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

Paul Griffiths, National Centre for Atmospheric Science, Cambridge University

Visiting Researcher of ESS-1, JAMSTEC since July 2022

paul.griffiths@ncas.ac.uk

Grateful thanks to the people listed here for their contribution:

Zosia Staniaszek, Ines Heimann, Alex Archibald, John Pyle - **Cambridge University & NCAS**

James Keeble, Nicola Warwick, N. Luke Abraham - - **Cambridge University & NCAS**

Fiona M. O'Connor, Gerd Folberth - **Met Office Hadley Centre, UK**

Keith Shine - **Reading University, UK**

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

National Centre for Atmospheric Science
NATURAL ENVIRONMENT RESEARCH COUNCIL

paultgriffiths

SCAN ME

AMIP studies of CH4 emissions

National Centre for Atmospheric Science
NATURAL ENVIRONMENT RESEARCH COUNCIL

Atmospheric methane is an important greenhouse gas

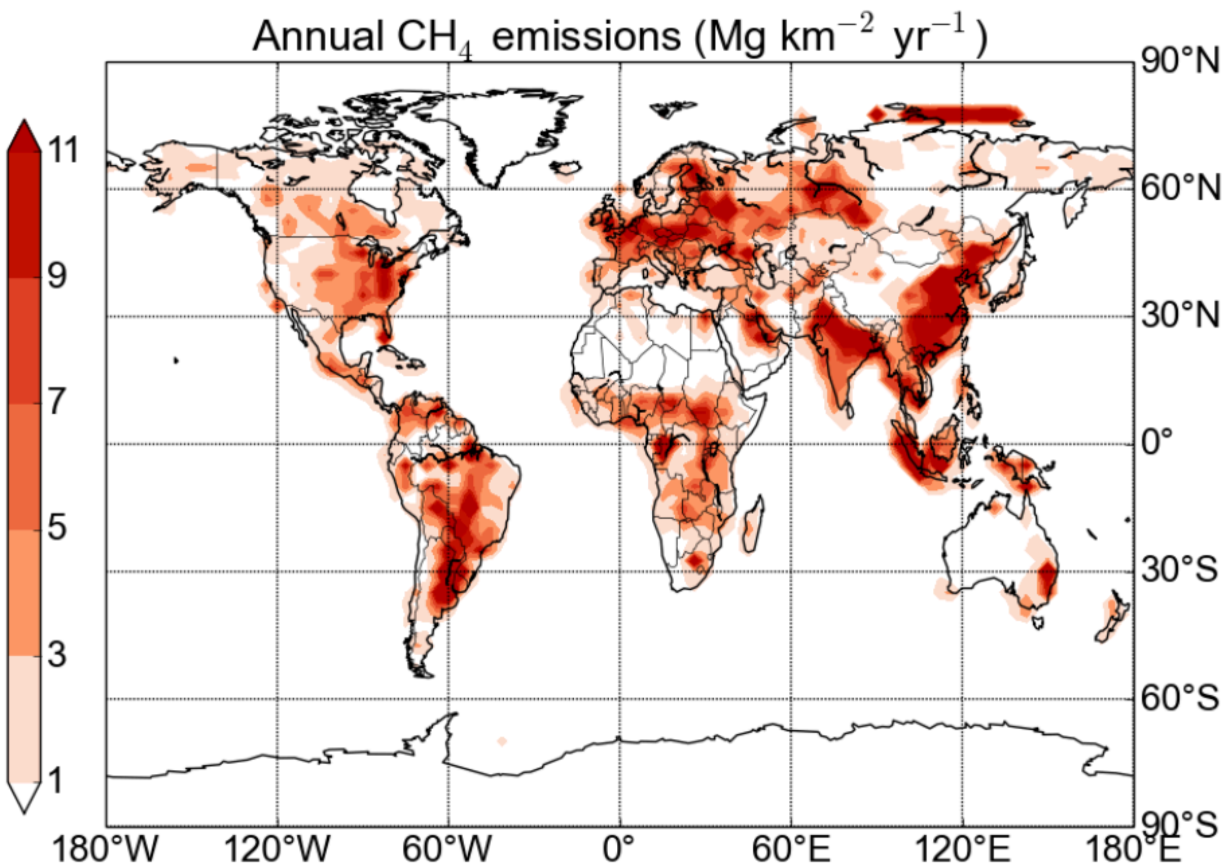
- Methane has a large (second largest) radiative forcing, making it an important anthropogenic greenhouse gas
  - CO_2 : 1.82 Wm^{-2} for an increase from 278 ppm (Pre-Industrial) to 391 ppm (Present-Day)
  - CH_4 : 0.48 Wm^{-2} [AR5] for an increase of 722 ppb to 1803 ppb (PI-PD)
  - O_3 : 0.4 (± 0.2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

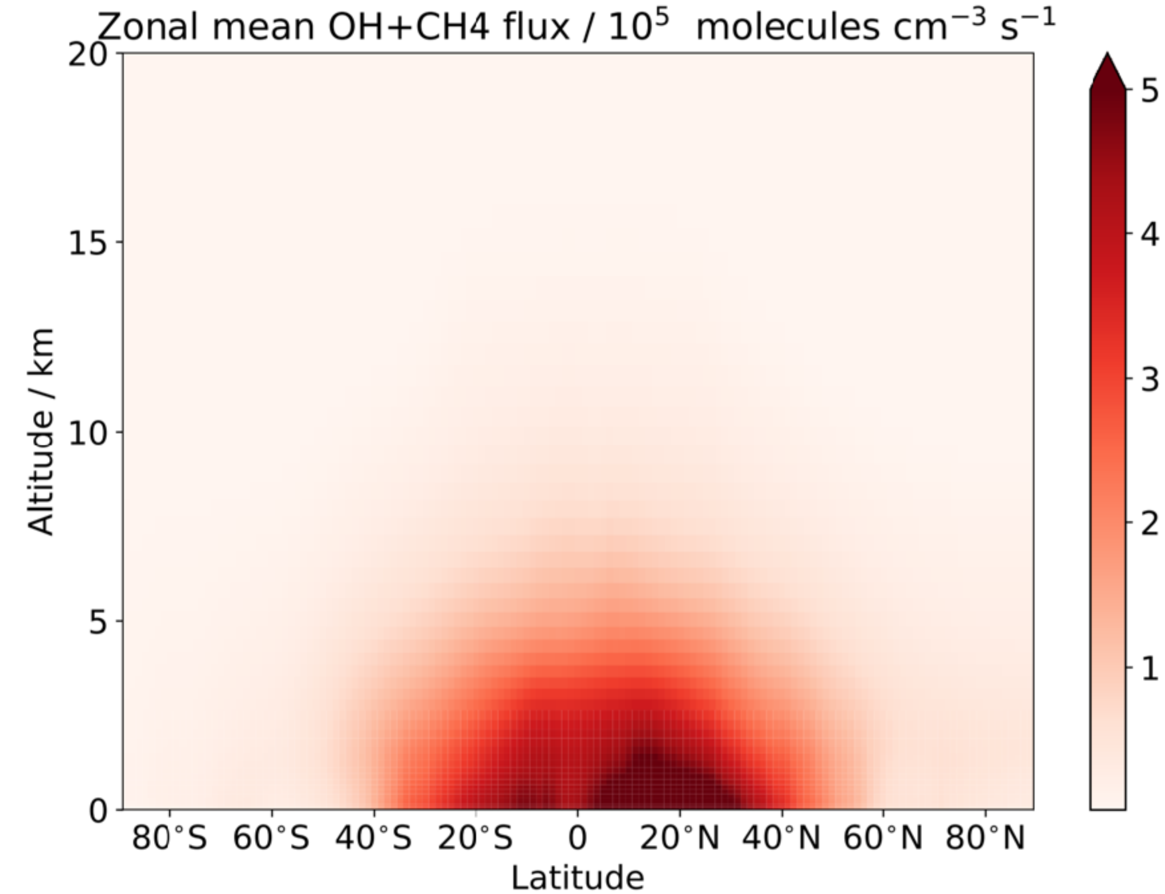
- A large Global Warming Potential – 28 on a 100-year horizon (per-molecule w.r.t. CO_2)
- Strong sources – 585 Tg CH_4 per year, with strong chemical sinks. Lifetime of 10 years
- Methane oxidation leads to ozone and water vapour – both greenhouse gases – with methane an important source of stratospheric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

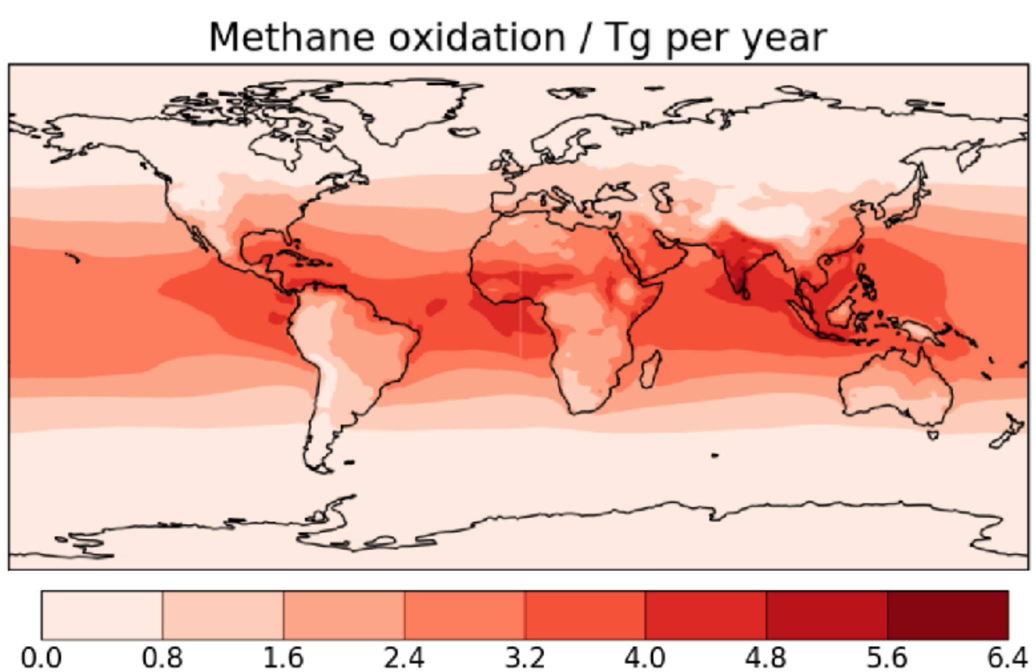
Sources	Wetlands	Fossile fuels gas and coal	Termites	Ruminants	Rice	Waste landfill	Biomass burning
Tg CH_4 per year	177-284	85-105	2-22	87-94	33-40	67-90	32-39

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



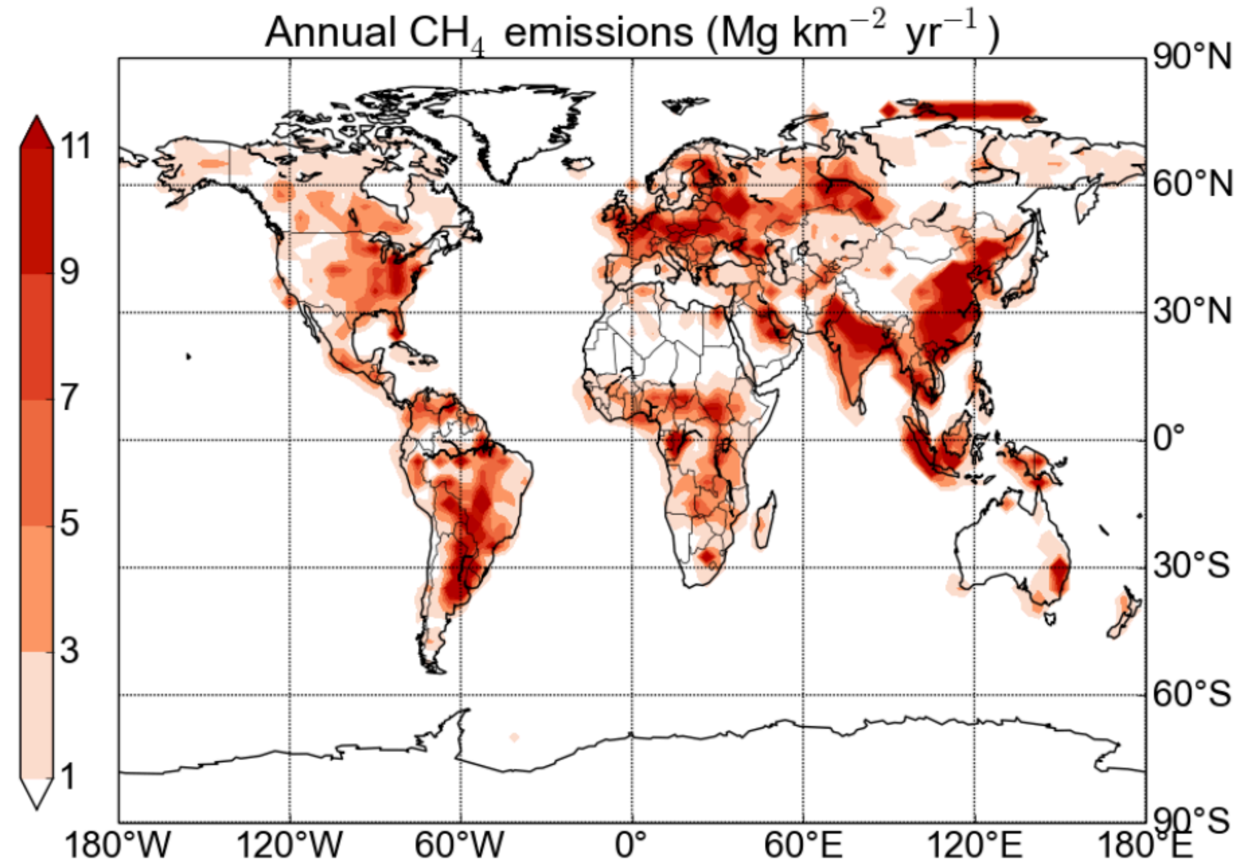


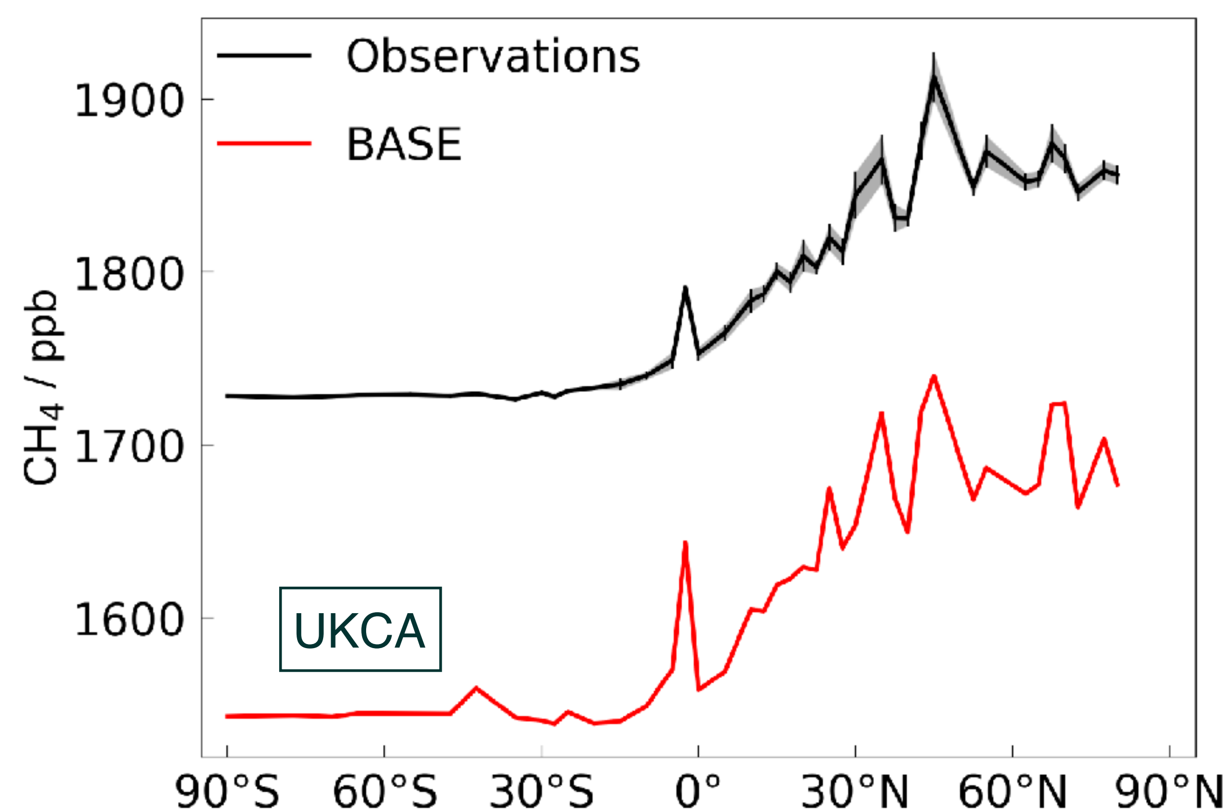


Methane sources are largest in the extra tropics, but oxidation rate is strongly temperature dependent, so peaks where T, humidity and OH high.

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?





JAMES | Journal of Advances in Modeling Earth Systems

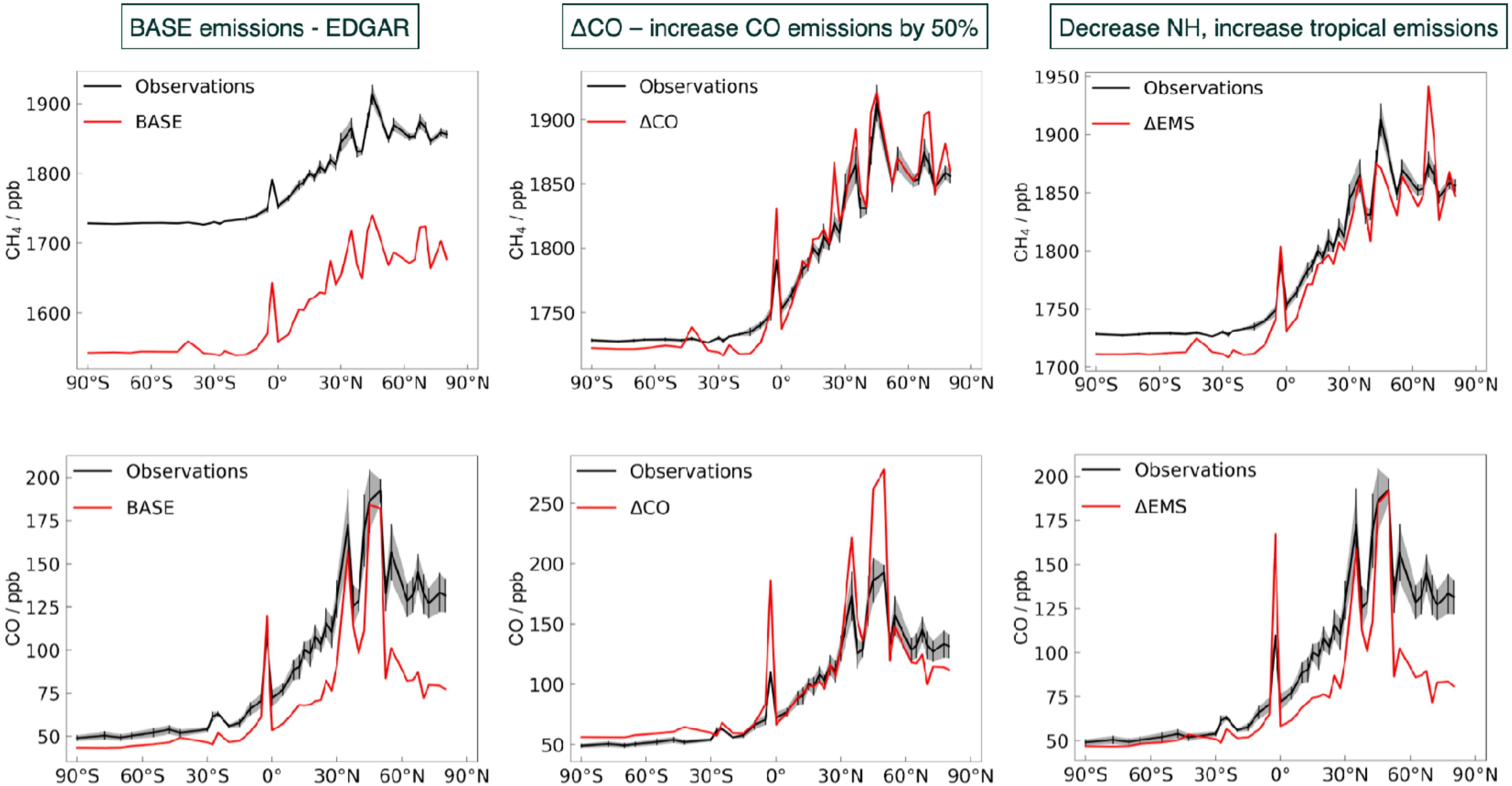
Research Article | Open Access

Methane Emissions in a Chemistry-Climate Model: Feedbacks and Climate Response

I. Heimann, P. T. Griffiths, N. J. Warwick, N. L. Abraham, A. T. Archibald, J. A. Pyle

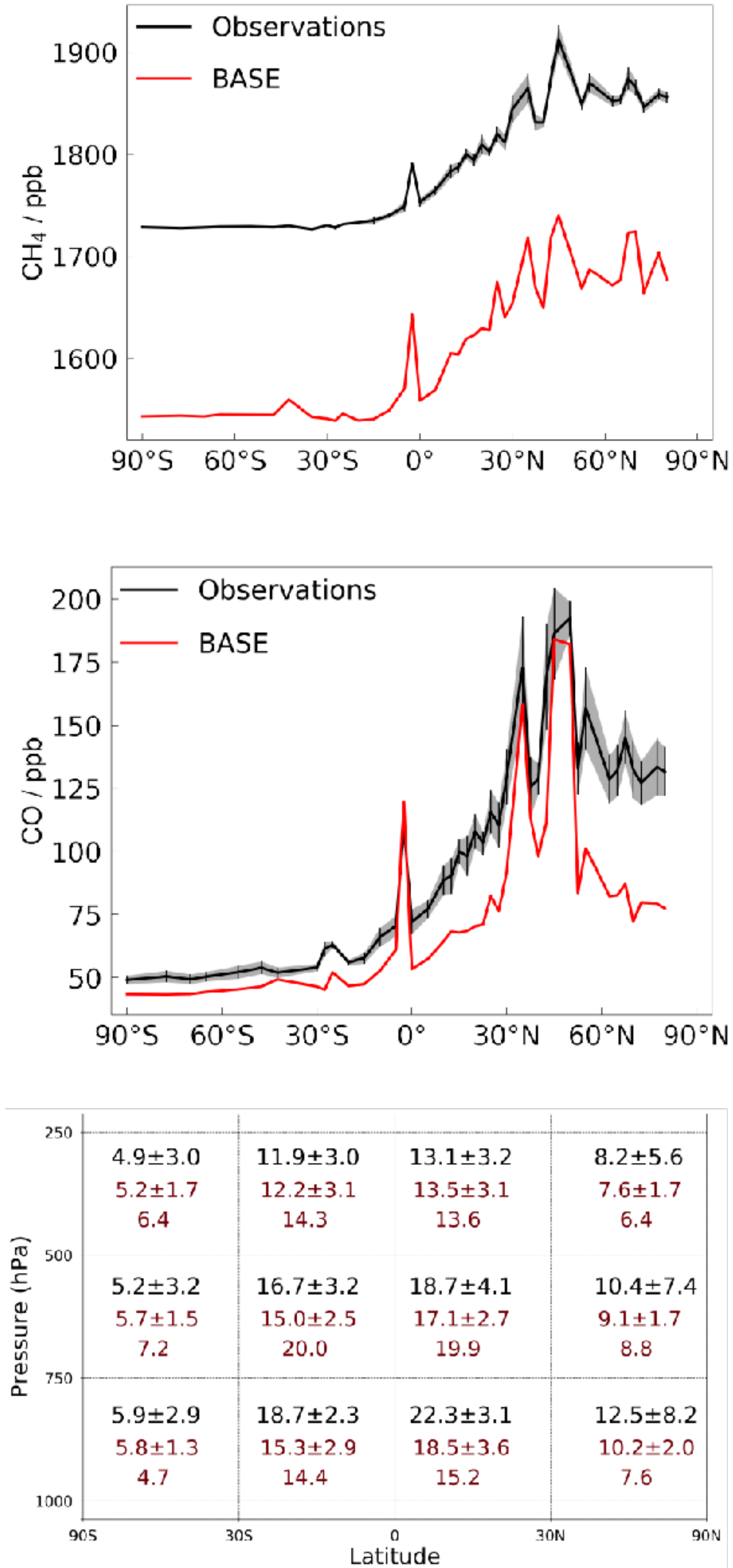
3 sensitivity experiments

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.

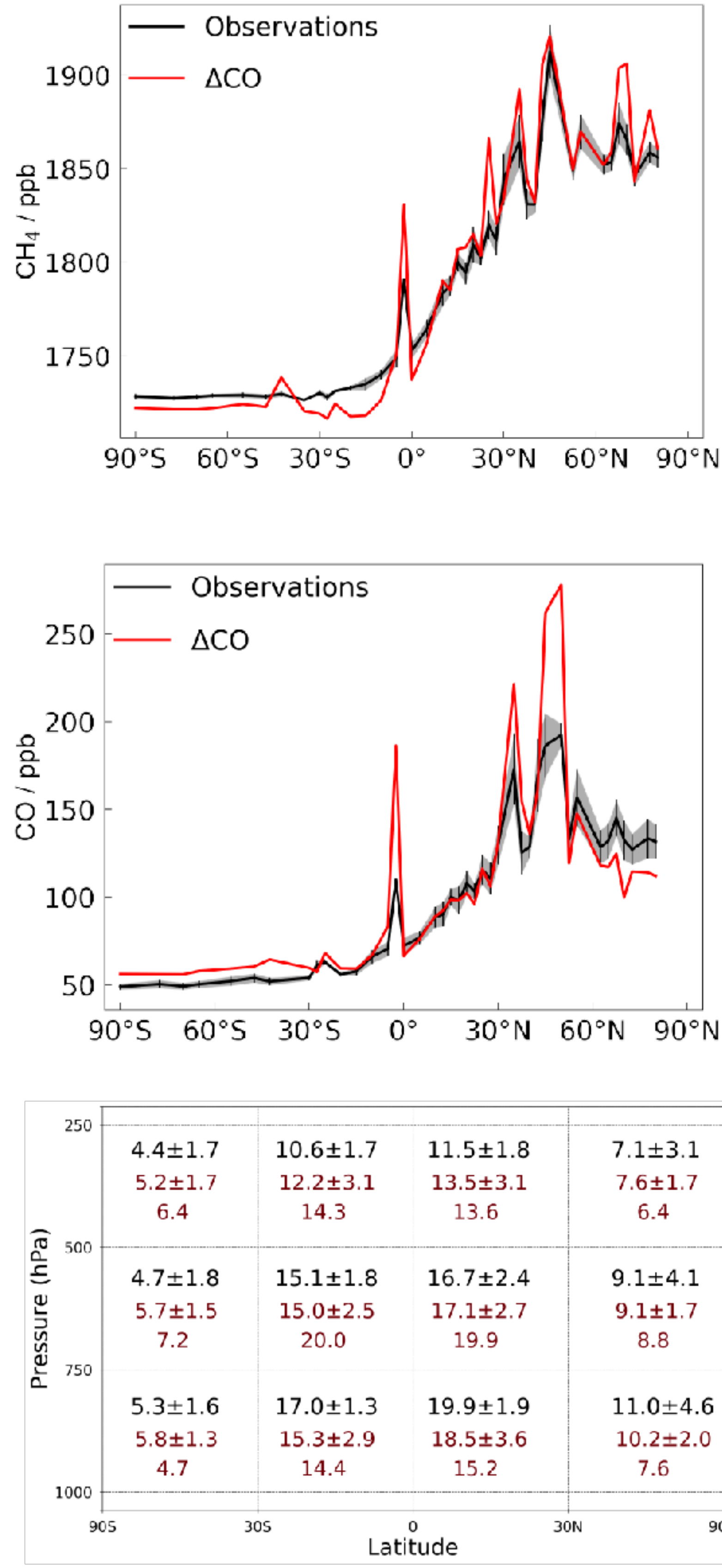


Sensitivity of UKCA to emissions – 3 global experiments

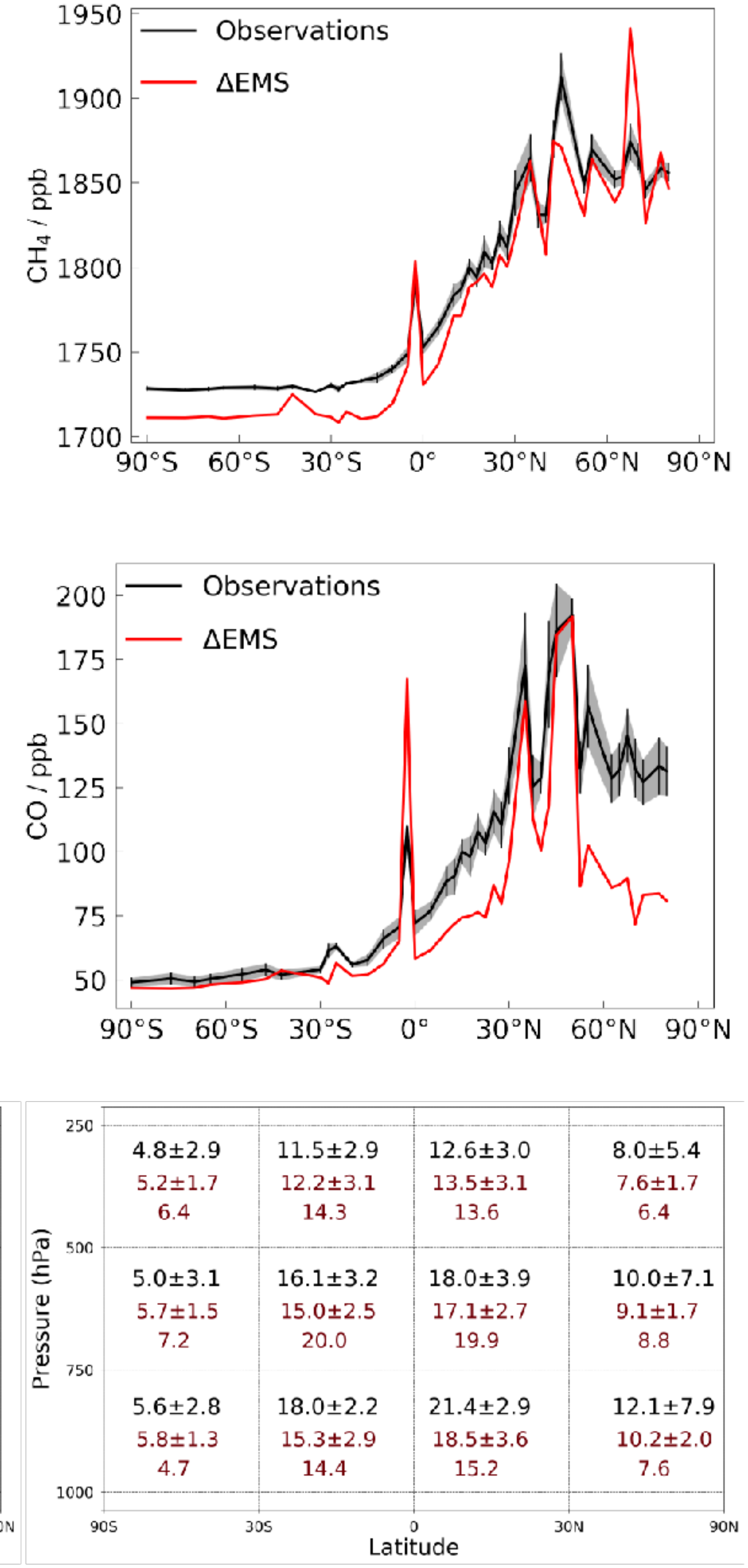
BASE emissions - EDGAR



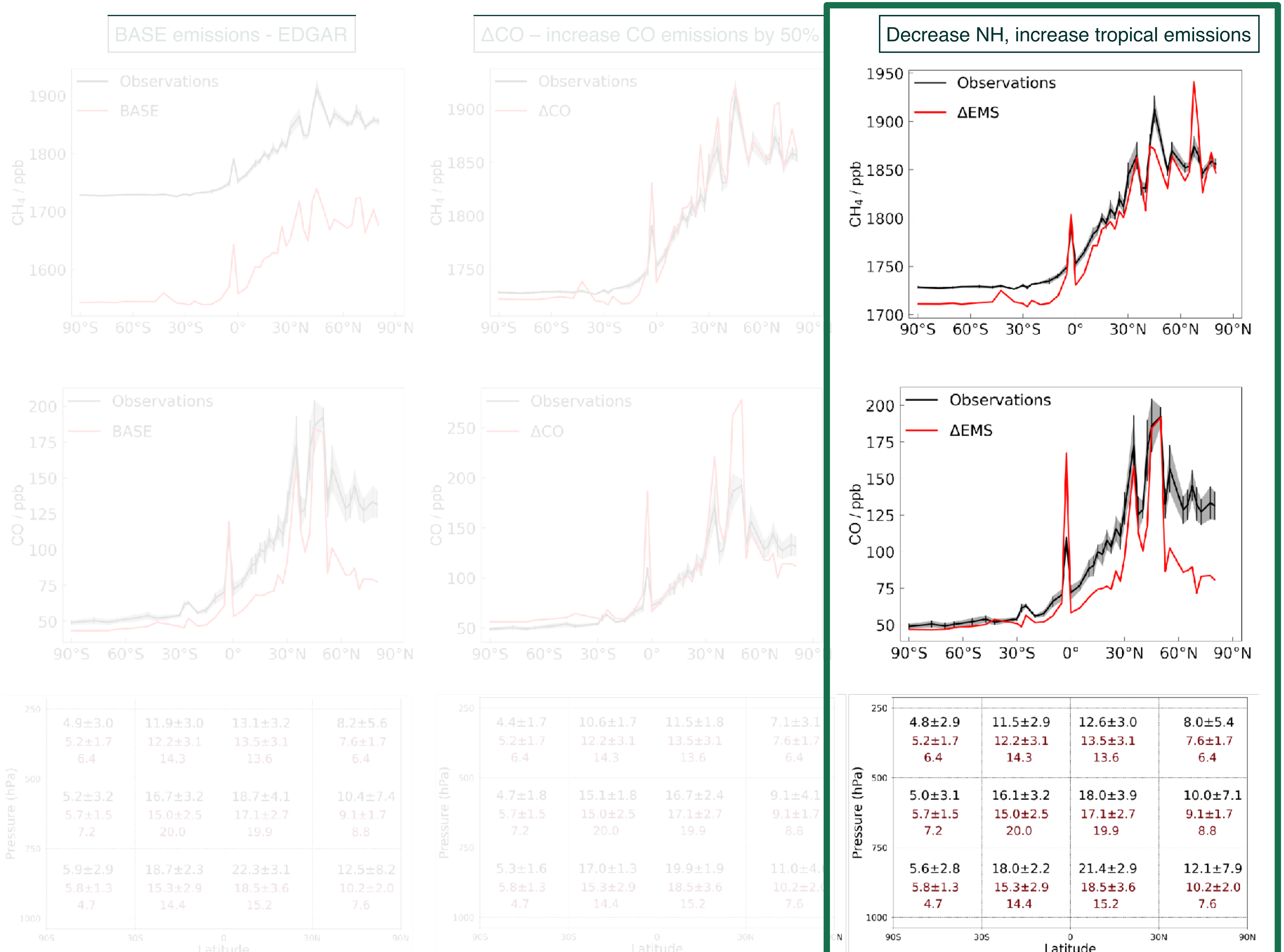
ΔCO – increase CO emissions by 50%



Decrease NH, increase tropical emissions



Sensitivity of UKCA to emissions – 3 global experiments

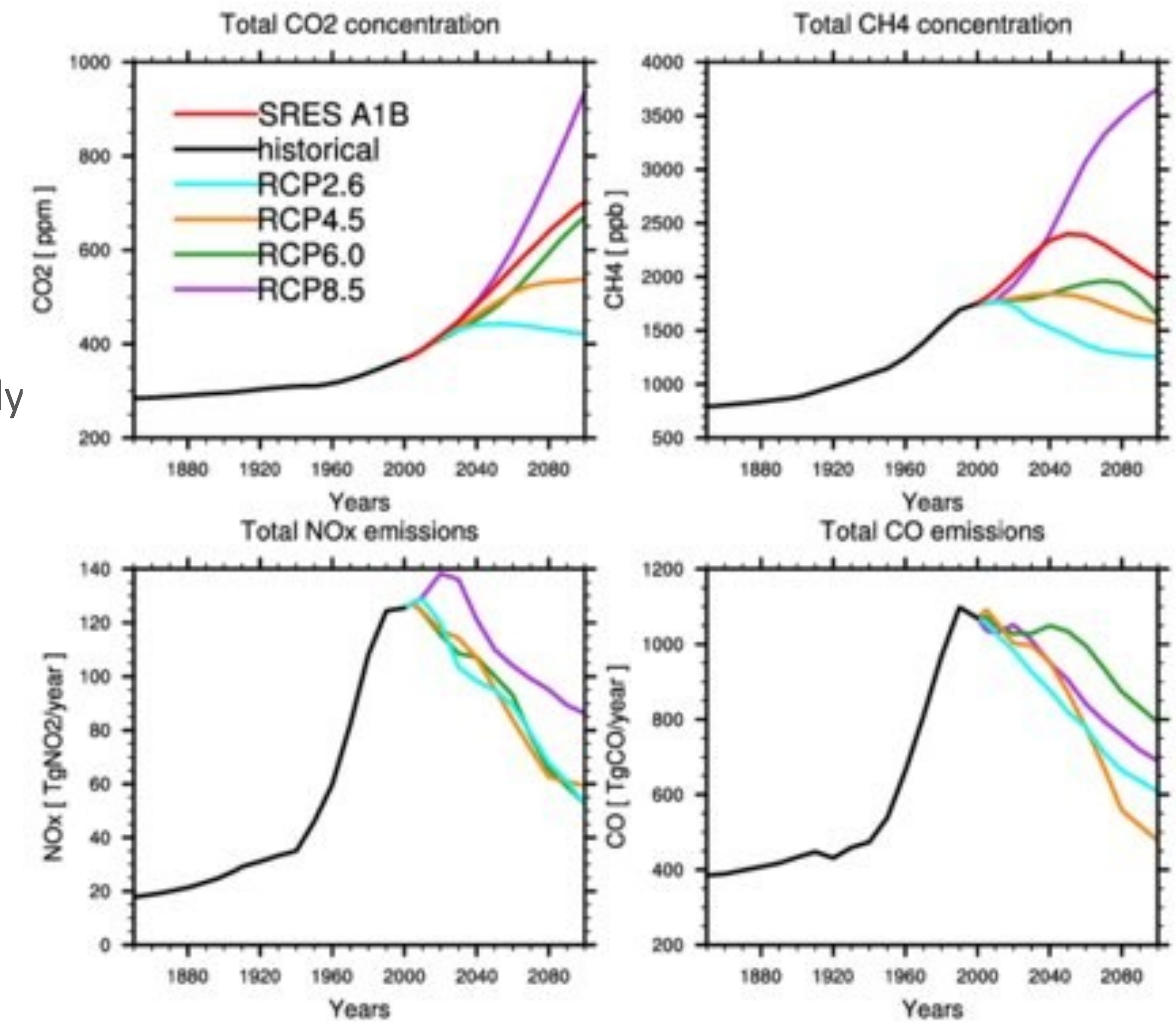


Drivers of methane levels in 2100

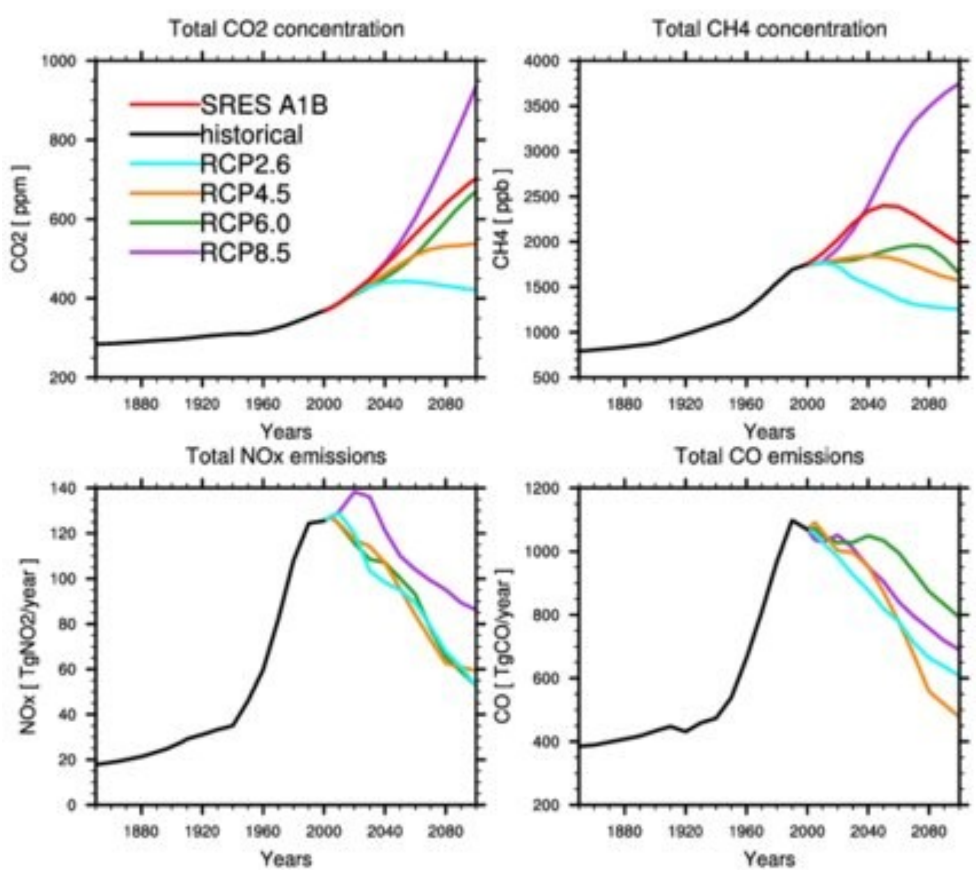
National Centre for Atmospheric Science
NATURAL ENVIRONMENT RESEARCH COUNCIL

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 methane emissions to RCP8.5
  - ΔCC+ALL – increase O3Pre to RCP8.5
- Bring all forcings together at the end

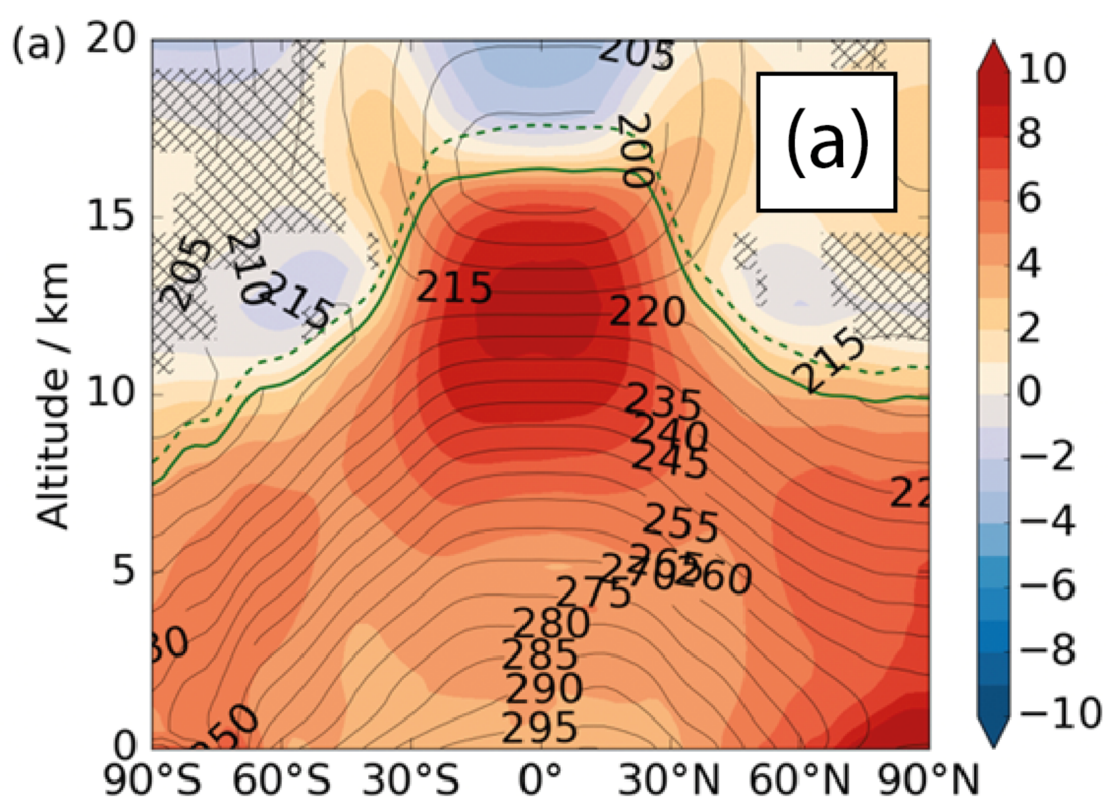


What happens to tropospheric oxidising capacity in future climate?

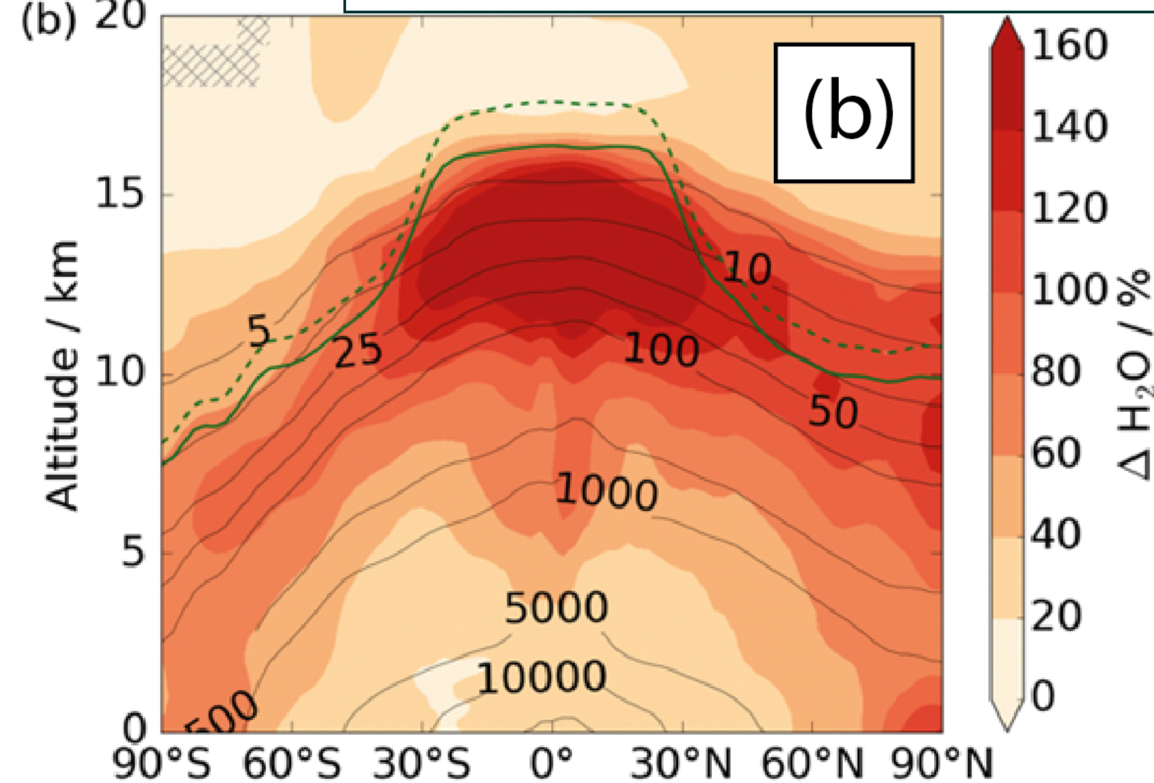


- In RCP8.5 there's a big increase in temperature throughout the troposphere by 2100.
- The warmer atmosphere can support more water vapour, so humidity increases.
- Tropospheric expansion means the upper troposphere experiences the biggest changes.

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



Temperature

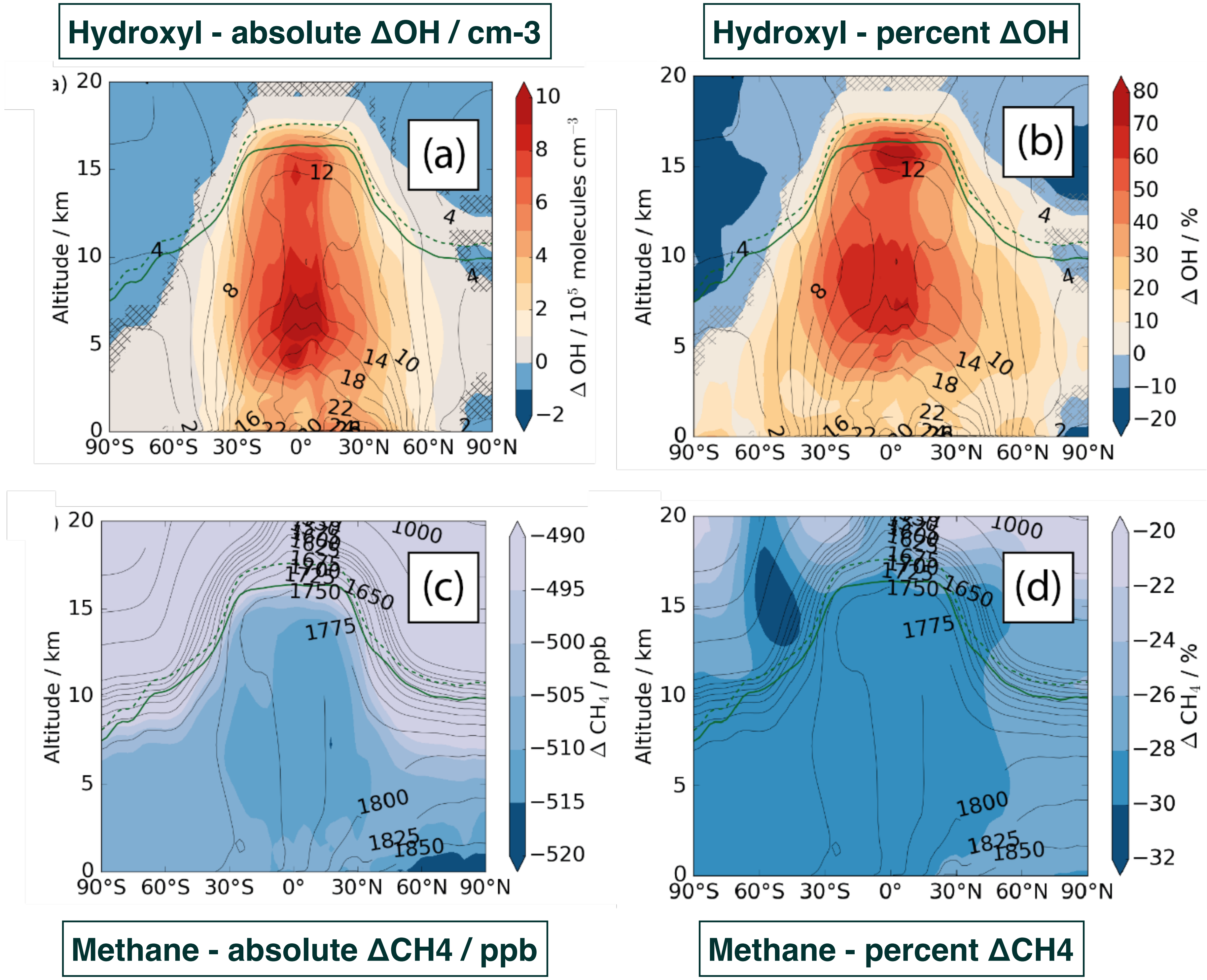


Water vapour

What happens to tropospheric oxidising capacity in future climate?

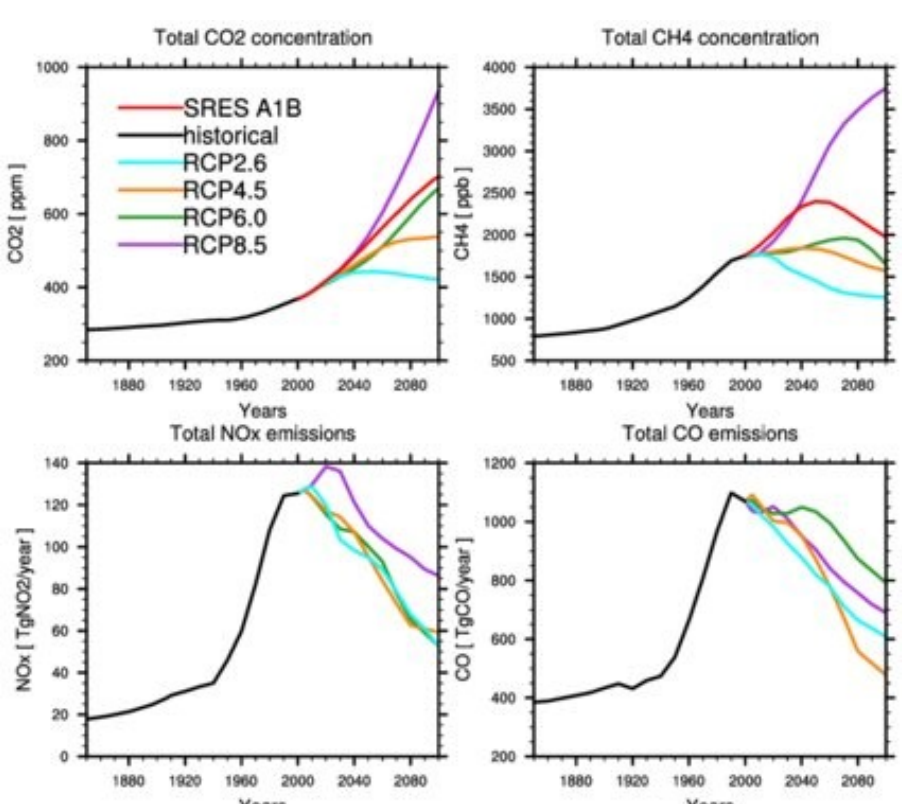
- OH – warmer, wetter atmosphere so OH increases
- Changes largest in tropical FT
- More OH means less CH4 (and k(OH+CH4) increases as T increases)
- Methane decrease large everywhere cf Year 2000.
- Methane lifetime reduced from 9 to 6 years.

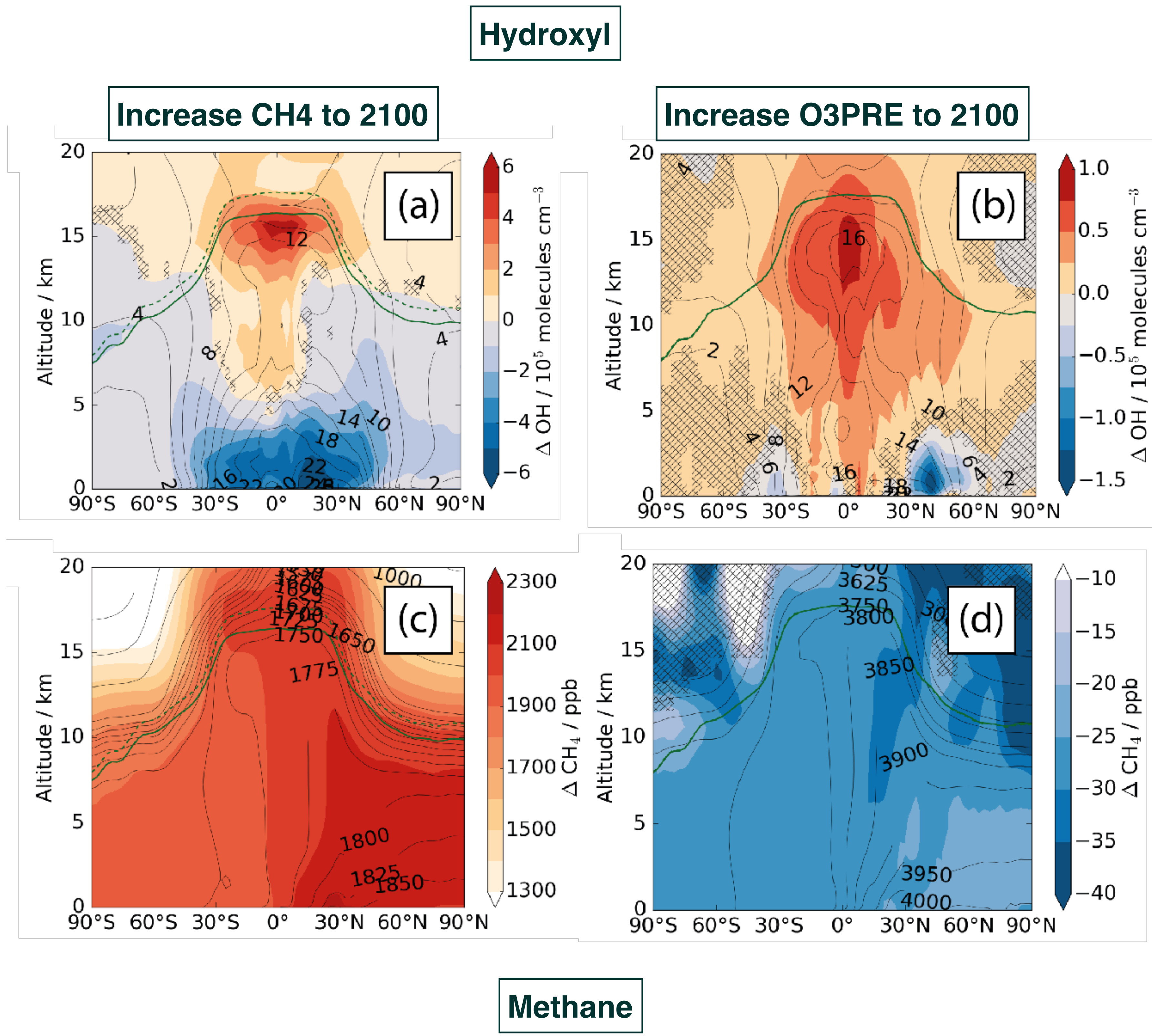
ΔCC with respect to year 2000



What happens to tropospheric oxidising capacity in future climate?

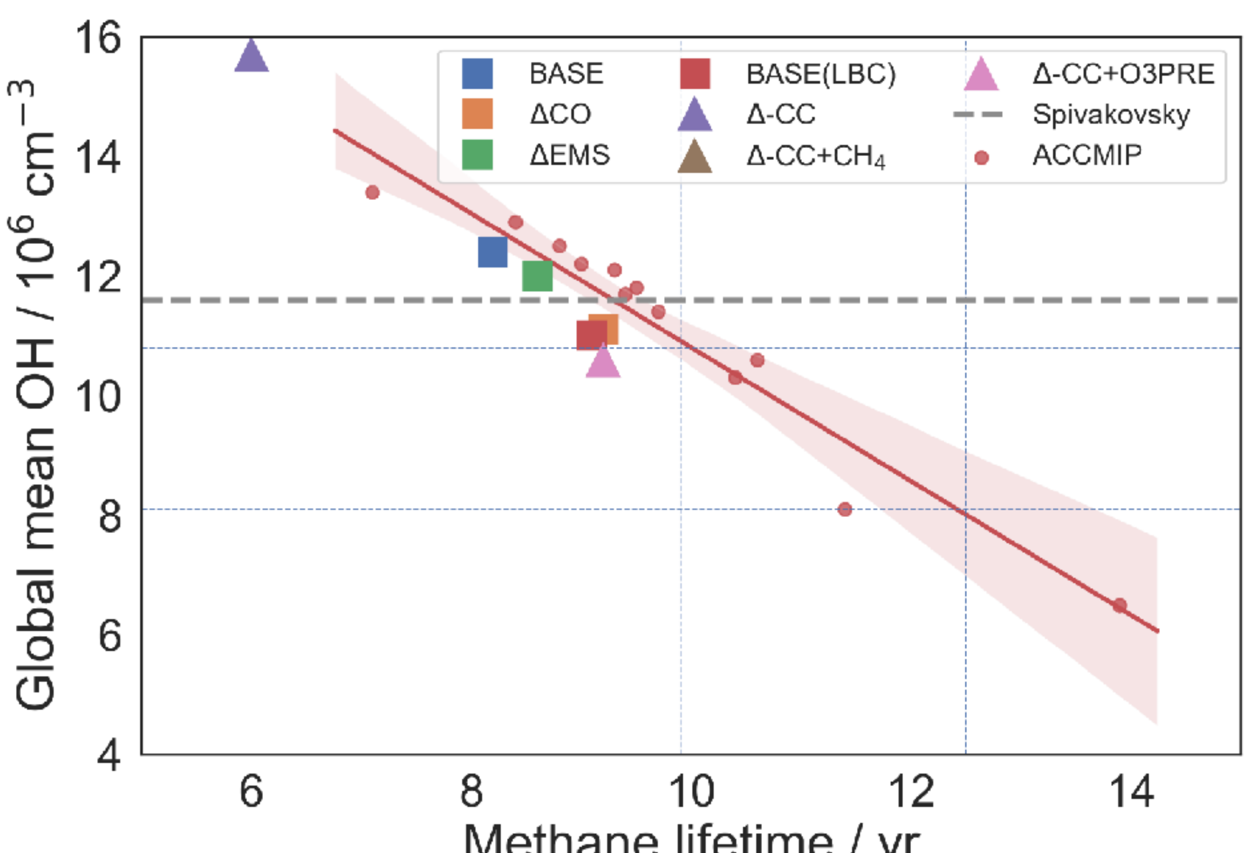
- Increasing CH_4 emissions to RCP8.5 levels gives
  - Large increase in CH_4
  - Large decrease in OH
- Increasing CO and NOx to RCP8.5 levels gives
  - Smaller change in OH
  - Small decreases in CH_4

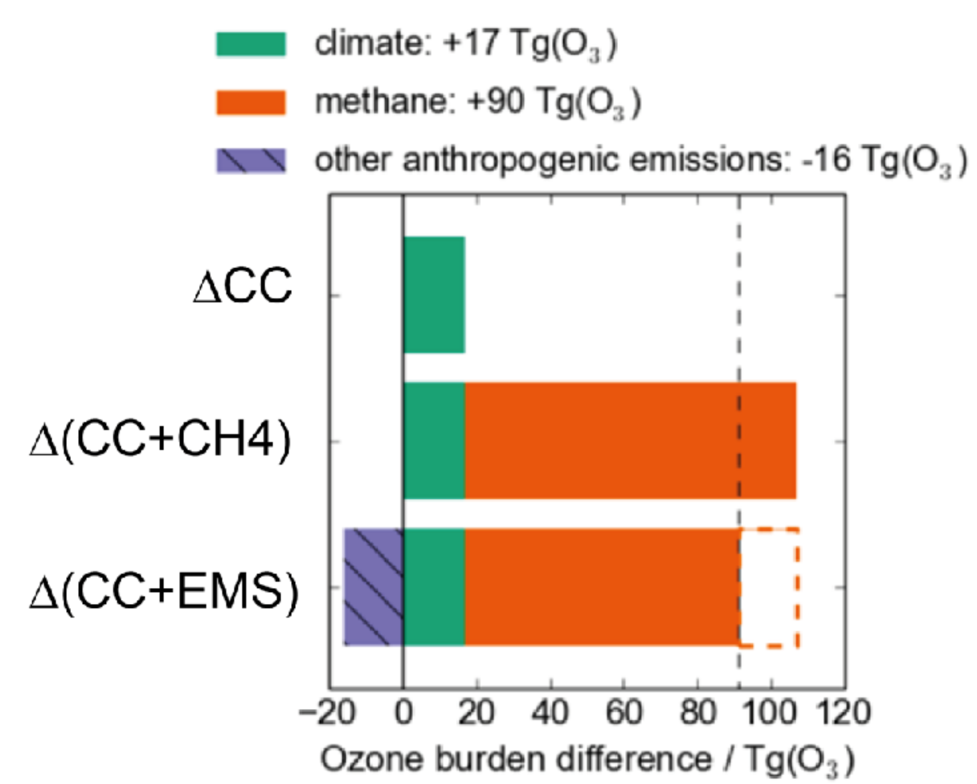




Methane in the UKCA chemistry-climate model - conclusions

- 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





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

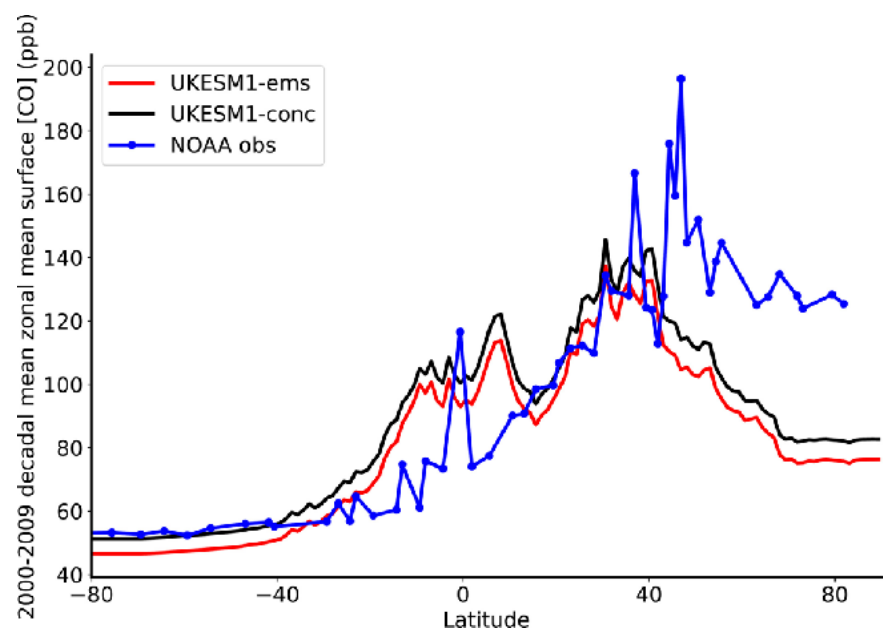
National Centre for Atmospheric Science
NATURAL ENVIRONMENT RESEARCH COUNCIL

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*

Research Article | Open Access

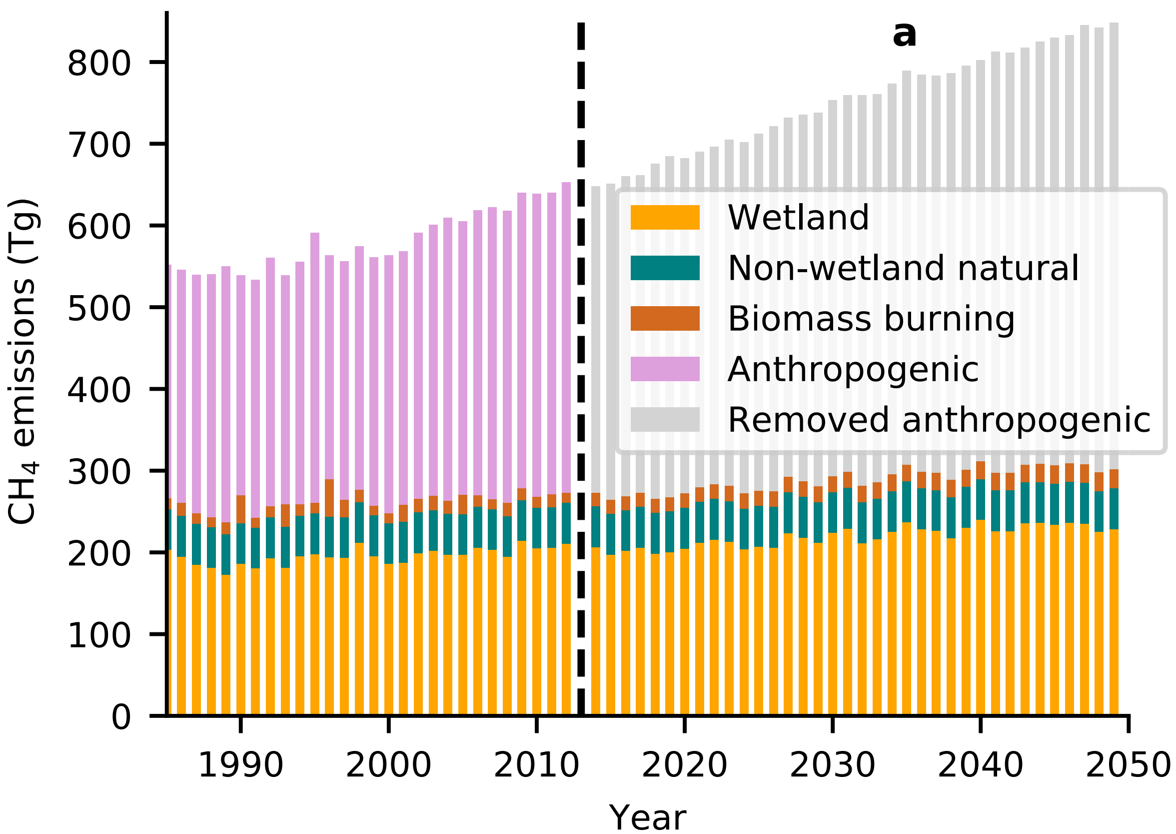
Description and Evaluation of an Emission-Driven and Fully Coupled Methane Cycle in UKESM1

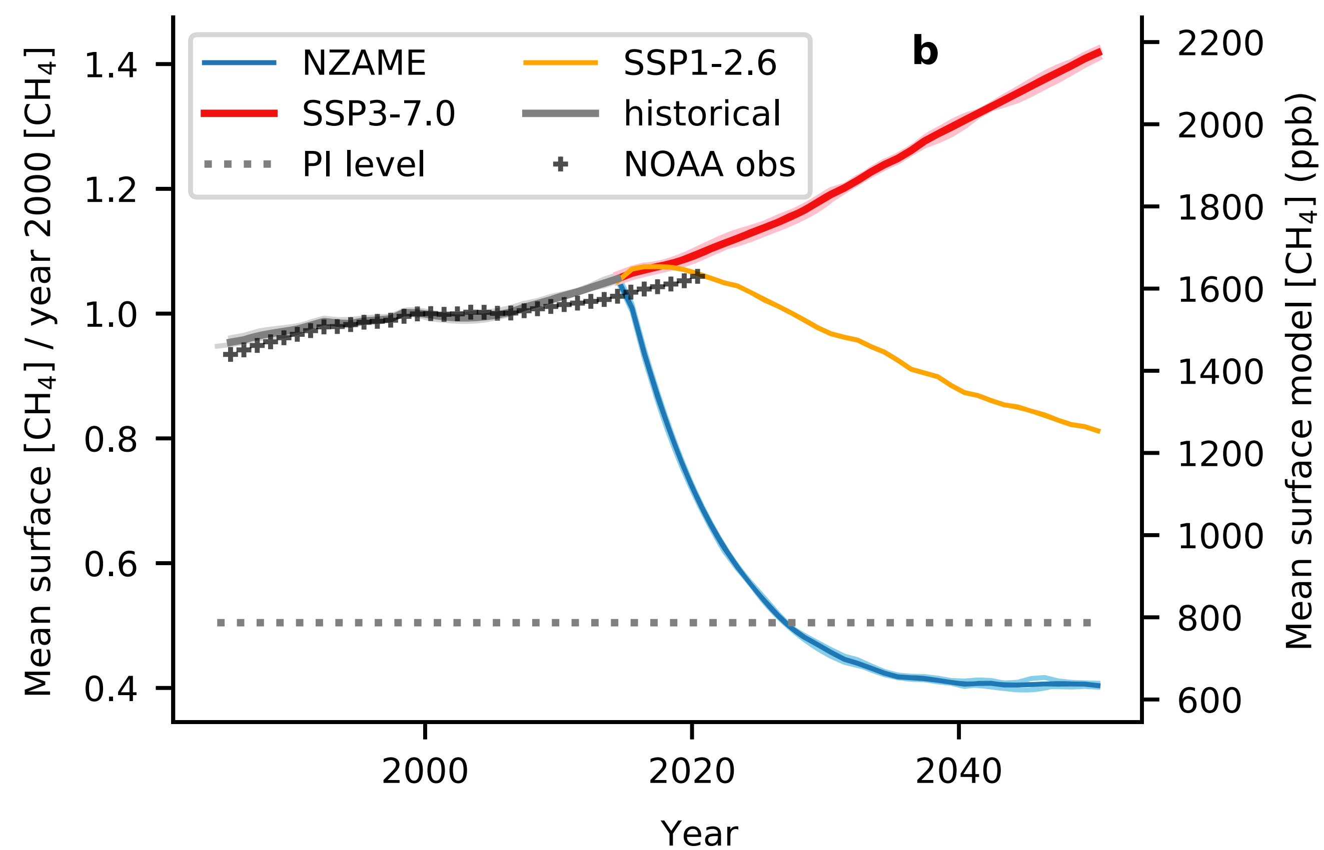
G. A. Folberth, Z. Staniaszek, A. T. Archibald, N. Gedney, P. T. Griffiths, C. D. Jones, F. M. O'Connor, R. J. Parker, A. A. Sellar, A. Wiltshire

First published: 07 July 2022 | https://doi.org/10.1029/2021MS002982

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



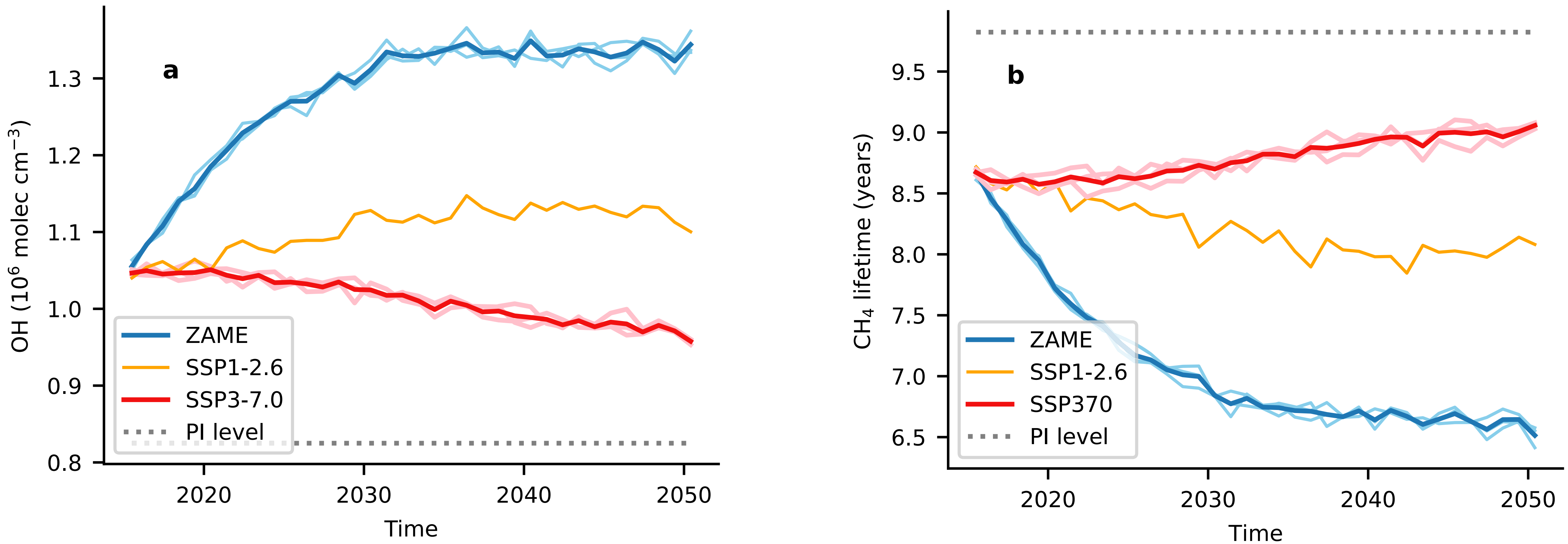


- Comparison with SSP3-7.0 and SSP1-2.6 allows them to function as a counterfactual
  - What are the risks of methane emissions?
  - What are the benefits of constraining future methane emissions?

Figures by Zosia Staniaszek

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

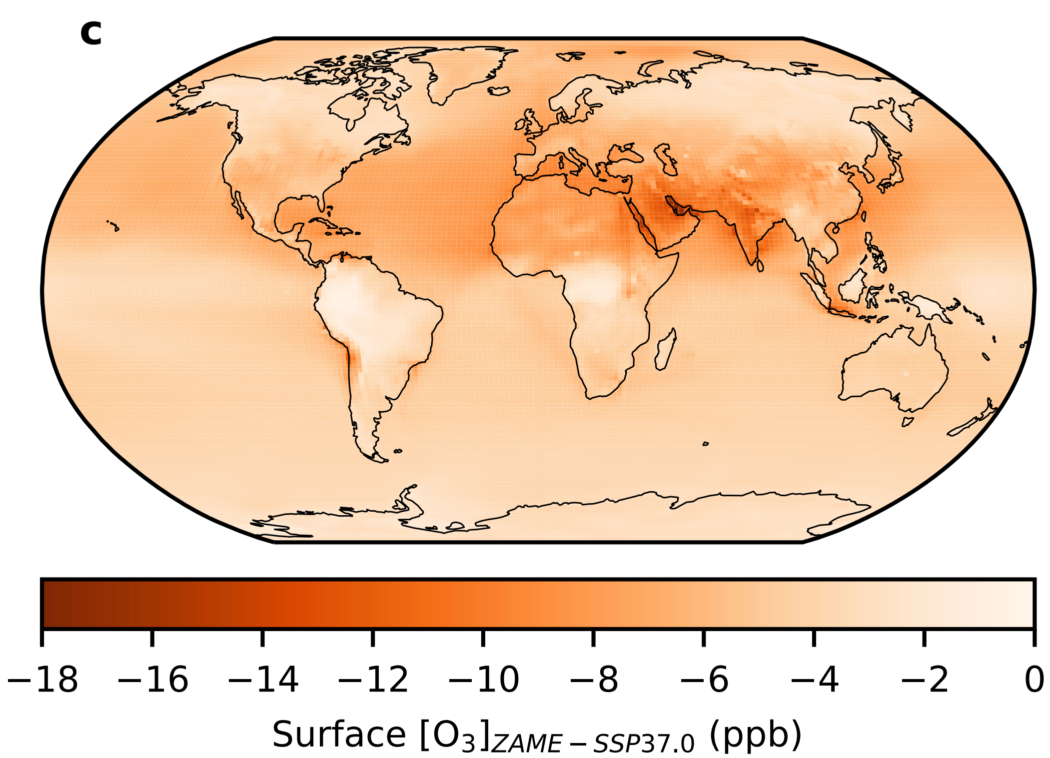


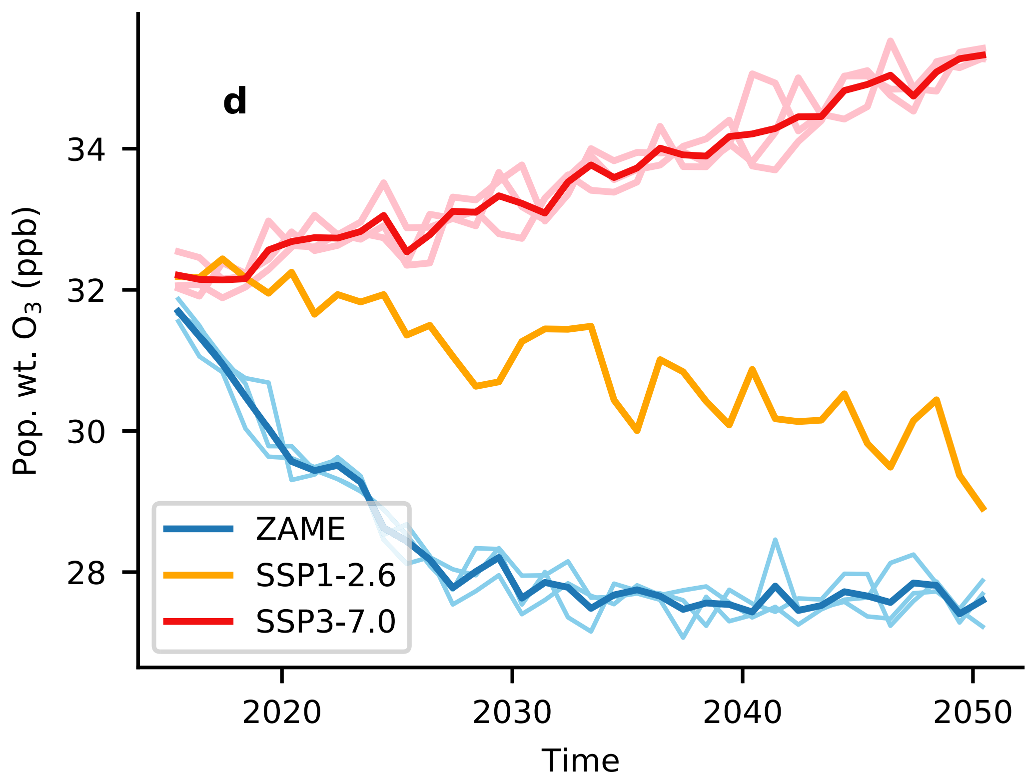
- Comparison with SSP3-7.0 and SSP1-2.6 allows them to function as a counterfactual
  - What are the risks of methane emissions?
  - What are the benefits of constraining future methane emissions?

Figures by Zosia Staniaszek

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



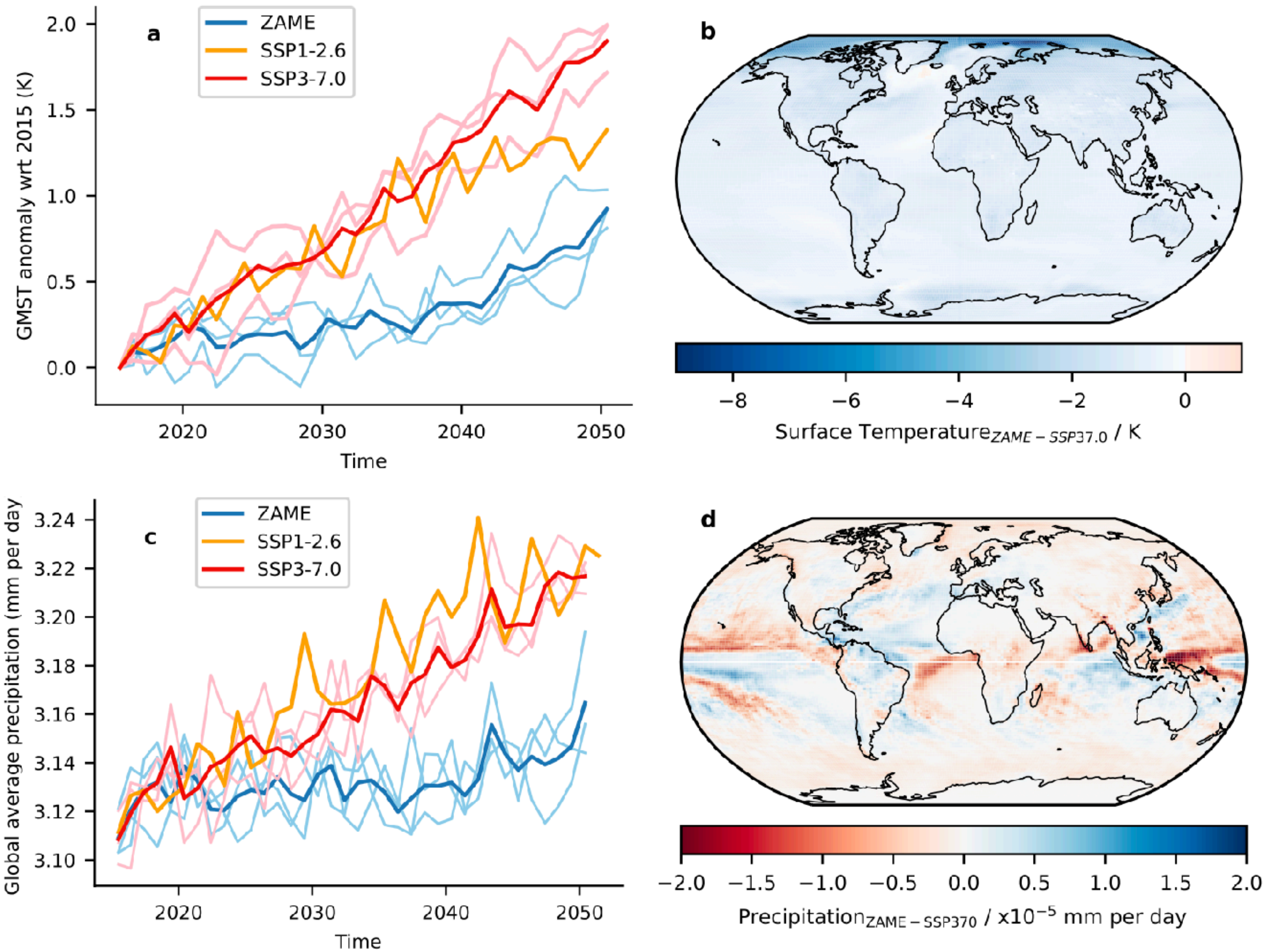


- Weighting the ozone field by human exposure shows ~10% decline
- Projected decrease in AQ-related mortality of the order of 500k per year

Figures by Zosia Staniaszek

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 → DT = 0.5 K
- Decline across the globe, strong regional variations, Arctic amplification



Figures by Zosia Staniaszek

Conclusions 2/3 - CH_4 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 O_3 RF.
  - Leads to more OH - shorter methane lifetime, reduced GWP.

Replacing CH_4 with H_2 as a fuel source

National Centre for Atmospheric Science
NATURAL ENVIRONMENT RESEARCH COUNCIL

Atmospheric chemistry of H_2

- Present-day sources

Sources	Fossil fuel	Biomass burning	N2 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 H_2, a mean mixing ratio of 550 ppb and a lifetime of 2.5 years
- H_2 affects
  - ozone levels (H_2 oxidation functions as a source of ozone)
  - methane levels (H_2 removes OH, decreasing the size of the CH_4 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)

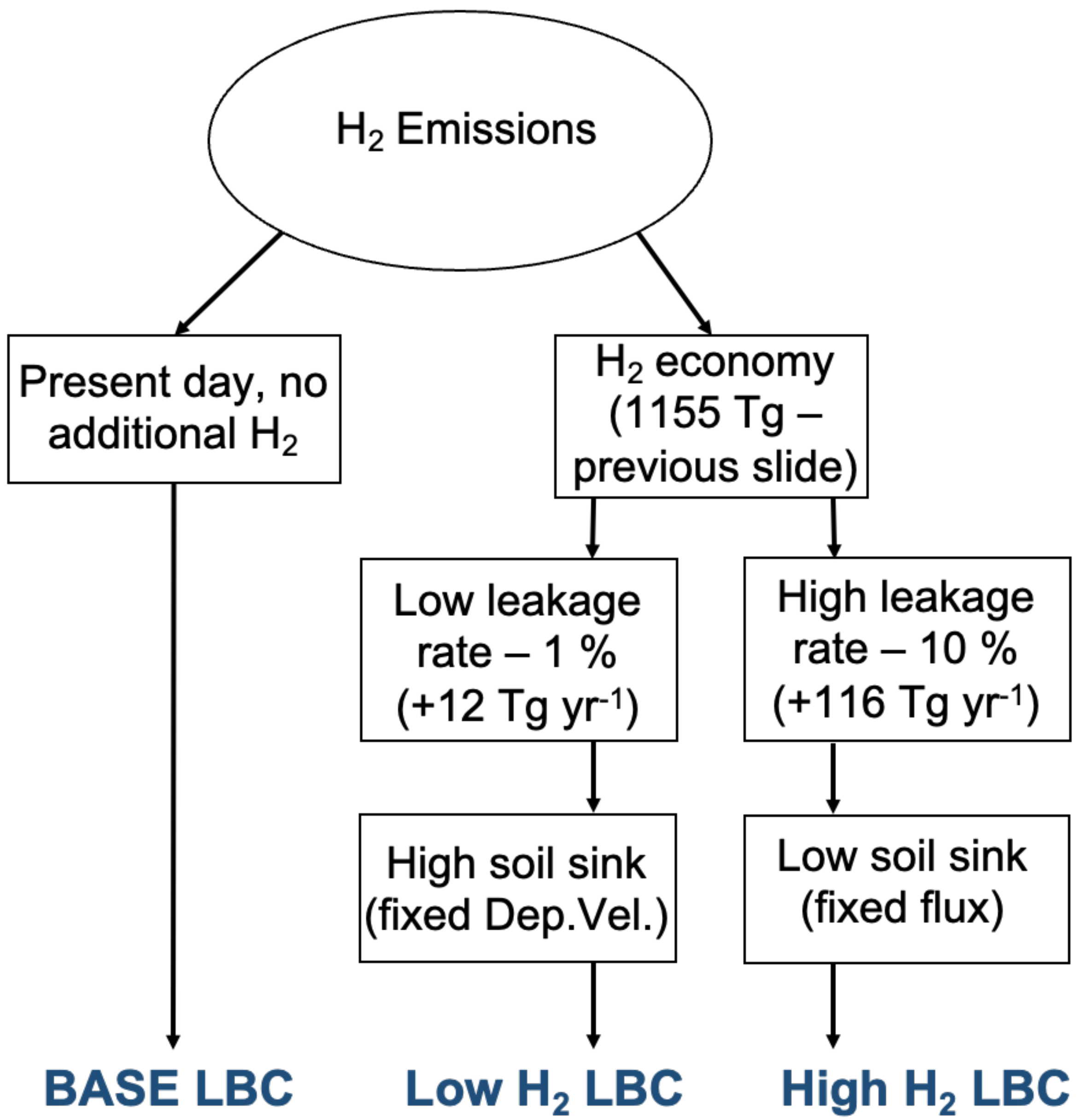
- Global capacity for power generation from H2 was ~0.01 % of total capacity in 2015
- Assuming continuing growth trend could be 0.5 % by 2030
- Assume 5% of power generation from H2 (~2 % of GED)

Total energy from H_2 = 22% of GED (BP) = 3.9×10^{13} kWh = 1155 Tg H_2

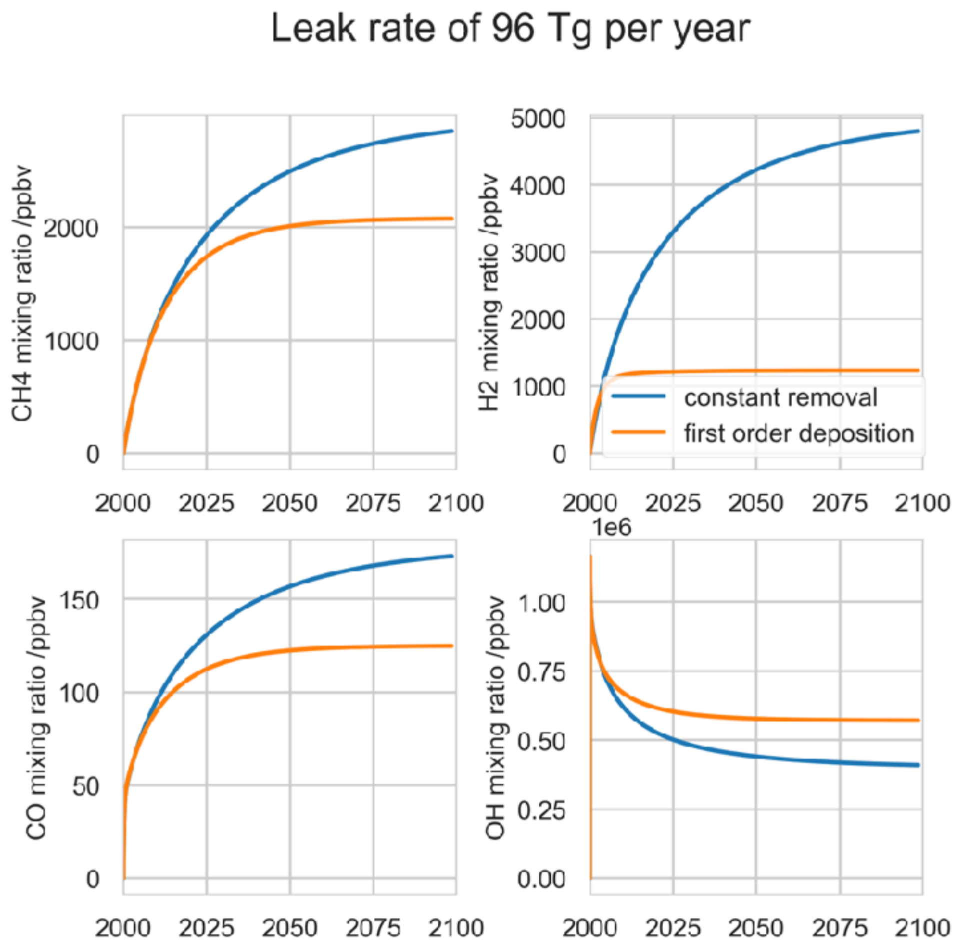
Atmospheric chemistry of H2

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



1. H_2 leakage emissions increase as a result of a move to H_2 as a fuel source.
    - 750 ppb, 1000 ppb and 2000 ppb (approx increase from 76 Tg to >200 Tg H_2 emissions)
2. Adoption of H_2 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 H_2 as a fuel source means CH4 emissions decrease and other other anthropogenic emissions such as CO, NOx, NMVOCs
    - Consider this under low-H_2 and high-H_2 leakage scenarios

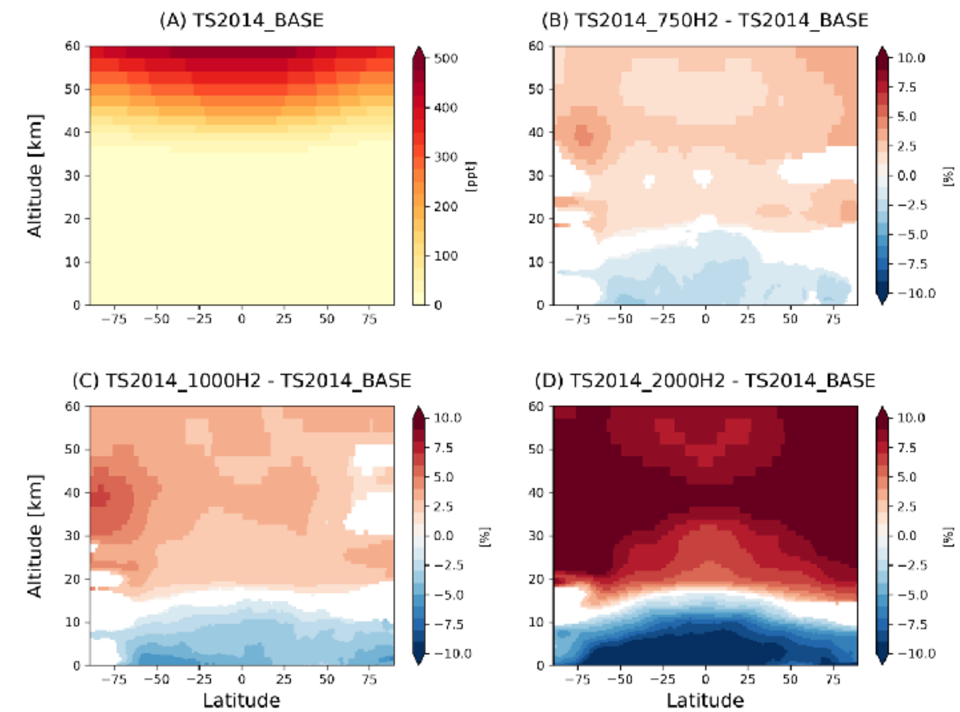
Scenario	H2	CH4	Notes
TS2014_BASE	500	1835	2000-2014 climatology from CMIP6 historical
TS2014_750H_2	750	1835	As TS2014_BASE
TS2014_1000H_2	1000	1835	As TS2014_BASE
TS2014_1500H_2	1500	1835	As TS2014_BASE
TS2014_2000H_2	2000	1835	As TS2014_BASE
TS2014_1500H_2_2058CH_4	1500	2058	As TS2014_BASE
TS2014_2000H_2_2171CH_4	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

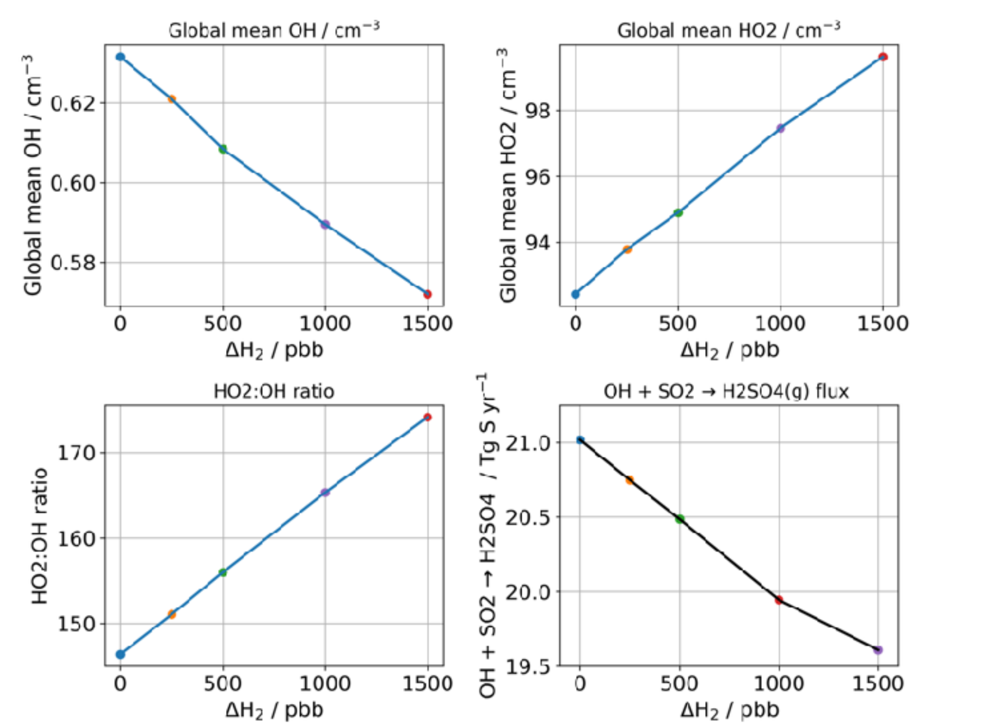
- UKCA in concentration-driven, atmosphere-only mode, 2014 timeslice: faster spin-up and use of fixed SSTs means can calculate ERF.
- Lots of scenarios to cover the range of potential H2 scenarios.
- Use a box model to estimate H2 levels resulting from various parametric uncertainties such as deposition.

Chemical effects of enhanced H2 levels

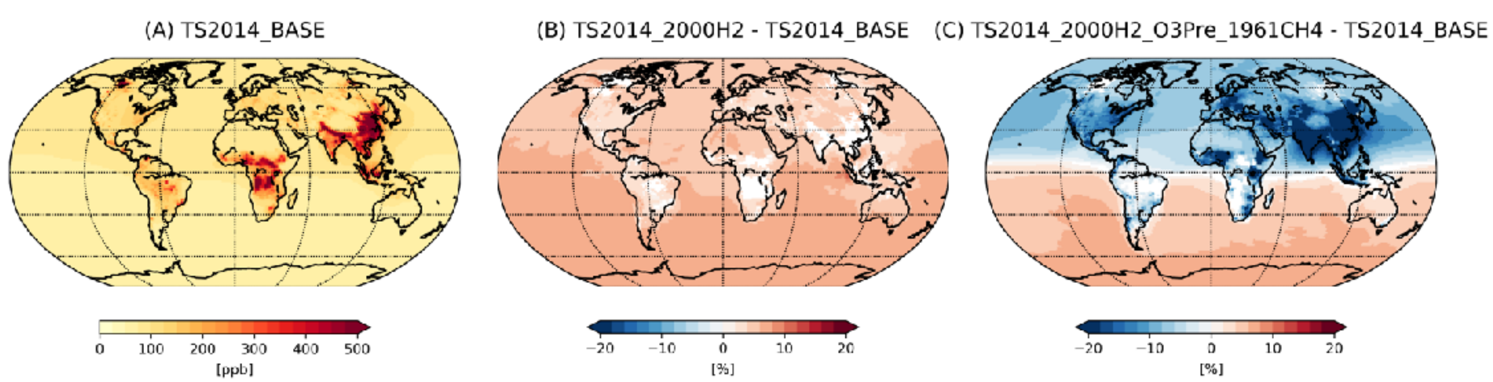
- Increased H_2 levels suppress OH via increase in $OH + H_2 \rightarrow H_2O + H$
  - Suppressed OH → enhanced CO
- Increased HO_2 levels enhance HO_2 via $H+O_2+M \rightarrow HO_2$
- Change in $OH:HO_2$ ratio, and changes to both O_3 Prod (HO_2+NO) and O_3 Loss (e.g. HO_2+O_3)
- Tropospheric ozone column mostly ozone increases, H2 functioning as O_3 precursors

Zonal Mean OH

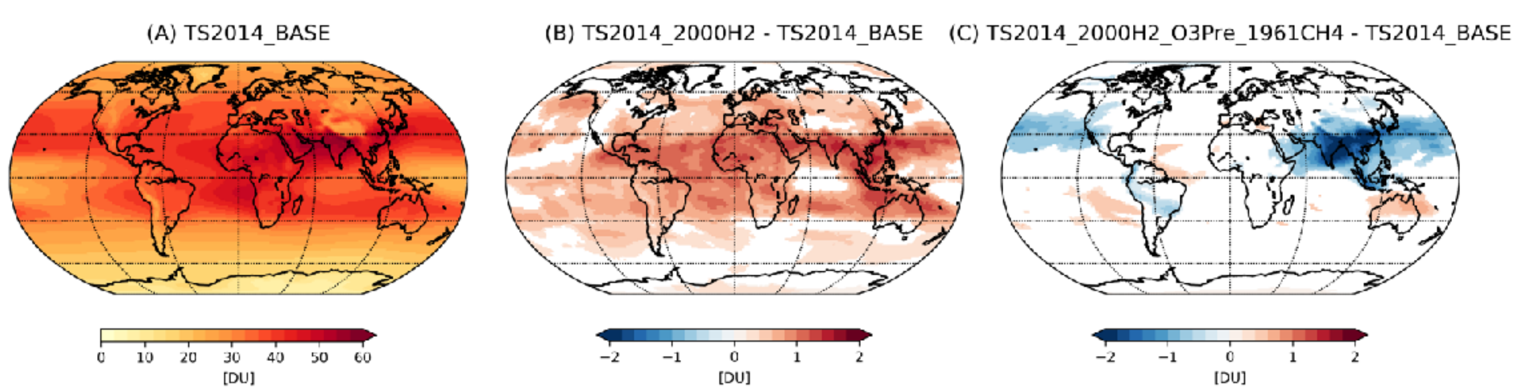




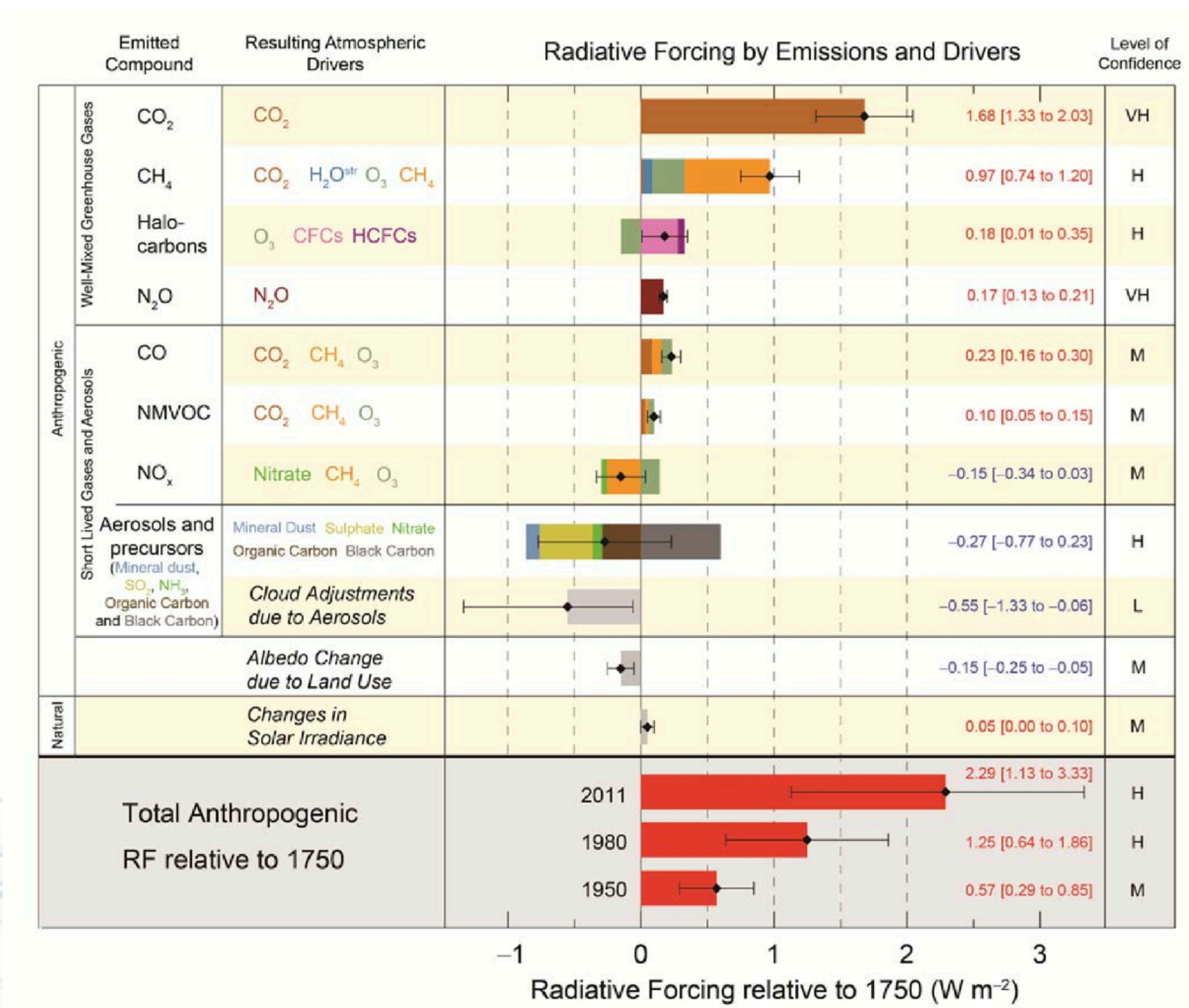
Surface CO



Tropospheric Column Ozone



Effective radiative forcing - CMIP5 picture

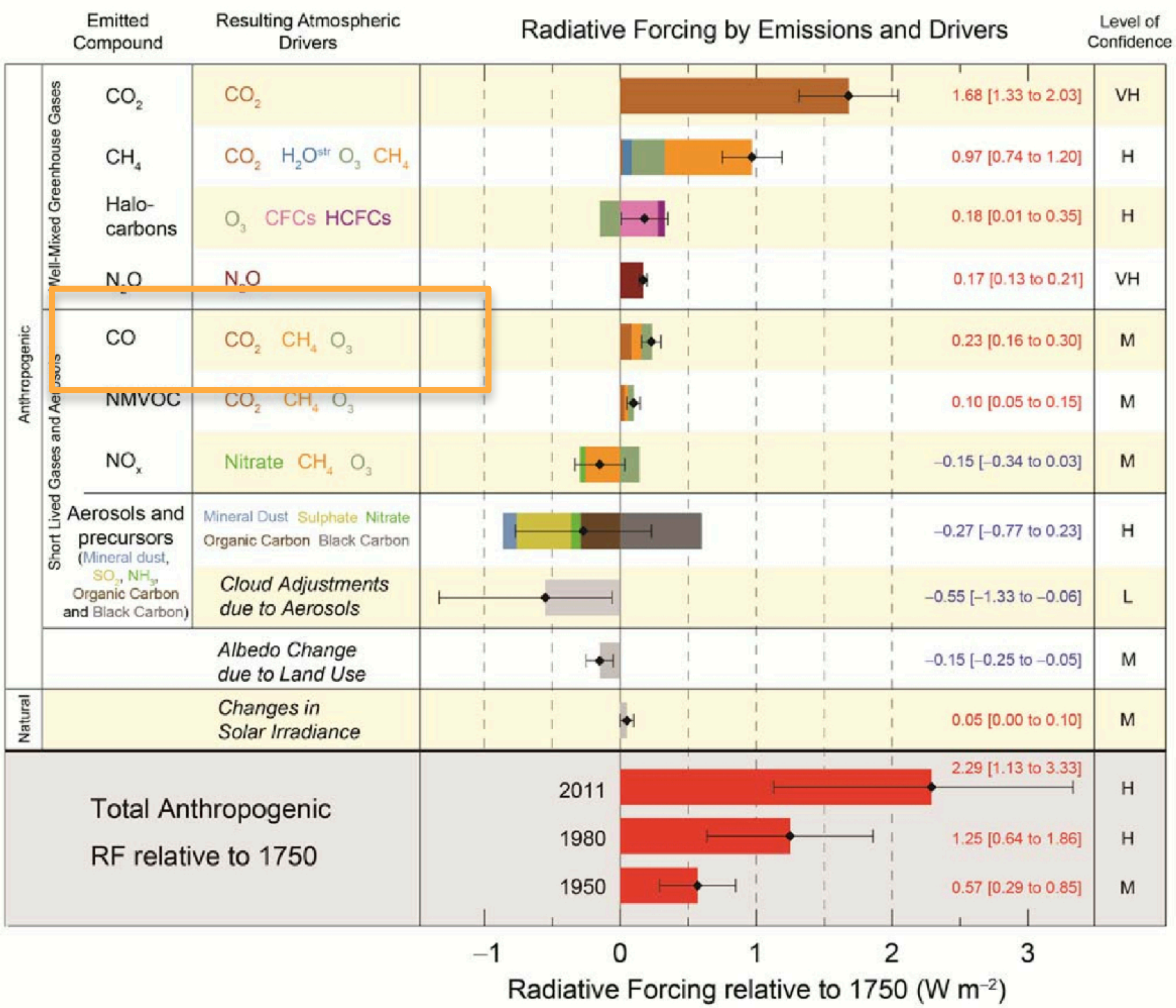


- The radiative forcing can be used to estimate the resulting global temperature change via

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

National Centre for Atmospheric Science
NATURAL ENVIRONMENT RESEARCH COUNCIL

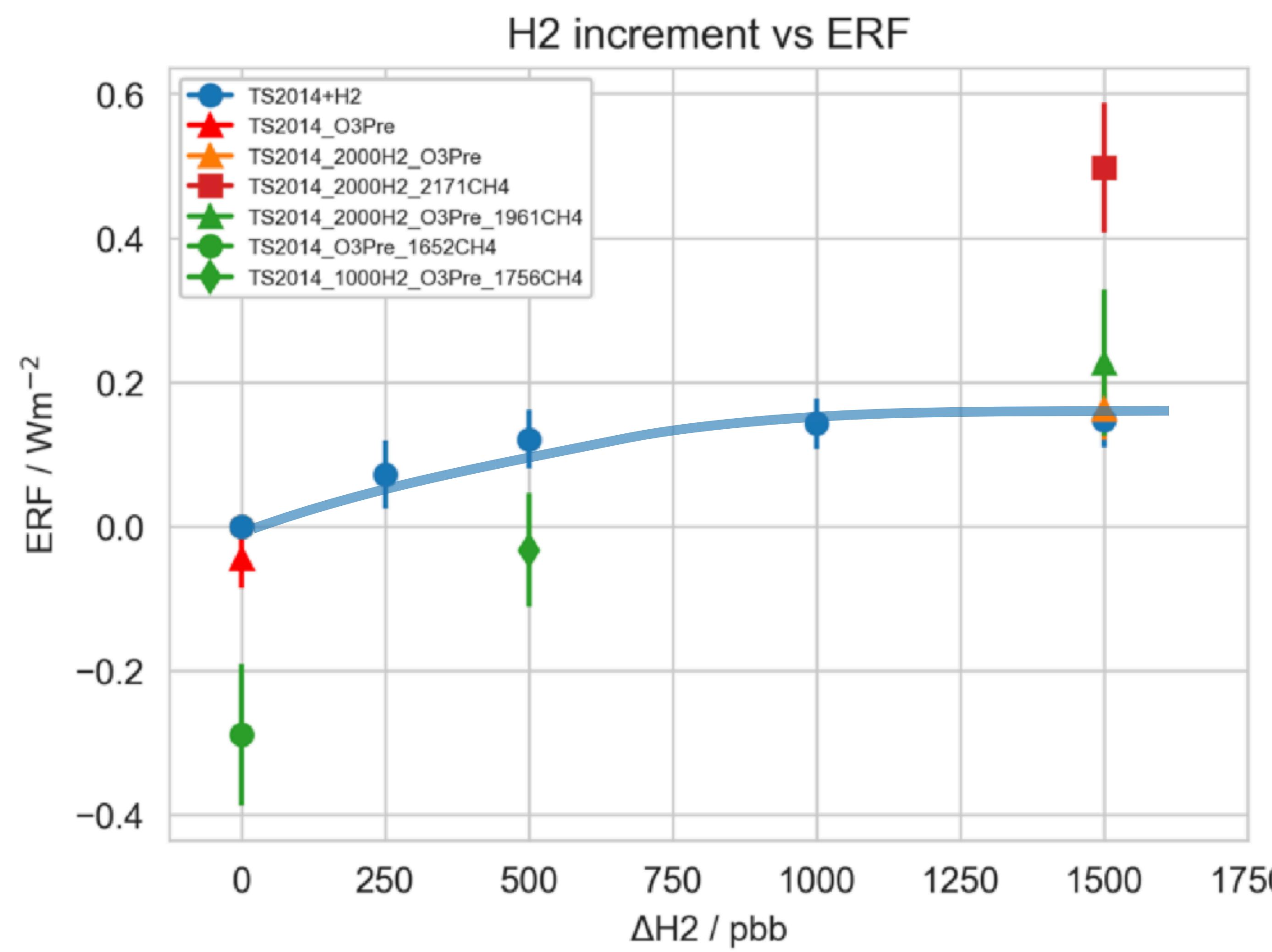
Effective radiative forcing - anthropogenic emissions



- Anthropogenic emissions affect the concentration of radiatively important gases such as CH4, O3
- Oxidants such as O3 also affect aerosol formation which can also perturb cloud properties
- ERF = ΔCS + ΔCRE - clear-sky (GG-dominated in the long wave) + Cloud Radiative Effects

Scenarios studied - what is the effect of H_2 fugitive emissions?

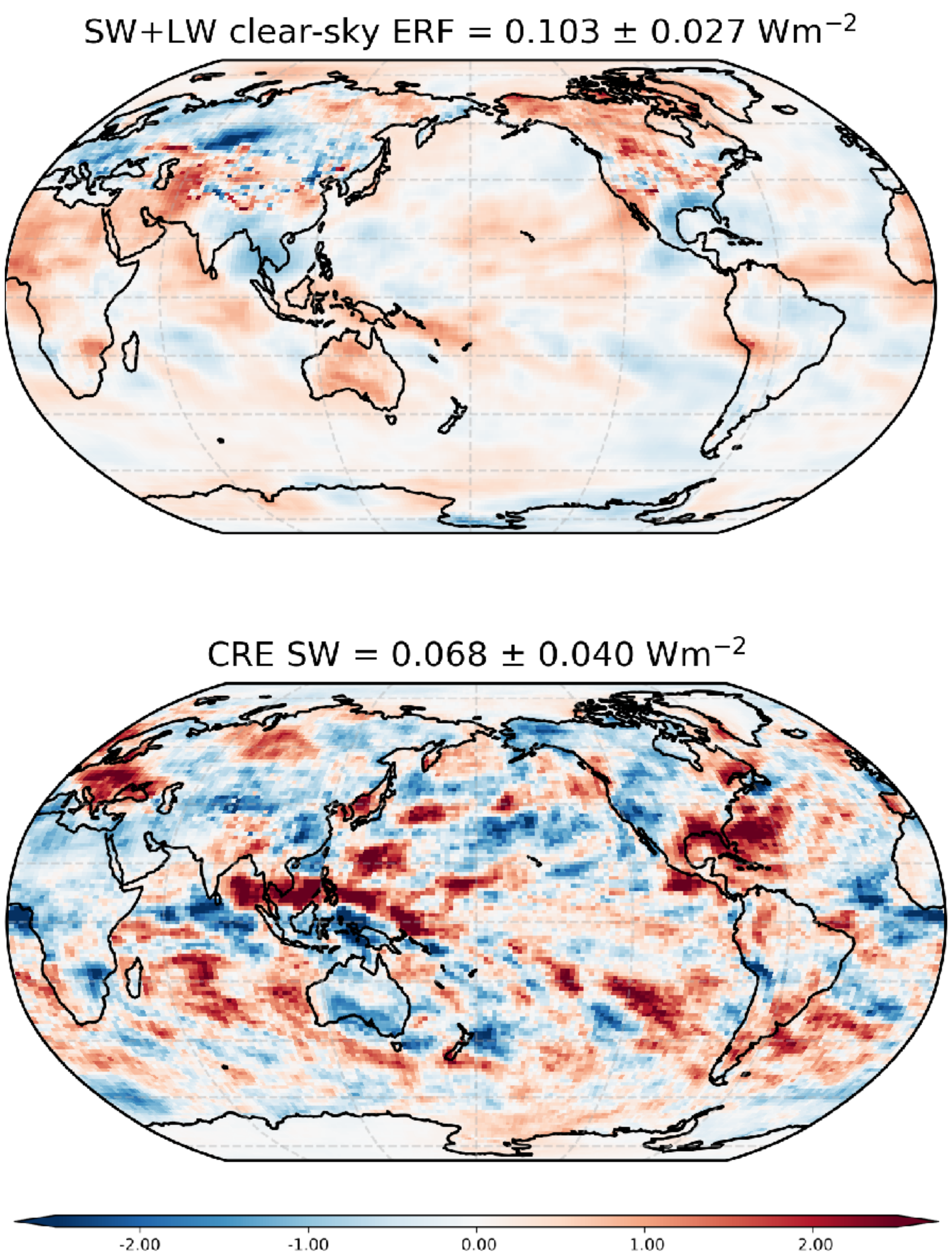
- Experiments with varying H_2 concentration in the atmosphere.
- The radiative forcing increases with increasing H_2 concentration, and is positive = a warming. Maybe a plateau?
- For the highest leak rates (an effective tripling of the global atmospheric H_2 source) ERF = 0.15 ± 0.08 Wm^{-2} which is approx 5% of the warming effect of CO_2
- Increasing H_2 levels see increases in methane lifetime and in ozone burden - can expect positive GG forcing.
- Increasing H_2 levels leads to decreased OH
- Potential impacts on stratospheric ozone.
- How to attribute the RF increase?



Experiment	H2 LBC	OH	TAU CH4	O3 Burden
	ppb	$10^6 cm^{-3}$	Years	Tg
Base	500	1.22	8.48	348.6
TS2014_750H2	750	1.20	8.67	347.3
TS2014_1000H2	1000	1.18	8.83	349.7
TS2014_2000H2	2000	1.11	9.46	353.5

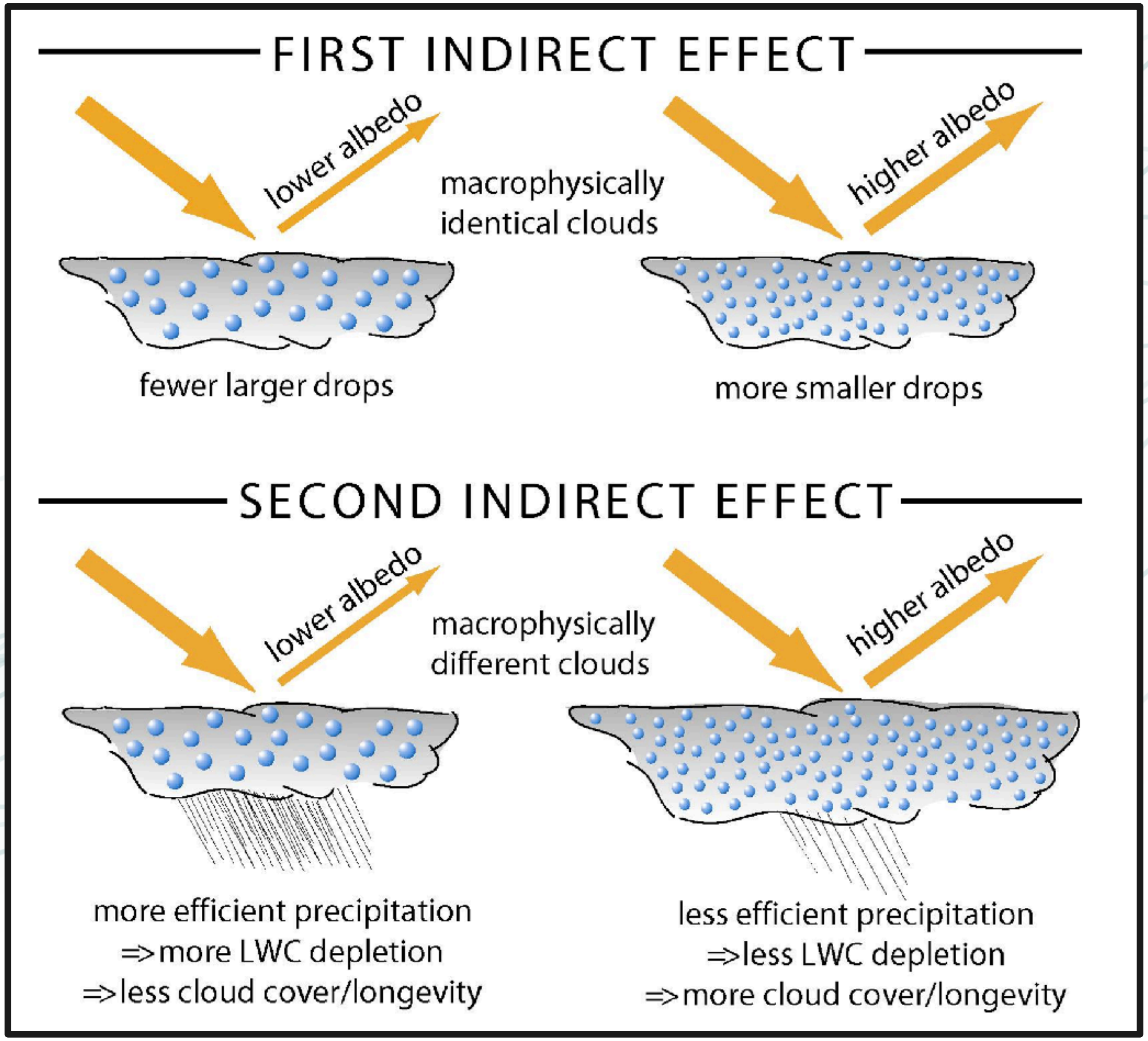
Breaking ERF down into clear-sky and cloud effects

- 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)
  - ΔCRE = 0.036 Wm^{-2}
- Which can be broken down further
  - Shortwave ΔCRE = 0.068 Wm^{-2}
  - Longwave ΔCRE = -0.032 Wm^{-2}
- i.e. the clear sky forcing is of the same order as the cloud radiative effect



Cloud radiative properties respond to aerosol changes

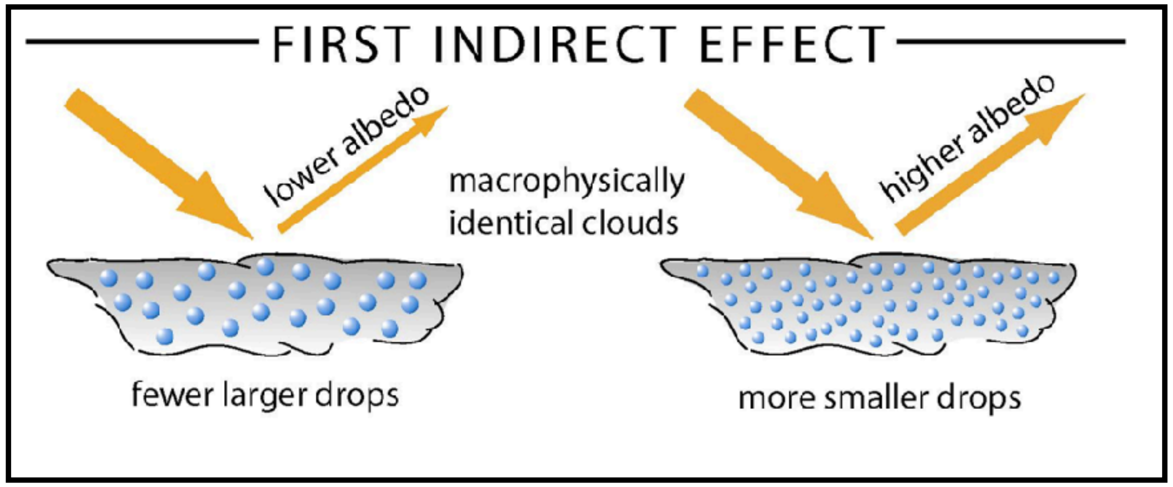
- Aerosol (CCN) controlled by atmospheric oxidation of gases like SO2, biogenic emissions, NOx.
- Clouds form on the aerosol (CCN) present in the atmosphere
- The cloud properties are sensitive to the number of aerosols
    - more aerosol → more cloud droplets
- More droplets means
    - a brighter cloud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

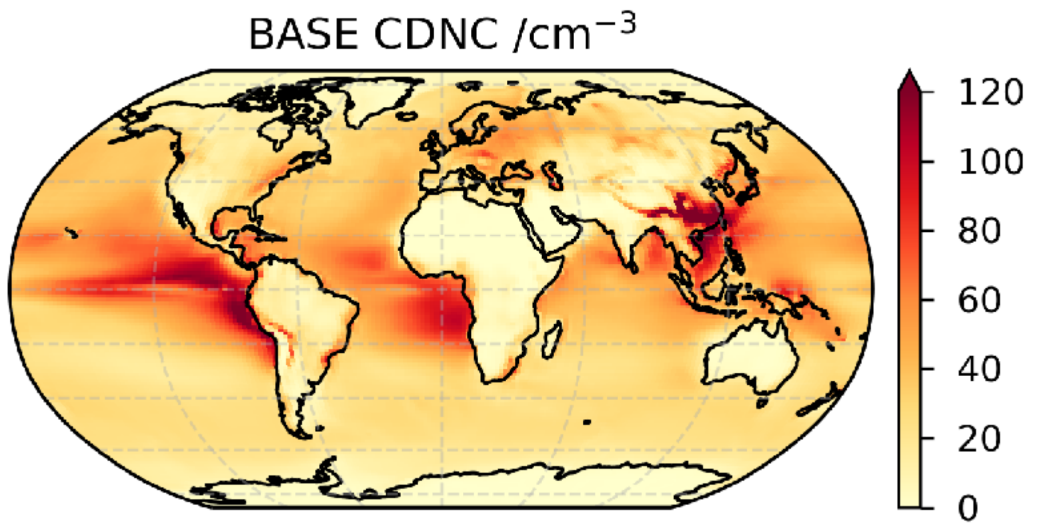


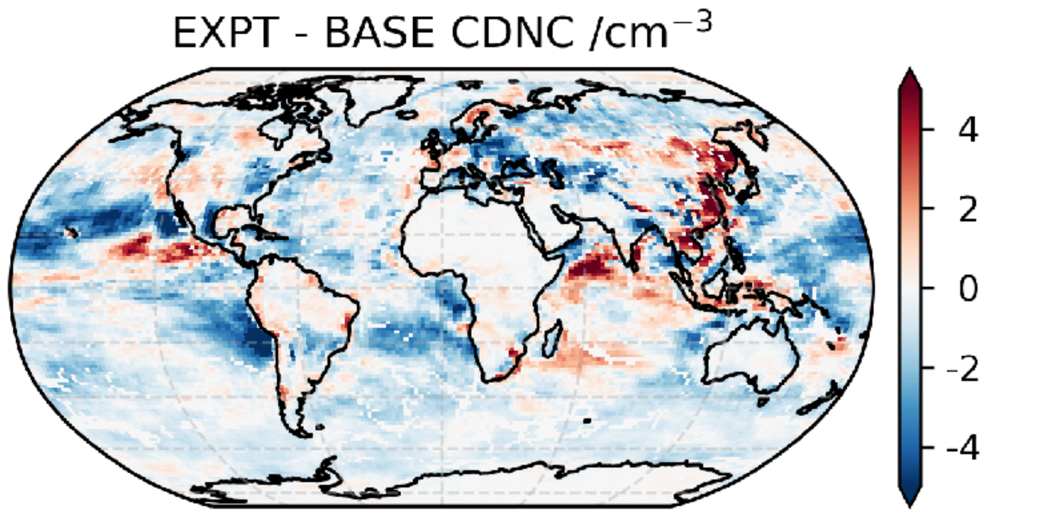
National Centre for Atmospheric Science
NATURAL ENVIRONMENT RESEARCH COUNCIL

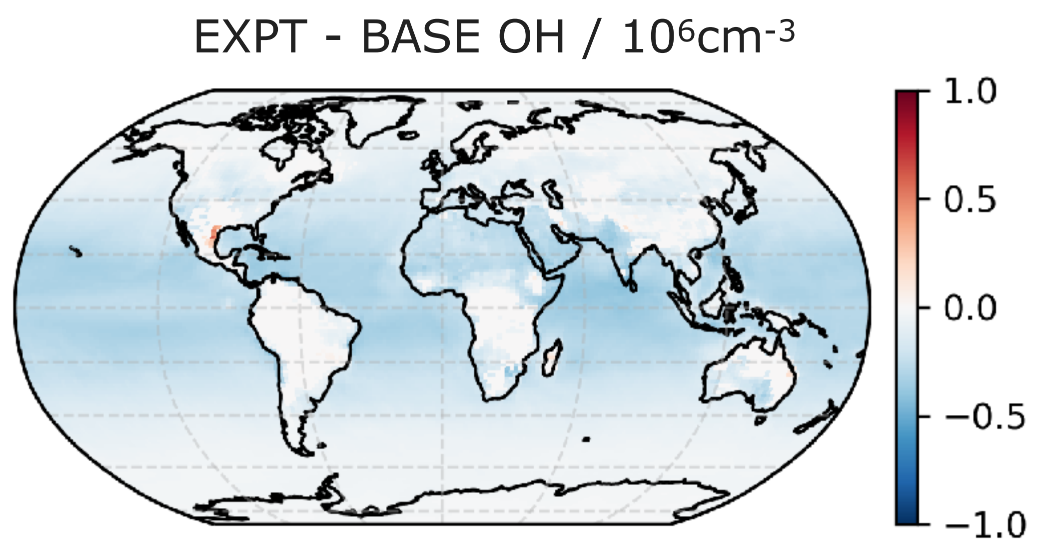
ERF - the coupling of gas phase oxidant to aerosol levels and cloud properties

- 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, NOx 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 H_2 (Tg H_2 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] $\pm$ 10
Tropospheric Lifetime, yr	2.1	1.9	2.2[b]	1.4	1.9	1.4	2.0

[a]Includes export to stratosphere of 1.9 Tg H_2 yr^{-1}.

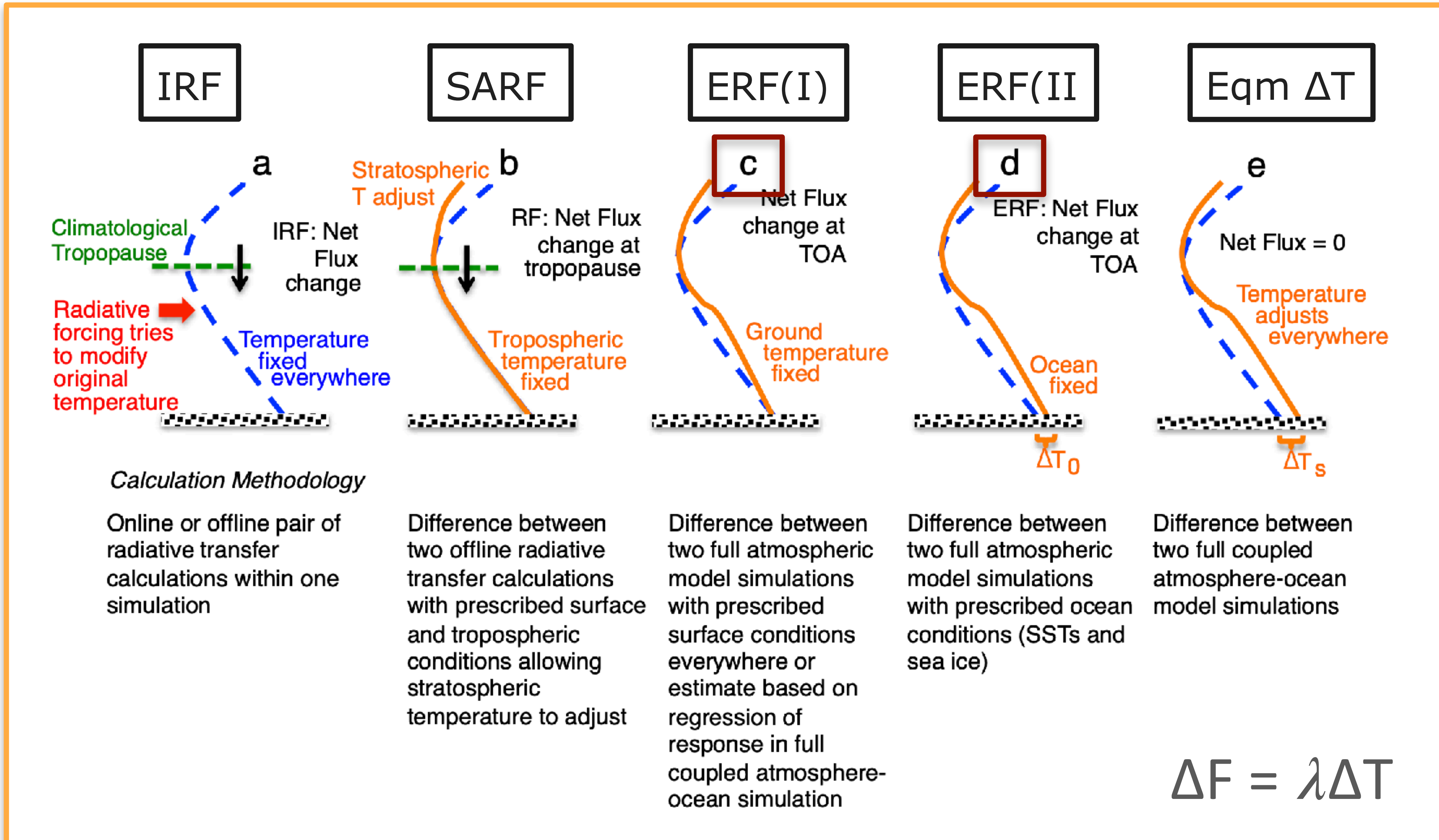
[b]Model domain reached 100 hPa; thus the burden includes about 1/2 of the stratosphere. Reduced to a troposphere holding 0.82 of the total air mass the burden would be 157 Tg H_2 and the tropospheric lifetime 2.0 yr.

[c]Calculated from sources and lifetime.

[d]From Novelli et al. (1999).

National Centre for Atmospheric Science
NATURAL ENVIRONMENT RESEARCH COUNCIL

Effective radiative forcing - definitions



- Calculation of ERF (Wm^{-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

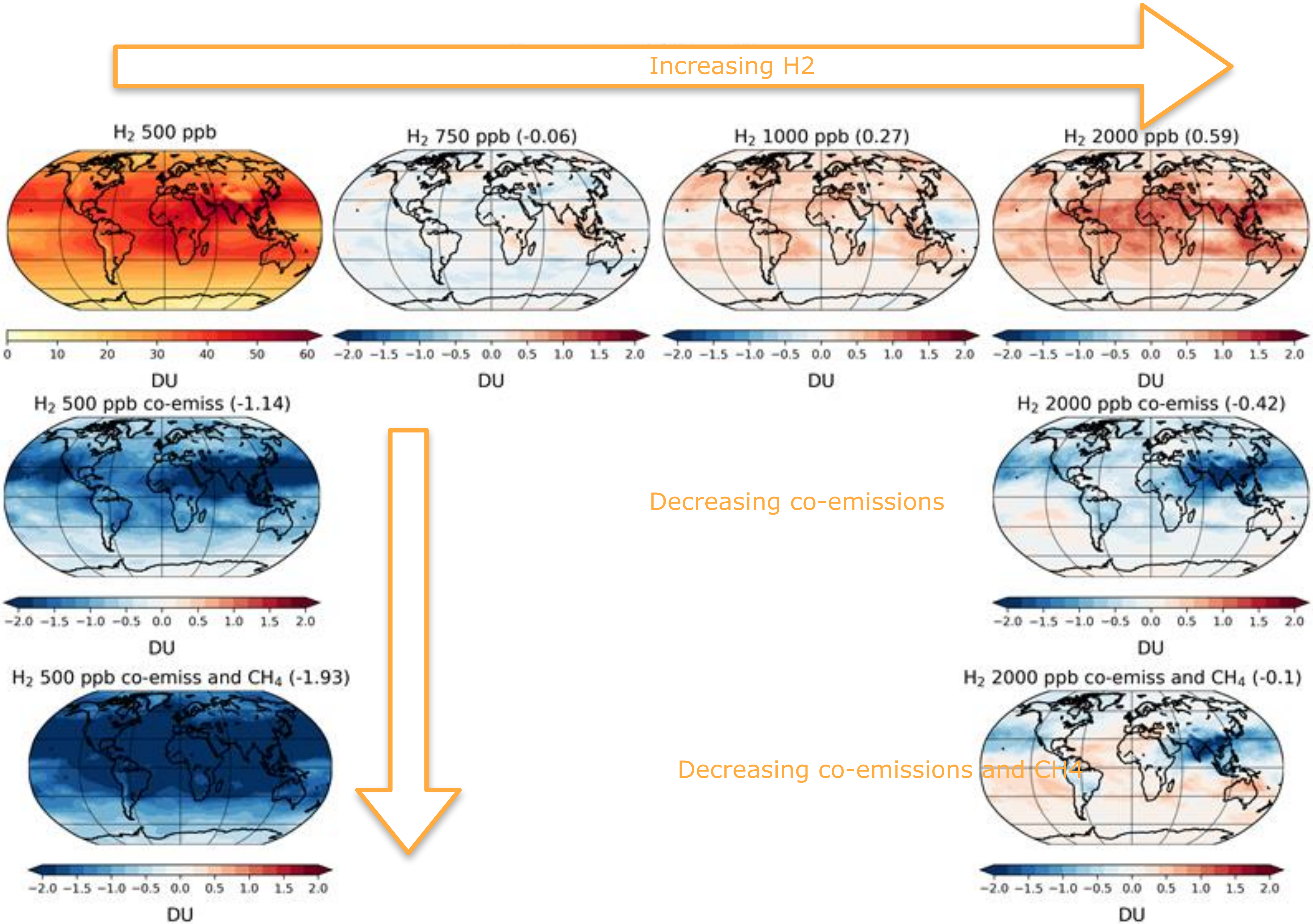


Figure by James Keeble