New insight into renewed methane increase: constraints by long-term evolution of ethane interhemispheric gradients

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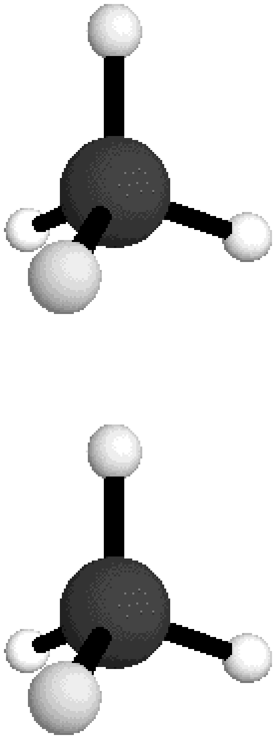
REGIONAL CLIMATE SYSTEMS – Atmospheric Variability and Trends

Motivation – why look at methane?

IR-active vibrations



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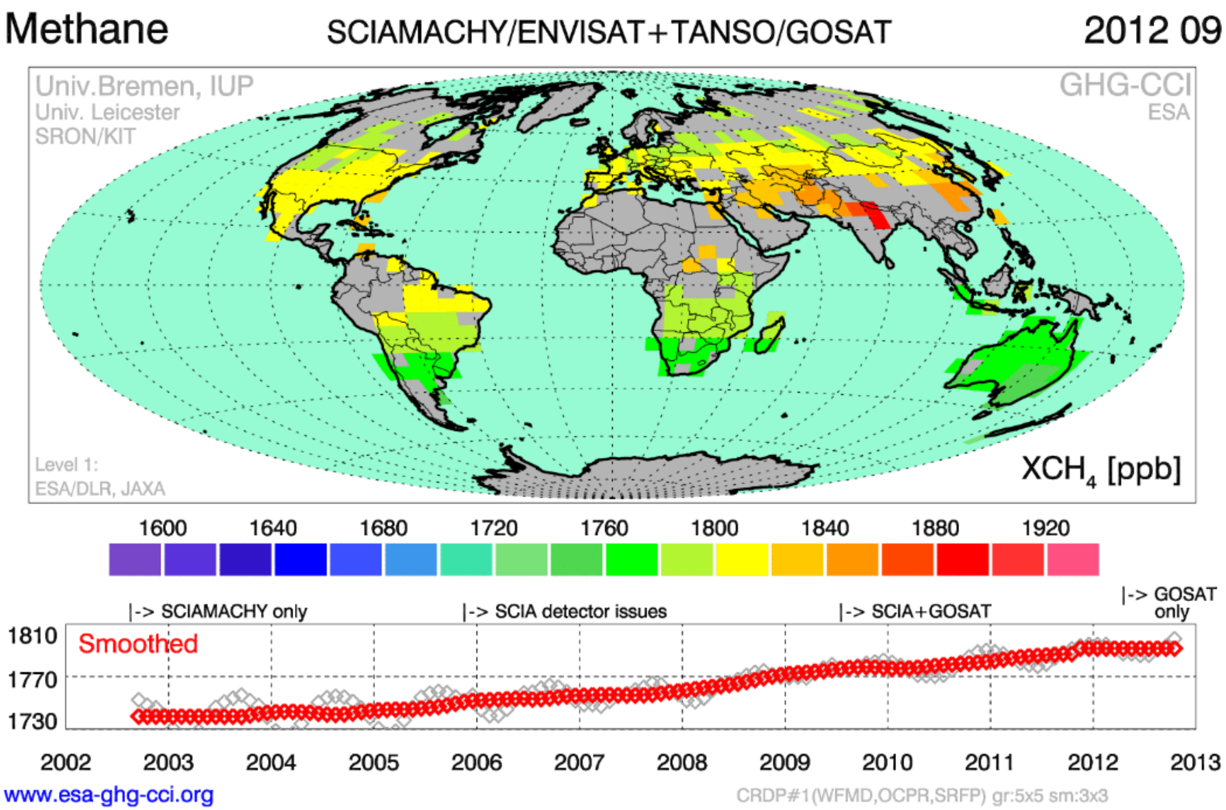
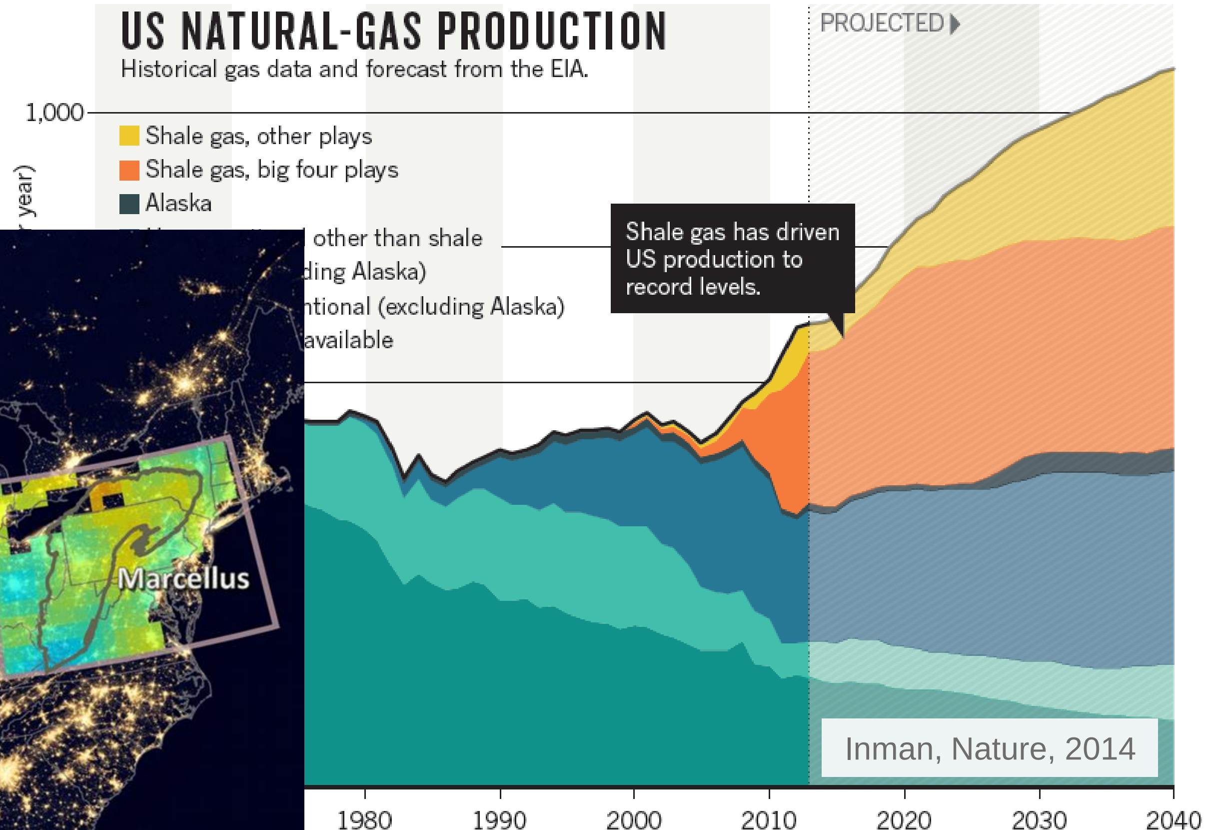
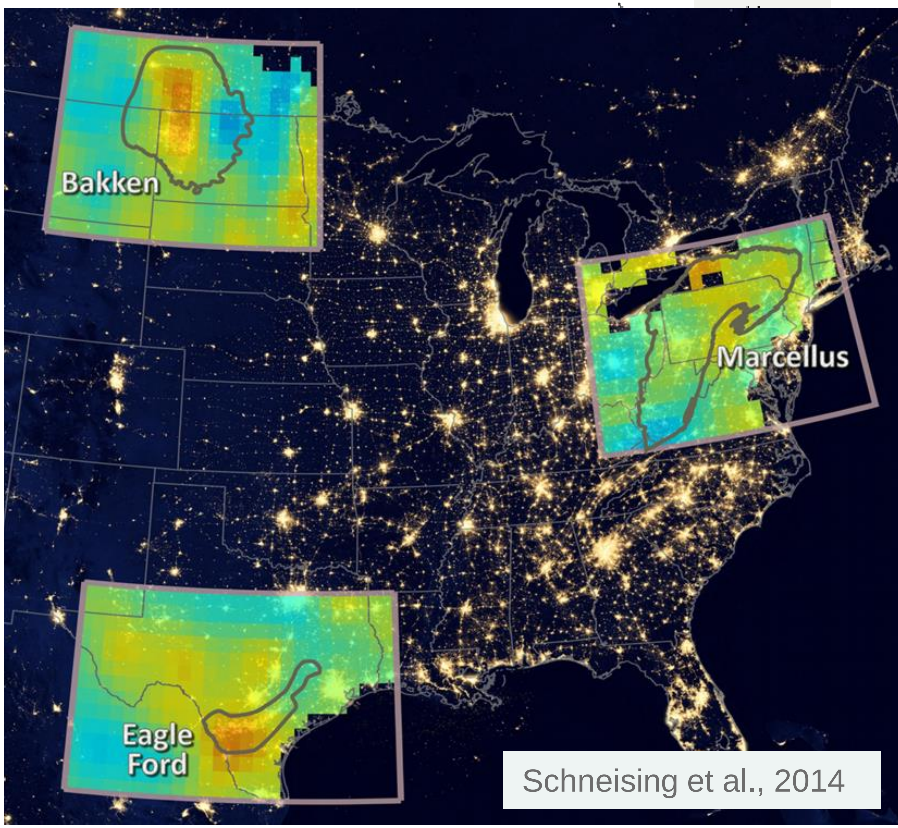
Long-term increase



Methane sources – US shale gas revolution



Inman, Nature, 2014



Schneising et al., 2014

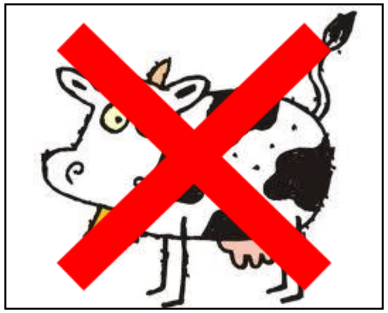
US CH_4 emissions underestimated + leakage rates highly uncertain

Methane sources – what does ethane tell us?

- shares major source with methane: fossil fuel production / distribution



- no significant biogenic ethane sources


→ C_2H_6 as valuable tracer for thermogenic methane

FTIR spectrometry – retrieval strategies

	CH_4	C_2H_6
strategy	Sussmann et al., 2011	NDACC recipe, 2014
micro windows [cm-1]	2613.70 – 2615.40 2835.50 – 2835.80 2921.00 – 2921.60	2976.66 - 2976.95 2983.20 - 2983.55
interfering species	H2O, HDO, NO_2	H_2O, O_3, CH_4
line list	HIT00 + 2001 update	C_2H_6 pseudo-lines (Franco et al., 2015) other species: HIT08 + 09
regularization **a priori profile**	Tikhonov-L_1, DOFS ~ 2.0 WACCM v6	Tikhonov-L_1, DOFS ~ 1.6 WACCM v6

- spectral fitting code: PROFFIT v9.6
- station-to-station harmonization of regularization strength

KIT-Campus Alpin
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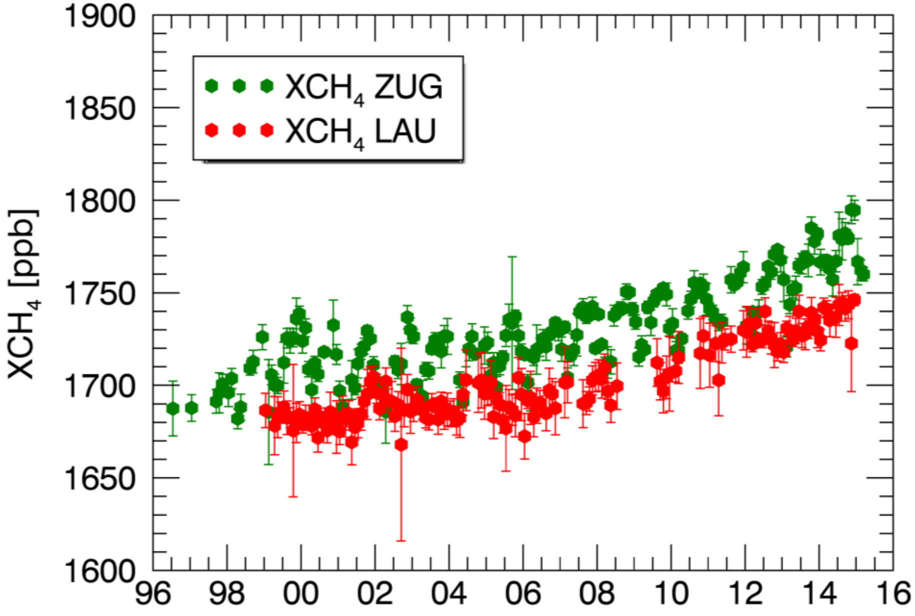
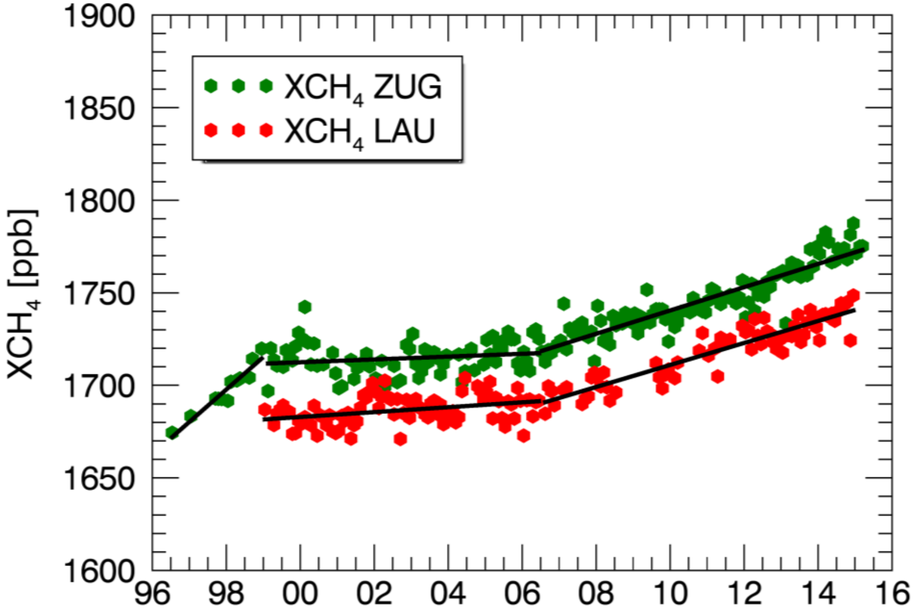
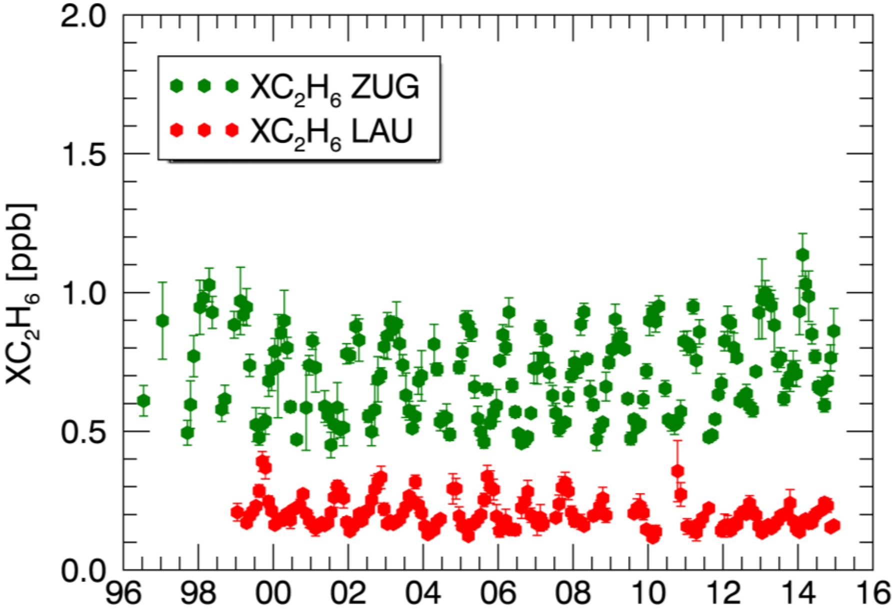
Trends – Zugspitze and Lauder methane trend

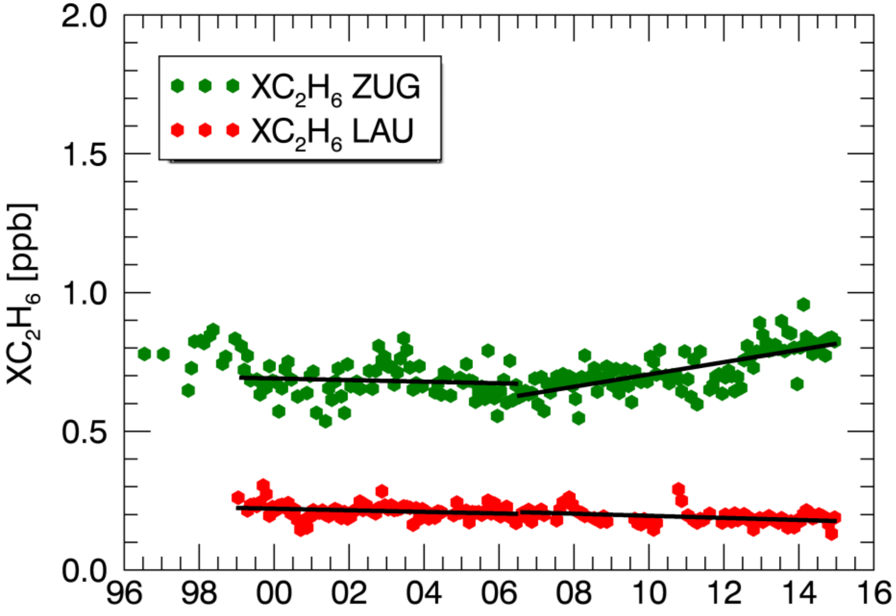




	trend [ppb/yr] Jan 1999 – Jun 2006	trend [ppb/yr] Jul 2006 – Dec 2014
Zugspitze (47° N)	0.76 [-0.14, 1.64]	6.29 [5.70, 6.87]
Lauder, NZ (45° S)	1.32 [0.61, 2.05]	5.94 [5.36, 6.52]

Trends – Zugspitze and Lauder ethane trend

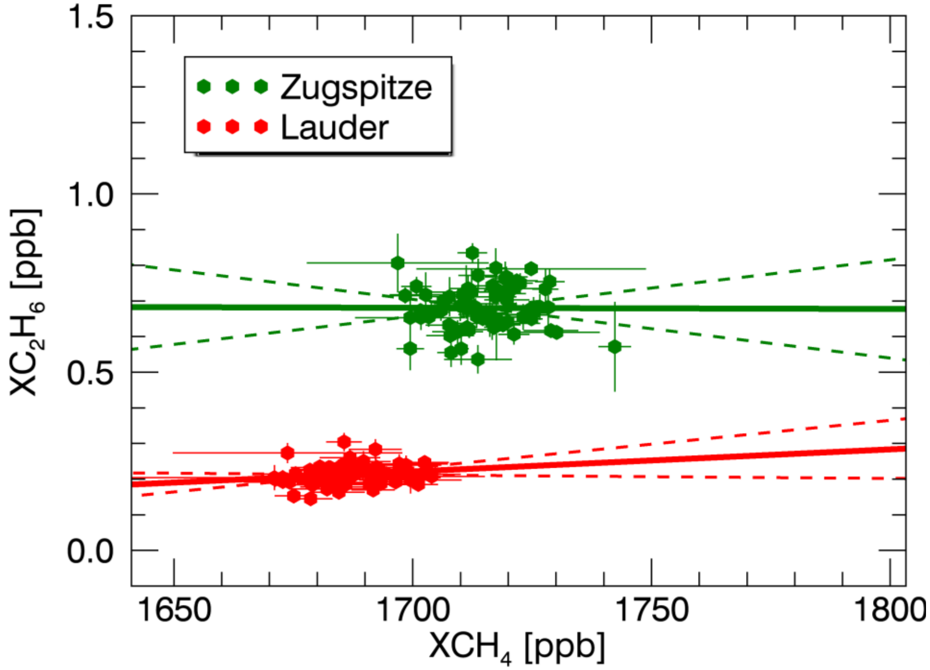




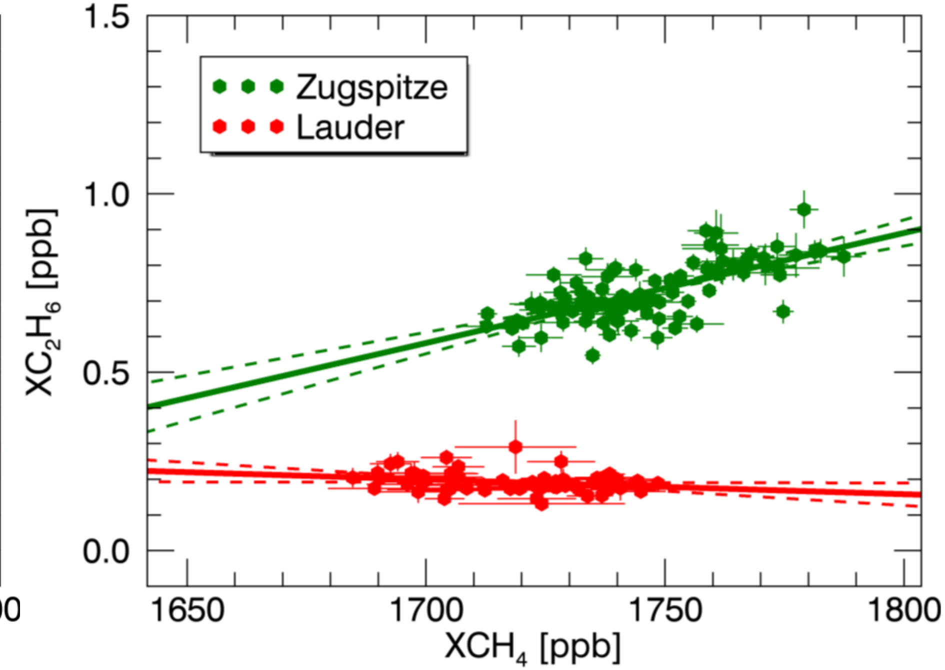
	trend [10^{-2} ppb/yr] Jan 1999 – Jun 2006	trend [10^{-2} ppb/yr] Jul 2006 – Dec 2014
Zugspitze (47° N)	-0.29 [-0.91, 0.35]	2.21 [1.77, 2.66]
Lauder, NZ (45° S)	-0.29 [-0.53, -0.04]	-0.38 [-0.60, -0.17]

Trends – ethane-to-methane ratio

Jan 1999 – Jun 2006



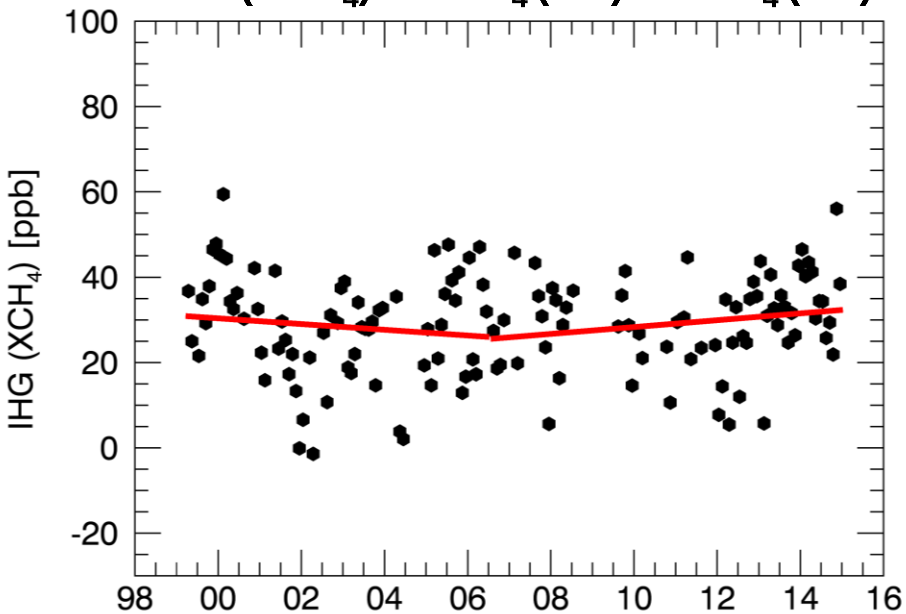
Jul 2006 – Dec 2014



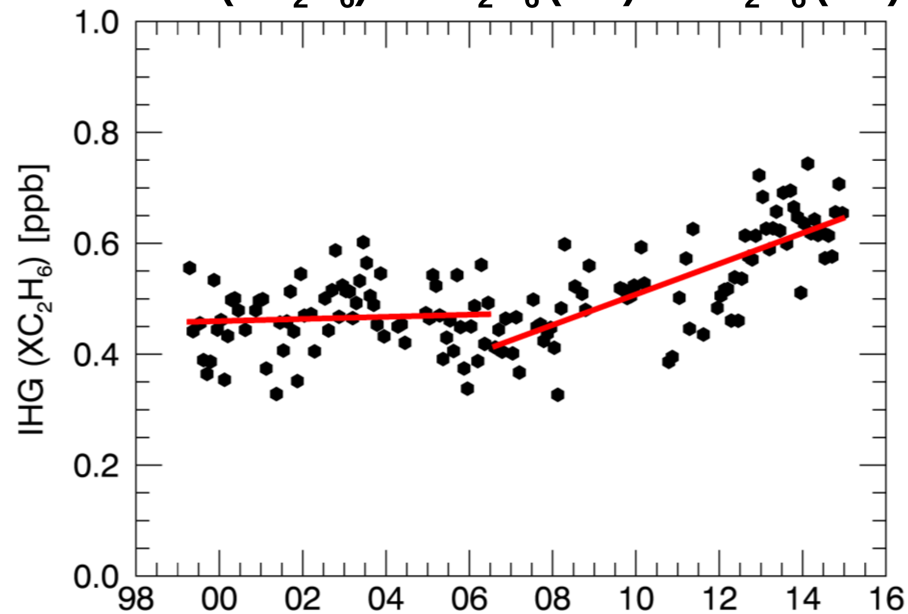
slope [%]	Jan 1999 – Jun 2006	Jul 2006 – Dec 2014
Zugspitze	-0.003 ± 0.161 (no)	0.309 ± 0.067 (yes)
Lauder	0.062 ± 0.072 (no)	-0.041 ± 0.039 (yes)

Trends – interhemispheric gradients

IHG (XCH_4) = XCH_4 (NH) – XCH_4 (SH)

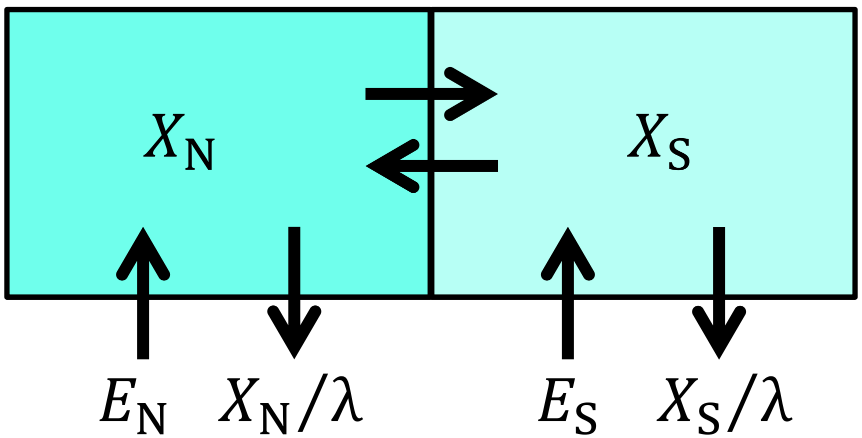


IHG (XC_2H_6) = XC_2H_6 (NH) – XC_2H_6 (SH)

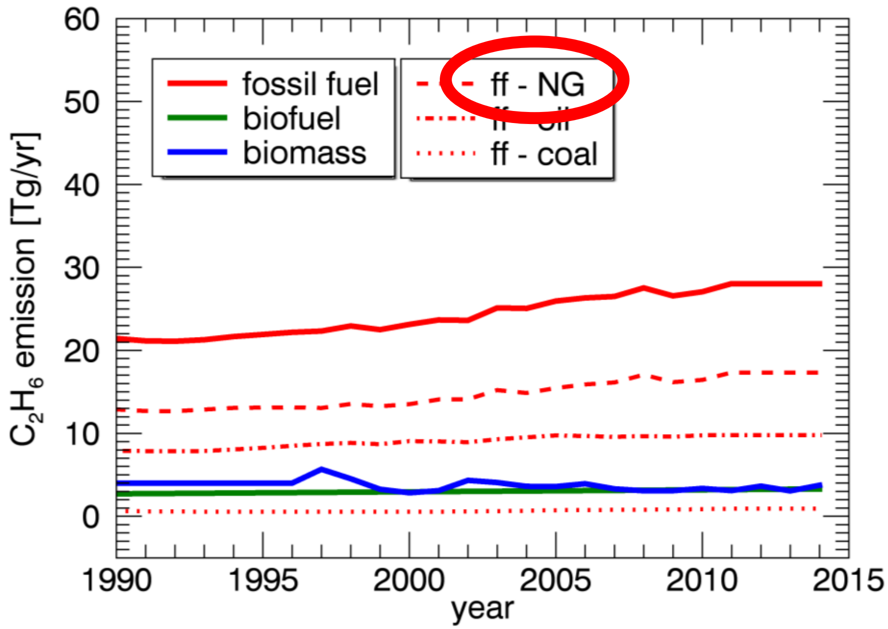


	Jan 1999 – Jun 2006	Jul 2006 – Dec 2014
IHG (XCH_4) [ppb/yr]	-0.67 [-2.01, 0.68]	0.80 [-0.18, 1.79]
IHG (XC_2H_6) [10^{-2} ppb/yr]	0.19 [-0.45, 0.83]	2.76 [2.20, 3.31]

Two-box model – global ethane budget



X_N, X_S	column-averaged mole fraction [ppb]
τ_{ex}	interhemispheric exchange time [yr]
E_N, E_S	emission [Tg/yr]
λ	atmos. lifetime [yr]

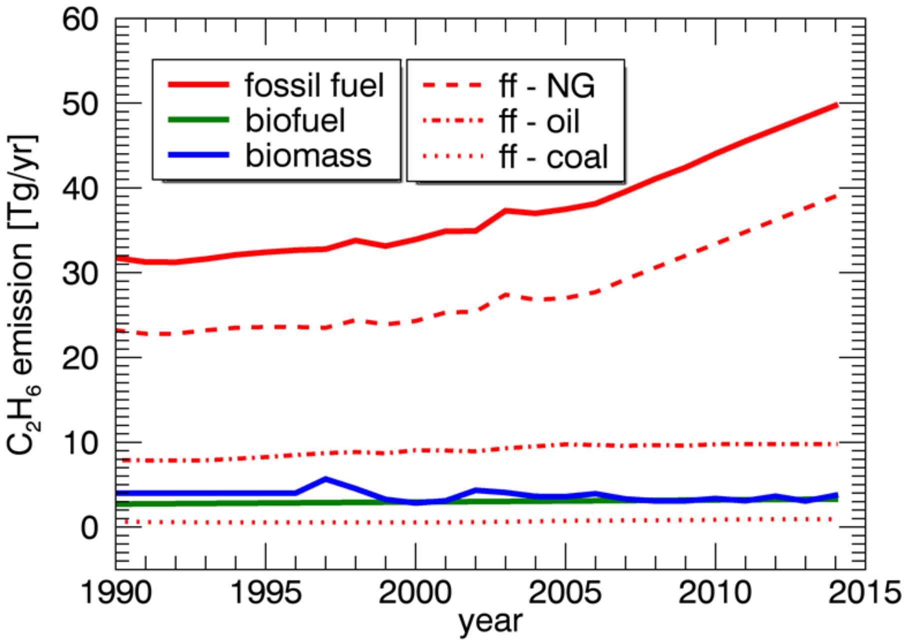


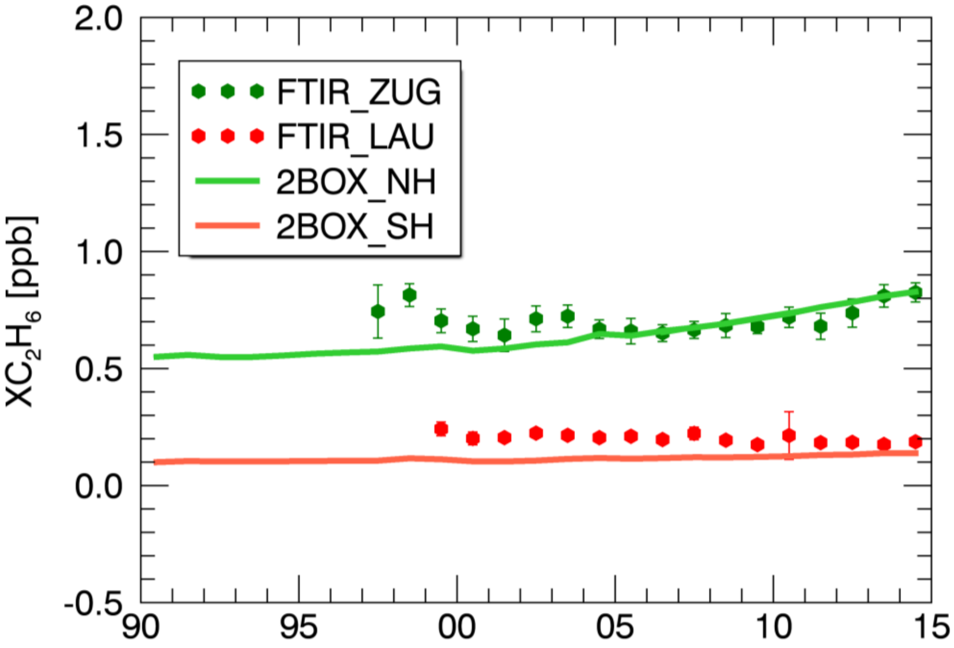
C_2H_6 emissions estimates:

- biomass burning (GFED4s)
- biofuel use × emiss. factor (Andreae and Merlet, 2001)
- fossil fuel prod. / distrib. (Schwietzke et al., 2014)

Two-box model – optimize ethane emissions

- minimize model-observation difference in XC_2H_6 trend from 2006 – 2014
- assumption: all "missing" emissions attributable to growing natural gas source





necessary change in C_2H_6 emissions from natural gas between 2006 and 2014:
$\Delta E_{\mathrm{gas}}^{\mathrm{max}}$ = 11.1 [3.9, 24.7] Tg/yr

Two-box model – renewed methane increase

Change in C_2H_6 natural gas emissions between 2006 and 2014
derived from ethane two-box model

Quantify associated methane emission change
using reasonable CH_4-C_2H_6-ratio

Quantify natural gas contribution to total renewed methane increase
applying analogous two-box model for methane

Summary and outlook

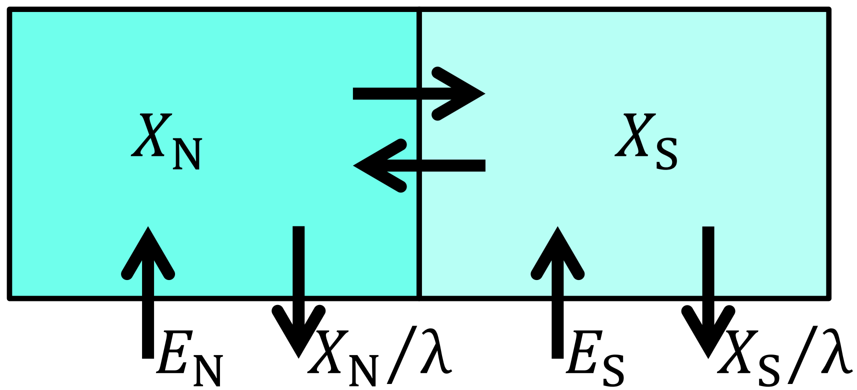
- Harmonized retrieval of methane and ethane for Zugspitze (47° N) and Lauder (45° S) time series
- Long-term trend analysis for methane and ethane (1995 - 2014):
  - Consistent renewed methane increase in both hemispheres
  - Significant ethane increase since 2006 in the northern hemisphere
- Two-box model for hemispheric ethane budgets:
  - Minimize model-observation difference of ethane trend since 2006
  - Optimize ethane emissions from natural gas production
- Methane two-box model: quantify contribution of fossil fuel emissions to renewed methane increase

Thank you for your attention!

OUTLINE

- Motivation
- FTIR trace gas retrieval
- Long-term trend analysis for ethane and methane
- Two-box model for global ethane budget
- Summary and outlook

Two-box model – global ethane budget



$$\frac{dX_N}{dt} = E_N - \frac{X_N}{\lambda} - \frac{(X_N - X_S)}{\tau_{ex}}$$

$$\frac{dX_S}{dt} = E_S - \frac{X_S}{\lambda} + \frac{(X_N - X_S)}{\tau_{ex}}$$

X_N, X_S	column-averaged mole fraction [ppb]
τ_{ex}	interhemispheric exchange time [yr]
E_N, E_S	emission [Tg/yr]
λ	atmos. lifetime [yr]

Two-box model – uncertainties

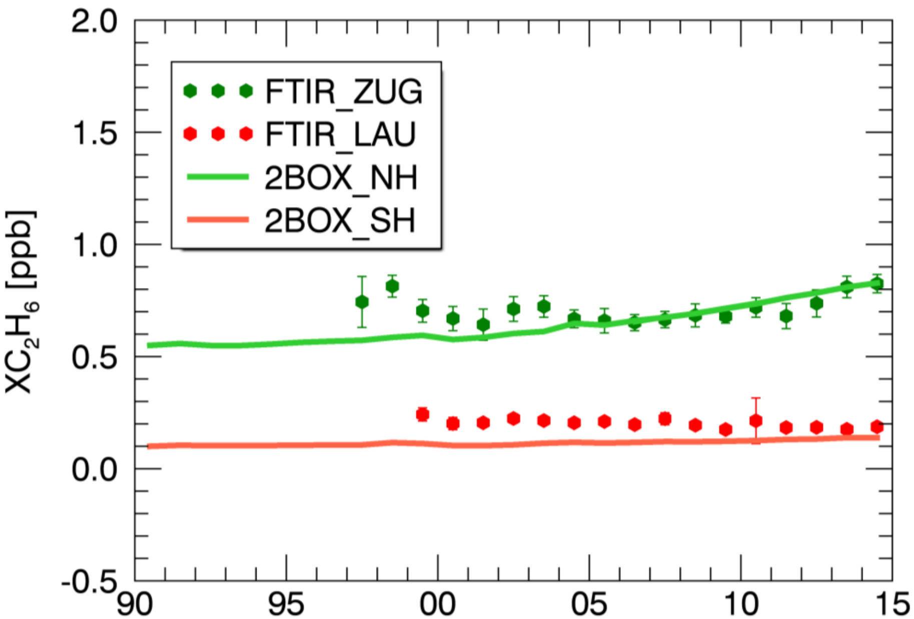
model parameters / ethane emissions / emission fraction in NH

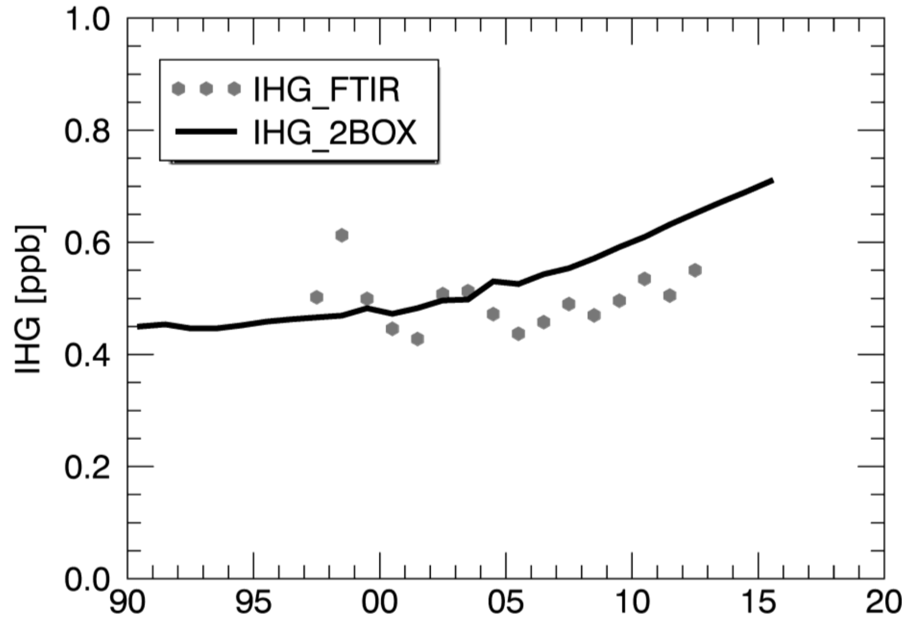
	λ [month]	τ_{ex} [yr]	c [Tg/ppb]
range	2.6 [2.0, 3.2]	0.98 [0.55, 1.41]	18 [10, 26]
slope [10^{-2} ppb/yr]	2.21 [1.75, 2.65]	2.21 [2.04, 2.30]	2.21 [3.98, 1.53]

	E_{bb} [Tg/yr]	E_{bf} [Tg/yr]	E_{coal} [Tg/yr]	E_{oil} [Tg/yr]
range	± 65%	± 75%	± 50%	± 50%
slope [10^{-2} ppb/yr]	[2.22, 2.20]	[2.19, 2.23]	[2.19, 2.23]	[2.20, 2.22]

	$f_{N,bb}$	$f_{N,bf}$	$f_{N,coal}$	$f_{N,oil}$	$f_{N,gas}$
range	0.53 ± 10%	0.81 ± 10%	0.90 ± 10%	0.93 ± 10%	0.96 ± 10%
s. [10^{-2} ppb/yr]	-	-	-	-	[2.03, 2.28]

Two-box model – ethane increase 2006-2014





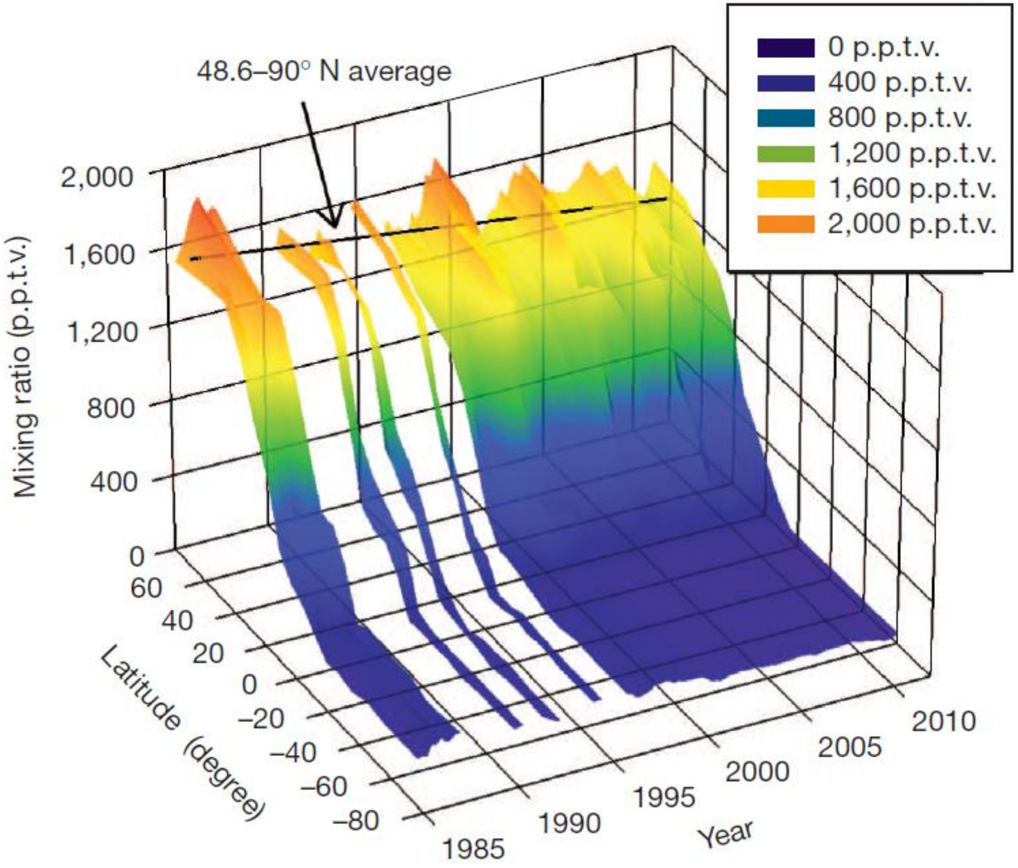
- optimization of C_2H_6 NG emission for 2006-2014

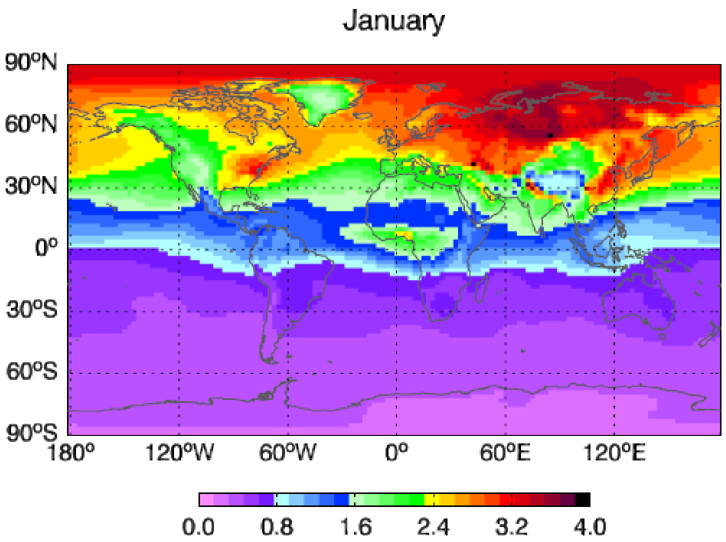
  $$E_{\mathrm{opt}} = E_{\mathrm{ini}} \times (2.0 \pm 0.1)$$

- change in C_2H_6 emission to reconstruct observed XC_2H_6 trend

  $$dE_{\mathrm{opt}} = 11.1\ [3.9, 24.7]\ \mathrm{Tg/yr}$$

Interhemispheric gradient of ethane





10^{16} molecules/cm^2

Simpson et al., Nature, 2012

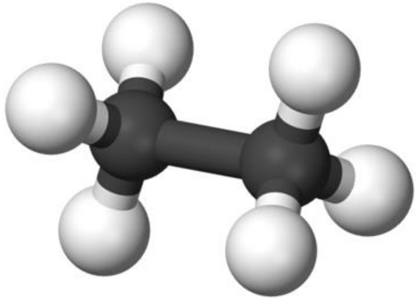
Xiao et al., 2008

Background – what does ethane tell us?

- second most abundant component of natural gas (~1-6%)
- no significant biogenic C_2H_6 sources
  → valuable tracer for thermogenic methane

	Methane	Ethane
Natural sources	40%	--
Anthrop. sources	50-65%	~ 100%
- Agriculture+waste	60%	--
- Fossil fuel prod.	30%	62%
- Biofuel use	10%	20%
- Biomass burning		18%
Sink: Oxidation by OH	90%	98%
Lifetime	9 years	2 months

IPCC, 2013; Xiao et al., 2008



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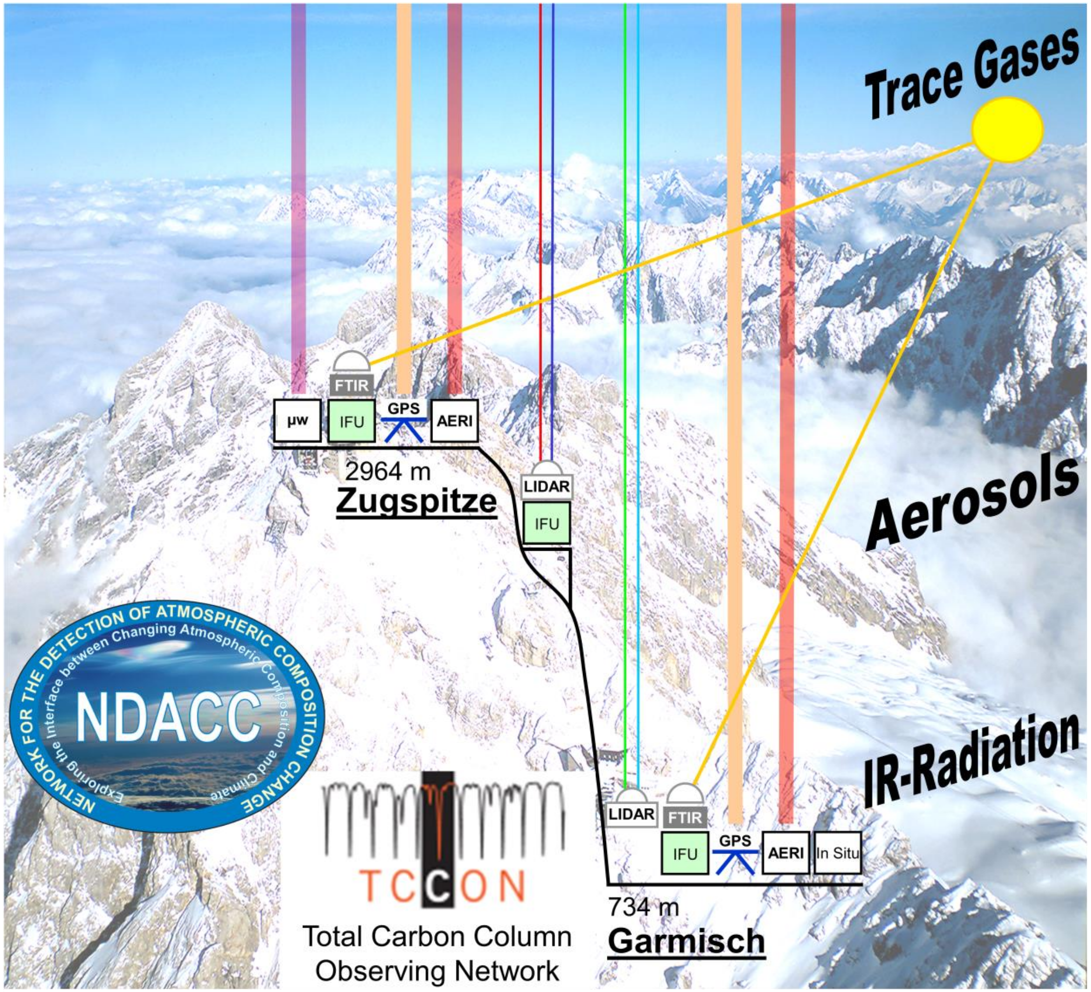
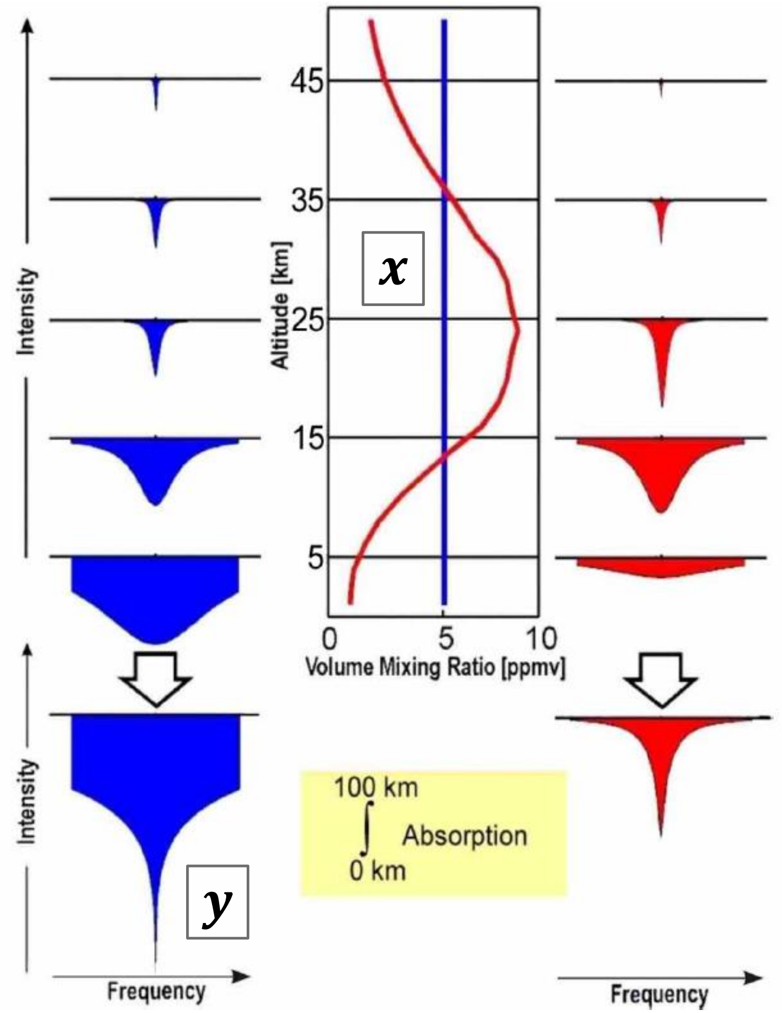
FTIR spectrometry – Zugspitze observatory



- Ground-based solar absorption FTIR measurements at Zugspitze (2964 m)
- Spectral resolution: ~ 0.005 cm^{-1}
- Mid-infrared region: CH_4 retrieval within 2600 - 2900 cm^{-1}

FTIR spectrometry – trace gas retrieval



x - trace gas vertical profile
y - measured spectrum

Forward model $y = F(x)$

Inverse model $x = F^{-1}(y)$

→ non-linear, ill-posed problem

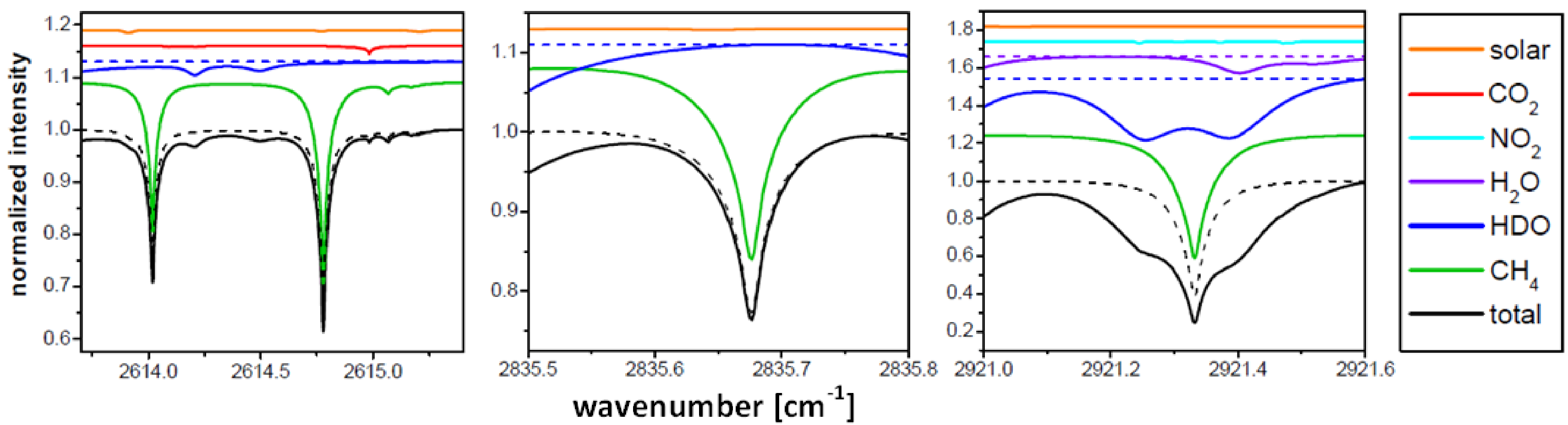
→ minimize cost function

$$\overbrace{(y - F(x))^T S_\varepsilon^{-1} (y - F(x))}^{\text{spectral error cost}} + \underbrace{(x - x_a)^T R (x - x_a)}_{\text{regularization cost}}$$

FTIR spectrometry – methane retrieval

Retrieval strategy (Sussmann et al., 2011):

- optimized for total column precision < 0.3% (1-σ diurnal variation)
- minimized H_2O/HDO-CH_4 interference error (seasonal bias < 0.14%)
- spectroscopic line list: HITRAN 2000 + updates
- altitude-constant Tikhonov-L_1 constraint ($DOFS \approx 2$)



Sussmann et al., 2011