What can the UKCA chemistry-climate model can tell us about ozone and methane?

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Thanks to: Alex Archibald, James Keeble, Ines Heimann and John Pyle

Atmospheric methane is an important greenhouse gas

- Methane has a large (second largest) radiative forcing, making it an important anthropogenic greenhouse gas
  - CO_2 : 1.82 Wm^{-2} for an increase from 278 ppm (Pre-Industrial) to 391 ppm (Present-Day)
  - CH_4 : 0.48 Wm^{-2} [AR5] for an increase of 722 ppb to 1803 ppb (PI-PD)
  - O_3 : 0.4 (± 0.2) Wm^{-2} for an increase of 10 ppb? to 50 ppb (PI ozone uncertain

- A large Global Warming Potential – 28 on a 100-year horizon (per-molecule w.r.t. CO_2)
- Strong sources – 585 Tg CH_4 per year, with strong chemical sinks. Lifetime of 10 years
- Methane oxidation leads to ozone and water vapour – both greenhouse gases – with methane an important source of stratospheric water vapor – modifies GWP up to 31 [Prather and Holmes, 2013].

Sources	Wetlands	Fossile fuels gas and coal	Termites	Ruminants	Rice	Waste landfill	Biomass burning
Tg CH_4 per year	177-284	85-105	2-22	87-94	33-40	67-90	32-39

Sinks	Tropospheric OH	Stratospheric loss	Tropospheric Cl	Methanotrophs
Tg CH_4 per year	454-617	40	13-37	9-47
Lifetime*	10 years	120 years	160 years	160 years

Questions for the study

- How do CH_4 and OH sources/sinks affect CH_4 concentration?
- How do they interact?
- What effect do these interactions have in a CCM such as UKCA?
- How large are these interactions?
- How do they evolve in the future?

“Using global and tropospheric statistics, we demonstrate that the decrease in CO abundance of about 20% (at the global scale) in 12 years has a significant impact on overall CO-OH-CH4 coupled system. “ [Gaubert, 2017].

Feedbacks in the methane system – different visualisations

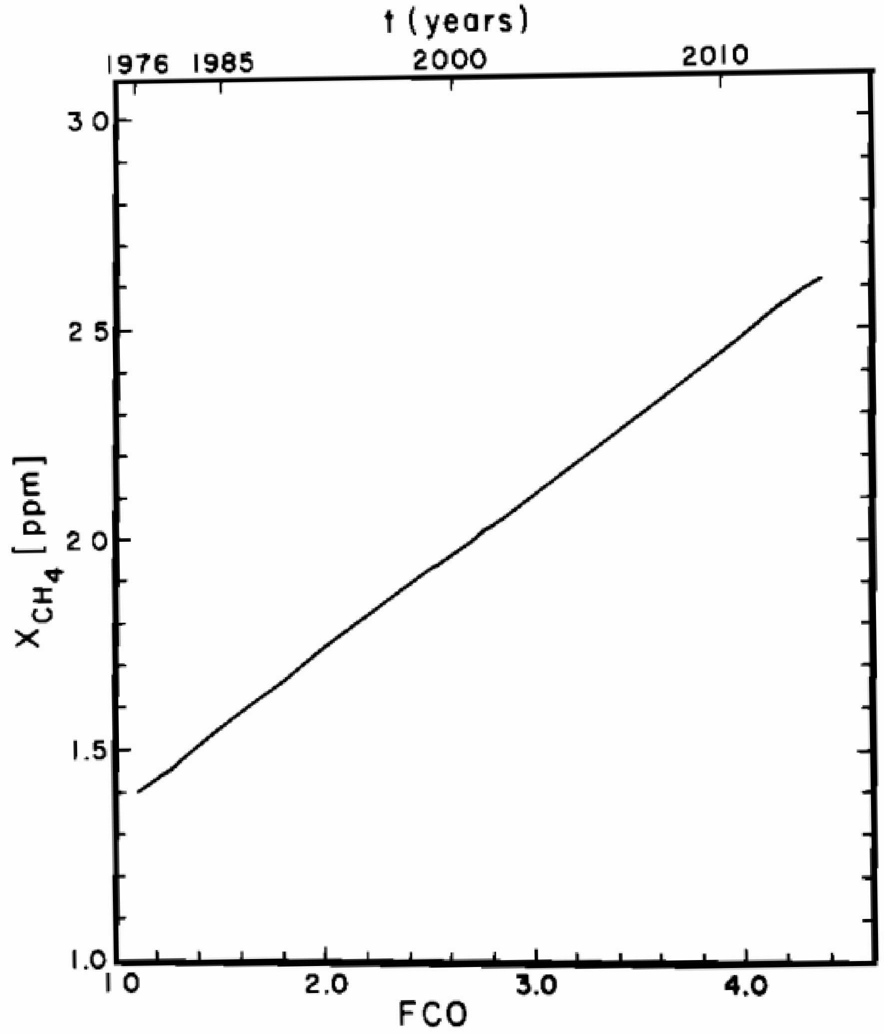


Fig. 1. The dependence of X_{CH_4}, the equilibrium CH_4 abundance, upon *FCO*, the non-CH_4 CO source strength, and upon time, where we assumed that $FCO = 3 \times 10^{10} + 8 \times 10^{10}(1.045)^{t-1976}$ cm^{-2} s^{-1}; i.e., the anthropogenic production rate is presently 8×10^{10} cm^{-2} s^{-1} and is increasing at an annual rate of 4.5%.

Chameides, Liu, Ciccerone, 1977

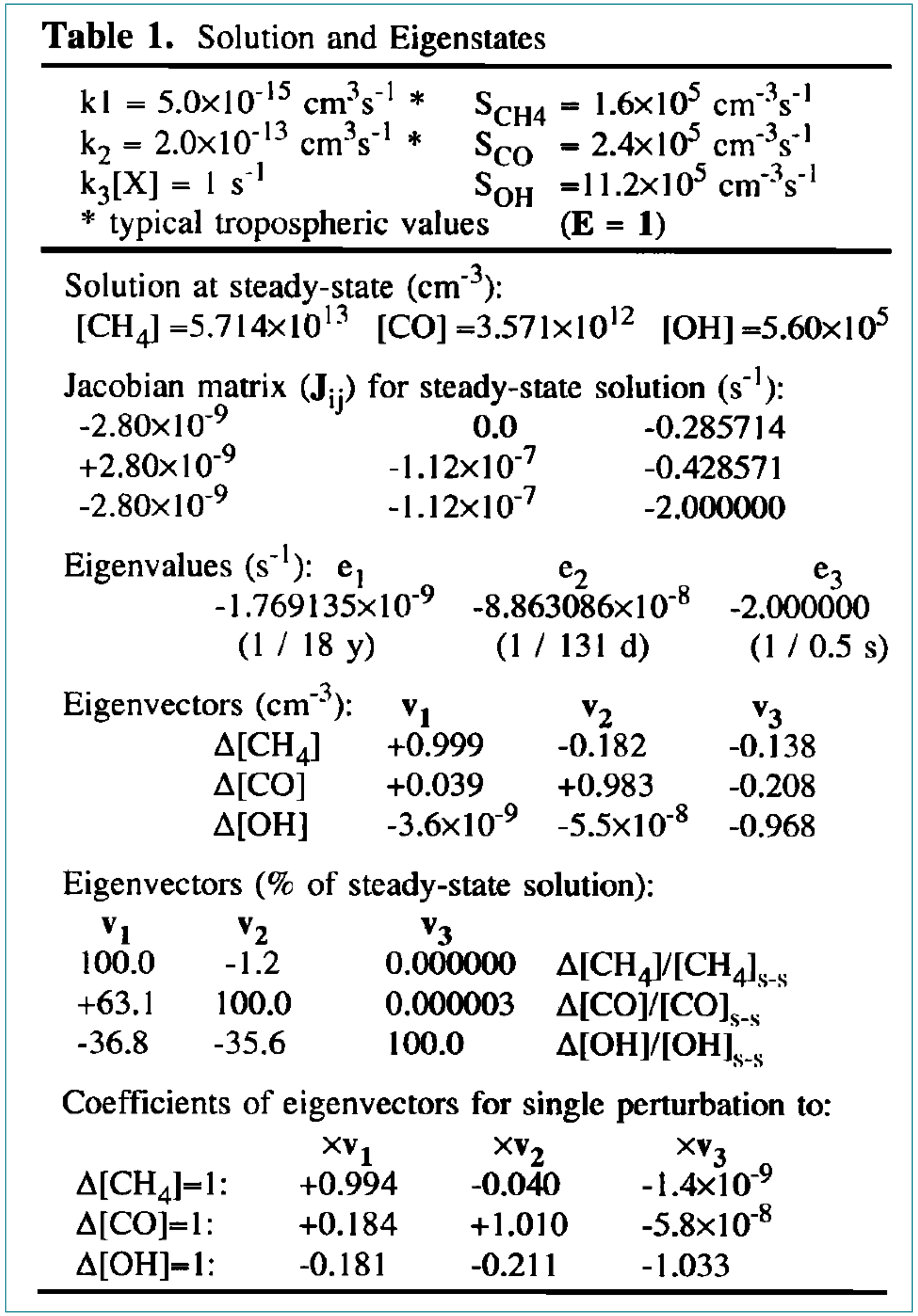


Table 1. Solution and Eigenstates

$k1 = 5.0\times10^{-15}$ cm^3s^{-1} * $S_{CH4} = 1.6\times10^5$ $cm^{-3}s^{-1}$
$k_2 = 2.0\times10^{-13}$ cm^3s^{-1} * $S_{CO} = 2.4\times10^5$ $cm^{-3}s^{-1}$
$k_3[X] = 1$ s^{-1} $S_{OH} = 11.2\times10^5$ $cm^{-3}s^{-1}$
* typical tropospheric values **(E = 1)**

Solution at steady-state (cm^{-3}):
$[CH_4] = 5.714\times10^{13}$ $[CO] = 3.571\times10^{12}$ $[OH] = 5.60\times10^5$

Jacobian matrix (**J**$_{ij}$) for steady-state solution (s^{-1}):

-2.80×10^{-9}	0.0	-0.285714
$+2.80\times10^{-9}$	-1.12×10^{-7}	-0.428571
-2.80×10^{-9}	-1.12×10^{-7}	-2.000000

Eigenvalues (s^{-1}):	e_1	e_2	e_3
	-1.769135×10^{-9}	-8.863086×10^{-8}	-2.000000
	(1 / 18 y)	(1 / 131 d)	(1 / 0.5 s)

Eigenvectors (cm^{-3}):	$\mathbf{v}_1$	$\mathbf{v}_2$	$\mathbf{v}_3$
$\Delta[CH_4]$	+0.999	-0.182	-0.138
$\Delta[CO]$	+0.039	+0.983	-0.208
$\Delta[OH]$	-3.6×10^{-9}	-5.5×10^{-8}	-0.968

Eigenvectors (% of steady-state solution):

$\mathbf{v}_1$	$\mathbf{v}_2$	$\mathbf{v}_3$	
100.0	-1.2	0.000000	$\Delta[CH_4]/[CH_4]_{s-s}$
+63.1	100.0	0.000003	$\Delta[CO]/[CO]_{s-s}$
-36.8	-35.6	100.0	$\Delta[OH]/[OH]_{s-s}$

Coefficients of eigenvectors for single perturbation to:

	$\times\mathbf{v}_1$	$\times\mathbf{v}_2$	$\times\mathbf{v}_3$
$\Delta[CH_4]=1$:	+0.994	-0.040	-1.4×10^{-9}
$\Delta[CO]=1$:	+0.184	+1.010	-5.8×10^{-8}
$\Delta[OH]=1$:	-0.181	-0.211	-1.033

Prather, 1994

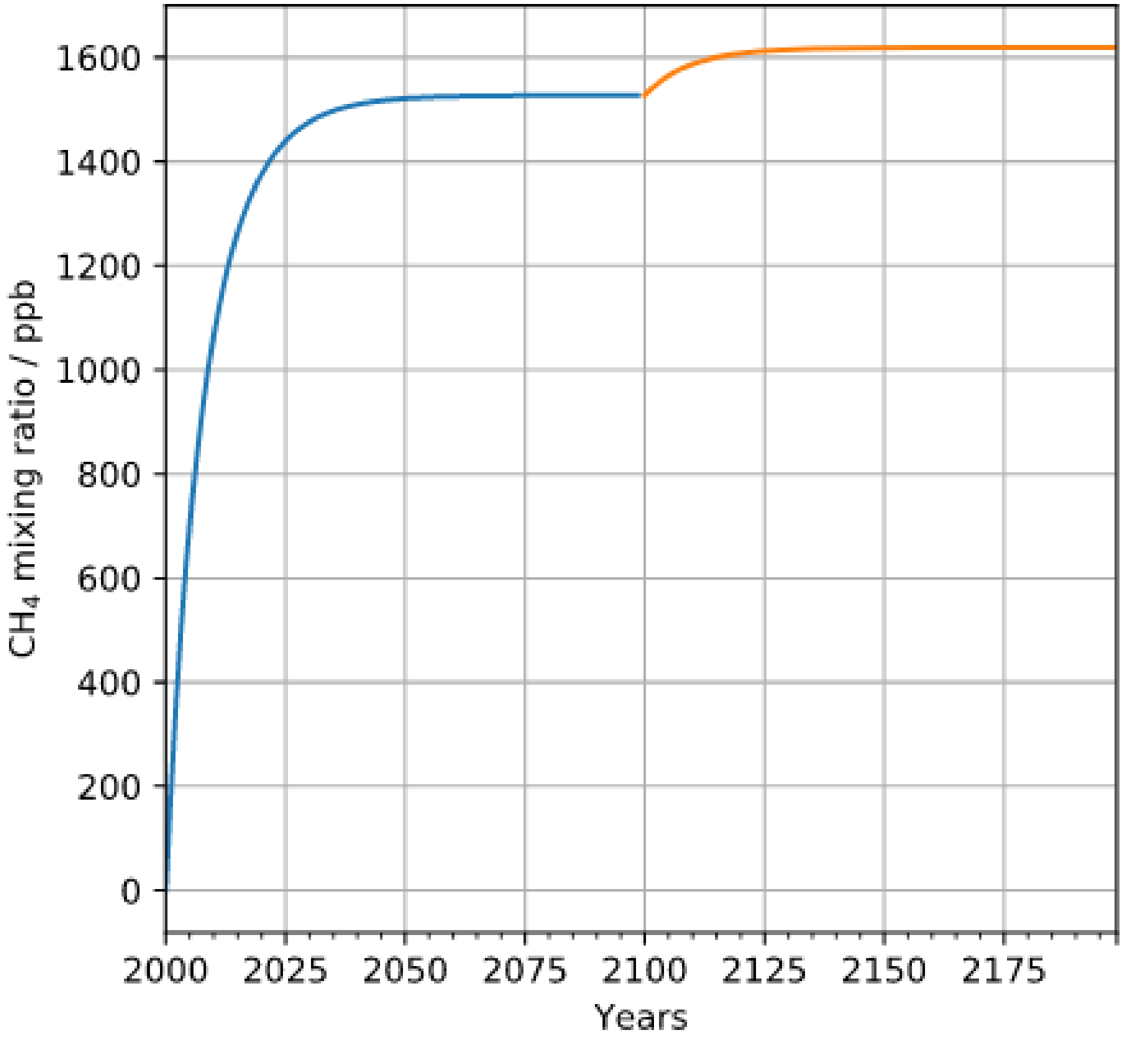
Atmospheric methane has important feedbacks – example model

$$\frac{d[CH_4]}{dt} = S_{CH_4} - k_1[OH][CH_4]$$

$$\frac{d[CO]}{dt} = S_{CO} - k_2[OH][CO] + k_1[OH][CH_4]$$

$$\frac{d[OH]}{dt} = S_{OH} - k_X[OH] - k_2[OH][CO] - k_1\,[OH][CH_4]$$

$k_1 = 5\times10^{-15}$ cm^{3}s^{-1}	S = 585 Tg CH_4 per year
$k_2 = 2\times10^{-13}$ cm^{3}s^{-1}	S = 1370 Tg CO per year
$k_X = 1$s^{-1}	S = 1.1×10^6 cm^{3}s^{-1}



- Initialise the model to zero
- The model spins up to steady state, with a time constant of 10 years.
- Once spun up, increase SCH4 by 5% and re-run to spin up.
- Derive a ‘feedback factor’ based on the increase in concentration per unit increase in emissions.

$$f = \frac{\Delta m/m_0}{\Delta E/E_0} = -\frac{d\ln\tau}{d\ln m}$$

- For these sources and sinks, a change of 5% gives a 7.6 % increase in mixing ratio, so f = 1.52

Atmospheric methane has important feedbacks – example model

$$\frac{d[CH_4]}{dt} = S_{CH_4} - k_1[OH][CH_4]$$

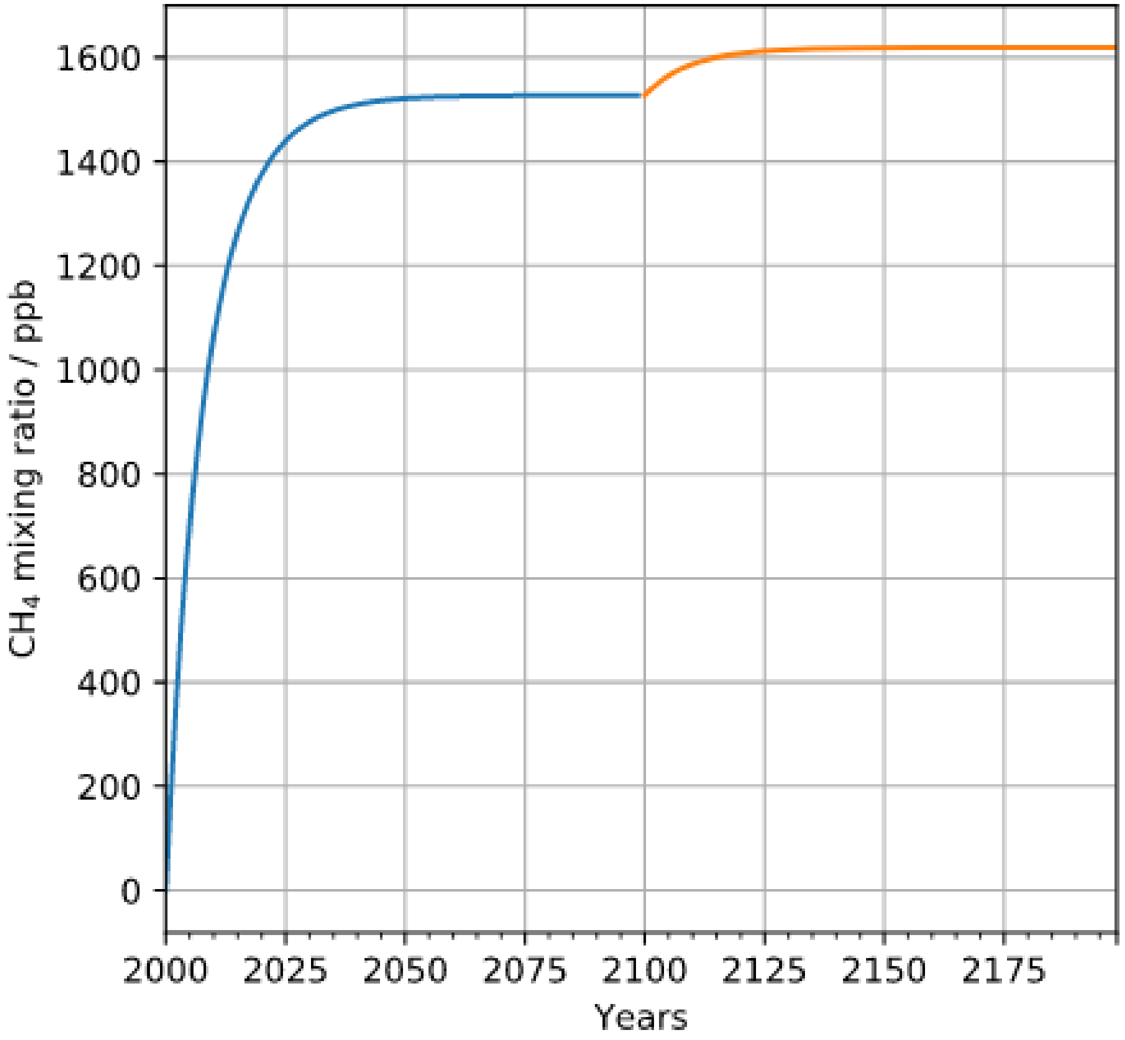
$$\frac{d[CO]}{dt} = S_{CO} - k_2[OH][CO] + k_1[OH][CH_4]$$

$$\frac{d[OH]}{dt} = S_{OH} - k_X[OH] - k_2[OH][CO] - k_1\,[OH][CH_4]$$

k_1= 5×10^{-15} cm^3s^{-1} S = 585 Tg CH_4 per year

k_2 = 2×10^{-13} cm^3s^{-1} S = 1370 Tg CO per year

k_X = $1s^{-1}$ S = 1.1×10^6 cm^3s^{-1}



- Initialise the model to zero
- The model spins up to steady state, with a time constant of 10 years.
- Once spun up, increase SCH4 by 5% and re-run to spin up.
- Derive a ‘feedback factor’ based on the increase in concentration per unit increase in emissions.
- The feedback factor governs both the final concentration and the timescale for equilibration to steady state
- $[CH_4(t)] = (1.05)^f \left\{1 - exp\left(\frac{t}{\tau \cdot f}\right)\right\}$

Atmospheric methane has important feedbacks – example model

$$\frac{d[CH_4]}{dt} = S_{CH_4} - k_1[OH][CH_4]$$

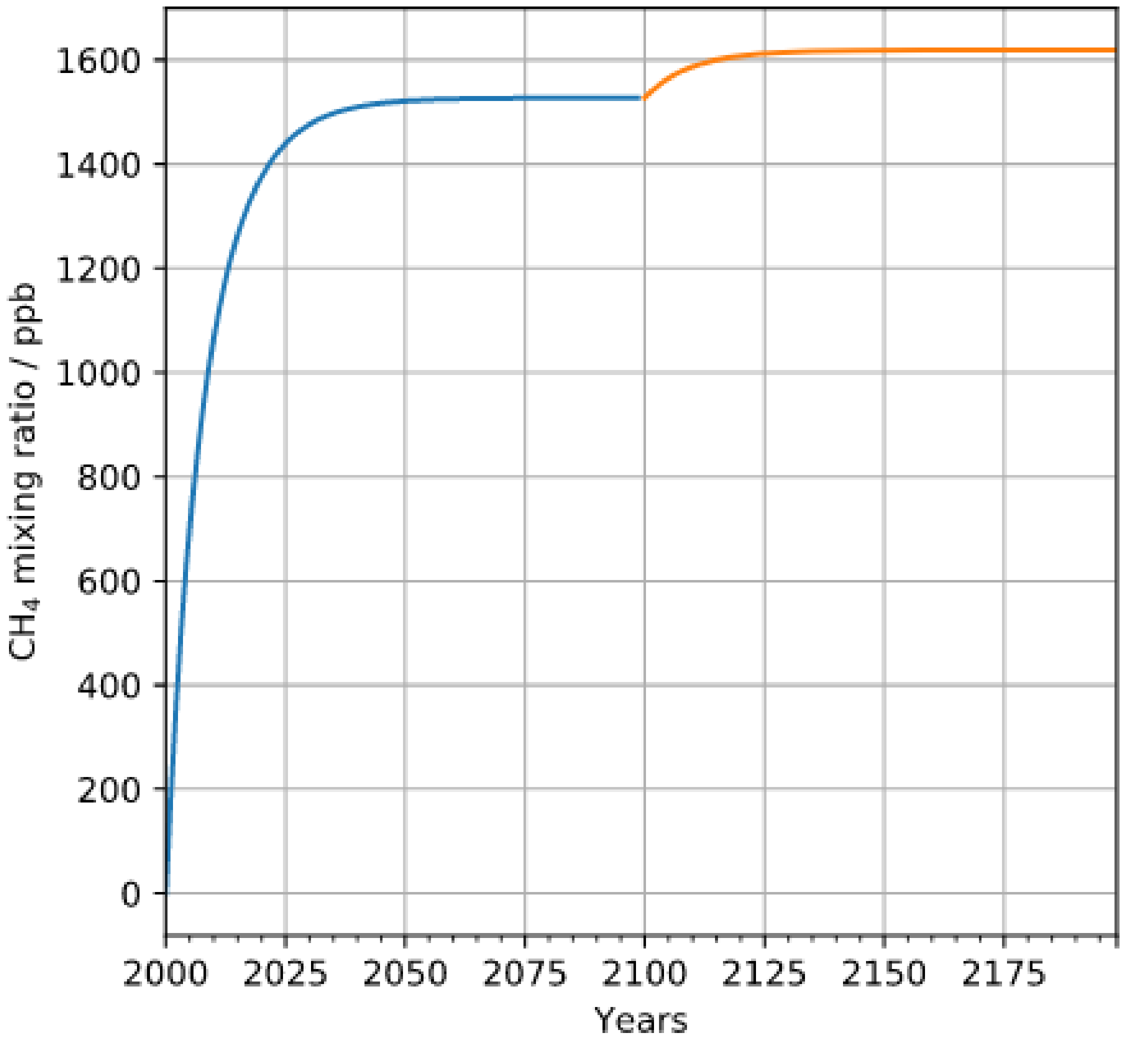
$$\frac{d[CO]}{dt} = S_{CO} - k_2[OH][CO]\boxed{+ k_1[OH][CH_4]}$$

$$\frac{d[OH]}{dt} = S_{OH} - k_X[OH]\boxed{- k_2[OH][CO]}\boxed{- k_1\,[OH][CH_4]}$$

$k_1 = 5\times10^{-15}$ cm^{3}s^{-1} S = 585 Tg CH_4 per year

$k_2 = 2\times10^{-13}$ cm^{3}s^{-1} S = 1370 Tg CO per year

$k_X = 1$s^{-1} S = 1.1×10^6 cm^{3}s^{-1}



- Experiment one: base with above numbers

- Experiments performed to test the strength of these feedbacks in turn

- **E1** – turn off all chemical feedbacks
- **E2** – increase S_{CO} by 50
- **E3** – remove CO production from CH_4
- **E4** - increase S_{OH} by 15%

Experiment two – remove all chemical feedbacks

$$\frac{d[CH_4]}{dt} = S_{CH_4} - k_1[OH][CH_4]$$

$$\frac{d[CO]}{dt} = S_{CO} - k_2[OH][CO]$$

$$\frac{d[OH]}{dt} = S_{OH} - k_X[OH]$$

$k_1 = 5\times10^{-15}\ cm^3s^{-1}$ S = 585 Tg CH_4 per year

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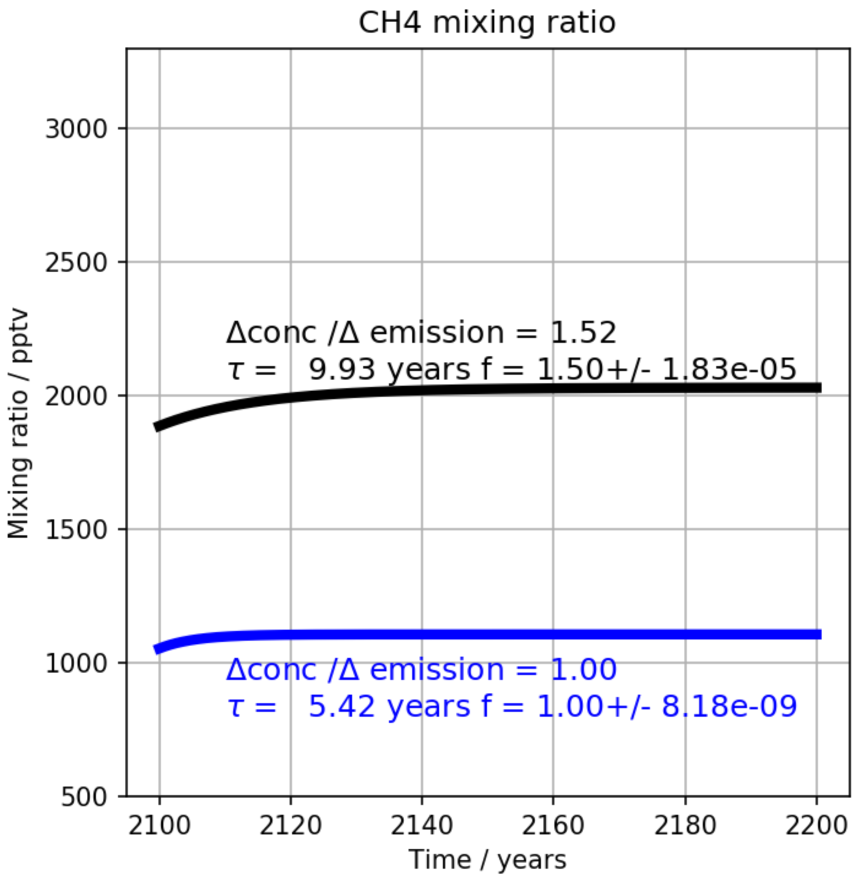
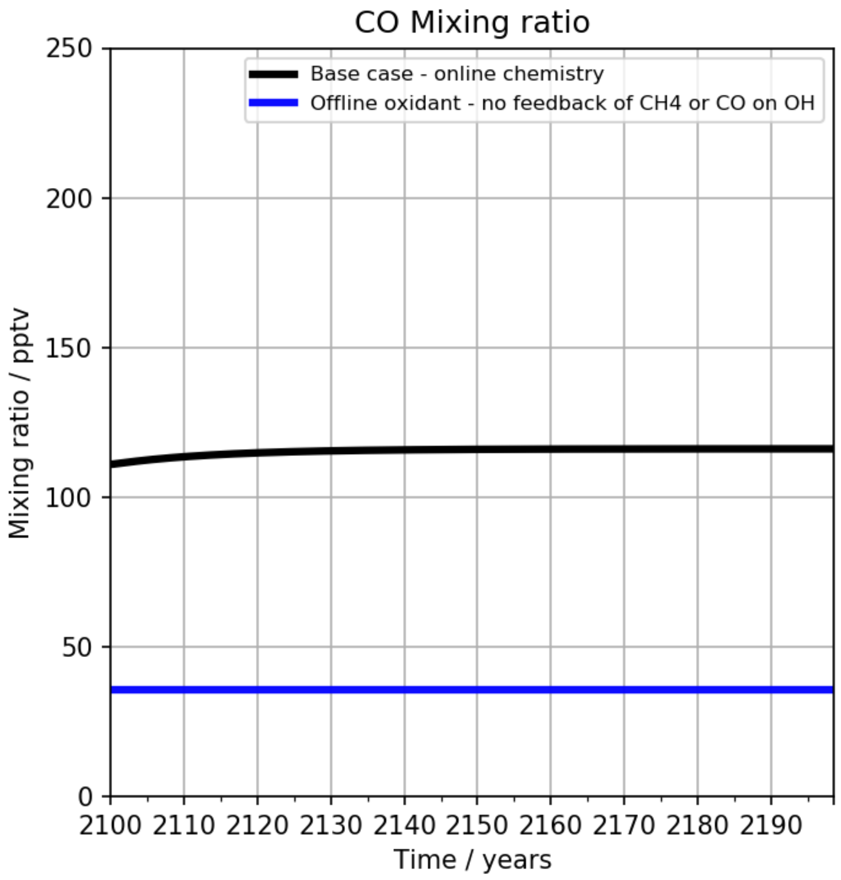
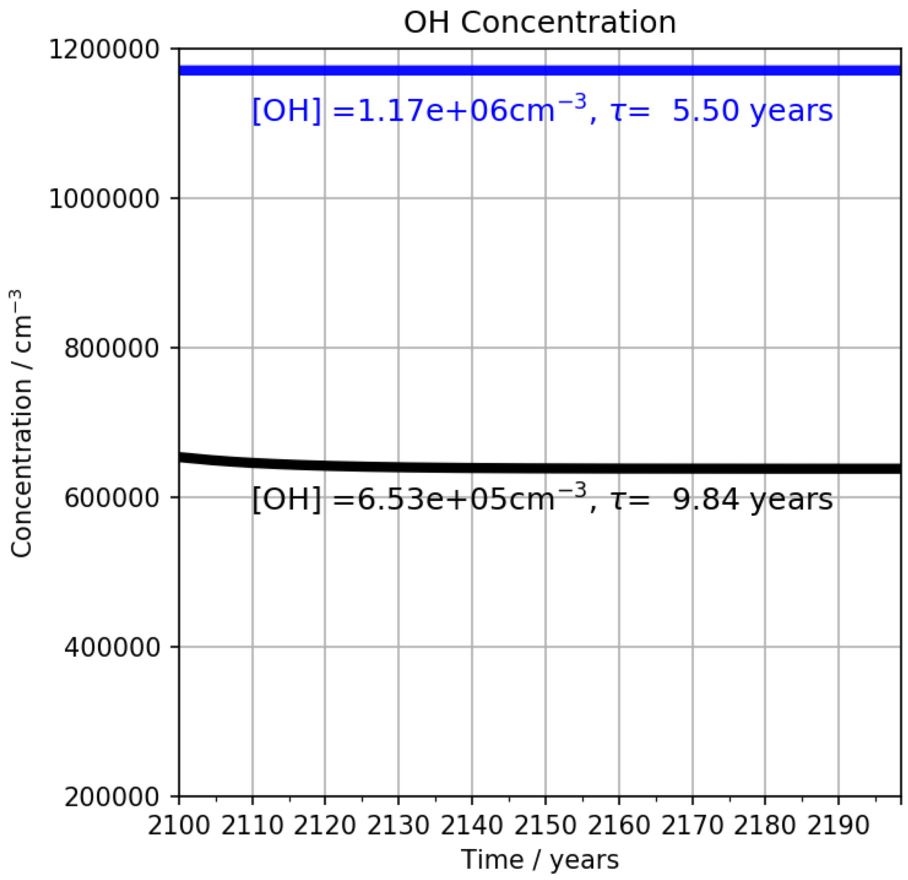
$k_X = 1s^{-1}$ $S = 1.1\times10^{6}\ cm^3s^{-1}$







$$f = 1.00$$

Experiment two – increase CO sources by 50%

$$\frac{d[CH_4]}{dt} = S_{CH_4} - k_1[OH][CH_4]$$

$$\frac{d[CO]}{dt} = \boxed{1.5\,S_{CO}} - k_2[OH][CO] + k_1[OH][CH_4]$$

$$\frac{d[OH]}{dt} = S_{OH} - k_X[OH] - k_2[OH][CO] - k_1\,[OH][CH_4]$$

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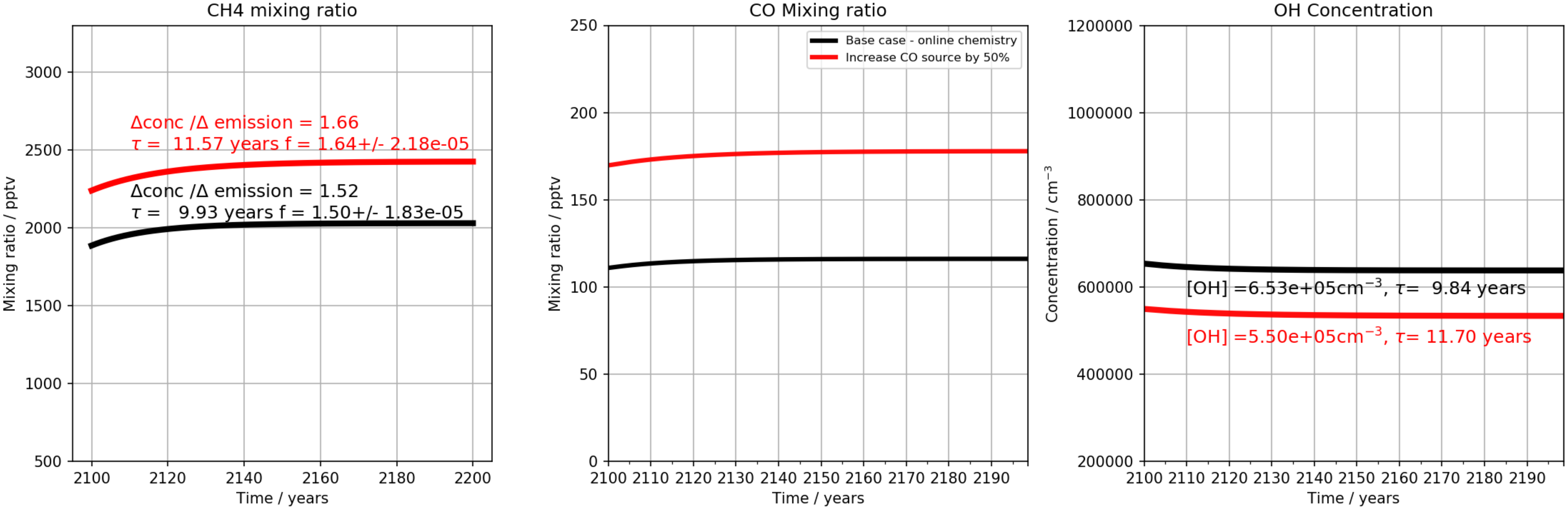

$$f = 1.66$$

Experiment three– decrease the CO production from CH_4 oxidation

$$\frac{d[CH_4]}{dt} = S_{CH_4} - k_1[OH][CH_4]$$

$$\frac{d[CO]}{dt} = \boxed{S_{CO} - k_2[OH][CO]} \qquad \cancel{+ k_1[OH][CH_4]}$$

$$\frac{d[OH]}{dt} = S_{OH} - k_X[OH] - k_2[OH][CO] - k_1[OH][CH_4]$$

$k_1 = 5\times10^{-15}\ cm^3s^{-1}$ $\quad$ S = 585 Tg CH_4 per year

$k_2 = 2\times10^{-13}\ cm^3s^{-1}$ $\quad$ S = 1370 Tg CO per year

$k_X = 1s^{-1}$ $\quad$ $S = 1.1\times10^6\ cm^3s^{-1}$

Simulation	S_{CH_4} Tg(CH_4) yr^{-1}	S_{CO} Tg(CO) yr^{-1}	S_{OH} $cm^{-3}\ s^{-1}$	Feedbacks	τ_{CH_4} years	f
Base	540	1370	1.15×10^6	Full	9.93	1.5
No feedbacks	540	1370	1.15×10^6	None	5.42	1
Inc 1º CO ems	540	2055	1.15×10^6	Full	11.57	1.64
No 2º CO	540	1370	1.15×10^6	No 2º CO	7.95	1.2
Inc S_{OH}	540	1370	1.44×10^6	Full	7.32	1.36

$$f = 1.20$$

Experiment four – increase S_{OH} by 25%

$$\frac{d[CH_4]}{dt} = S_{CH_4} - k_1[OH][CH_4]$$

$$\frac{d[CO]}{dt} = S_{CO} - k_2[OH][CO] + k_1[OH][CH_4]$$

$$\frac{d[OH]}{dt} = \boxed{1.25\ S_{OH}} - k_X[OH] - k_2[OH][CO] - k_1\ [OH][CH_4]$$

$k_1 = 5\times10^{-15}\ cm^3s^{-1}$ S = 585 Tg CH_4 per year

$k_2 = 2\times10^{-13}\ cm^3s^{-1}$ S = 1370 Tg CO per year

$k_X = 1s^{-1}$ S = $1.4\times10^{6}\ cm^3s^{-1}$

Simulation	S_{CH_4} Tg(CH_4) yr^{-1}	S_{CO} Tg(CO) yr^{-1}	S_{OH} $cm^{-3}\ s^{-1}$	Feedbacks	τ_{CH_4} years	f
Base	540	1370	1.15×10^6	Full	9.93	1.5
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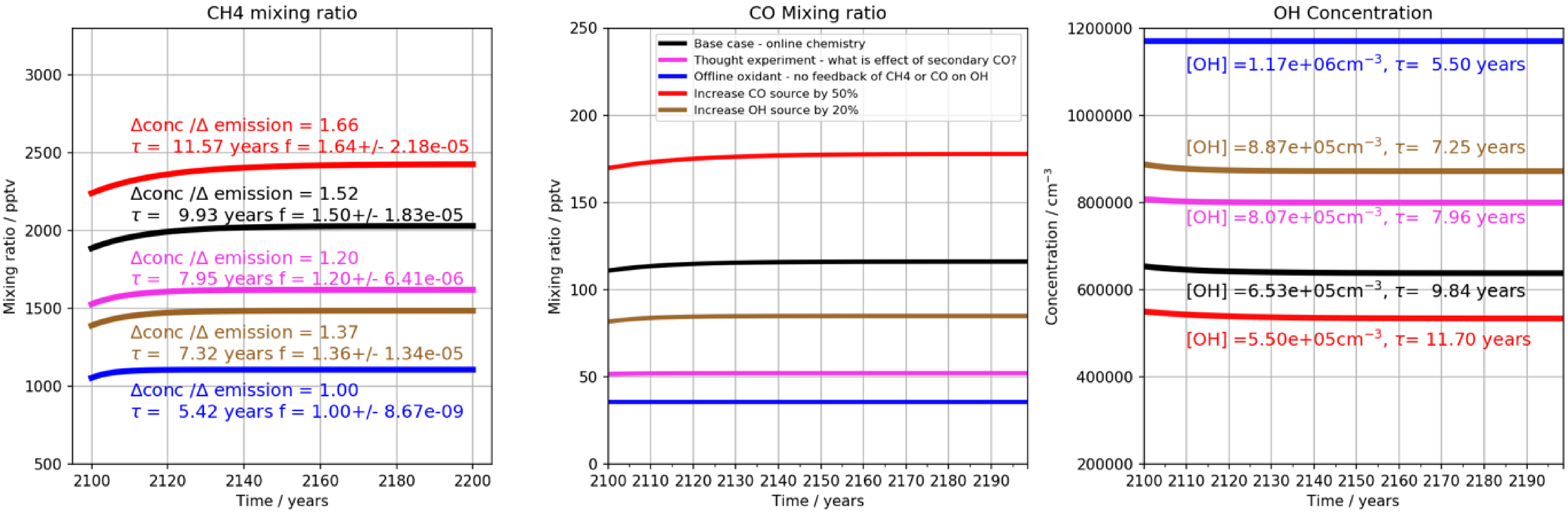
Inc S_{OH}	540	1370	1.44×10^6	Full	7.32	1.36

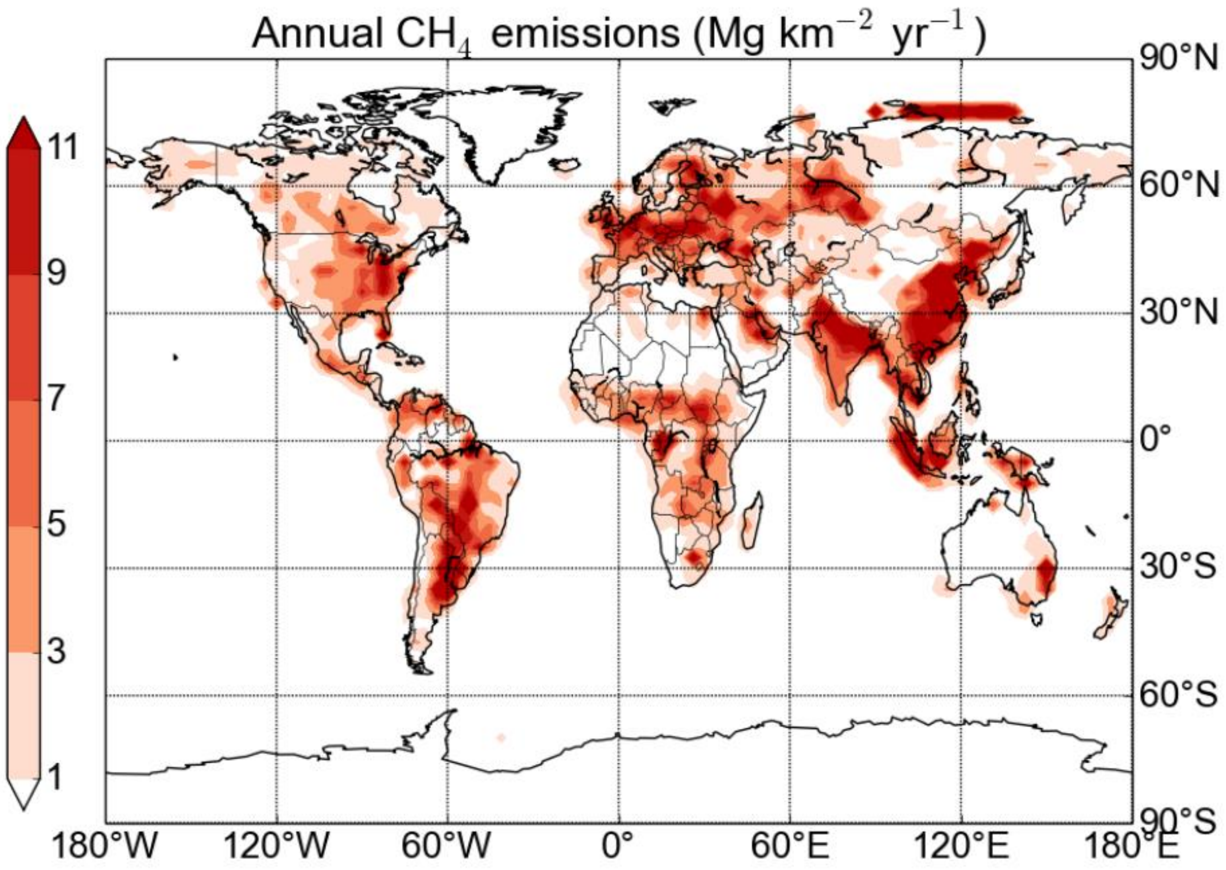
$$f = 1.36$$

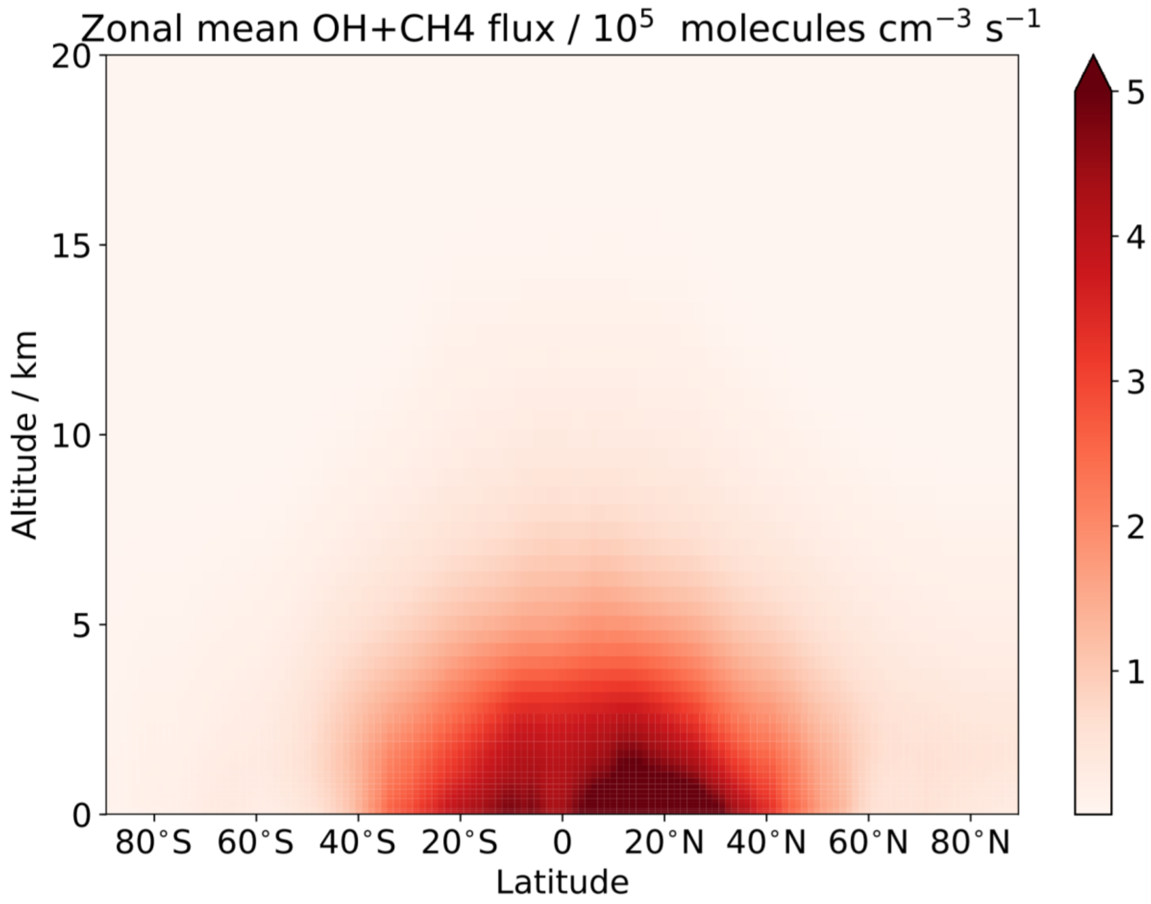
Box modelling to determine feedback factors

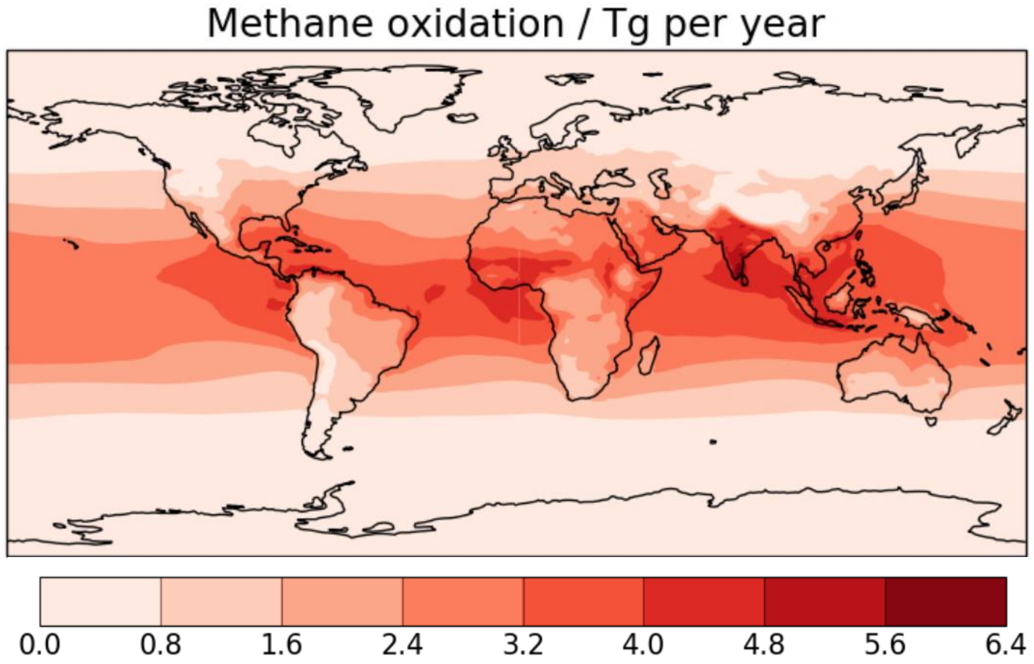
- CH_4-CO-OH system is strongly coupled. Changes to CH_4 source produces changes in CO and OH
- CO production from CH_4 oxidation is coupled via OH to CH_4 concentration and lifetime
- Increasing CO emissions decreases both CH_4 and OH, changes feedback
- Increasing OH source also modifies CH_4 and leads to a decrease in CH4 per unit increase in CH4 emissions (ie decreased sensitivity, lower feedback)
- Both CO and OH sources modify the lifetime of CH_4 and hence its GWP



Methane in UKCA - emissions vs OH sink



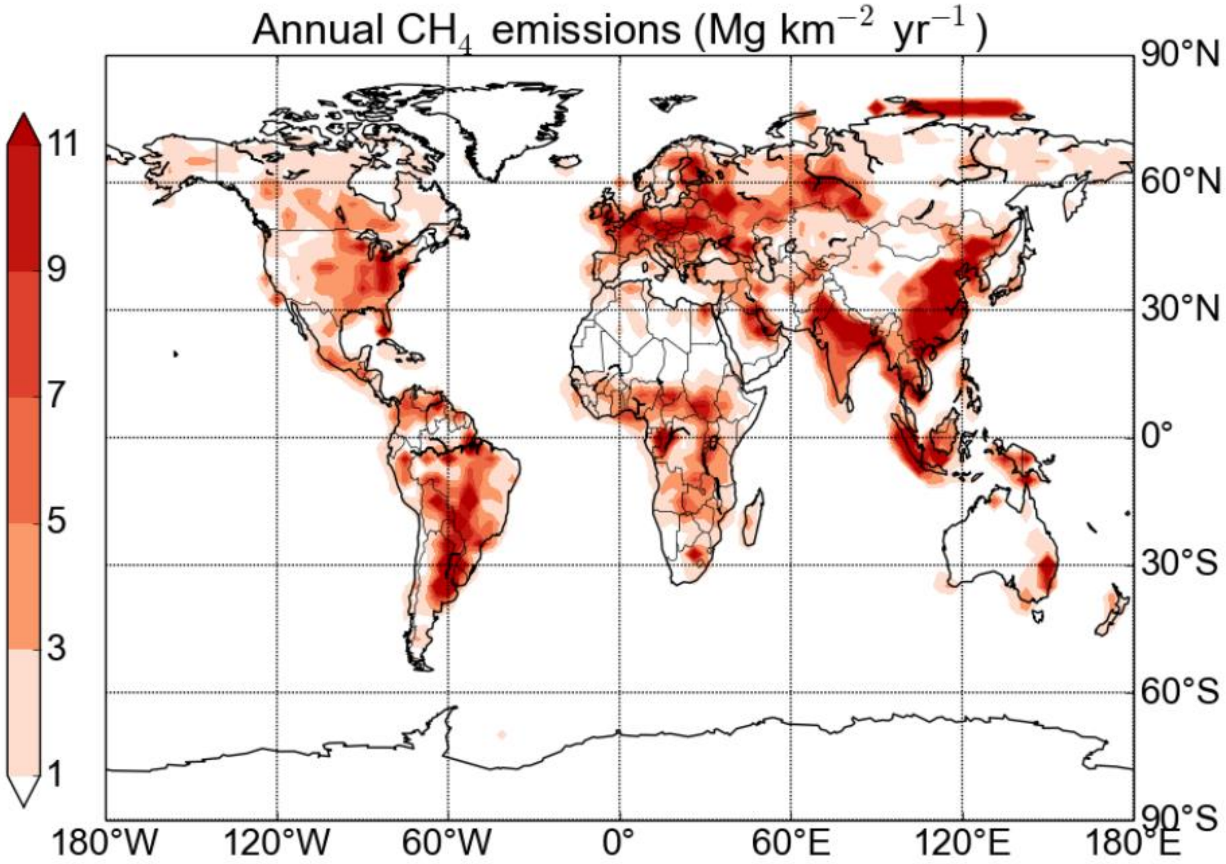


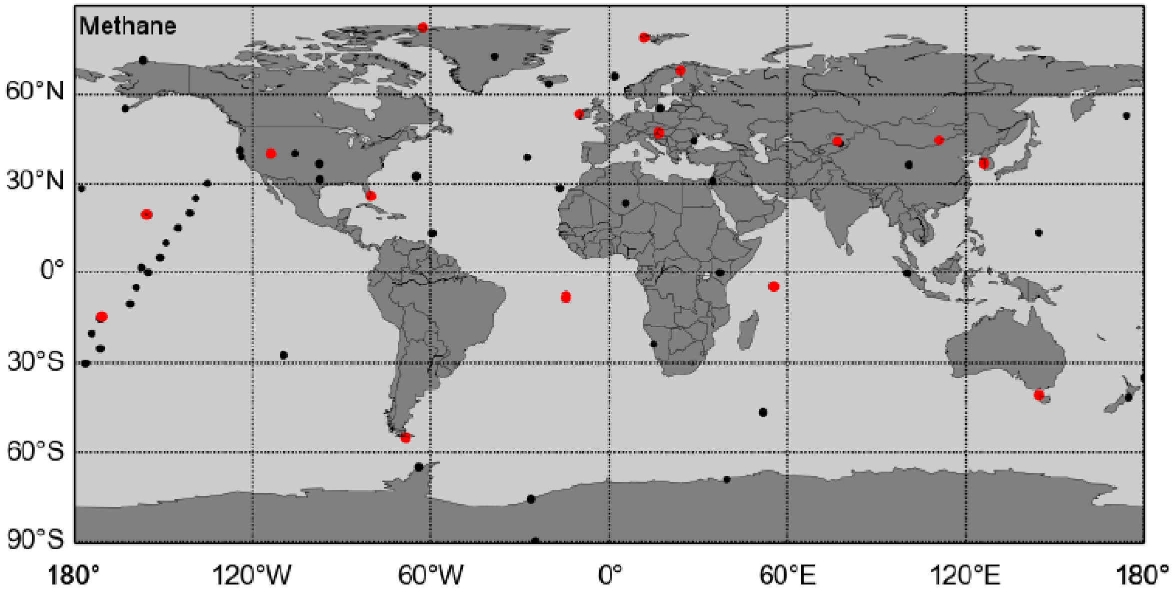


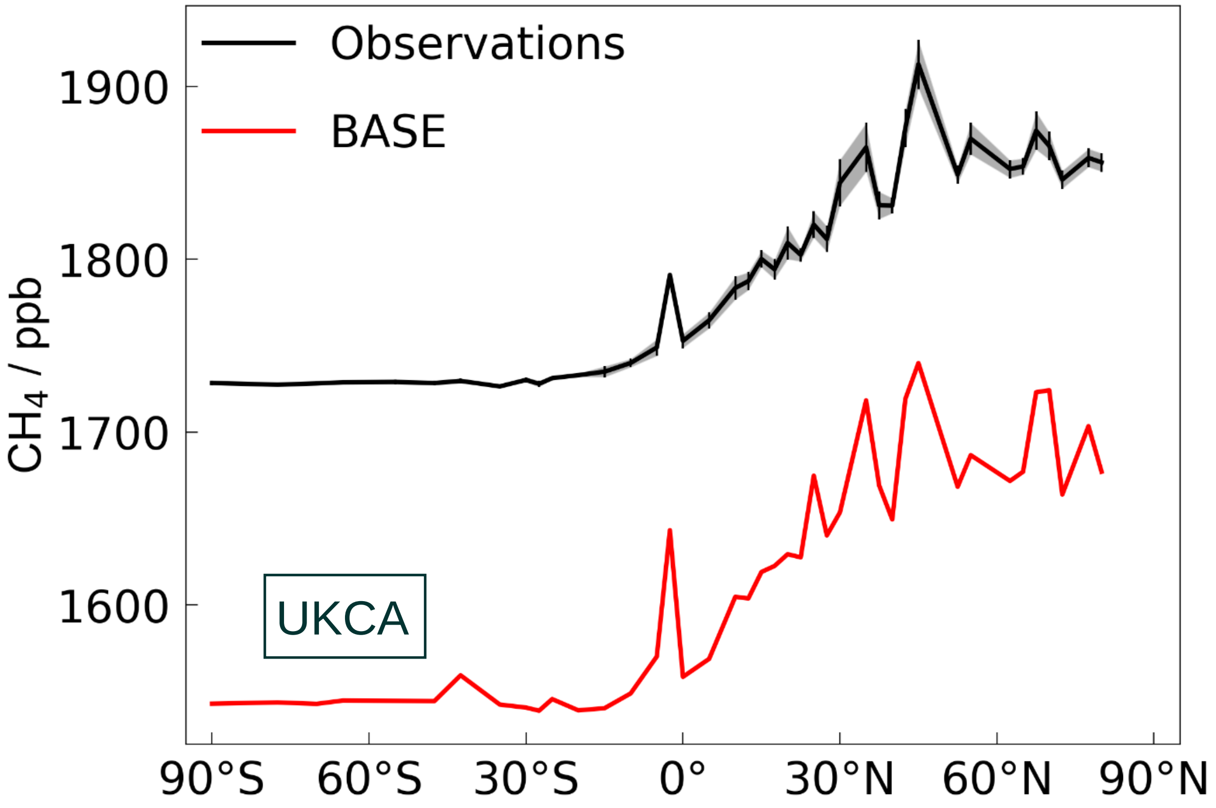
Methane sources are largest in the extra tropics, but oxidation rate is strongly temperature dependent, so peaks where T, humidity and OH high.

Methane in UKCA - comparison with observations

- Using methane emissions derived from EDGAR emissions database.
- Methane concentrations substantially low-biased. **Why**?
- NB latitudinal gradient looks good!
- Are emissions *wrong* (low-biased) ?
- Are the sinks *wrong* – is the OH not correctly represented and high-biased?
    - If OH is too high, are its sinks too low?





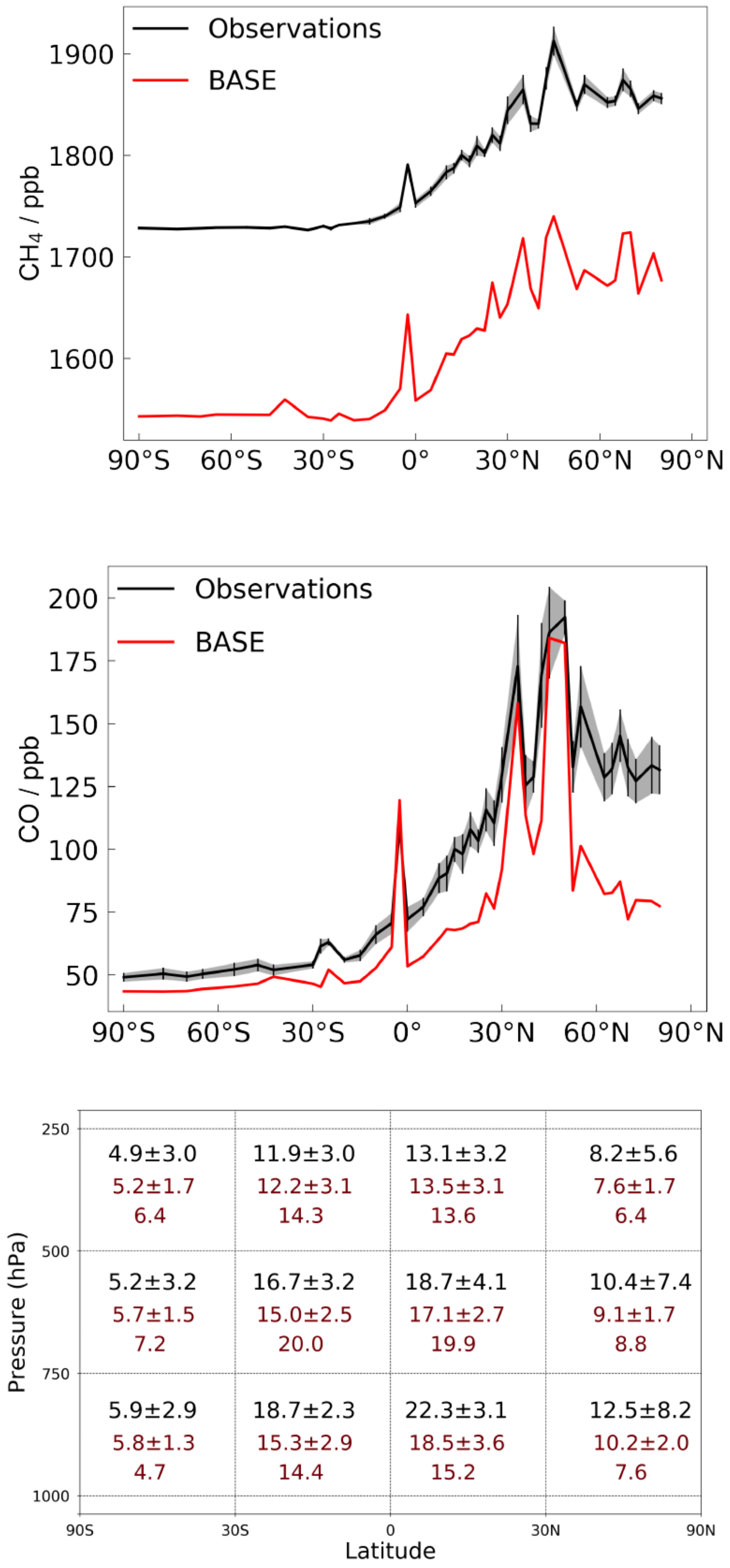


3 sensitivity experiments

1. Our BASE run using methane emissions derived from EDGAR emissions database.
2. A second experiment in which CO emissions are increased everywhere by 50%
3. An experiment in which we use a different emissions dataset with lower emissions in NH midlatitudes higher emissions in tropics.

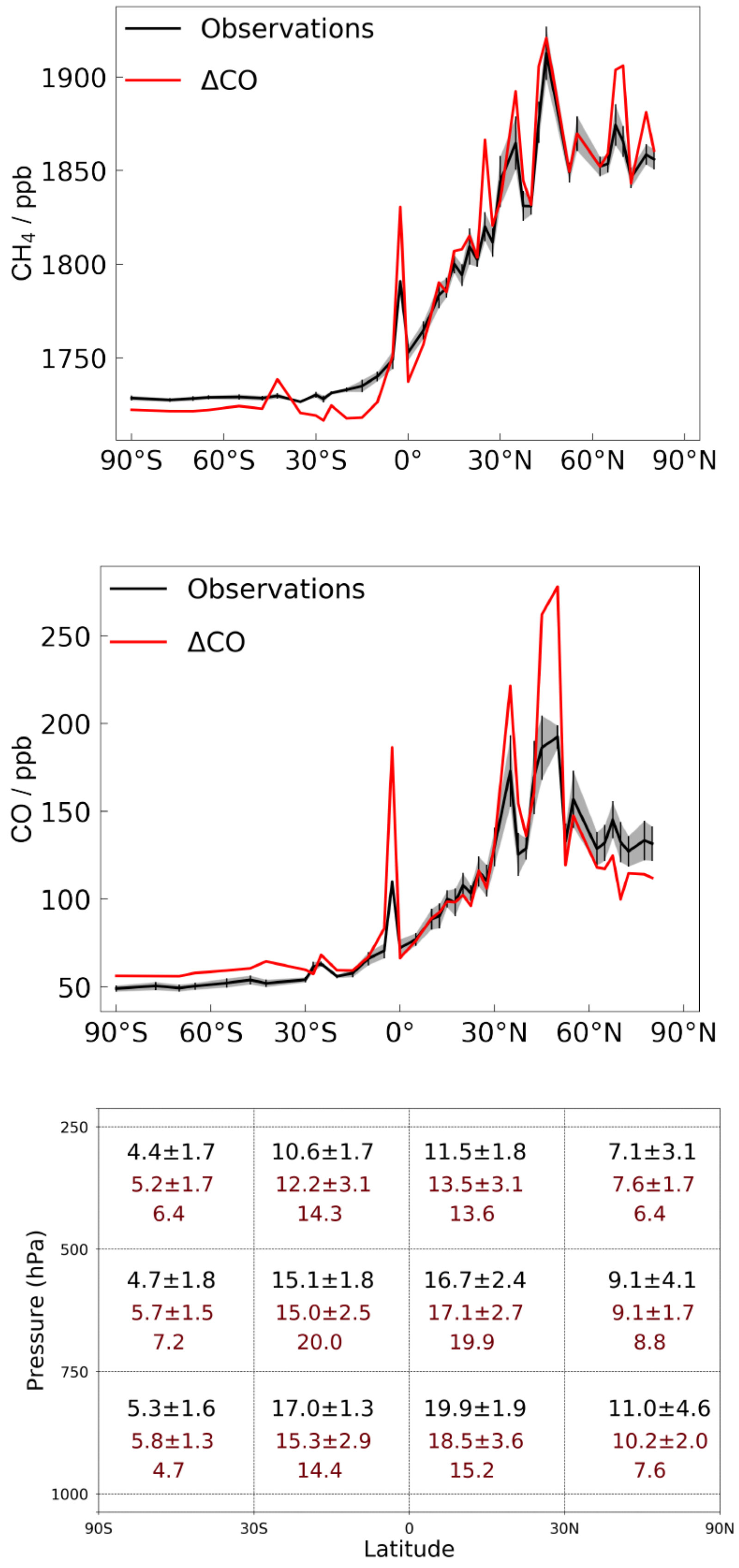
Sensitivity of UKCA to emissions – 3 global experiments

BASE emissions - EDGAR



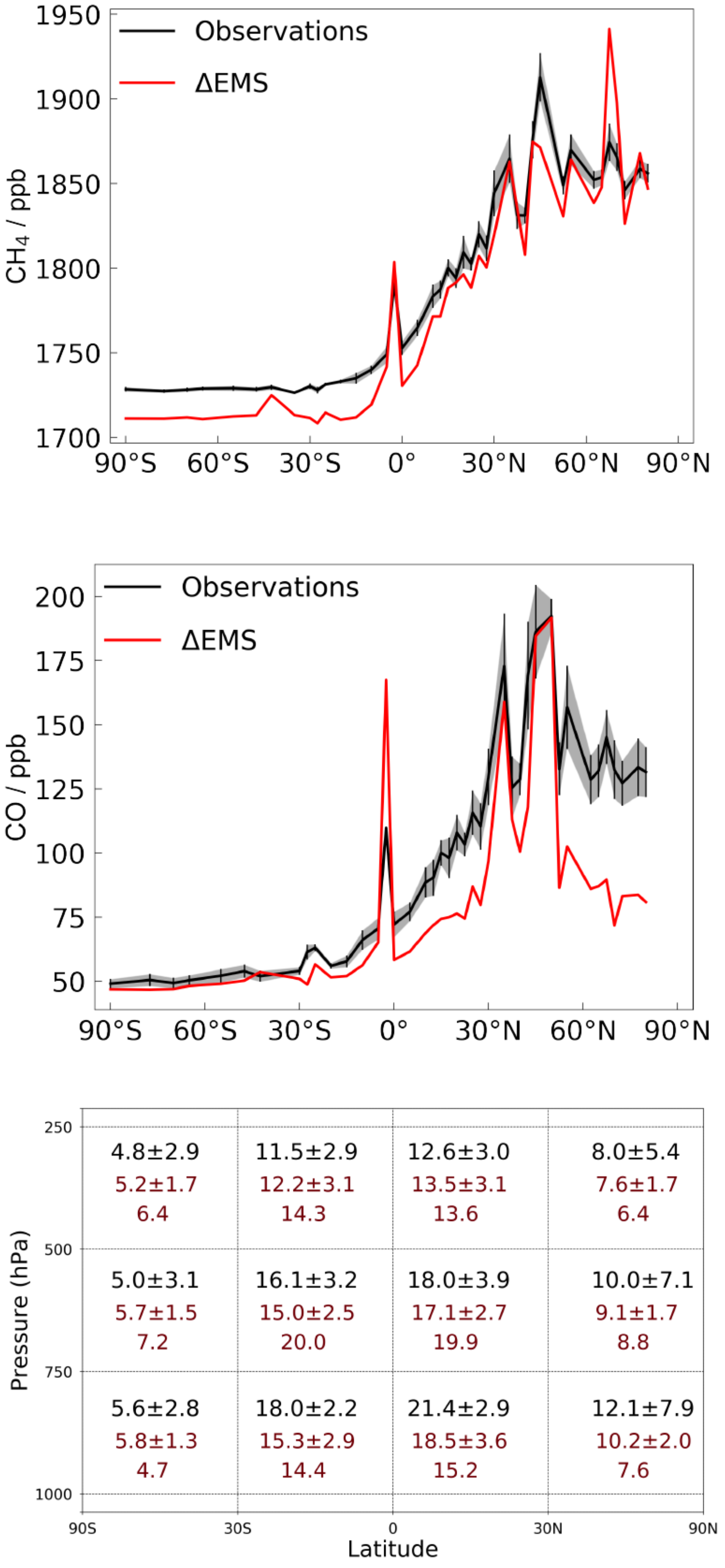
4.9±3.0 5.2±1.7 6.4	11.9±3.0 12.2±3.1 14.3	13.1±3.2 13.5±3.1 13.6	8.2±5.6 7.6±1.7 6.4
5.2±3.2 5.7±1.5 7.2	16.7±3.2 15.0±2.5 20.0	18.7±4.1 17.1±2.7 19.9	10.4±7.4 9.1±1.7 8.8
5.9±2.9 5.8±1.3 4.7	18.7±2.3 15.3±2.9 14.4	22.3±3.1 18.5±3.6 15.2	12.5±8.2 10.2±2.0 7.6

ΔCO – increase CO emissions by 50%



4.4±1.7 5.2±1.7 6.4	10.6±1.7 12.2±3.1 14.3	11.5±1.8 13.5±3.1 13.6	7.1±3.1 7.6±1.7 6.4
4.7±1.8 5.7±1.5 7.2	15.1±1.8 15.0±2.5 20.0	16.7±2.4 17.1±2.7 19.9	9.1±4.1 9.1±1.7 8.8
5.3±1.6 5.8±1.3 4.7	17.0±1.3 15.3±2.9 14.4	19.9±1.9 18.5±3.6 15.2	11.0±4.6 10.2±2.0 7.6

Decrease NH, increase tropical emissions



4.8±2.9 5.2±1.7 6.4	11.5±2.9 12.2±3.1 14.3	12.6±3.0 13.5±3.1 13.6	8.0±5.4 7.6±1.7 6.4
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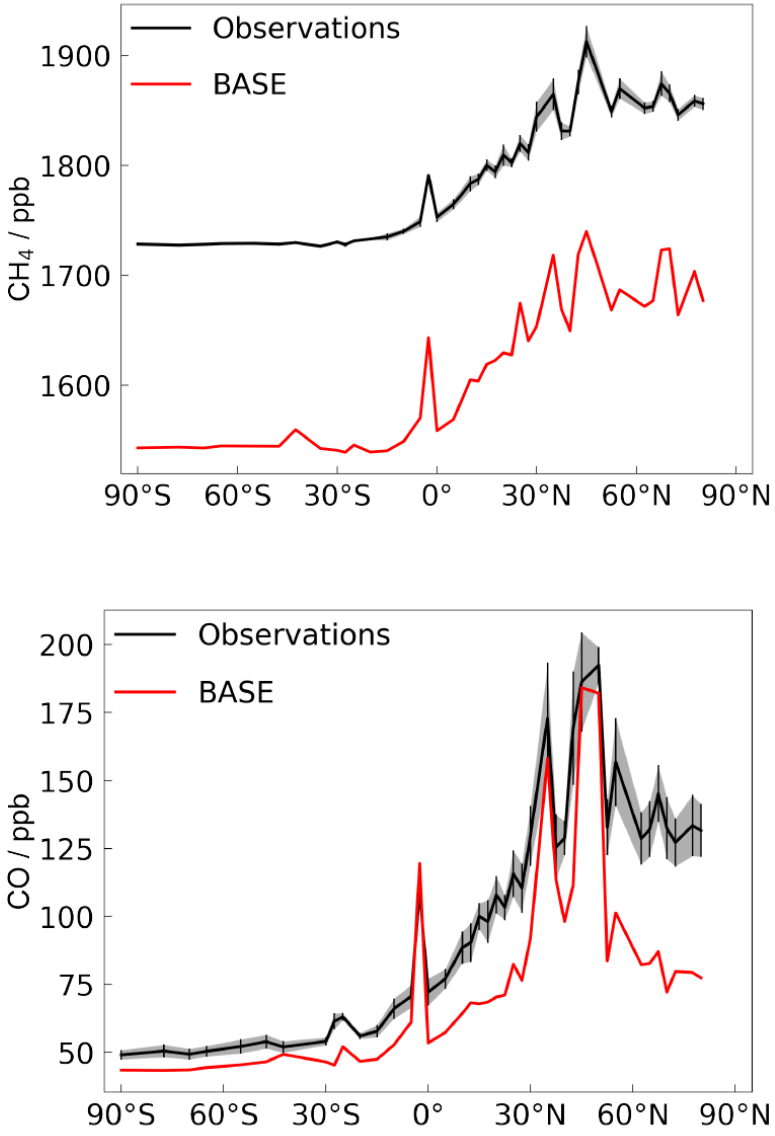
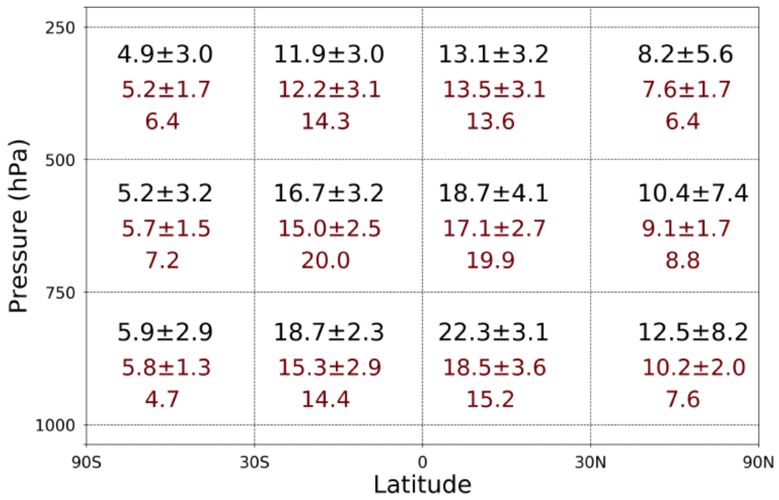
Sensitivity of UKCA to emissions – 3 global experiments

BASE emissions - EDGAR





4.9±3.0 5.2±1.7 6.4	11.9±3.0 12.2±3.1 14.3	13.1±3.2 13.5±3.1 13.6	8.2±5.6 7.6±1.7 6.4
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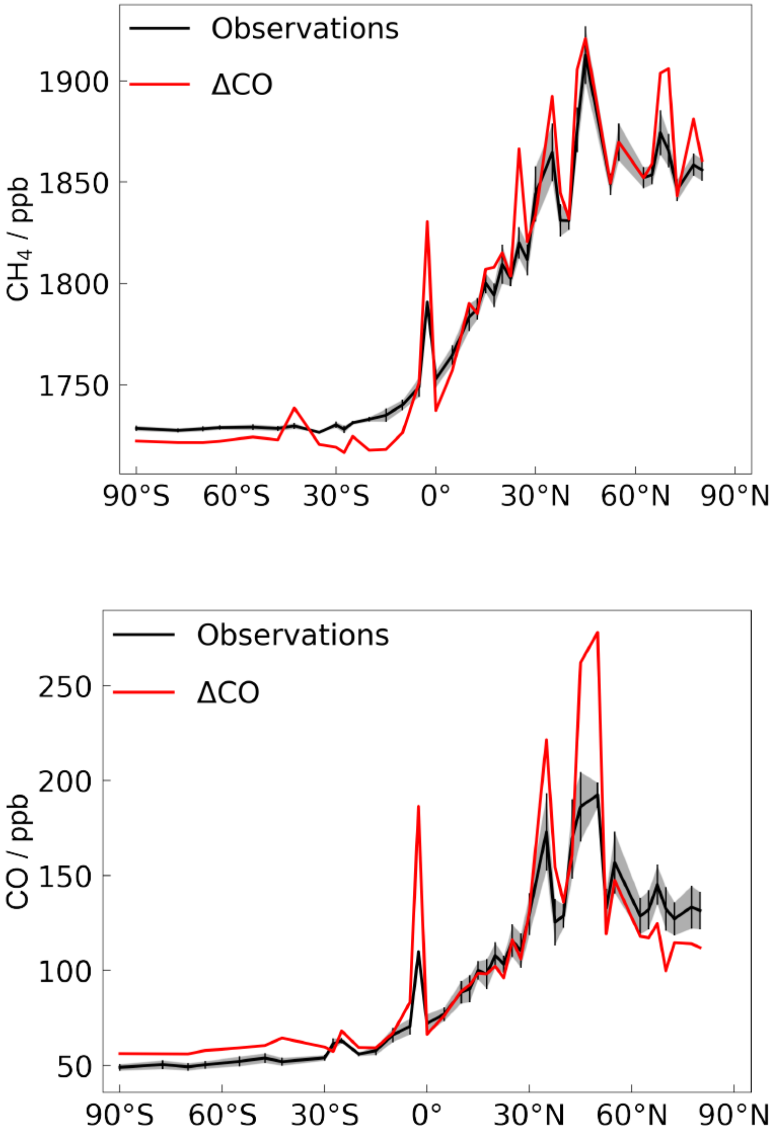
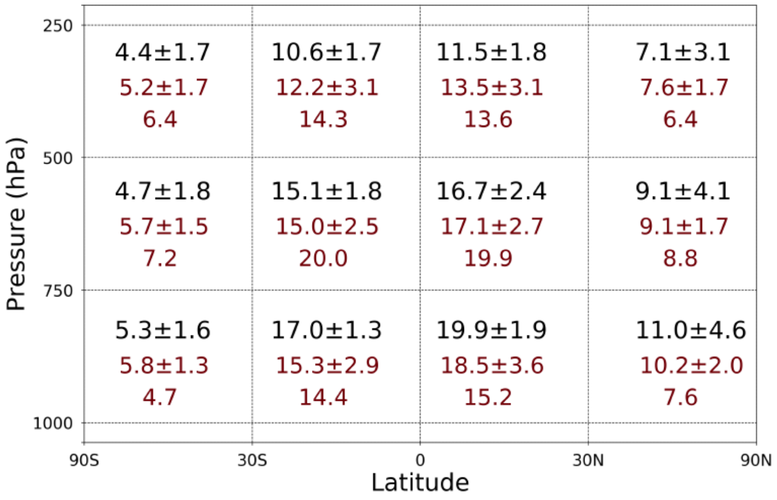
5.9±2.9 5.8±1.3 4.7	18.7±2.3 15.3±2.9 14.4	22.3±3.1 18.5±3.6 15.2	12.5±8.2 10.2±2.0 7.6

ΔCO – increase CO emissions by 50%





4.4±1.7 5.2±1.7 6.4	10.6±1.7 12.2±3.1 14.3	11.5±1.8 13.5±3.1 13.6	7.1±3.1 7.6±1.7 6.4
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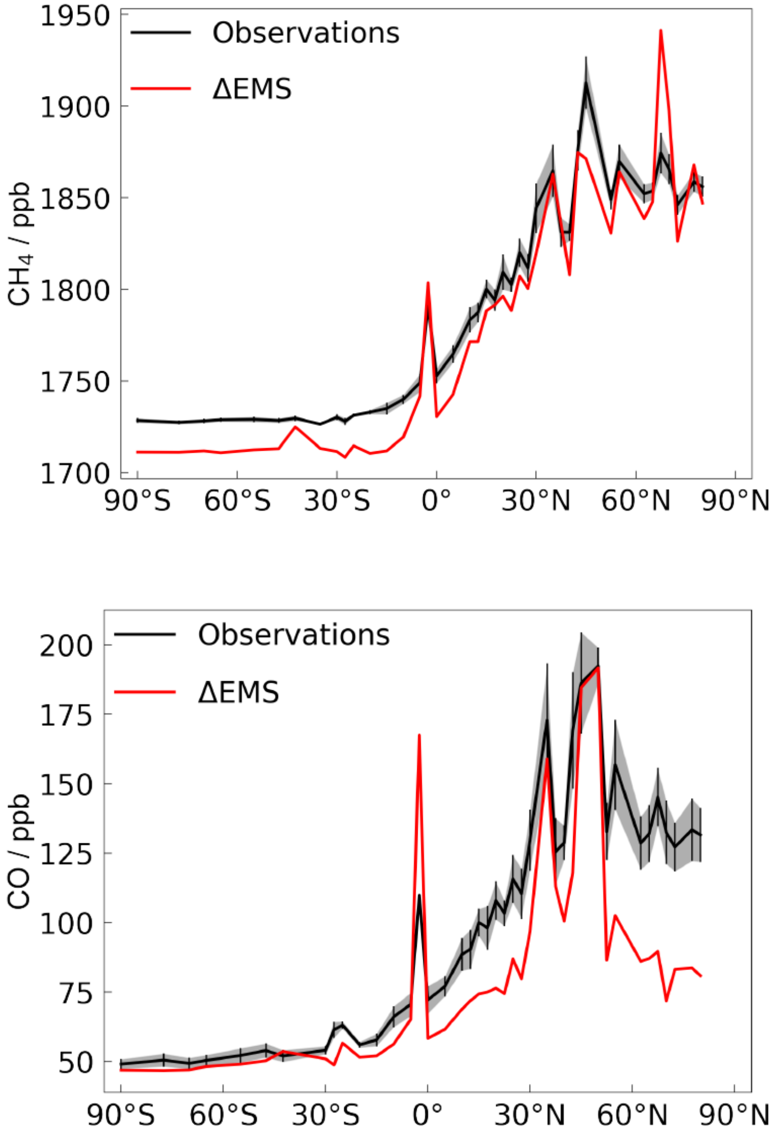
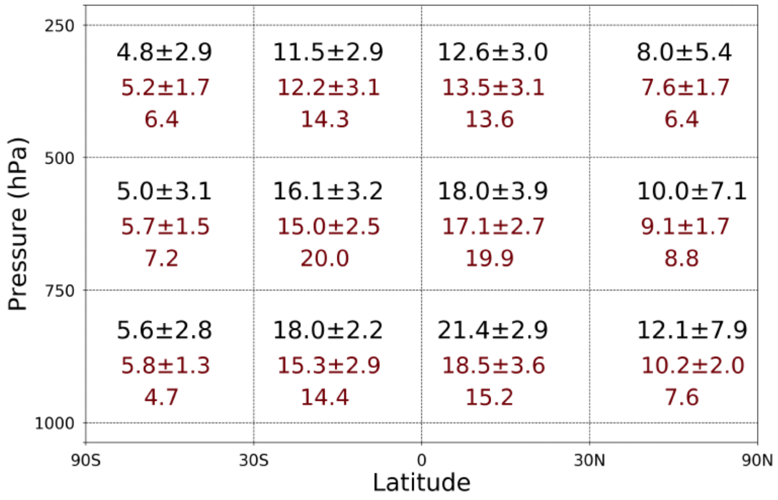
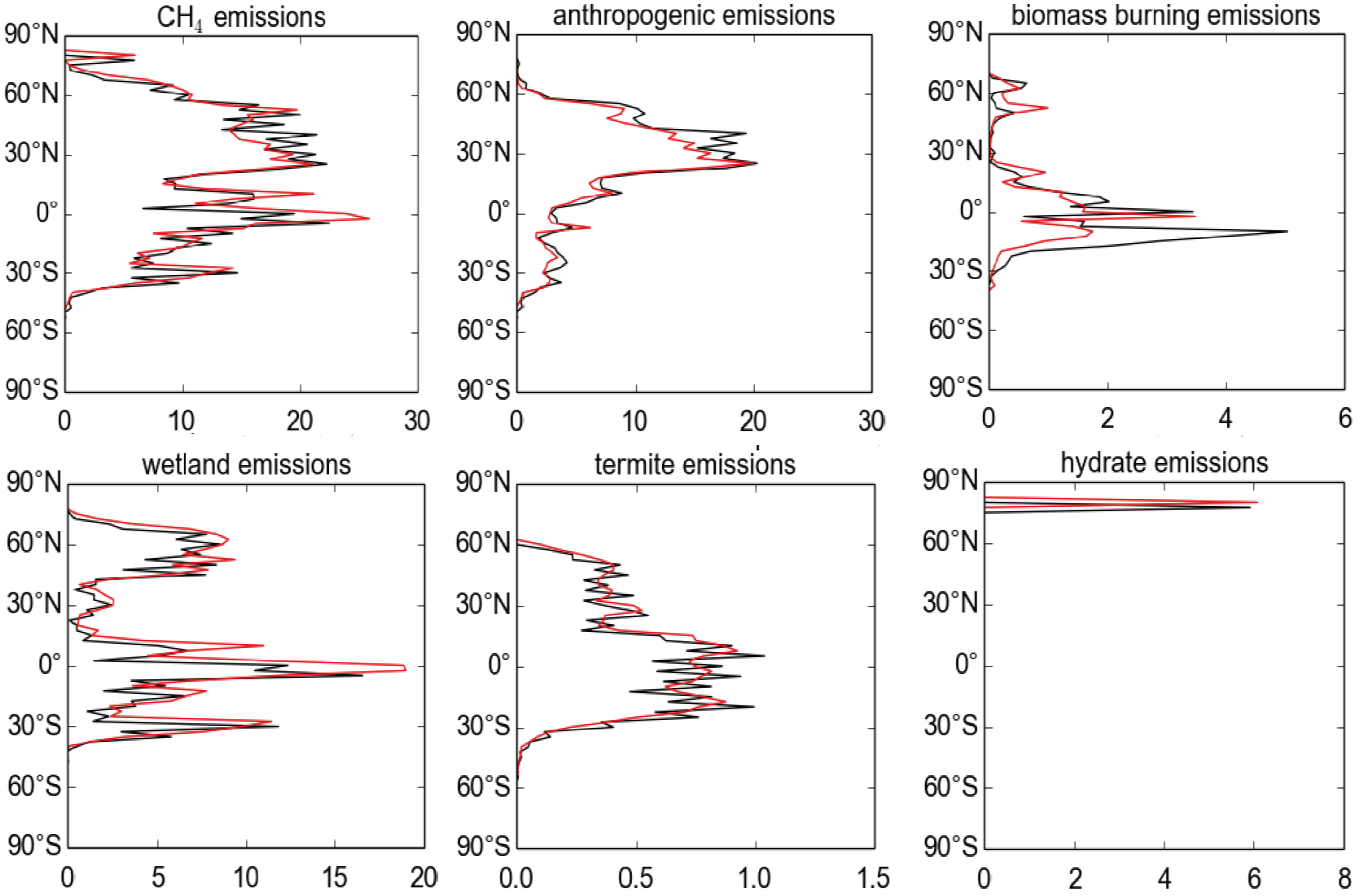




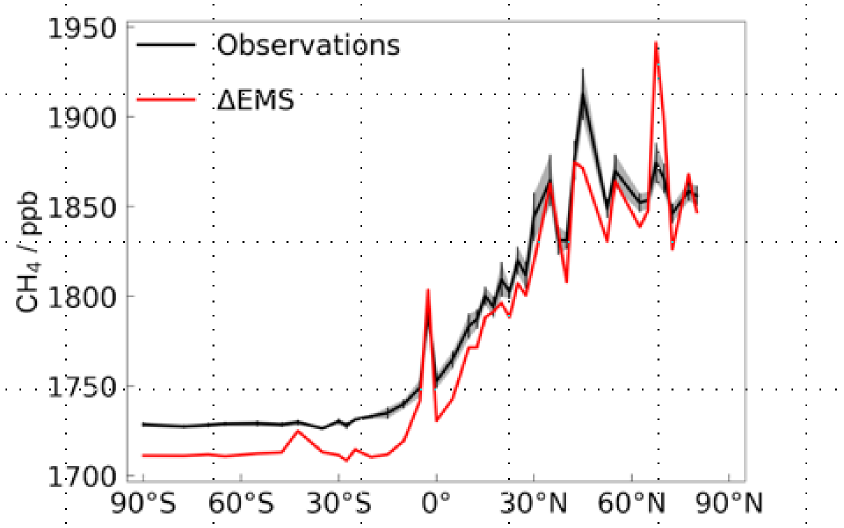
4.8±2.9 5.2±1.7 6.4	11.5±2.9 12.2±3.1 14.3	12.6±3.0 13.5±3.1 13.6	8.0±5.4 7.6±1.7 6.4
5.0±3.1 5.7±1.5 7.2	16.1±3.2 15.0±2.5 20.0	18.0±3.9 17.1±2.7 19.9	10.0±7.1 9.1±1.7 8.8
5.6±2.8 5.8±1.3 4.7	18.0±2.2 15.3±2.9 14.4	21.4±2.9 18.5±3.6 15.2	12.1±7.9 10.2±2.0 7.6

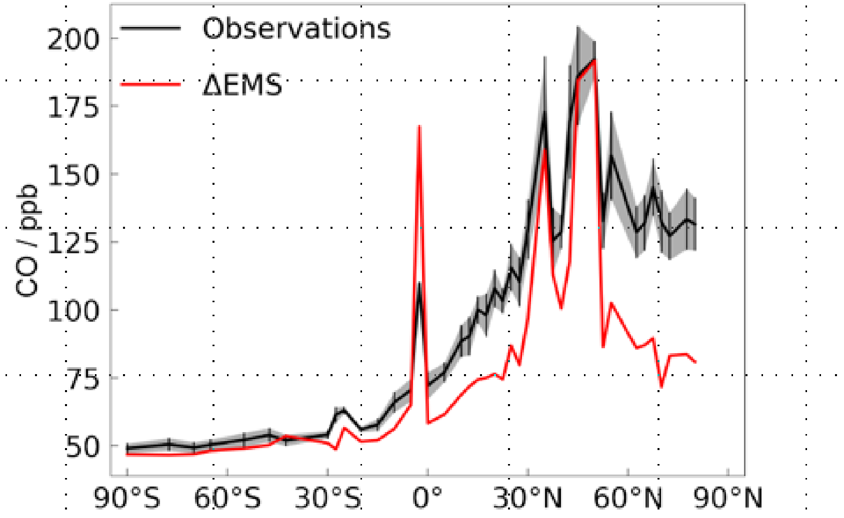
What are the changes that drive the improvement in agreement?

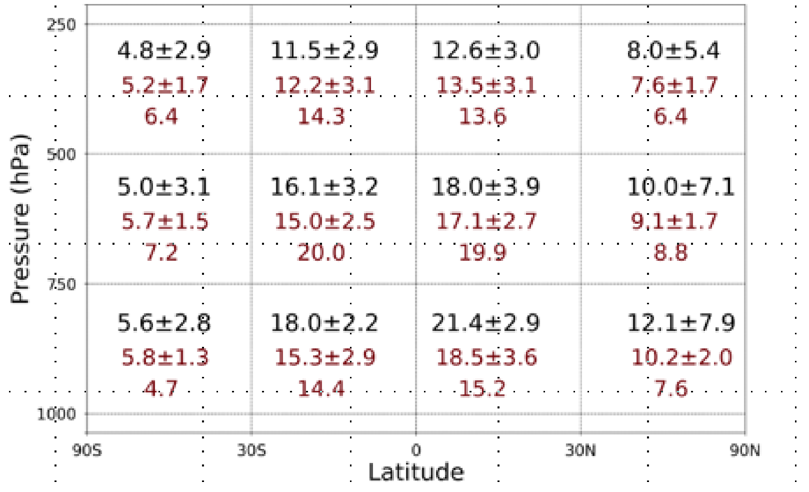
Source	Strength / Tg		ΔEMS - BASE	
	BASE	ΔEMS	Tg	Percentage
Anthropogenic	322	275	-49	-15%
Biomass burning	35	25	-10	-29%
Wetlands	190	259	-69	+36%
Other biogenic	26	26	0	0
Total	548	585	+37	+7%



Decrease NH, increase tropical emissions







Pressure (hPa)	90S–30S	30S–0	0–30N	30N–90N
250–500	4.8±2.9 5.2±1.7 6.4	11.5±2.9 12.2±3.1 14.3	12.6±3.0 13.5±3.1 13.6	8.0±5.4 7.6±1.7 6.4
500–750	5.0±3.1 5.7±1.5 7.2	16.1±3.2 15.0±2.5 20.0	18.0±3.9 17.1±2.7 19.9	10.0±7.1 9.1±1.7 8.8
750–1000	5.6±2.8 5.8±1.3 4.7	18.0±2.2 15.3±2.9 14.4	21.4±2.9 18.5±3.6 15.2	12.1±7.9 10.2±2.0 7.6

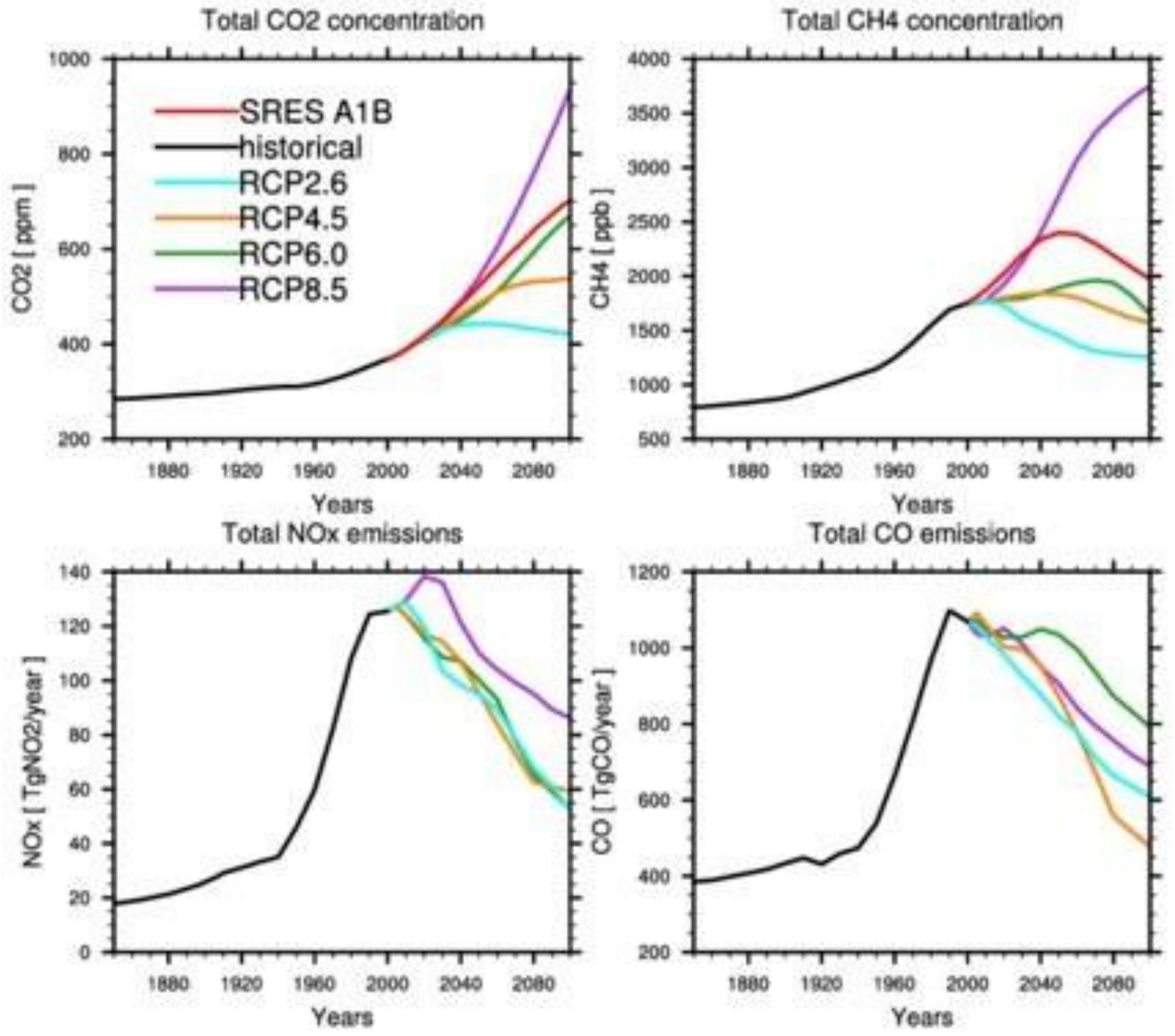
Summary of the year 2000 experiments

	BASE	ΔCO	ΔEMS
Tropospheric CH_4 emissions / Tg(CH_4) per year	548	548	585
Tropospheric CO emissions / Tg (CO) per year	1113	1660	1113
Whole Atmospheric CH_4 burden / Tg(CH_4)	4325	4790	4789
Tropospheric global mean CH_4 / ppb	1590 vs obs 1780	1787	1760
N:S methane mixing ratio gradient / ppb	104 vs obs 97	105	103
Tropospheric OH / 10^5 molecules cm^{-3}	12.4	11.1	12.0
Tropospheric global mean CO / ppb	77 vs obs 102	107	81
N:S CO mixing ratio gradient / ppb	39 vs obs 67	59	38
OH + CH_4 flux / Tg(CH_4) yr^{-1}	526	521	580
Tau_{OH+CH4} / years	8.2	9.2	8.6
Ozone burden / Tg	331	329	336
Feedback factor, R	1.55	-	-

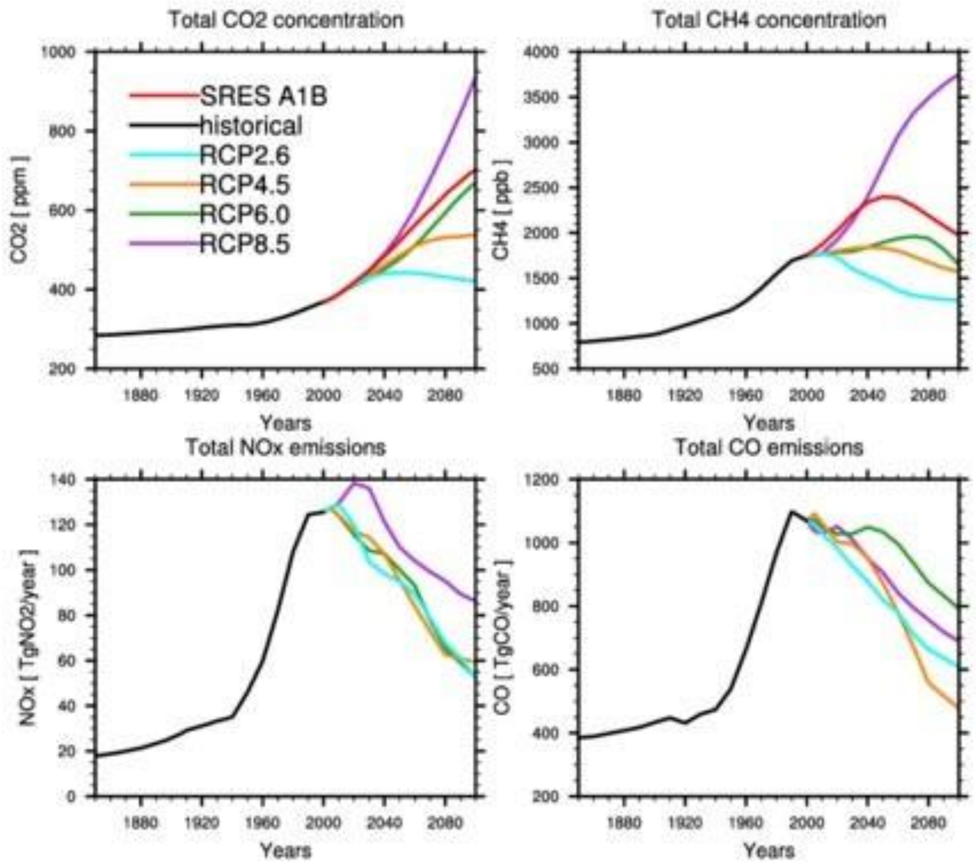
Methane in 2100

What happens to tropospheric oxidising capacity in future climate?

- We chose RCP8.5 – ODS, CO_2 and other emissions increased to give 8.5 Wm^{-2} radiative forcing.
- RCP8.5 also features
  - Large increases in methane by the end of the century
  - NOx and CO decreasing after 2050
- Our approach was to look at these climate drivers individually
  - ‘What is the effect of the temperature driver?’
  - ΔCC – climate forcings only
  - ‘And emissions?’
  - ΔCC+CH4 – increase **anthropogenic methane** emissions to RCP8.5
- Bring all forcings together at the end
  - ΔCC+ALL – increase (NTCF) O3Pre to RCP8.5

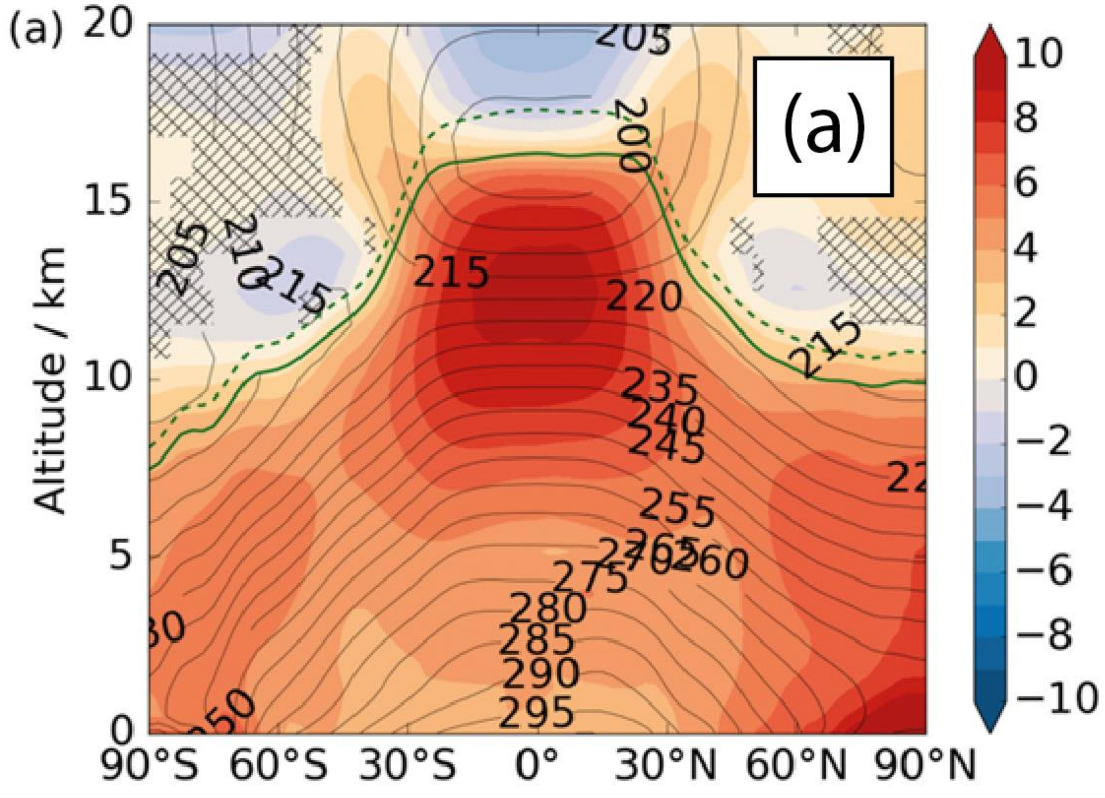


What happens to tropospheric oxidising capacity in future climate?

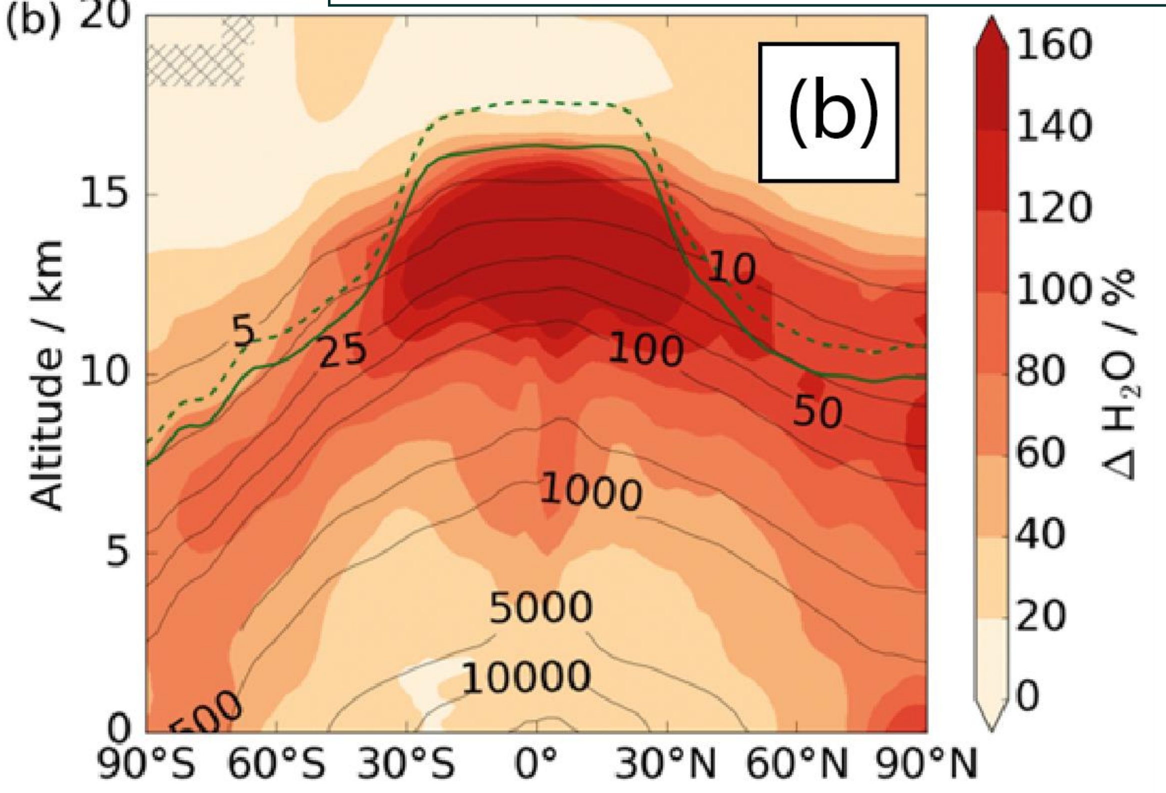


- ○ In RCP8.5 there's a big increase in temperature throughout the troposphere by 2100.
- ○ The warmer atmosphere can support more water vapour, so humidity increases.
- ○ Tropospheric expansion means the upper troposphere experiences the biggest changes.

Dashed green line – year 2100 tropopause
Solid green line – year 2000 tropopause



Temperature

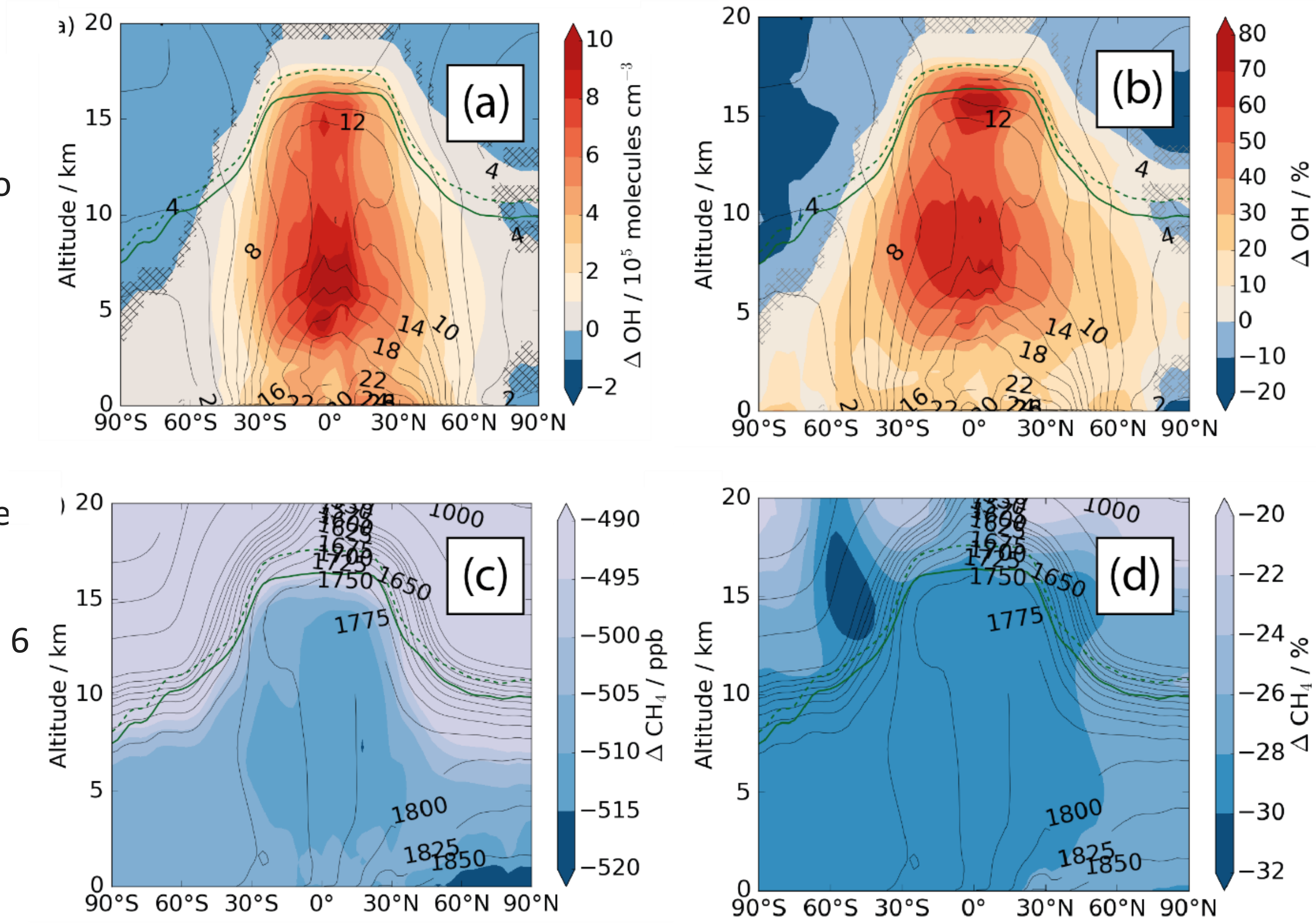


Water vapour

What happens to tropospheric oxidising capacity in future climate?

- OH – warmer, wetter atmosphere so OH increases
- Changes largest in tropical FT
- More OH means less CH4 (and k(OH+CH4) increases as T increases)
- Methane decrease large everywhere cf Year 2000.
- Methane lifetime reduced from 9 to 6 years.

Hydroxyl



Methane

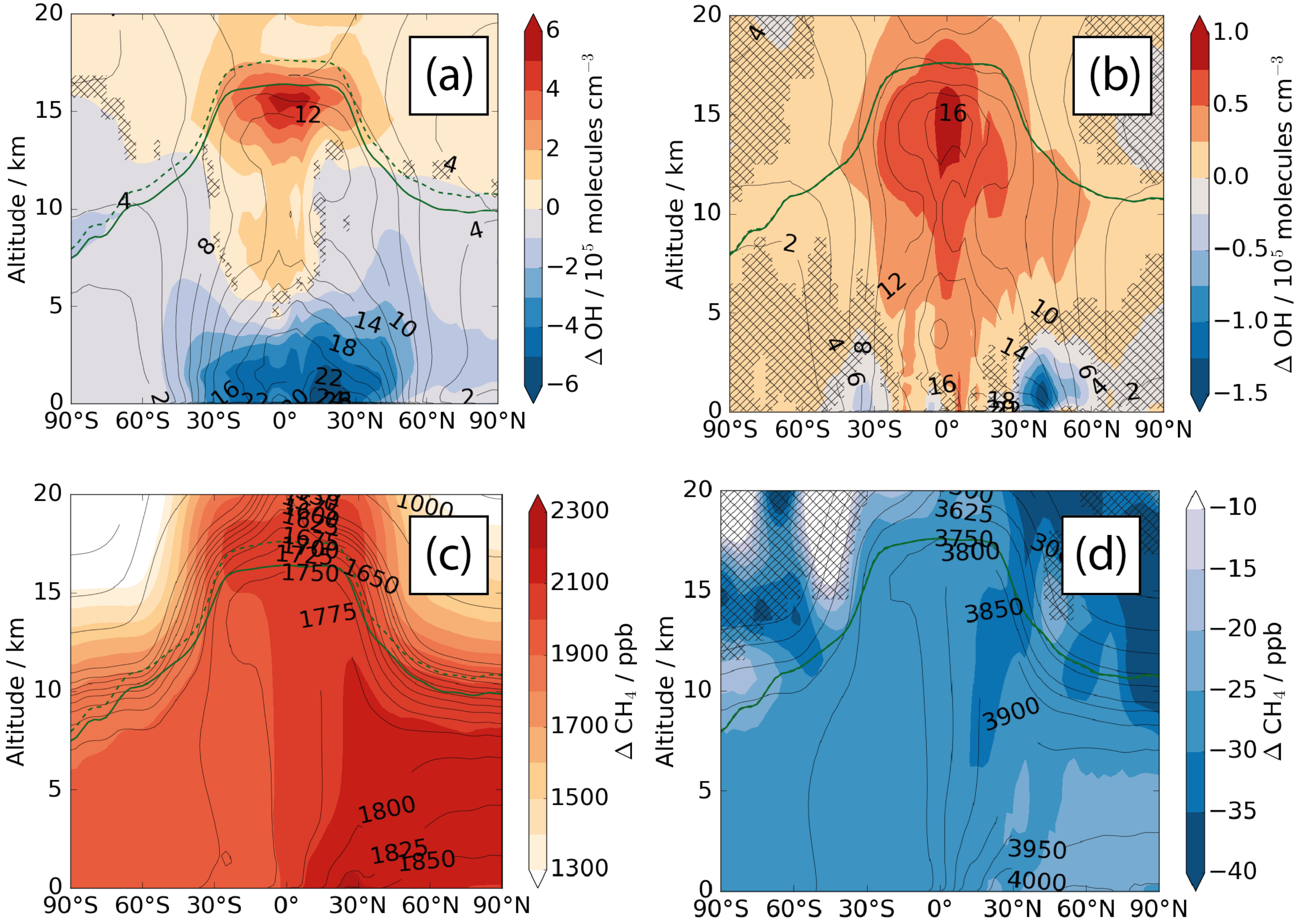
What happens to tropospheric oxidising capacity in future climate?

- Increasing CH_4 emissions to RCP8.5 levels gives
  - Large increase in CH_4
  - Large decrease in OH
- Increasing CO and NOx to RCP8.5 levels gives
  - Smaller change in OH
  - Small decreases in CH_4

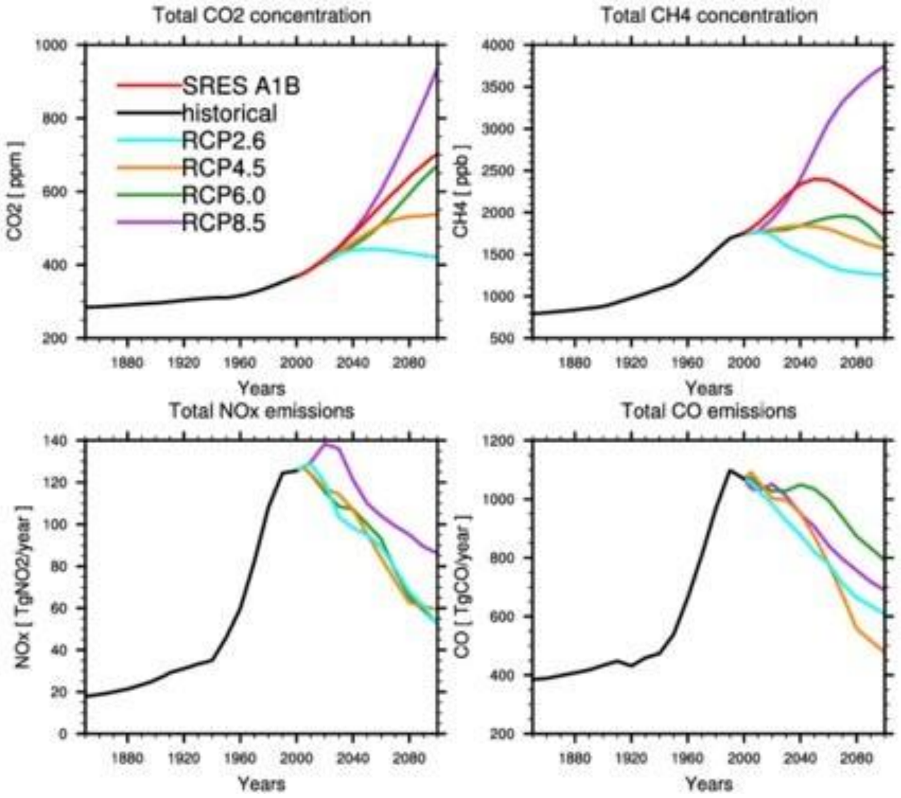
Hydroxyl

Increase CH4 to 2100

Increase O3PRE to 2100



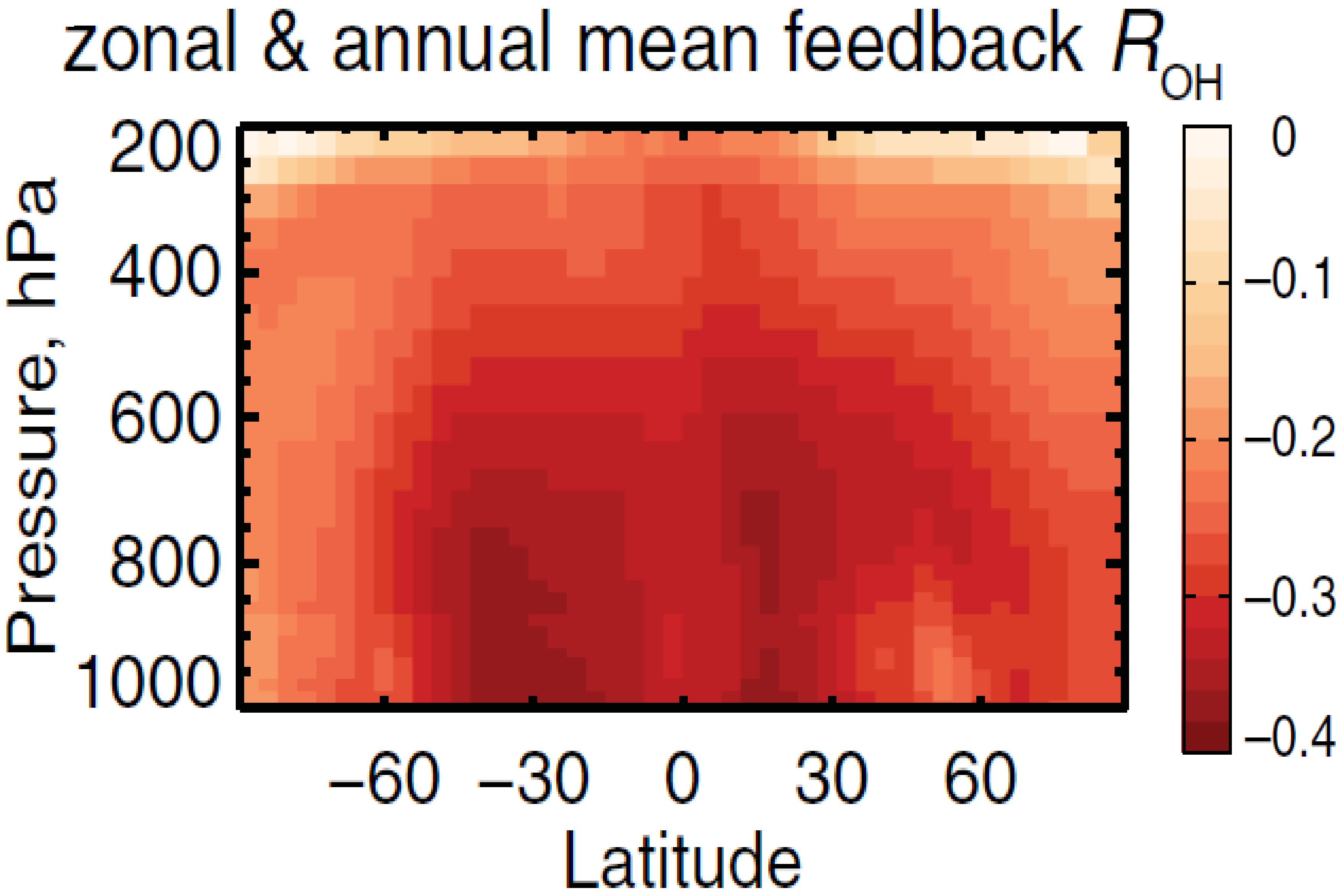
Methane



Summary of the CC experiments

	ΔCC	$\Delta(CC+CH_4)$	Δ(CC+EMS)
Tropospheric CH_4 emissions / Tg(CH_4) per year	548	1170	1170
Tropospheric CO emissions / Tg (CO) per year	1113	1113	734
Anthropogenic NOx emissions / Tg N per year	44	44	30
Whole Atmospheric CH_4 burden / Tg(CH_4)	3421	10336	10260
Tropospheric global mean CH_4 / ppb	1275	3828	3746
Tropospheric OH / 10^5 molecules cm^{-3}	15.7	10.5	10.6
OH + CH_4 flux / Tg(CH_4) yr^{-1}	568	1120	1121
$Tau_{(OH + CH4)}$ / years	6.0	9.2	9.2
Tropospheric O_3 burden / Tg	350	443	427
Feedback factor, R	1.62	1.44	1.43

Spatial variation in feedback – not constant through the troposphere!



Zero feedback f =1/(1+R) = 1

Weak feedback f =1/(1+R) = 1.25

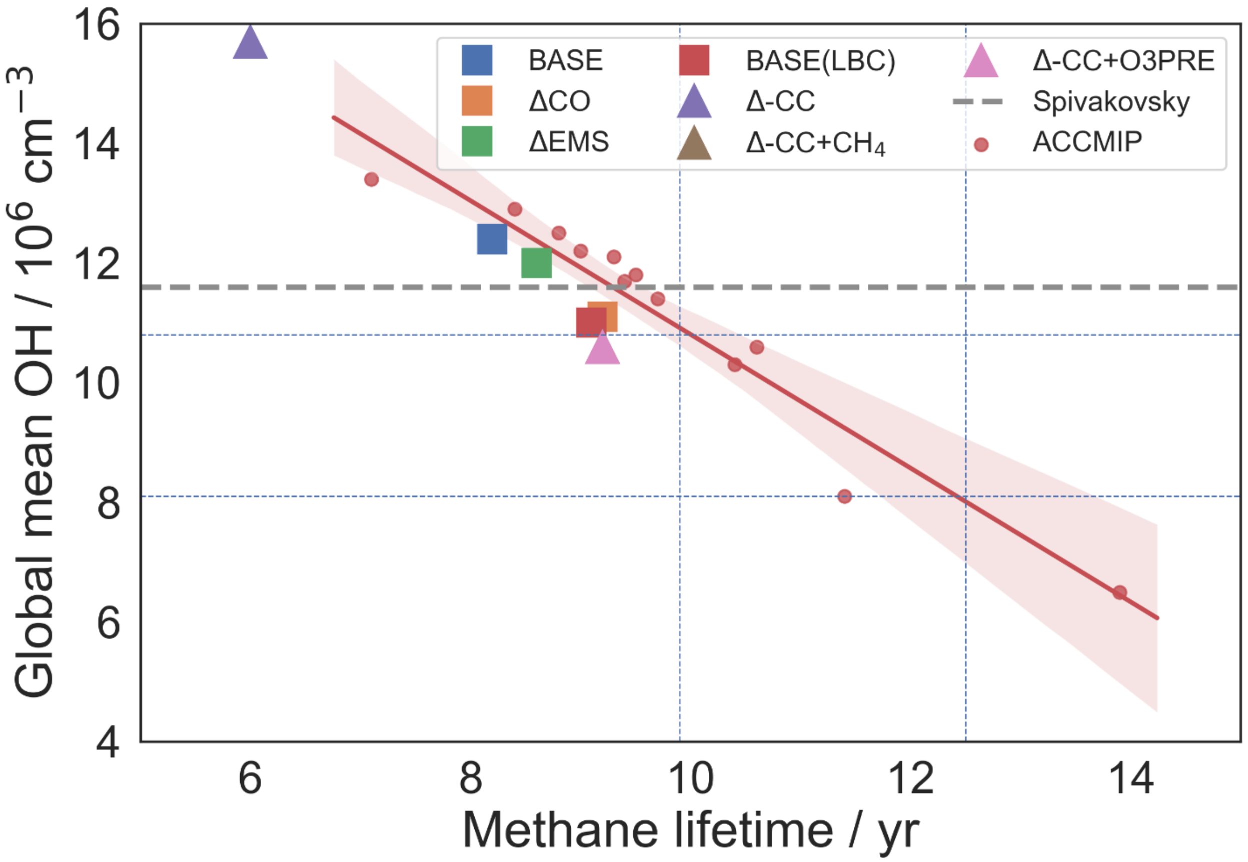
Medium feedback f =1/(1+R) = 1.42

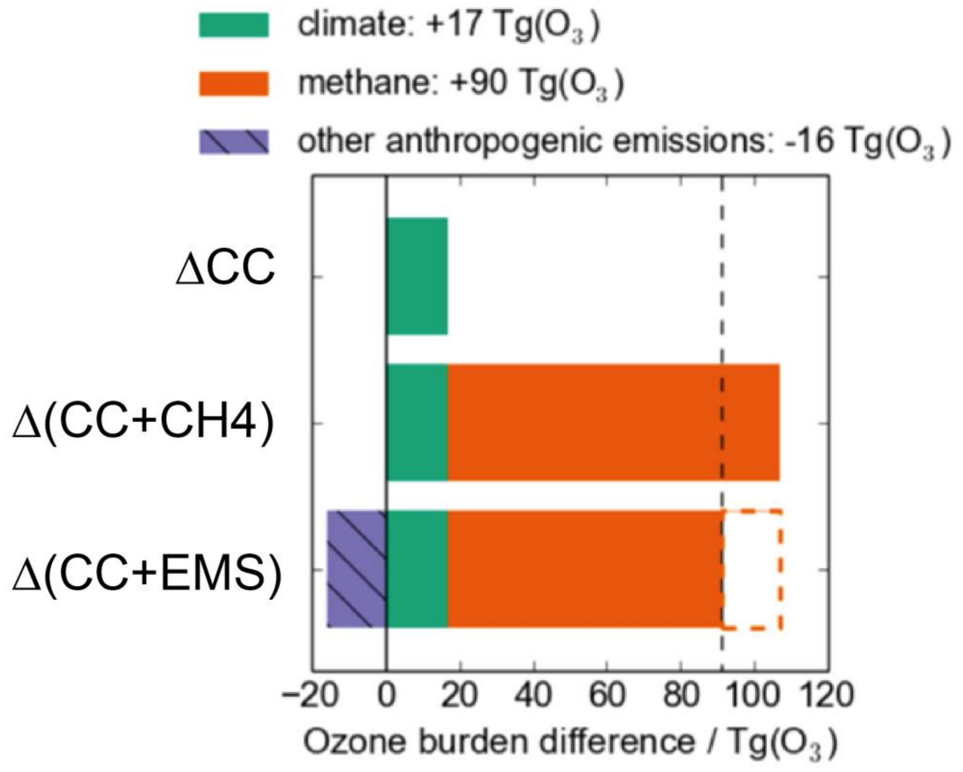
Strong feedback f=1/(1+R)=1.67

NB Year 2000 conditions/OH/temp

Methane in the UKCA chemistry-climate model - conclusions

- Every emissions dataset can probably be *tweaked* to compare well with obs when implemented in a 3D model
  - Tropical CH_4 emissions slightly low biased, boreal emissions high biased [UKCA]
  - CO emissions may be low, but secondary CO production from VOC oxidation important and under-represented
  - In future climate, warmer temperatures act to increase OH, oxidising capacity
  - Methane emissions produce a large change in oxidizing capacity
  - Suppresses OH but increases ozone





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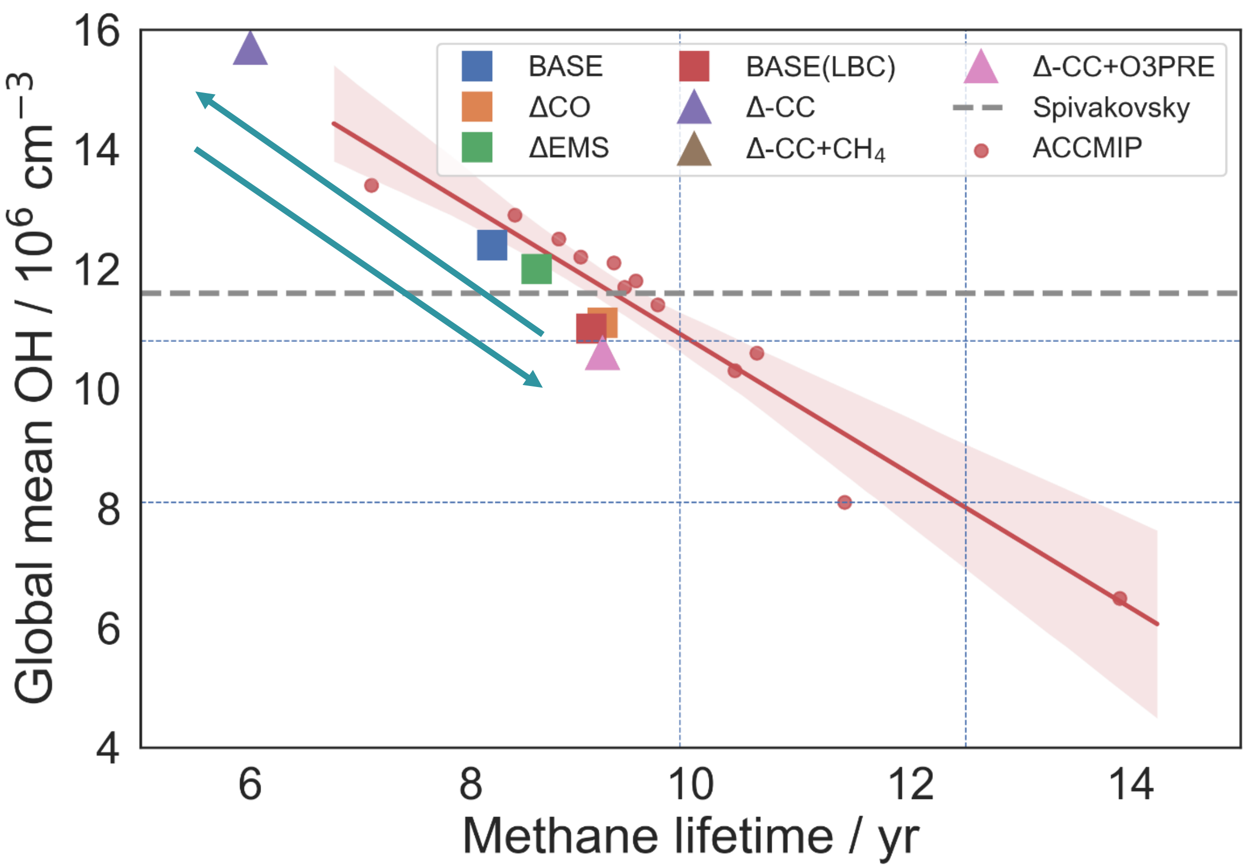

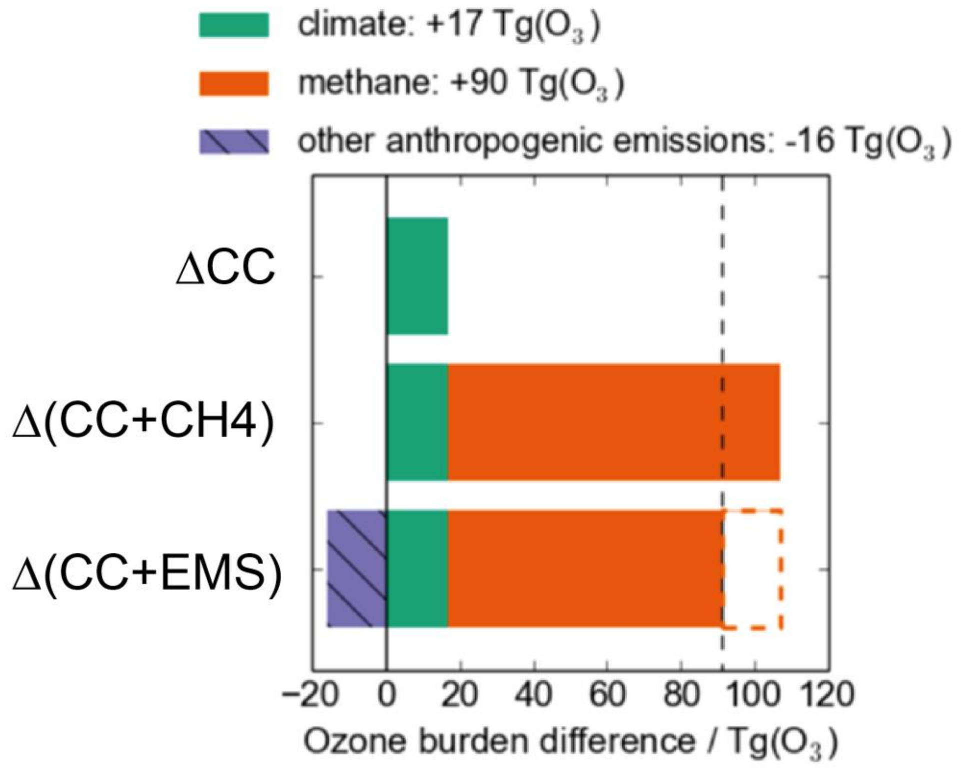

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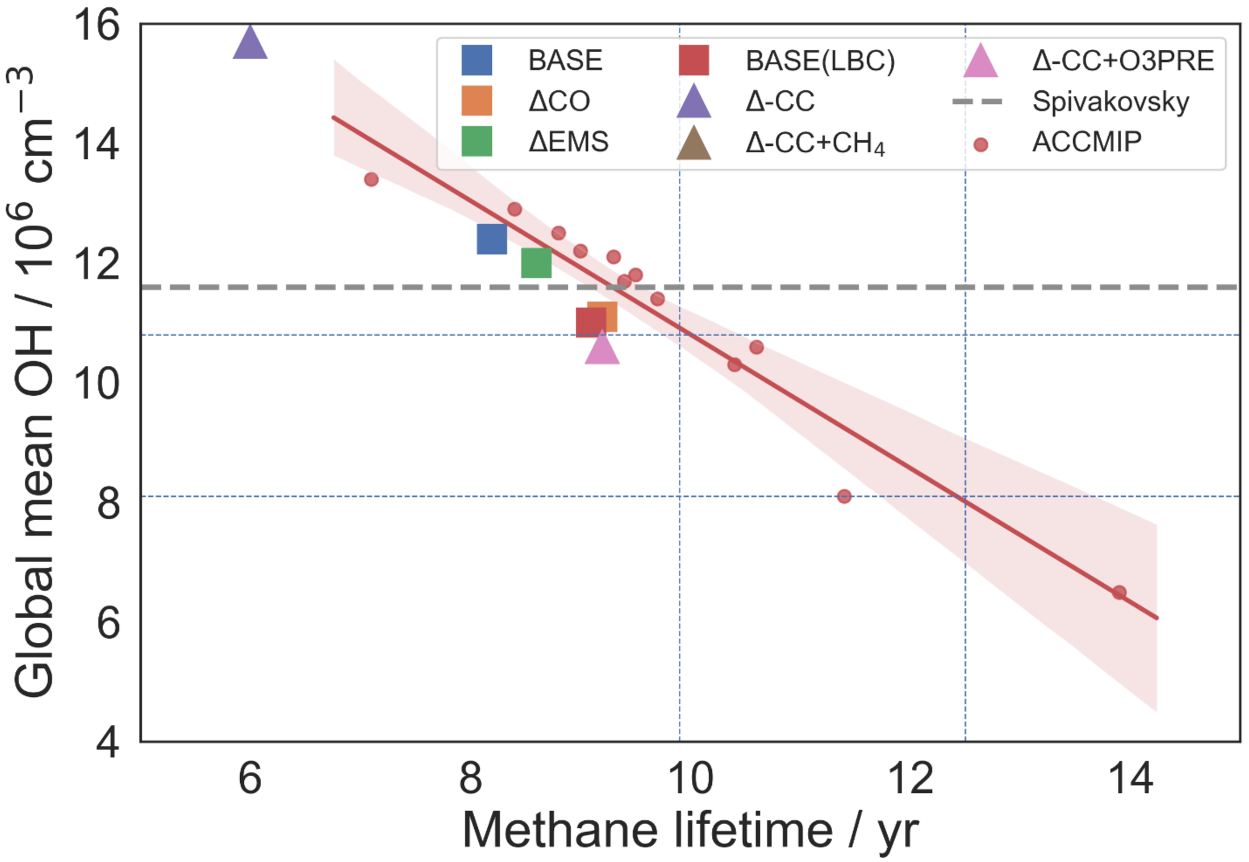

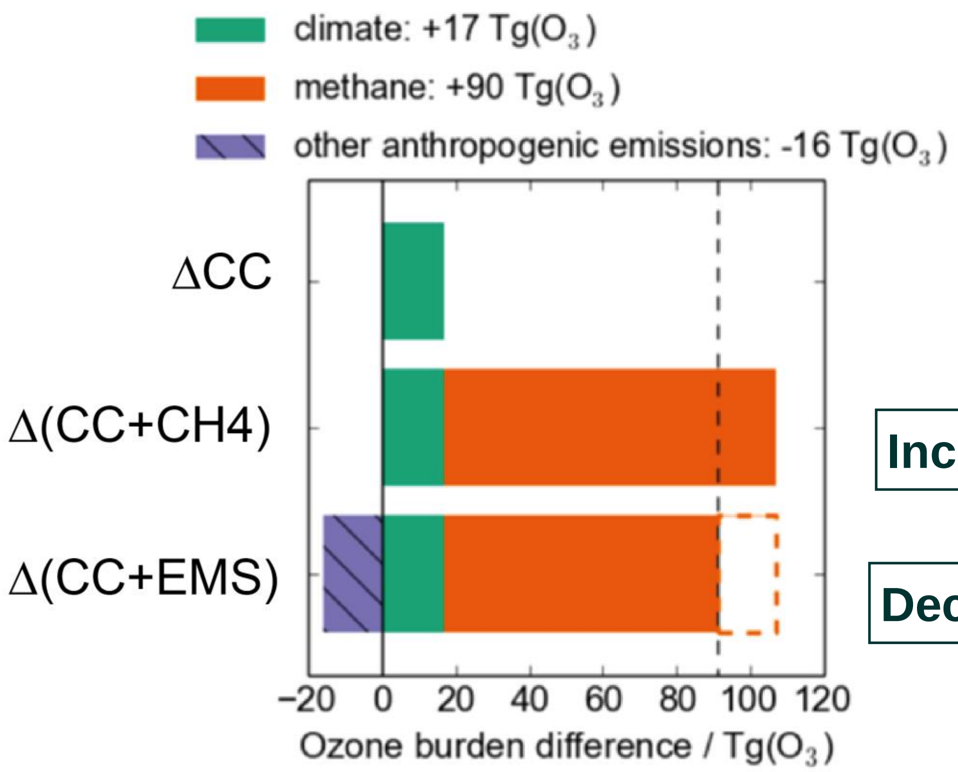

Inc in O3

Dec in O3PRE

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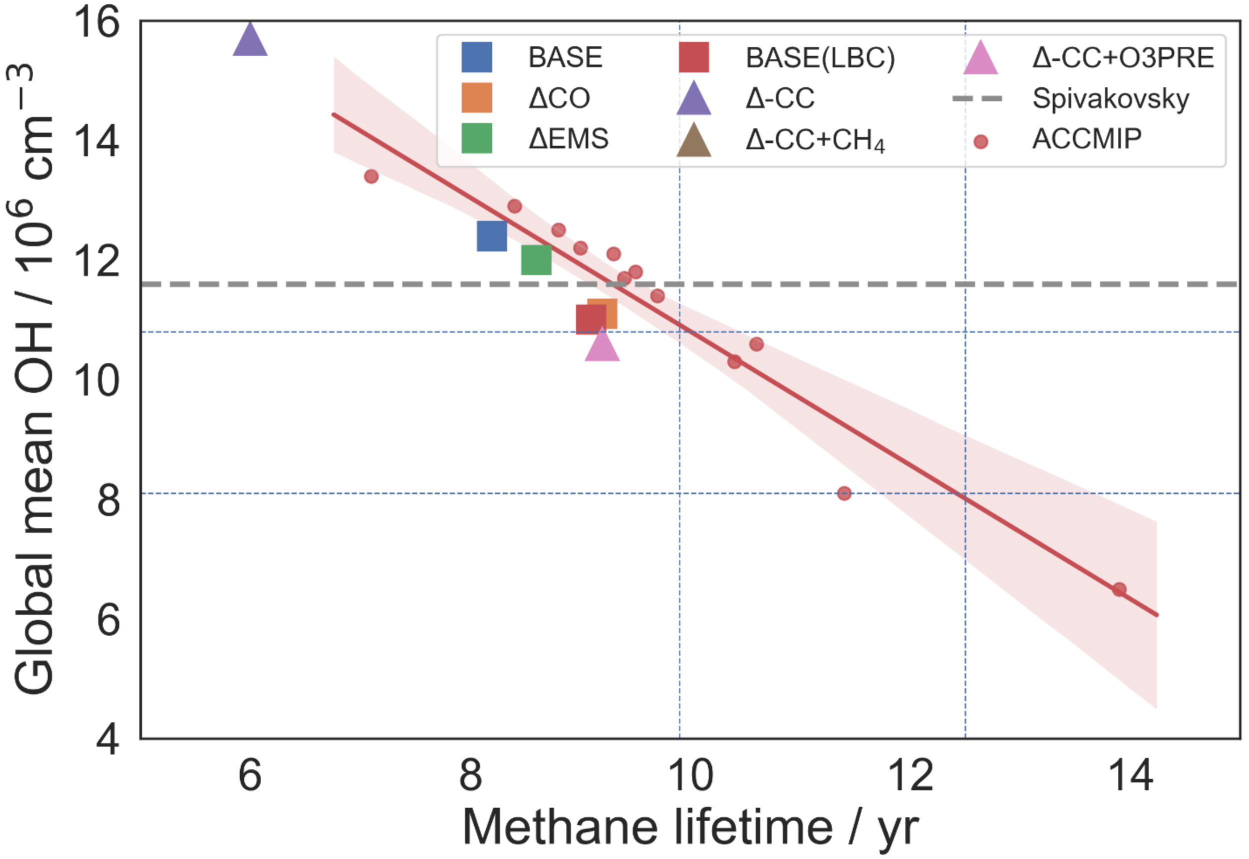
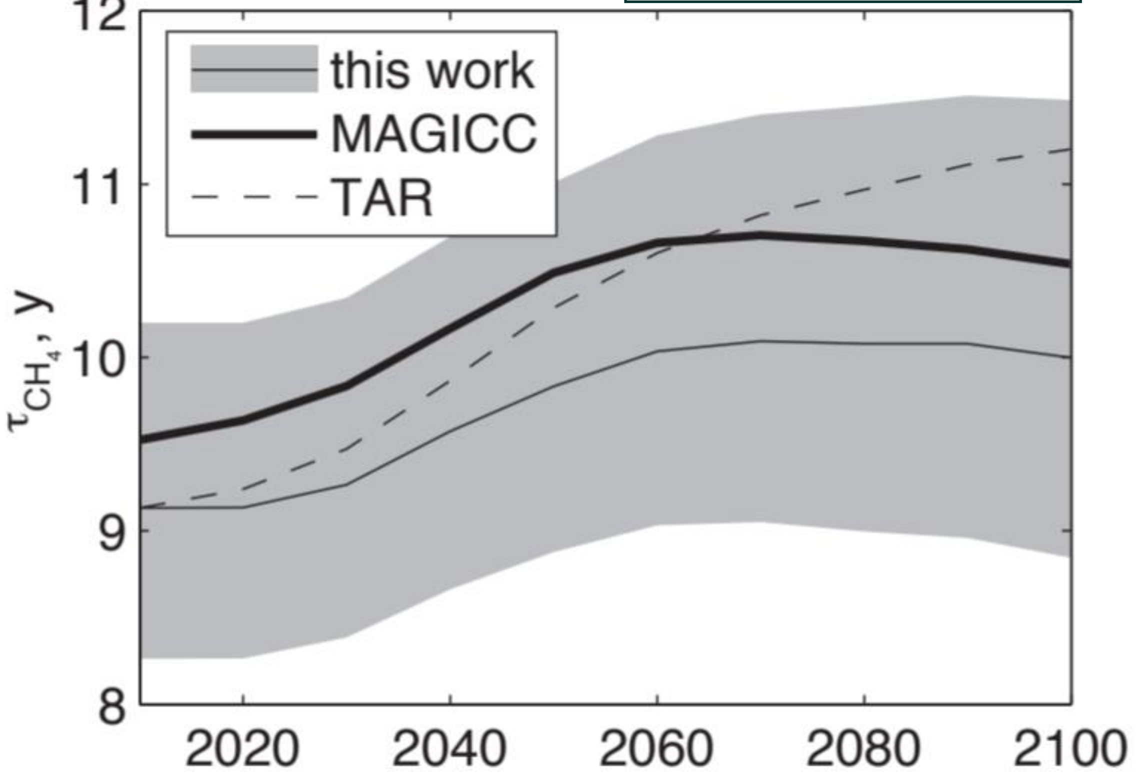


Holmes et al., 2013



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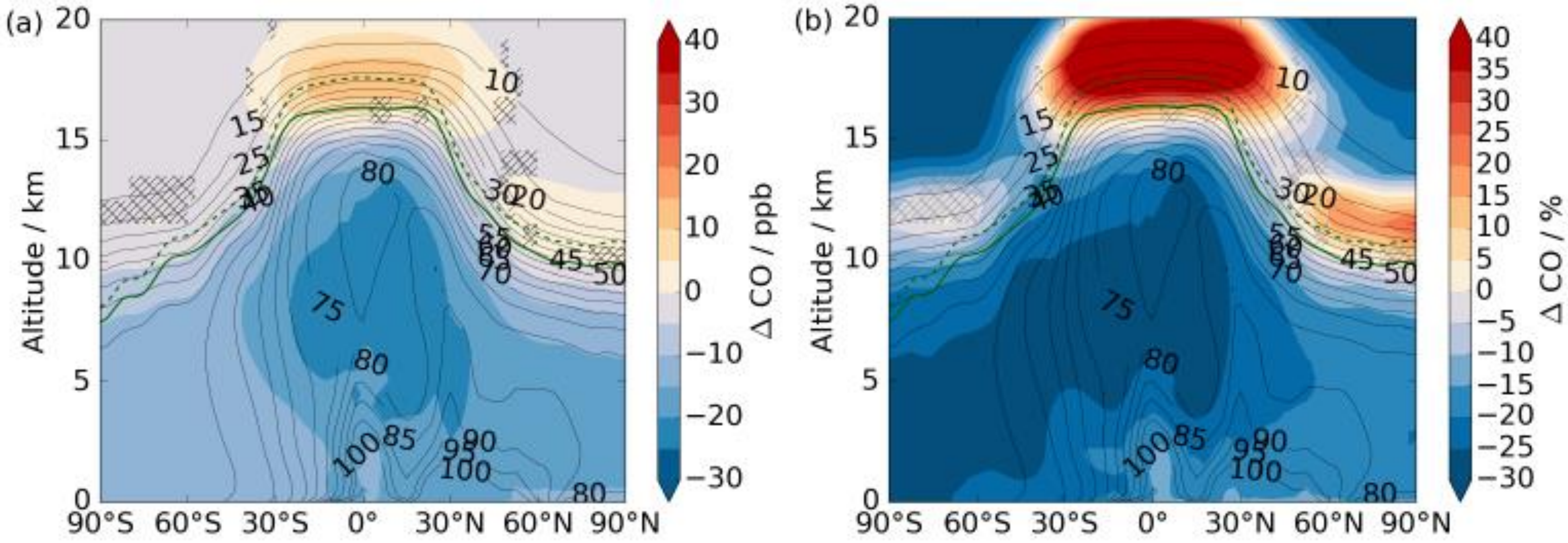
Tropospheric O3 burden / Tg	350	443	427	329	336

Conclusions

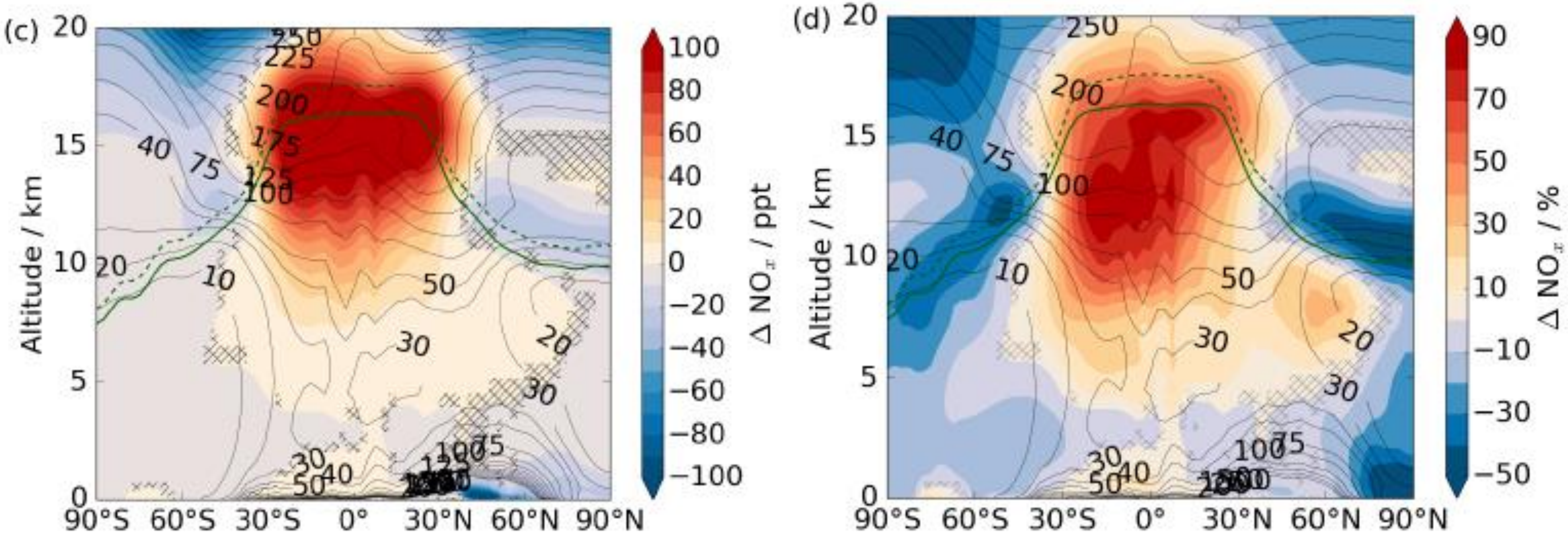
- Assessing methane emissions in a chemistry-climate model poses problems of constraint
- CO is a big part of the story as CO, CH4 and OH are coupled together
- Playing slightly fast and loose with the methane emissions enables good model-measurement agreement

- RCP8.5 Year 2100 show large differences from present day (!)
  - Increases in OH due to temperature decrease methane lifetime by 3 years
  - Including methane emissions pushes methane lifetime back up to 9 years
    - Large increase in O3 burden due to methane increases
  - RCP8.5 small decreases in O3PRE have small effect on methane lifetime, OH

CO in ΔCC experiments



NOx in ΔCC experiments



O3 in ΔCC experiments