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

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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**

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paultgriffiths

Hello, my name is Paul Griffiths

- Senior Research Fellow (Grade 9) at Cambridge University
- Adjunct lecturer in atmospheric science.
- National Centre for Atmospheric Science, based in Cambridge
- IPCC Contributing Author, AR6
  - Co-chair UK Atmospheric Science Special Interest Group for Royal Met Soc
  - Co-chair Model Evaluation Working Group for UKCA chemistry-climate model
  - Visiting Scientist (NARIT, Chiang Mai, Thailand 2016-2020)
- Co-supervising three PhD students
  - Seb Hickman (Machine Learning/Ozone; Causal analysis)
  - Zosia Staniaszek (methane in future climate, COP26 Methane Pledge)
  - Vichawan Sakulsupich (climate forcing by aerosols)

Work to date

- You can find me on Google Scholar - can see my background

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Atmospheric chemistry

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TITLE	CITED BY	YEAR
An overview of current issues in the uptake of atmospheric trace gases by aerosols and clouds CE Kolb, RA Cox, JPD Abbatt, M Ammann, EJ Davis, DJ Donaldson, ... Atmospheric Chemistry and Physics 10 (21), 10561-10605	363	2010
UKESM1: Description and evaluation of the UK Earth System Model AA Sellar, CG Jones, JP Mulcahy, Y Tang, A Yool, A Wiltshire, ... Journal of Advances in Modeling Earth Systems 11 (12), 4513-4558	321	2019
Photochemical production of aerosols from real plant emissions TF Mentel, J Wildt, A Kiendler-Scharr, E Kleist, R Tillmann, MD Maso, ... Atmospheric Chemistry and Physics 9 (13), 4387-4406	134	2009
Methane mitigation: methods to reduce emissions, on the path to the Paris agreement EG Nisbet, RE Fisher, D Lowry, JL France, G Allen, S Bakkaloglu, ... Reviews of Geophysics 58 (1), e2019RG000675	114	2020
A comprehensive evaluation of water uptake on atmospherically relevant mineral surfaces: DRIFT spectroscopy, thermogravimetric analysis and aerosol growth measurements RJ Gustafsson, A Orlov, CL Badger, PT Griffiths, RA Cox, RM Lambert Atmospheric Chemistry and Physics 5 (12), 3415-3421	114	2005
Phase transitions and hygroscopic growth of aerosol particles containing humic acid and mixtures of humic acid and ammonium sulphate CL Badger, I George, PT Griffiths, CF Braban, RA Cox, JPD Abbatt Atmospheric Chemistry and Physics 6 (3), 755-768	111	2006

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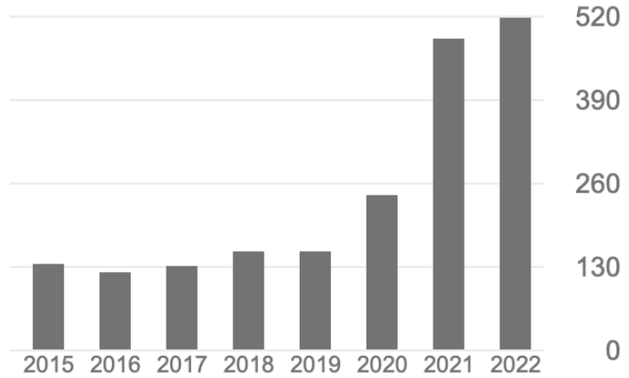

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Co-authors EDIT

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Work to date

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Reactive uptake coefficients for heterogeneous reaction of N 2 O 5 with submicron aerosols of NaCl and natural sea salt DJ Stewart, PT Griffiths, RA Cox Atmospheric Chemistry and Physics 4 (5), 1381-1388	57	2004	
The intriguing steric dependence and interfering behavior of rotationally inelastic NO-Ar, He and D {sub 2} collisions M de Lange, M Wisse, G Boon, P Griffiths, S Stolte, C Taatjes, M Drabbles, ...		2002	Molecular dynamics days
The photodissociation of physisorbed alkyl nitrites.: Λ-doublet population and alignment of desorbed NO PT Griffiths, C Simpson, S Stolte, M Towrie Chemical Physics Letters 315 (3-4), 158-166	4	1999	
Steric asymmetry in state-resolved NO–Ar collisions MJL de Lange, M Drabbels, PT Griffiths, J Bulthuis, S Stolte, JG Snijders Chemical physics letters 313 (3-4), 491-498	71	1999	Chemical physics days
Steric asymmetries of fine structure conserving collisions of NO and Ar S Stolte, MJL de Lange, M Drabbels, PT Griffiths, J Bulthuis, JG Snijders Faraday discussions of the Chemical Society 113, 484		1999	
Photodissociation of physisorbed molecules. PT Griffiths University of Oxford		1998	
Photodissociation of alkyl nitrites adsorbed on an MgF2 surface. Rotational and translational energy distributions of product NO (v, J) molecules C Simpson, PT Griffiths, HL Wallaart, M Towrie Chemical physics letters 263 (1-2), 19-24	22	1996	

Talk outline

- Ozone in the troposphere
  - Is formed from Volatile Organic Compounds (VOC) and nitrogen oxide emissions
  - Is a non-linear chemical system: high NOx causes a decrease in ozone production
- Ozone in the CMIP6 era
  - Natural emissions - LNOx and VOC
  - Methane and oxidants
  - The role of the stratosphere
- Outlook

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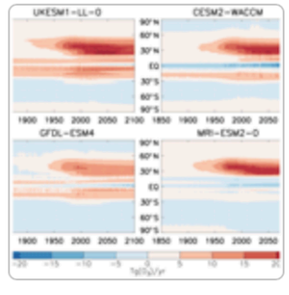
Tropospheric Ozone in CMIP6

Atmos. Chem. Phys., 21, 4187–4218, 2021
https://doi.org/10.5194/acp-21-4187-2021

Research article

18 Mar 2021

Tropospheric ozone in CMIP6 simulations



Paul T. Griffiths[1,2,★], Lee T. Murray[3,★], Guang Zeng[4], Youngsub Matthew Shin[1], N. Luke Abraham[1,2], Alexander T. Archibald[1,2], Makoto Deushi[5], Louisa K. Emmons[6], Ian E. Galbally[7,8], Birgit Hassler[9], Larry W. Horowitz[10], James Keeble[1,2], Jane Liu[11], Omid Moeini[12], Vaishali Naik[10], Fiona M. O'Connor[13], Naga Oshima[5], David Tarasick[12], Simone Tilmes[6], Steven T. Turnock[13], Oliver Wild[14], Paul J. Young[14,15], and Prodromos Zanis[16]

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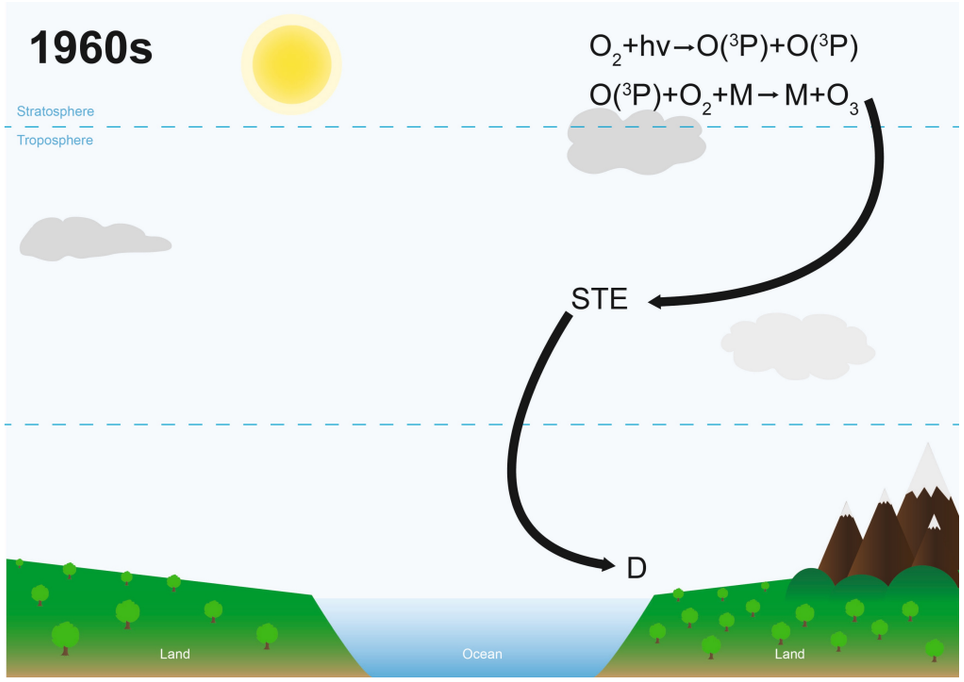
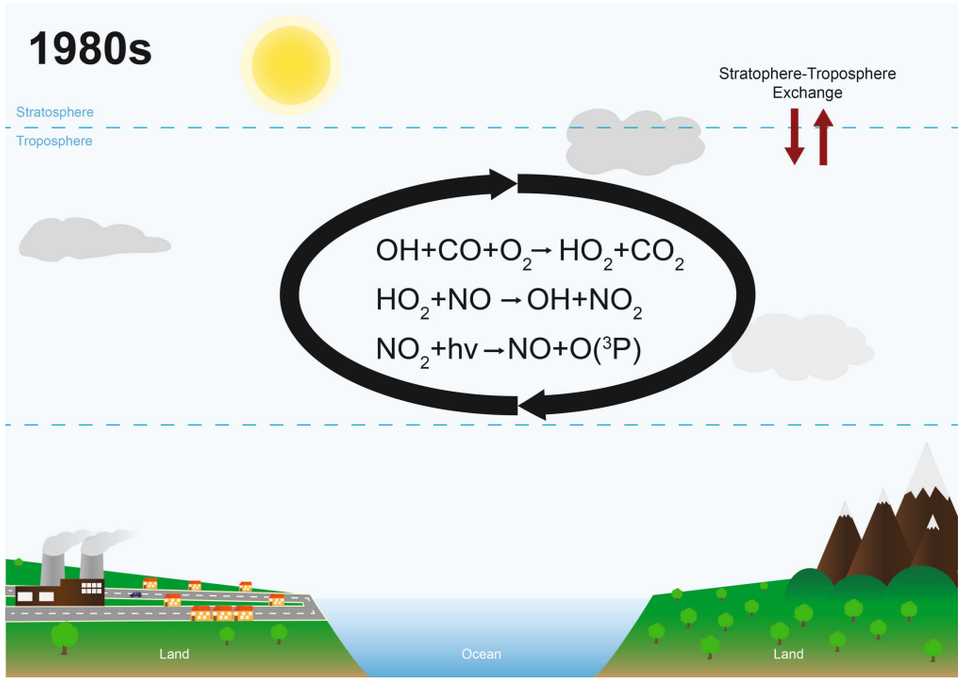
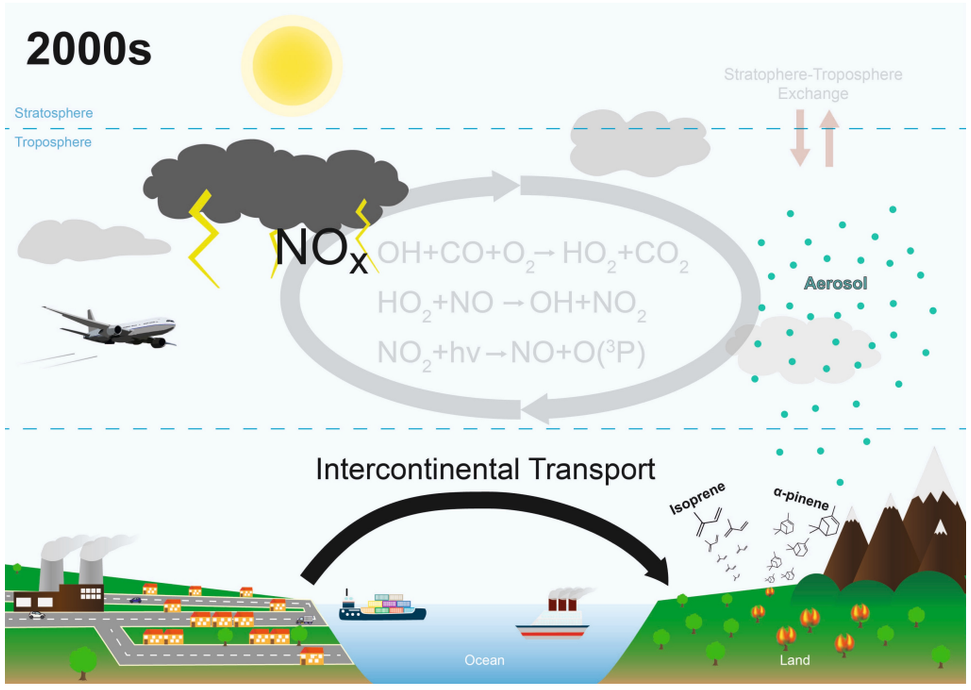
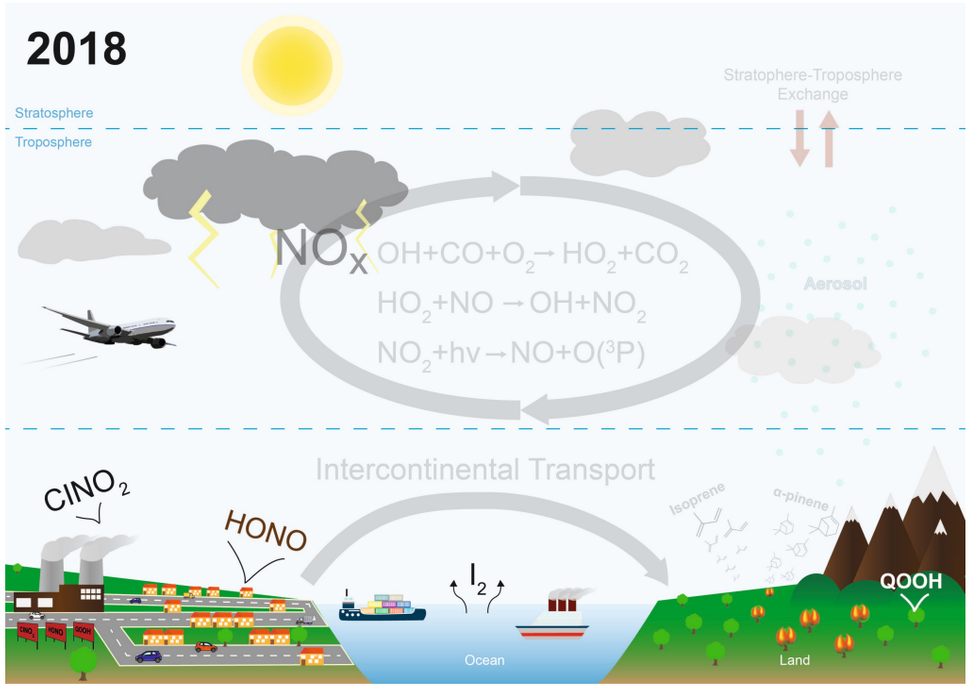
Ozone in CCMs – developing complexity









Archibald et al., TOAR “Budget”, Elementa 2021

How does tropospheric ozone evolve in CMIP6?

- How do emissions changes influence climate (and vice versa)?
- Multi model assessments provide us with an estimate of uncertainty.
- CMIP6 featured coupled atmosphere-ocean models with online, whole-atmosphere chemistry.
- **Transient experiments** (AR5 and ACCMIP relied mostly on timeslice experiments)
- **Whole atmosphere models** - interactive stratosphere, captures the effec of stratospheric ozone depletion and recovery

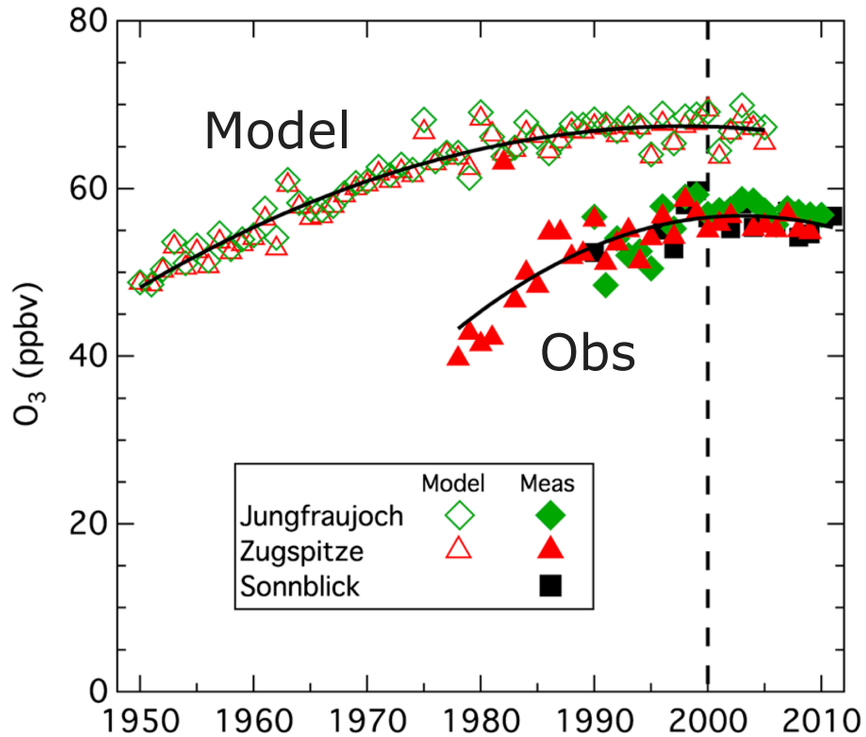


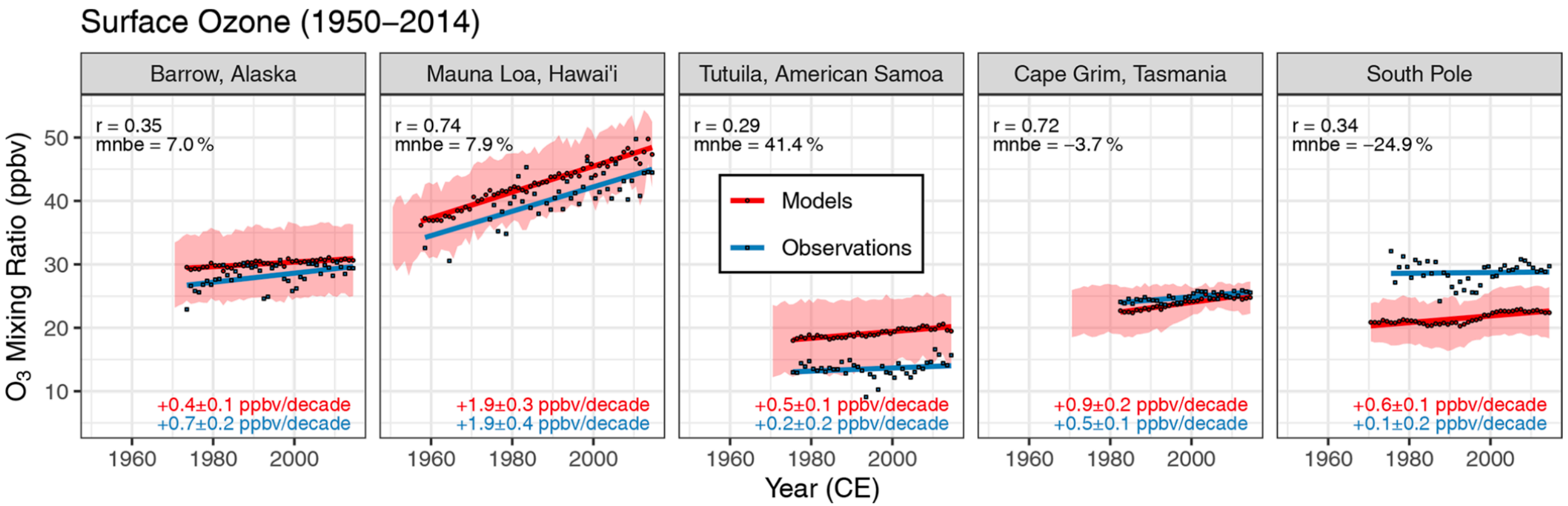
Figure 1. Seasonally averaged springtime (March, April, and May) O_3 concentrations at alpine sites in Europe. Closed and open symbols give measurements and GFDL CCM results, respectively. The solid lines give quadratic fits to respective results. The vertical dashed line indicates the year 2000 reference.

- **Earth System models -** online BVOC and NOx, vegetation sinks for ozone
- **Interactive aerosol formation** - secondary aerosols responding to changes in oxidants
- **AR6 deadline** - submission by December 31st 2019; acceptance by January 31st 2020 - not all models available!!

Models with online whole-atmosphere chemistry featured in CMIP6 with data on BADC as of 2021-12-04

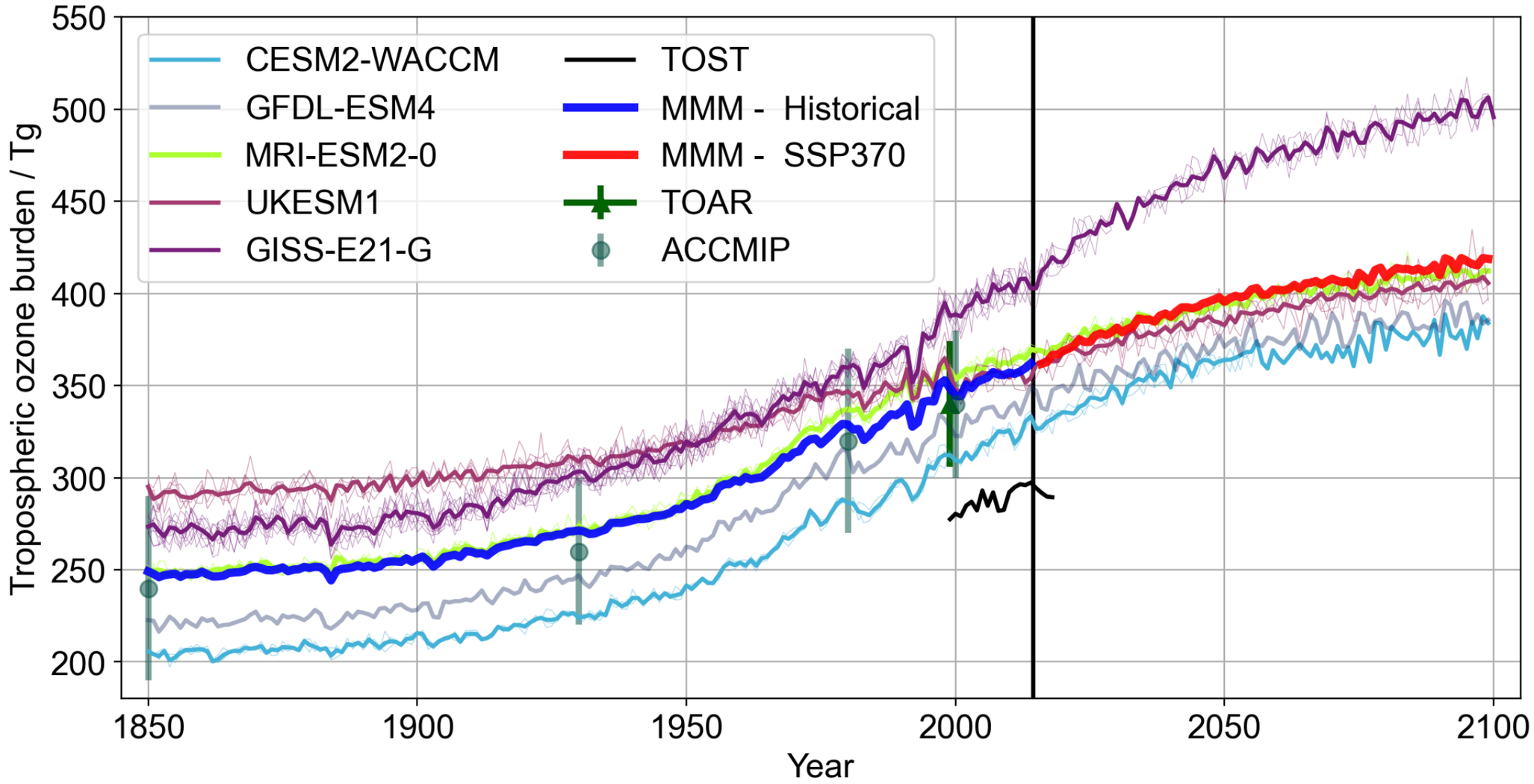
Model	SSP119	SSP126	SSP245	SSP370	SSP434	SSP460	SSP534	SSP585
UKESM1-0-LL								
CESM2-WACCM								
MRI-ESM2-0								
GFDL-ESM4								
GISS-ES2-1-G								

How does tropospheric ozone evolve in CMIP6? Comparison with obs



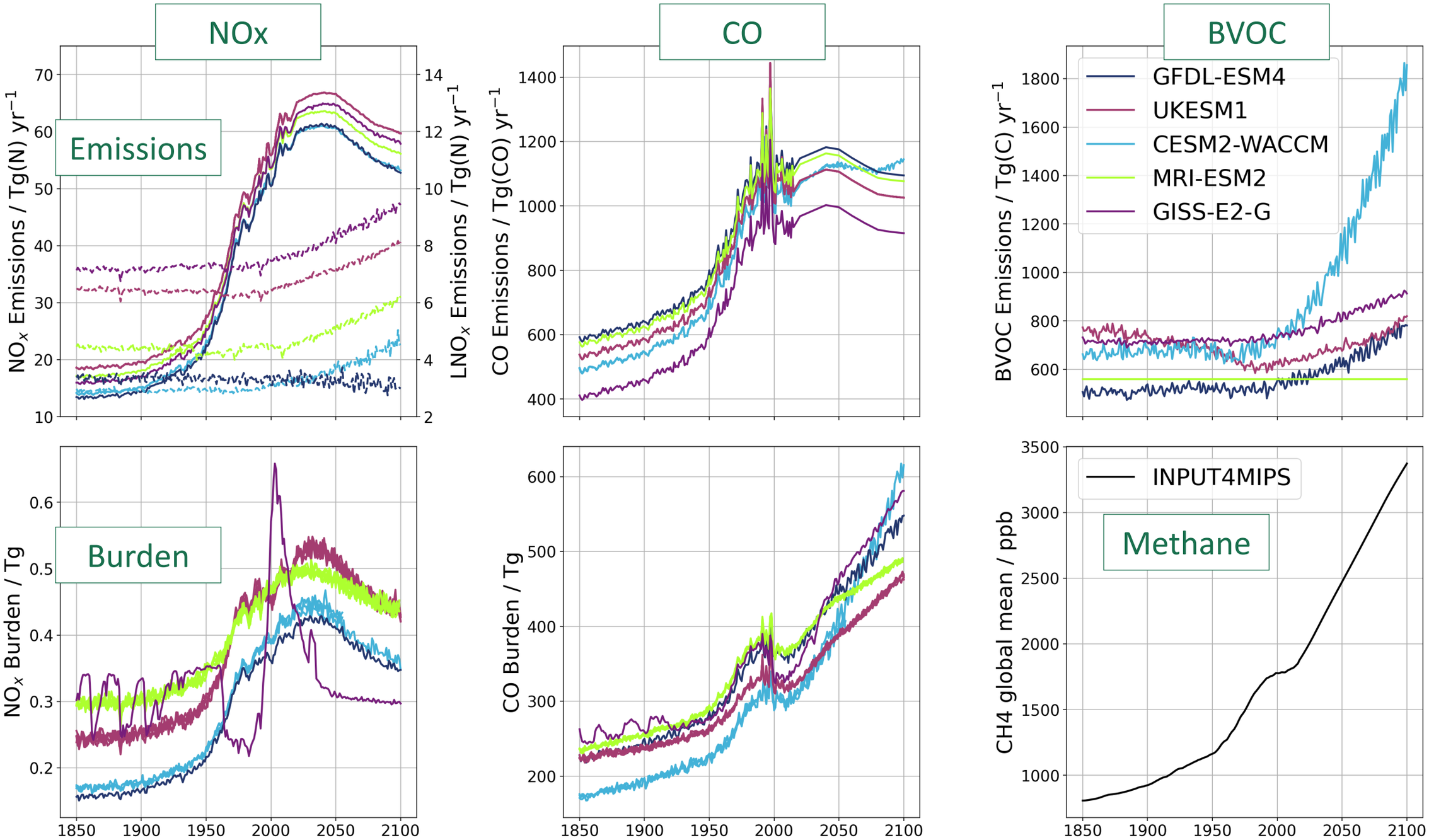
- CMIP6 featured coupled atmosphere-ocean models with online, whole-atmosphere chemistry.
- **Good agreement between models and observations for the remote sites studied here.**
- Also found nice agreement between in-situ ozone sonde measurements.
- **Assessment using EO products more of a challenge** - tropopause definition?
- Consistent model biases in simulating the seasonality of free-tropospheric ozone in equatorial America, Japan and northern high latitudes and near-surface ozone over northern and north-eastern Europe.

How does UKESM1 tropospheric ozone evolve in CMIP6?



- CMIP Historical and ScenarioMIP SSP3-70 experiments, for which suitable diagnostic output was available.
- Picture has changed little since CMIP5/CCMI, MM range is also similar.
- Ozone burden increased by about 40% from 1850 levels of 240 Tg (MMM) with steepest rate of increase around 1960.
- In SSP3-70, the rate of growth of the burden declines further, as NOx emissions start to fall along this pathway after 2050.

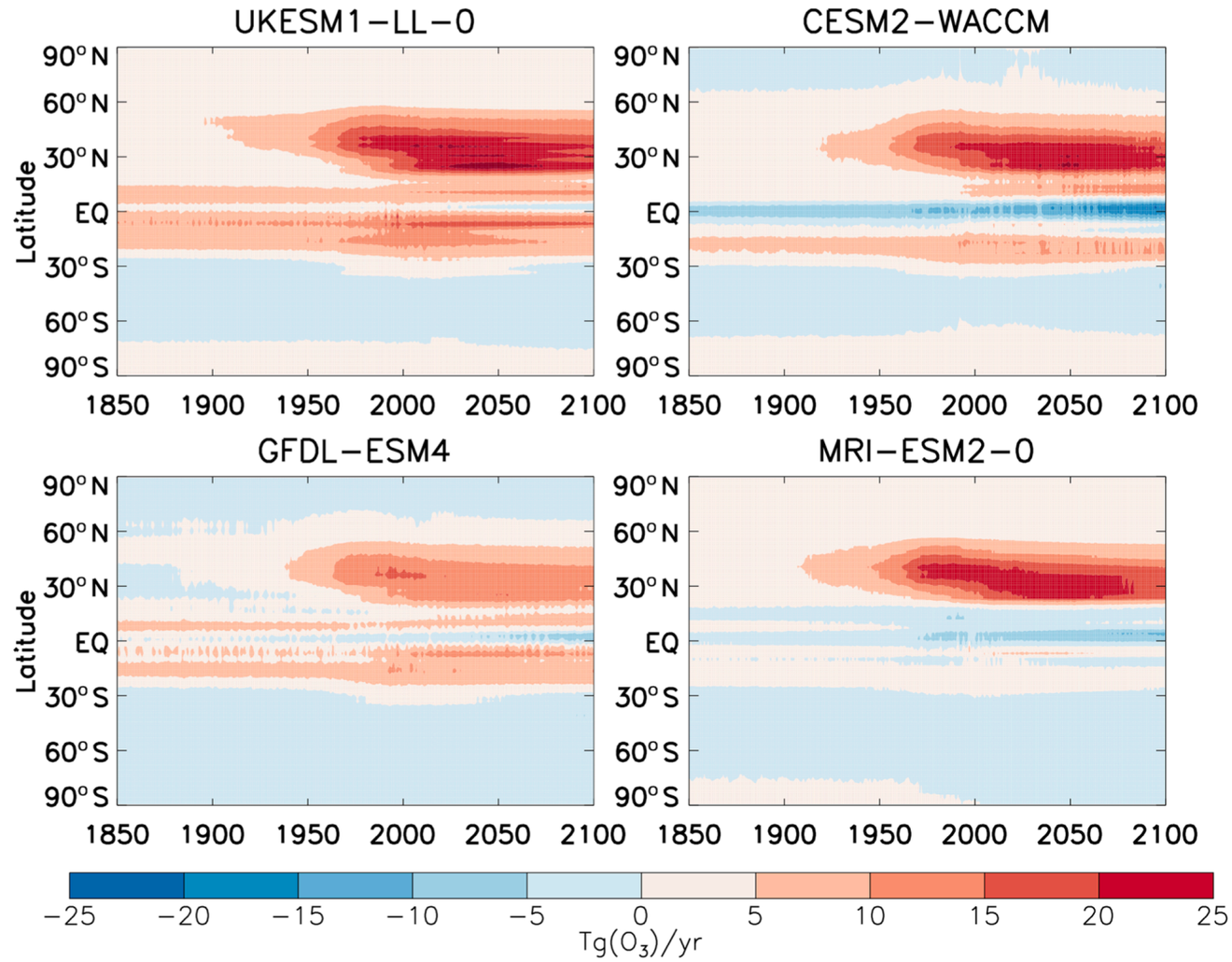
What drives tropospheric ozone in CMIP6?

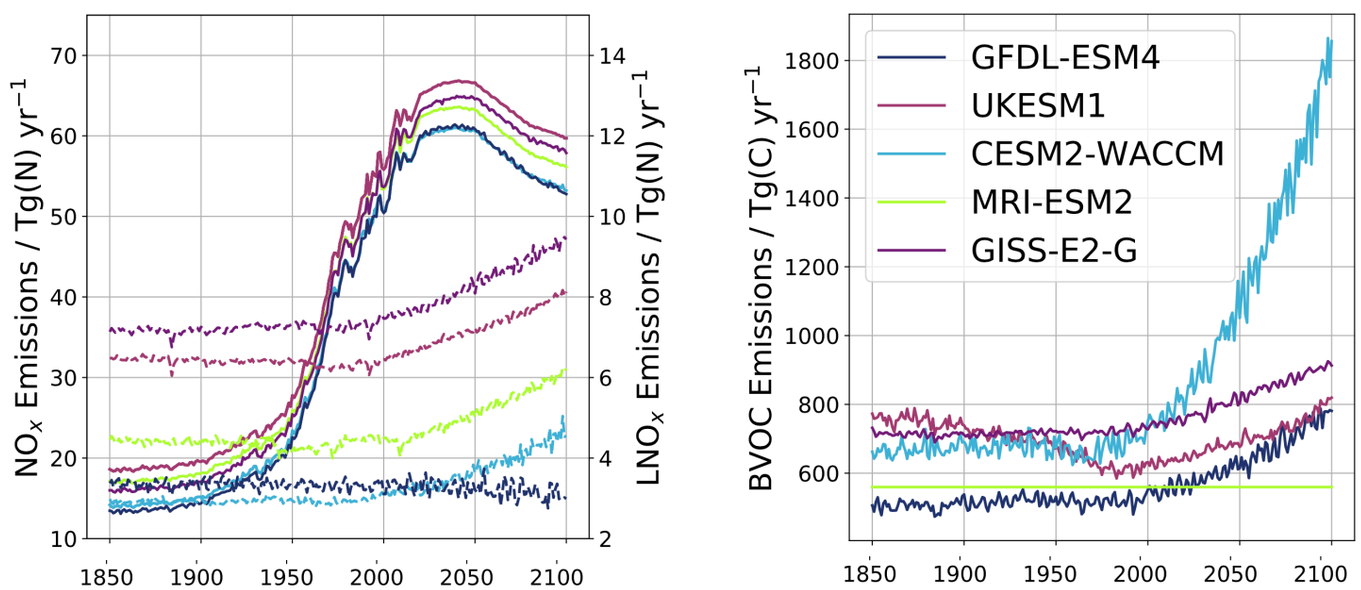


- Decline in precursor emissions in SSP3-70 experiments
- steady increase in ozone burden - Strat O3 recovery increasing role + LiNOx

What drives tropospheric ozone budget in CMIP6?

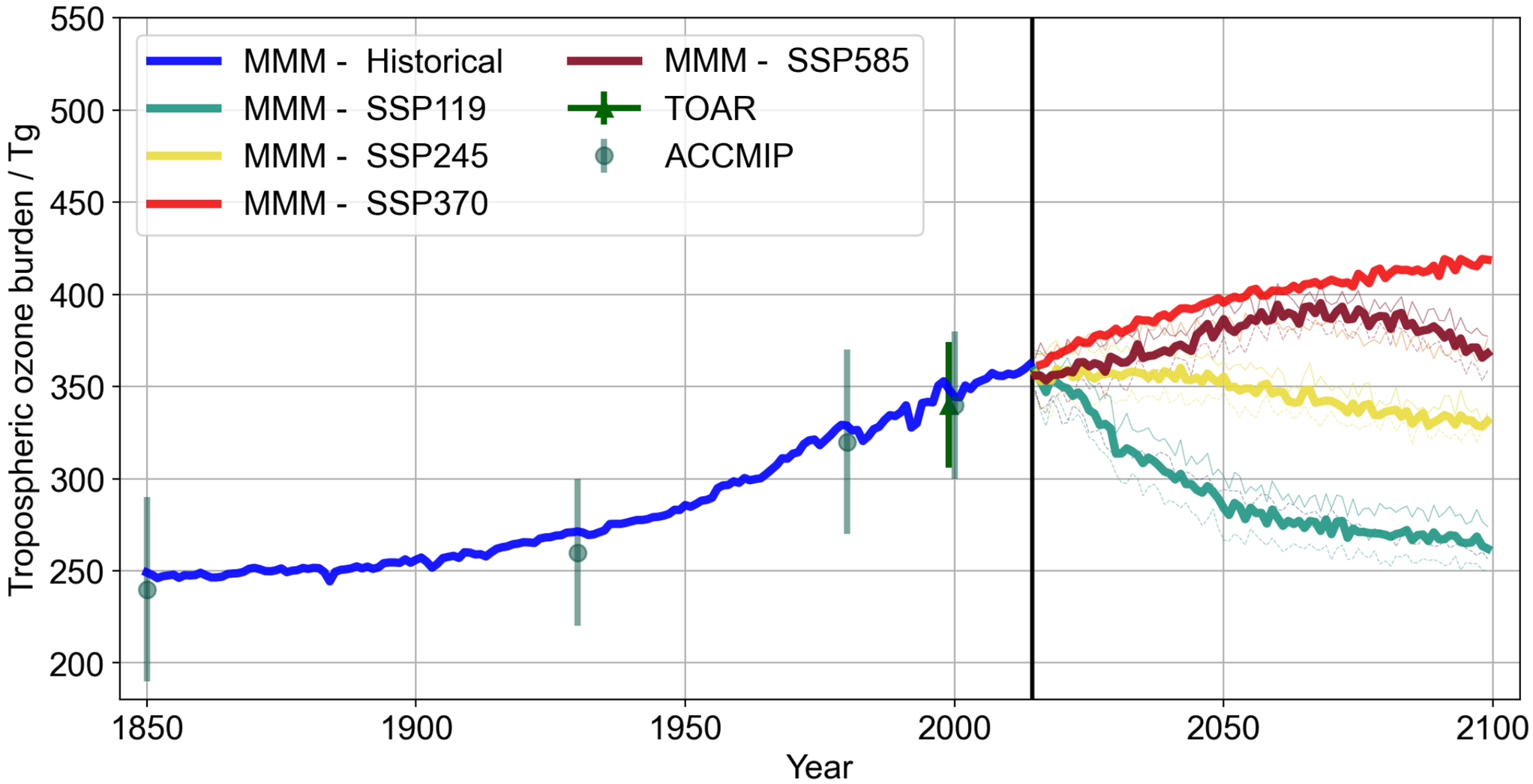
- Analysis so far has focused on CMIP Historical and ScenarioMIP SSP3-70 experiments, for which suitable diagnostic output was available.
- Quite a strong diversity in net ozone production: UKESM1 and MRI-ESM2 show O3 production throughouth the NH in 1850.
- Equatorward shift in emissions after 1980
- Maximum of in-situ chemical production in the period 2000-2050,
- Strong local changes in ozone seen regionally at the end of the century.
- EMIBVOC rather diverse!
- LNOx increasing in importance





Tropospheric ozone precursor emissions

Database of of tropospheric ozone burden changes



- Initial results (dataset is rather incomplete)

Models with online whole-atmosphere chemistry featured in CMIP6 with data on BADC as of 2021-12-04								
Model	**SSP119**	**SSP126**	**SSP245**	**SSP370**	**SSP434**	**SSP460**	**SSP534**	**SSP585**
UKESM1-0-LL				■				
CESM2-WACCM				■				
MRI-ESM2-0	■	■	■	■	■	■	■	■
GFDL-ESM4	■	■	■	■				■
GISS-ES2-1-G				■				

Conclusions 1/4 - Trop O3 in CMIP6

- CMIP6 historical experiments performed well against observations for both trends and absolute amounts
- CMIP6 exercise was limited by data availability - hard to define outliers.
- Picture changed little from CMIP5
- Online model components - LNOX, BVOC emissions - drive model differences in the PI/1850.
    - Models with higher PI BVOC have higher ozone, lower PI-PD changes
    - Evaluation of processes becomes more critical for ESMs
- Future ozone depends on the SSP - co-benefits of SSP126/SSP245 seen
- Evaluation still rather limited by the CMIP6 timeline - most centres now moved on to CCMI2022

The role of methane and oxidants

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Methane is important to climate forcing

- 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

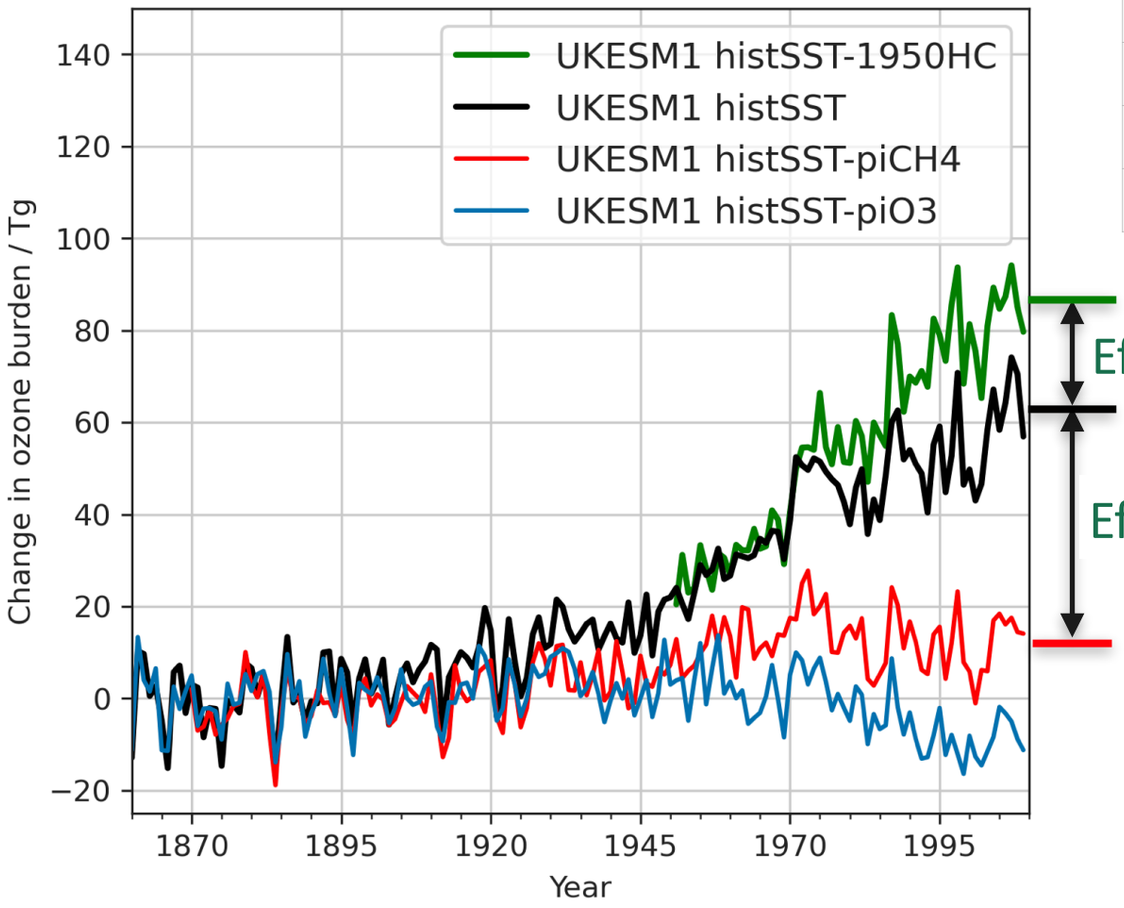
- A large Global Warming Potential – 28 on a 100-year horizon (per-molecule w.r.t. CO_2)
- Strong sources – 585 Tg CH_4 per year, with strong chemical sinks. Lifetime of 10 years
- Methane oxidation leads to ozone and water vapour – both greenhouse gases – with methane an important source of stratospheric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

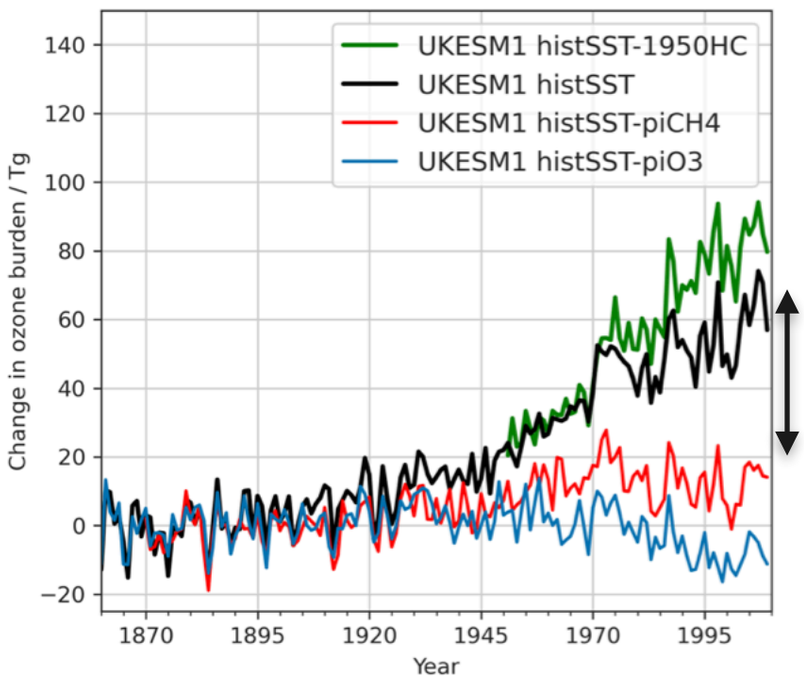
Sources	Wetlands	Fossile fuels gas and coal	Termites	Ruminants	Rice	Waste landfill	Biomass burning
Tg CH_4 per year	177-284	85-105	2-22	87-94	33-40	67-90	32-39

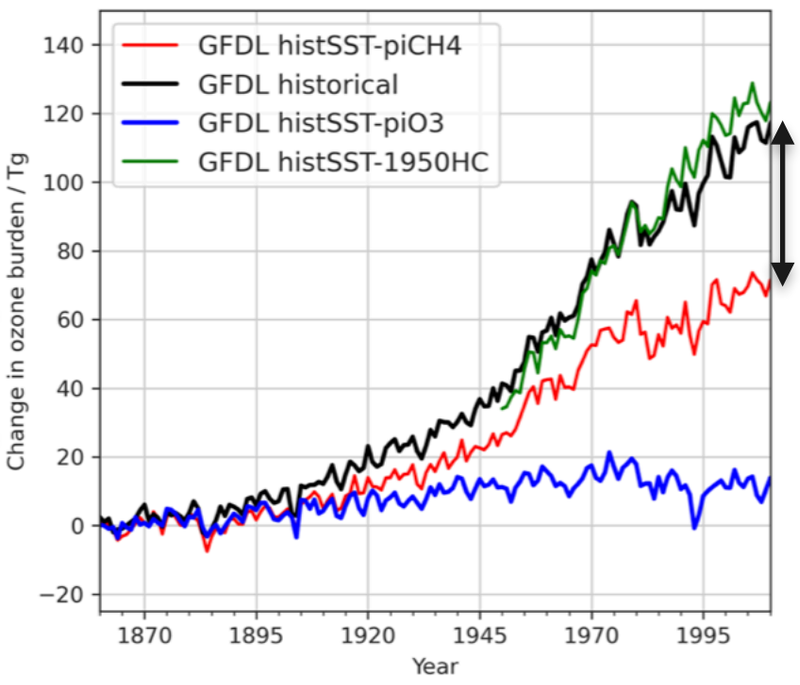
Sinks	Tropospheric OH	Stratospheric loss	Tropospheric Cl	Methanotrophs
Tg CH_4 per year	454-617	40	13-37	9-47
Lifetime*	10 years	120 years	160 years	160 years

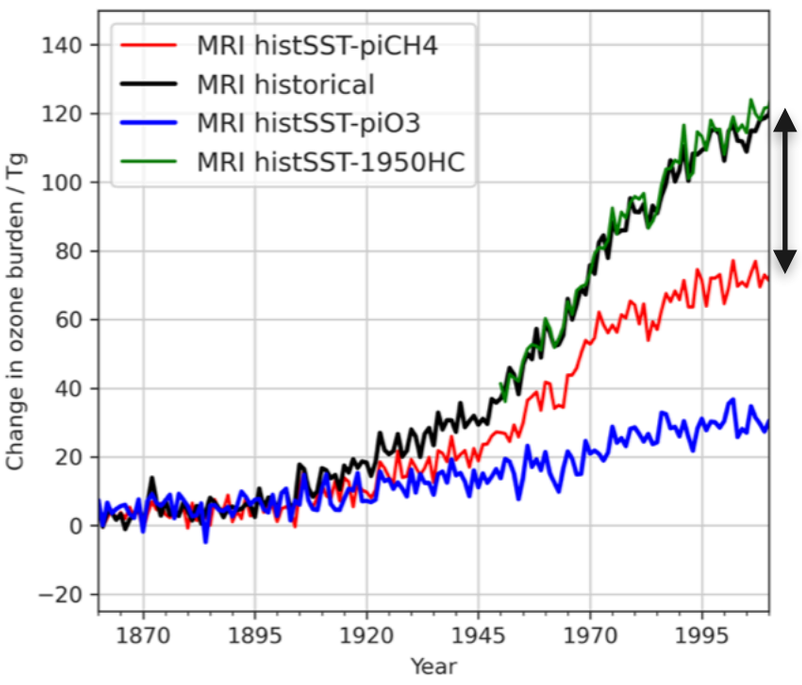
Methane is important to tropospheric ozone

Experiment_ID	CH4	N2O	AERPRE	O3PRE	CFC/HCFC
histSST	Hist	Hist	Hist	Hist	Hist
histSST-piAer	Hist	Hist	1850	Hist	Hist
histSST-piO3	Hist	Hist	Hist	1850	Hist
histSST-piCH4	1850	Hist	Hist	Hist	Hist

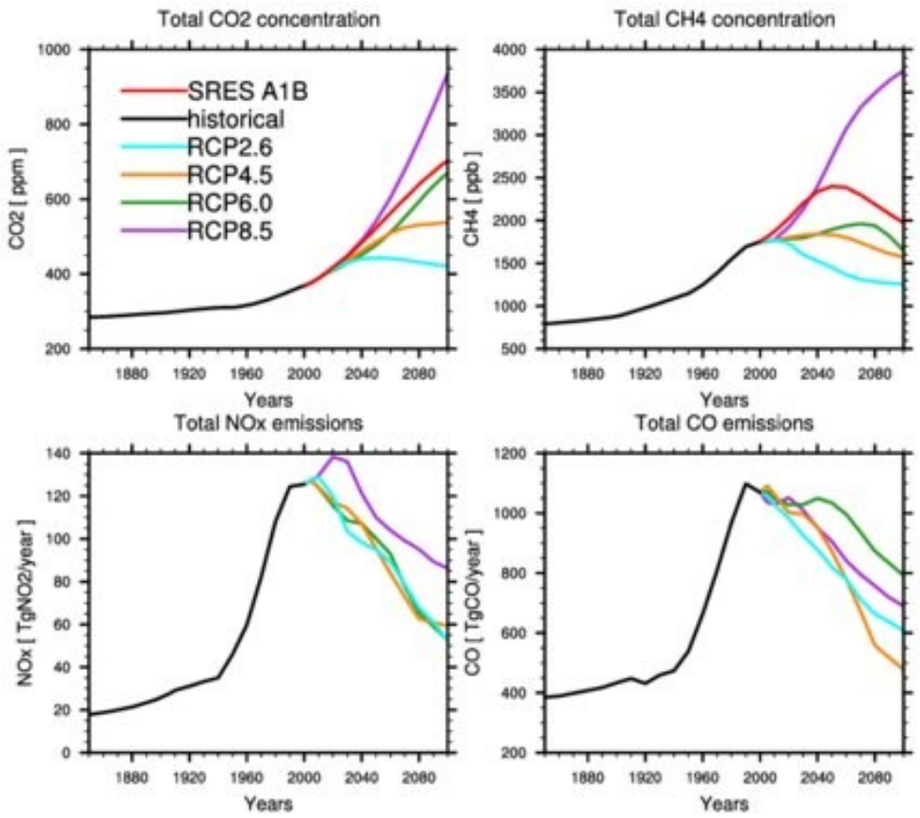






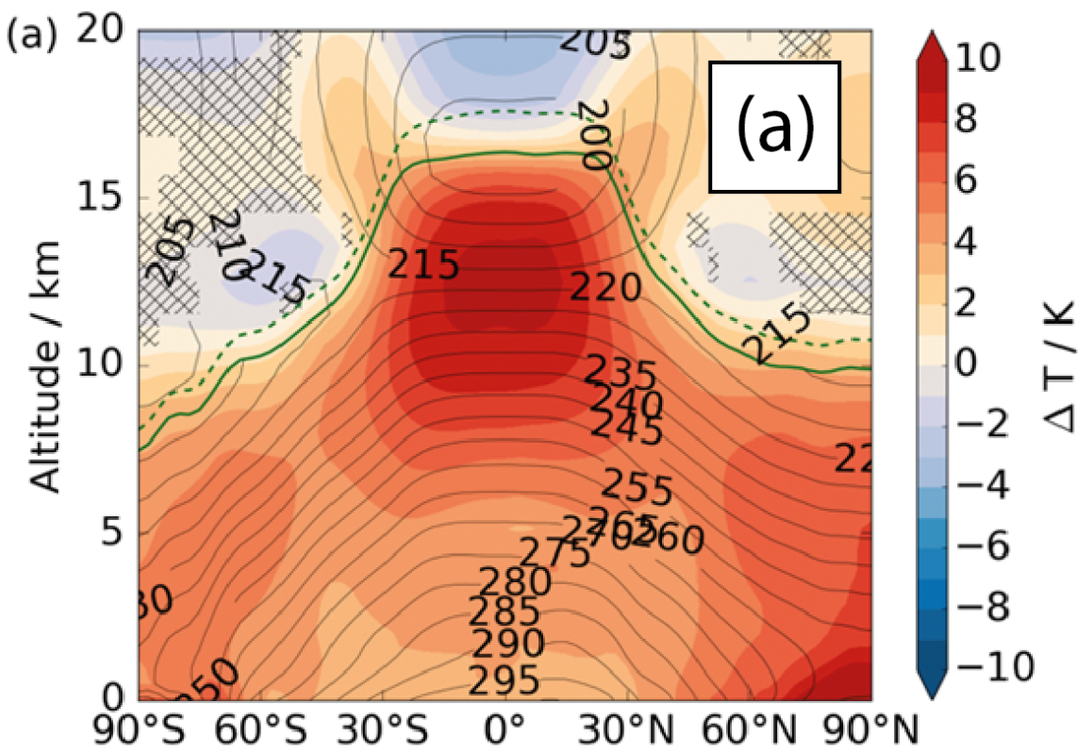


Climate change is important to methane - sinks

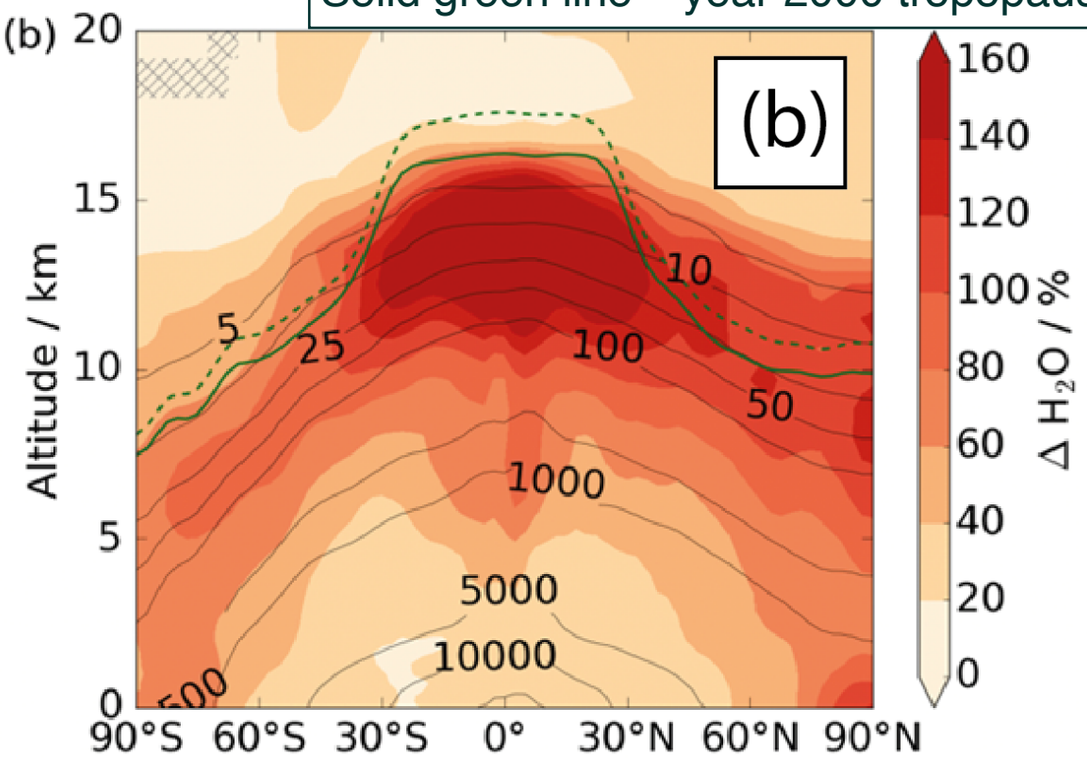


- In RCP8.5 there's a big increase in temperature throughout the troposphere by 2100.
- The warmer atmosphere can support more water vapour, so humidity increases.
- Tropospheric expansion means the upper troposphere experiences the biggest changes.

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



Temperature

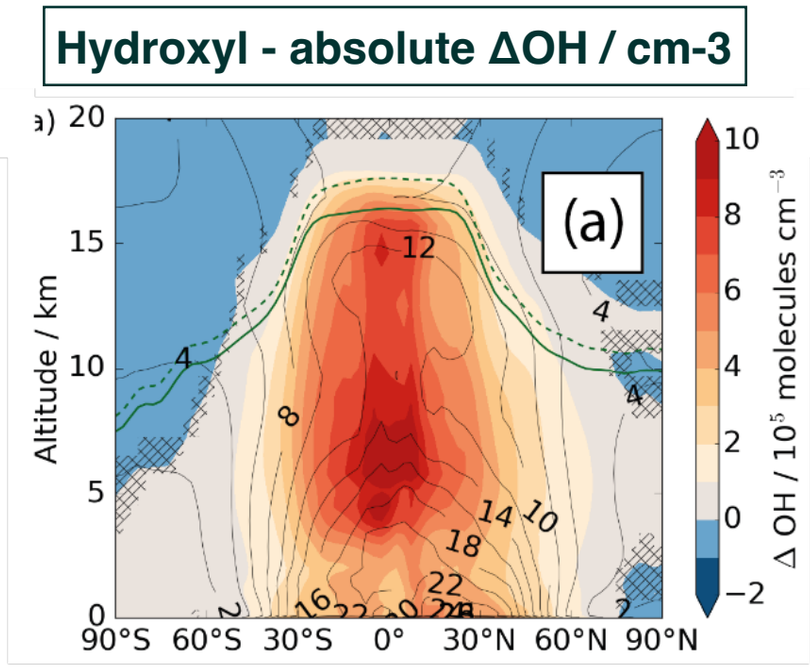


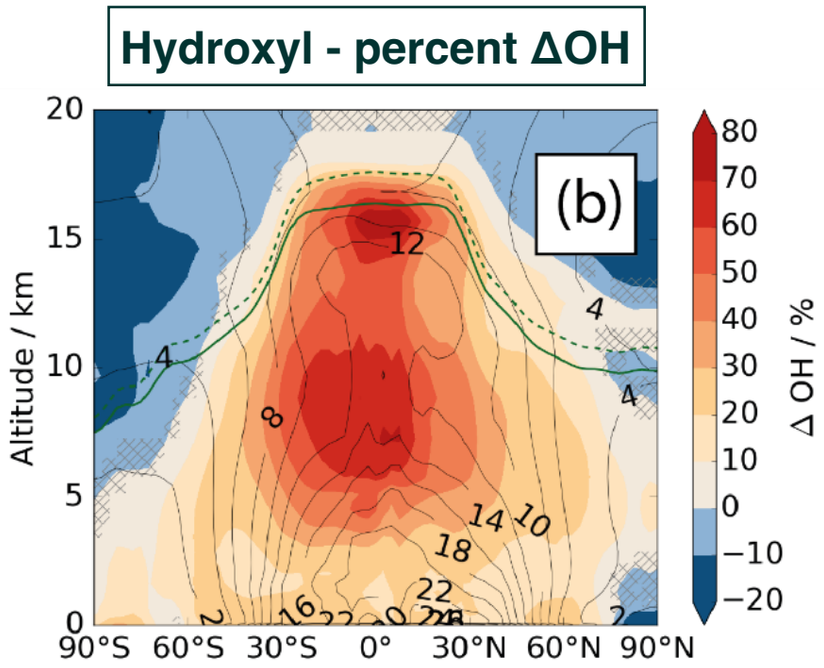
Water vapour

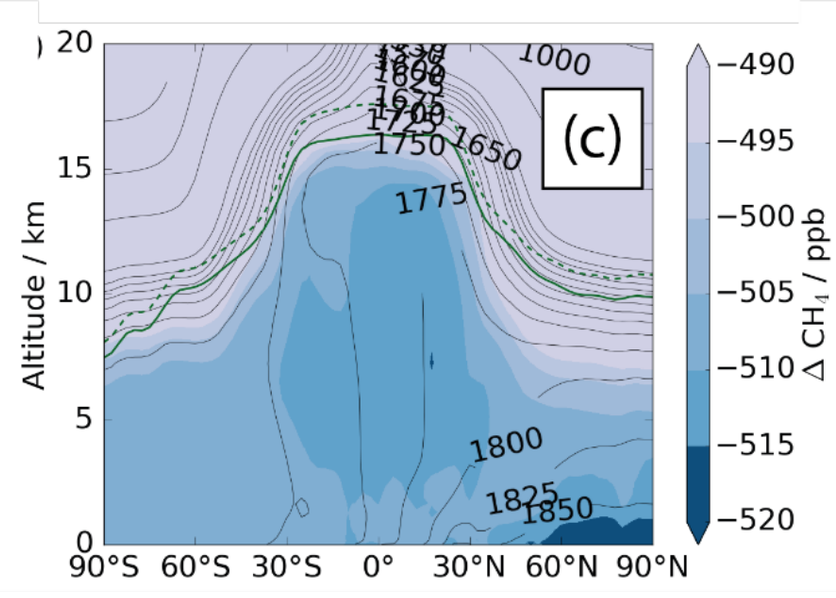
What happens to tropospheric oxidising capacity in future climate?

- 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.
- O1D+H2O drives increase, contributions from HO2+O3?

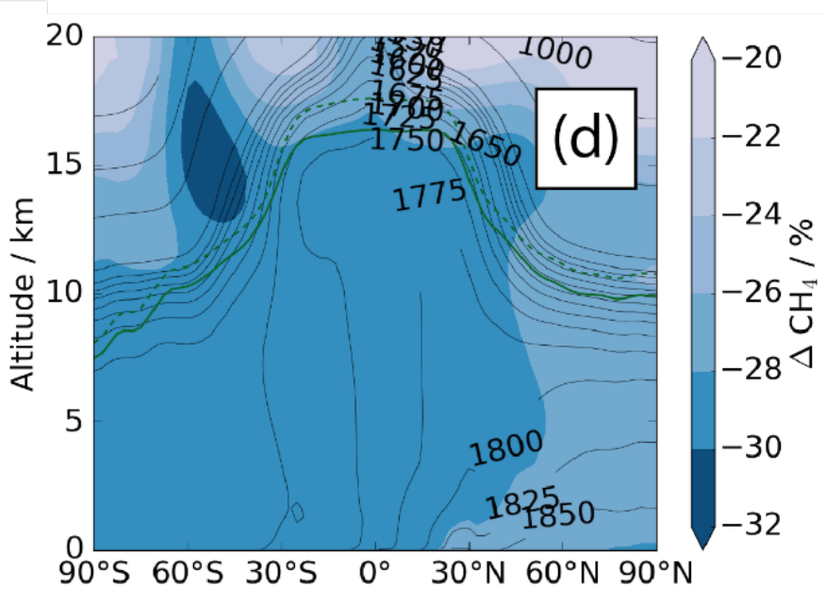
ΔCC with respect to year 2000







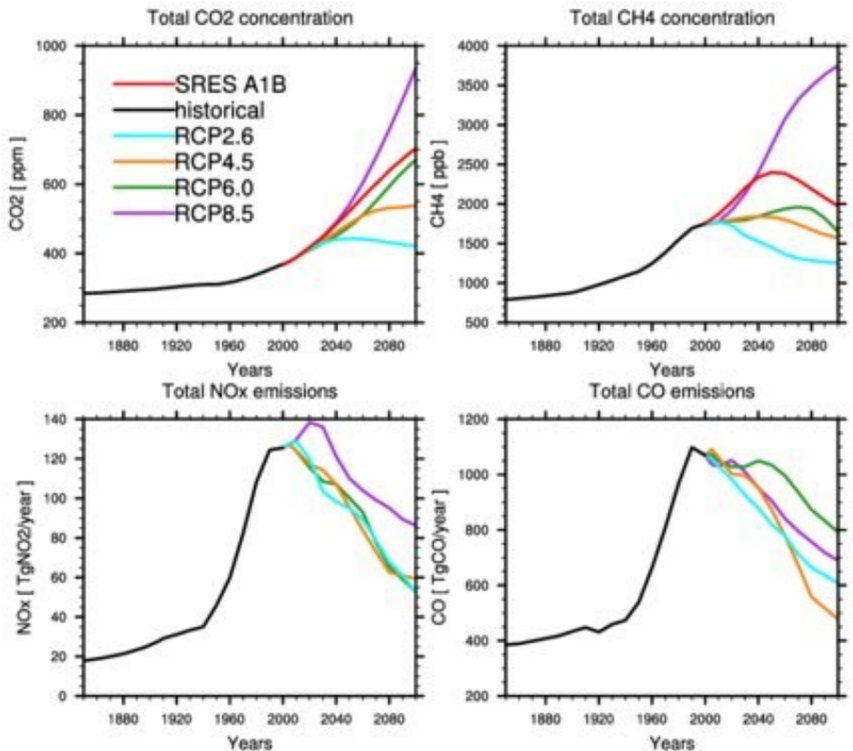
Methane - absolute ΔCH4 / ppb



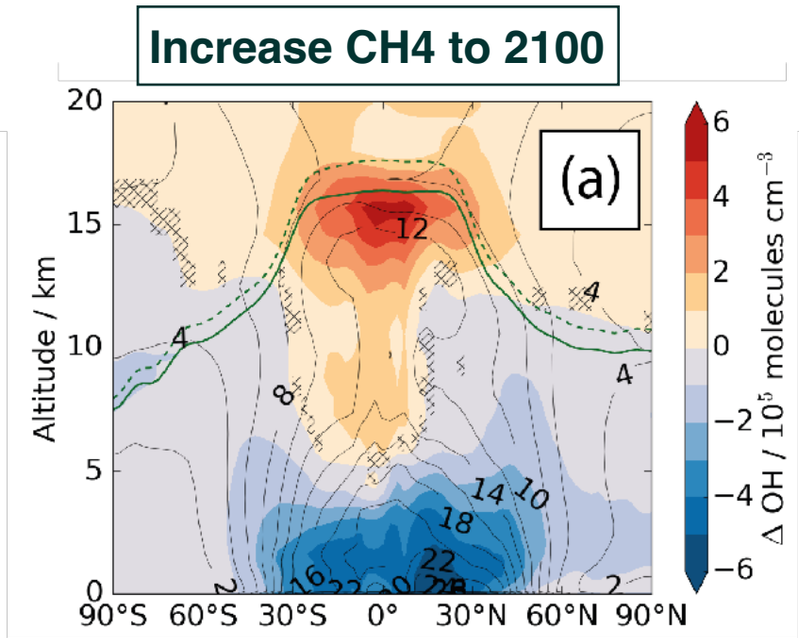
Methane - percent ΔCH4

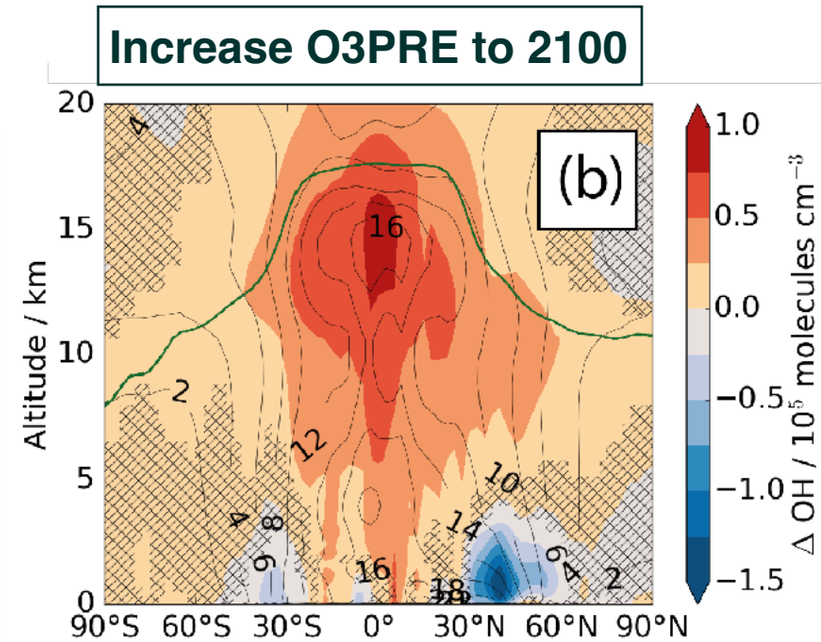
What happens to tropospheric oxidising capacity in future climate?

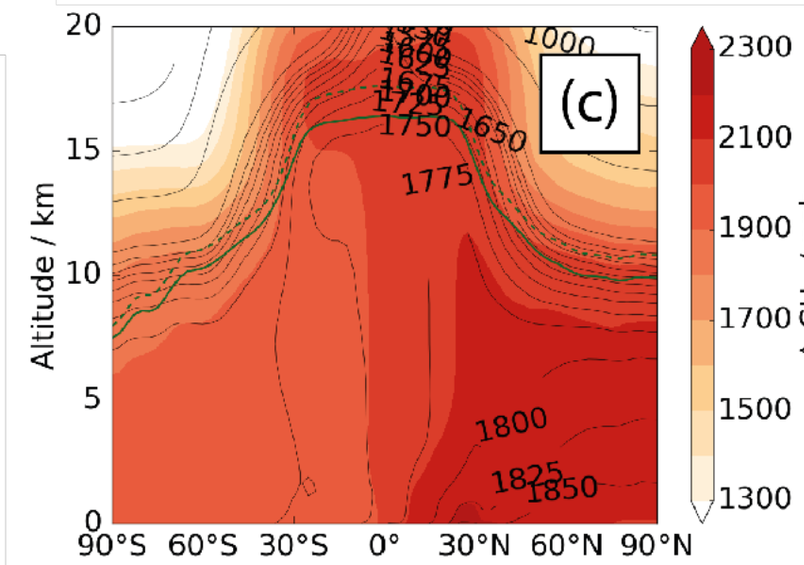
- Increasing CH_4 emissions to RCP8.5 levels gives
  - Large increase in CH_4
  - Large decrease in OH
- Increasing CO and NOx to RCP8.5 levels gives
  - Smaller change in OH
  - Small decreases in CH_4

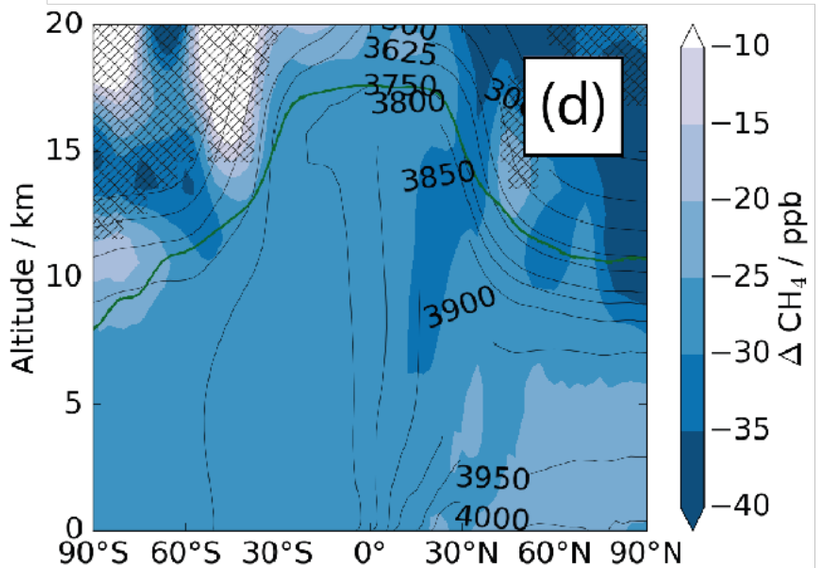


Hydroxyl









Methane

Conclusions 2/4 - CH4 in future climate

- AerChemMIP histSST experiments provide idealised experiments
- Can inter-compare model responses to same idealised forcing changes
- ODS depletion caused a decrease in tropospheric ozone
- 1850-2015 increase in methane levels caused an ~40 Tg increase in O3 burden
  - Model sensitivity to this change is different: 40% to 80% (!)
- Climate change leads to higher temp and humidity
  - Increased OH production higher levels of OH - shorter methane lifetime, reduced GWP.
  - Increased methane offsets this - OH levels suppressed by methane
- What are the co-benefits to mitigating methane emissions?

AGU100 ADVANCING EARTH AND SPACE SCIENCE

Reviews of Geophysics

REVIEW ARTICLE
10.1029/2019RG000675

Key Points:
- The atmospheric methane burden is rising fast; this growth is an increasing threat to the Paris Agreement of the UN Framework Convention on Climate Change (UNFCCC)

Methane Mitigation: Methods to Reduce Emissions, on the Path to the Paris Agreement

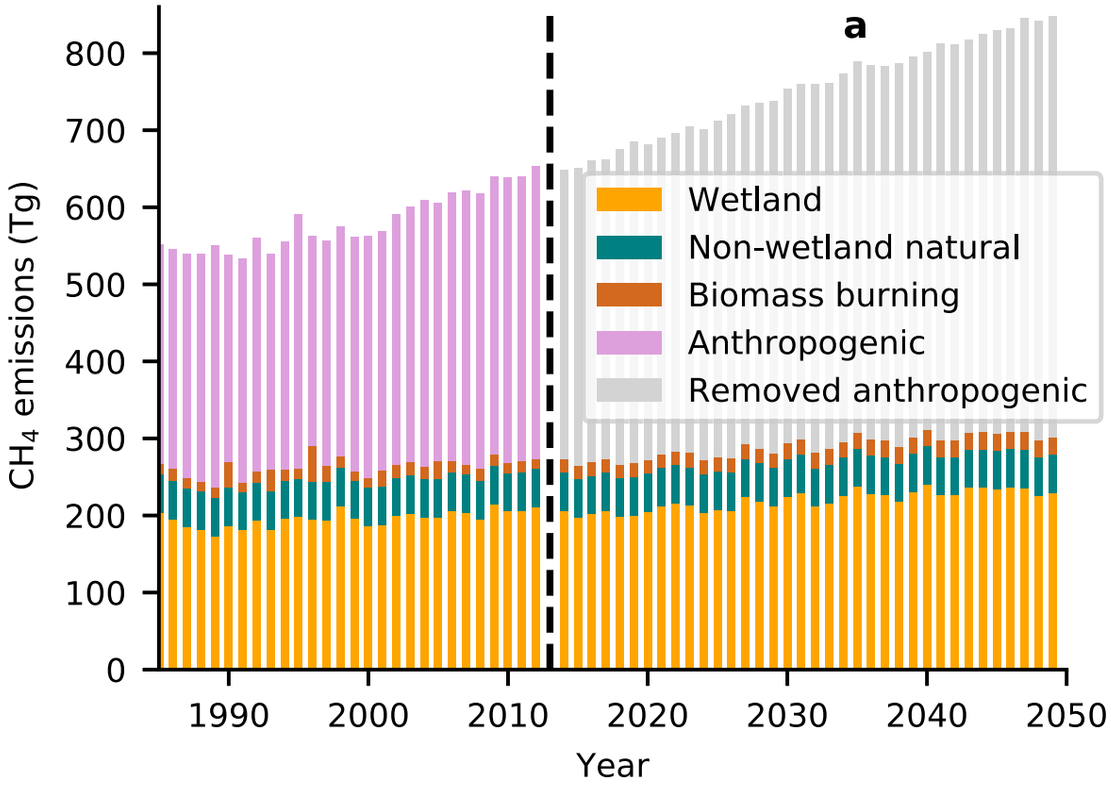
E. G. Nisbet[1], R. E. Fisher[1], D. Lowry[1], J. L. France[1], G. Allen[2], S. Bakkaloglu[1], T. J. Broderick[3], M. Cain[4], M. Coleman[5], J. Fernandez[1], G. Forster[6], P. T. Griffiths[7], C. P. Iverach[8], B. F. J. Kelly[8], M. R. Manning[9], P. B. R. Nisbet-Jones[1], J. A. Pyle[7], A. Townsend-Small[10], A. al-Shalaan[1], N. Warwick[7], and G. Zazzeri[11]

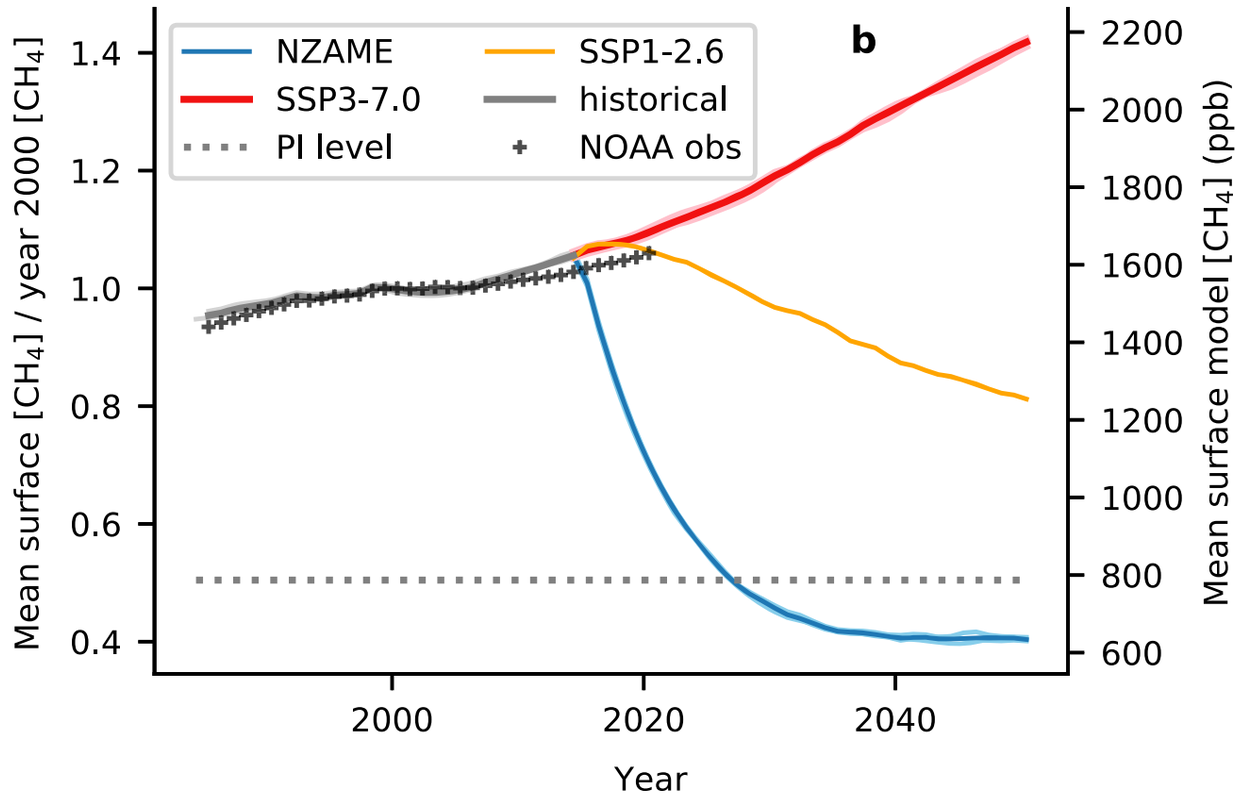
Coupled atmosphere-ocean studies of the role of methane in future climate

National Centre for Atmospheric Science
NATURAL ENVIRONMENT RESEARCH COUNCIL

Methane emissions in a fully coupled atmosphere-ocean model

- 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



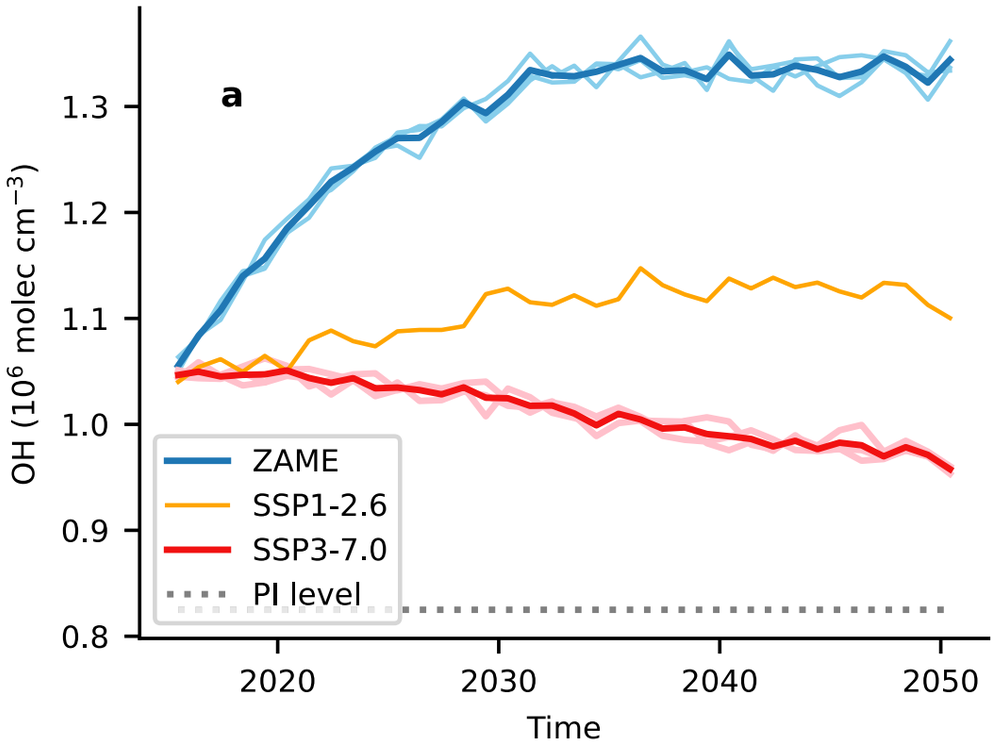


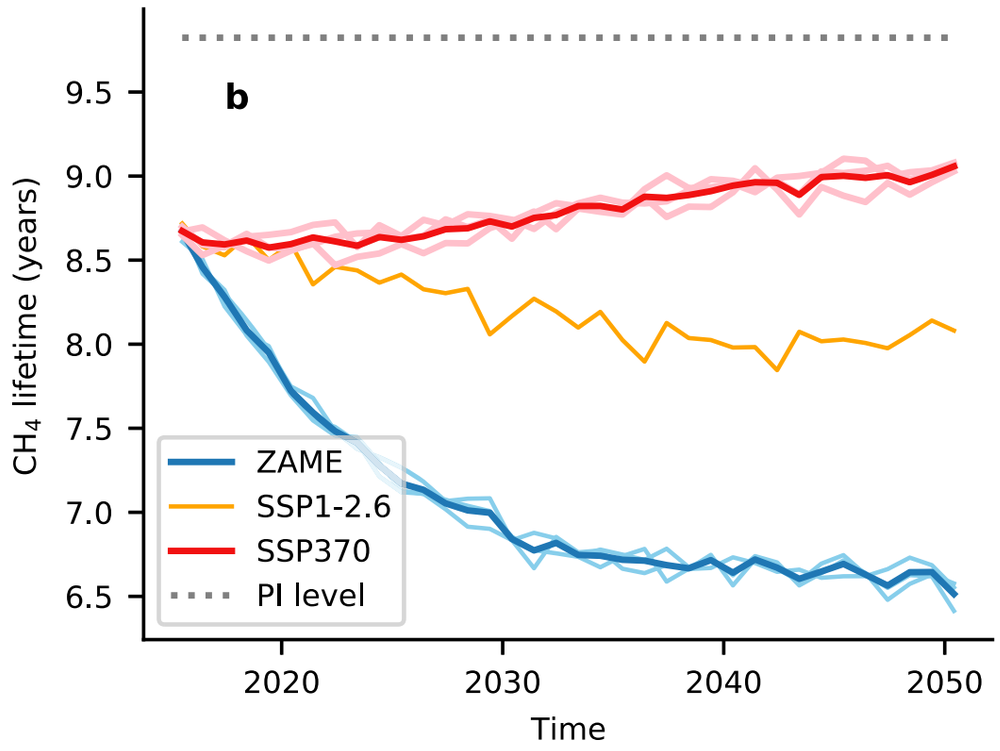
- Comparison with SSP3-7.0 and SSP1-2.6 allows them to function as a counterfactual
  - What are the risks of methane emissions?
  - What are the benefits of constraining future methane emissions?

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



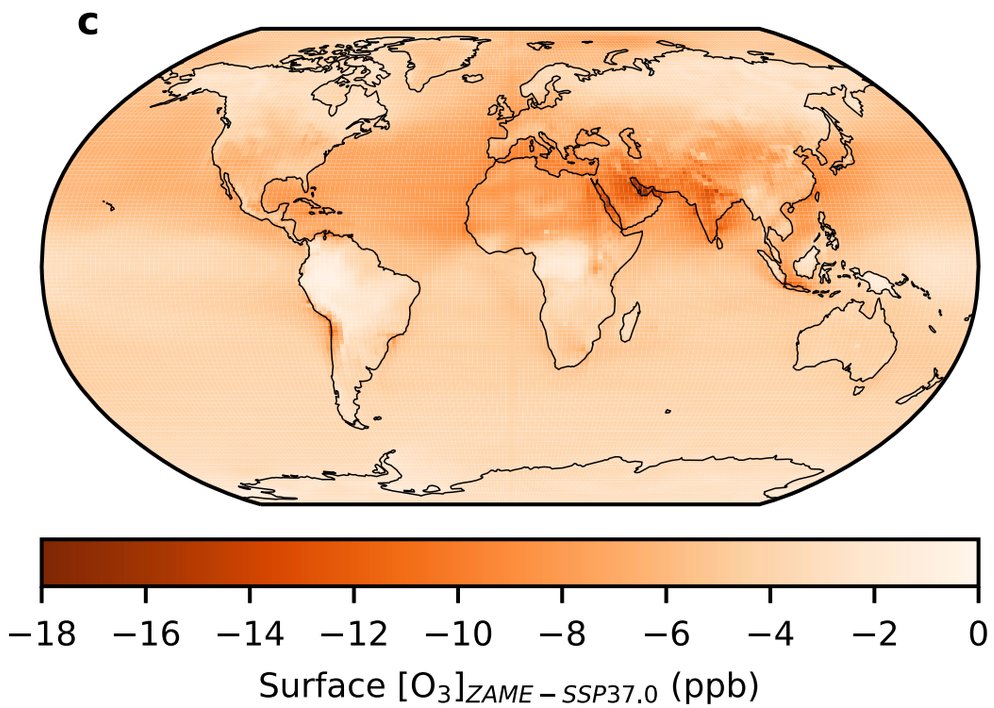


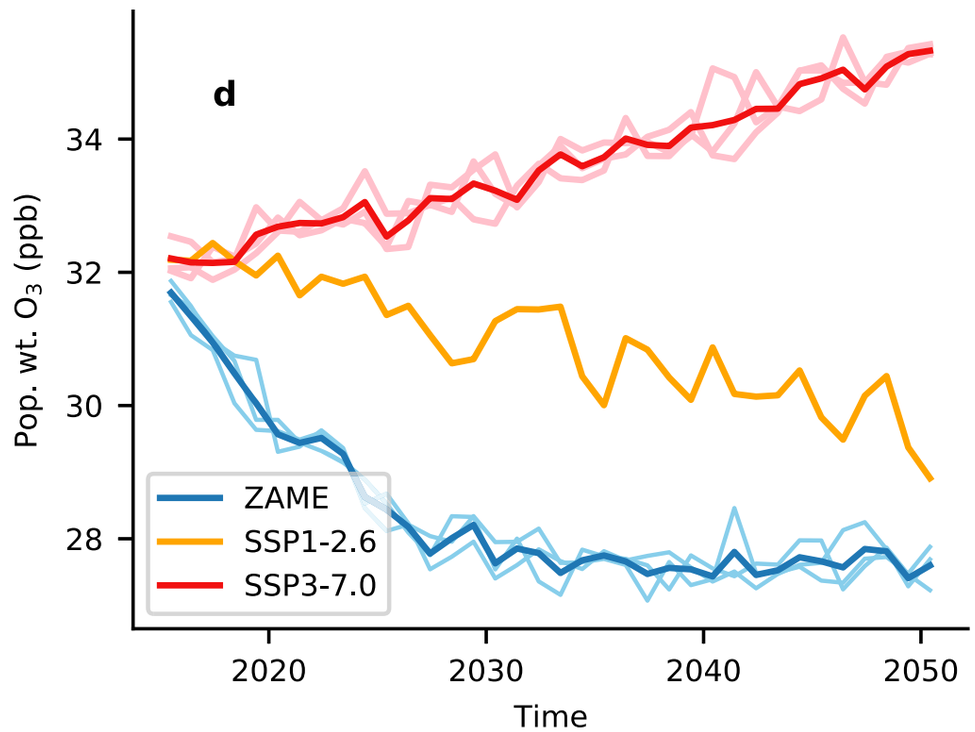
- Comparison with SSP3-7.0 and SSP1-2.6 allows them to function as a counterfactual
  - What are the risks of methane emissions?
  - What are the benefits of constraining future methane emissions?

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



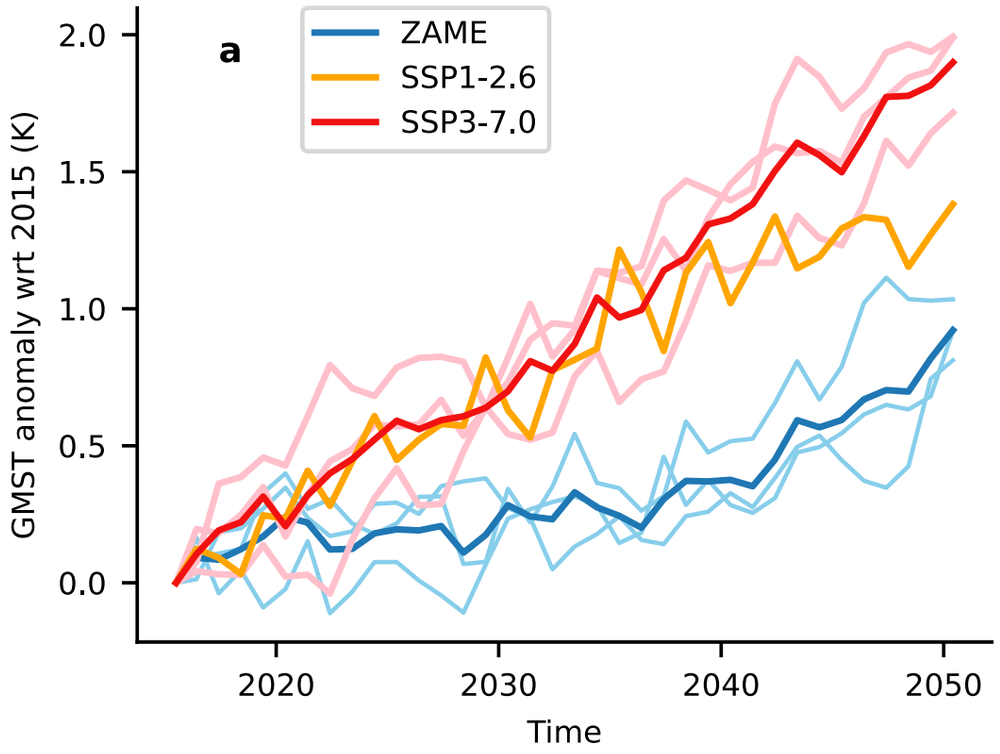


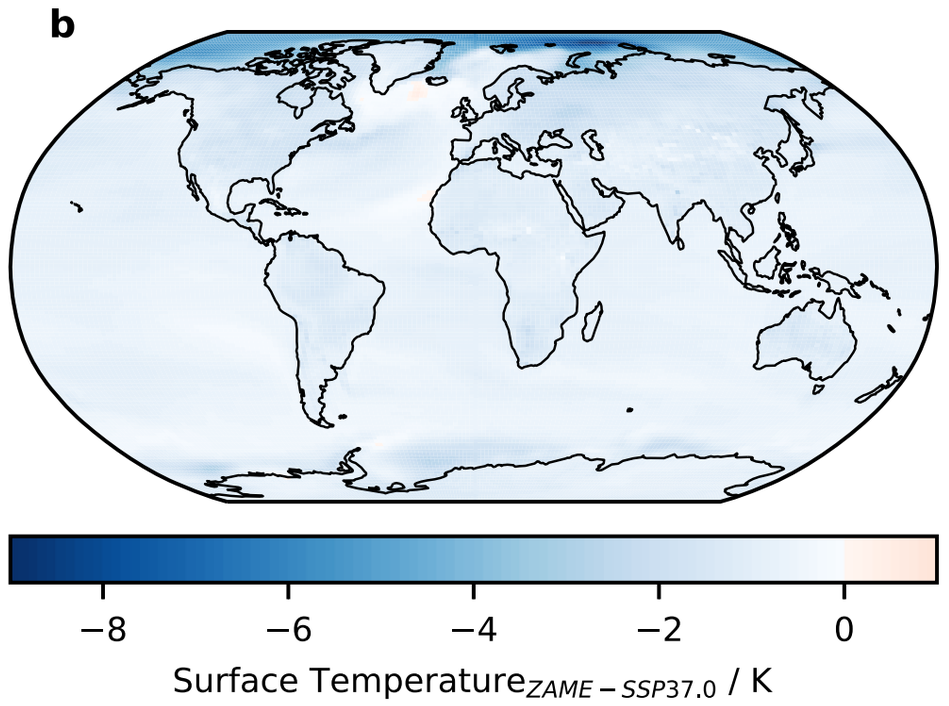
- Weighting the ozone field by human exposure shows ~10% decline
- Projected decrease in AQ-related mortality of the order of 500k per year

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 → $\Delta T = 0.5$ K
- Decline across the globe, strong regional variations, Arctic amplification





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ARTICLE OPEN

Check for updates

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

Zosia Staniaszek[1]✉, Paul T. Griffiths[1,2], Gerd A. Folberth[3], Fiona M. O'Connor[3], N. Luke Abraham[1,2] and Alexander T. Archibald[1,2]✉

Figures by Zosia Staniaszek

Conclusions 2/4- 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.

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www.nature.com/npjclimatsci

ARTICLE OPEN

Check for updates

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

Zosia Staniaszek[1]✉, Paul T. Griffiths[1,2], Gerd A. Folberth[3], Fiona M. O'Connor[3], N. Luke Abraham[1,2] and Alexander T. Archibald[1,2]✉

The role of oxidant in radiative forcing - replacing CH4 with H2 as a fuel source

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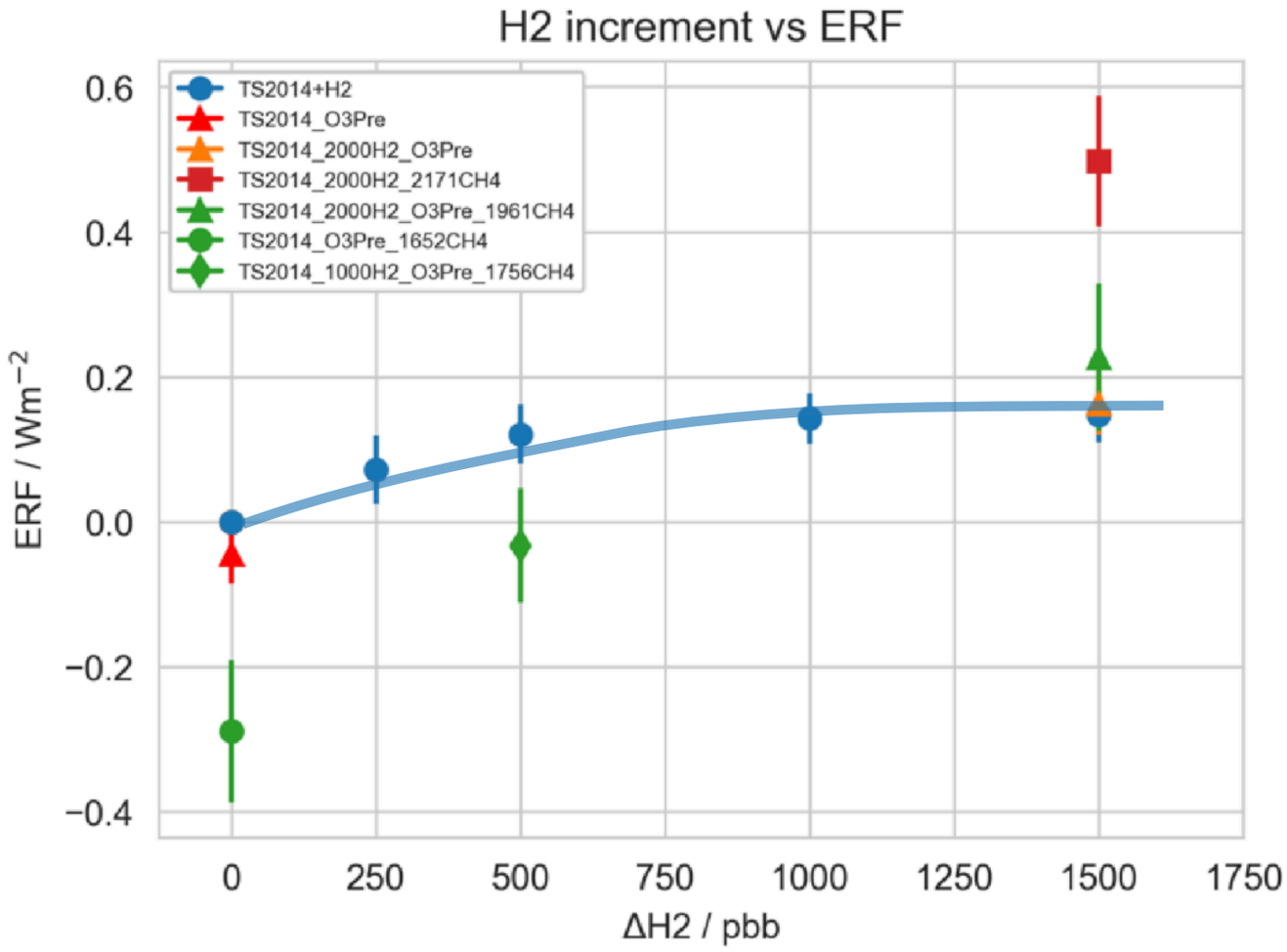
NATURAL ENVIRONMENT RESEARCH COUNCIL

Climate effects of oxidant changes - 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 ± 0.08 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	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

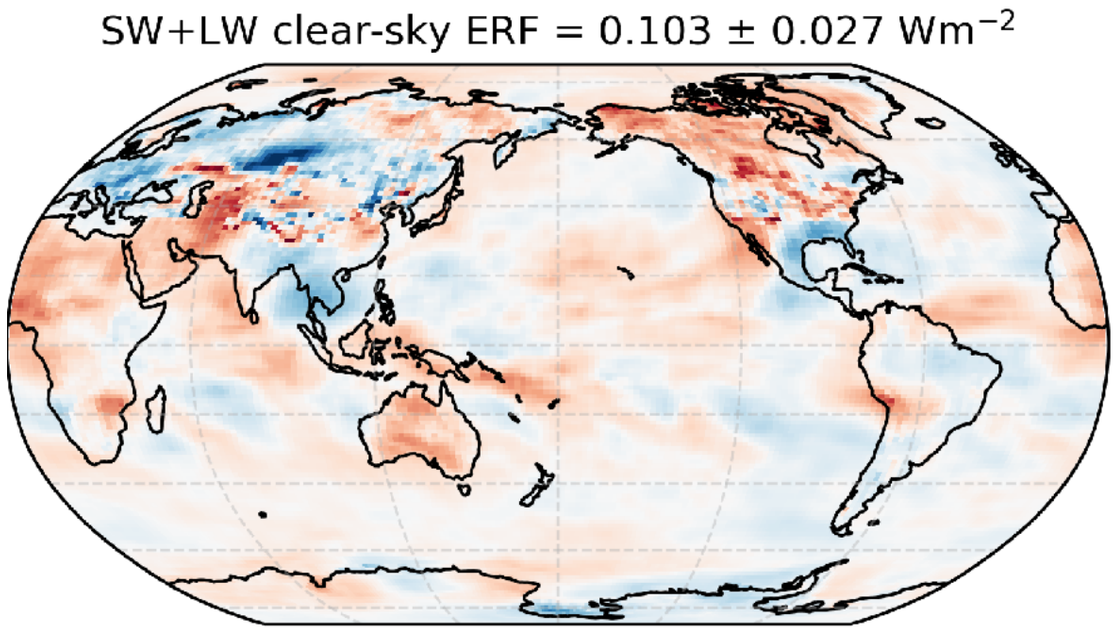
Breaking ERF down into clear-sky and cloud effects

- 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)

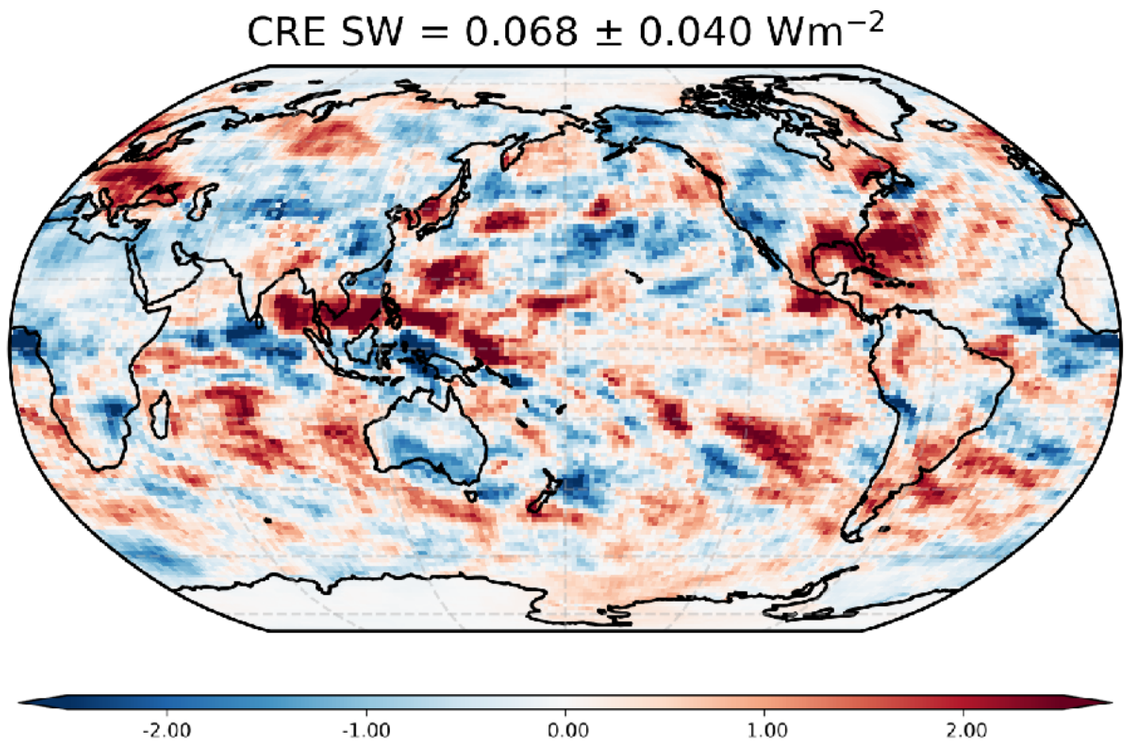
  - ΔCRE = 0.036 Wm^{-2}

- Which can be broken down further

  - Shortwave ΔCRE = 0.068 Wm^{-2}

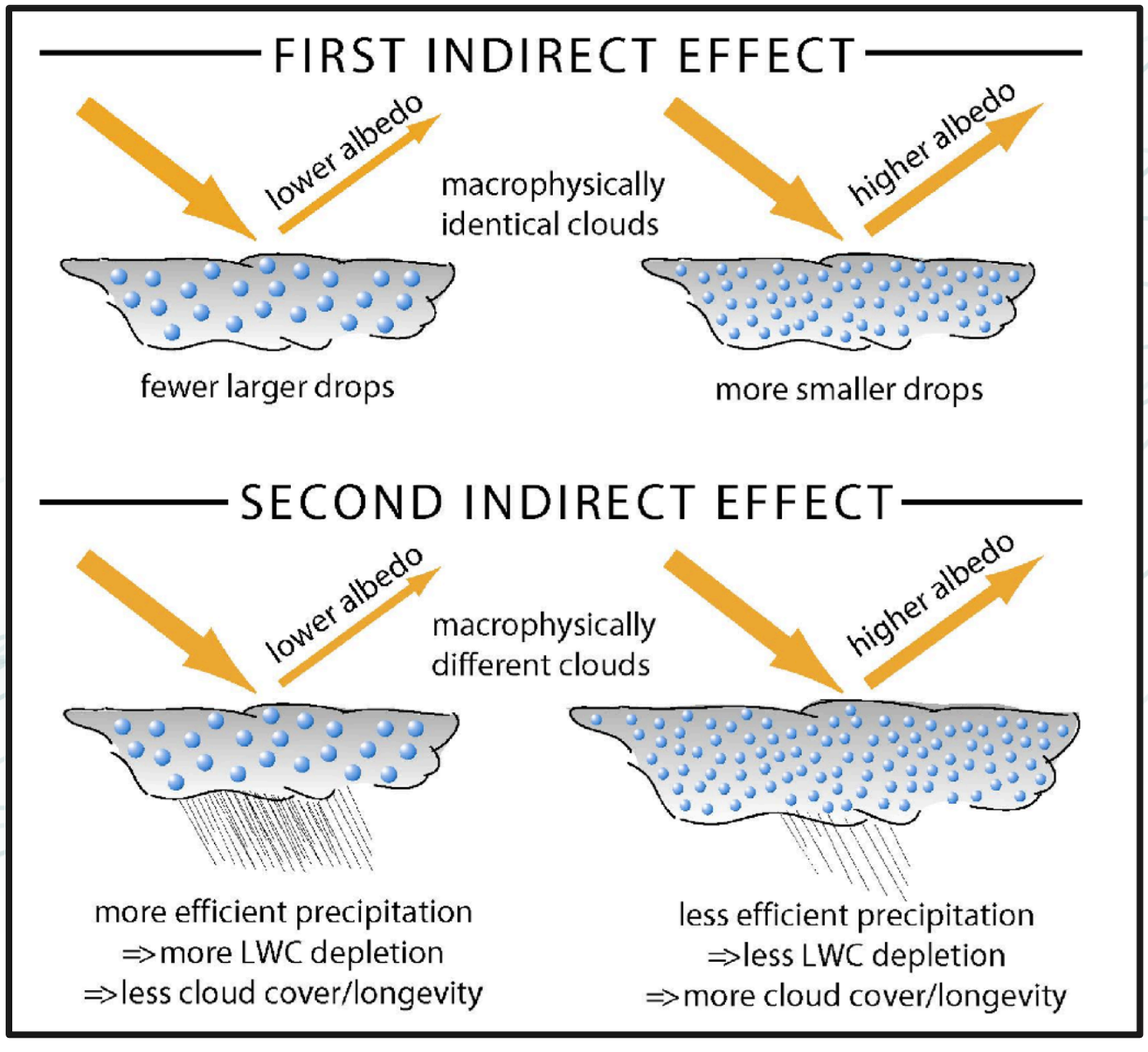
  - Longwave ΔCRE = -0.032 Wm^{-2}

- 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



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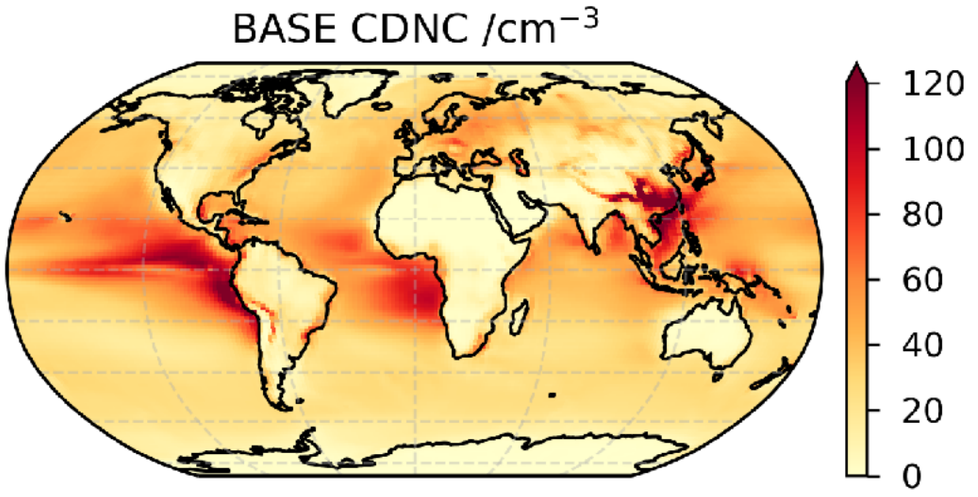
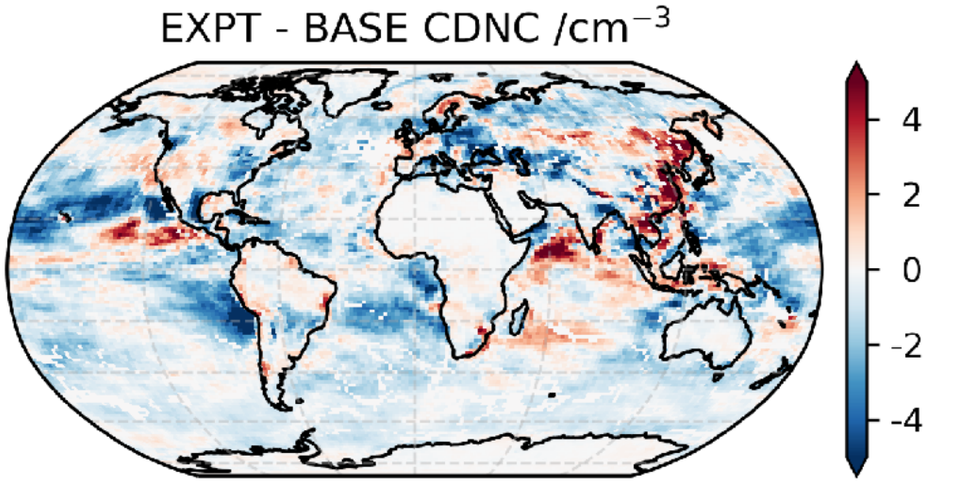
ERF - the coupling of gas phase oxidant to aerosol levels and cloud properties

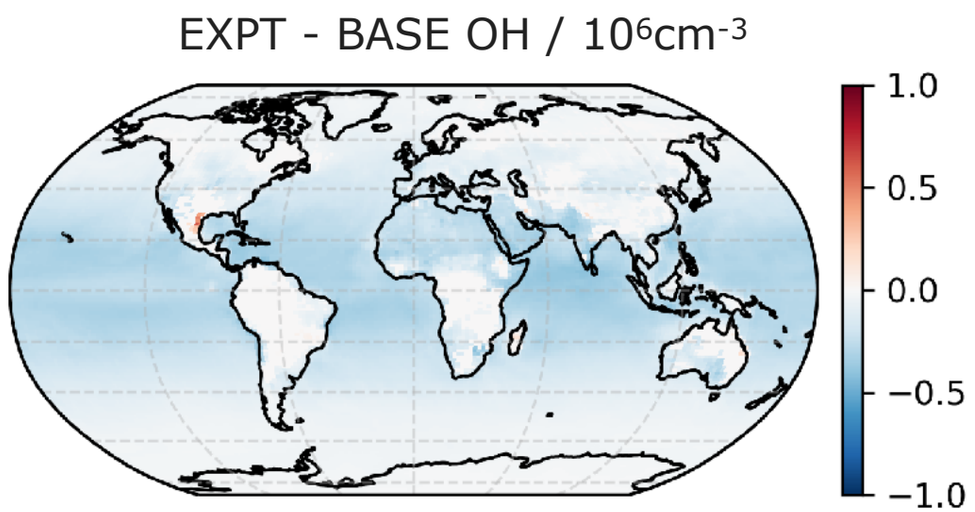
- The additional H2 has caused a decrease in cloud droplet number concentration (CDNC). Seen here as a decrease in cloud droplet number with respect to our low H2 base case.

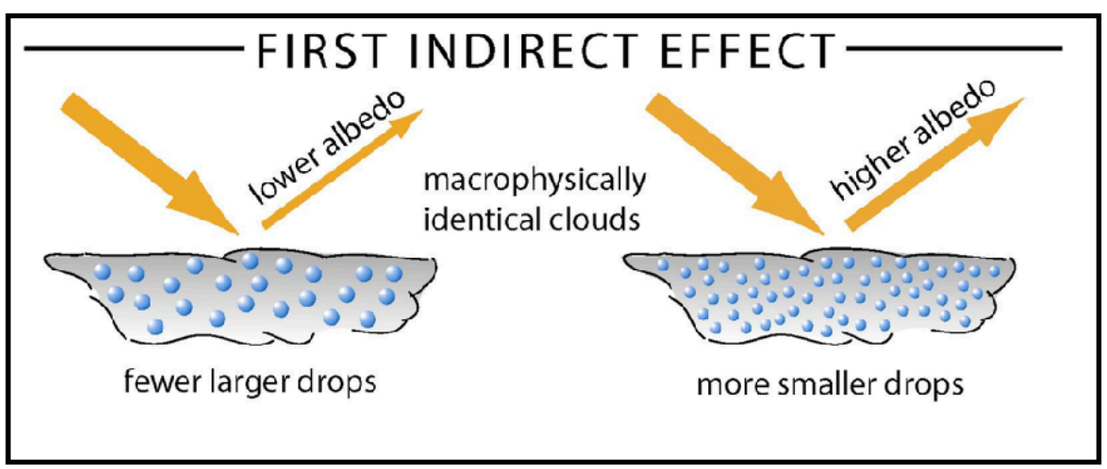
- We can associate this decrease with the lower levels of the OH free radical oxidant in the region where aerosol is formed. There are fewer aerosol particles as a result.

- 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 3/4 - oxidant and RF

- Ozone is itself a greenhouse gas - approx. 0.3 Wm^{-2} of forcing
- Oxidant is also important - couples e.g. CO, NOx emissions into ozone RF
- Secondary aerosol is also important, both direct (scattering/absorption) and indirect (cloud albedo/lifetime) depend on oxidant levels.
- Emissions of H2 produce two effects
  - Increase levels of ozone via HO2+NO → NO2 → → O3
  - Changes aerosol size and number distribution, e.g sulfate aerosol
  - More H2 → less OH → less aerosol nucleation → decreased cloud albedo
- Both of these function as a warming
- Impact depends on ‘fugitive’ emissions - i.e. leaks prior to use.
- High leakage rates can have negative consequences which may offset lower CH4 and CO2 emissions (But the debate goes on).

The role of the stratosphere on tropospheric ozone

National Centre for Atmospheric Science
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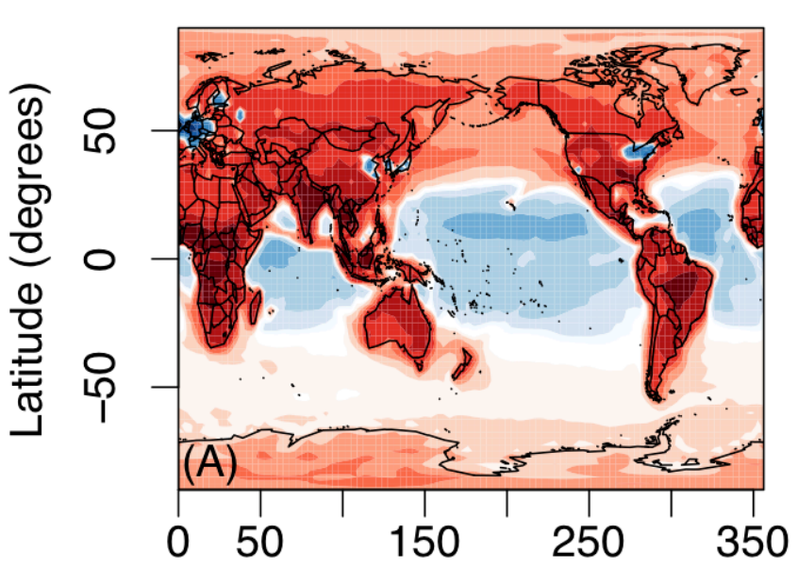
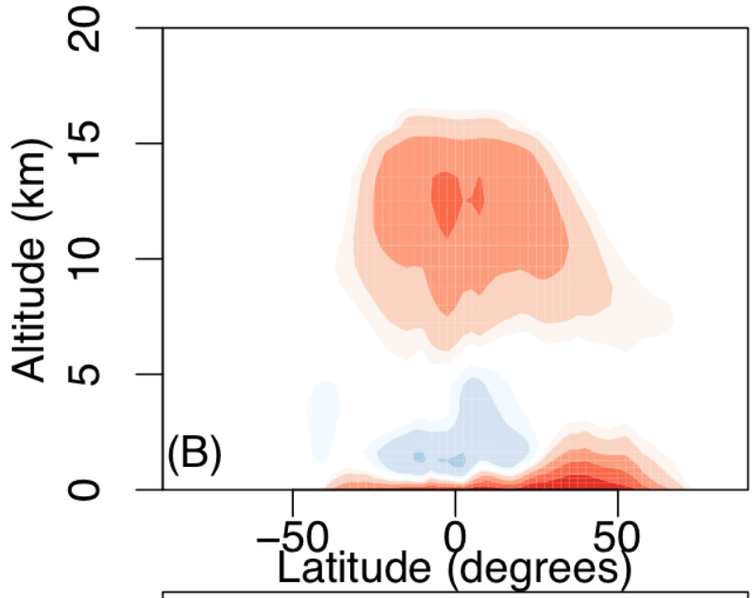
Multimodel ozone tendency - TOAR Budget

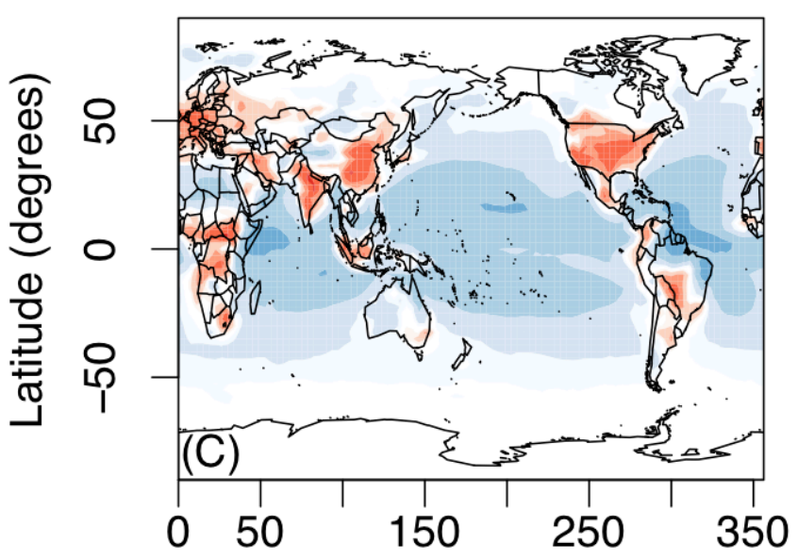
Surface

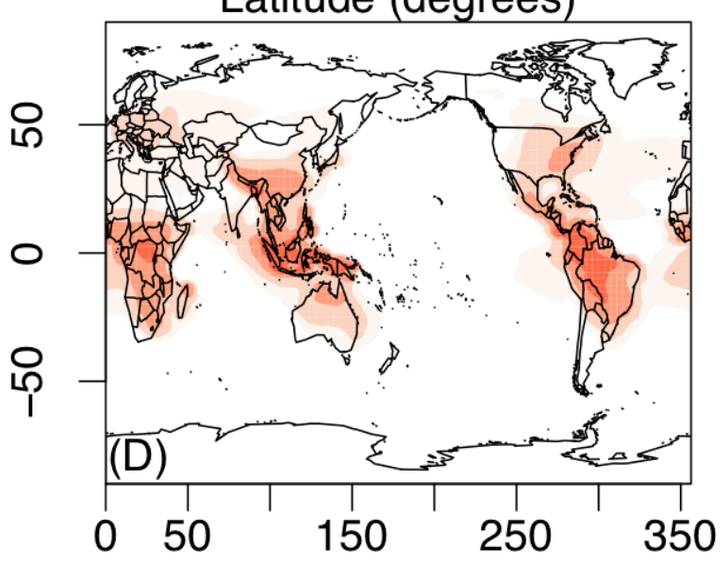




ZM

FT





UT

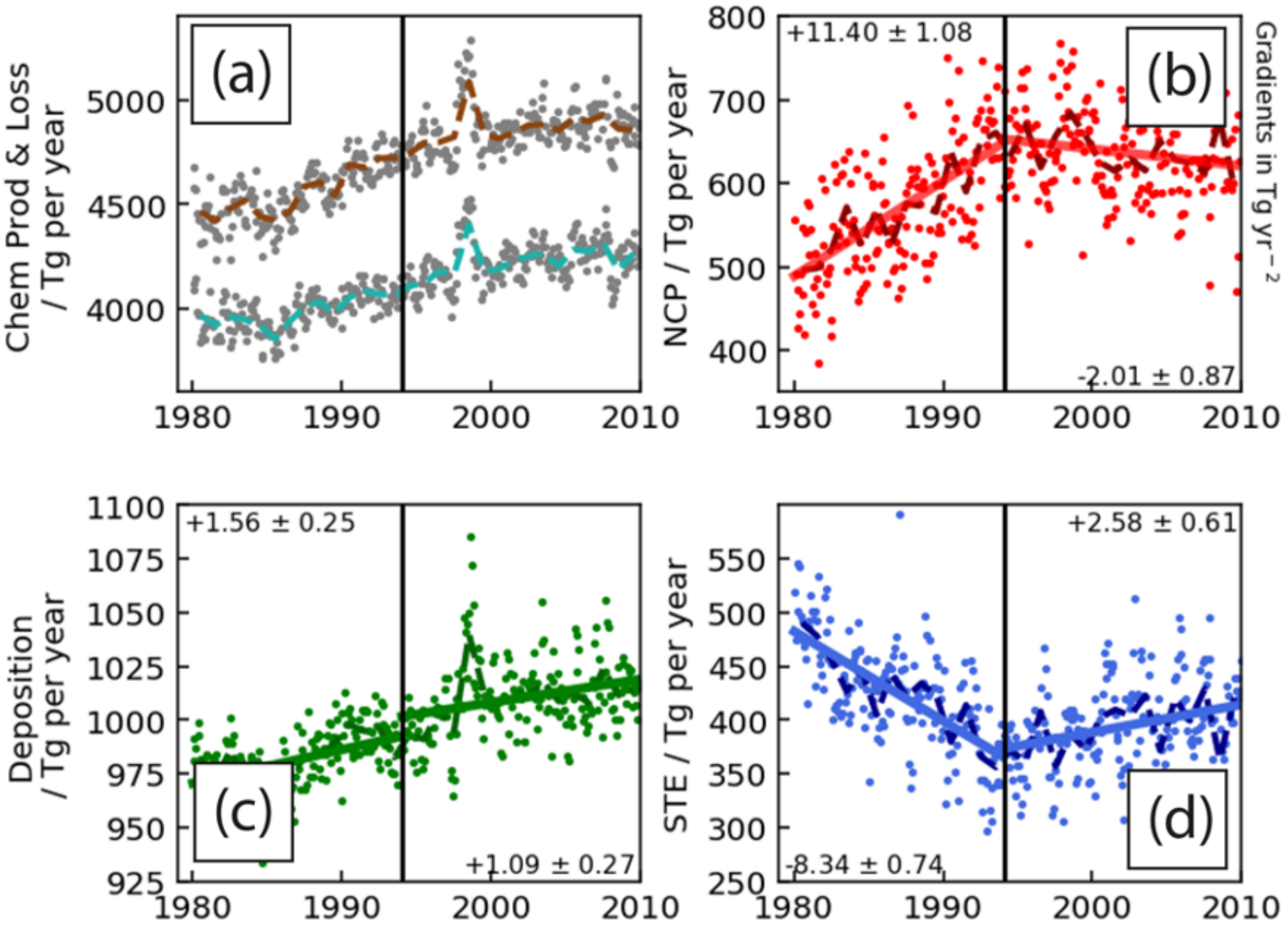
Longitude (degrees)

ΔO_3 ppb/hr

−5 −1 −0.5 −0.2 −0.1 −0.05 −0.02 −0.01 0.01 0.02 0.05 0.1 0.2 0.5 1 5

Archibald et al., TOAR “Budget”, Elementa 2021

Tropospheric ozone budget in CCMs - large, opposing terms



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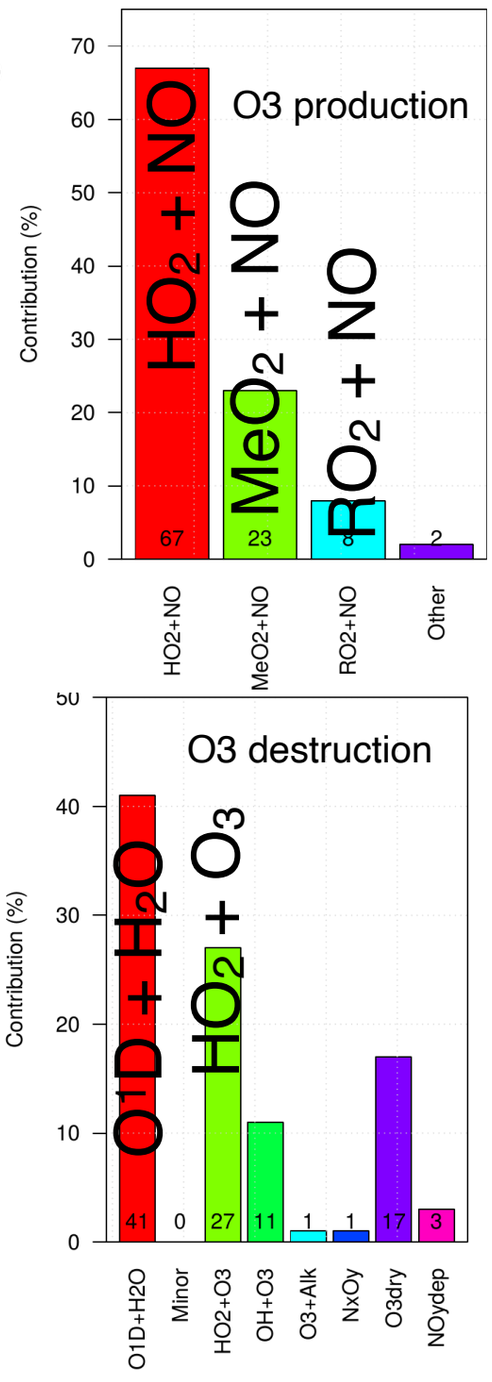


Geophysical Research Letters

Research Letter | Open Access

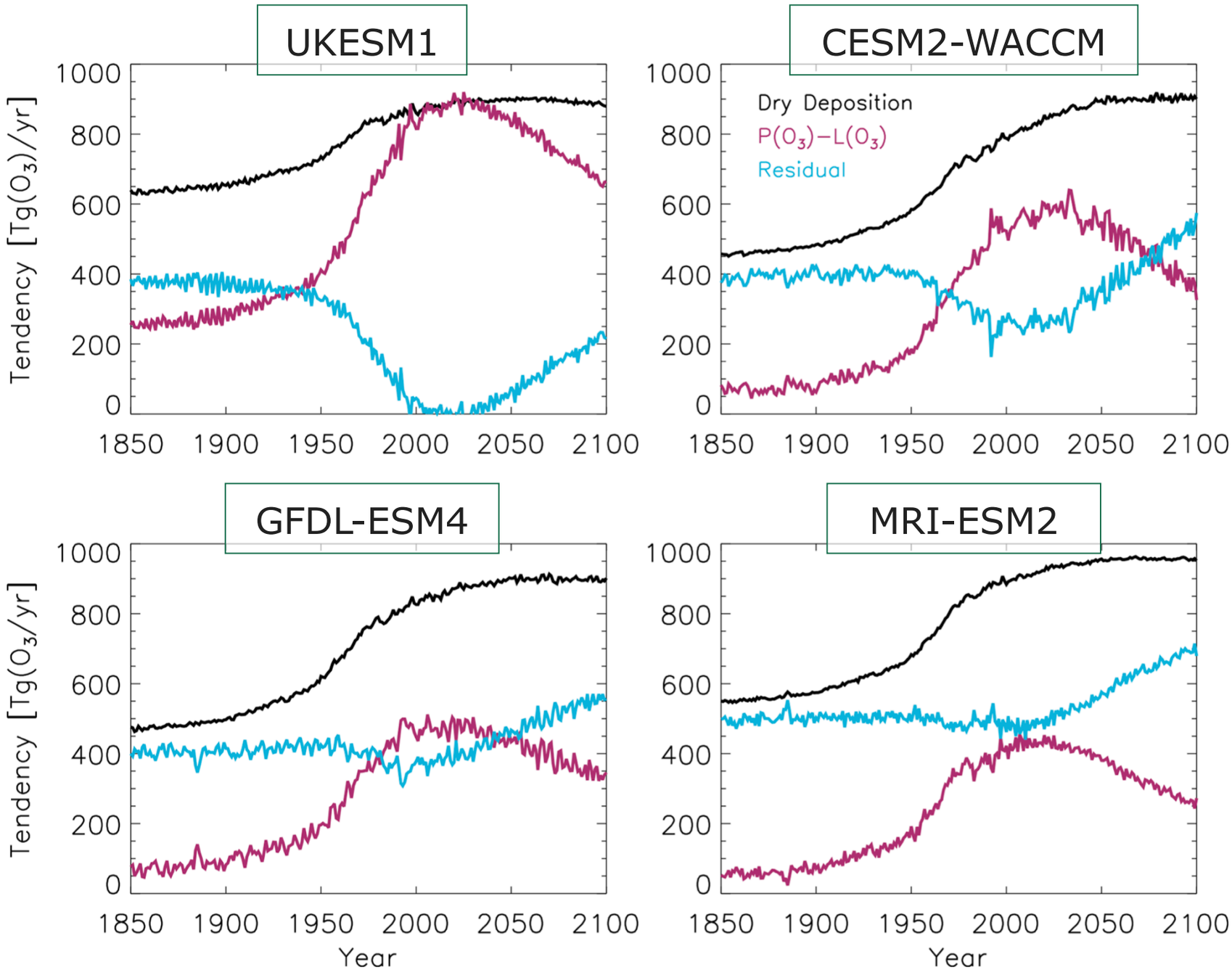
On the Changing Role of the Stratosphere on the Tropospheric Ozone Budget: 1979–2010

P. T. Griffiths, J. Keeble, Y. M. Shin, N. L. Abraham, A. T. Archibald, J. A. Pyle

Inferred STE in CMIP6 models varies widely

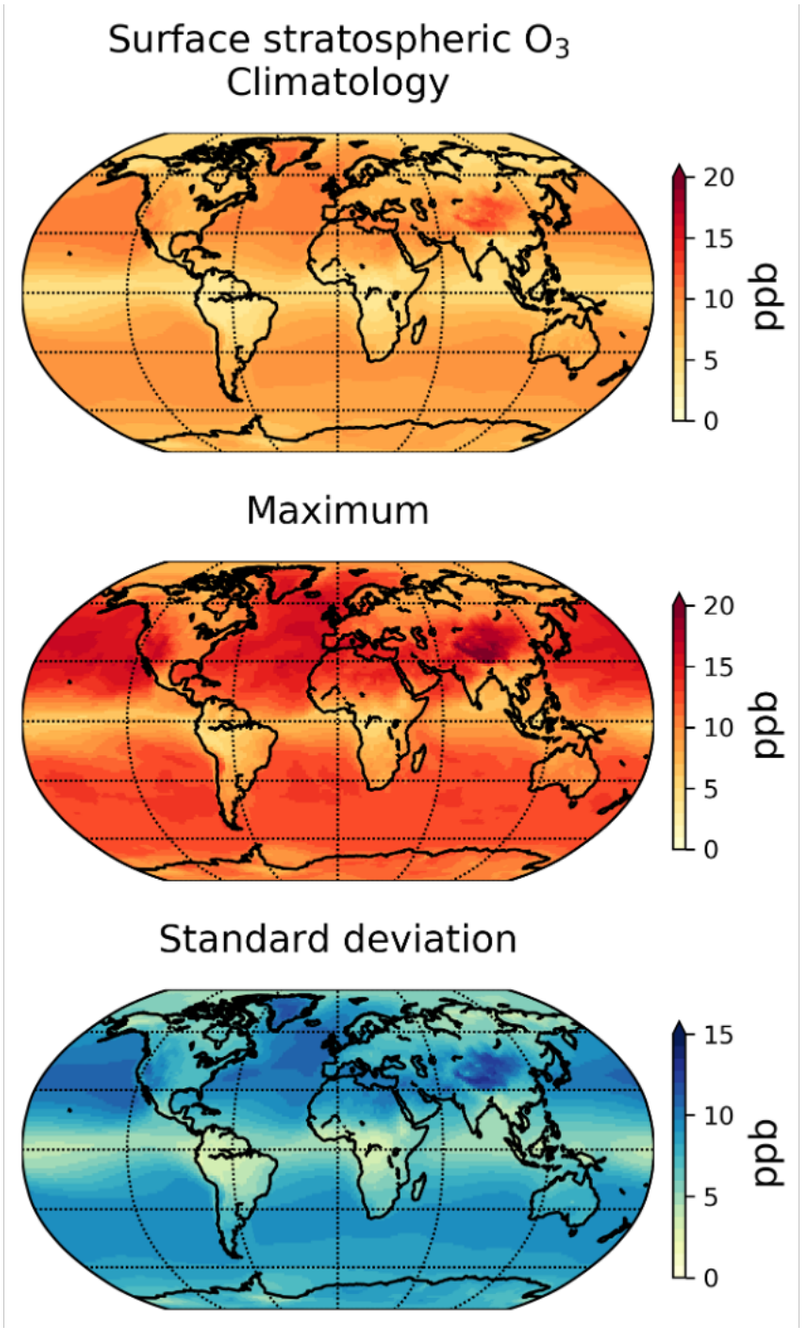
- For a closed Ozone budget, in-situ production and downward transport from the stratosphere are balanced by in-situ destruction and chemical loss, ie P+S=D+L

- From which Sinf = Deposition - (Production - Loss) = 1000 Tg/ yr - 500 Tg/ yr = 500 Tg/ yr



TOAR-II ROSTEES project

- James Keeble and I are now leading a IGAC TOAR-II endorsed project “The role of the stratosphere in the Earth system”
- Review the role of stratospheric ozone recovery in controlling future ozone levels, due 2024.
- Improved estimates of strat-trop transport of ozone in chemistry-climate models using CCMI2022 data.
- For more stratospheric ozone work see Pyle et al. 2022



Article | Published: 24 August 2022

Integrated ozone depletion as a metric for ozone recovery

John A. Pyle, James Keeble, Nathan Luke Abraham, Martyn P. Chipperfield & Paul T. Griffiths

Nature **608**, 719–723 (2022) | Cite this article

Conclusions 4/4 - the role of the stratosphere

- Ozone is produced and destroyed in large amounts in the troposphere, these reactions buffer each other [Wild & Palmer 2008]
- Deposition at the surface and downward transport from the stratosphere close the budget
- Stratospheric ozone depletion produced a significant change in the tropospheric ozone budget and oxidant [Murray et al. 2022]
- Stratospheric ozone recovery will change the budget again -
  - Less photolysis as UV levels decrease [e.g. Zhang et al. 2014]
  - Increased ozone as stratospheric ozone recovers and downward transport increases - particularly in SH where STE has largely shut down [Ruiz & Prather 2022].
  - Increased Brewer-Dobson circulation? [Zanis et al., 2021]
- Impacts on air quality are important
  - EPA routinely considers stratospheric intrusions in its assessments

Summary - tropospheric ozone in CMIP6

- Ozone is buffered - produced and destroyed in large amounts in the troposphere and these respond similarly to emissions changes.
- Climate change drives significant changes in chemistry
- Assessment is a challenge - O3 and STE best constraints
- Increasing complexity of ESMs makes assessment harder and more important to understanding multi-model differences - CMIP7?
- Fewer models taking part - need a strategy to increase model participation and e.g. CTM involvement for greater process-level diversity
- Understanding model diversity requires a good quantification of
  - Stratospheric ozone
  - Methane
  - Oxidant-aerosol coupling

Thank you

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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

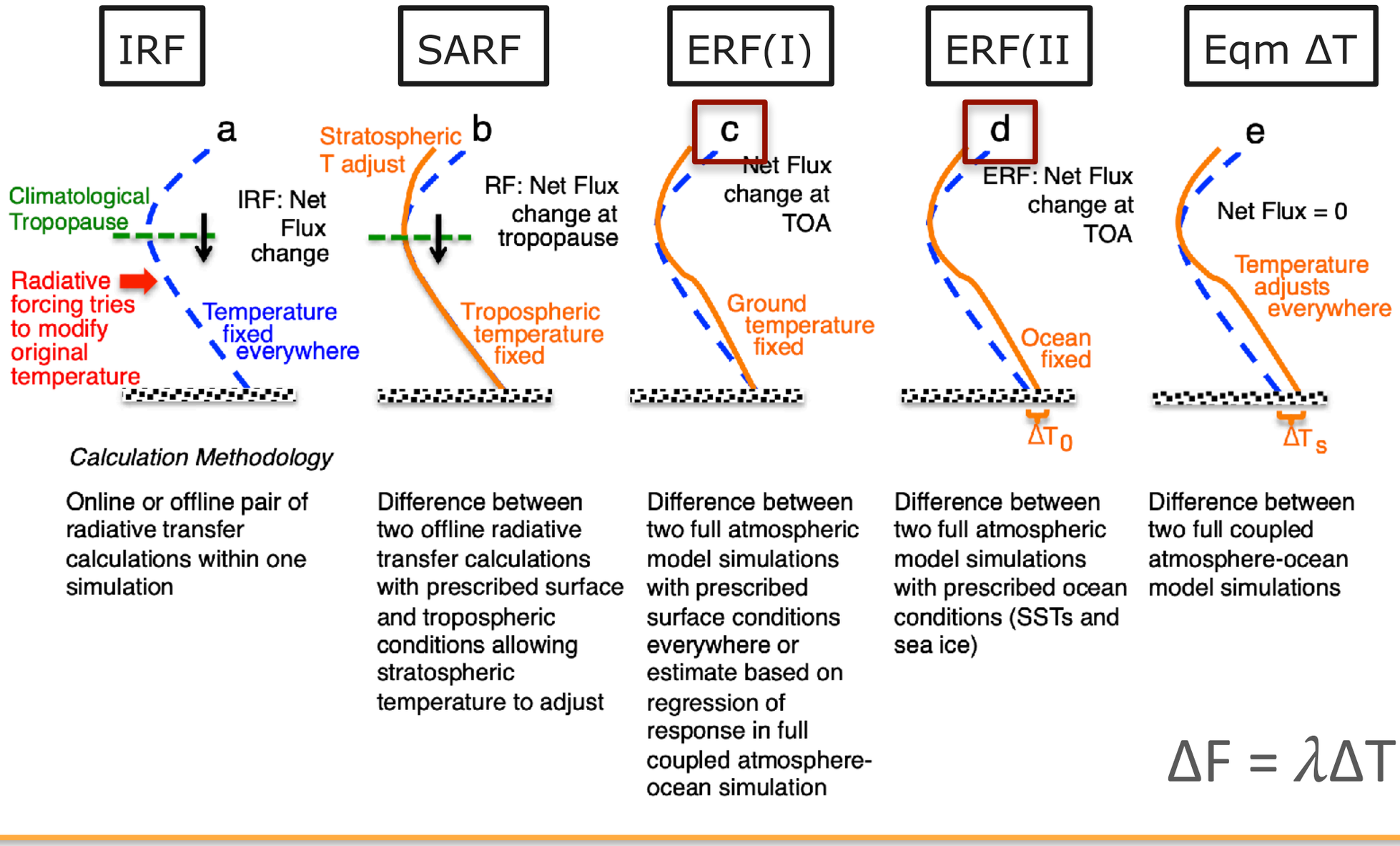
[a]Includes export to stratosphere of 1.9 Tg H_2 yr^{-1}.

[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).

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

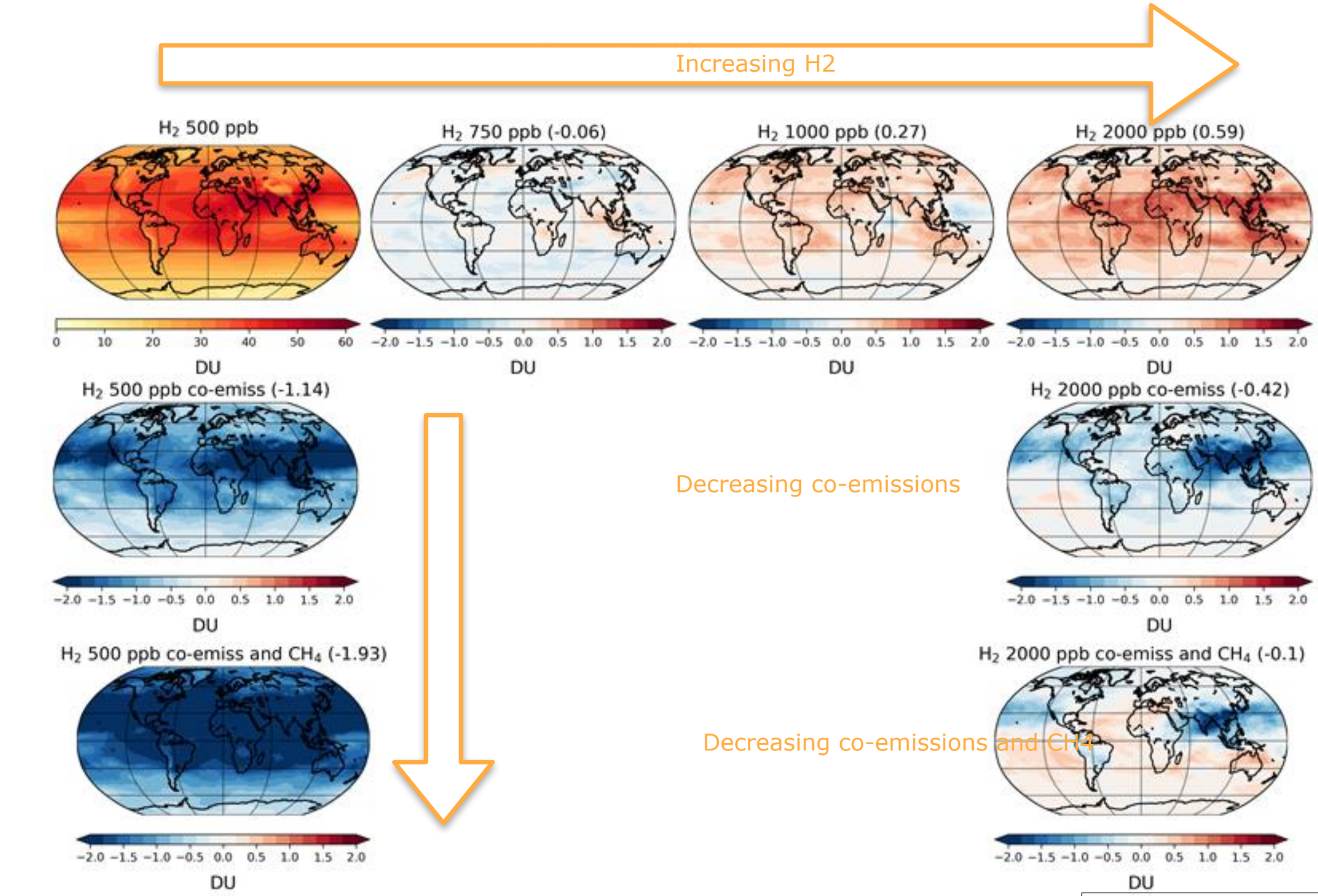
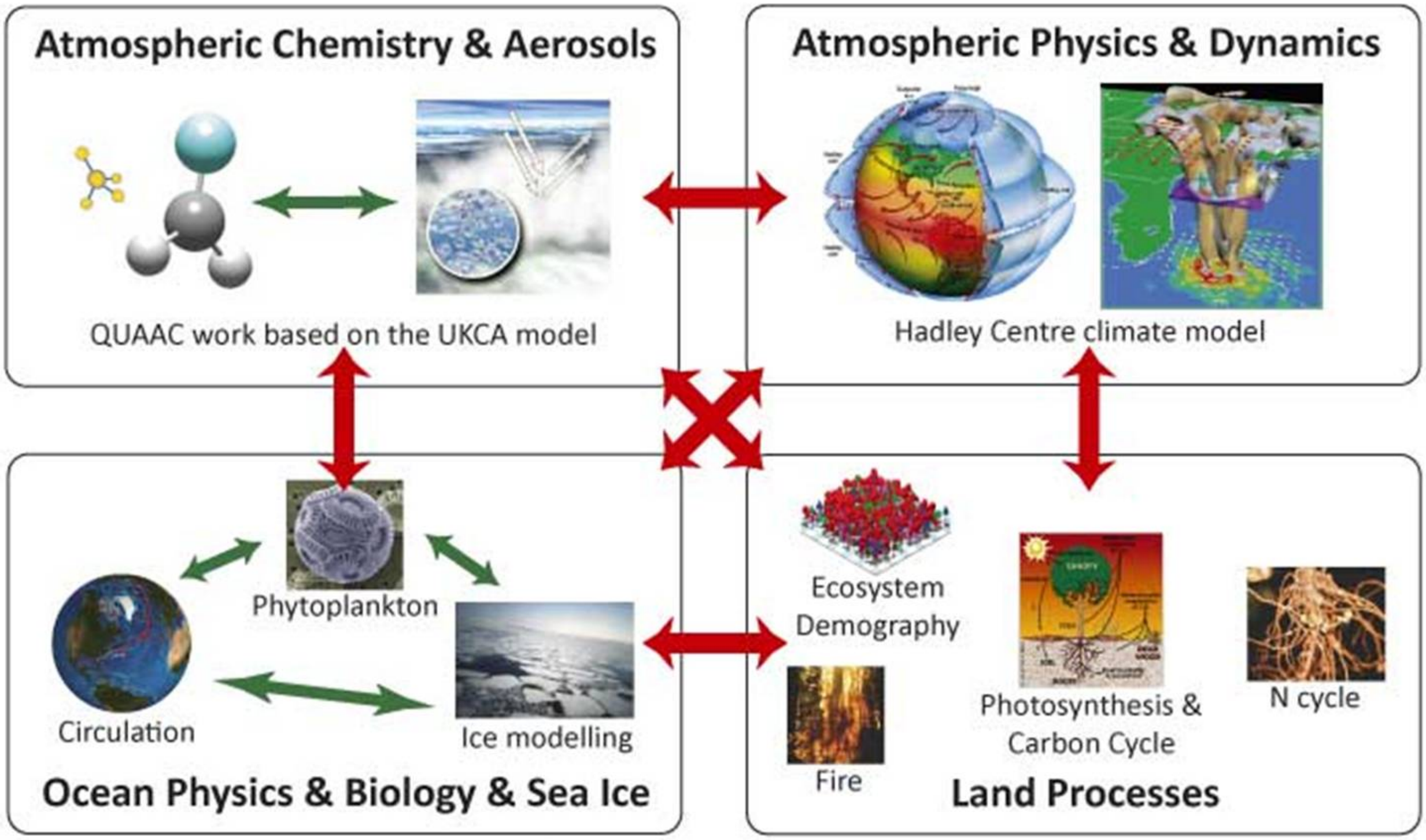


Figure by James Keeble

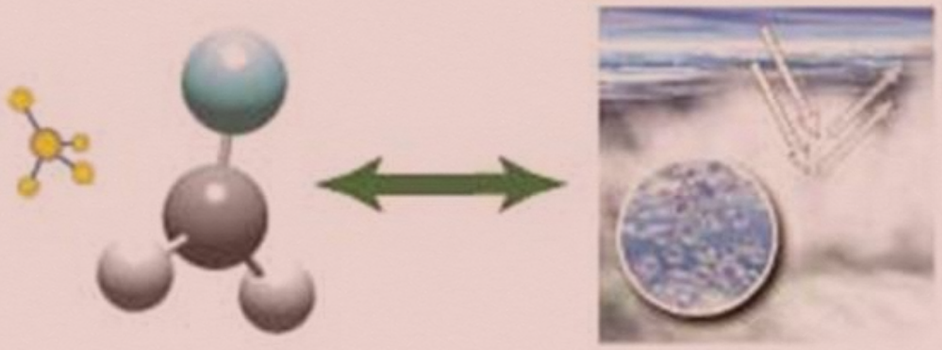
Model components of Earth System



Earth system modelling within QUEST. Based on a diagram by M. Joshi

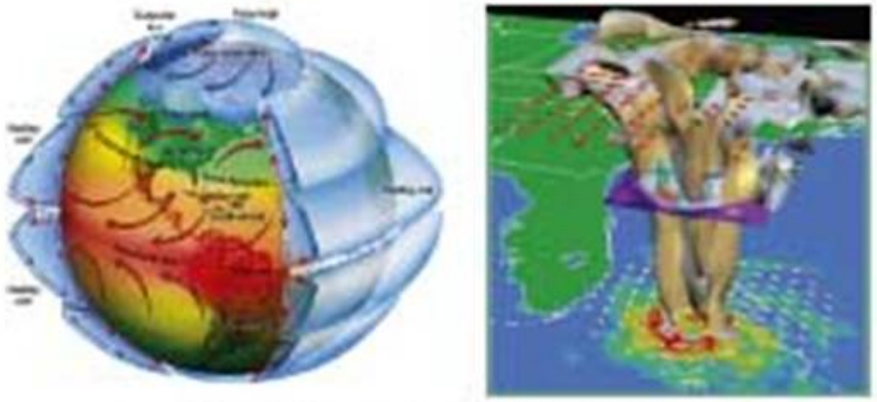
Model components of Earth System

Atmospheric Chemistry & Aerosols

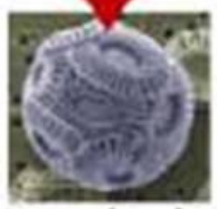


QUAAC work based on the UKCA model

Atmospheric Physics & Dynamics



Hadley Centre climate model

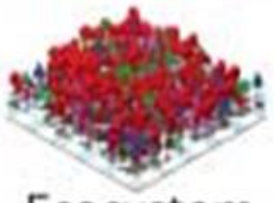


Phytoplankton

Circulation

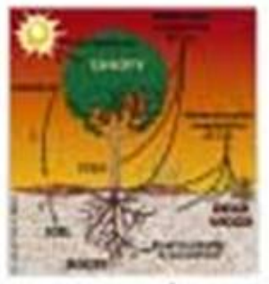
Ice modelling

Ocean Physics & Biology & Sea Ice



Ecosystem Demography

Fire



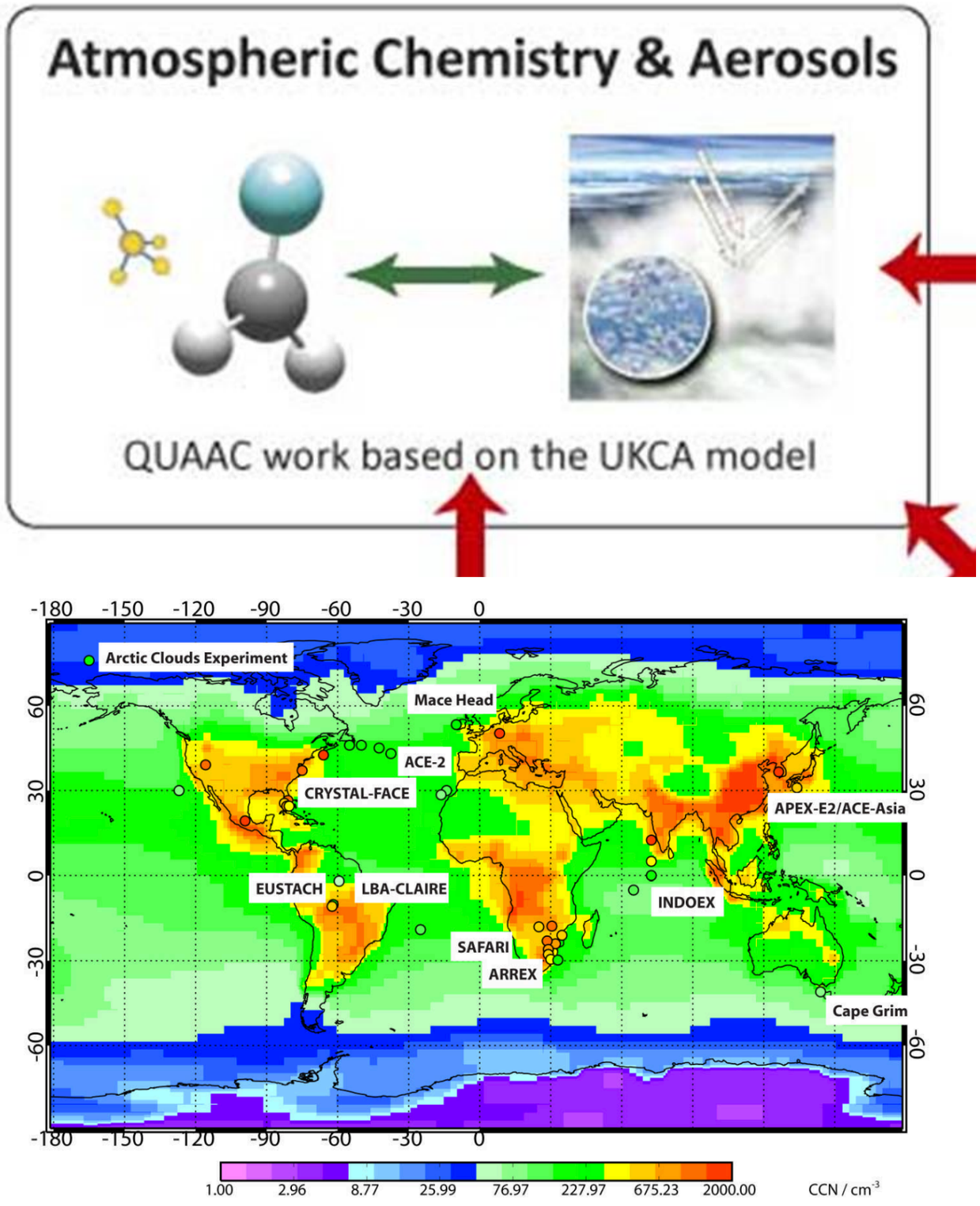
Photosynthesis & Carbon Cycle

N cycle

Land Processes

Earth system modelling within QUEST. Based on a diagram by M. Joshi

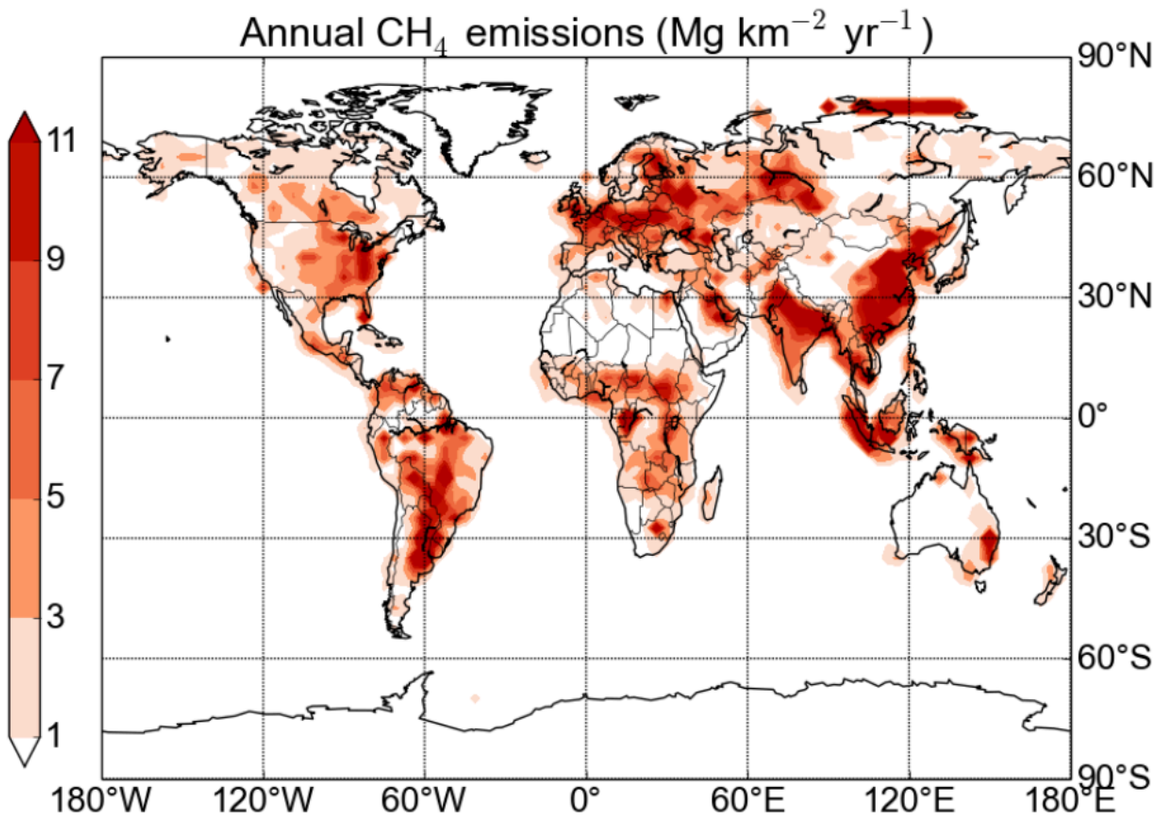
Model components of Earth System

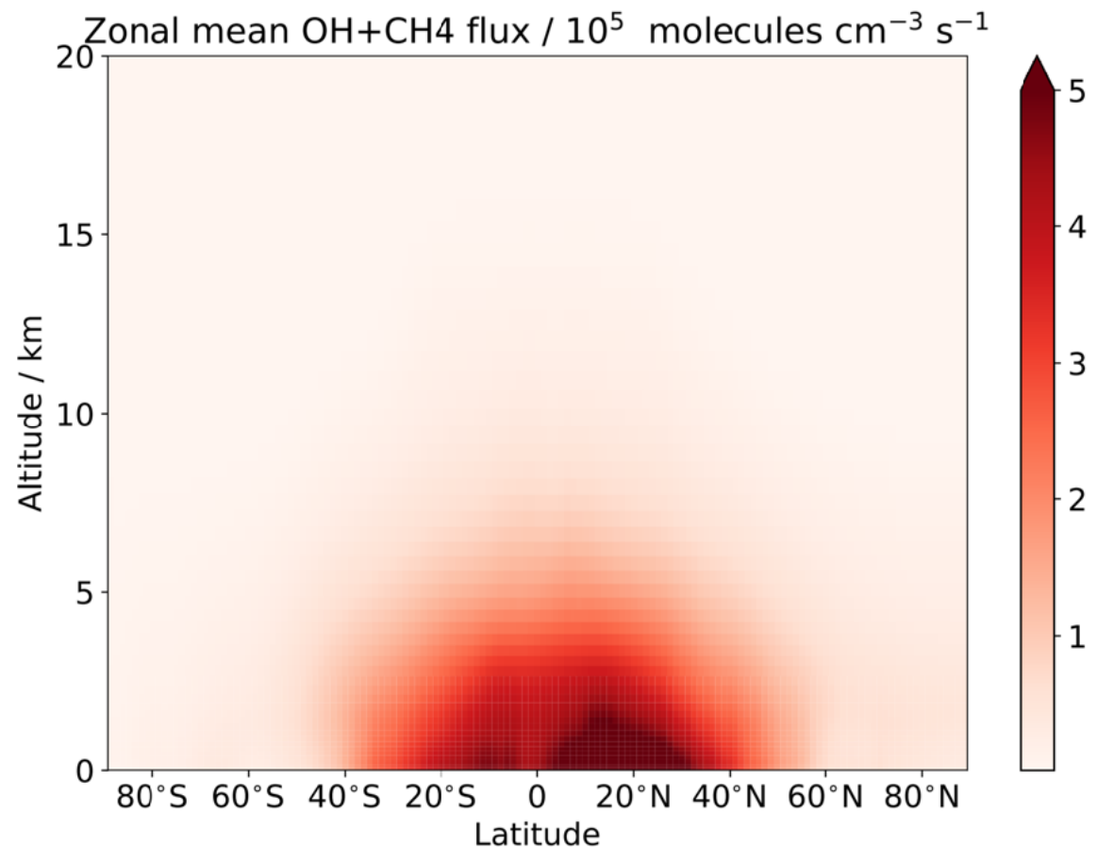


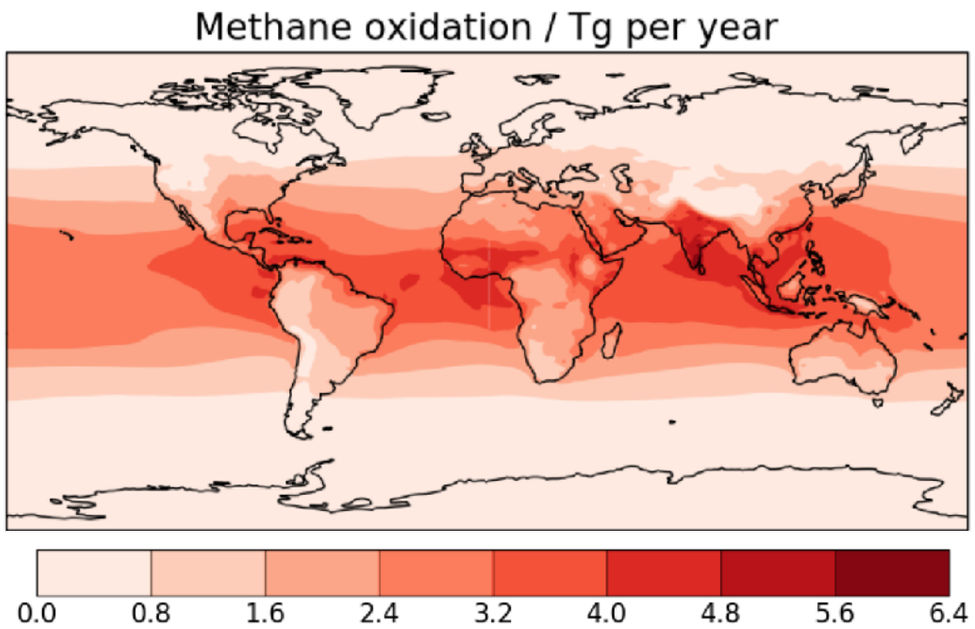
Note: map shows CCN at 0.2% supersaturations.
Coloured circles show observations at range of supersaturations

- Our chemistry module sits inside the UK Met Office Unified Model (UM) and in HadGEM/HadES models
- See Wikipedia (search 'Unified Model')
- Accurate coupling between aerosols and chemistry. Aim to capture feedbacks,
- e.g. SO2 oxidation →sulfate aeroosl → photolysis → OH → sulphate oxidation
- Radiation also included for photochemistry
- GLOMAP-MODE predicts aerosol [Mann, 2010]

Methane in UKCA - emissions vs OH sink



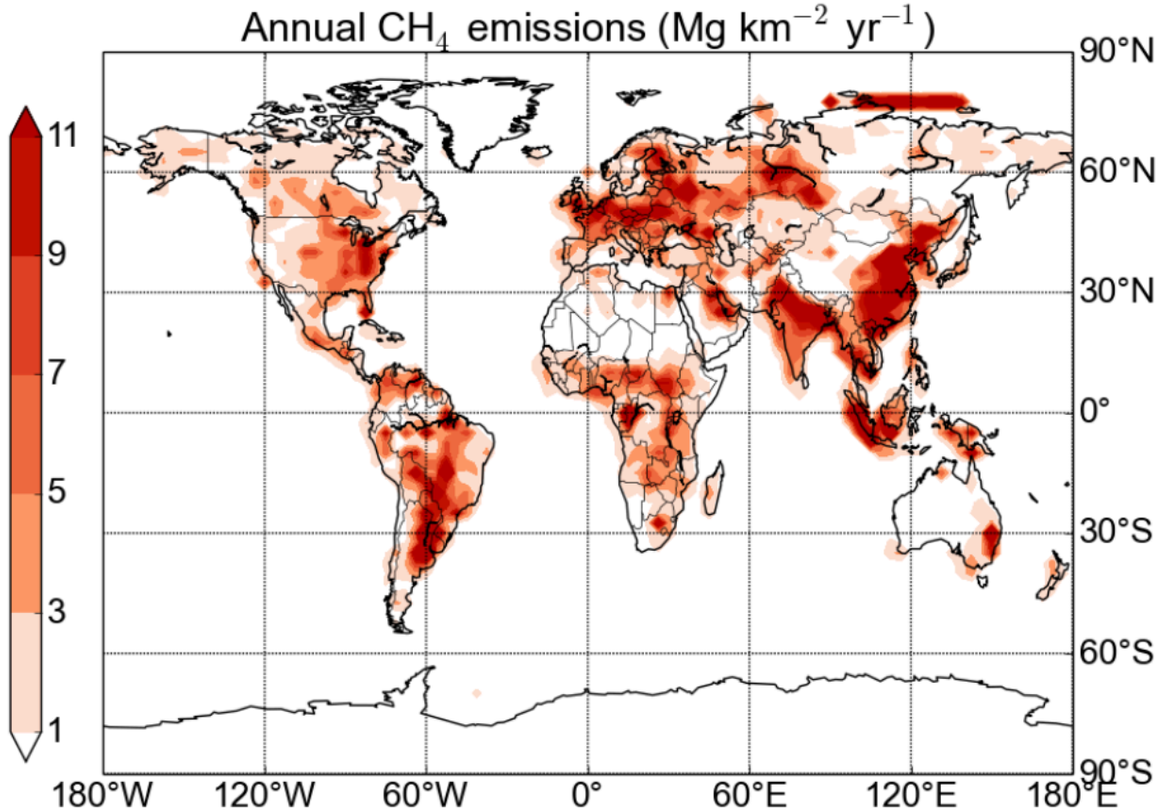


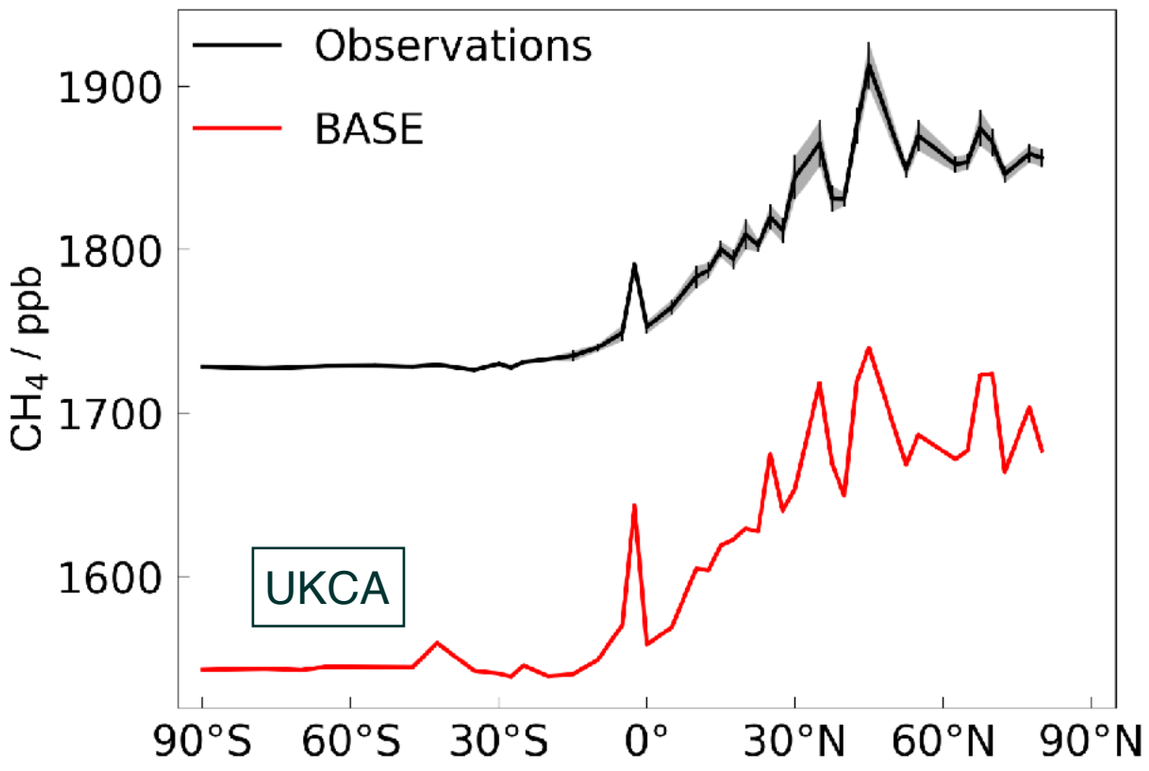


Methane sources are largest in the extra tropics, but oxidation rate is strongly temperature dependent, so peaks where T, humidity and OH high.

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?





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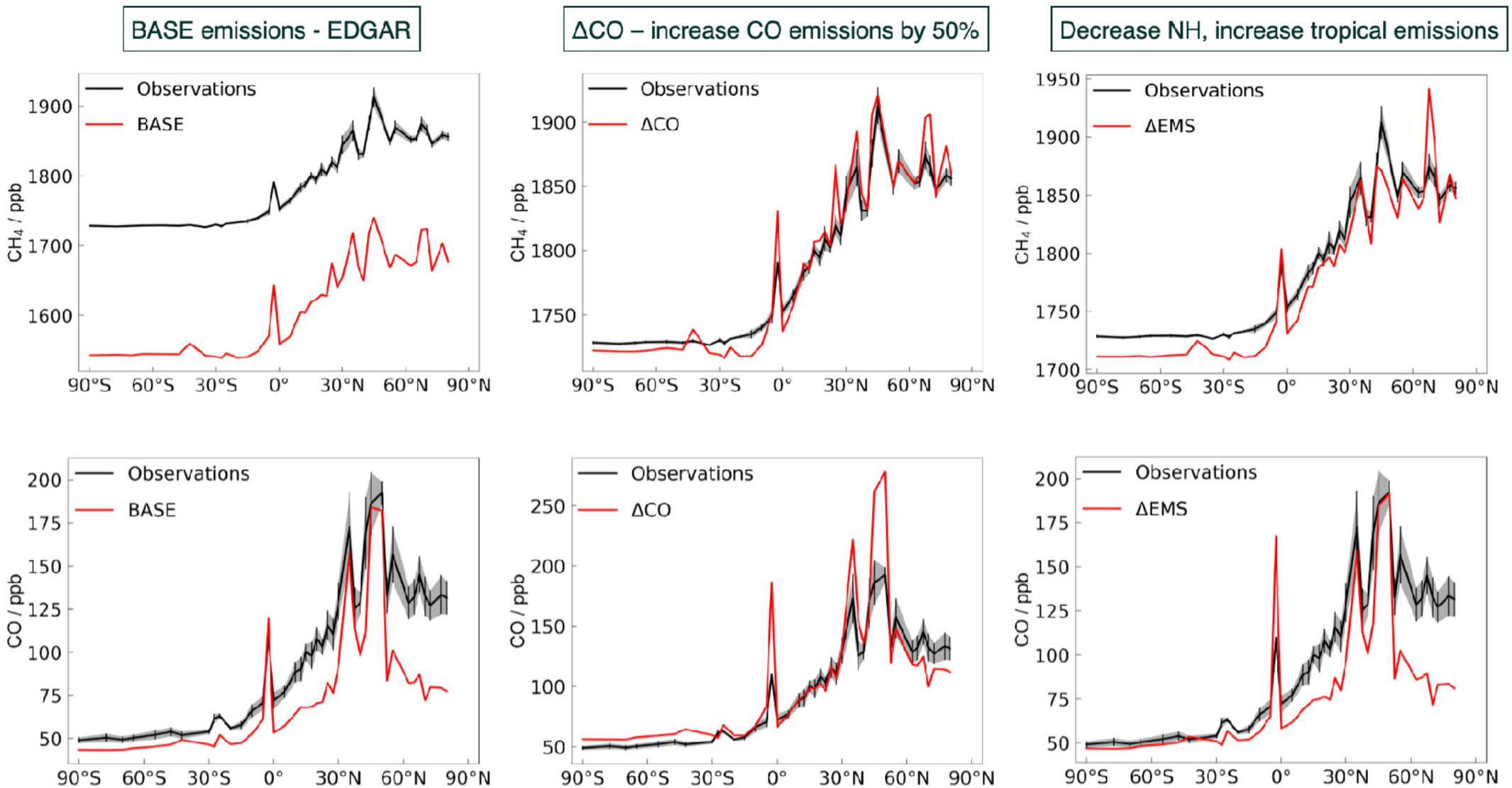
Research Article | Open Access

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

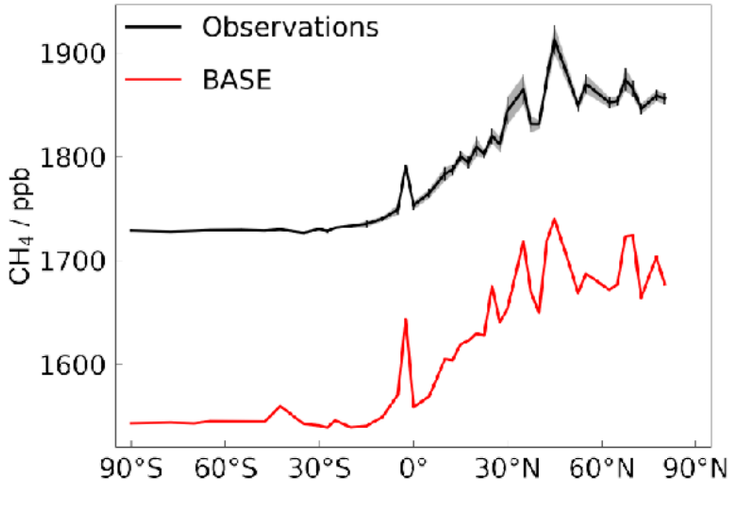
3 sensitivity experiments

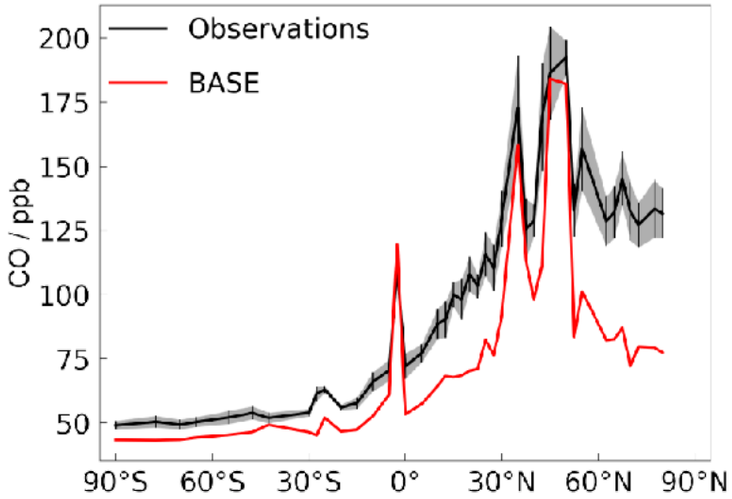
1. Our BASE run using methane emissions derived from EDGAR emissions database.
2. A second experiment in which CO emissions are increased everywhere by 50%
3. An experiment in which we use a different emissions dataset with lower emissions in NH midlatitudes higher emissions in tropics.

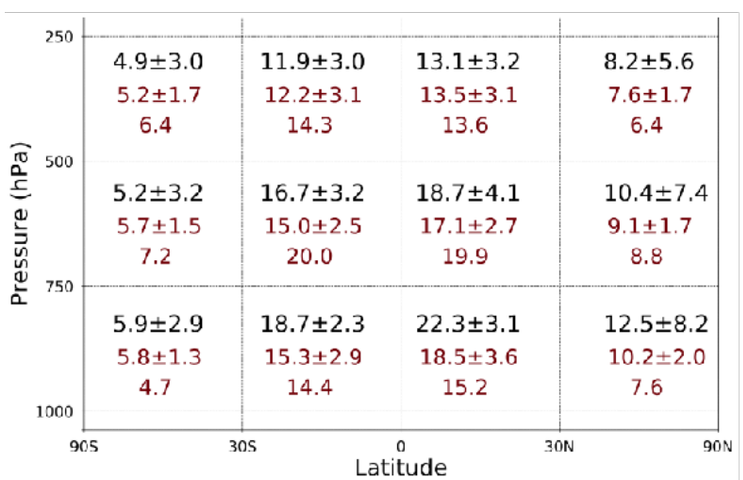


Sensitivity of UKCA to emissions – 3 global experiments

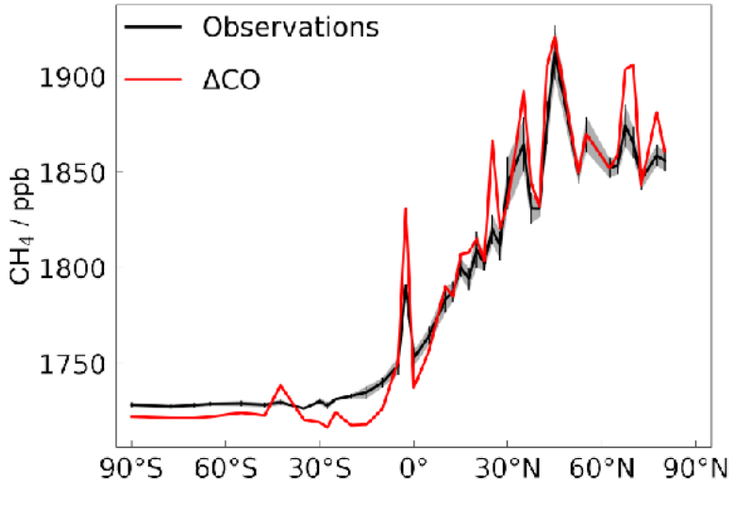
BASE emissions - EDGAR

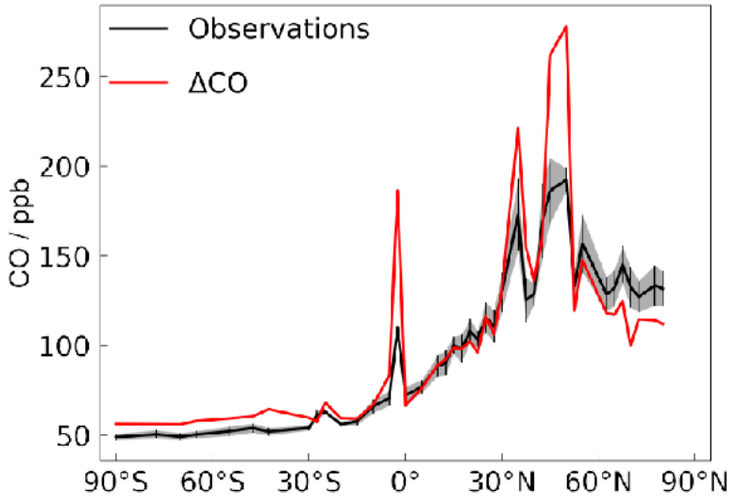


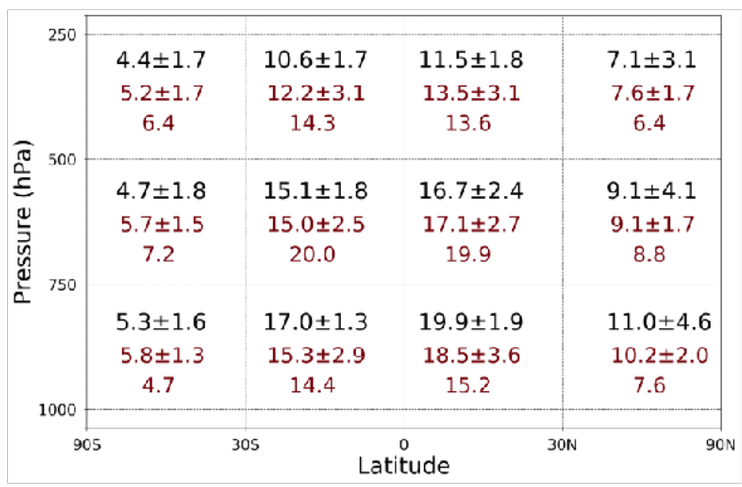




ΔCO – increase CO emissions by 50%







Decrease NH, increase tropical emissions