

Studies of chemistry-climate interactions using UKESM1: near-term climate forcers of the recent past and near future

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Keith Shine - **Reading University, UK**

Peter Coleman - **UK Govt Dept for Business, Energy and Industrial Strategy**

AMIP studies of CH₄ emissions

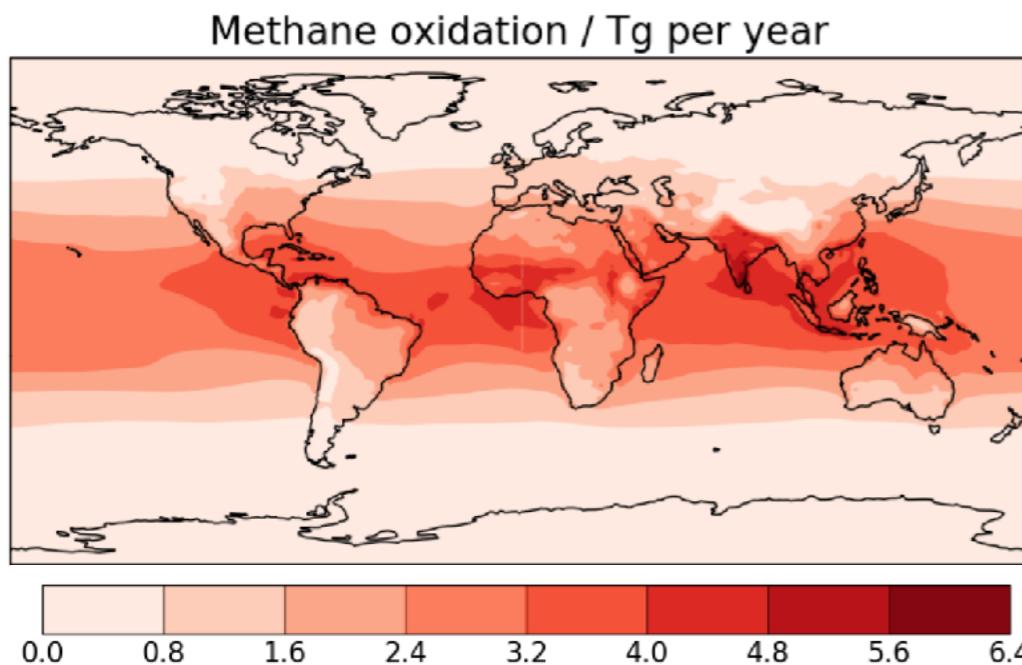
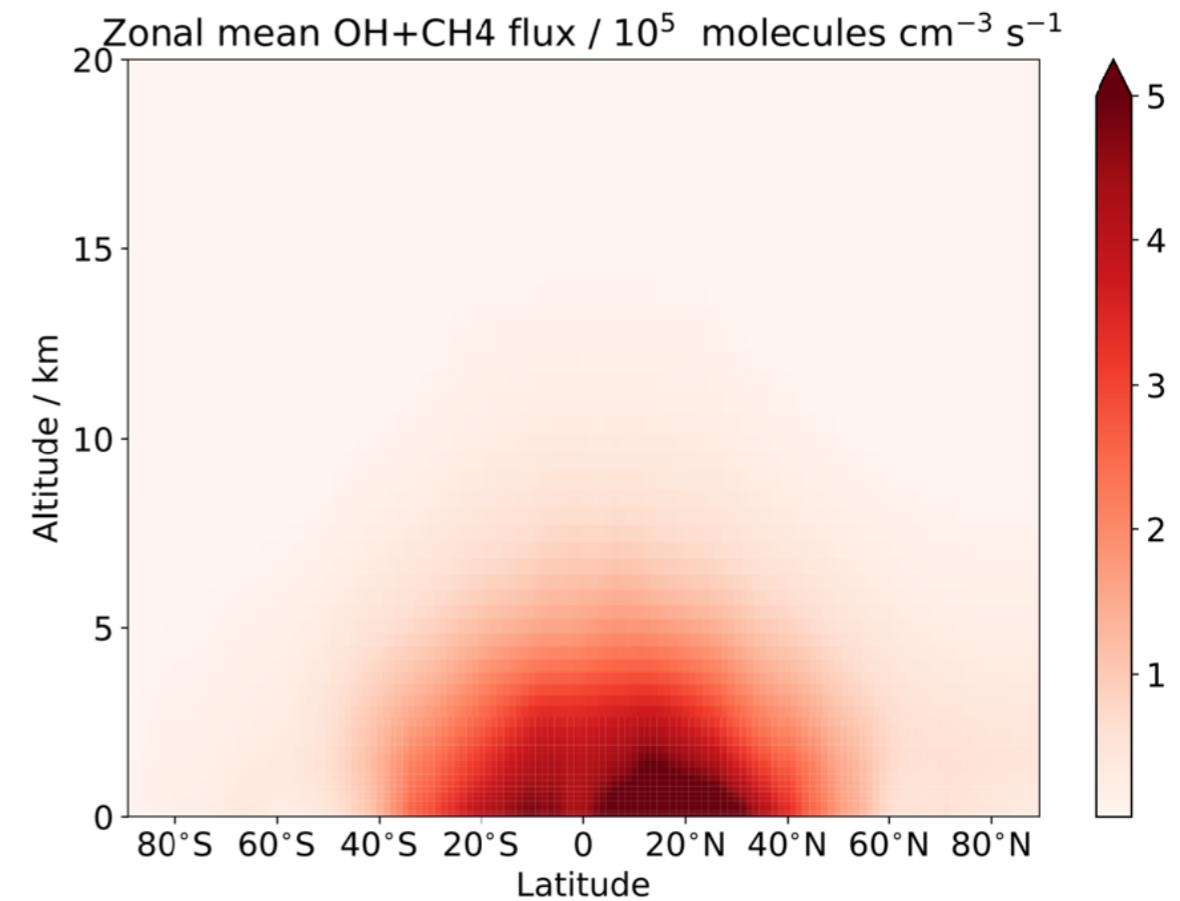
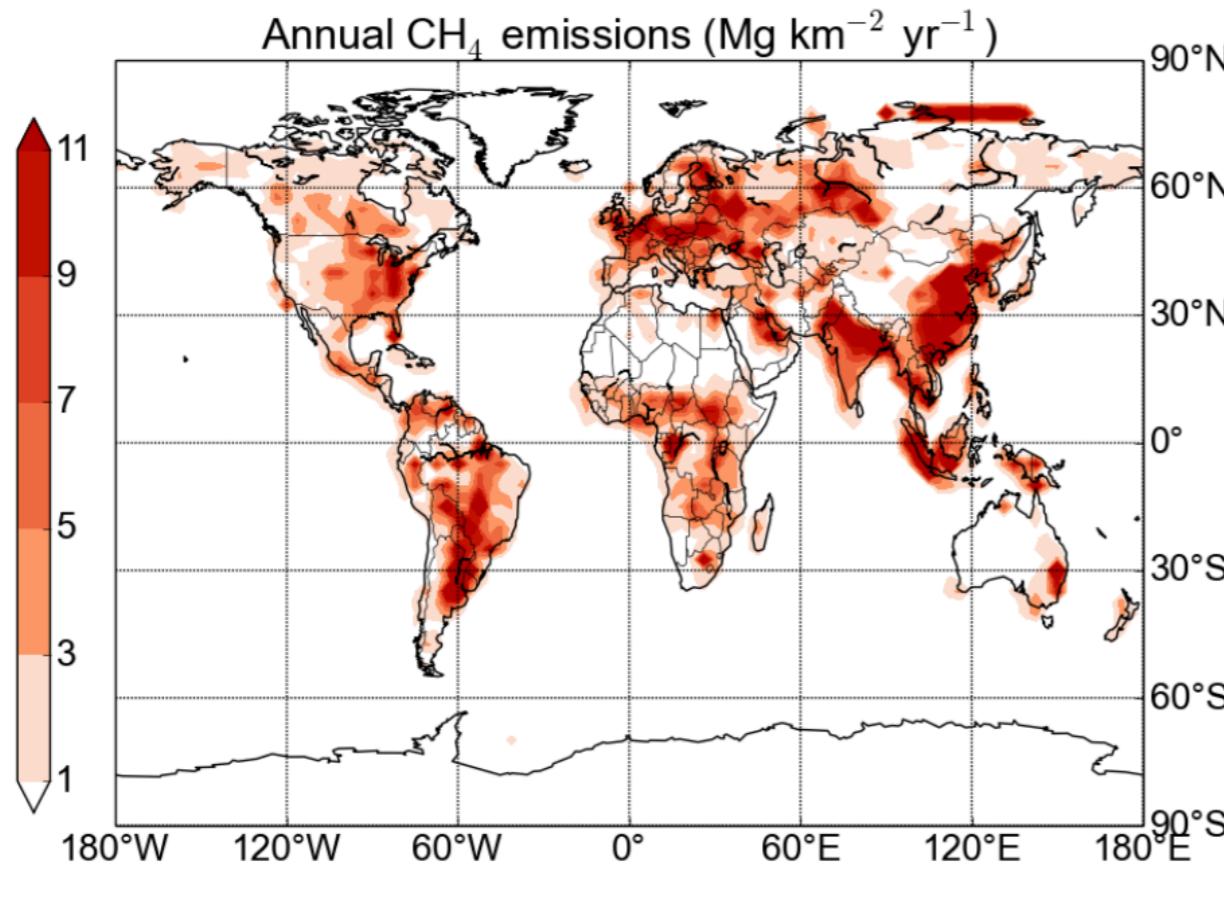
Atmospheric methane is an important greenhouse gas

- 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

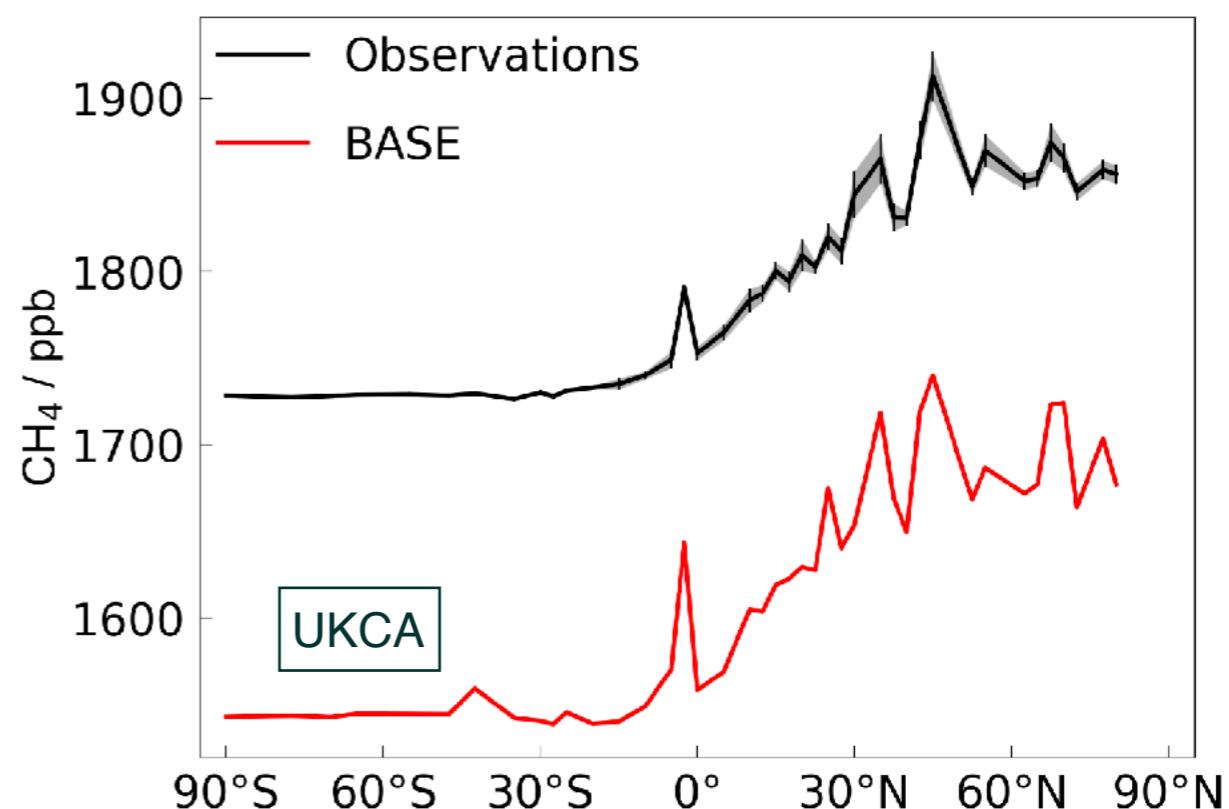
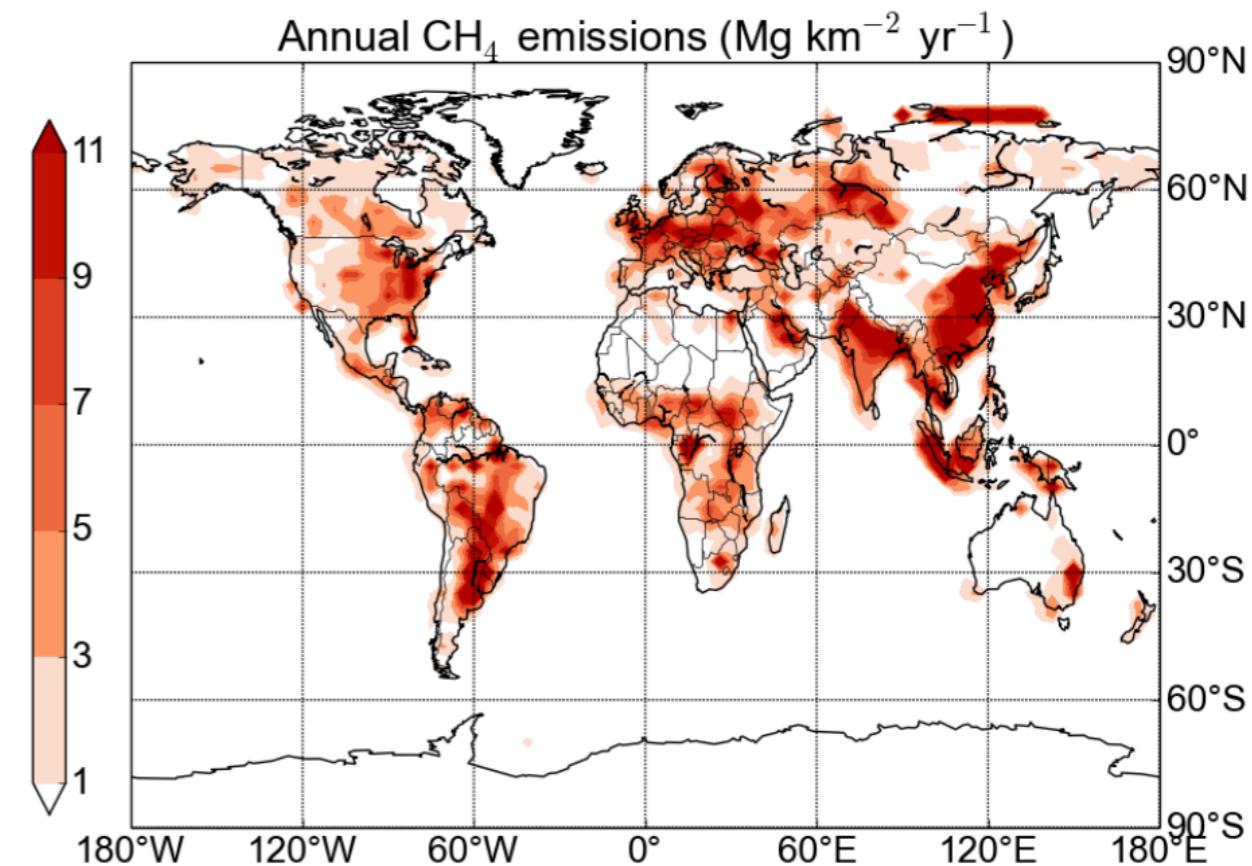
Methane in UKCA - emissions vs OH sink



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

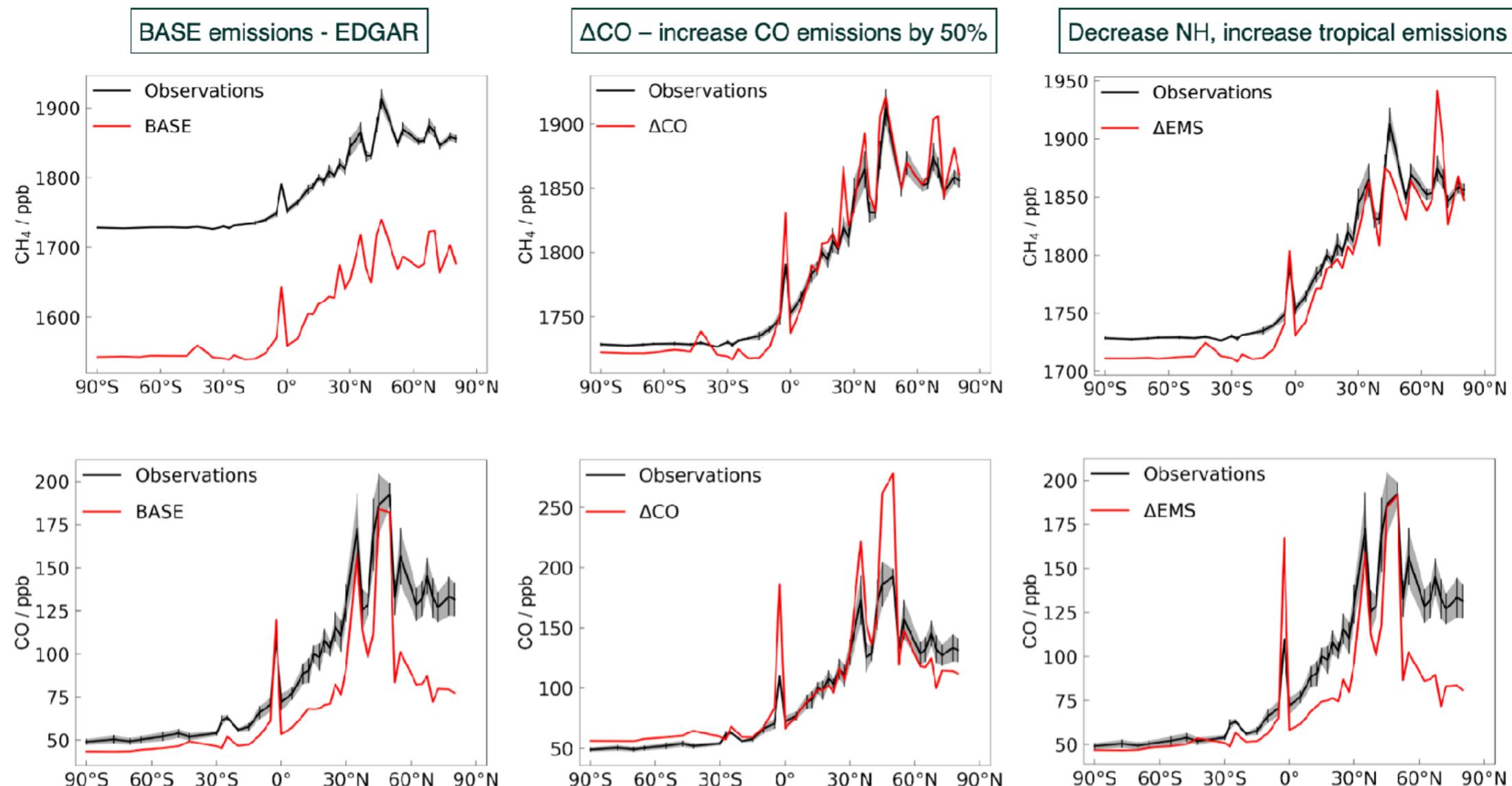
Methane in UKCA - comparison with observations

- 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?

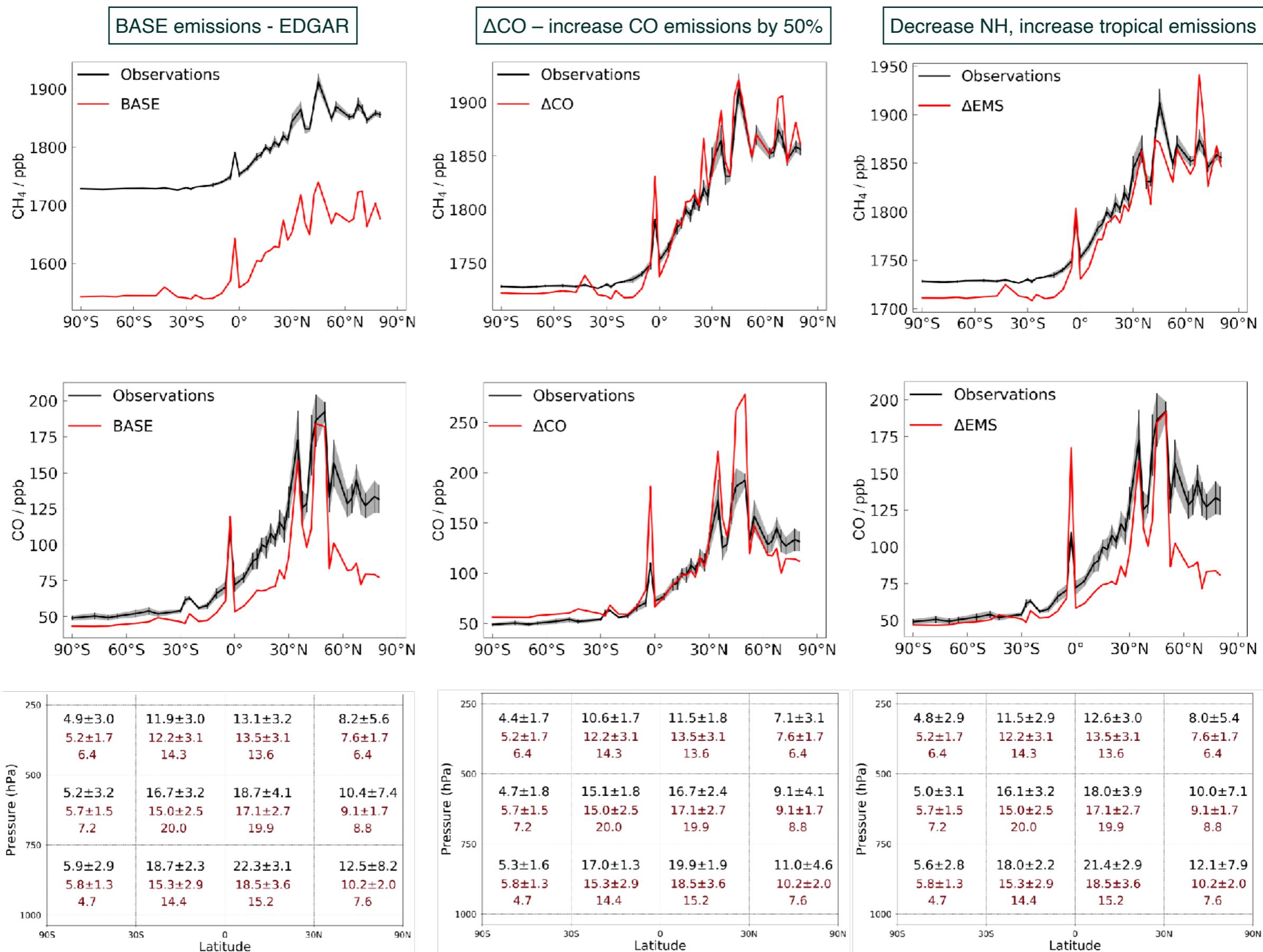


3 sensitivity experiments

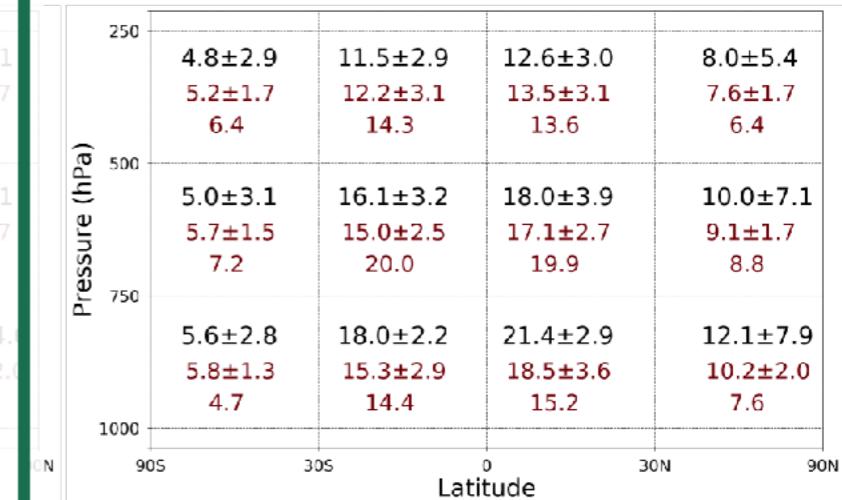
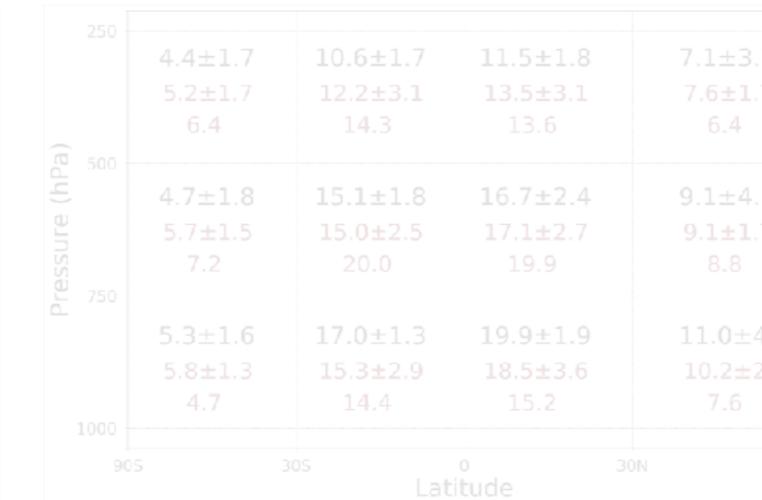
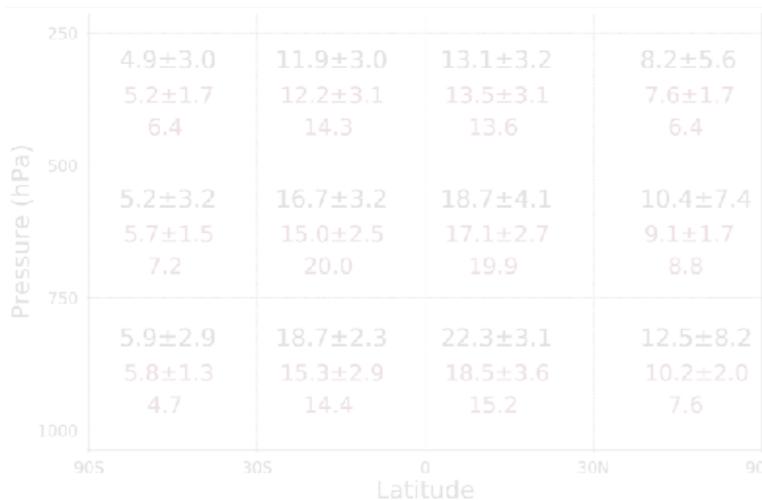
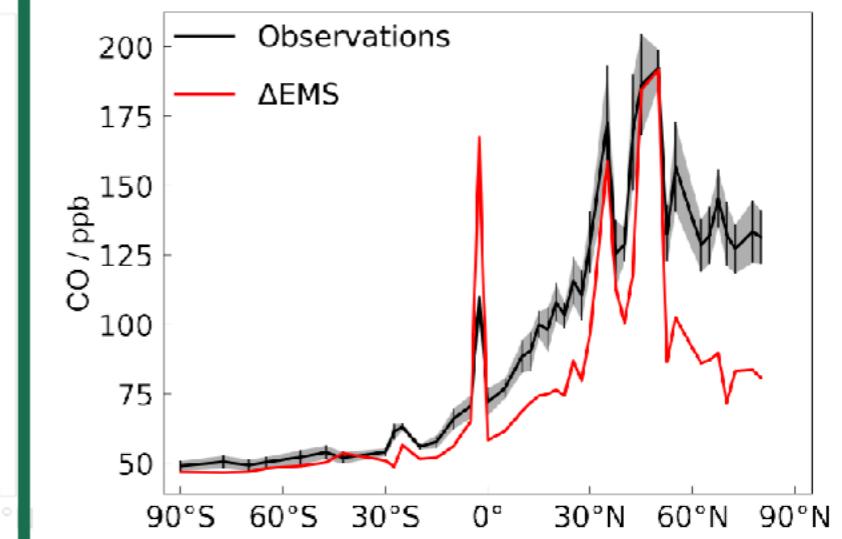
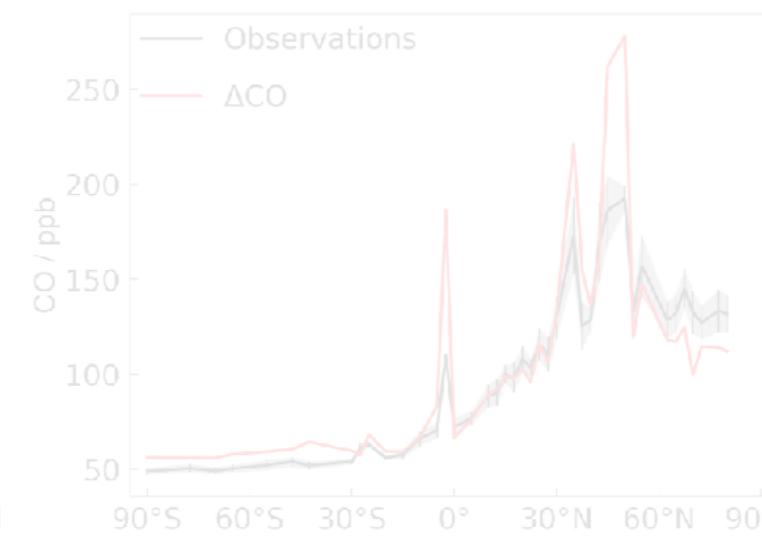
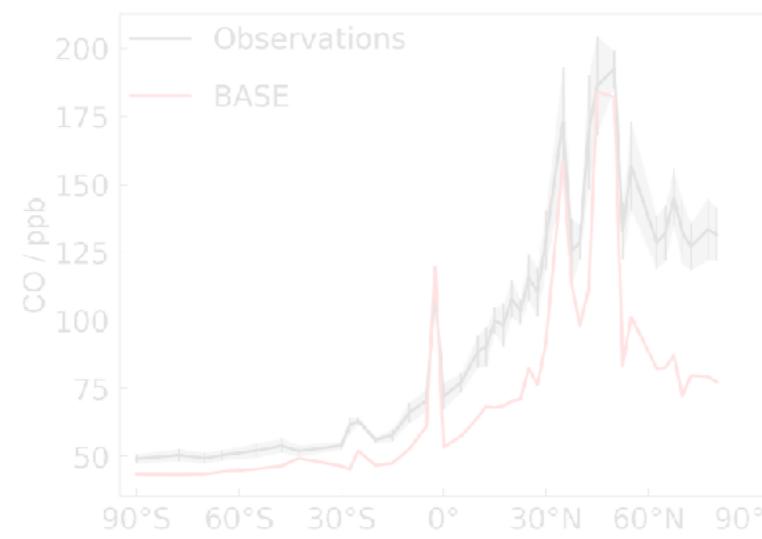
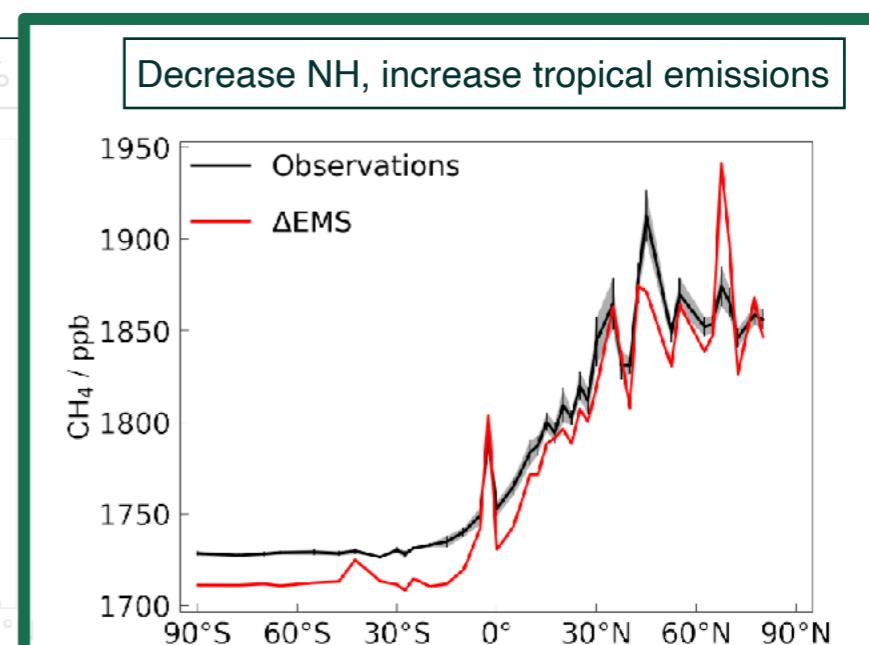
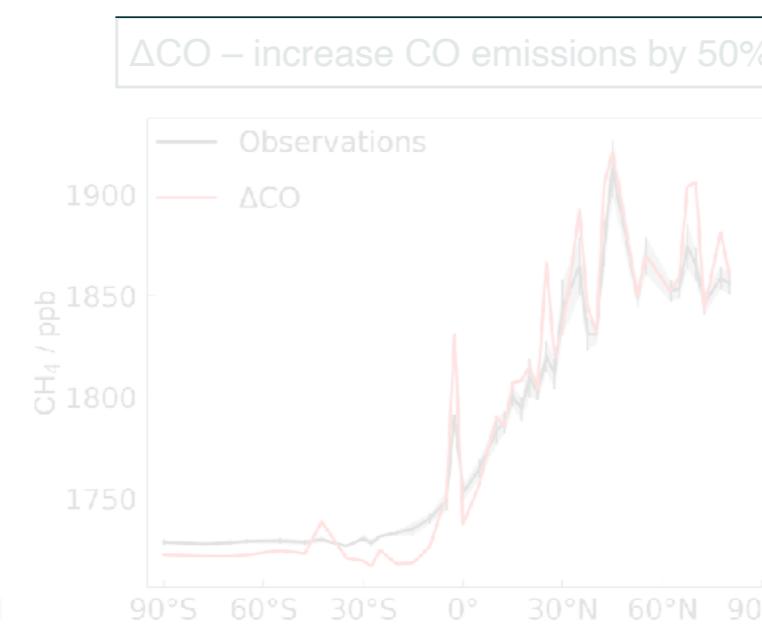
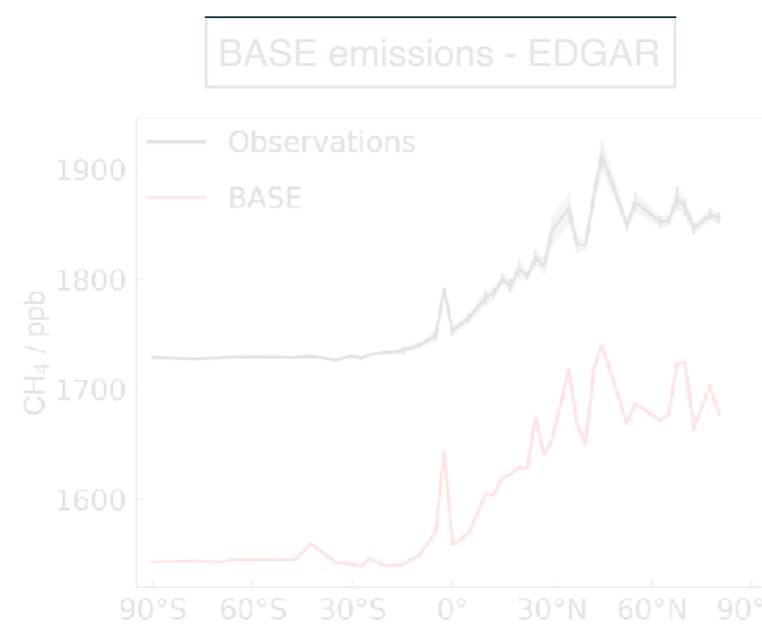
1. Our BASE run using methane emissions derived from EDGAR emissions database.
2. A second experiment in which CO emissions are increased everywhere by 50%
3. An experiment in which we use a different emissions dataset with lower emissions in NH midlatitudes higher emissions in tropics.

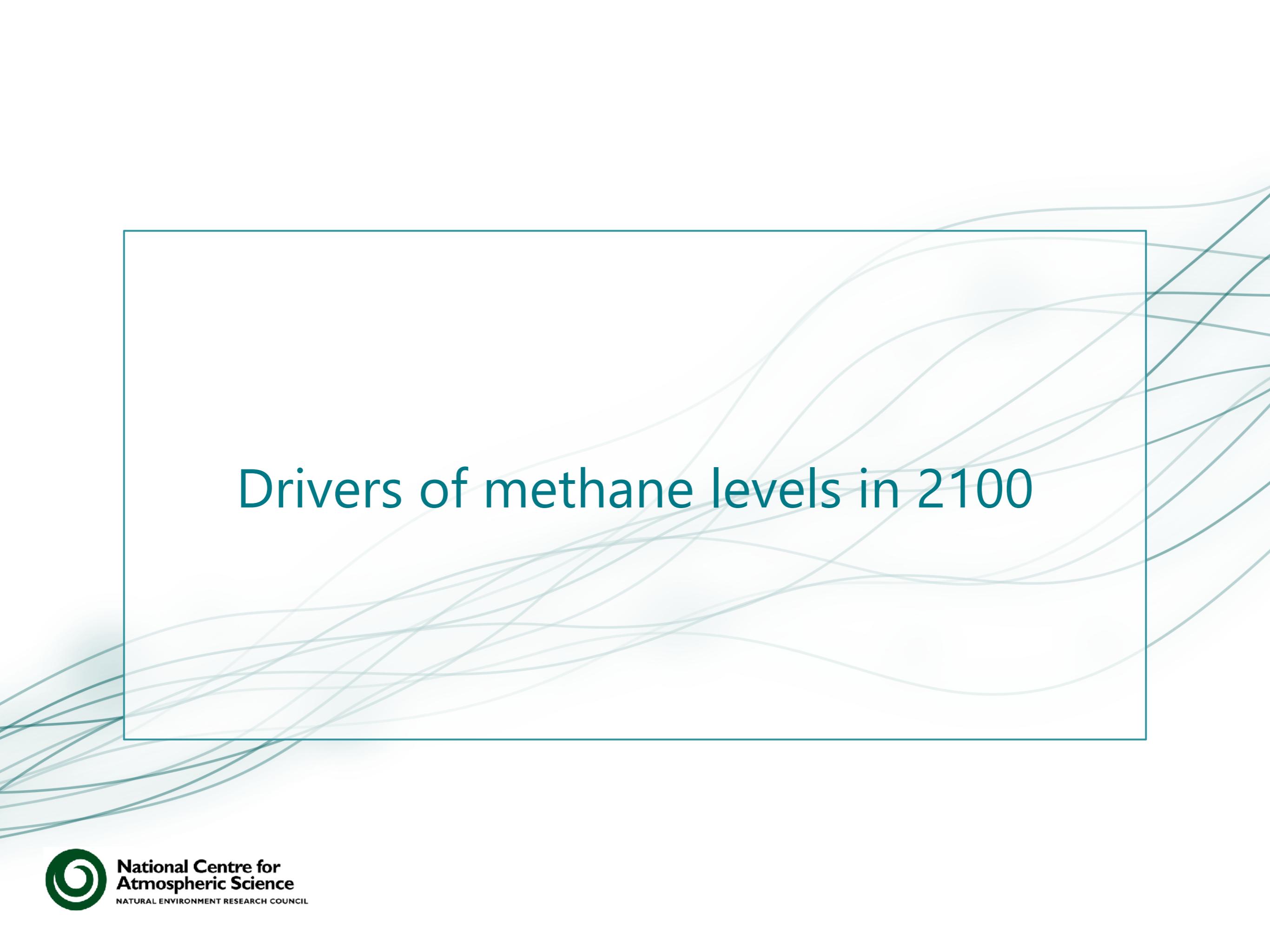


Sensitivity of UKCA to emissions – 3 global experiments



Sensitivity of UKCA to emissions – 3 global experiments

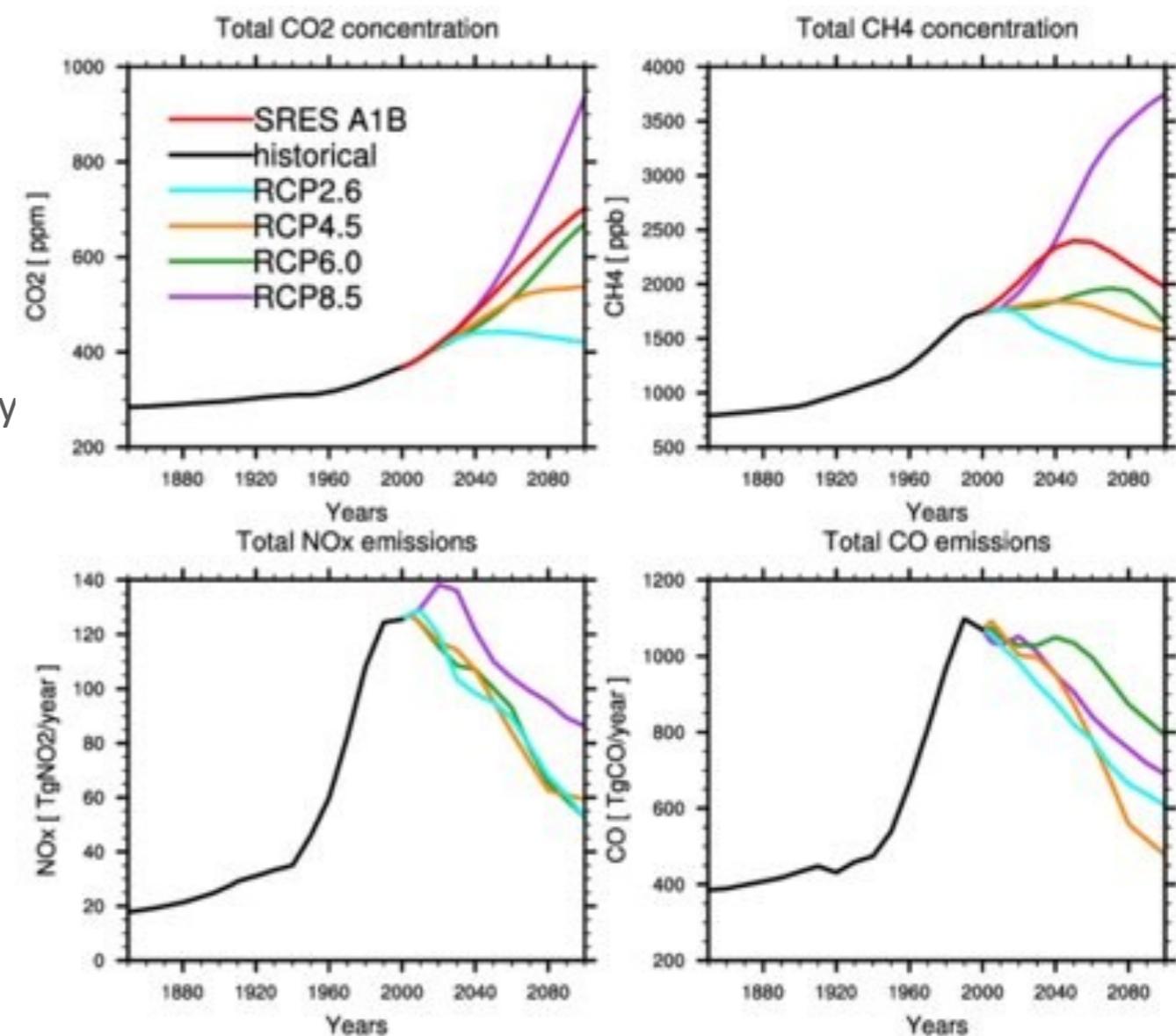




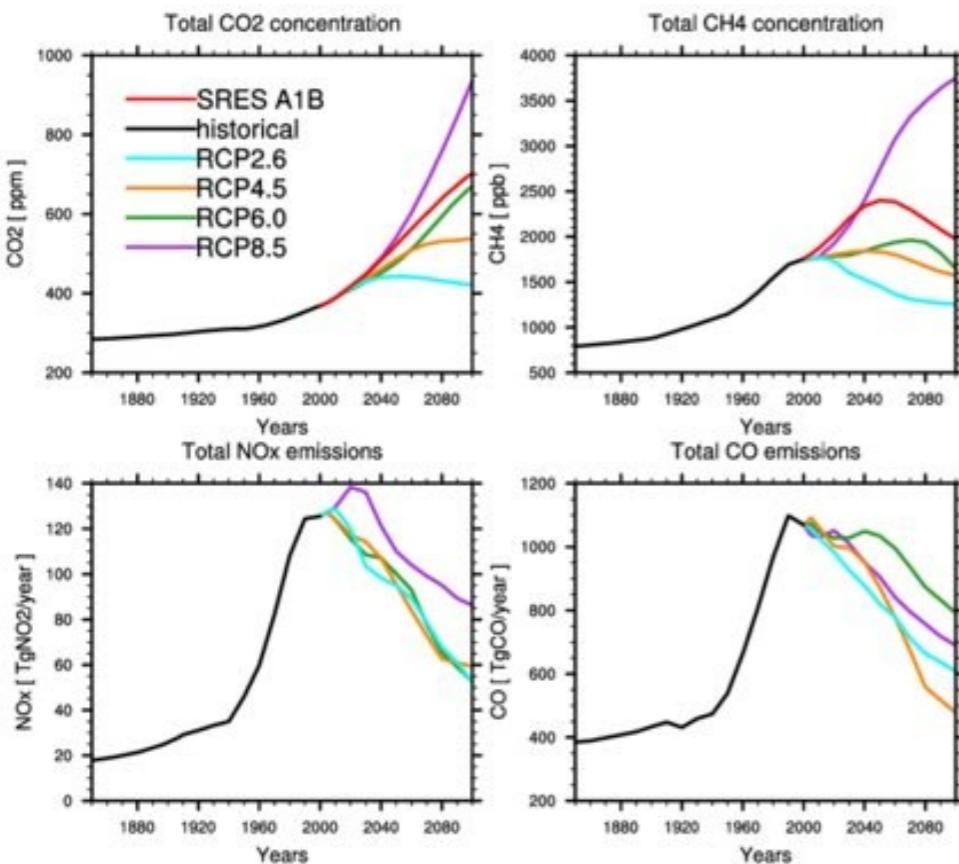
Drivers of methane levels in 2100

What happens to tropospheric oxidising capacity in future climate?

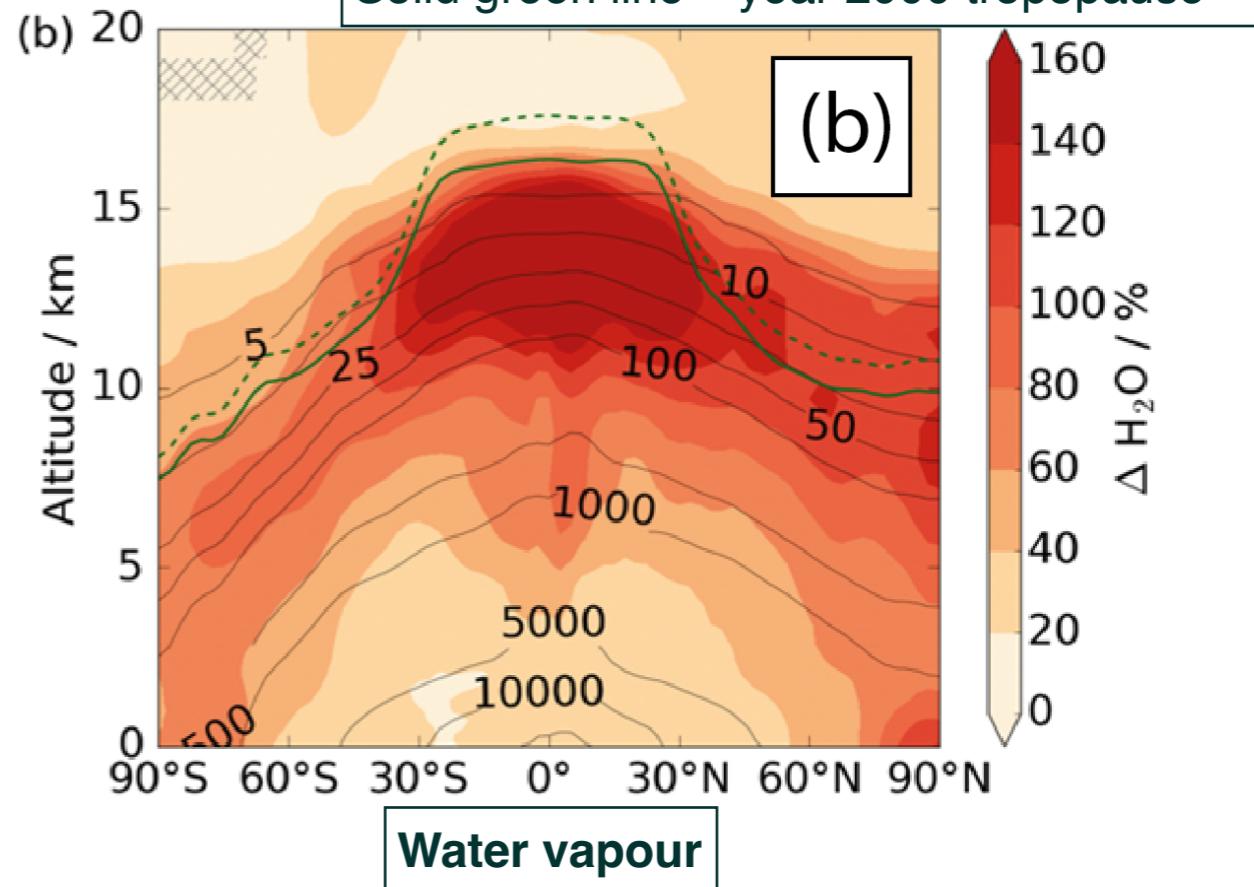
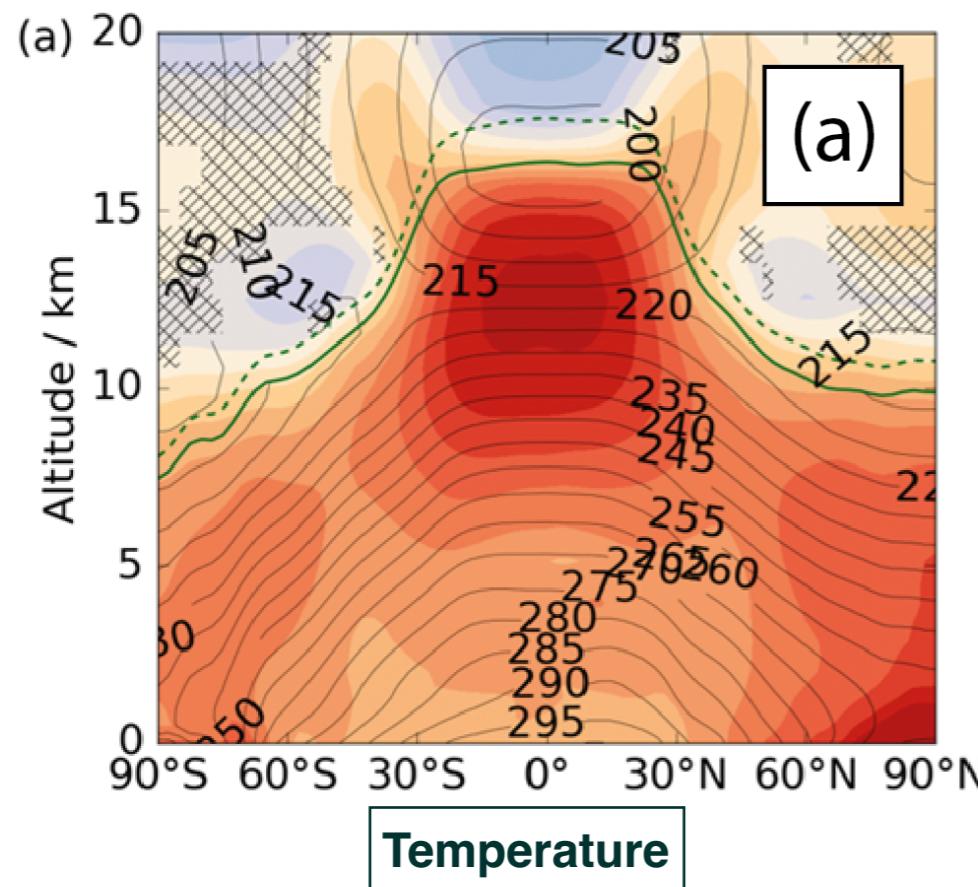
- We chose RCP8.5 – ODS, CO₂ and other emissions increased to give 8.5 Wm⁻² radiative forcing.
- RCP8.5 also features
 - Large increases in methane by the end of the century
 - NOx and CO decreasing after 2050
- Our approach was to look at these climate drivers individually
 - ‘What is the effect of the temperature driver?’
 - ΔCC – climate forcings only
 - ‘And emissions?’
 - ΔCC+CH4 – increase methane emissions to RCP8.5
 - ΔCC+ALL – increase O3Pre to RCP8.5
- Bring all forcings together at the end



What happens to tropospheric oxidising capacity in future climate?



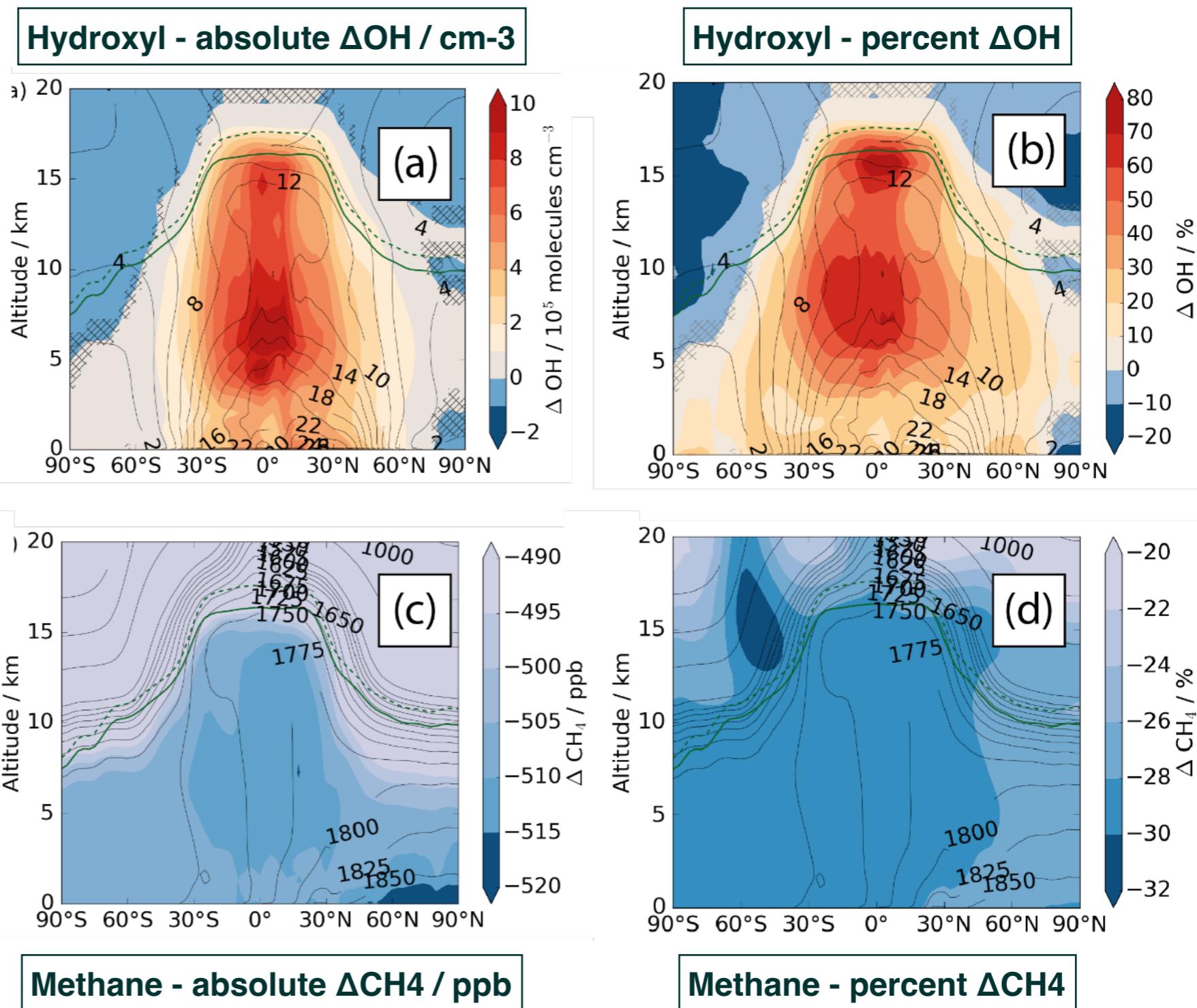
- In RCP8.5 there's a big increase in temperature throughout the troposphere by 2100.
- The warmer atmosphere can support more water vapour, so humidity increases.
- Tropospheric expansion means the upper troposphere experiences the biggest changes.



What happens to tropospheric oxidising capacity in future climate?

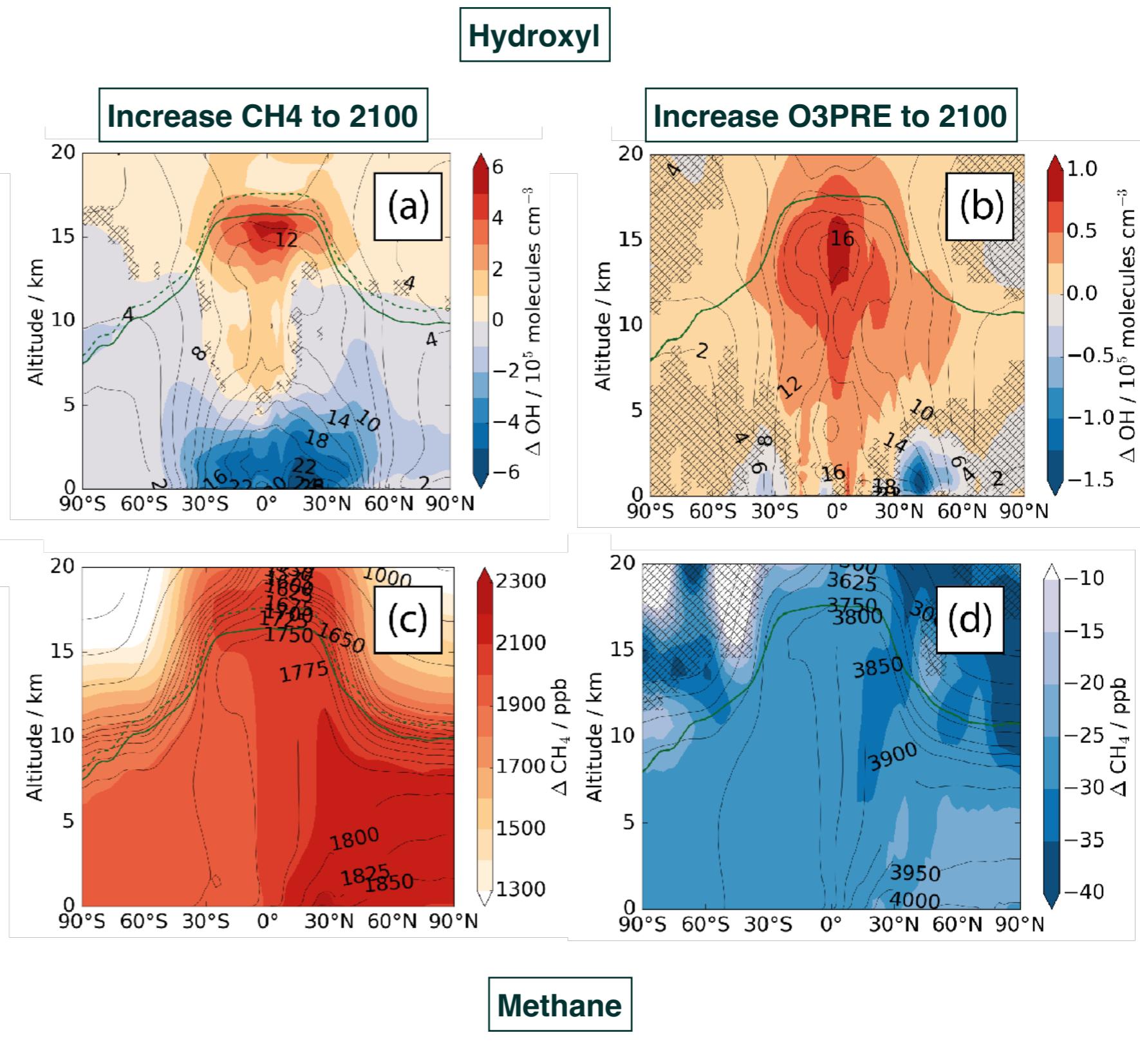
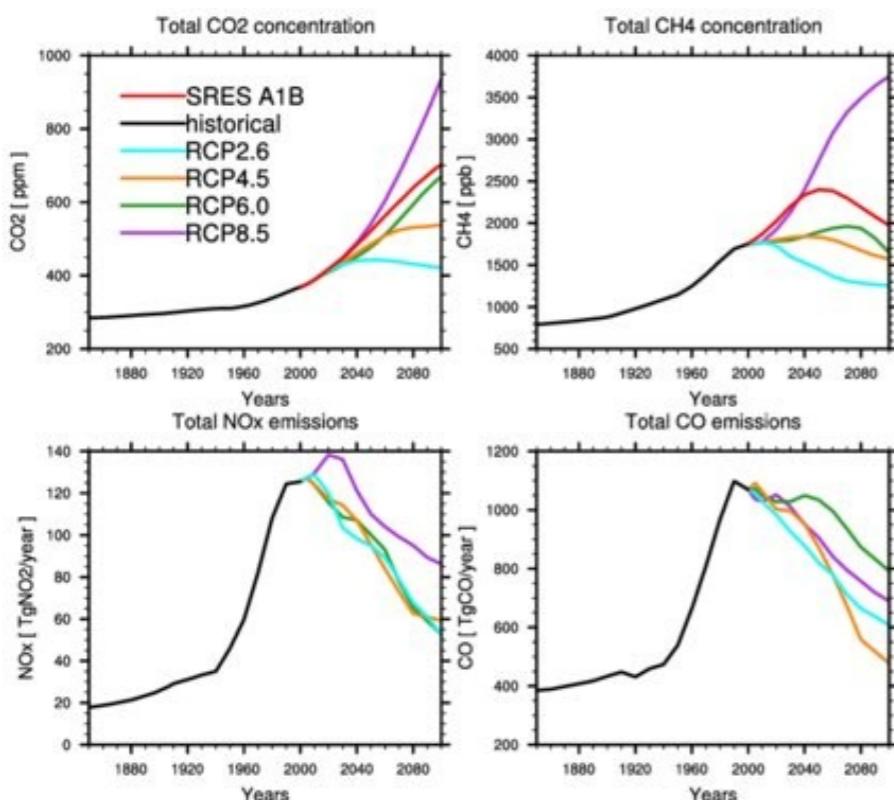
ΔCC with respect to year 2000

- OH – warmer, wetter atmosphere so OH increases
- Changes largest in tropical FT
- More OH means less CH₄ (and $k(OH+CH_4)$ increases as T increases)
- Methane decrease large everywhere cf Year 2000.
- Methane lifetime reduced from 9 to 6 years.



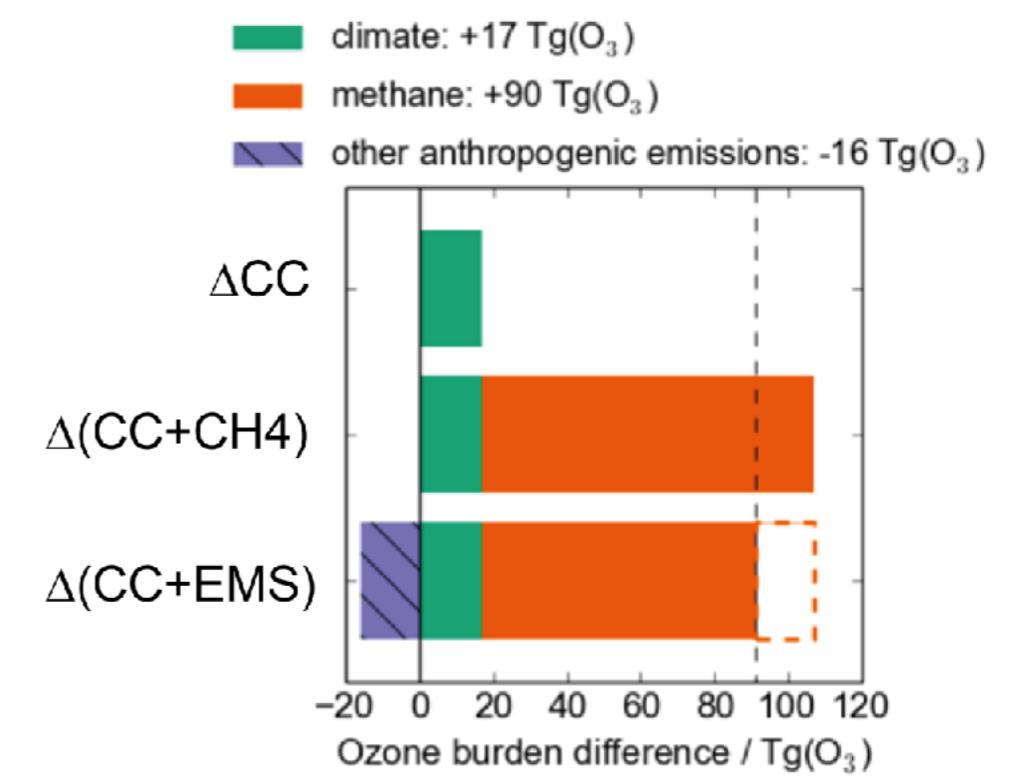
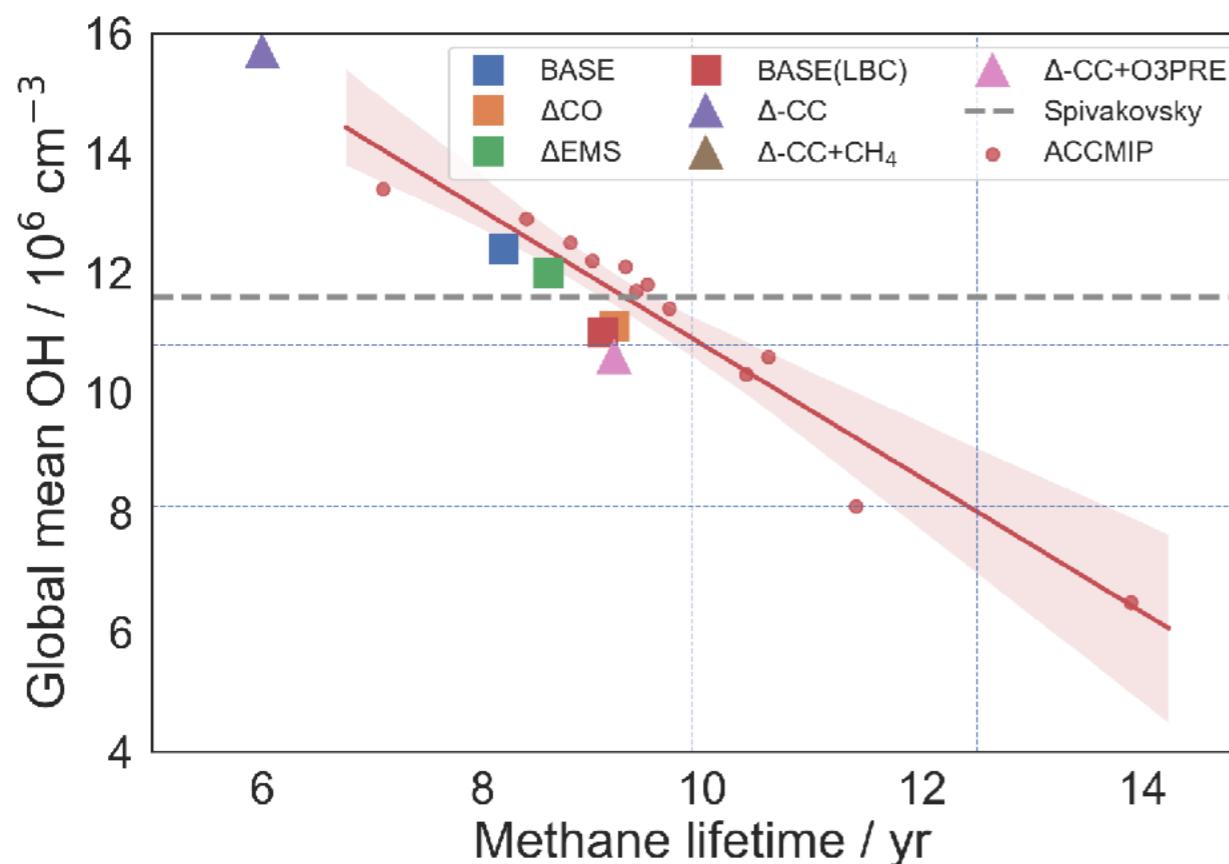
What happens to tropospheric oxidising capacity in future climate?

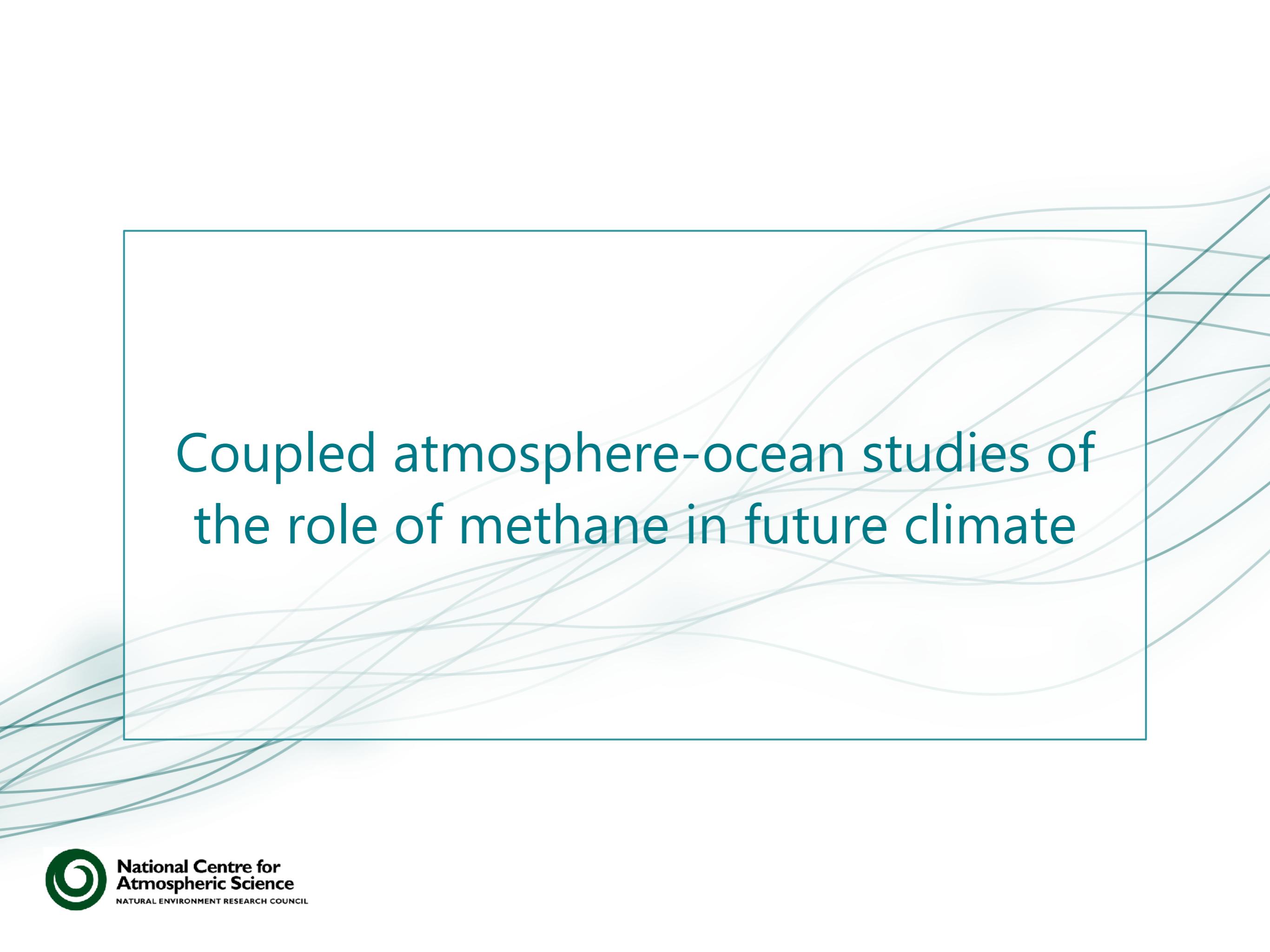
- Increasing CH₄ emissions to RCP8.5 levels gives
 - Large increase in CH₄
 - Large decrease in OH
- Increasing CO and NOx to RCP8.5 levels gives
 - Smaller change in OH
 - Small decreases in CH₄



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 CH₄ 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



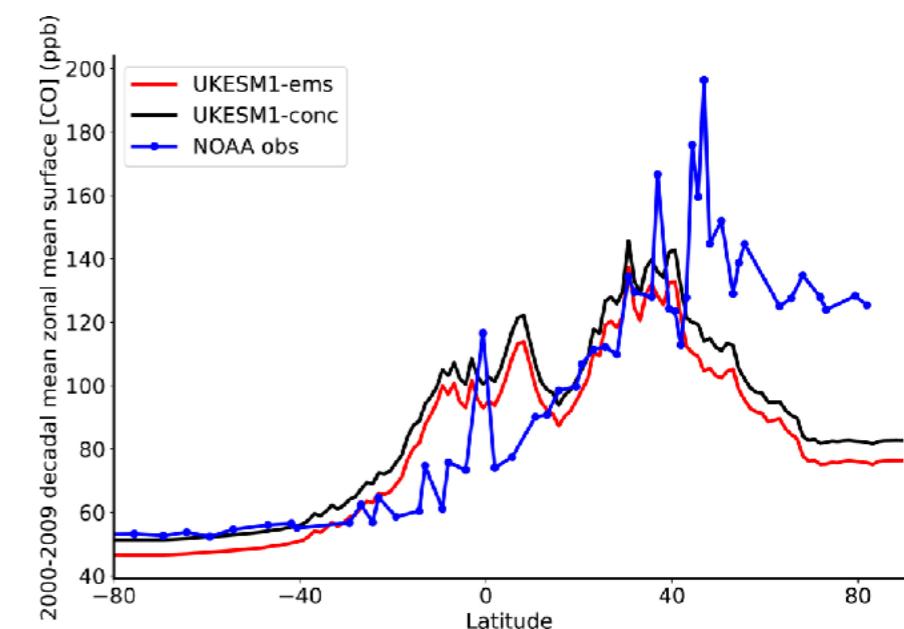


Coupled atmosphere-ocean studies of the role of methane in future climate

Methane emissions in a fully coupled atmosphere-ocean model

- See Folberth et al. for further details of UKCA-CH4
- Anthropogenic/biomass burning emissions from CEDS database but now JULES wetland emissions coupled.
 - Adds response of wetlands to changing climate + IAV from precipitation.
 - Interactive deposition

Methane budget in UKCA-CH4					
Wetlands	197	217 [177–284]	175 [142–208]	147 [102–179]	180 [153–196]
Anthropogenic	333	331 [304–368]	335 [273–409]	334 [321–358]	332 [312–347]
Wildfires	11	n/a	n/a	3 [1–5] ^a	n/a
Termites	20	11 [2–22]	n/a	9 [3–15]	n/a
Oceanic sources	21	18 [2–40]	n/a	13 [9–22]	n/a
Methane hydrates	9	0	n/a	2 [0–5]	n/a
Sinks					
Total chemical loss	549	604 [483–738]	528 [510–538]	595 [489–749]	505 [459–516]
Tropospheric OH	525	528 [454–617]	n/a	553 [476–677]	n/a
Tropospheric O(1D)	1	n/a	n/a	n/a	n/a
Stratospheric OH, O(1D)	23	51 [16–84]	n/a	31 [12–37]	n/a
Tropospheric Cl	n/a	25 [13–37]	n/a	11 [1–35]	n/a
Soil uptake	31	28 [9–47]	32 [26–42]	30 [11–49]	34 [27–41]
Overall Budget					
Sum of sources	591	678 [542–852]	553 [526–569]	703 [500–842]	552 [488–590]
Sum of sinks	580	632 [592–785]	550 [514–560]	625 [500–798]	540 [486–556]
Imbalance	11	n/a	3 [-4–19]	78	3 [-10–38]
Atmospheric growth	9.3	n/a	6	n/a	5.8 [4.9–6.6]



JAMES | Journal of Advances in Modeling Earth Systems^{*}

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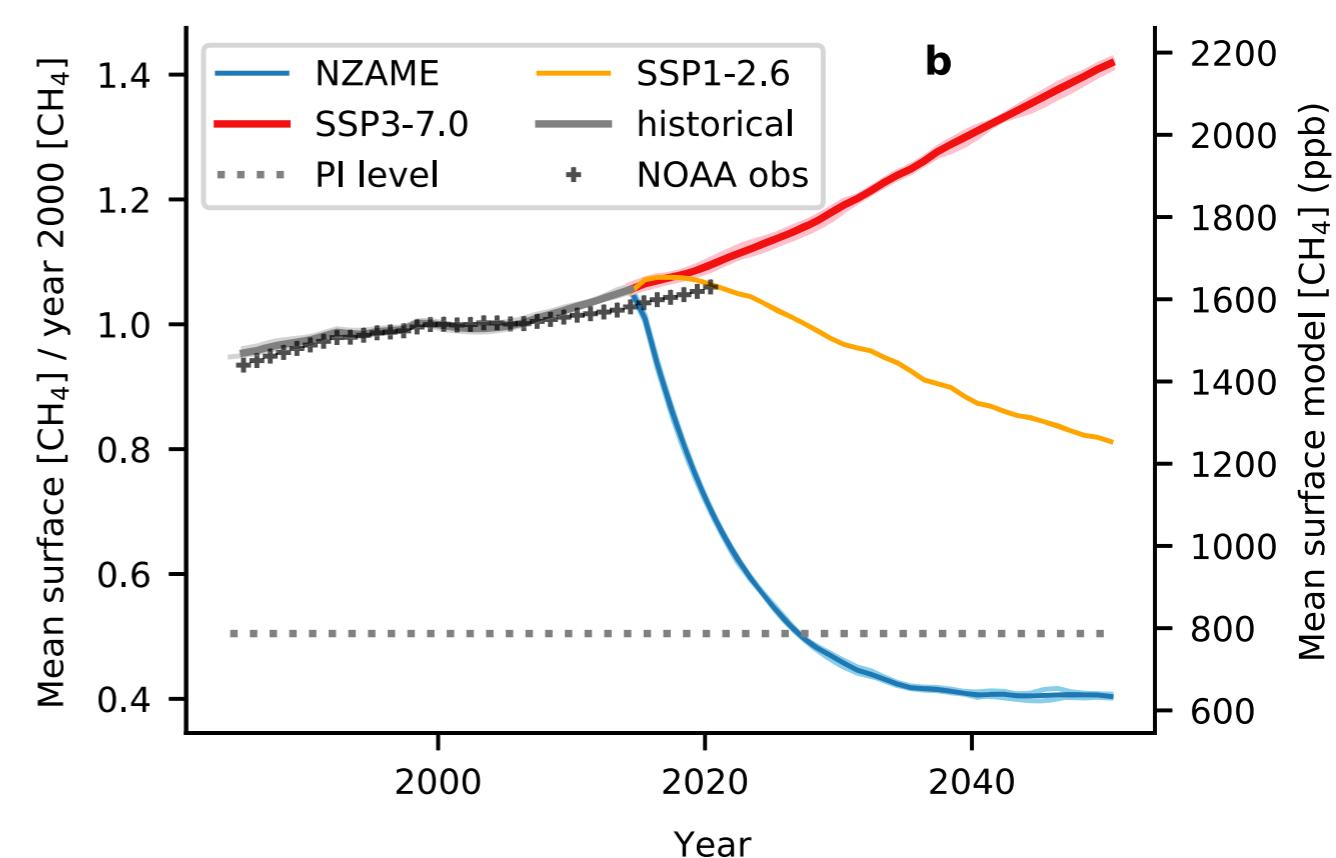
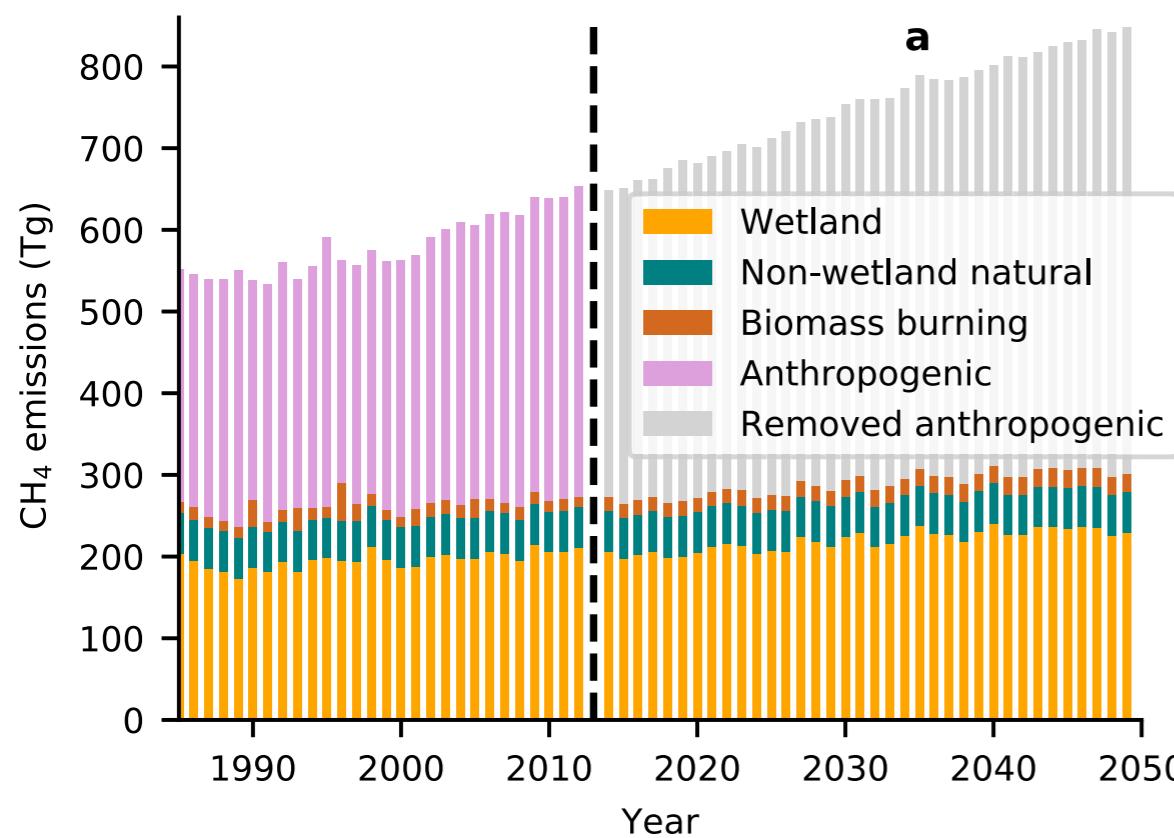
Description and Evaluation of an Emission-Driven and Fully Coupled Methane Cycle in UKESM1

G. A. Folberth , Z. Staniaszek, A. T. Archibald, N. Gedney, P. T. Griffiths, C. D. Jones, F. M. O'Connor, R. J. Parker, A. A. Sellar, A. Wiltshire

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Methane emissions in a fully coupled atmosphere-ocean model

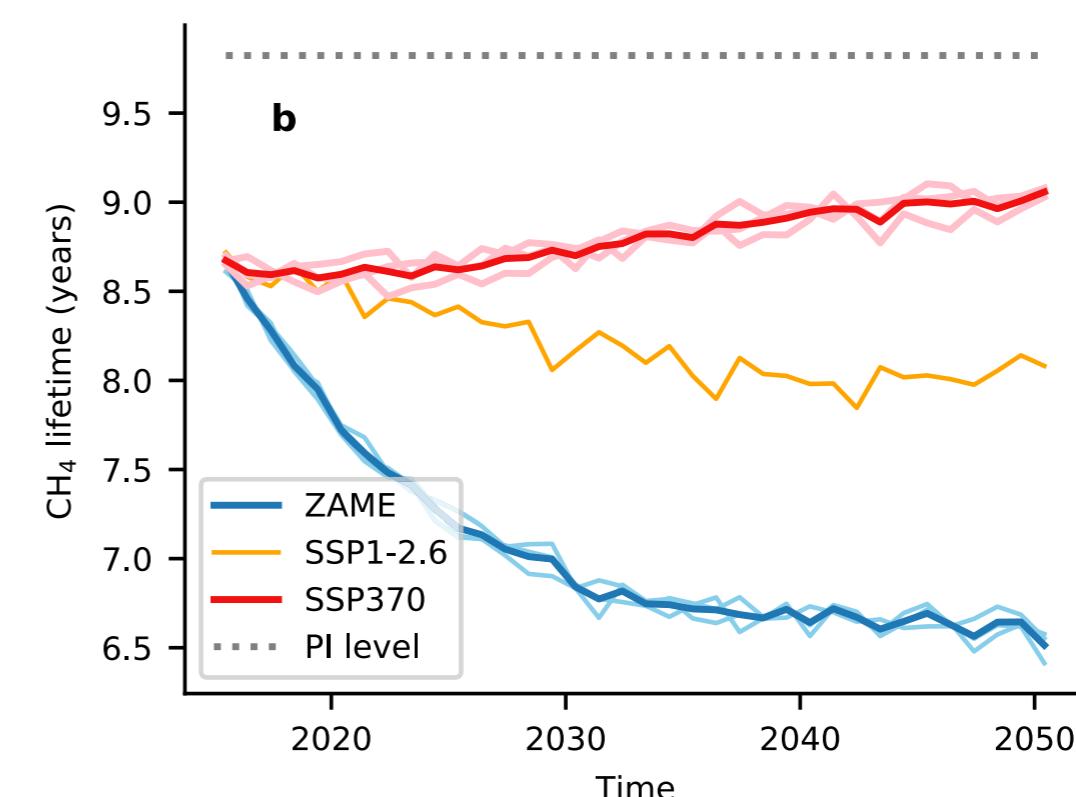
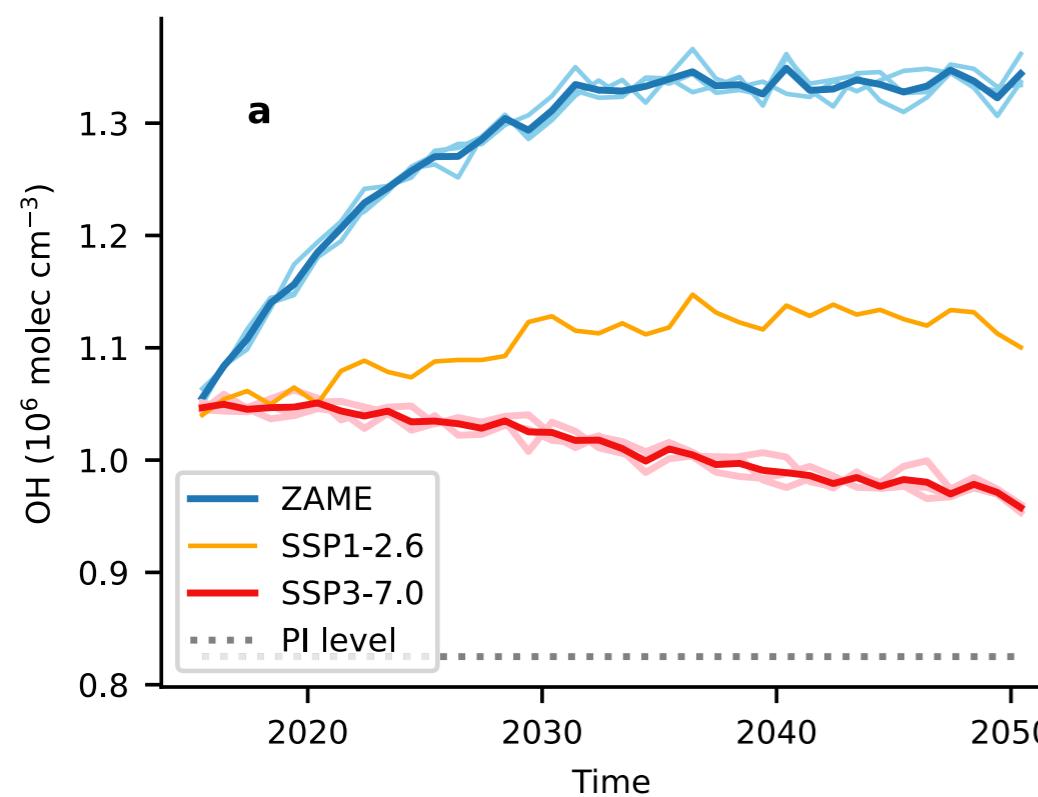
- What are the risks of unconstrained future methane emissions?
- For an upper bound, set anthropogenic emissions to net-zero - “NZAME” scenario
- Comparison with SSP3-7.0 and SSP1-2.6



- Comparison with SSP3-7.0 and SSP1-2.6 allows them to function as a counterfactual
 - What are the risks of methane emissions?
 - What are the benefits of constraining future methane emissions?

The role of future anthropogenic methane emissions in air quality and climate

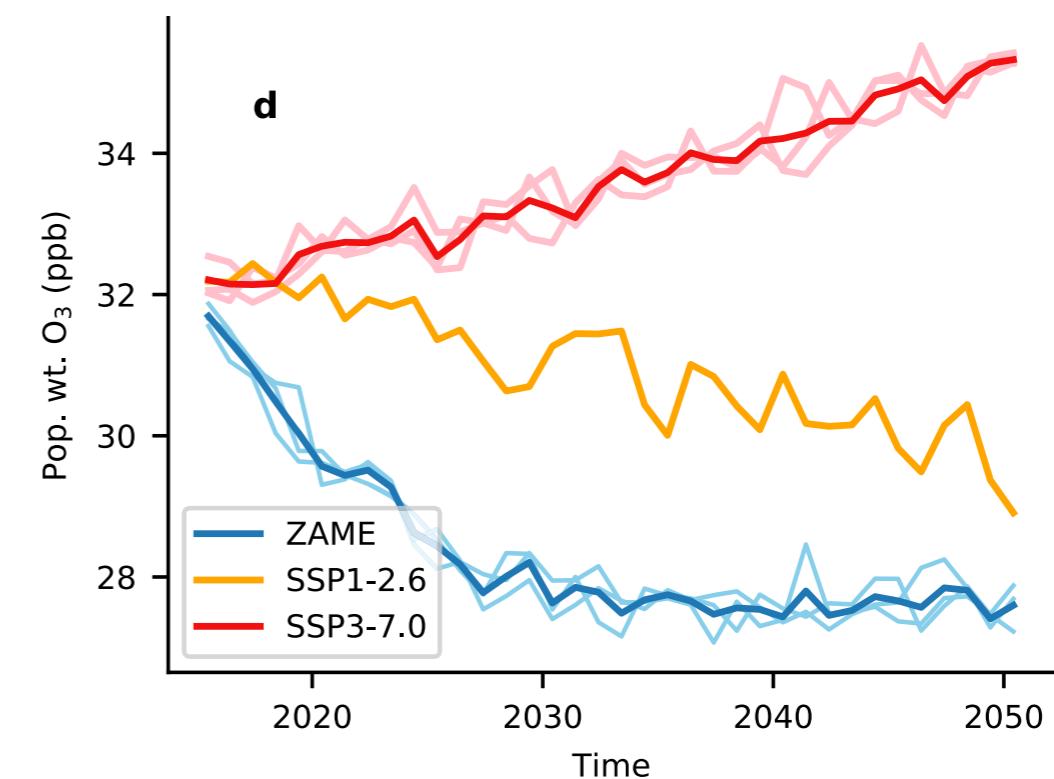
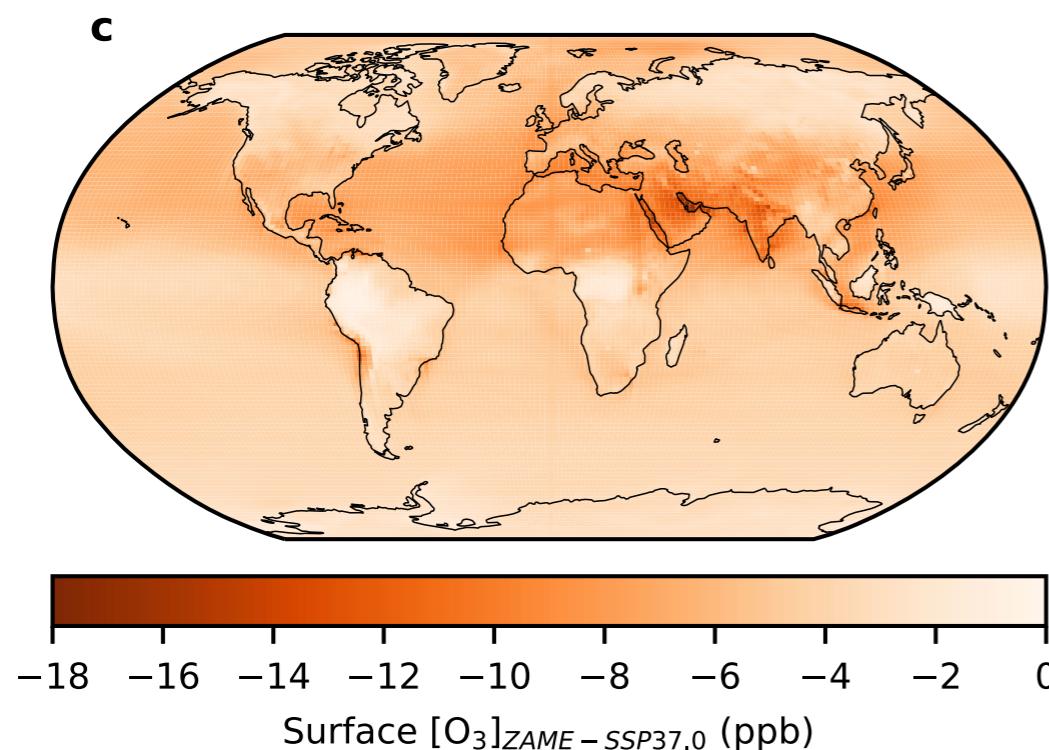
- What are the impacts of lower methane emissions on OH and methane lifetime?
- OH increases significantly - warmer climate, wetter, more OH production, increase of 30%
- Methane lifetime declines rapidly



- Comparison with SSP3-7.0 and SSP1-2.6 allows them to function as a counterfactual
 - What are the risks of methane emissions?
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The role of future anthropogenic methane emissions in air quality and climate

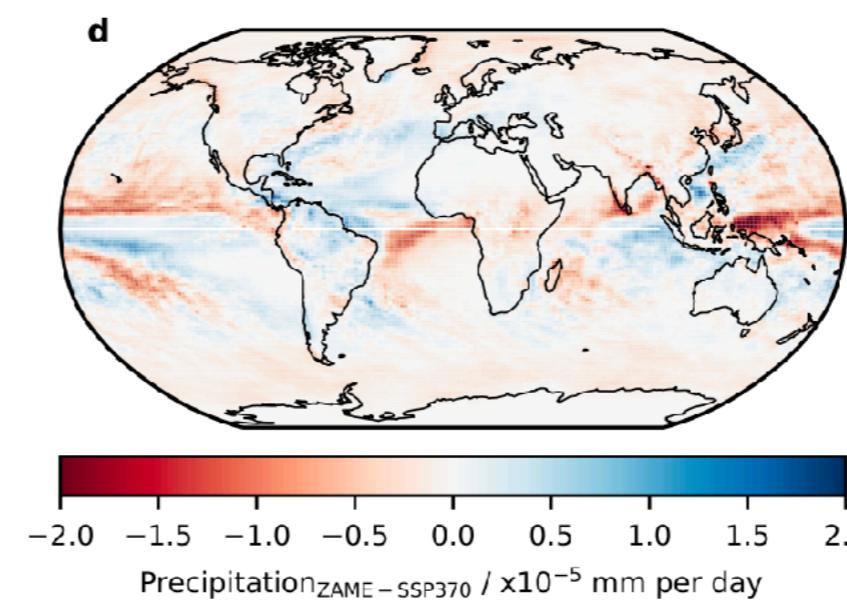
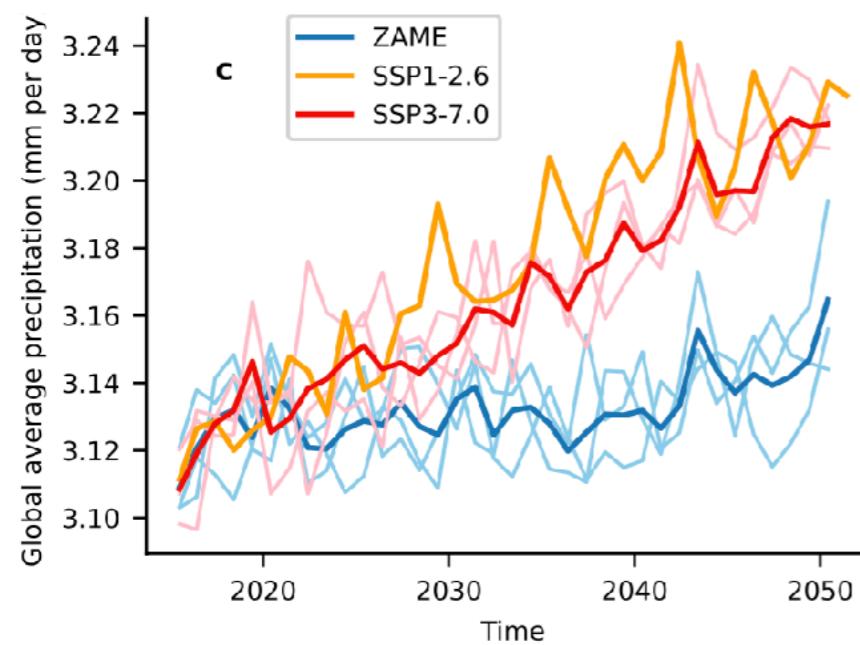
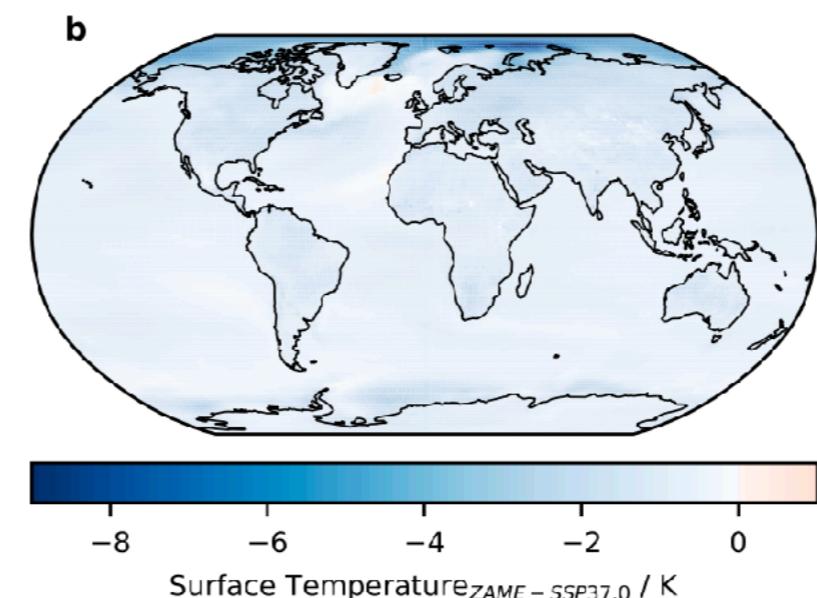
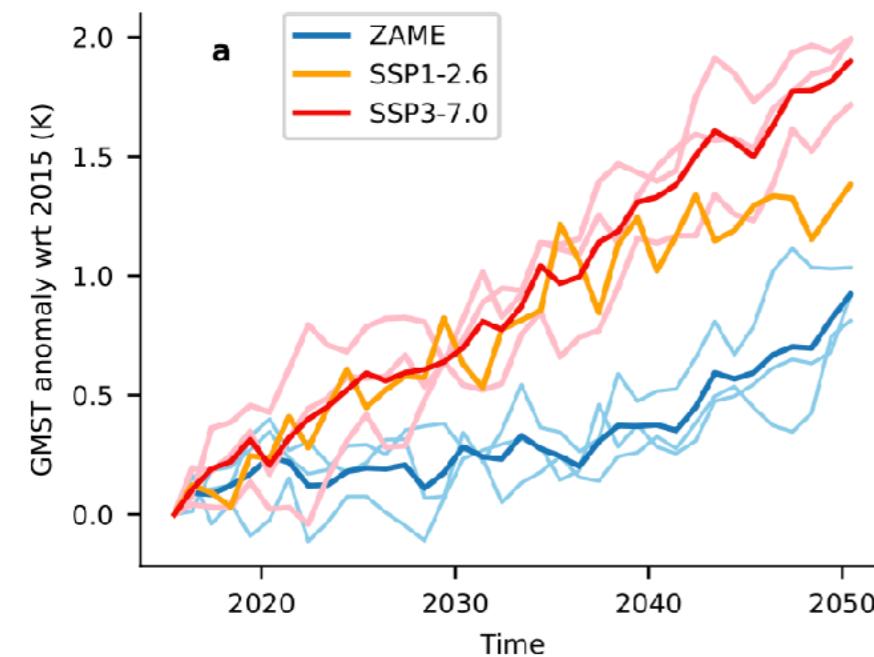
- What are the impacts of lower methane emissions on OH and methane lifetime?
- CH₄ is an important O₃ precursor - decreased CH₄ → decreased O₃
- Decline across the globe, strong regional variations



- Weighting the ozone field by human exposure shows ~10% decline
- Projected decrease in AQ-related mortality of the order of 500k per year

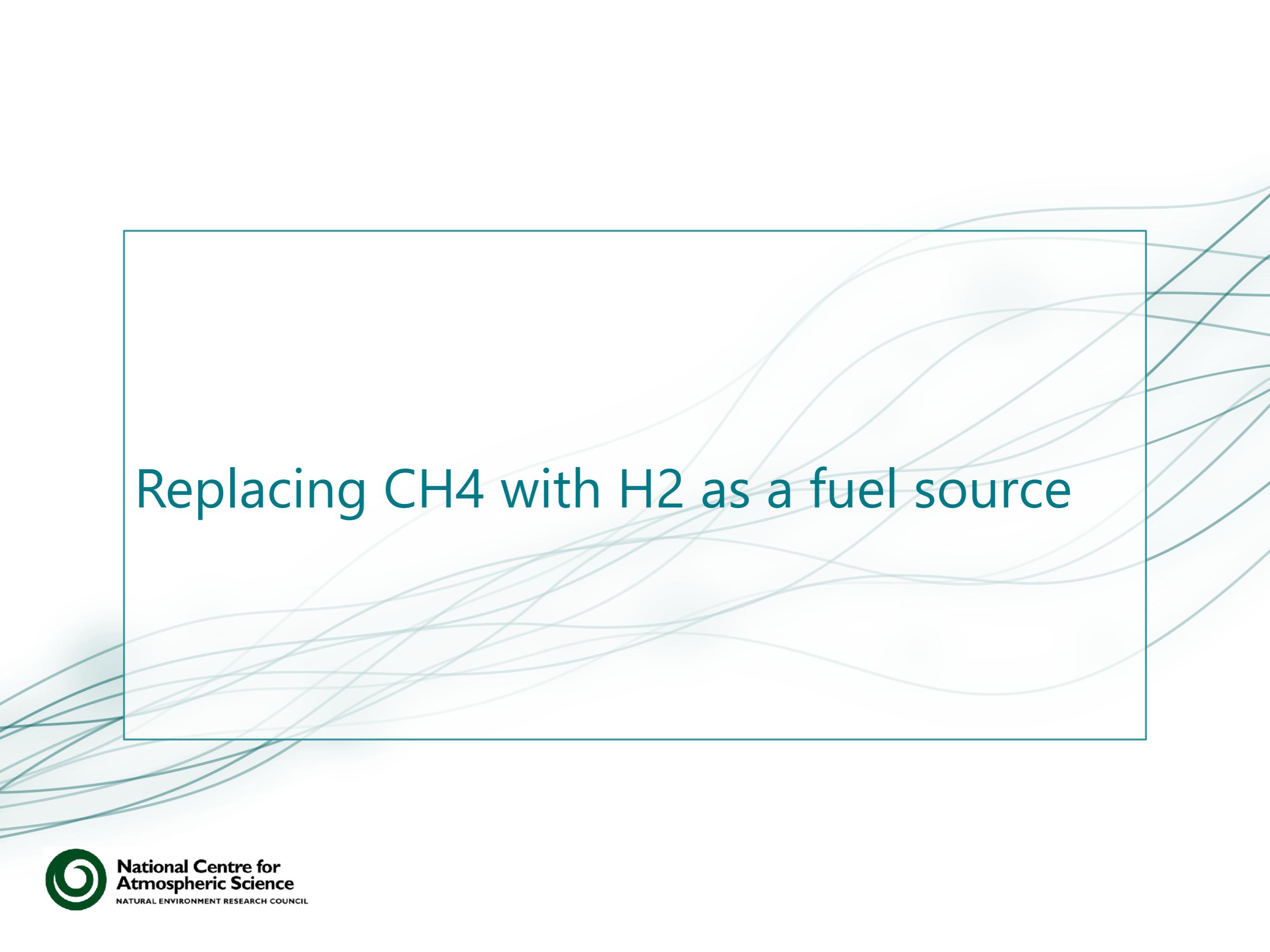
The role of future anthropogenic methane emissions in air quality and climate

- What are the impacts of lower methane emissions on global surface temperature
- Decreased radiative forcing → $\Delta T = 0.5 \text{ K}$
- Decline across the globe, strong regional variations, Arctic amplification



Conclusions 2/3 - CH₄ in future climate

- Net Zero Anthropogenic Methane Emissions ('NZAME') experiment shows that the maximum feasible (...) reduction in emissions would
 - Prevent approx. 0.5°C of global surface temperature rise
 - Reduce tropospheric ozone levels (any improvement in WHO 8hr levels?) with benefits to O₃ RF.
 - Leads to more OH - shorter methane lifetime, reduced GWP.



Replacing CH₄ with H₂ as a fuel source

Atmospheric chemistry of H₂

- Present-day sources

Sources	Fossil fuel	Biomass burning	N ₂ fixation	Photochemical production	Total
Strength / Tg per yr	17 ± 4	15 ± 6	9 ± 3	36 ± 7	76 ± 10

- Present day sinks

Sinks	Photochemical removal	Uptake by soil	Total
Strength / Tg per yr	23 ± 8	50 +30 / -20	70 ± 30

- Low temperature combustion in the atmosphere (without the ‘squeaky pop’)
- Giving an atmospheric burden of 155 Tg H₂, a mean mixing ratio of 550 ppb and a lifetime of 2.5 years
- H₂ affects
 - ozone levels (H₂ oxidation functions as a source of ozone)
 - methane levels (H₂ removes OH, decreasing the size of the CH₄ sink)
 - aerosol and cloud properties via removal of OH and modification of sulfate aerosol number

Aims of the study

UK Govt Business, Energy and Industrial Strategy commissioned a study into the impacts of a ‘global’ hydrogen economy.

Specific questions:

- Impact of H₂ on tropospheric and stratospheric composition
- Calculation of GWP for inter-comparison of interventions
- Calculation of radiative forcing

Specific issues:

- Design of scenarios - esp. energy mix, leakage rates, lack of detail on proposed technology (!)
- Uncertainty in process-level data, esp. H₂ deposition at global scale

Scenario design - thanks to Nicola Warwick

Buildings sector (~15 % Global Energy Demand, GED):

- Assume all fossil energy from the buildings sector converted to H₂ (~10% of GED)

Transport (~20% GED):

- Half of energy demand for global transport from light duty vehicles
- H₂ avoids land use/air quality impact of biofuels & limited range/recharging times of EVs
- Assume 50% road transport converts to H₂ (~10 % of GED)

Power generation (~40% GED, 25% of GED from gas & coal)

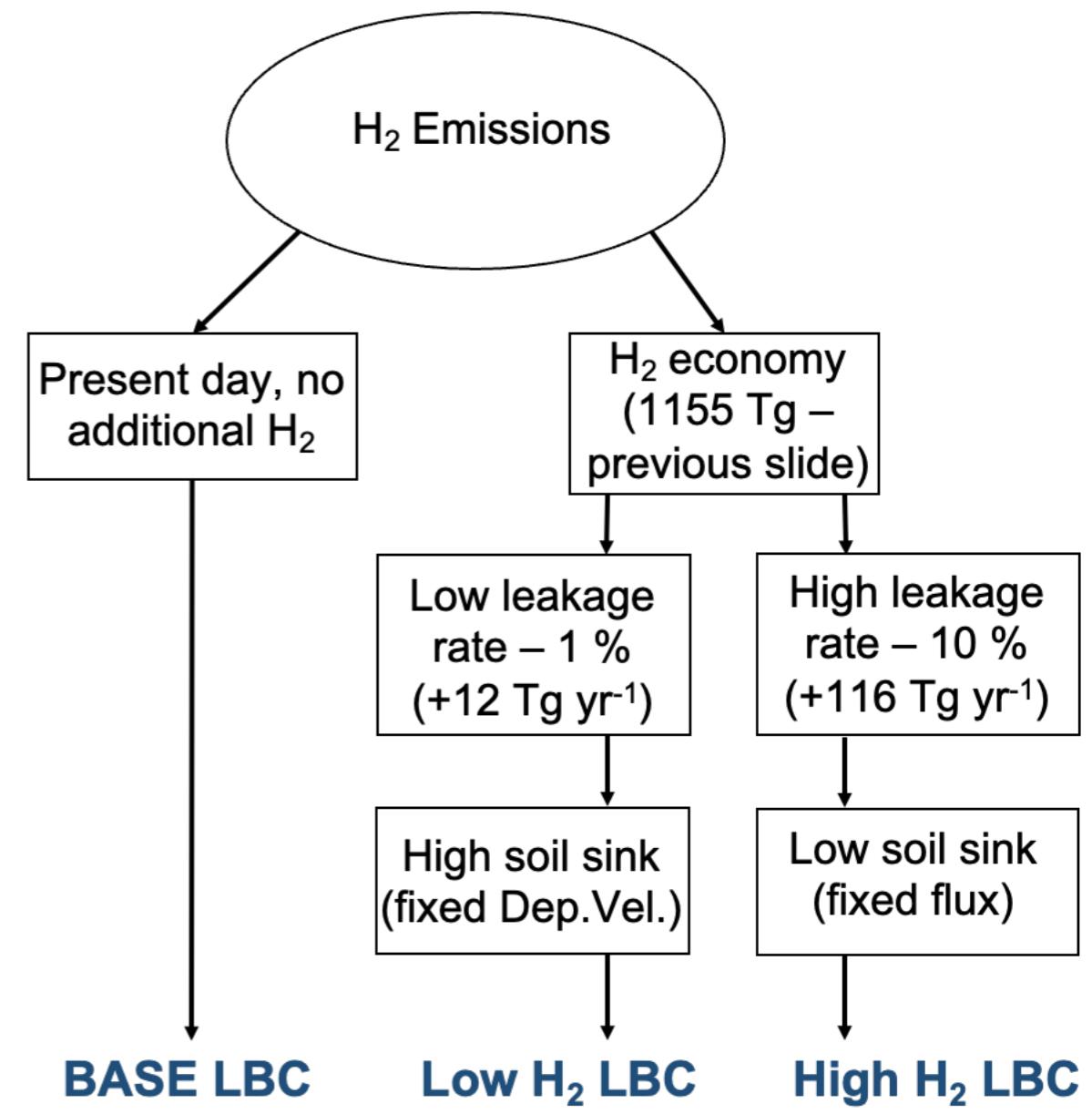
- Global capacity for power generation from H₂ was ~0.01 % of total capacity in 2015
- Assuming continuing growth trend could be 0.5 % by 2030
- Assume 5% of power generation from H₂ (~2 % of GED)

Total energy from H₂ = 22% of GED (BP) = 3.9 x 10¹³ kWh = 1155 Tg H₂

Atmospheric chemistry of H₂

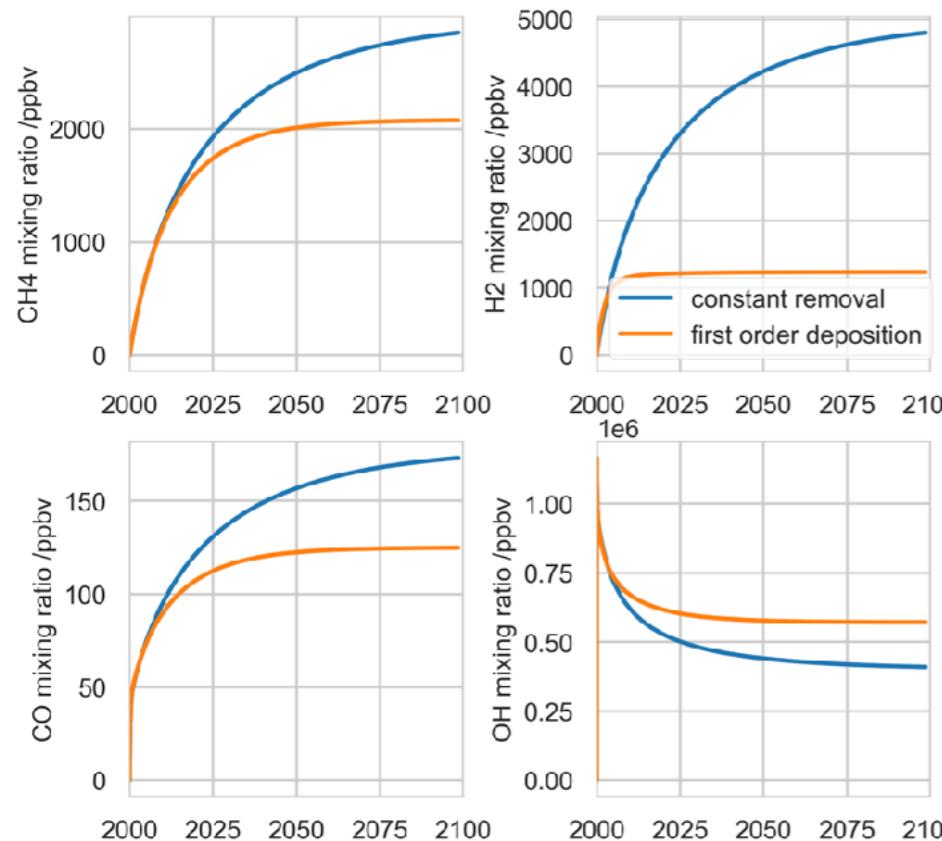
H₂ leakages rates of 1 % and 10 %

- Very few estimates available: truck transport – 1 to 2.3 %, US gas grid ~ 1 %, but likely to be underestimated, gas grids – up to 4.5%
- Schultz et al., 2003 used 3%, Tromp et al., 2003 used 10-20%, Warwick et al. 2004 used 1-10 %.
- No emissions associated with H₂ generation
- Soil sink: include both fixed flux and fixed deposition velocity to account for uncertainty in how the soil sink may respond to large changes in H₂
- Future 2050 scenarios based on today's energy demand (use the same H₂ lower boundary conditions as the present day scenarios)
- Future co-benefit emissions CO, NOx and NMHCs calculated based on sector % replaced
- CO₂ changes not considered



Chemical effects of enhanced H₂ levels

Leak rate of 96 Tg per year



1. H₂ leakage emissions increase as a result of a move to H₂ as a fuel source.
 - 750 ppb, 1000 ppb and 2000 ppb (approx increase from 76 Tg to >200 Tg H₂ emissions)
2. Adoption of H₂ as a fuel source means that there is a co-benefit of reduction in other anthropogenic emissions such as CO, NOx, NMVOCs.
3. Adoption of H₂ as a fuel source means CH₄ emissions decrease and other other anthropogenic emissions such as CO, NOx, NMVOCs
 - Consider this under low-H₂ and high-H₂ leakage scenarios

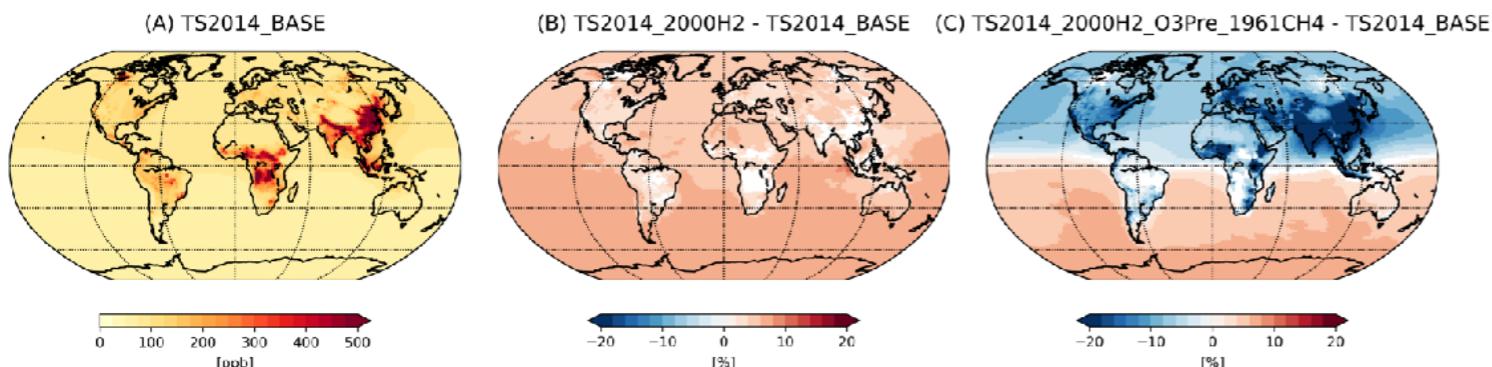
Scenario	H ₂	CH ₄	Notes
TS2014_BASE	500	1835	2000-2014 climatology from CMIP6 historical
TS2014_750H ₂	750	1835	As TS2014_BASE
TS2014_1000H ₂	1000	1835	As TS2014_BASE
TS2014_1500H ₂	1500	1835	As TS2014_BASE
TS2014_2000H ₂	2000	1835	As TS2014_BASE
TS2014_1500H ₂ _2058CH ₄	1500	2058	As TS2014_BASE
TS2014_2000H ₂ _2171CH ₄	2000	2171	As TS2014_BASE
TS2014_O3Pre	500	1835	Reduced O ₃ precursor emissions
TS2014_1500H ₂ _O3Pre	1500	1835	Reduced O ₃ precursor emissions
TS2014_2000H ₂ _O3Pre	2000	1835	Reduced O ₃ precursor emissions
TS2014_O3Pre_1652CH ₄	500	1652	Reduced O ₃ precursor emissions
TS2014_1000H ₂ _O3Pre_1756CH ₄	1000	1756	Reduced O ₃ precursor emissions
TS2014_2000H ₂ _O3Pre_1961CH ₄	2000	1961	Reduced O ₃ precursor emissions

- UKCA in concentration-driven, atmosphere-only mode, 2014 timeslice: faster spin-up and use of fixed SSTs means can calculate ERF.
- Lots of scenarios to cover the range of potential H₂ scenarios.
- Use a box model to estimate H₂ levels resulting from various parametric uncertainties such as deposition.

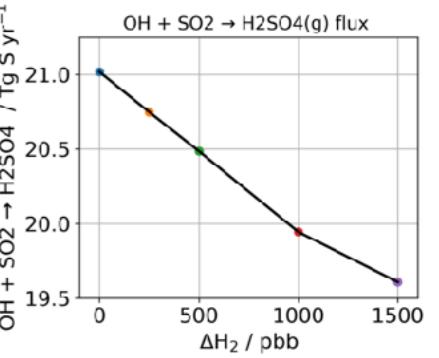
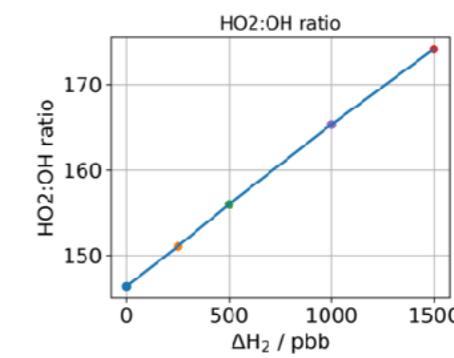
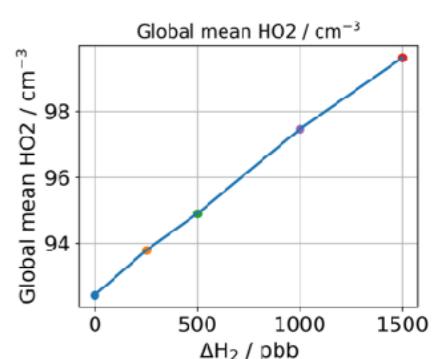
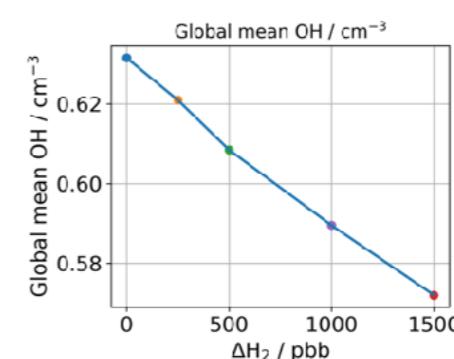
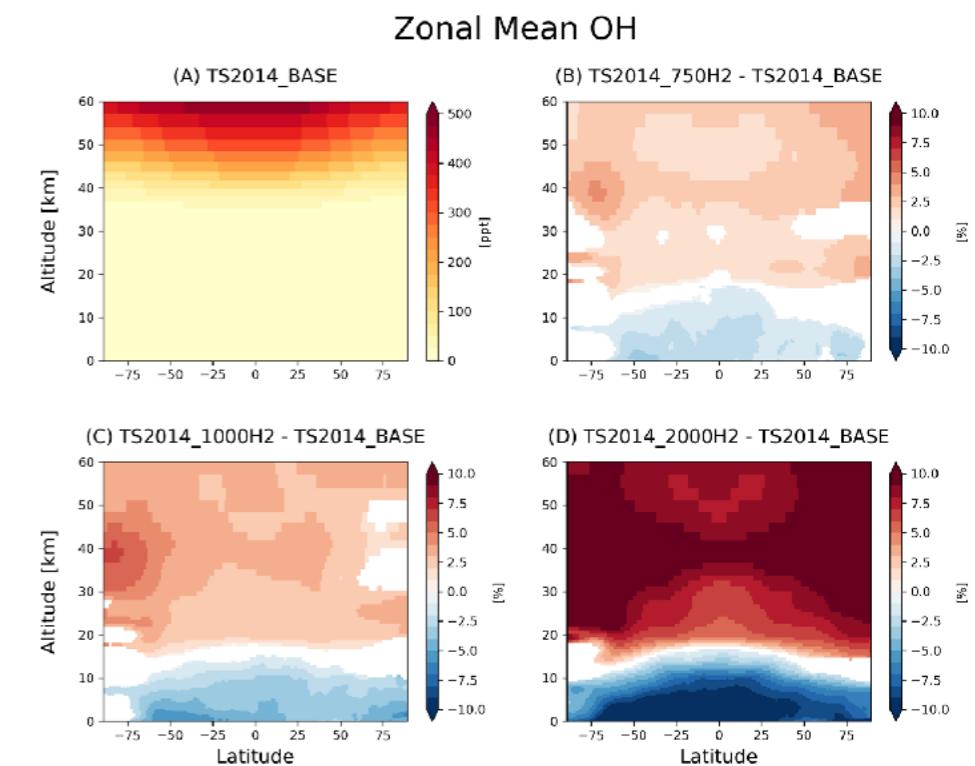
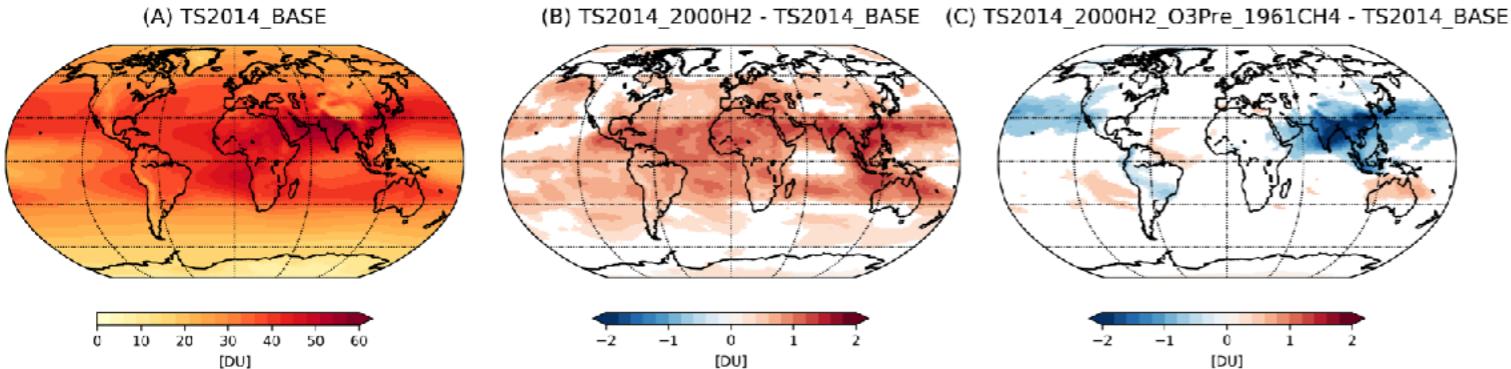
Chemical effects of enhanced H₂ levels

- Increased H₂ levels suppress OH via increase in OH + H₂ → H₂O + H
 - Suppressed OH → enhanced CO
- Increased HO₂ levels enhance HO₂ via H+O₂+M → HO₂
- Change in OH:HO₂ ratio, and changes to both O₃ Prod (HO₂+NO) and O₃ Loss (e.g. HO₂+O₃)
- Tropospheric ozone column mostly ozone increases, H₂ functioning as O₃ precursors

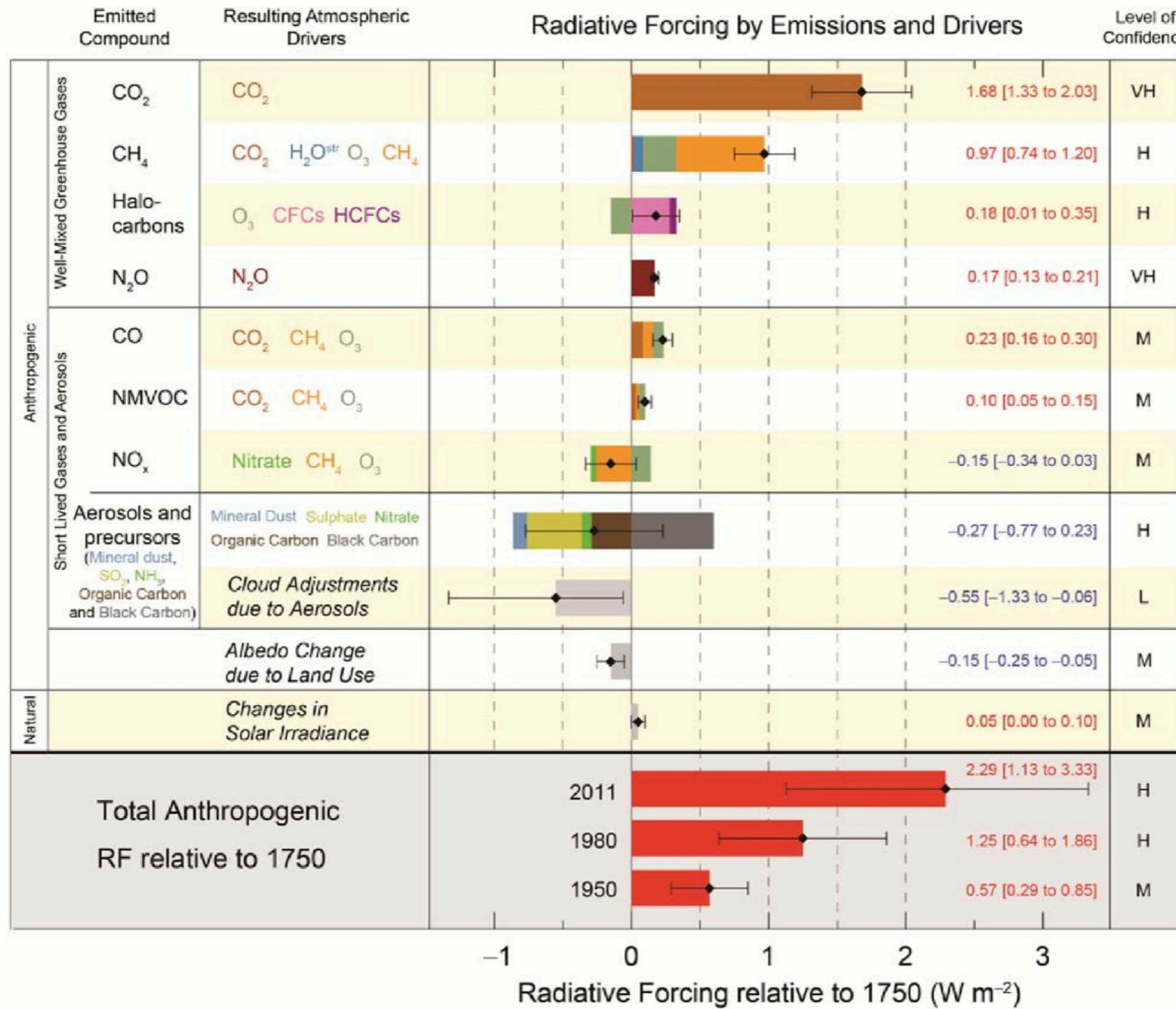
Surface CO



Tropospheric Column Ozone



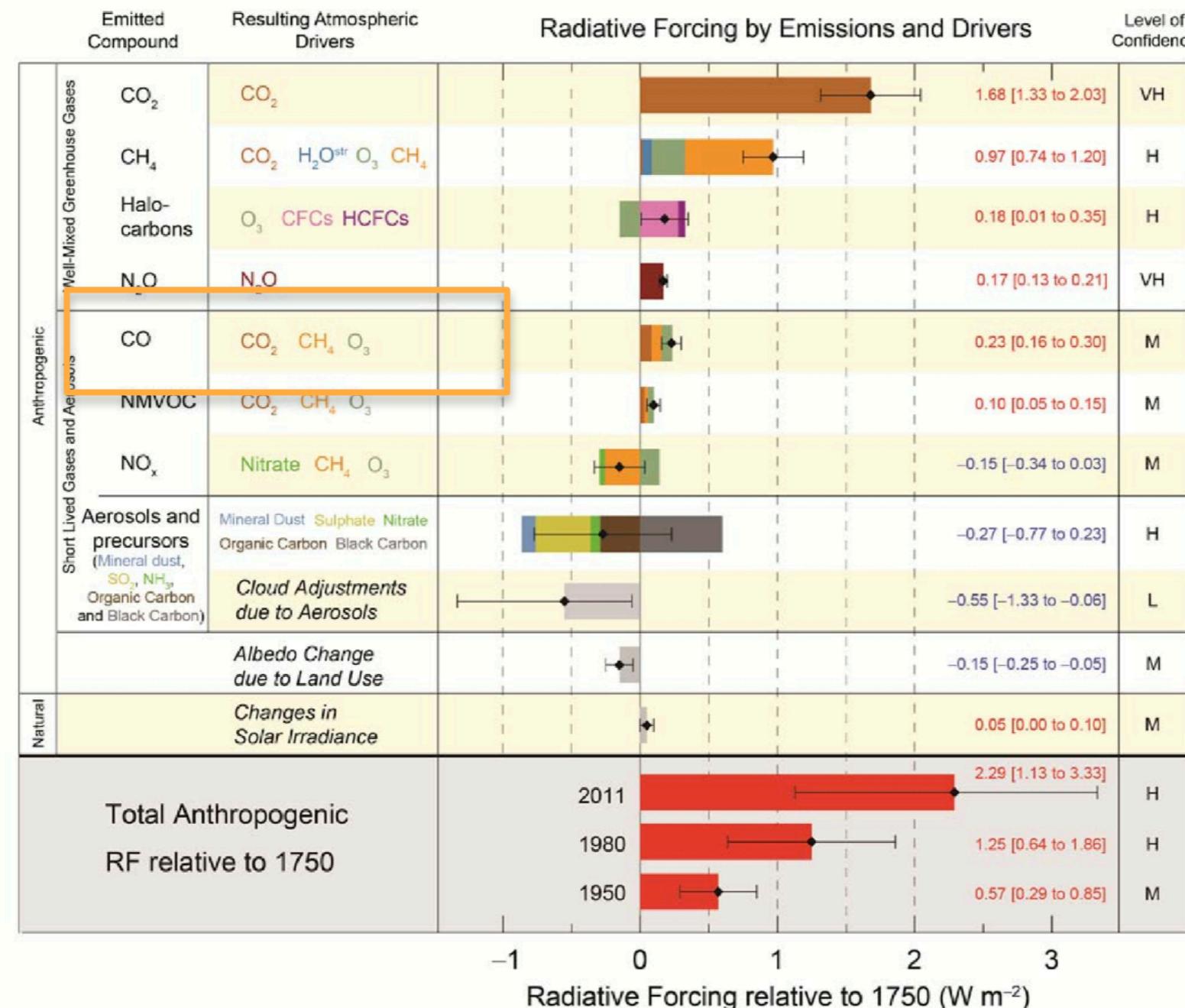
Effective radiative forcing - CMIP5 picture



- The radiative forcing can be used to estimate the resulting global temperature change via

$$\Delta F = \lambda \Delta T$$

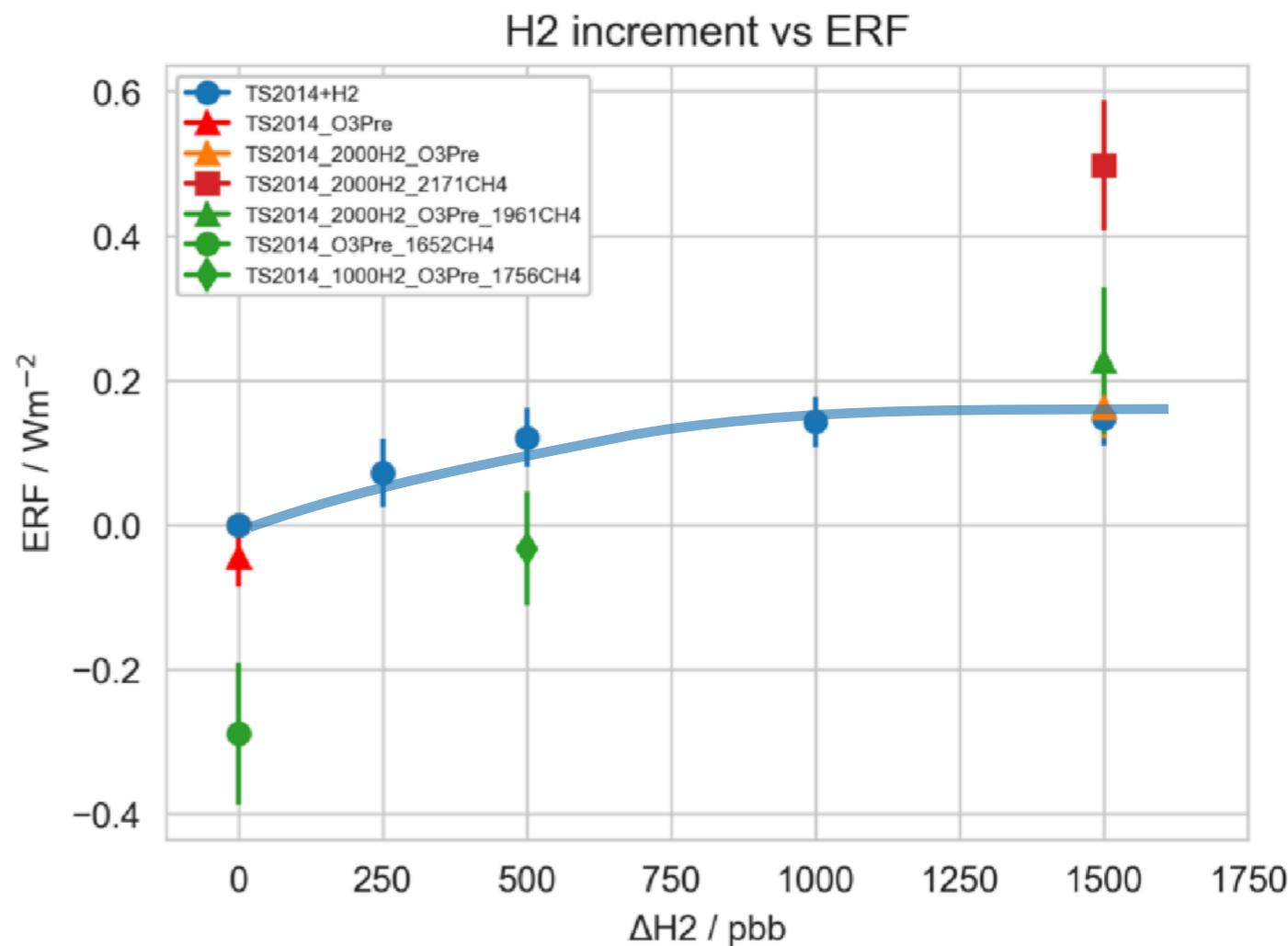
Effective radiative forcing - anthropogenic emissions



- Anthropogenic emissions affect the concentration of radiatively important gases such as CH_4 , O_3
- Oxidants such as O_3 also affect aerosol formation which can also perturb cloud properties
- $\text{ERF} = \Delta\text{CS} + \Delta\text{CRE} - \text{clear-sky (GG-dominated in the long wave)} + \text{Cloud Radiative Effects}$

Scenarios studied - what is the effect of H₂ fugitive emissions?

- Experiments with varying H₂ concentration in the atmosphere.
- The radiative forcing increases with increasing H₂ concentration, and is positive = a warming. Maybe a plateau?
- For the highest leak rates (an effective tripling of the global atmospheric H₂ source) ERF = $0.15 \pm 0.08 \text{ Wm}^{-2}$ which is approx 5% of the warming effect of CO₂
- Increasing H₂ levels see increases in methane lifetime and in ozone burden - can expect positive GG forcing.
- Increasing H₂ levels leads to decreased OH
- Potential impacts on stratospheric ozone.
- How to attribute the RF increase?

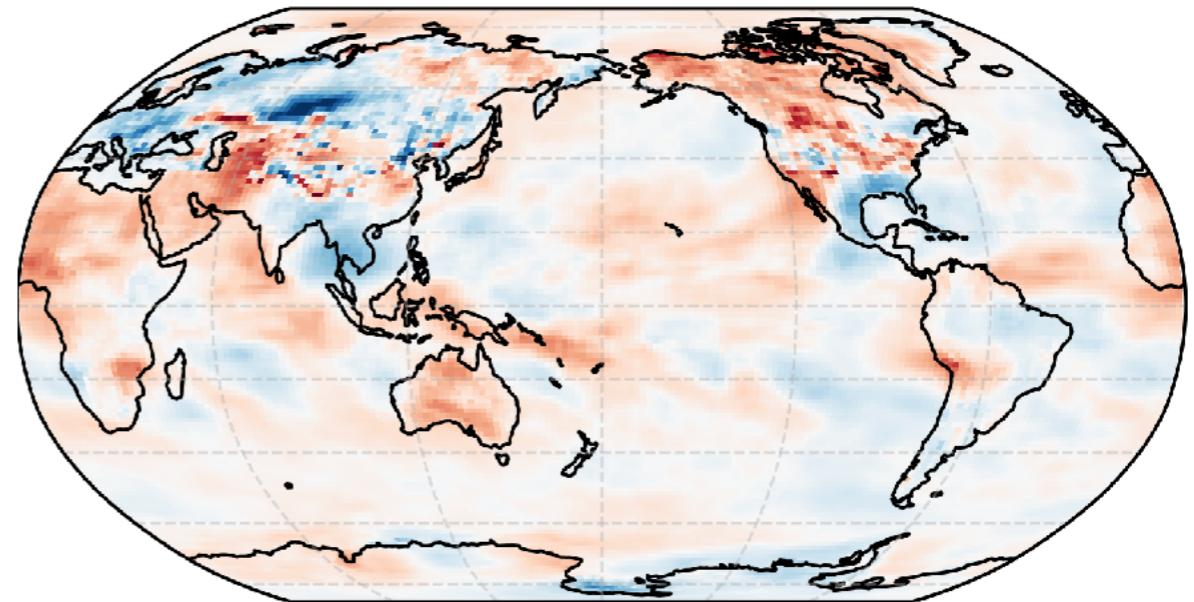


Experiment	H ₂ LBC	OH	TAU CH ₄	O ₃ Burden
	ppb	10^6 cm^{-3}	Years	Tg
Base	500	1.22	8.48	348.6
TS2014_750H ₂	750	1.20	8.67	347.3
TS2014_1000H ₂	1000	1.18	8.83	349.7
TS2014_2000H ₂	2000	1.11	9.46	353.5

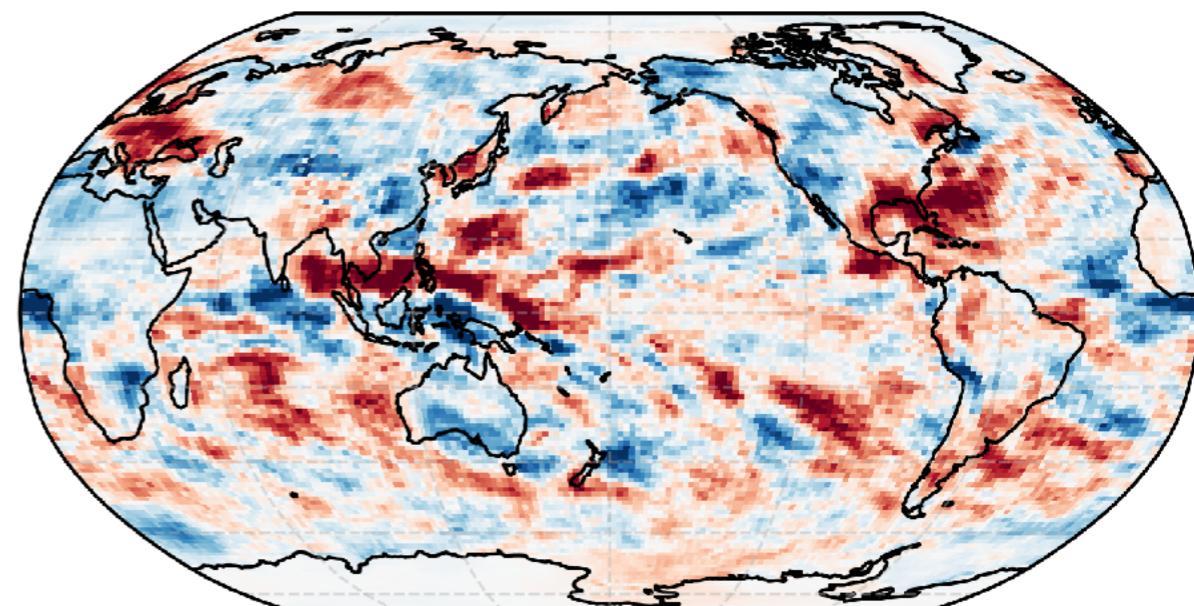
Breaking ERF down into clear-sky and cloud effects

- Can break the change in radiative flux at the top of the atmosphere down further. Focusing here on the 2000 ppb H₂ case.
- The change in the greenhouse gas forcing, a.k.a. the Clear Sky (cloud-free) forcing
 - ERF = 0.103 Wm⁻²
 - Presumably from the small increase in tropospheric ozone (a greenhouse gas)
- The change in the radiative properties of the clouds (global averaged effects)
 - $\Delta\text{CRE} = 0.036 \text{ Wm}^{-2}$
- Which can be broken down further
 - Shortwave $\Delta\text{CRE} = 0.068 \text{ Wm}^{-2}$
 - Longwave $\Delta\text{CRE} = -0.032 \text{ Wm}^{-2}$
- i.e. the clear sky forcing is of the same order as the cloud radiative effect

SW+LW clear-sky ERF = $0.103 \pm 0.027 \text{ Wm}^{-2}$



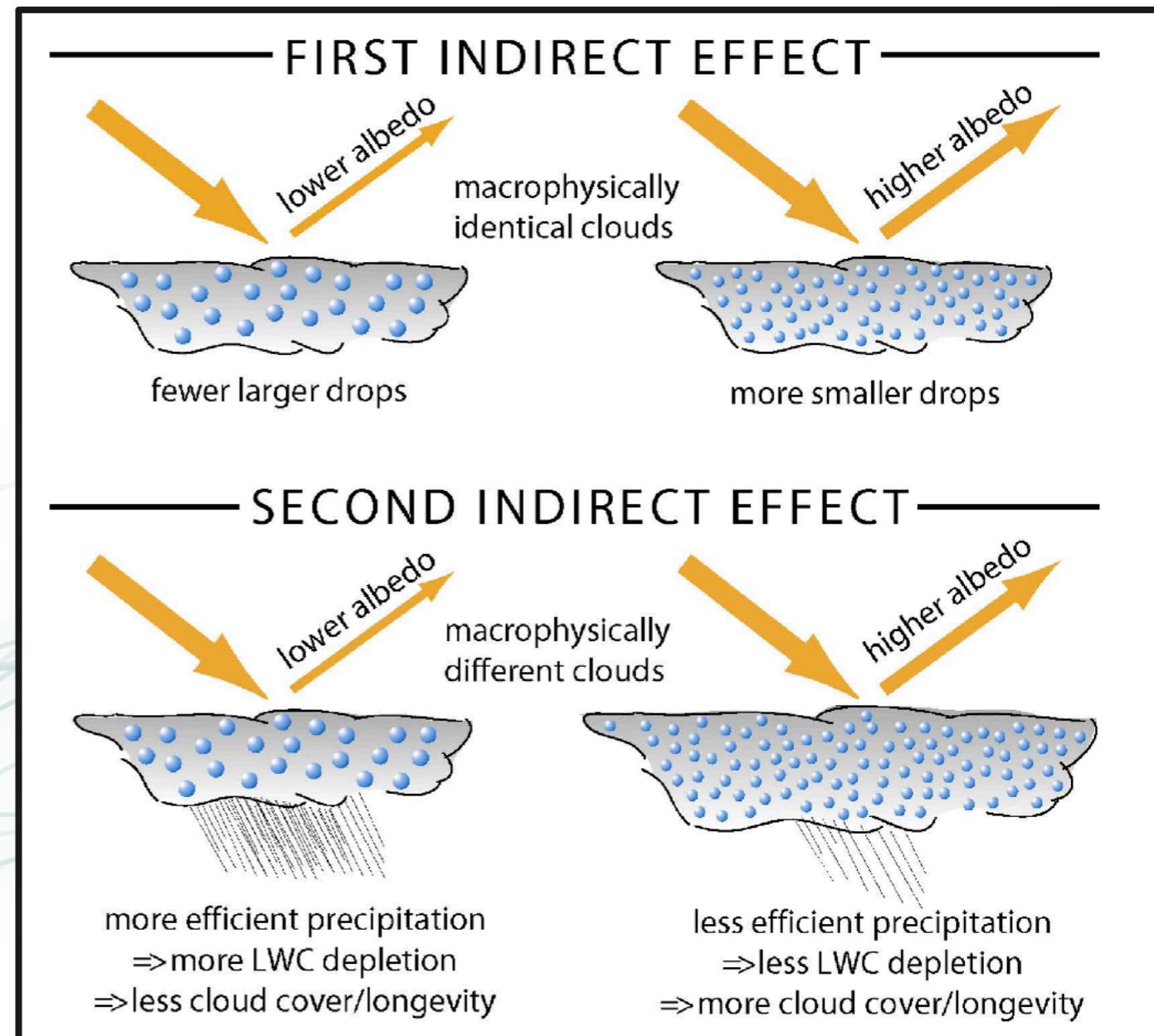
CRE SW = $0.068 \pm 0.040 \text{ Wm}^{-2}$



-2.00 -1.00 0.00 1.00 2.00

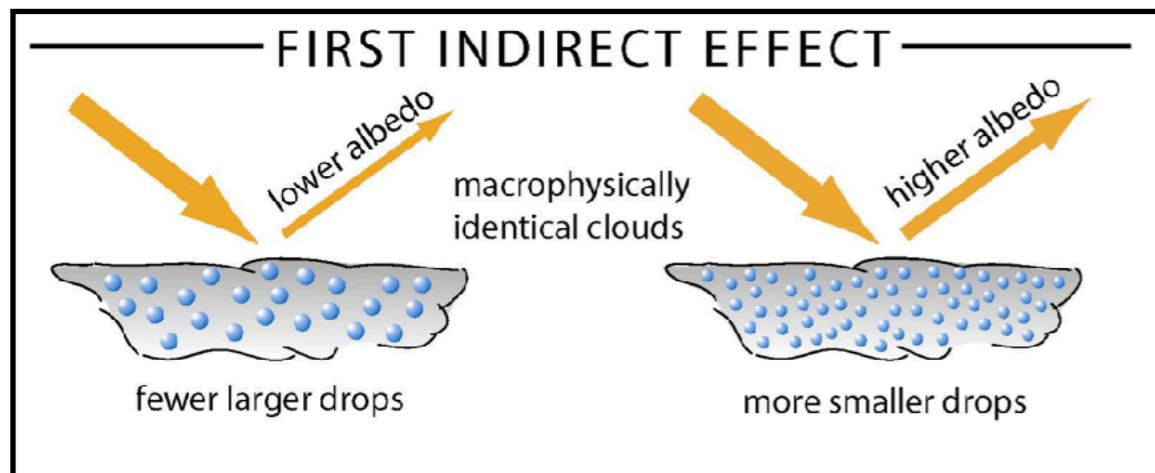
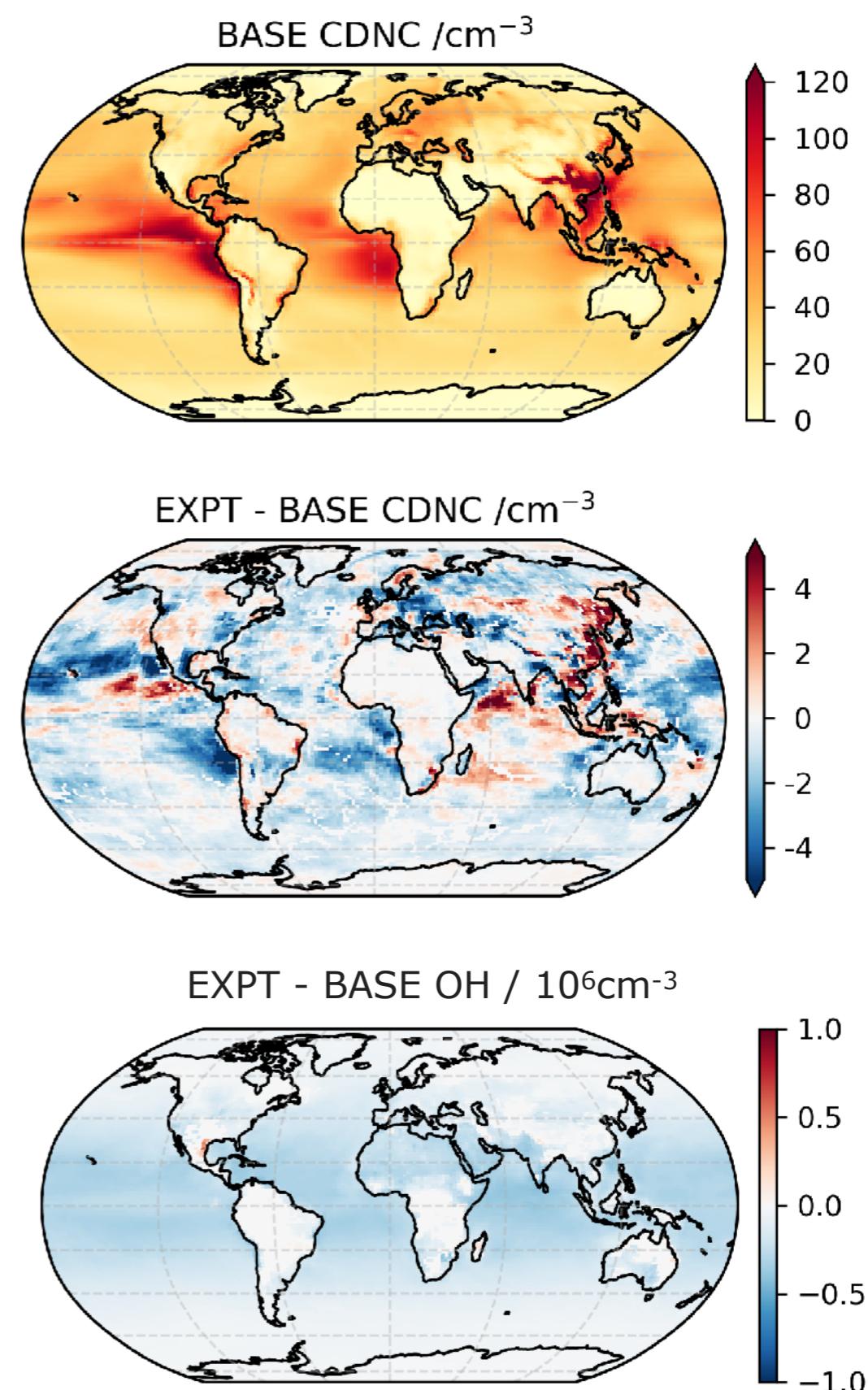
Cloud radiative properties respond to aerosol changes

- Aerosol (CCN) controlled by atmospheric oxidation of gases like SO₂, biogenic emissions, NO_x.
- Clouds form on the aerosol (CCN) present in the atmosphere
- The cloud properties are sensitive to the number of aerosols
 - more aerosol → more cloud droplets
- More droplets means
 - a brighter cloud
 - a longer cloud lifetime
- Leading to negative forcing (increased energy at the top of the atmosphere) and less energy reaching the surface



ERF - the coupling of gas phase oxidant to aerosol levels and cloud properties

- The additional H₂ has caused a decrease in cloud droplet number concentration (CDNC). Seen here as a decrease in cloud droplet number with respect to our low H₂ base case.
- We can associate this decrease with the lower levels of the OH free radical oxidant in the region where aerosol is formed. There are fewer aerosol particles as a result.
- The effect of elevated H₂ is to suppress OH, and this is having knock-on effects on aerosol and on other components (e.g. CH₄ and O₃).



Conclusions 3/3 - H₂ economy

- H₂ couples into the radiative budget of the Earth via its effect on atmospheric oxidants
 - Overall, H₂ functions as a source of ozone which is a greenhouse gas
 - H₂ is also a sink for OH, which is an important atmospheric oxidant
 - This affects the aerosol formation process - increased H₂ leads to less OH and so less efficient formation of CCN. This has decreases cloud albedo and is a positive forcing.
- From our studies of other scenarios, we conclude
 - Controlling H₂ fugitive emissions is important
 - The effect of H₂ on CH₄ can be strong - for 2000 ppbv H₂, the H₂ is affecting CH₄ lifetime and increasing CH₄ levels significantly.
 - H₂ use with strongly controlled leaks can lead to significant benefits, due to reduce co-emissions of CO, NO_x and NMVOCs.
 - Not all bad news: with reductions in CH₄ we may achieve a reduction in forcing of 0.3 Wm⁻².



Thank you

Table 1. Major global tropospheric sources and sinks of H₂ (Tg H₂ yr⁻¹) from various authors

	Novelli et al. (1999)	Hauglustaine and Ehhalt (2002)	Sanderson et al. (2003)	Rhee et al. (2006a)	Price et al. (2007)	Xiao et al. (2007)	This work
Fossil fuel	15 ± 10	16	20.0	15 ± 6	18.3	15 ± 10	11 ± 4
Biomass burning	16 ± 5	13	20.0	16 ± 3	10.1	13 ± 3	15 ± 6
Biofuel					4.4		
N ₂ fixation, ocean	3 ± 2	5	4.0	6 ± 5	6.0		6 ± 3
N ₂ fixation, land	3 ± 1	5	4.0	6 ± 5	0		3 ± 2
Photochemical production							
from methane	26 ± 9		15.2		24.5		23 ± 8
from VOC	14 ± 7		15.0		9.8		18 ± 7
total	40	31	30.2	64 ± 12	34.3	77 ± 10	41 ± 11
Sources total	77 ± 16	70	78.2	107 ± 15	73	105 ± 10	76 ± 14
Oxidation by OH	19 ± 5	15	17.1	19 ± 3	18	18 ± 3	19 ± 5
Soil uptake	56 ± 41	55	58.3	88 ± 11	55 ± 8.3	85 ± 5	60 ⁺³⁰ ₋₂₀
Sinks total	75 ± 41	70	75.4	107 ± 11	73	105 ^a	79 ⁺³⁰ ₋₂₀
Tropospheric Burden, Tg H ₂	155 ± 10	136	172 ^b	150 ^c	141	149 ± 23	155 ^d ± 10
Tropospheric Lifetime, yr	2.1	1.9	2.2 ^b	1.4	1.9	1.4	2.0

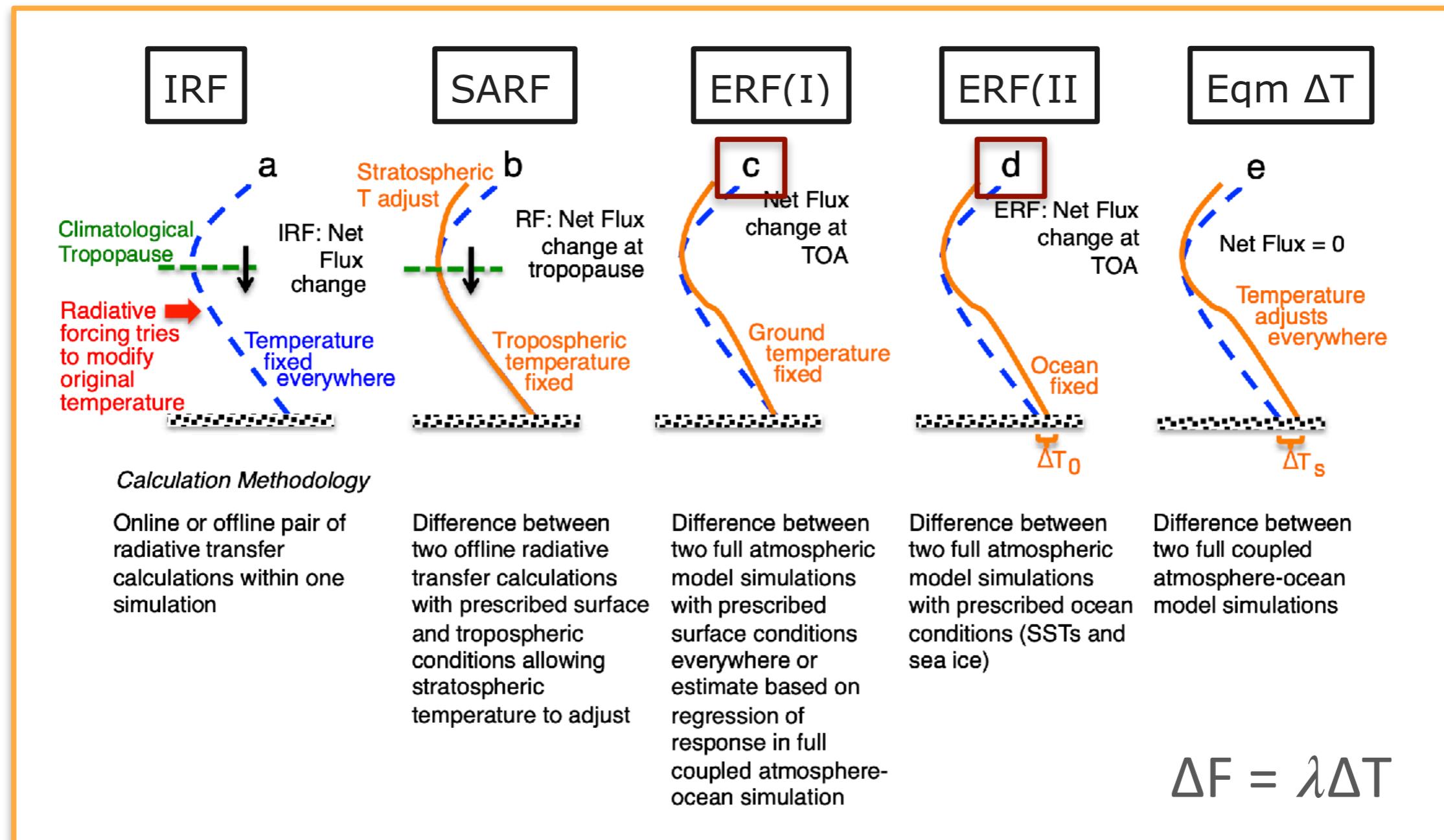
^aIncludes export to stratosphere of 1.9 Tg H₂ yr⁻¹.

^bModel domain reached 100 hPa; thus the burden includes about 1/2 of the stratosphere. Reduced to a troposphere holding 0.82 of the total air mass the burden would be 157 Tg H₂ and the tropospheric lifetime 2.0 yr.

^cCalculated from sources and lifetime.

^dFrom Novelli et al. (1999).

Effective radiative forcing - definitions



- Calculation of ERF (W m^{-2}) as the change in energy flux at the top of the atmosphere following a perturbation (natural or anthropogenic).
- ERF includes all the tropospheric and land-surface adjustments - all the responses on a short timescale that occur as a result of the forcing agent, distinct from the slow feedbacks that arise due to temperature perturbations.

Chemical effects of enhanced H₂ levels

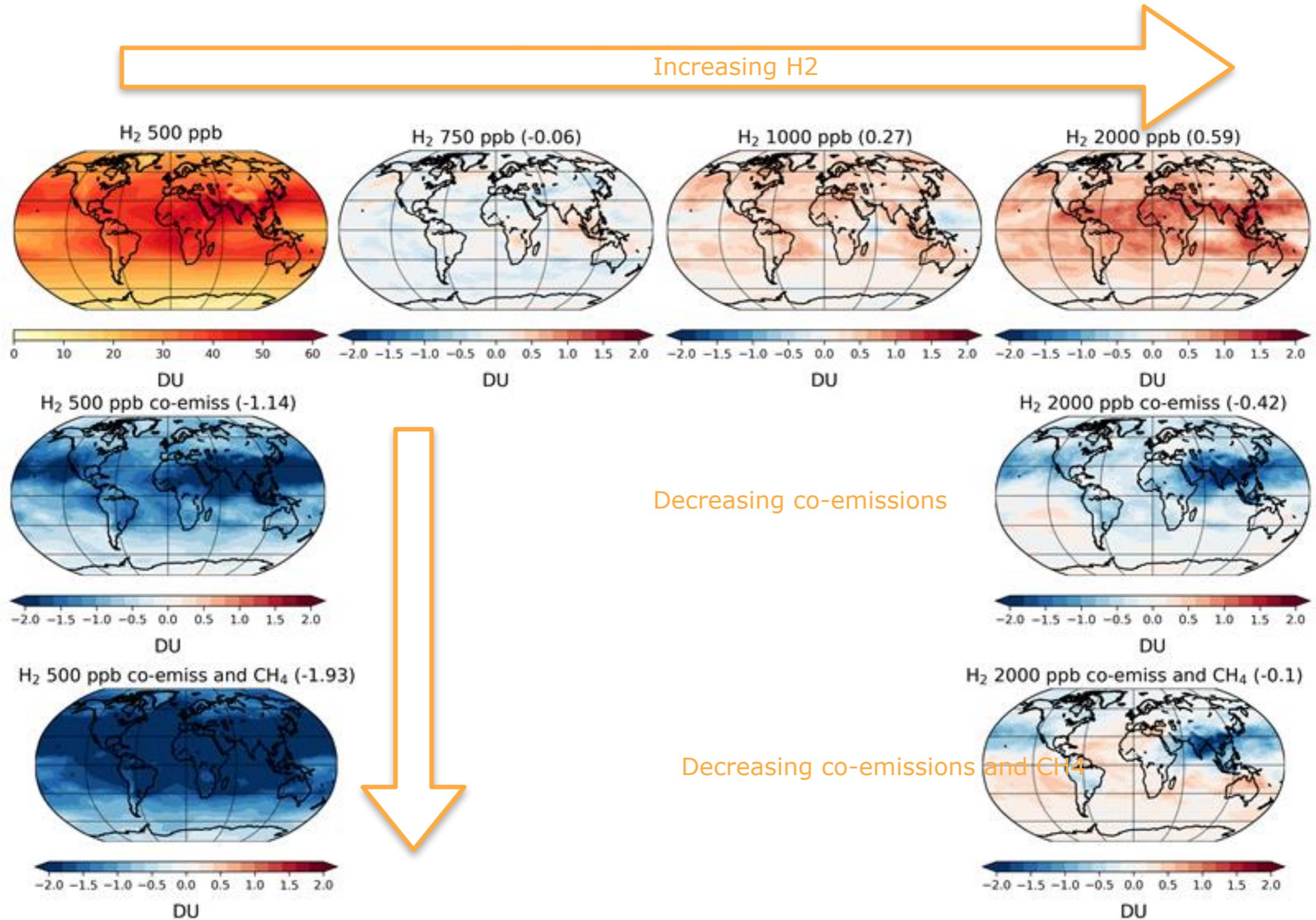


Figure by James Keeble