

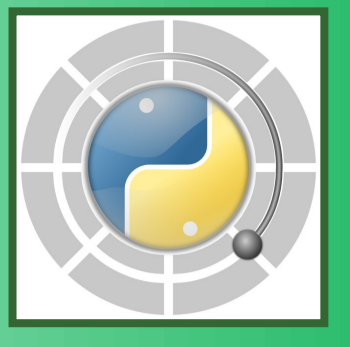
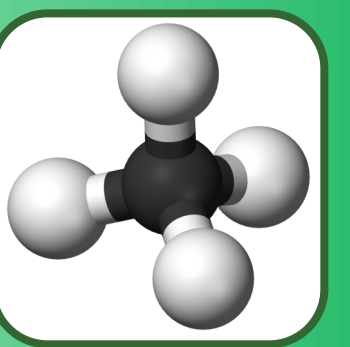
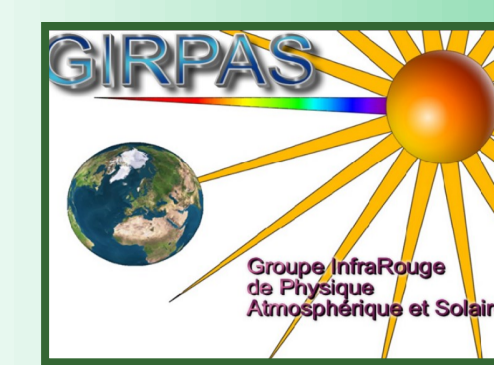
Seeking for causes of recent methane increase: comparison between GEOS-Chem tagged simulations and FTIR column measurements above Jungfraujoch.

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GEOS-CHEM MODEL : TAGGED SIMULATIONS

- GEOS-Chem CTM v9-02
 - A resolution in 2° latitude and 2.5° longitude and 47 vertical levels
 - Time step of 3 hours
 - 11 tracers listed in *Table 1* representing the contribution of each emission source and sink in the total CH₄.
 - GEOS5 - GFED3 - OH_v5-07-08 - EDGAR V4.2 (2004-2008)
 - Simulation from 2005 to 2012
- A detailed description of the methane simulation is on K. Wecht et al., 2014

- TABLE 1 - TRACERS -

Tracer	Source
CH4-tot	Total
CH4-ga	Gas and oil
CH4-co	Coal
CH4-ef	Livestock
CH4-wa	Waste
CH4-bf	Biofuels
CH4-ri	Rice fields
CH4-oa	Other anthropogenic emissions
CH4-bb	Biomass burning
CH4-wl	Wetlands
CH4-sa	Absorption by soils
CH4-on	Other natural emissions

METHANE RECENT INCREASE ABOVE JUNGFRAUJOCH (4x5 SIMULATION)

- *Figure 1* - Daily mean total column time series for the FTIR measurements (blue open circles) and the 4x5 GEOS-Chem simulation (filled orange circles) for the 794 available coincidences over the 2005-2011 time period. Lines show linear component of trend calculation. The upper frame displays fractional differences $[(FTIR - GC)/((FTIR+GC)/2)]$, in %.

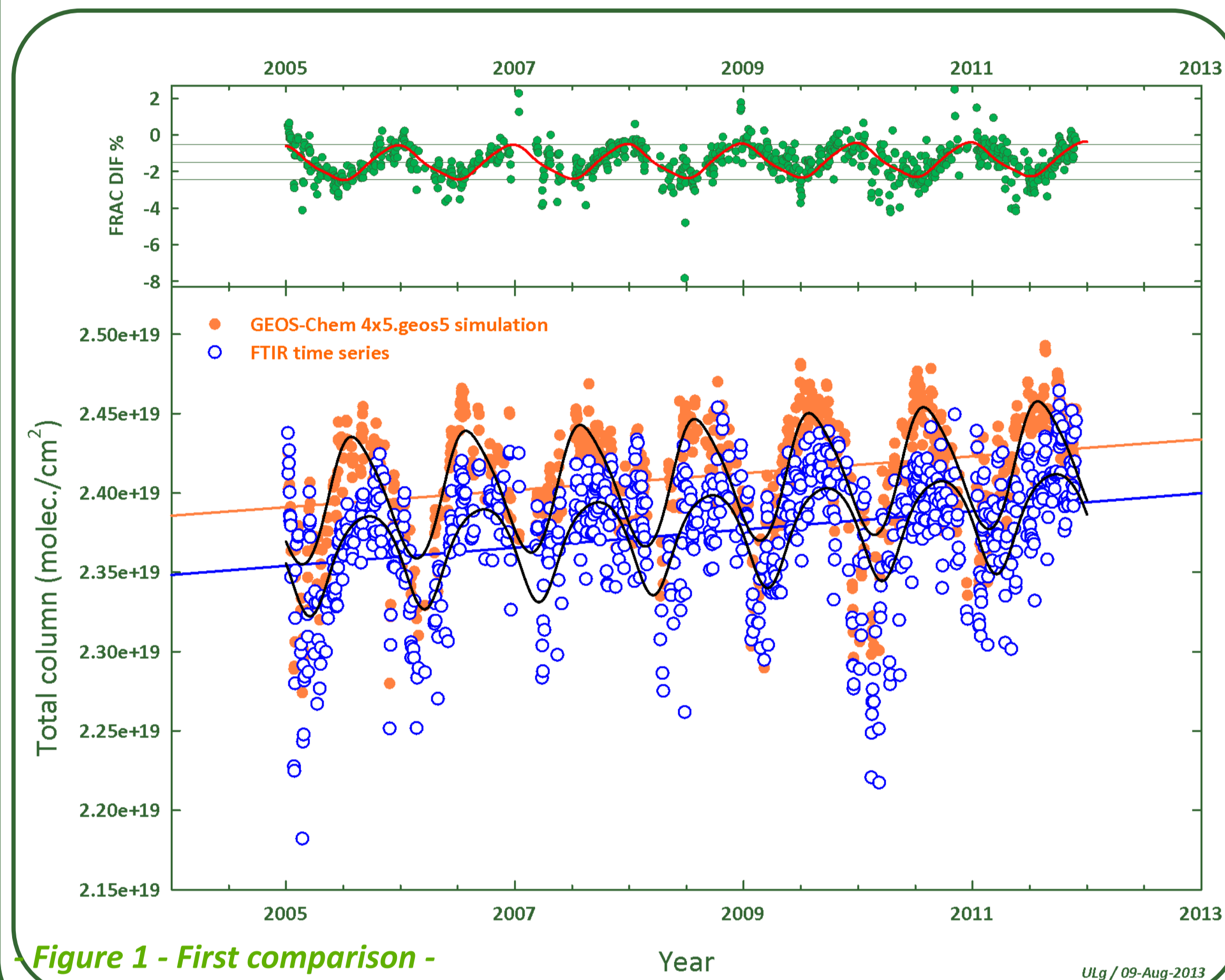


Figure 1 - First comparison -

* Bias between the two data sets : mean fractional difference of $-1.49 \pm 0.96\%$ (1 σ) with a clear seasonal signal, with a peak-to-peak amplitude of 1.89%. Both time series show a slight but significant increase of methane over the 6-year time interval. Trends have been evaluated and relative column changes are listed in *Table 2*. Both trend values are significant and in agreement within the 2 σ uncertainty level.

* The seasonal bias should not be observed anymore with the 2x2.5 simulation as issues on the wetland emissions have been dealt with. Since wetland emissions is responsible for ~20% of atmospheric methane, the recent changes have the largest impact on the magnitude and seasonality of high-latitude methane emissions.

* A slight imbalance between the sources and sinks relative to the world leading to a potential global mean bias of up to ~30ppbv has been identified when compared to observations and should be taken into account. (Alex Turner, personal communication).

- Table 2 -

		2.5 percentile	Trend	97.5 percentile
Column Trend (molec./cm ²)/yr	FTIR	3.29E+16	4.43E+16	5.55E+16
	GEOS-Chem (4x5)	2.88E+16	3.72E+16	4.56E+16
Relative Change (%/yr, wrt 2005)	FTIR	0.14	0.19	0.24
	GEOS-Chem (4x5)	0.12	0.16	0.19

- *Table 2* - Column trend and relative changes for both FTIR and GEOS-Chem time series computed by a bootstrap resampling method.

METHANE RECENT INCREASE ABOVE JUNGFRAUJOCH (46.5°N, 8.0°E, 3580M)

Inversion Strategy

- 6 windows approach described by Sepúlveda et al., 2012
- > 4 « methane dedicated »
- > 2 auxiliary for the joint retrieval of H₂O and HDO profiles
- 7 species CH₄, H₂O, HDO, CO₂, O₃, NO₂, HCl
- Linelist : hitran08 - updated by F. Hase (Personal Communication, 2013)
- VMR a priori : WACCM v.6
- H₂O and HDO profile scaling as pre-fit
- On about 5600 spectra (1920 days) recorded with a Bruker IFS120-HR

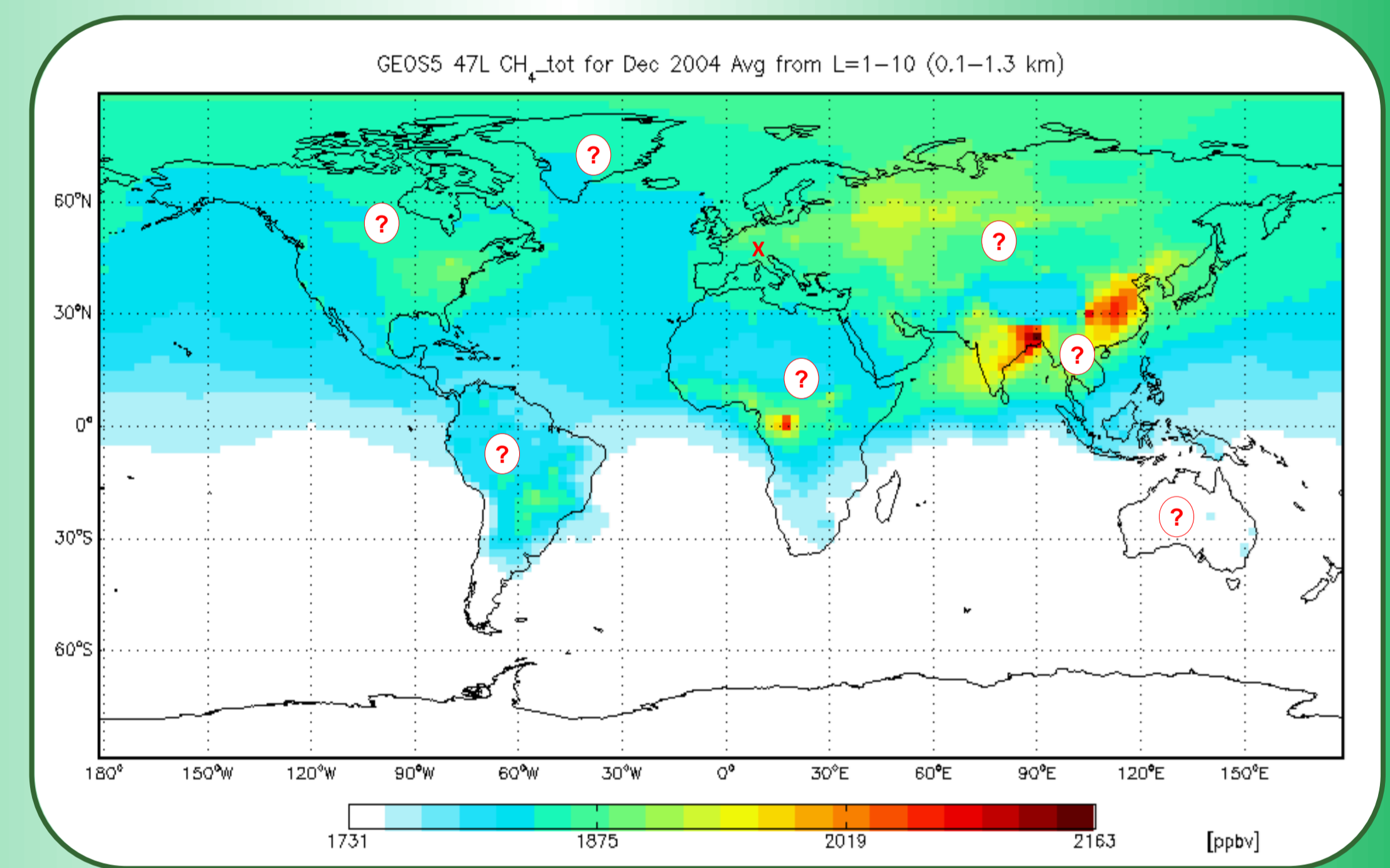
Methane Trends

Atmospheric CH₄ reached 260% of the pre-industrial level (~700 ppb) due to increased emissions from anthropogenic sources. Globally averaged CH₄ reached a new high of 1819 ± 1 ppb in 2012, an increase of 6 ppb with respect to the previous year (WMO, Greenhouse gas Bulletin N.9, 2013). CH₄ above Jungfraujoch increases at $0.53 \pm 0.19\%$ /year during the late 90s to stabilize and reach a non significant trend from 2000 to 2005. Since 2006, atmospheric methane has been continuously increasing with a rate of $0.19 \pm 0.05\%$ /year. The attribution of this increase to any CH₄ source is difficult since the current network is insufficient to characterize emissions by region and source process, emphasizing the need for source-tagged model simulations as it should provide us information on processes causing the increase of atmospheric methane since 2005/2006.

OUTLINE

The idea would be to do a multi-site study of the recent increase in atmospheric methane. The future work would evolve around three points :

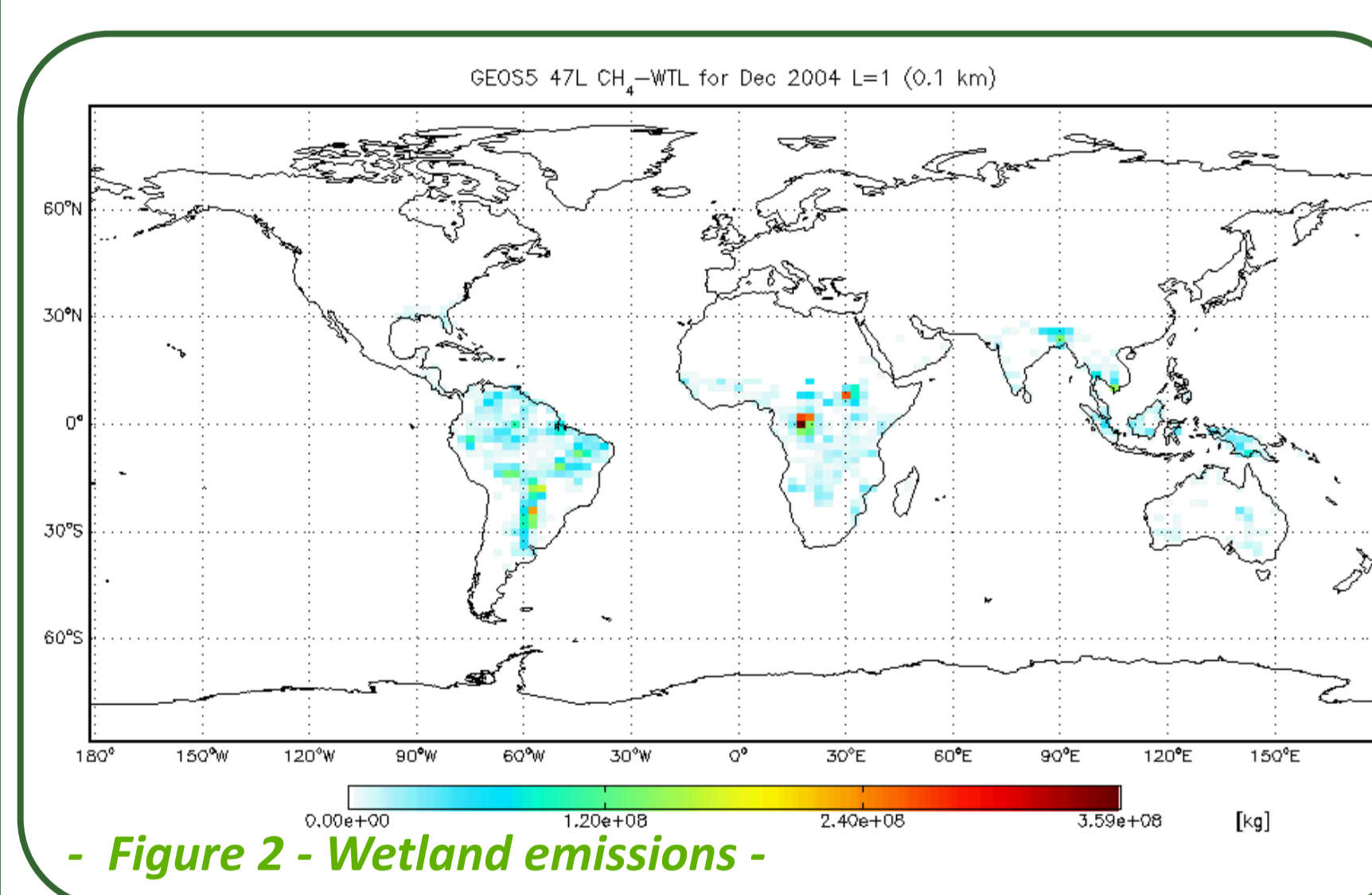
- * The 2x2.5 source-tagged simulation
- * Comparisons between GEOS-Chem model outputs and FTIR observations
- > Does the model satisfyingly appraise CH₄ total and partial columns above the station ?
- > Is the methane increase well reproduced ?
- * Study of trends for each CH₄ tracer



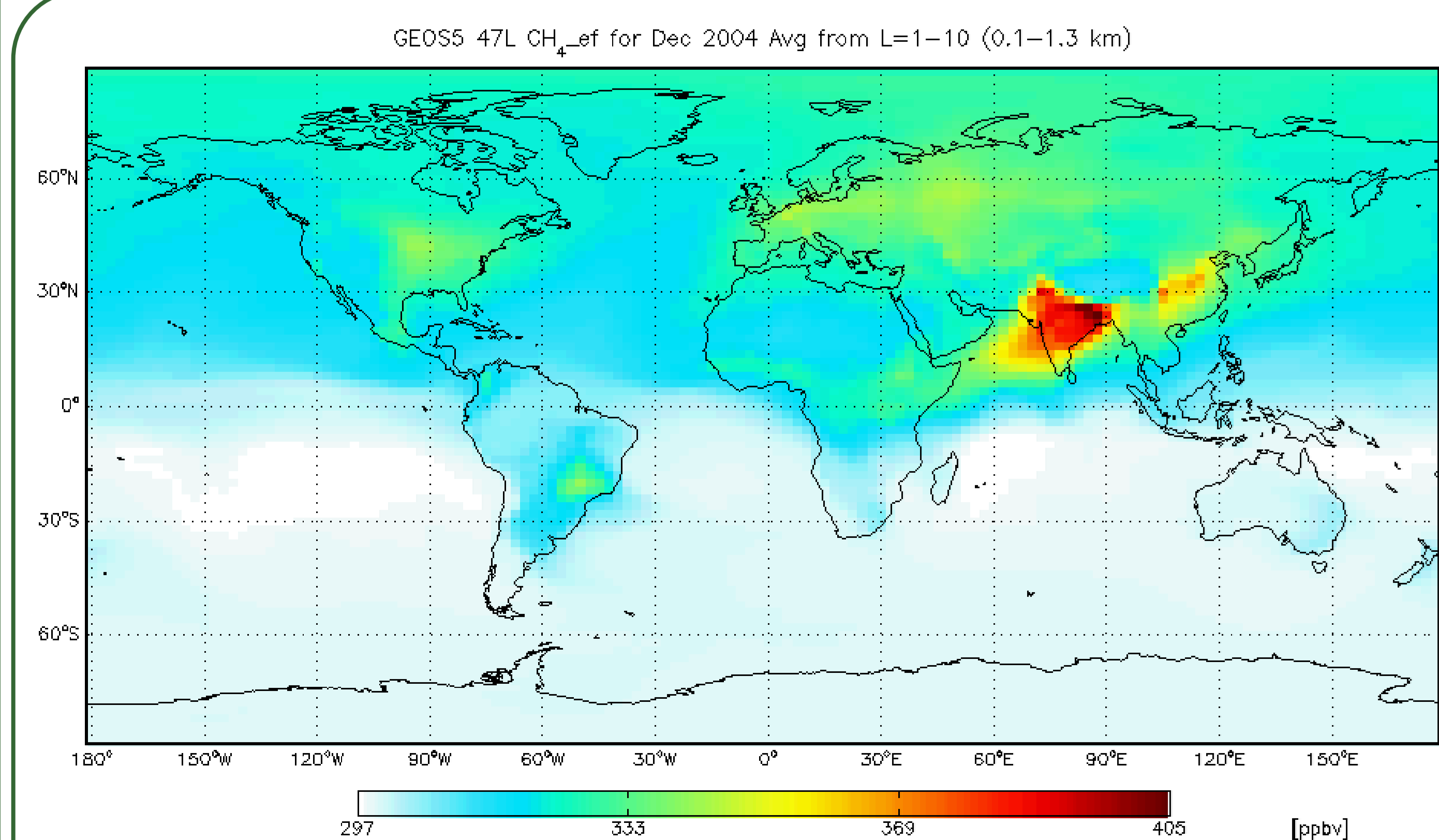
- *Figure 2* - Global distribution of methane emissions (in kg) from wetlands in December 2004 (spin Run) from the GEOS-Chem 2x2.5 simulation.

Retrievals from FTIR solar observations are defined on a fixed vertical grid while GEOS-Chem uses vertical grids (GEOS5) defined by hybrid coordinates and completed by external data (i.e., elevation of the base levels of the GEOS-Chem vertical grids). Therefore, in order to compare GEOS-Chem outputs and methane columns retrieved from FTIR observations, a 3D regridding is necessary.

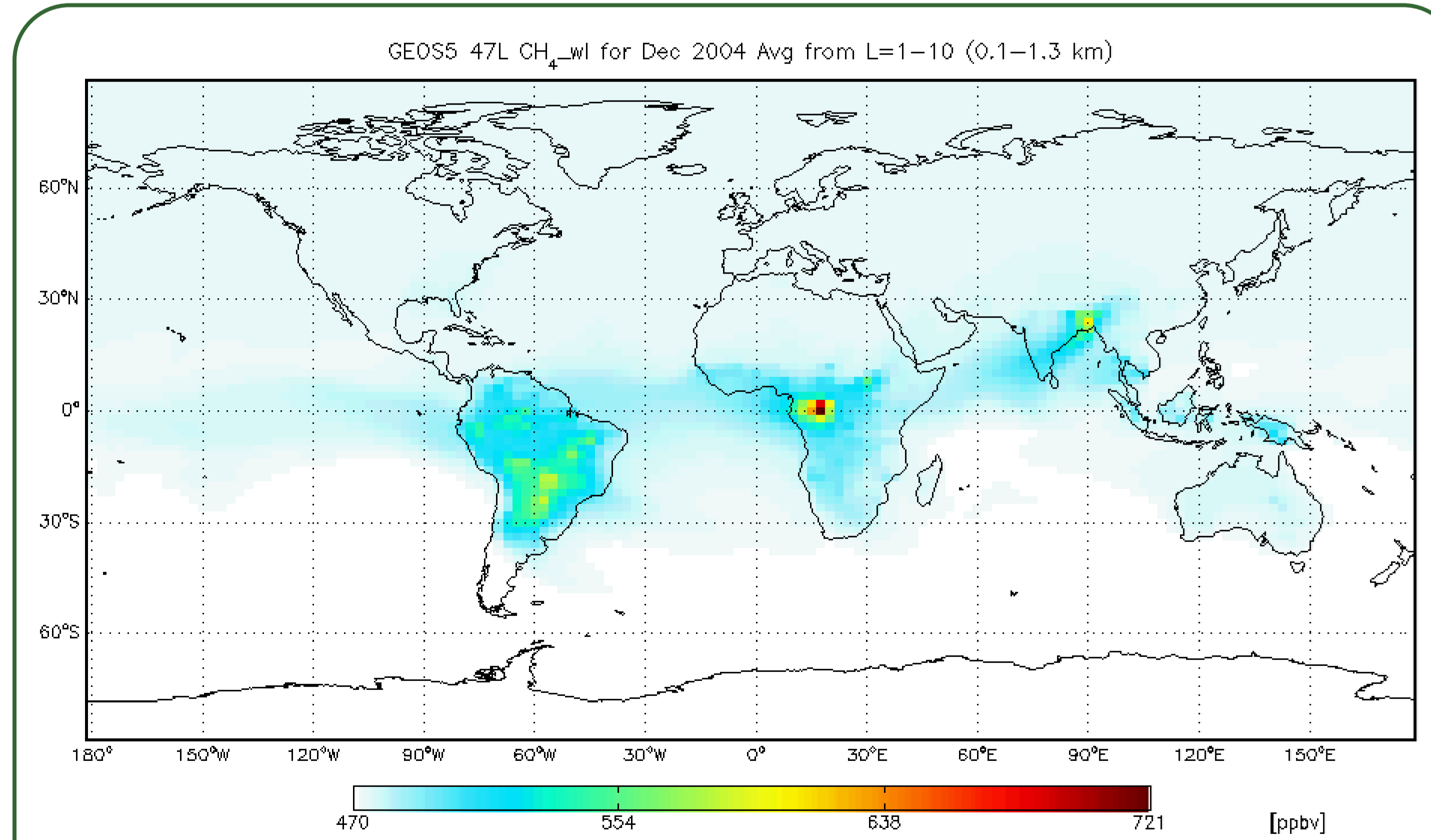
GEOS-CHEM TAGGED SIMULATIONS (2x2.5 SIMULATION)



- *Figure 2* - Wetland emissions -



- *Figure 4* - Livestock tracer -



- *Figure 3* - Wetland tracer -

GEOS-Chem outputs are converted into partial and total columns above a given station thanks to a nearest-neighbour interpolation scheme for the horizontal component and a conservative vertical regridding scheme so that the total mass of the tracer is preserved.

- *Figure 3* - Methane mixing ratio (in ppbv) global distribution issued from wetland emissions in December 2004 (spin Run) from the GEOS-Chem 2x2.5 simulation.

The 2x2.5 GEOS-Chem tagged simulation provides us a powerful tool to evaluate the impact of the methane emissions on its worldwide distribution in the atmosphere as well as the impact of transport and the many methane sources on the measured columns at a given station. We clearly see on *Figure 3* the impact of transport of wetland emissions (*Figure 2*) from Central Africa along the equator. The worshipping of Indian sacred cows is quite obvious when modeling the methane emissions due to livestock (see *Figure 4*).

- *Figure 4* - Methane mixing ratio (in ppbv) global distribution issued from livestock emissions in December 2004 (spin Run) from the GEOS-Chem 2x2.5 simulation.

Determining methane trends for each tracer will allow us to identify which parameter impact the recent increase in methane, e.g. whether one or a group of tracer predominantly increase through time or loss by soil absorption changes over time.

REFERENCES

- * Sepúlveda E. et al., Long-term validation of tropospheric column-averaged CH₄ mole fractions obtained by mid-infrared ground-based FTIR spectrometry, Atmospheric Measurement and Techniques, 5, 1425-1441, 2012, doi: 10.519/amt-5-1425-2012.
- * Wecht K. et al., Mapping of North American methane emissions with high spatial resolution by inversion of SCIAMACHY satellite data, submitted to the Journal of Geophysical Research, 2014, download manuscript on <http://goo.gl/ao6CAZ>.
- * WMO Greenhouse gas bulletin N.9, 2013.

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