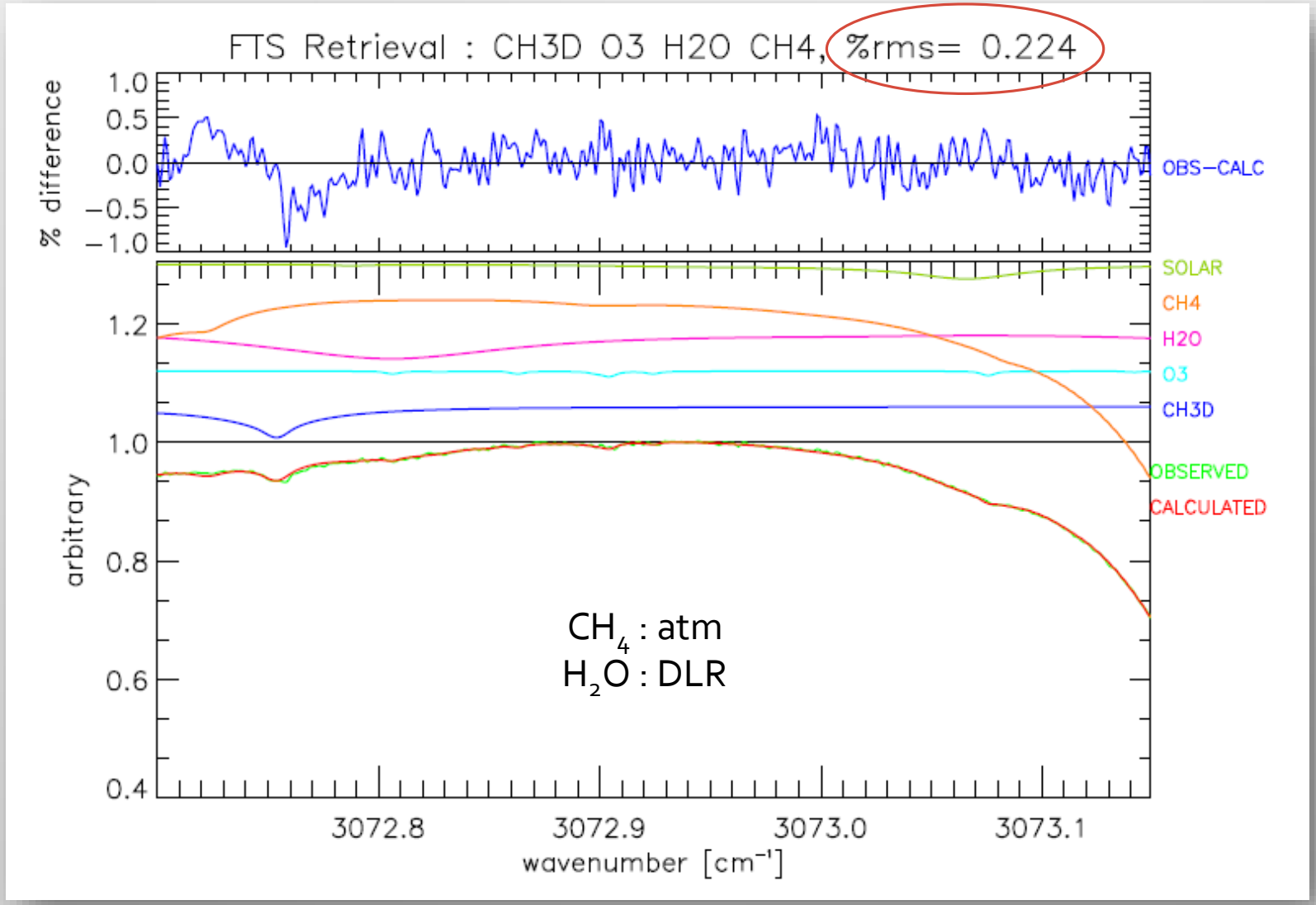
Spectroscopy : example of Lauder (C)



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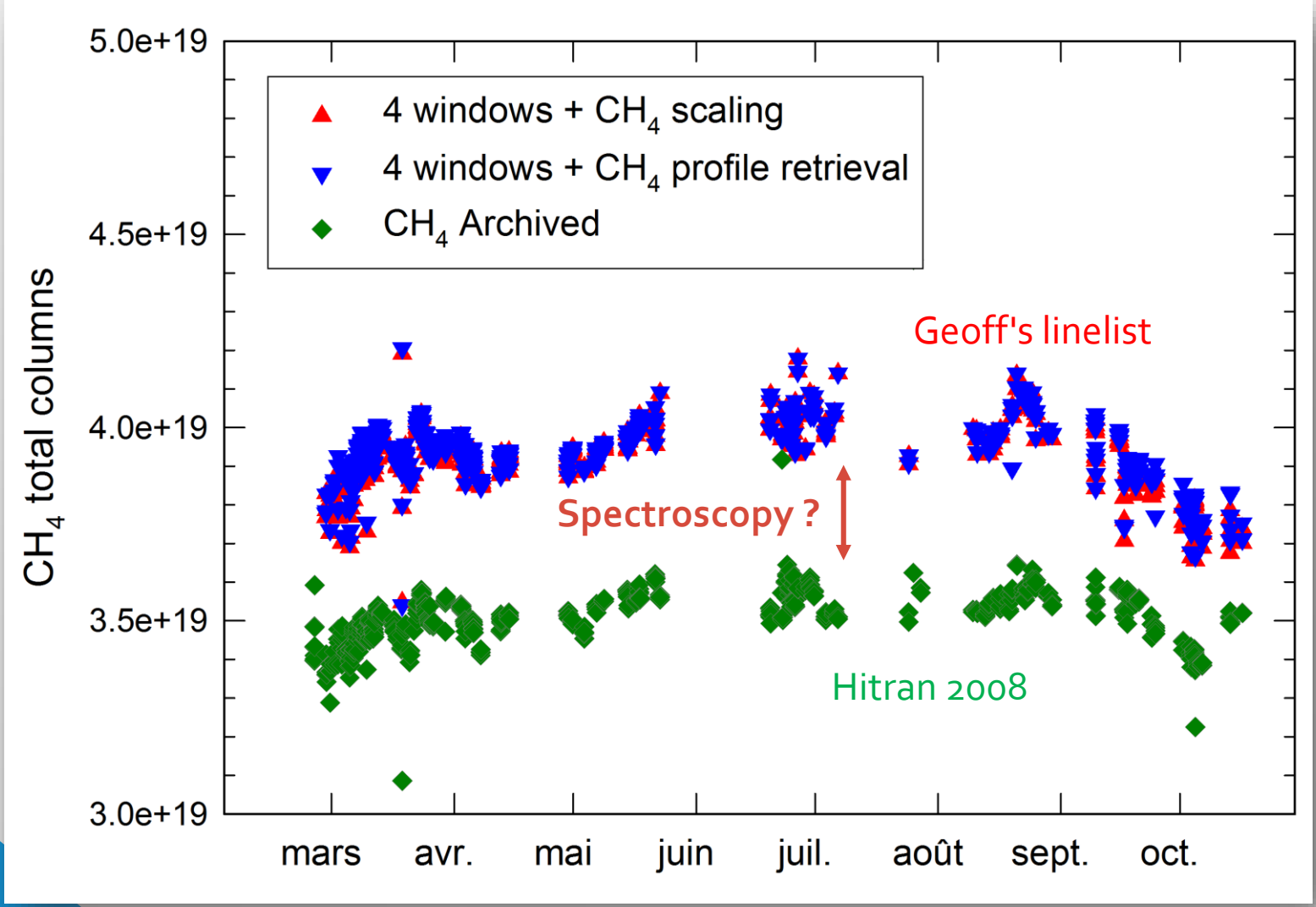
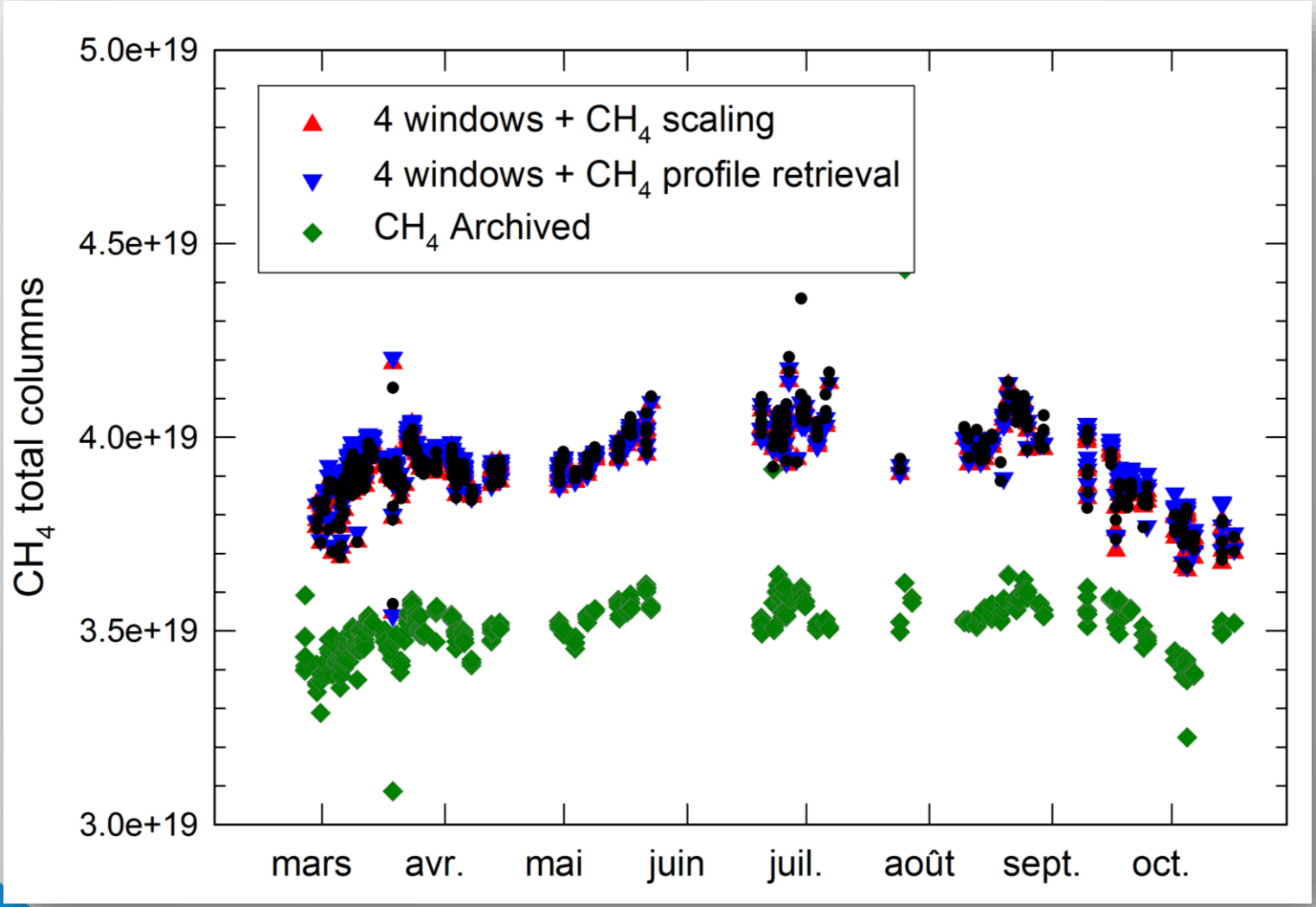
CH_4 approach

Need proper CH_4 for ‰ computation



CH_4 approach

What's causing this gap ?



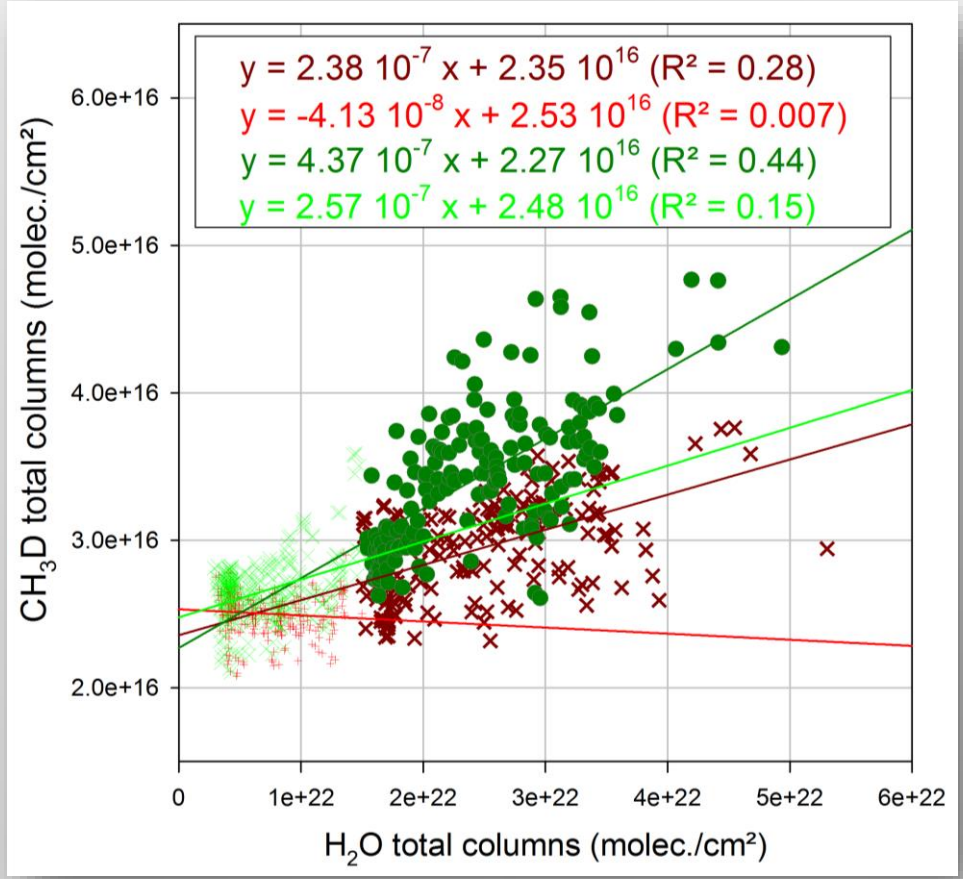
H_2O approach

Use of a water vapor specific window as 5th window ?

(2941.65 –2941.89 cm^{-1})

Similar to strategy for CH_4 at Jungfraujoch, Kiruna and Izaña

H_2O approach

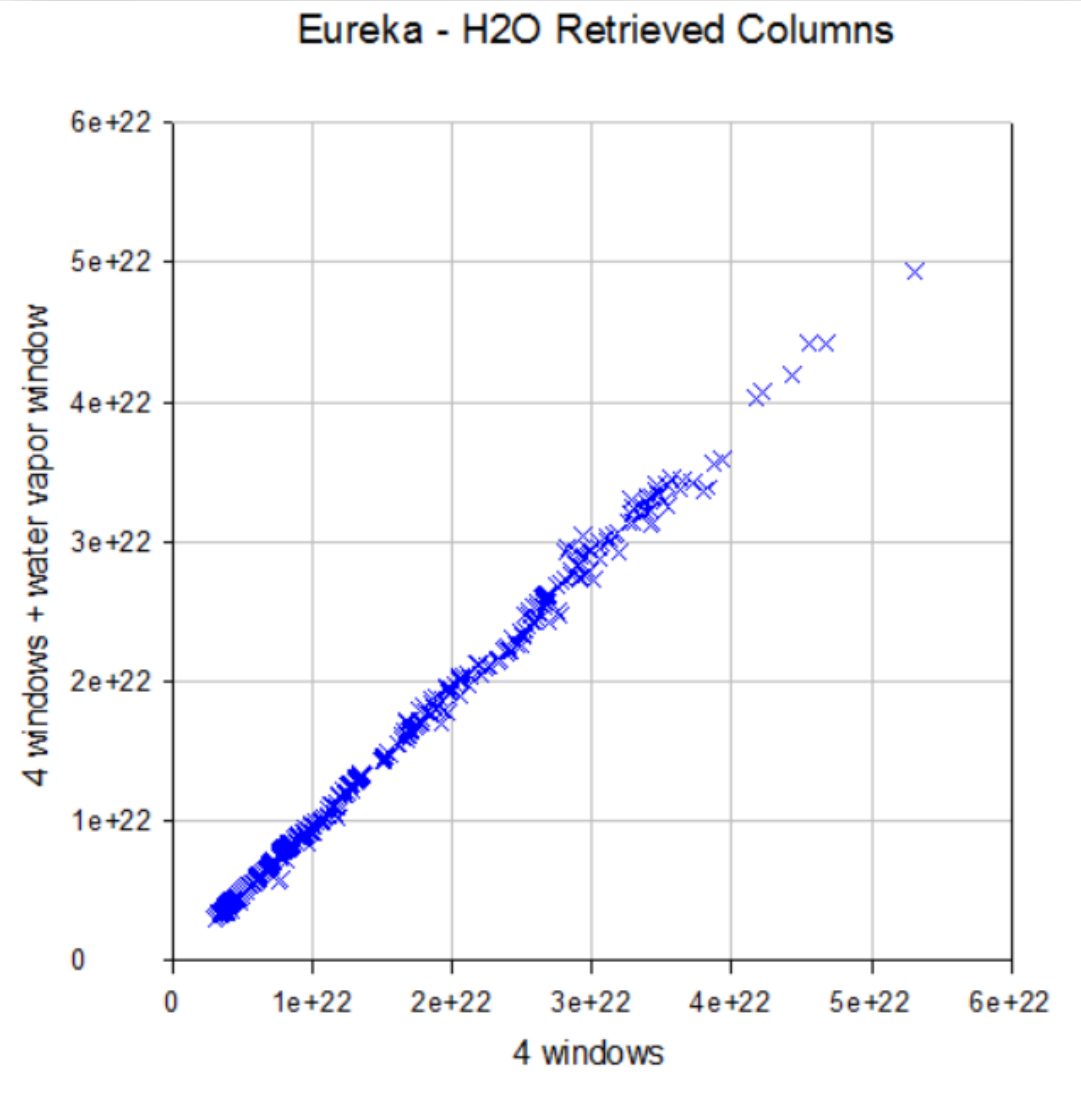


4 windows

+ water vapor window

4 windows

H_2O approach



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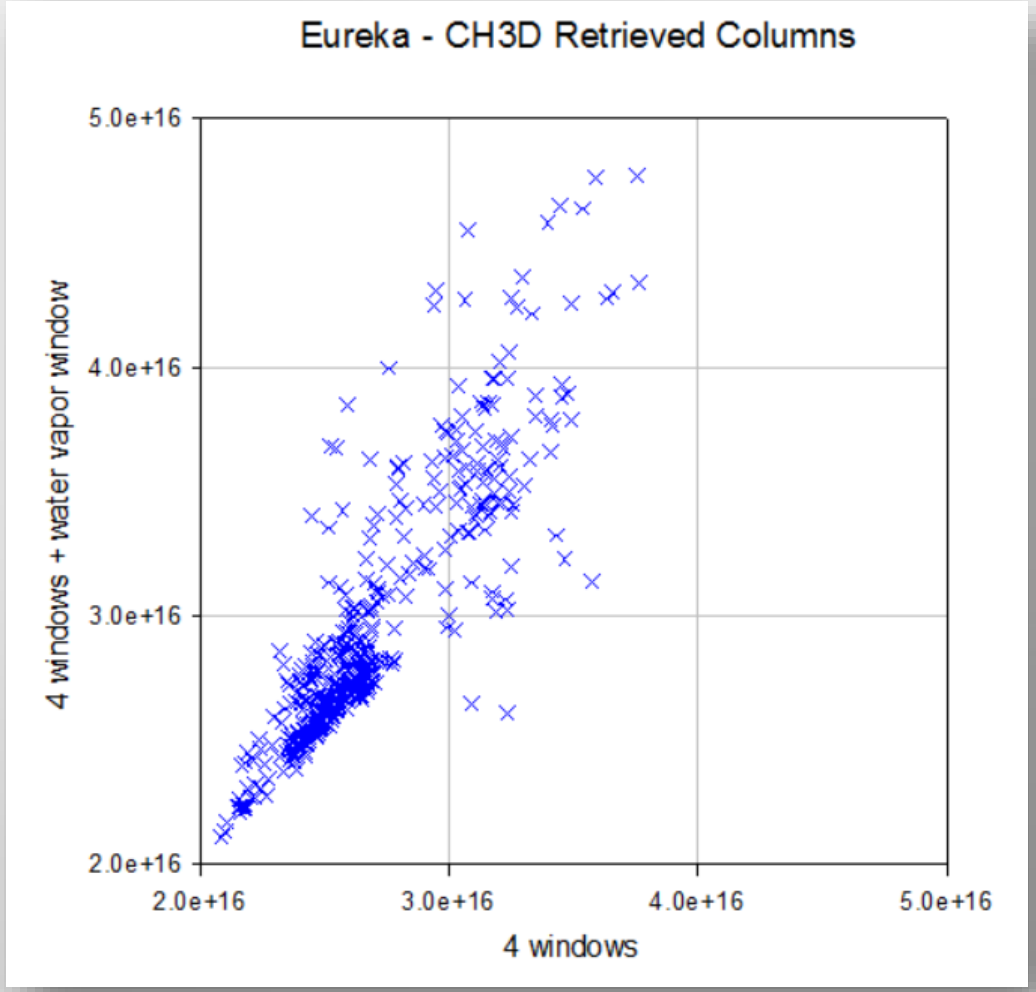
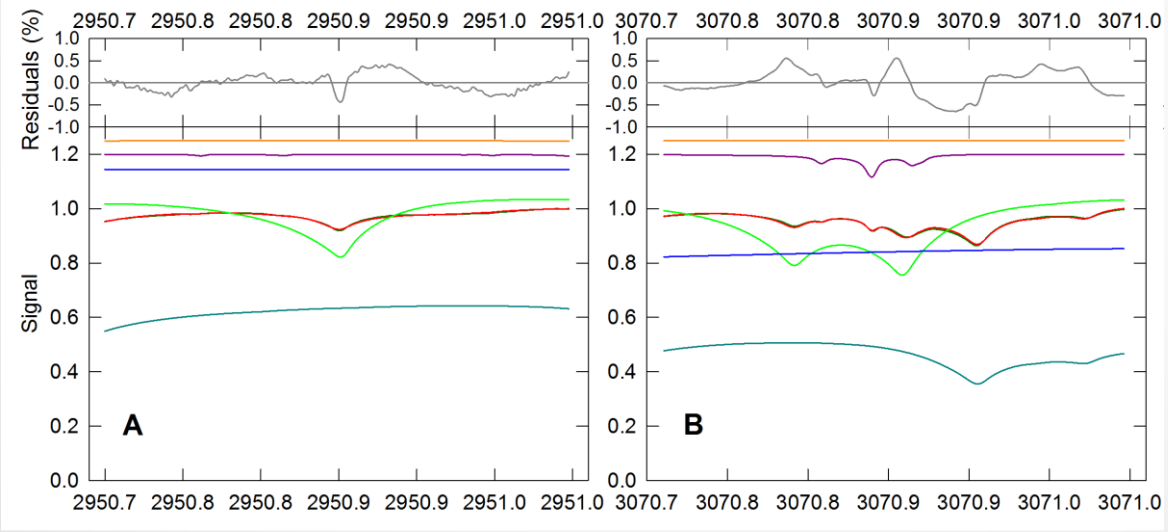
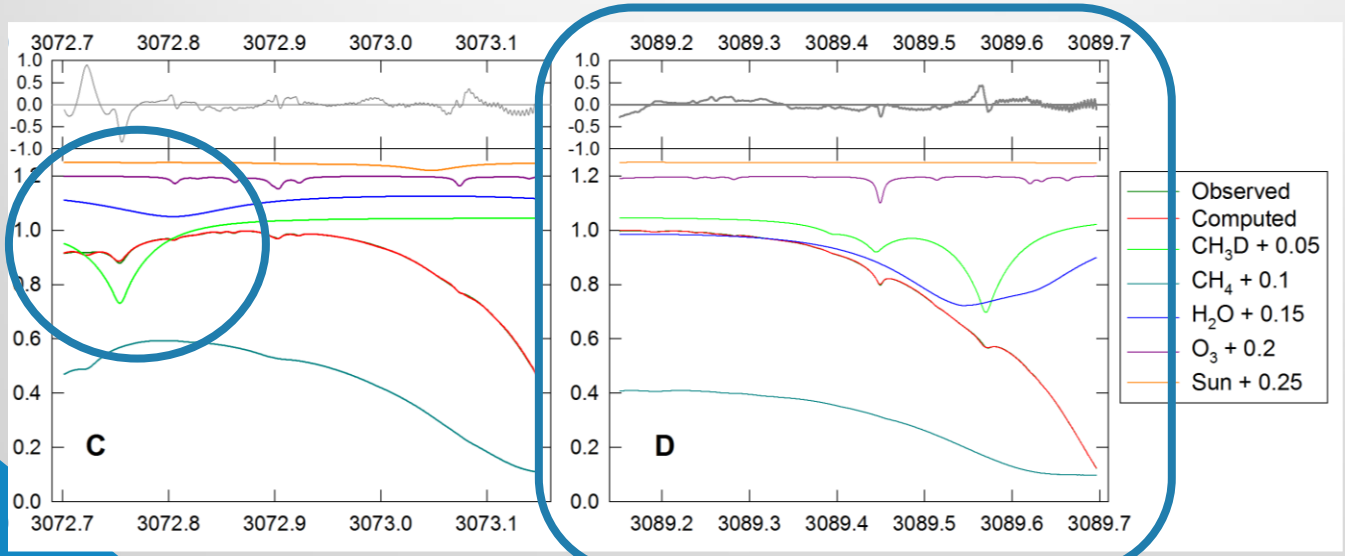


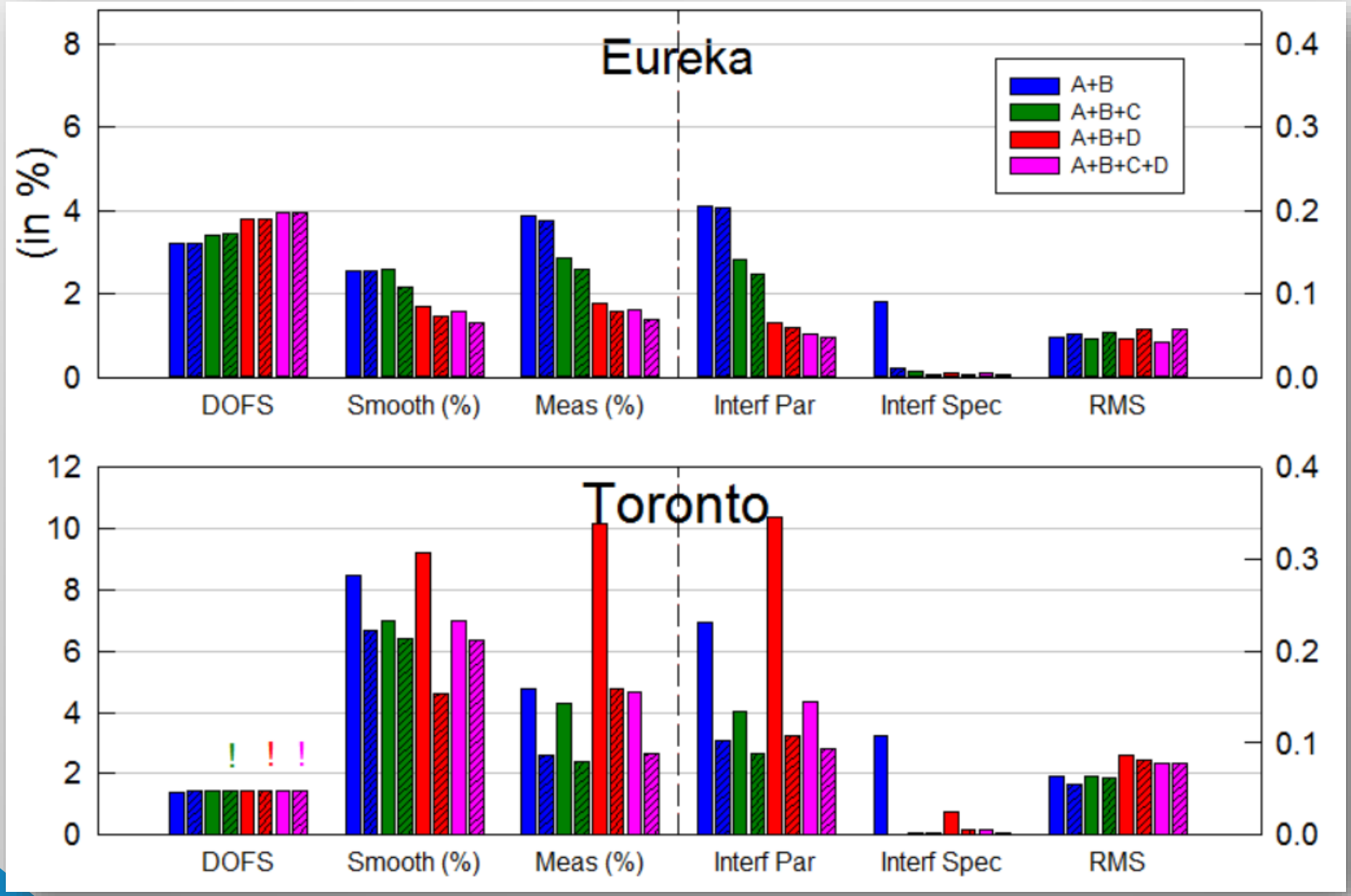
H2O approach

4 windows ?



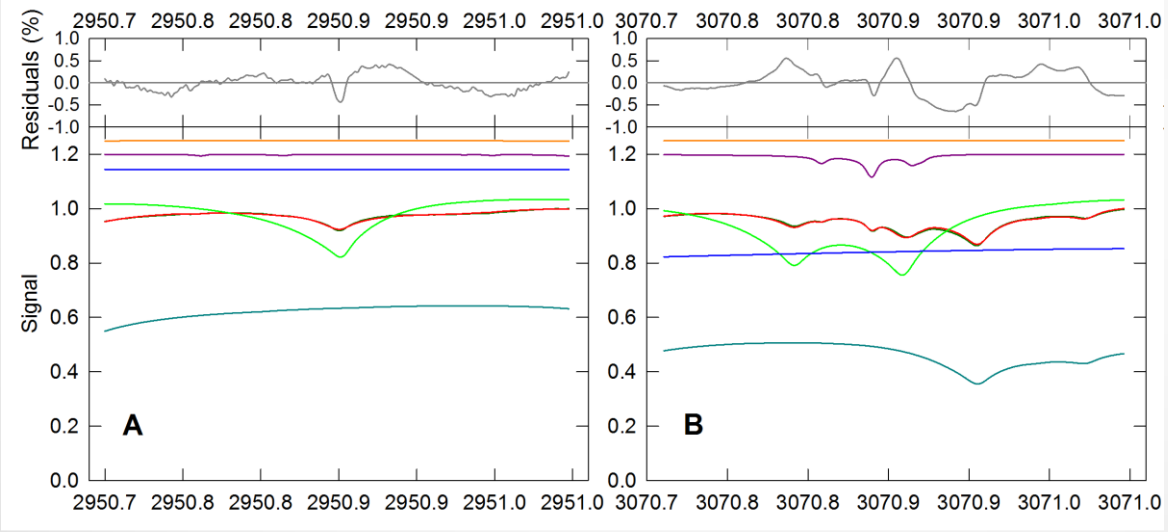


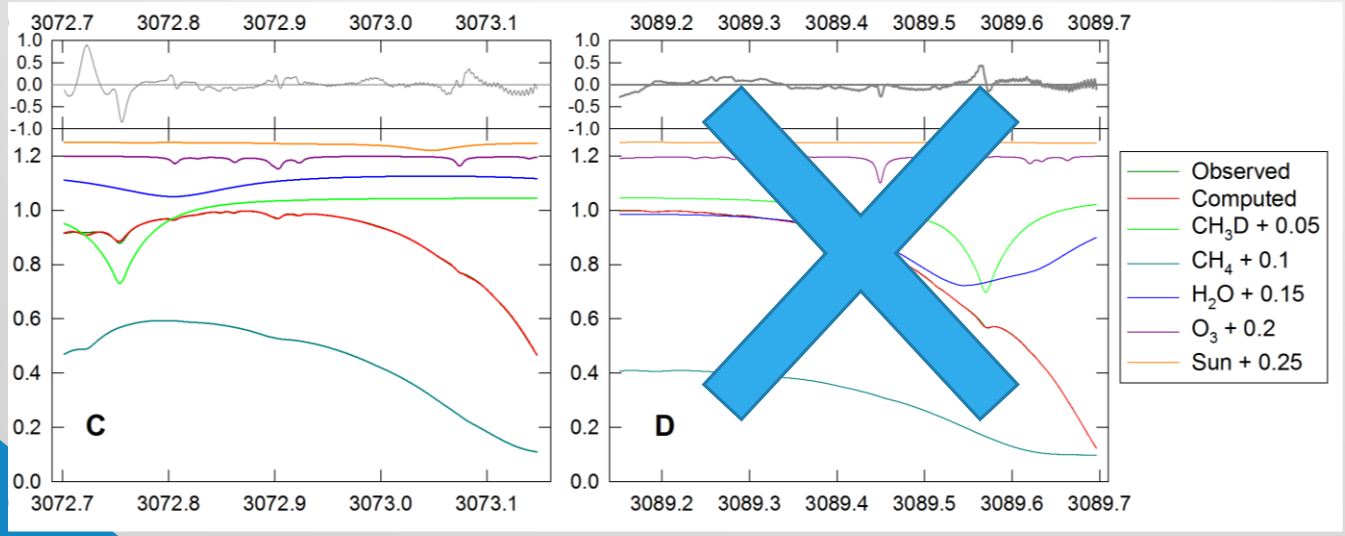
4 windows ? Error budget



H2O approach

4 windows ?





Retrieval strategy : Summary

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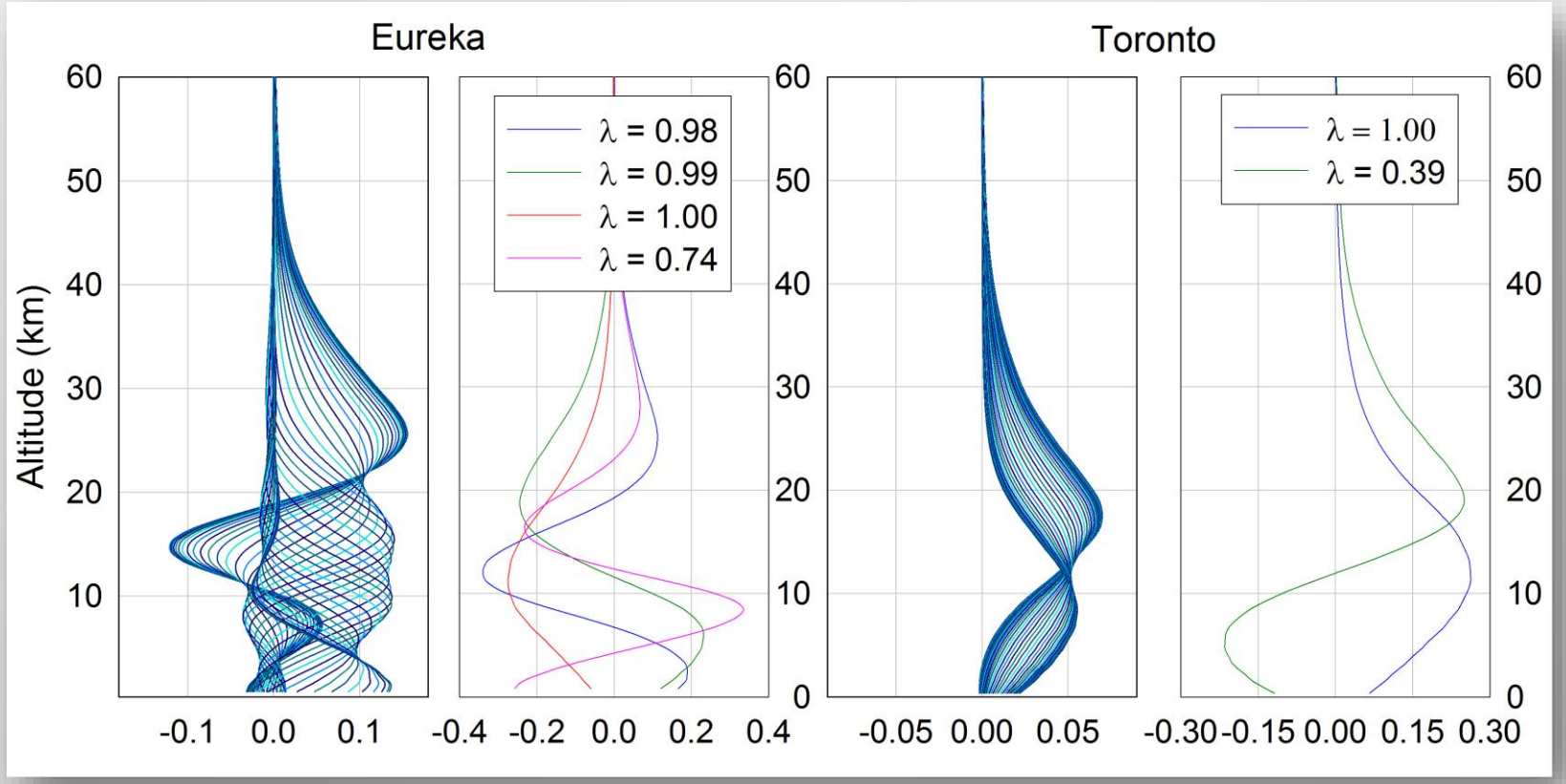
- Total of **205 tests** for Toronto and Eureka station
- Best strategy at the moment
  - 3 windows combination
  - Intefering species : $^{S}CH_4$, $^{R}O_3$, $^{S}H_2O$ + retrieval of solar lines
  - Geof Toon's CH_4 linelist and DLR's H_2O linelist
  - H_2O approach can be improved
  - CH_4 retrieval of profile doesn't bring better results
- Tests with a really dry and a really humid site would help making a decision…
- Encouraging results (considered it's a first)
- I only showed issues : e.g. good correlation between O_3 simultaneously retrieved and archived O_3 columns

Current "final" strategy

Eureka 2007 - 2015

Toronto 2002 - 2016

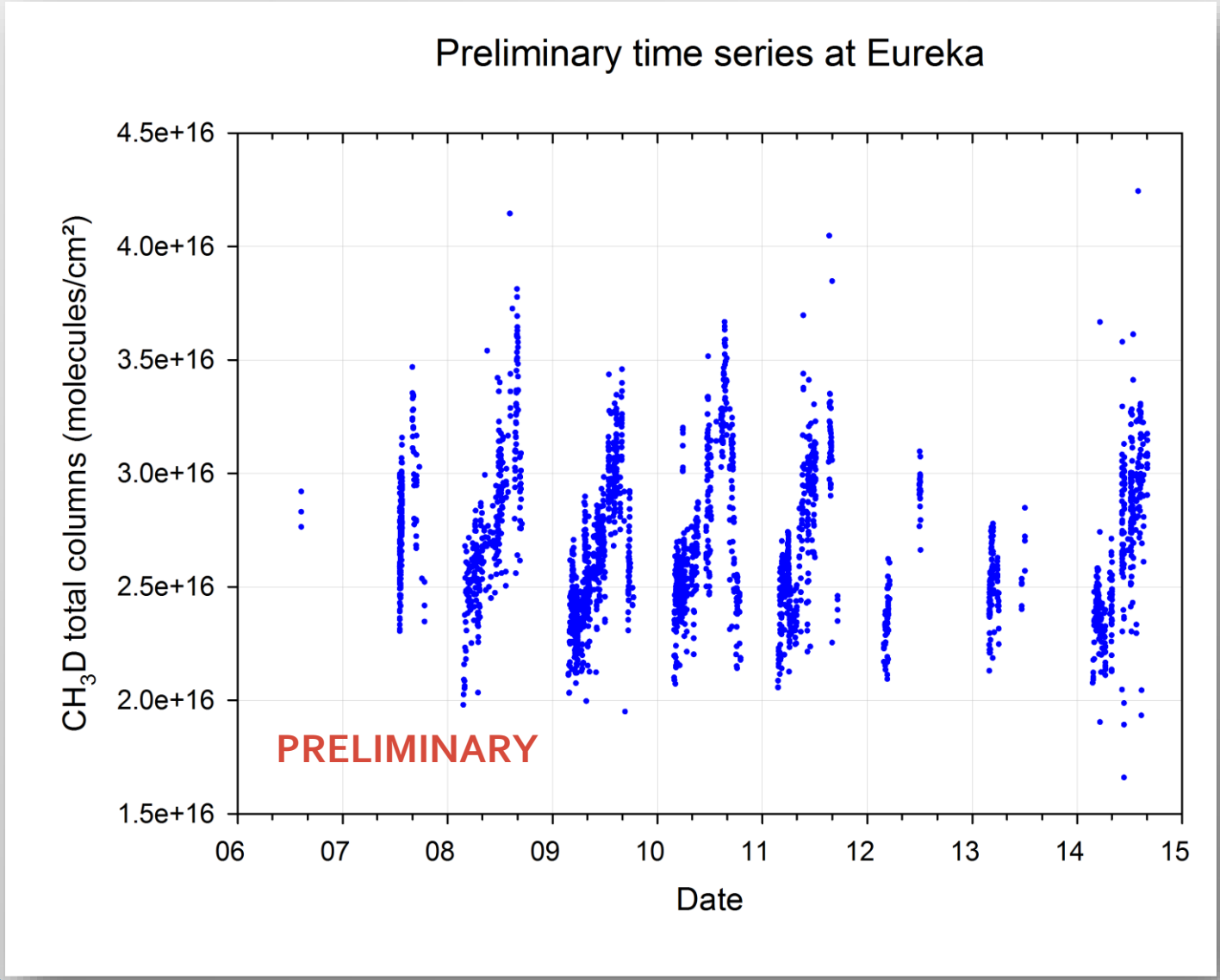
Information content & Error budget



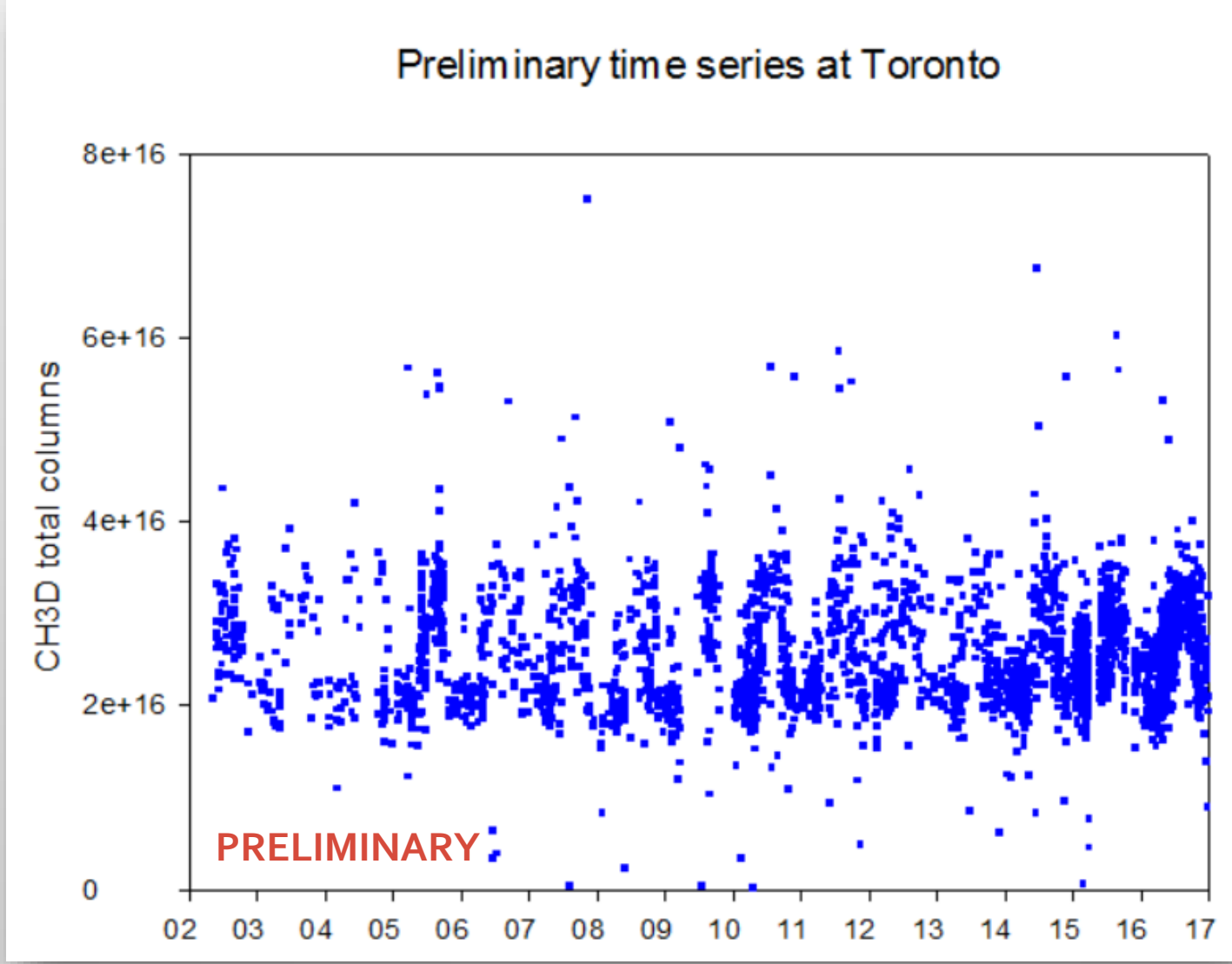
Mean averaging kernels and associated eigen vectors and eigen values.

DOFS = 3.81 (Eureka) and 1.55 (Toronto)

Time series @ Eureka



Time series @ Toronto



Conclusions

- CH_3D
- 3 windows strategy (SFIT-4)
- 1.5 < DOFS < 3.5
- A priori WACCMvEB
- Tikhonov regularization
- $^{S}CH_4$: Geoff's linelist
- $^{S}H_2O$: DLR linelist (thanks M. Palm)
- $^{R}O_3$ columns vs. Archived is good
- H_2O approach ?
- CH_4 approach ?

- $^{13}CH_4$
- Starting soon…
- WACCM profile available
- 6 windows available

The infrared working group has valuable information to share in the context of the methane increase question and needs to take part in this discussion.

Thank you for your attention.

If you have any suggestions or wish to contribute, come talk to me !

SFIT4 Control files and Tikhonov matrices (and document to determine α consistently) can be available as soon as tomorrow.

WACCM a priori profile can be available in ~ 1 week.
(cfr. Eric Buzan)

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Why is CH_4 increasing ?

- Large discrepancies between bottom-up and top-down estimates.
- From C2H6 measurements : oil and gas sector (Franco et al., 2016; Hausmann, et al. 2016; Helmig et al., 2016)
- From GEOS-Chem tagged simulation : oil and gas, coal and livestock
- From $^{13}CH_4$ in situ measurements : isotopic signature (~60‰) suggest a renewed rise due to biogenic sources (wetlands and agriculture)
- New studies question the importance of the role of OH : McNorton et al., 2016, Rigby et al., 2017, Turner et al., 2017