The imprint of stratospheric transport on column-averaged methane (XCH_4)

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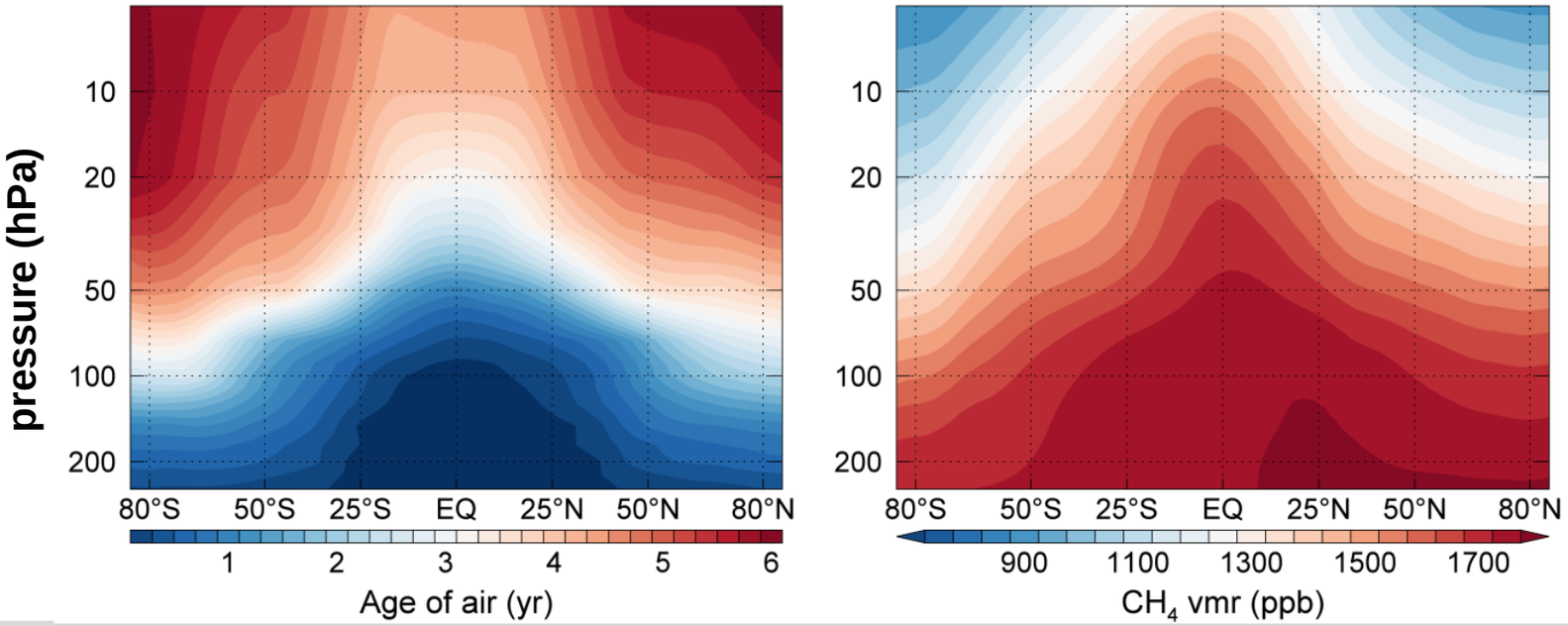
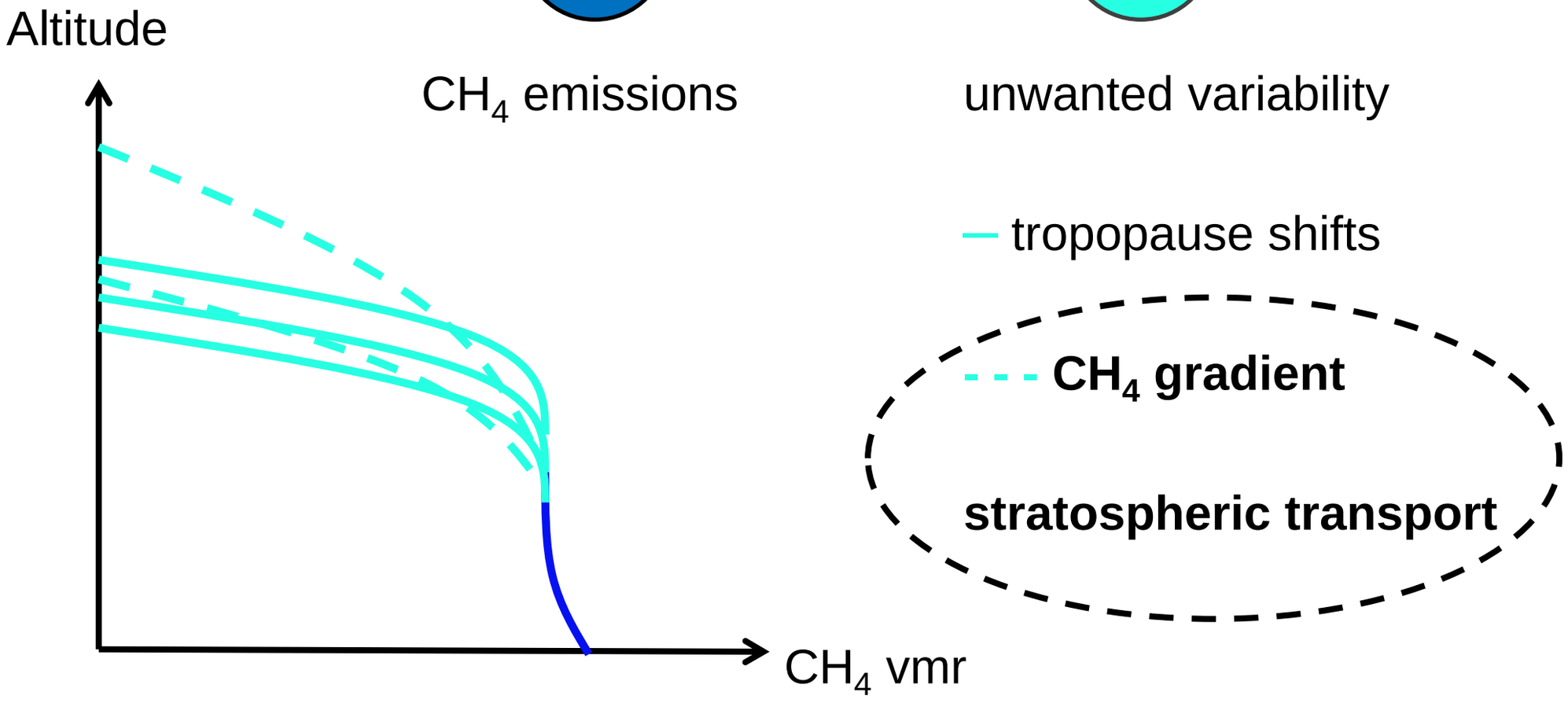


What controls XCH_4?

XCH_4 = tropospheric CH_4 + stratospheric CH_4

CH_4 emissions

unwanted variability



Stratospheric transport

Diagnostics for stratospheric transport:

Mean age of stratospheric air

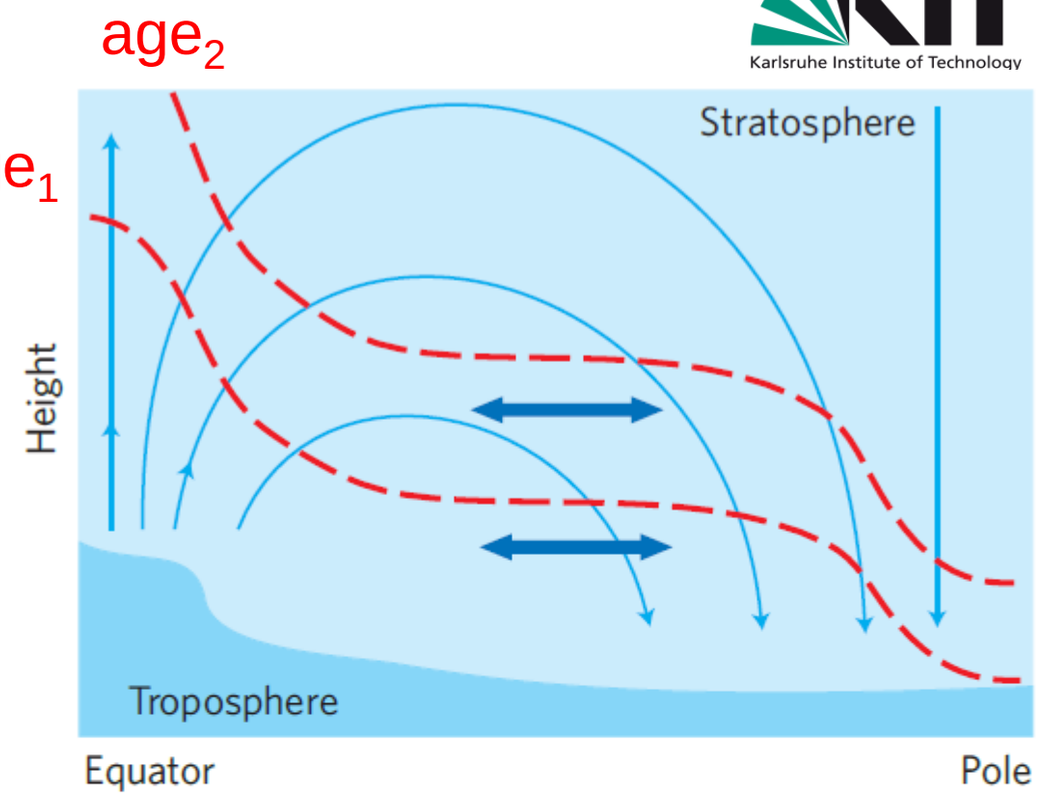
= Transport time

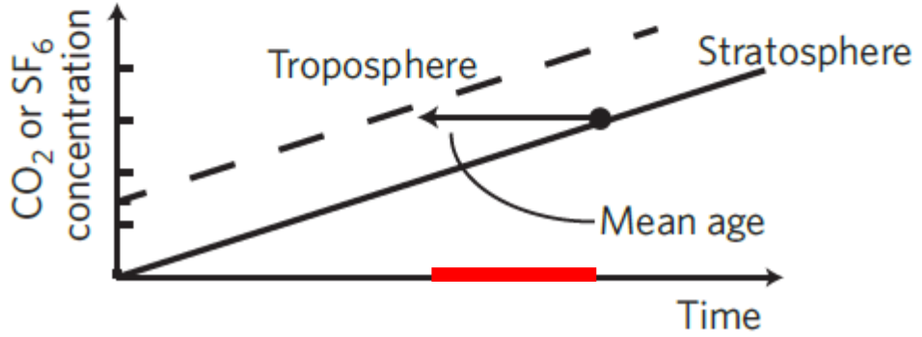
from tropical tropopause

to stratospheric location

mean age data:

observations vs. simulations





from Waugh et al. (2009)

Methodology

Intention: Describe sensitivity of XCH_4 to stratospheric transport

Approach: $CH_4(z) \rightarrow CH_4(age)$

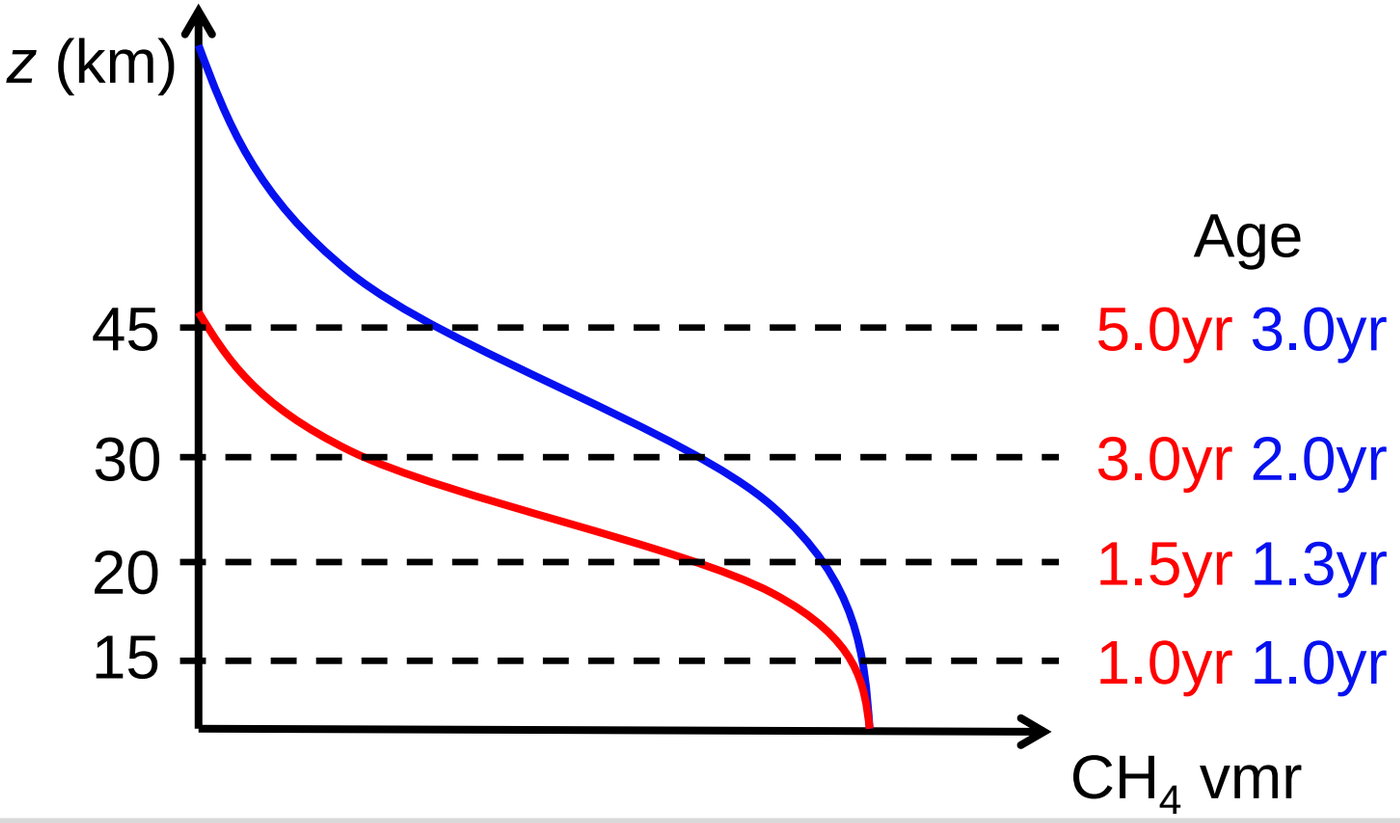
Data:	CH_4 and Age simulations:	ACTM (Patra et al. 2014)
	Age observations:	balloone-borne SF_6 profiles
	XCH_4 observations:	TCCON GGG2014

Stratospheric correction

Stratospheric model-transport error: modeled age ≠ observed age

Stratospheric correction: $CH_4(z) = CH_4(\text{tropopause}) + F(\text{observed age})$

→ Volk et al. (1997)

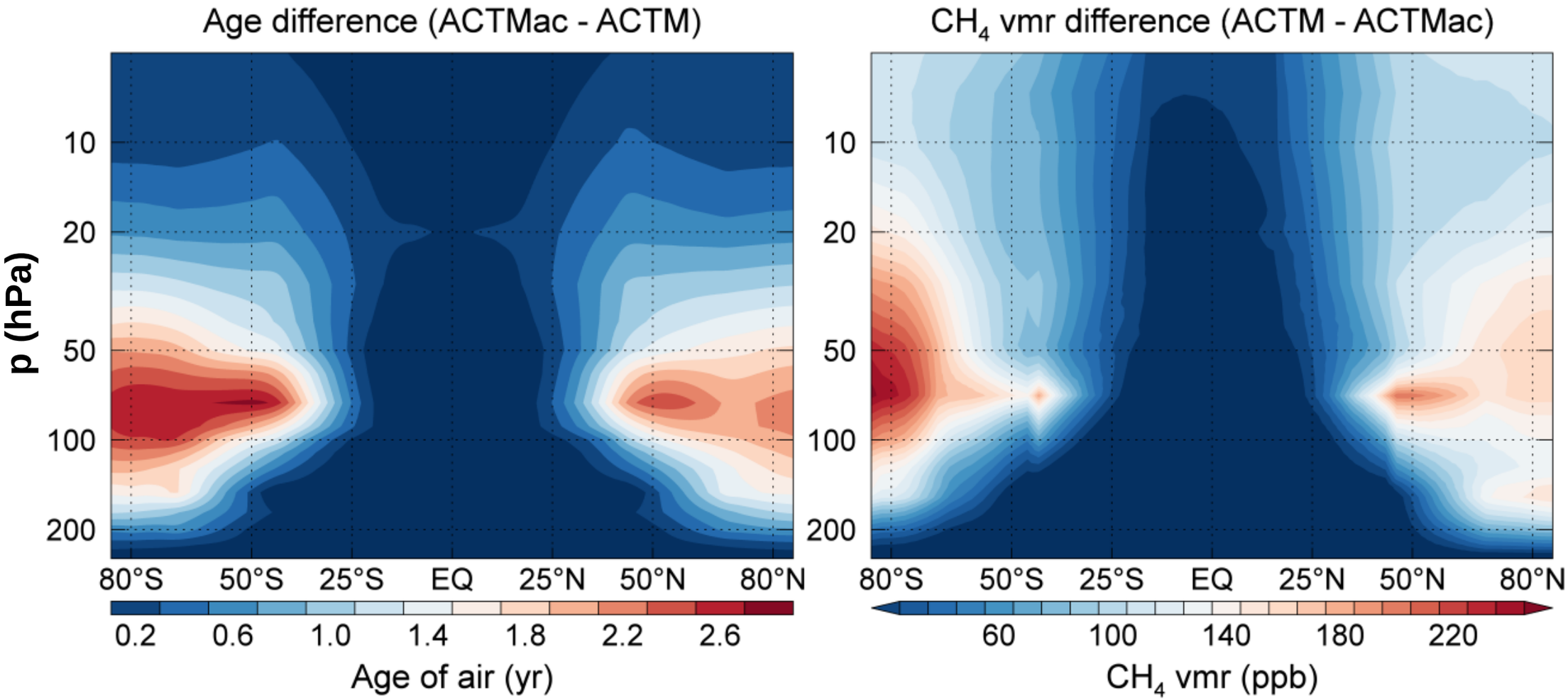


CH_4 simulations:

ACTM (original)
ACTMac (age-corrected)

Stratospheric zonal mean distributions

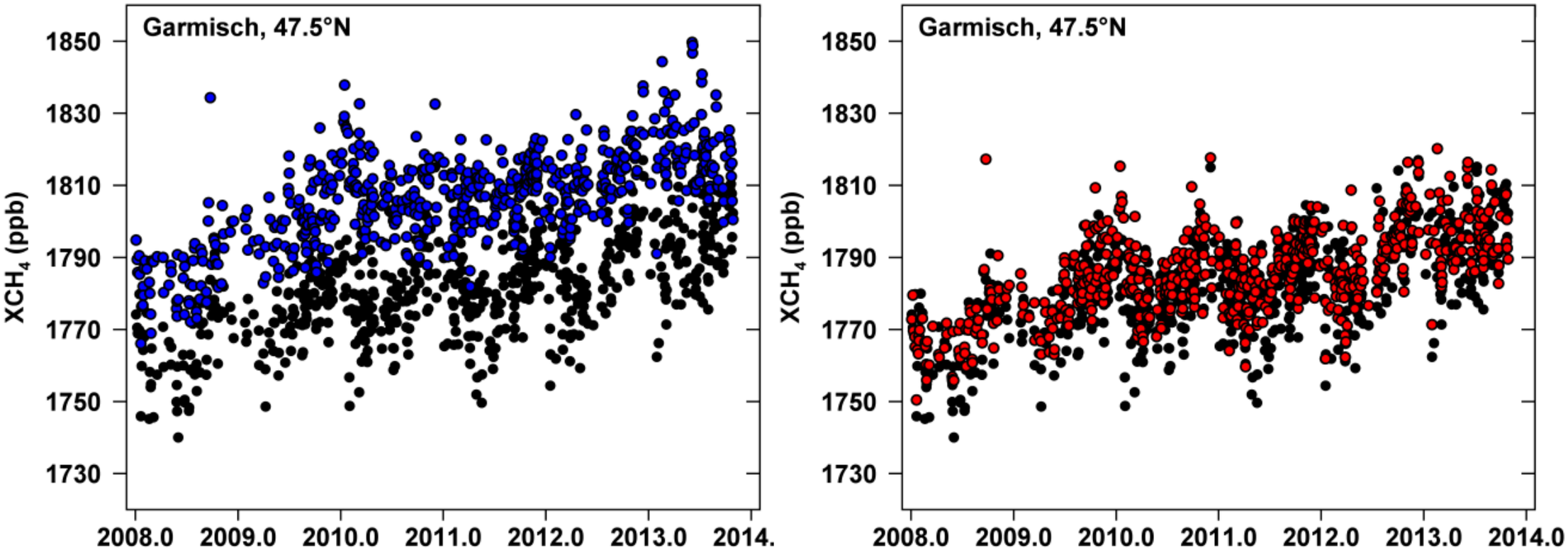
Age differences → CH_4 differences



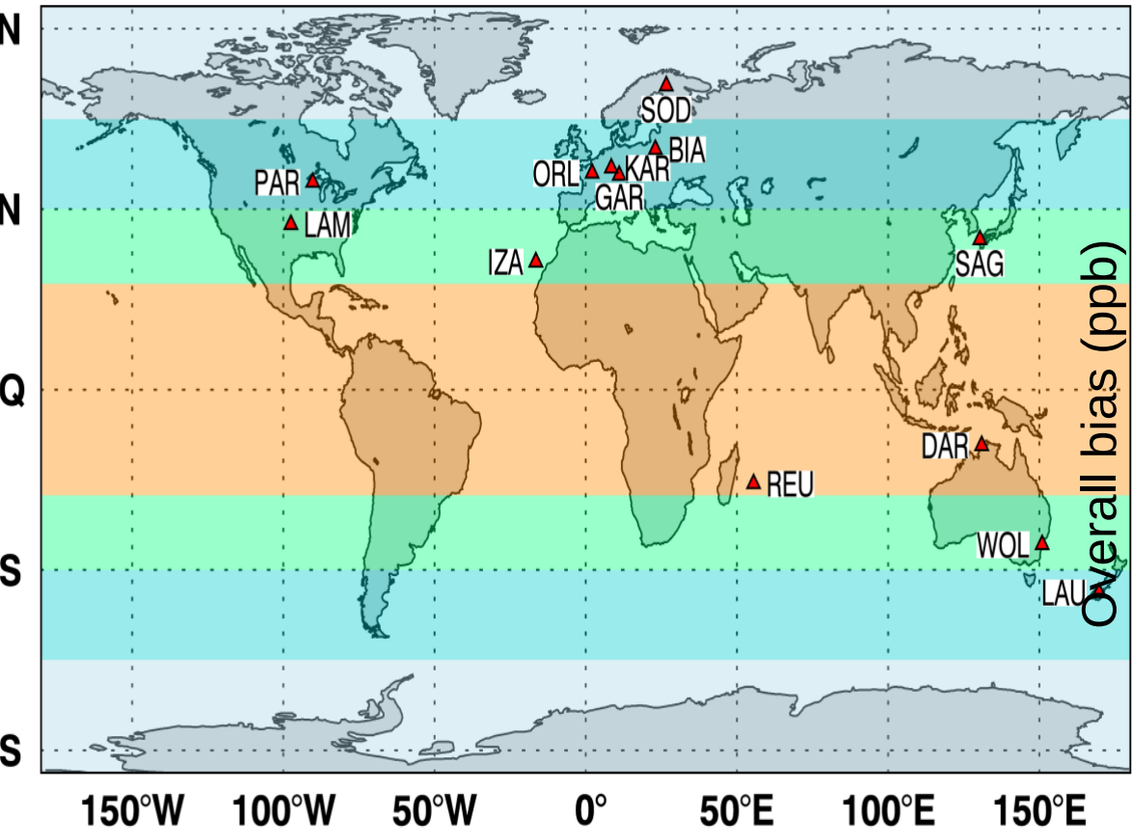
Evaluation of model simulations with TCCON

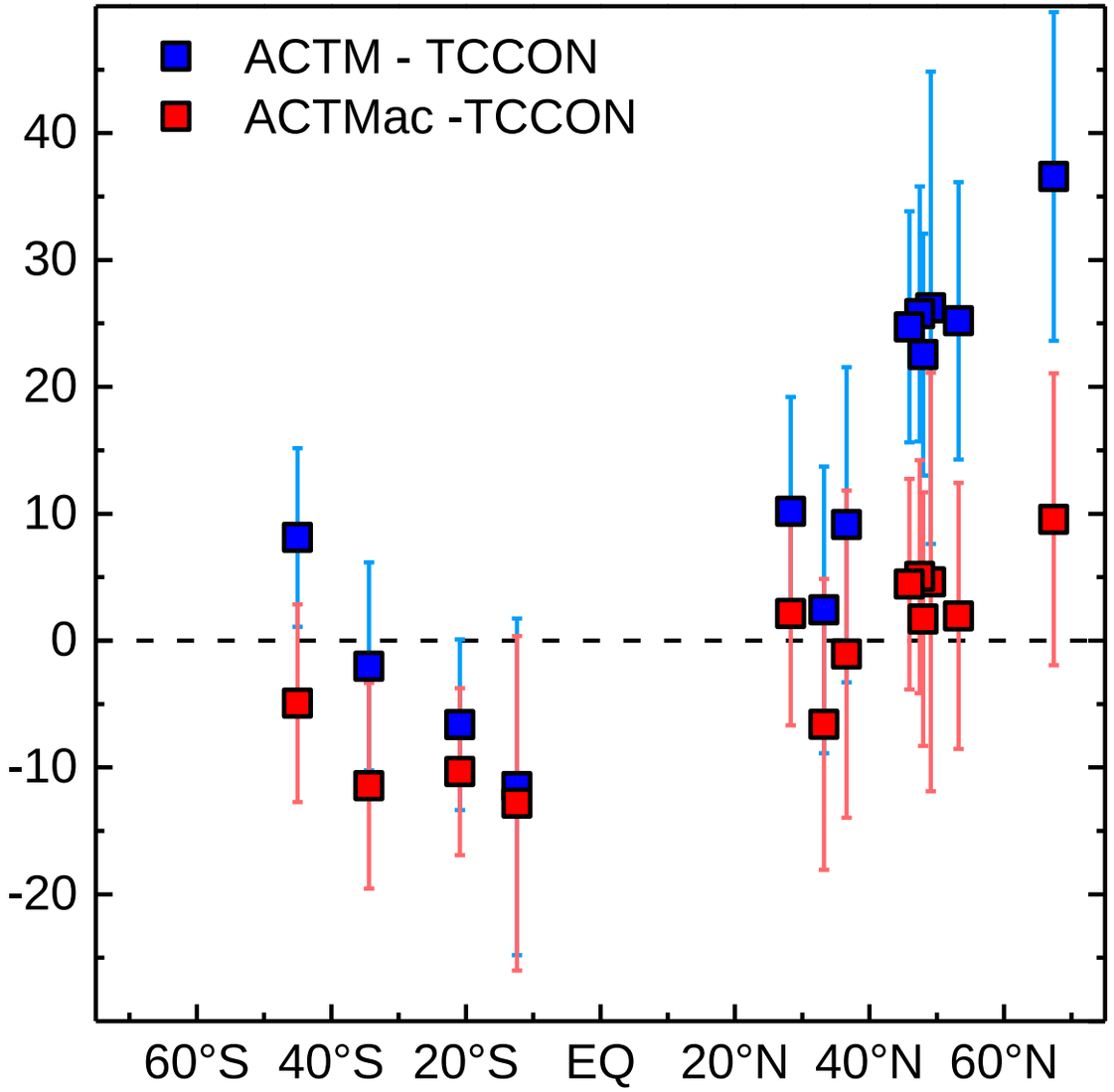
- Convert modeled CH_4 vmr profiles into XCH_4 (account for TCCON a priori and kernels!)

ACTM **TCCON** **ACTMac**

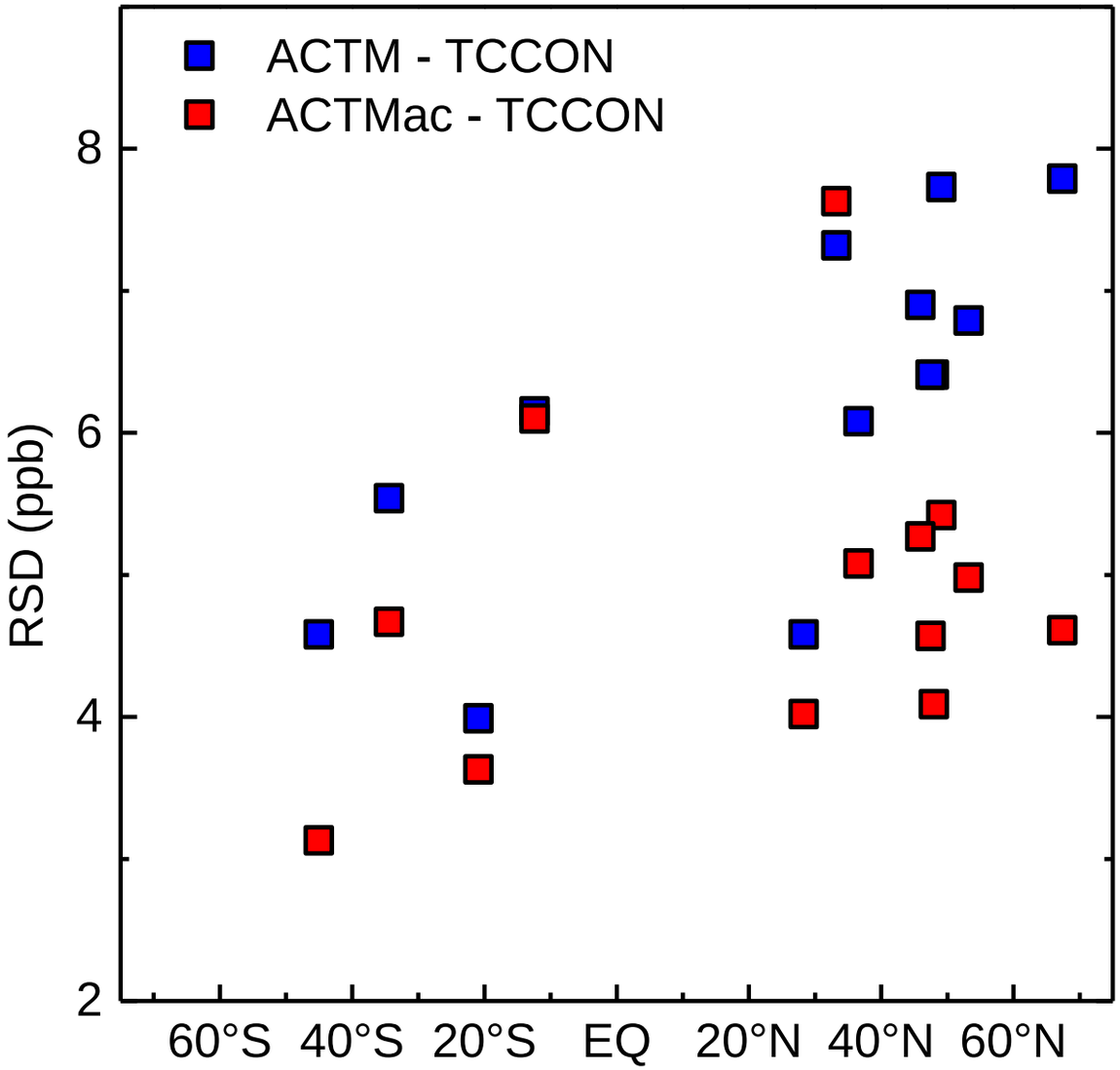


Model-data agreement XCH_4: Overall bias



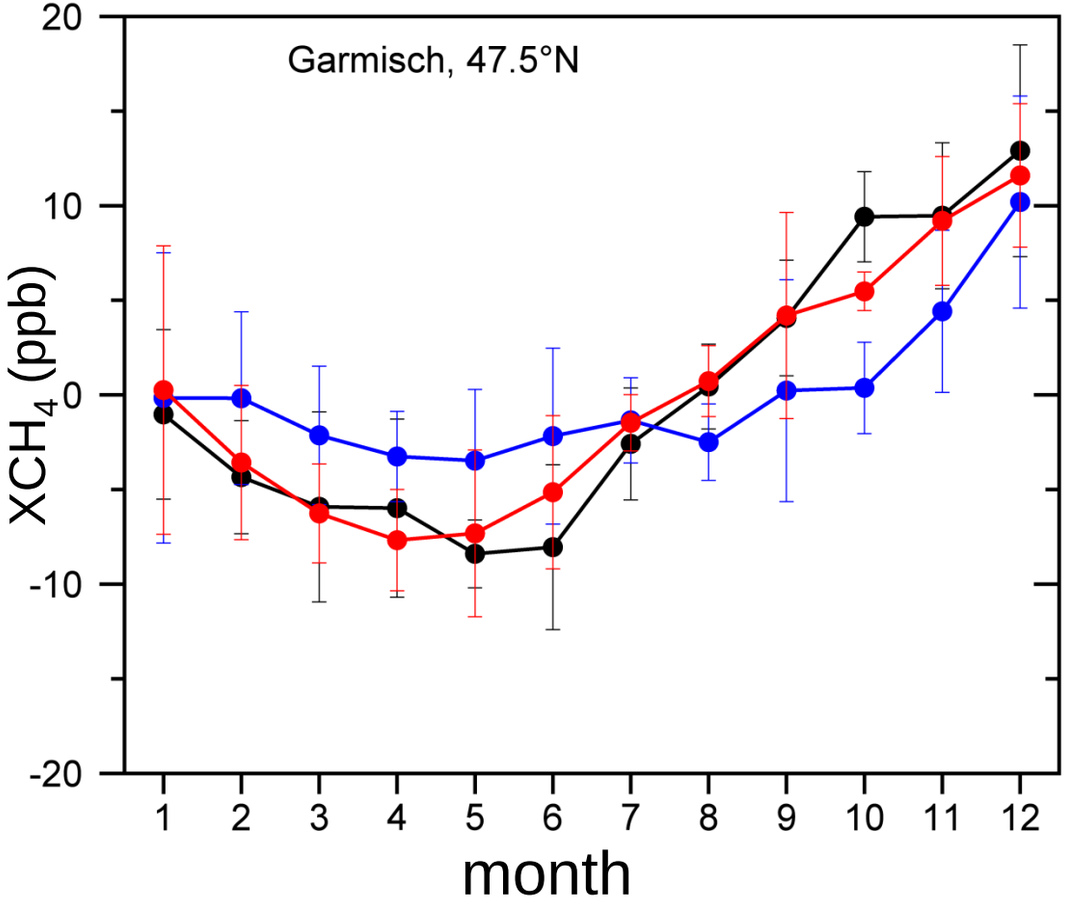


Model-data agreement: Seasonal bias

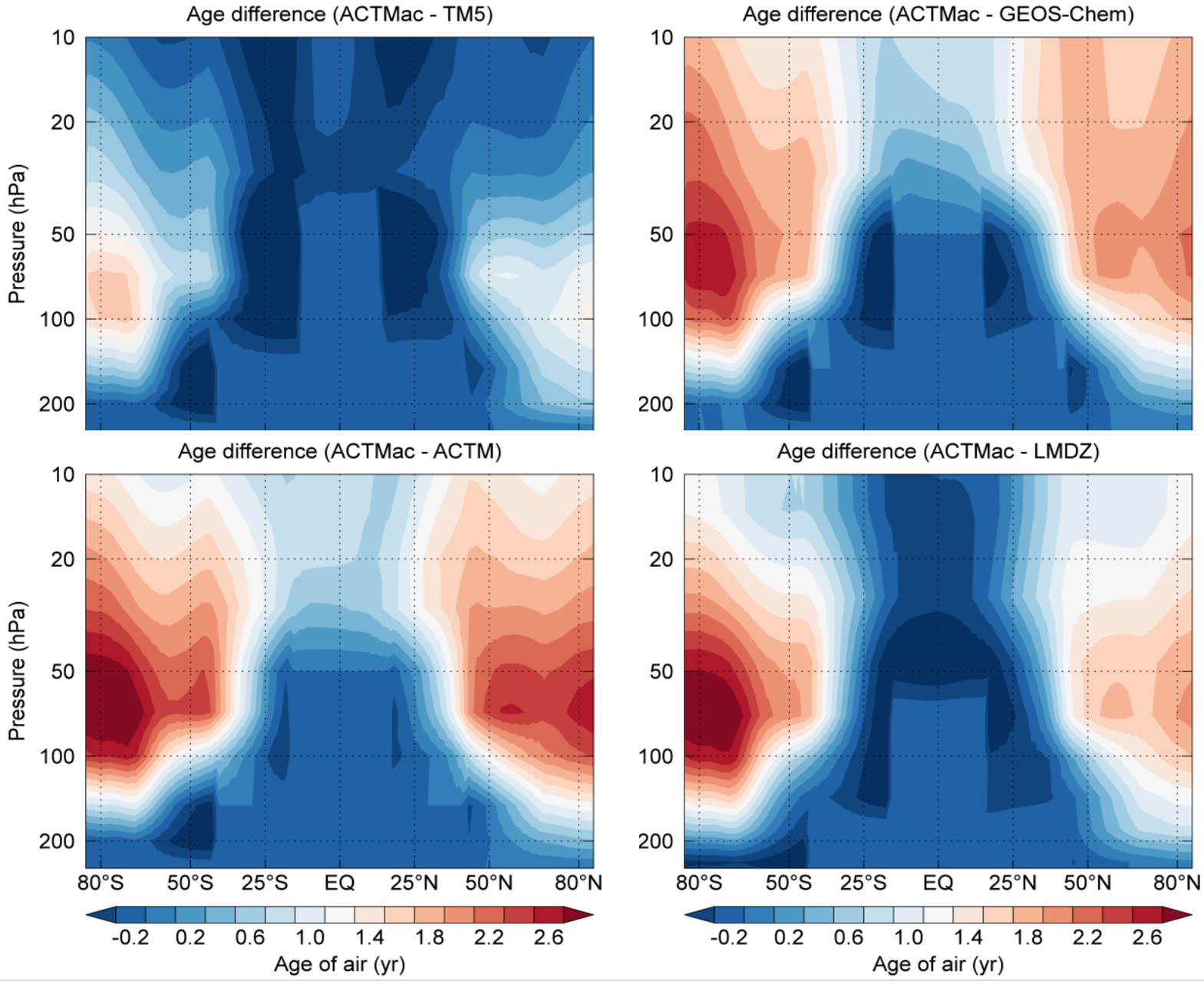


XCH_4 mean seasonal cycle

ACTM TCCON ACTMac



Evaluation of additional CTMs



Age differences: observational data – model simulations

TM5 GEOS-Chem

ACTM LMDZ

Impact of stratospheric model-transport error on inverting CH_4 fluxes

CH_4 distribution → CH_4 burden: $[CH_4]$

Original – Corrected → $[\Delta CH_4] = [CH_4] - [CH_4]$

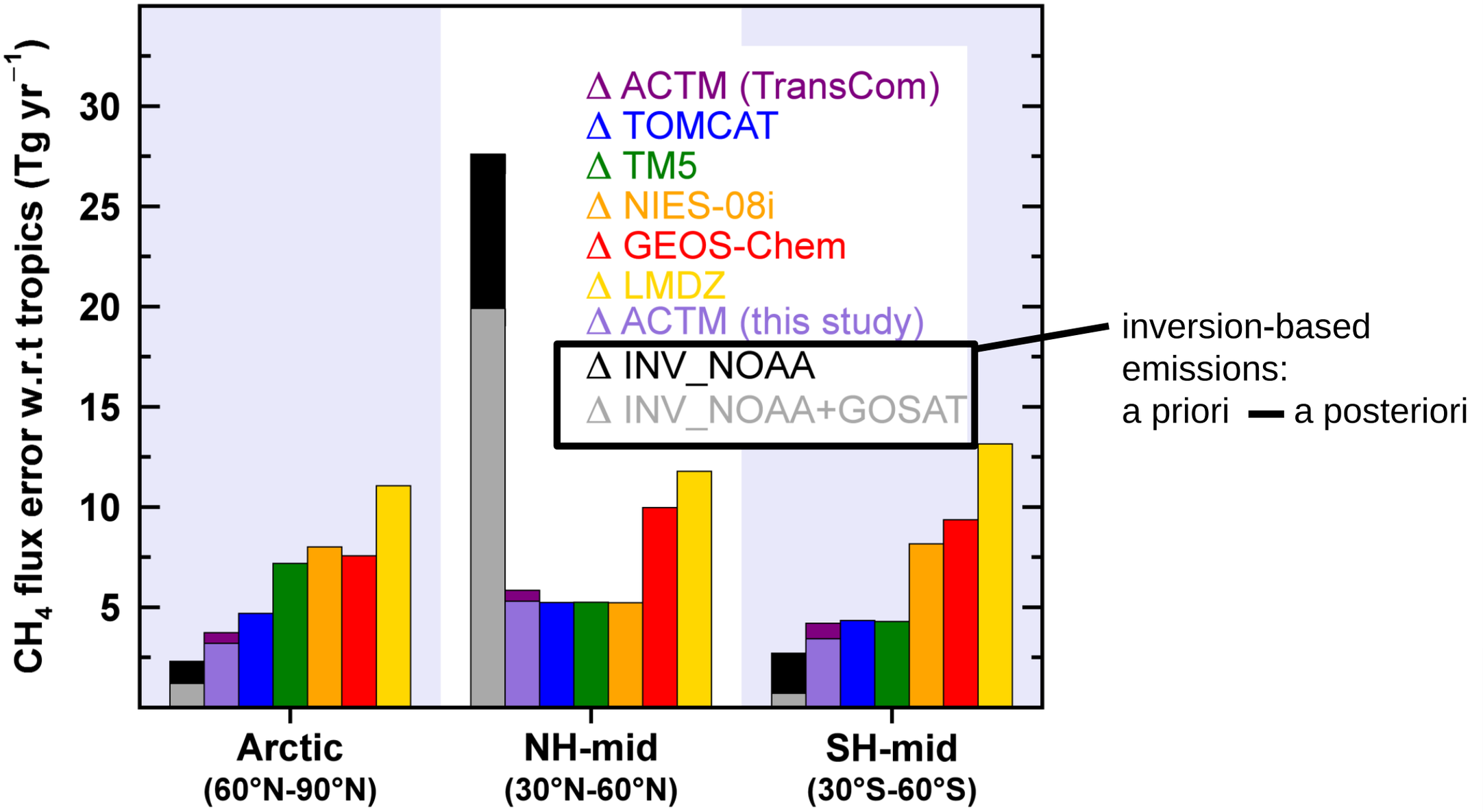
How much (additional) CH_4 has to be emitted
to produce global burden difference $[\Delta CH_4]$?

$[\Delta CH_4]$ → CH_4 emissions (= flux error)

Method: one-box model

$E = d[CH_4]/dt + [CH_4]/\tau$ τ = mean lifetime of atmospheric CH_4

Stratospheric model-transport error — flux error



Summary

► stratospheric CH_4 depends on stratospheric mean age

► stratospheric model-transport error lead to bias in stratospheric CH_4

► impact of stratospheric model-transport error on XCH_4 depends on latitude
(twofold: model bias × stratospheric contribution to total column)

► model errors in stratospheric CH_4 correspond to overestimation of CH_4 emissions

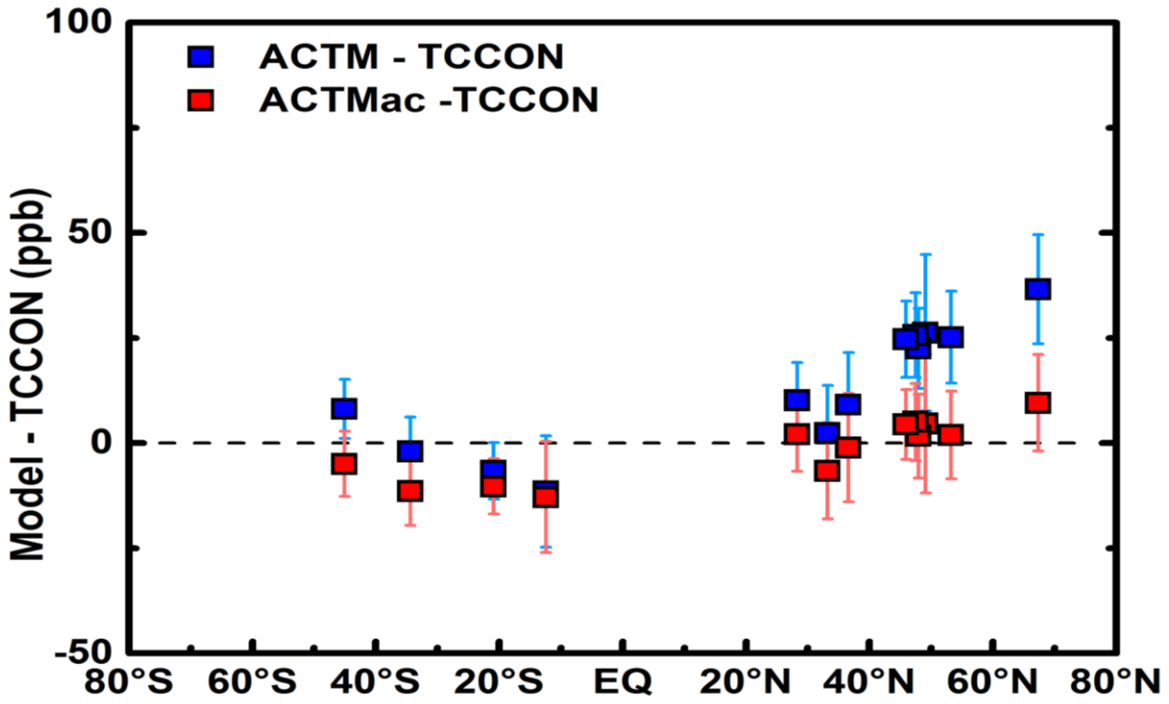
Conclusions

►stratospheric transport is an import controlling factor of XCH_4

►using XCH_4 data in atmospheric inversions requires accurate modeling of stratospheric transport

► Solving the stratospheric problem in inversions:

Present: ad hoc bias correction
Intermediate: stratospheric age correction
Future: improved modeling



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End — Thank you!

Additional material

- References
- Age observations
- Age correction
- Comparison between ACTM and satellite climatologies

Additional material: References

Waugh, D. Atmospheric dynamics: The age of stratospheric air. *Nat. Geosci.* **2,** 14 - 16 (2009).

Patra, P. K. *et al.* Observational evidence for interhemispheric hydroxyl-radical parity. *Nature* **513,** 219–223 (2014).

Volk, C. M. *et al.* Evaluation of source gas lifetimes from stratospheric observations. *J. Geophys. Res.: Atmos.* **102(D21),** 25543–25564 (1997).

Turner, A. J. *et al.* Estimating global and North American methane emissions with high spatial resolution using GOSAT satellite data. *Atmos. Chem. Phys. Discuss.* **15,** 4495-4536 (2015).

Additional material: Age observations

Harnisch, J., Borchers R., Fabian P. & Maiss M. Tropospherictrends for CF_4 and $C2F_6$ since 1982 derived from SF_6 dated stratospheric air. *Geophys. Res. Lett.* **23,** 1099–1102 (1996).

► 5 balloone flights between 8 – 34 km (MPAE cyrosampler) at 3 locations:

17°N (India, 1987); 44°N (France, 1993); 68°N (Sweden, 1992/1995)

Patra, P., Lal S., Subbaraya B., Jackman C. H. & Rajaratnam P. Observed vertical profile of sulfur hexafluoride (SF_6) and its atmospheric applications. *J. Geophys. Res.* **102,** 8855–8859 (1997).

► 1 balloone flight between 8 – 37 km (cyrosampler) at 3 locations:

17°N (India, 1994)

Additional material: Age correction

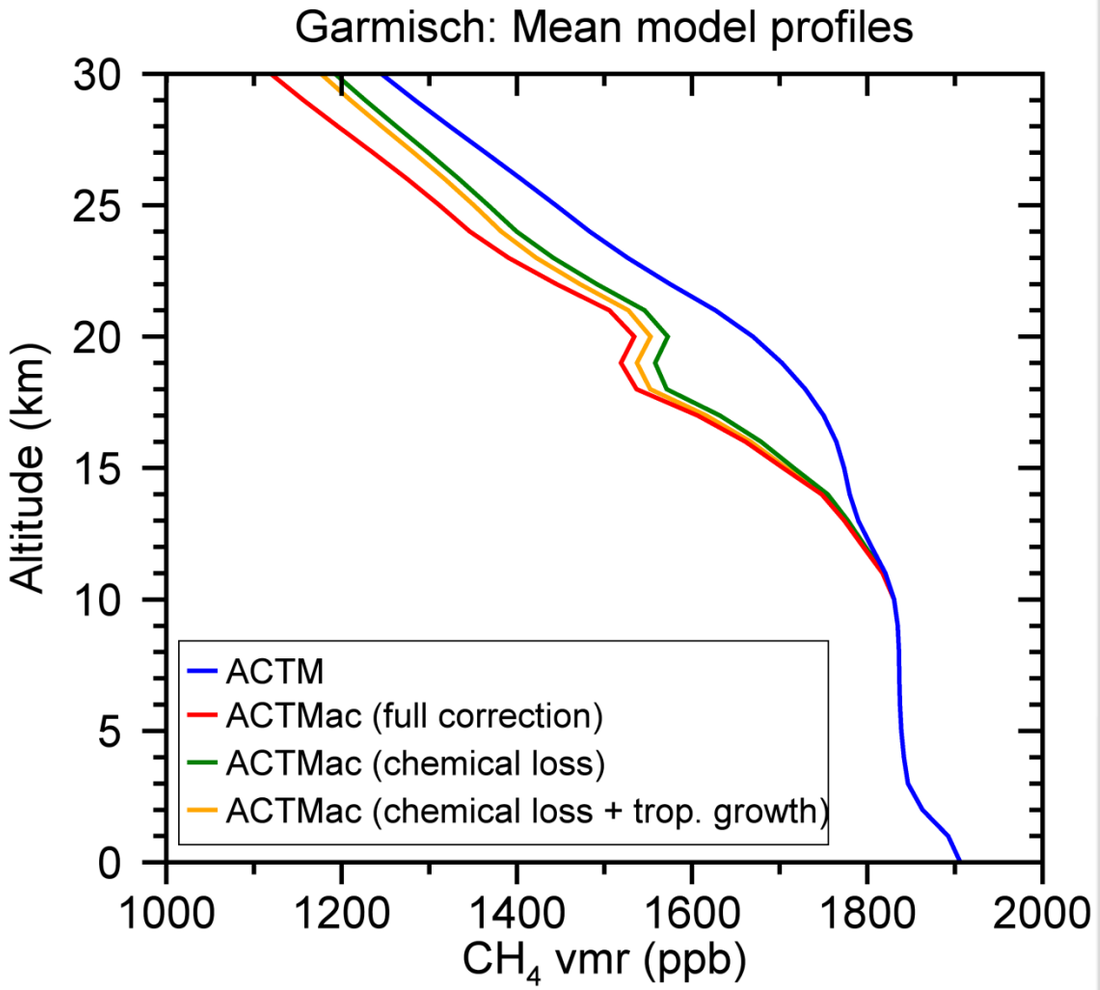
CH_4 mixing ratio profiles $\boldsymbol{x}$ as a function of mean age ($\boldsymbol{\Gamma}$).

$$\boldsymbol{x}(\boldsymbol{\Gamma}) = \boldsymbol{x}_0\,[1 - \beta_0\boldsymbol{\Gamma} - \gamma_0\boldsymbol{\Gamma} + \beta_0\gamma_0(\boldsymbol{\Gamma}^2 + 2\boldsymbol{\Delta}^2)]$$

$$\beta_0 = -\frac{1}{\boldsymbol{\Gamma}_{tp}}\frac{\mathrm{d}\boldsymbol{x}}{\mathrm{d}\boldsymbol{\Gamma}}\Big|_{\boldsymbol{\Gamma}_{\mathrm{tp}}}$$

Δ is the width of the age spectrum.

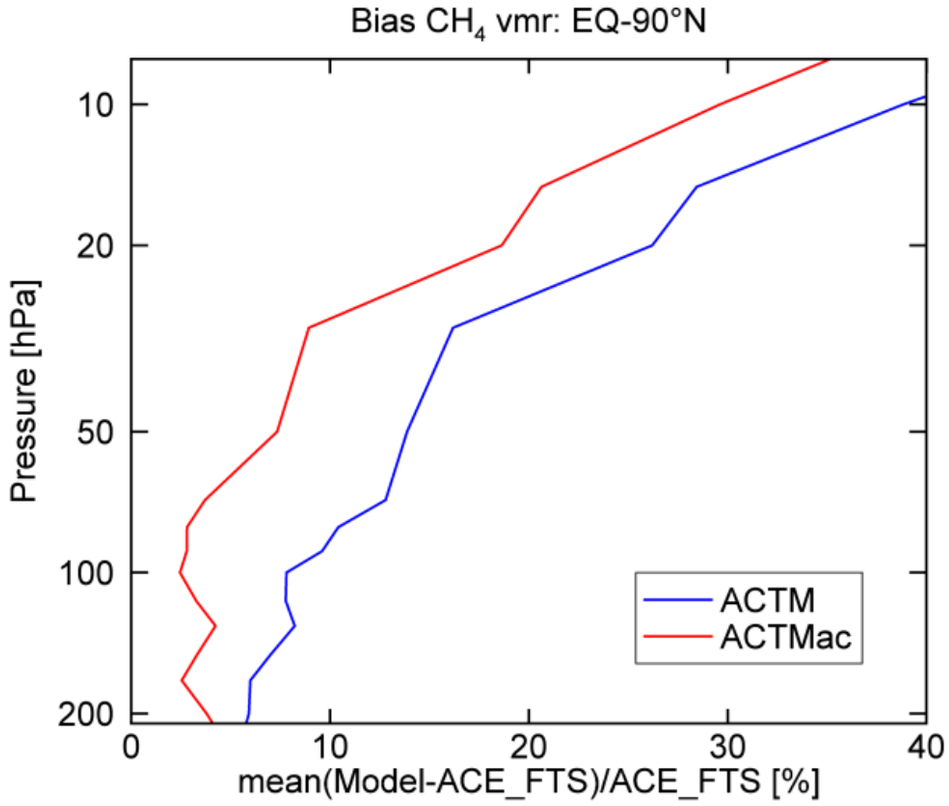
β_0 = original CH_4 model profiles.
γ_0 = 6 ppb yr^{-1} since the year 2006
$\Delta^2 = 1.25\,(\boldsymbol{\Gamma} + 0.5)$



Additional material:
Evaluation using ACE/HALOE climatology

Two-year model climatology vs. satellite climatology

Model vs. ACE



Model vs. HALOE