Renewed methane increase (2007–2014):

Contribution of oil and natural gas emissions determined from methane and ethane column observations

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1. Motivation

Methane (CH_4) is important anthropogenic GHG:

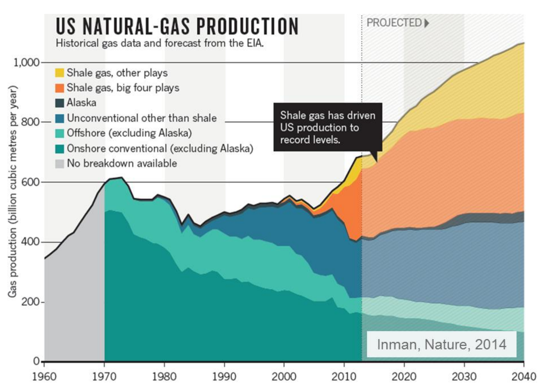
- Global warming potential: 84 (20 years)
- 20 % of global warming since 1750
- Relatively short lifetime of about 9 years
- ➢ Attractive target for climate-change mitigation

Renewed methane increase since 2007:

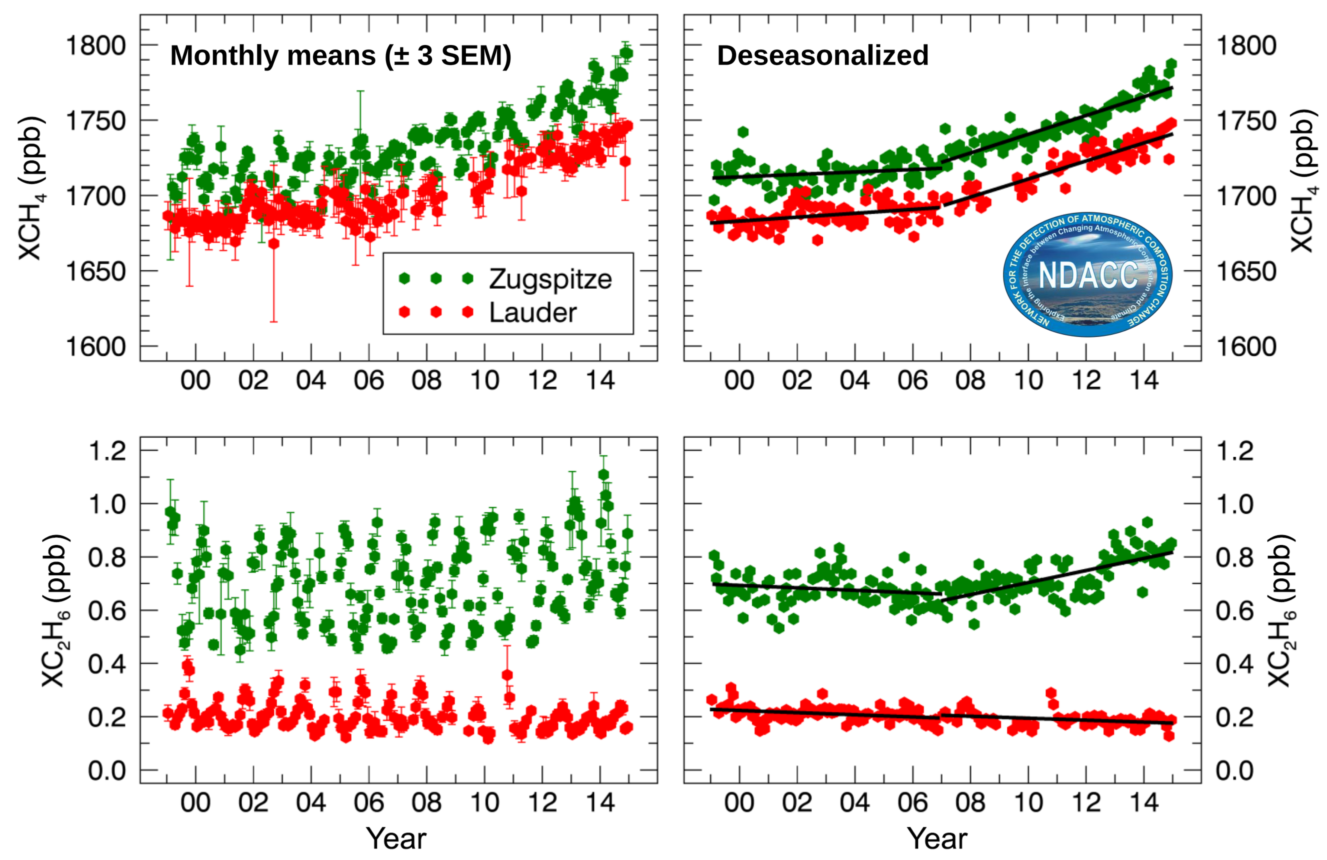
- Dominant drivers are likely growing emissions
  (i) from natural wetlands (biogenic) and
  (ii) from fossil fuel production (thermogenic)
- But: their relative contribution is uncertain
- ➢ Source attribution: ethane (C_2H_6) provides valuable constraint (no biogenic sources)

Strong increase in US oil & natural gas production:

- Leakage rates highly uncertain
- Climate benefit?
- Likely under-estimated CH_4 emissions from oil & gas sector



2. Long-term FTIR observations and trend analysis



High-resolution mid-infrared spectrometry:

- Ground-based solar FTIR measurements at Zugspitze (47° N) and Lauder (45° S)
- Representative of free tropospheric background conditions in each hemisphere
- Harmonized retrieval of column-averaged dry-air mole fractions (XCH_4 and XC_2H_6):

Retrieval	CH_4	C_2H_6
Strategy	Sussmann et al., 2011	NDACC IRWG, 2014
Micro-windows (cm^{-1})	2613.7 – 2615.4 2835.5 – 2835.8 2921.0 – 2921.6	2976.7 – 2977.0 2983.2 – 2983.6
Line list	HITRAN 2000 (+ 2001 update)	C_2H_6 pseudo-lines (Franco et al., 2015)
Regularization	Tikhonov-L_1 DOFS ~ 2.1 (1.8)	Tikhonov-L_1 DOFS ~ 1.6 (1.2)

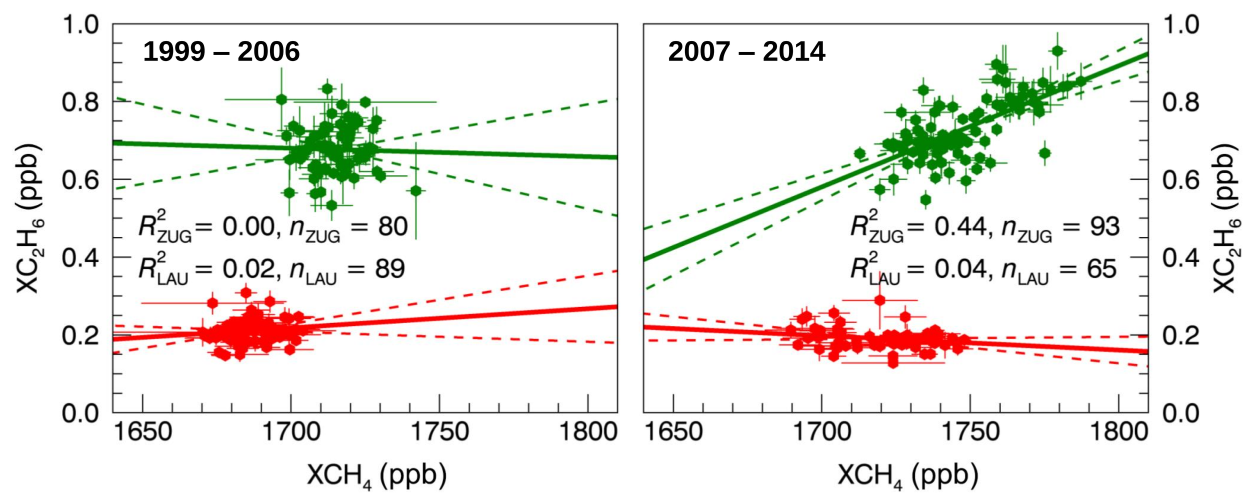
Long-term trend analysis (method in Sussmann et al., 2012):

- Linear trend estimate (uncertainty from bootstrap resampling of residuals)
- Consistent renewed methane increase since 2007 in both hemispheres
- Significant positive ethane trend in NH since 2007, but continuing decline in SH

Trend (ppb yr^{-1}) with 95 % confidence interval	1999 – 2006		2007 – 2014	
	Zugspitze	Lauder	Zugspitze	Lauder
Methane	0.8 [0.0, 1.6]	1.3 [0.6, 1.9]	6.2 [5.6, 6.9]	6.0 [5.3, 6.7]
Ethane ($\times 10^{-2}$)	-0.5 [-1.0, 0.1]	-0.4 [-0.7, -0.2]	2.3 [1.8, 2.8]	-0.4 [-0.6, -0.1]

3. Ethane – methane correlation

Correlation of XCH_4 and XC_2H_6 time series:



Correlation / Linear regression	1999 – 2006		2007 – 2014	
	Zugspitze	Lauder	Zugspitze	Lauder
Significant correlation?	no	no	yes	no
Regression slope (±2σ)	-0.02 ± 0.16 %	0.05 ± 0.08 %	0.31 ± 0.07 %	-0.04 ± 0.04 %

Source emission ratio (molar) from instantaneous mixing model:

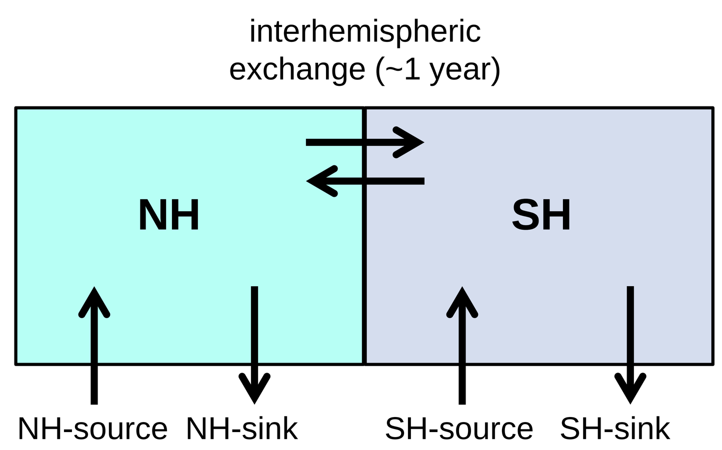
$EMR_{src} = EMR_{bg} \times k_{C2H6}/k_{CH4}$ = 12 – 19 % → $EMR_{oil\ \&\ gas}$ = 1 – 25 %

4. Two-box model

Well-mixed hemispheres:

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

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



X_N, X_S – hemispheric mean XCH_4 or XC_2H_6 (ppb)
E_N, E_S – hemispheric emissions (ppb yr^{-1})
τ_{ex} – interhemisph. exchange (yr)
λ – atmospheric lifetime (yr)

Lifetimes vs. mixing timescales:

- CH_4: 9 years → zonal and interhemispheric mixing
- C_2H_6: 2.6 months → zonal mixing, no interhemisph. exchange

Ethane emission inventories (~ 80 % in NH):

- Fossil fuel production (oil, gas, coal; Schwietzke et al., 2014)
- Biomass burning (Global Fire Emission Database 1997–2014)
- Biofuel use × emission factor (Andreae and Merlet, 2001)

Methane emissions (~ 70 % in NH):

- Decadal total emissions (IPCC, 2013)

5. Contribution of oil and natural gas emissions

Emission optimization (ethane):

- Simulate ethane increase since 2007 at Zugspitze with two-box model
- Add linear emission increase
- Assume: all "missing" emissions can be attributed to underestimated oil and natural gas emissions

Emission optimization (methane):

- Simulate methane increase since 2007
- Optimize total methane emissions

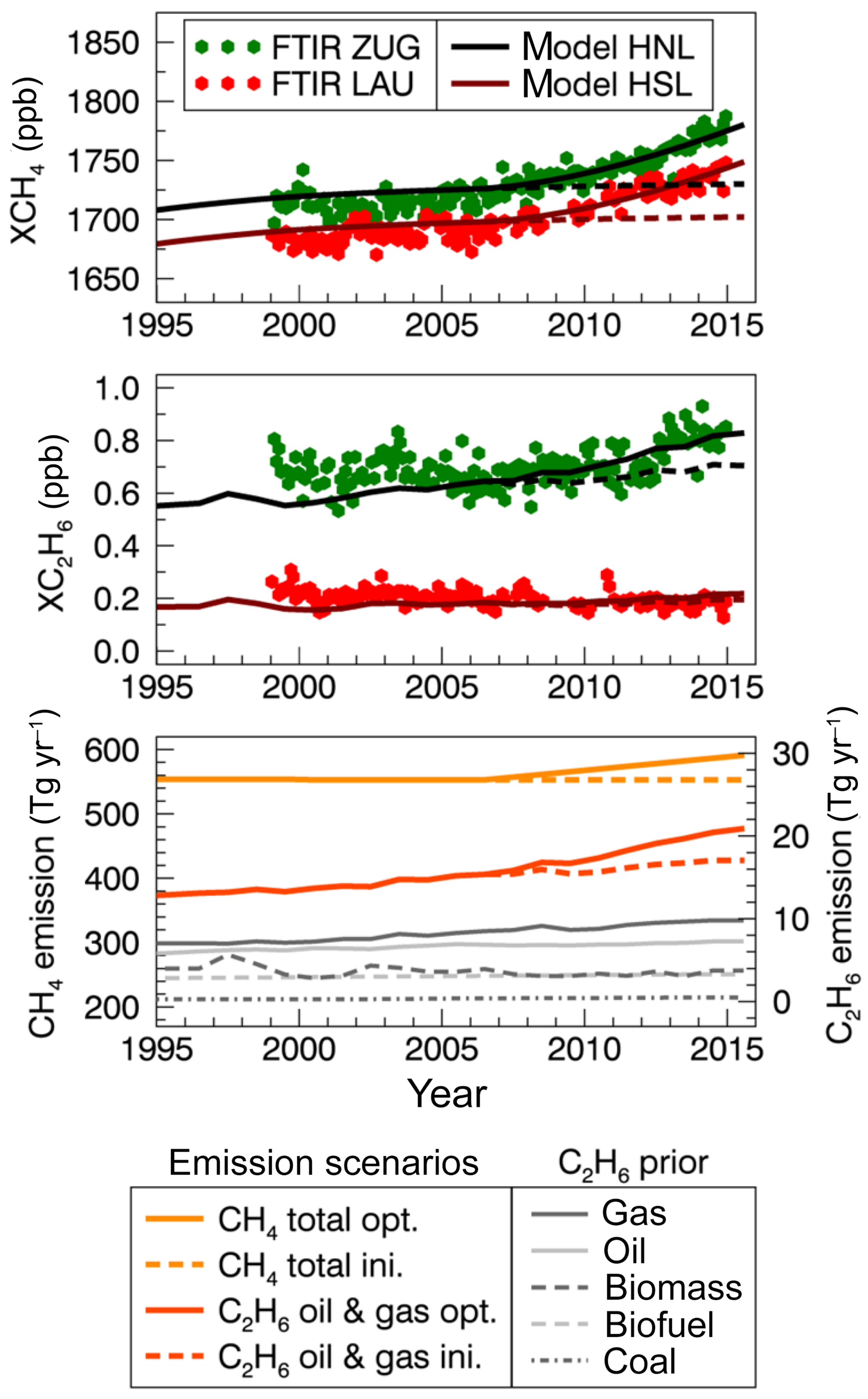
Overall emission change 2007 – 2014:

- Ethane oil & gas emission increase
  $\Delta E_{C2H6,\ oil\ \&\ gas,\ opt.}$ = 1 – 11 Tg yr^{-1}
- Methane total emission increase
  $\Delta E_{CH4,\ total,\ opt.}$ = 24 – 45 Tg yr^{-1}

Contribution of oil & natural gas emissions:

- Use methane-to-ethane ratio (MER) to get associated methane oil & gas emission increase (3 scenarios)
- Quantify contribution $C = \frac{\Delta E_{C2H6,\ oil\ \&\ gas,\ opt.} \times MER}{\Delta E_{CH4,\ total,\ opt.}}$

At least 39 % (18 %, 73 %) contribution of emissions from oil & natural gas production to renewed methane increase (2007 – 2014)



Scenario	MER	C (2.5th – 97.5th %ile)
Oil & Gas	3.3 – 7.6	**39** – 160 %
Oil (limit)	1.7 – 3.3	**18** – 72 %
Gas (limit)	7.6 – 12.1	**73** – 280 %

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