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

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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

- I'm a Guest Professor at the Centre for Climate Systems Research, Tokyo University (end Sep – end Dec).
- Undergrad in chemistry; PhD in chemical physics.

- Since 2016, I work in the National Centre for Atmospheric Science, based in Cambridge
- IPCC Contributing Author, AR6, 2020.
- Co-chair UK Atmospheric Science Special Interest Group for Royal Met Soc, since 2020
- Co-chair Model Evaluation Working Group for UKCA chemistry-climate model since 2018
- Visiting Scientist (NARIT, Chiang Mai, Thailand 2016-2020)

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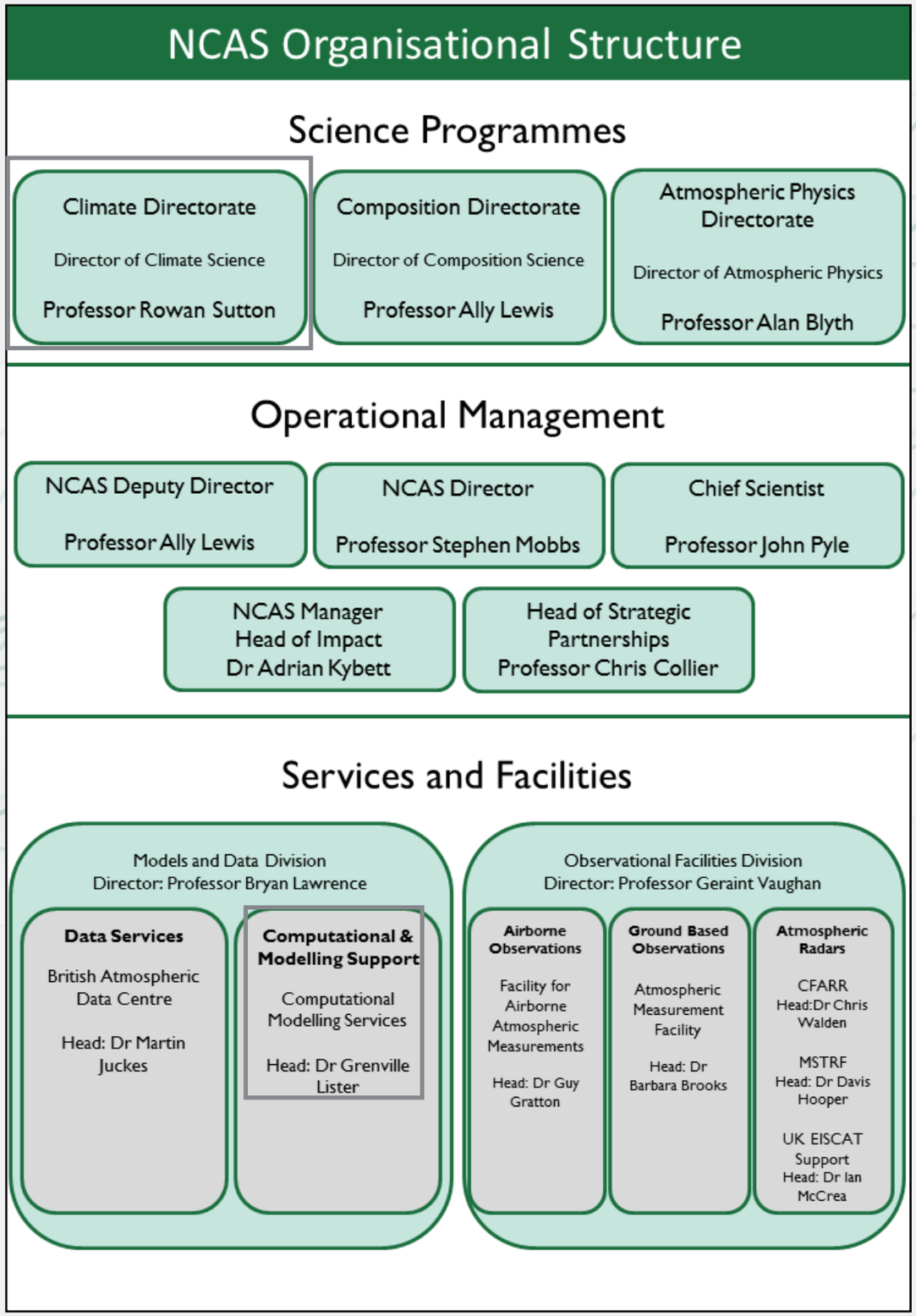
Atmospheric Science in the UK - NCAS

- Funding comes from the UK Natural Environment Research Council (**NERC**).
- **NERC** funds research centres like the **British Antarctic Survey** (Cambridge), **National Oceanographic Centre** (Southampton) and the **Centres for Hydrology and Ecology.**
- I work in the **National Centre for Atmospheric Science** (NCAS).
- **NCAS** is the UK centre for Atmospheric Science, researchers are distributed across the UK. Budget £9M.
- I am employed in NCAS **Climate**, working on modelling atmospheric working on air quality and aerosols.



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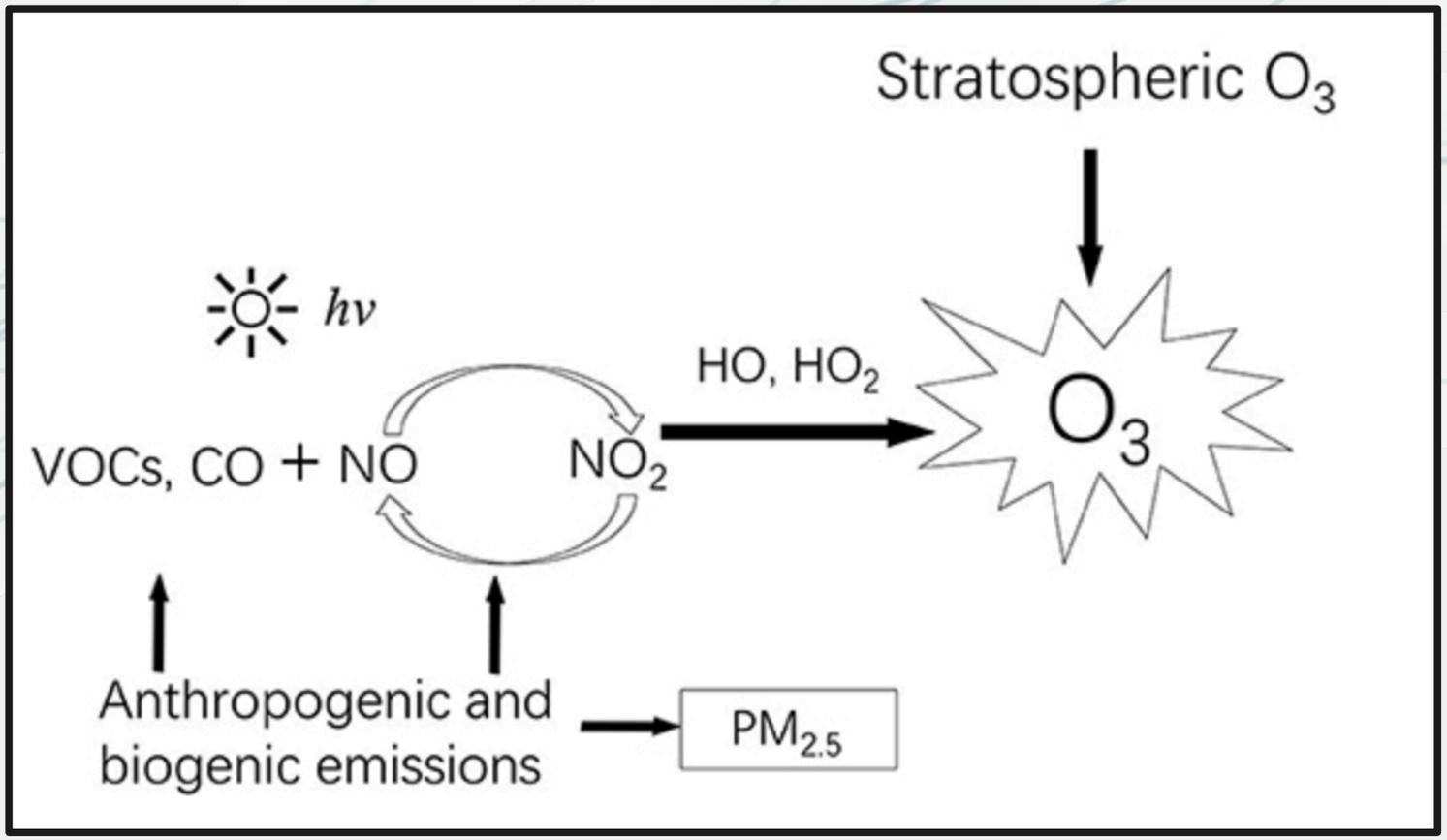
Ozone in the troposphere

- Ozone is interesting to a chemist because it’s not emitted directly, but is formed in the atmosphere via atmospheric chemistry processes.
- Also need to include dynamical transport of ozone into the stratosphere (STE)
- Ozone precursors are from anthropogenic and biogenic sources: both hydrocarbons and NOx
- Sunlight/humidity/temperature are all important to ozone formation
- Ozone deposition at the surface to vegetation - connection to land cover



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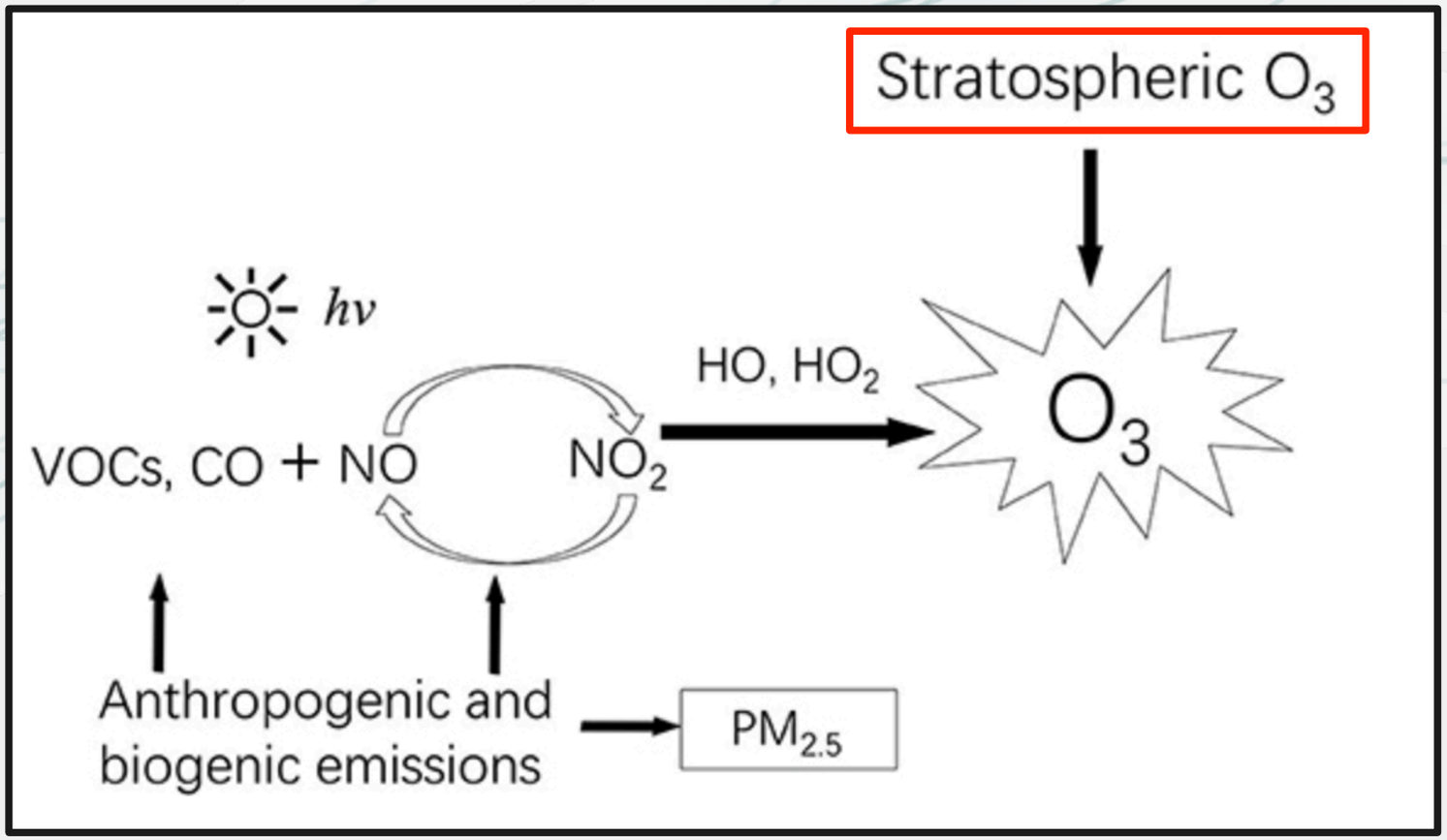

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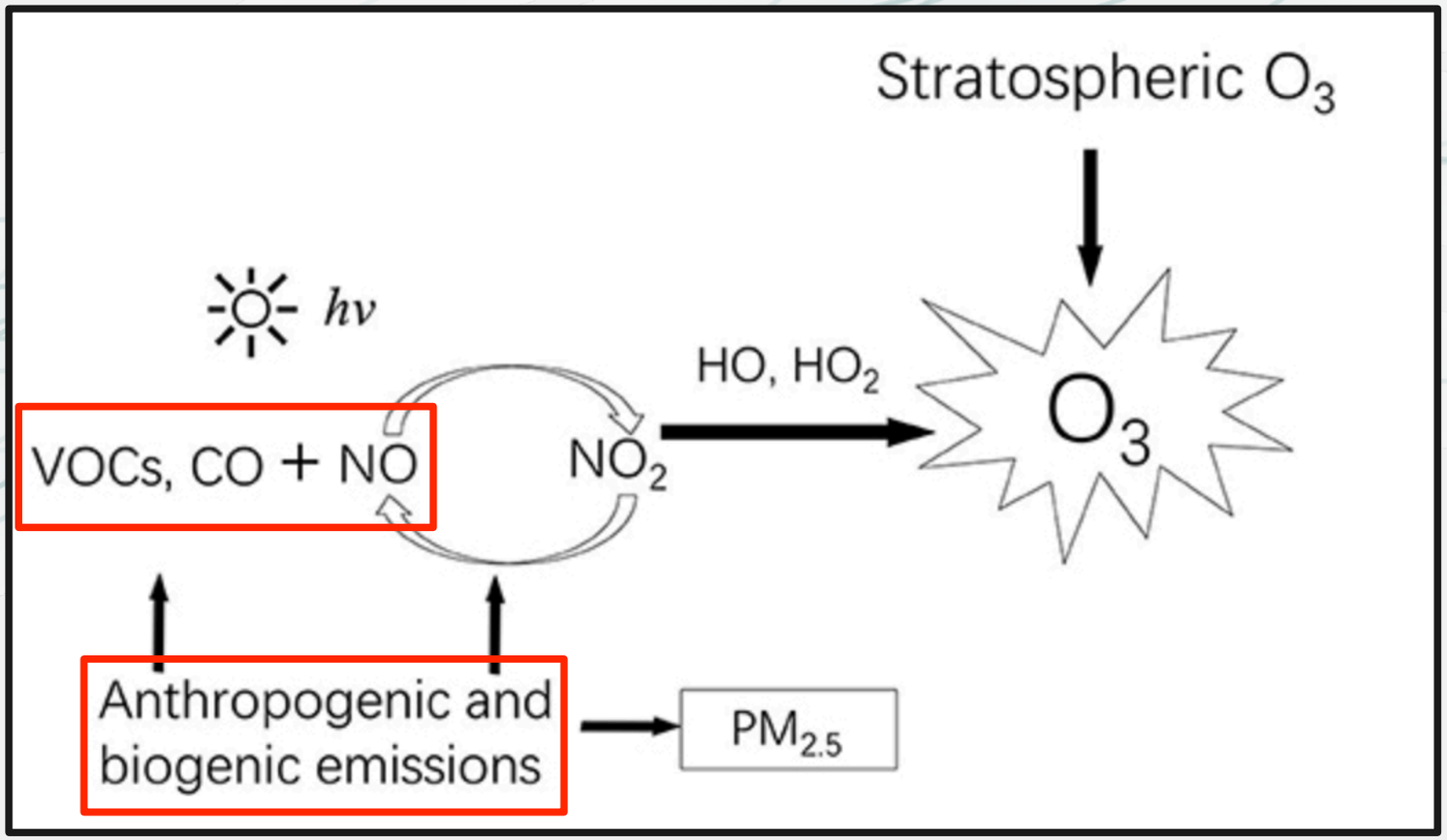

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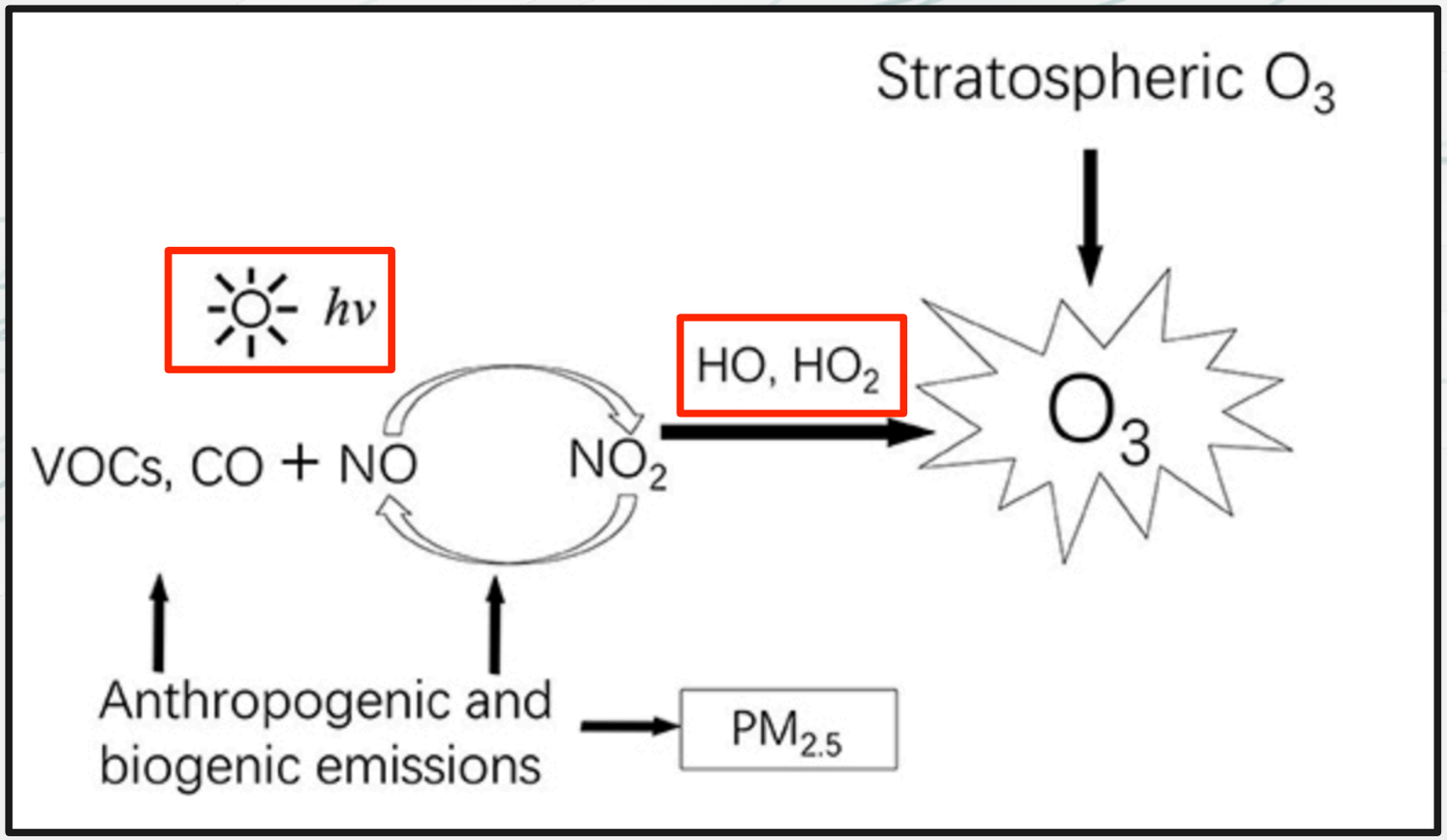

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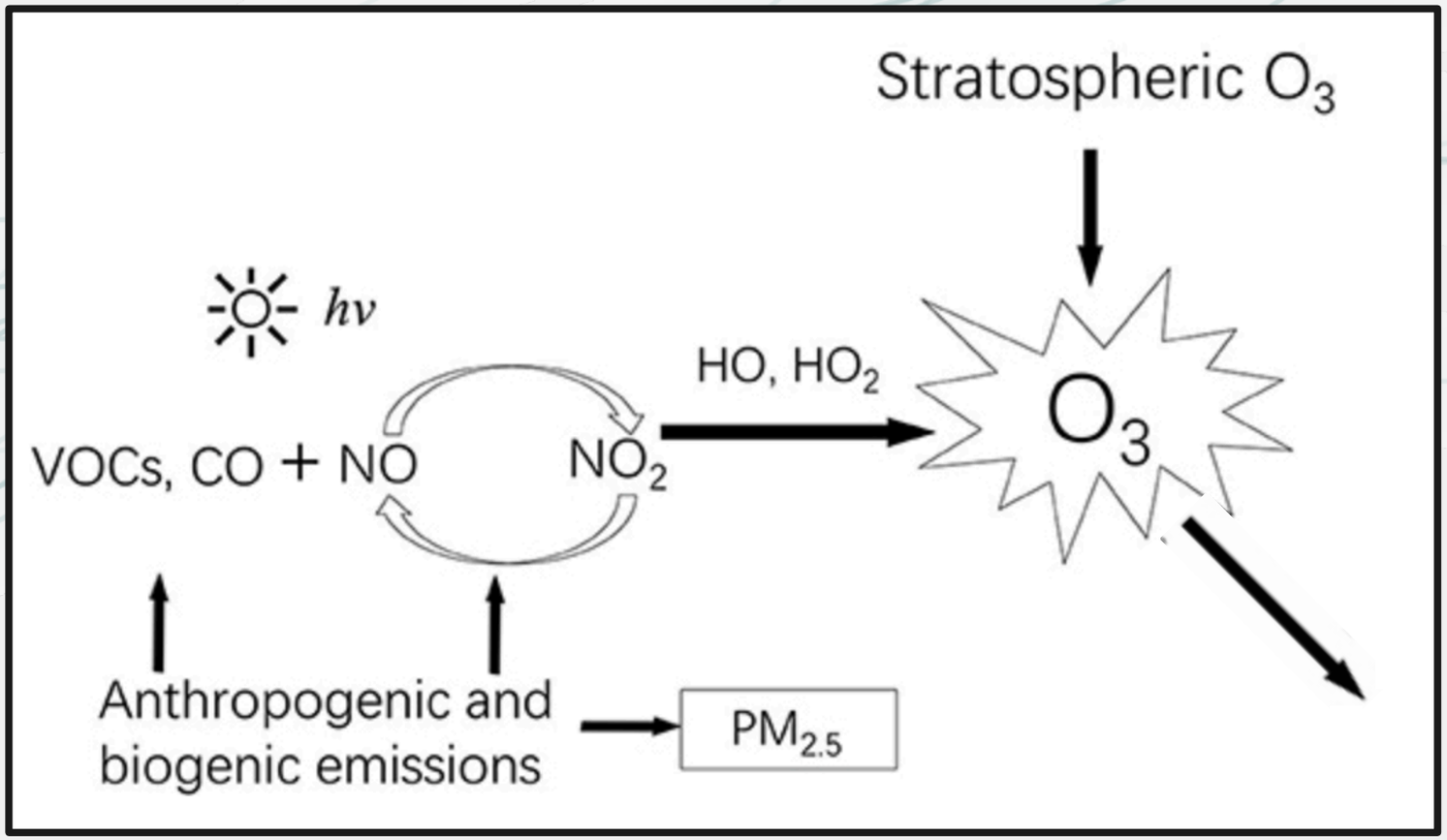

Tropospheric Ozone in CMIP6

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

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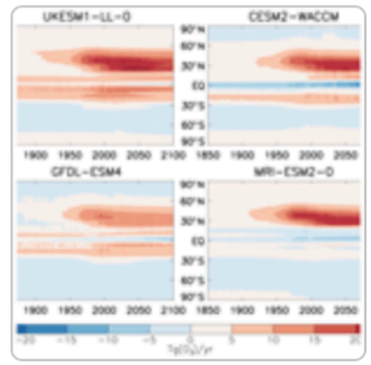
18 Mar 2021

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]

Many thanks to the centres for their hard work in preparing the data to a very tight deadline!

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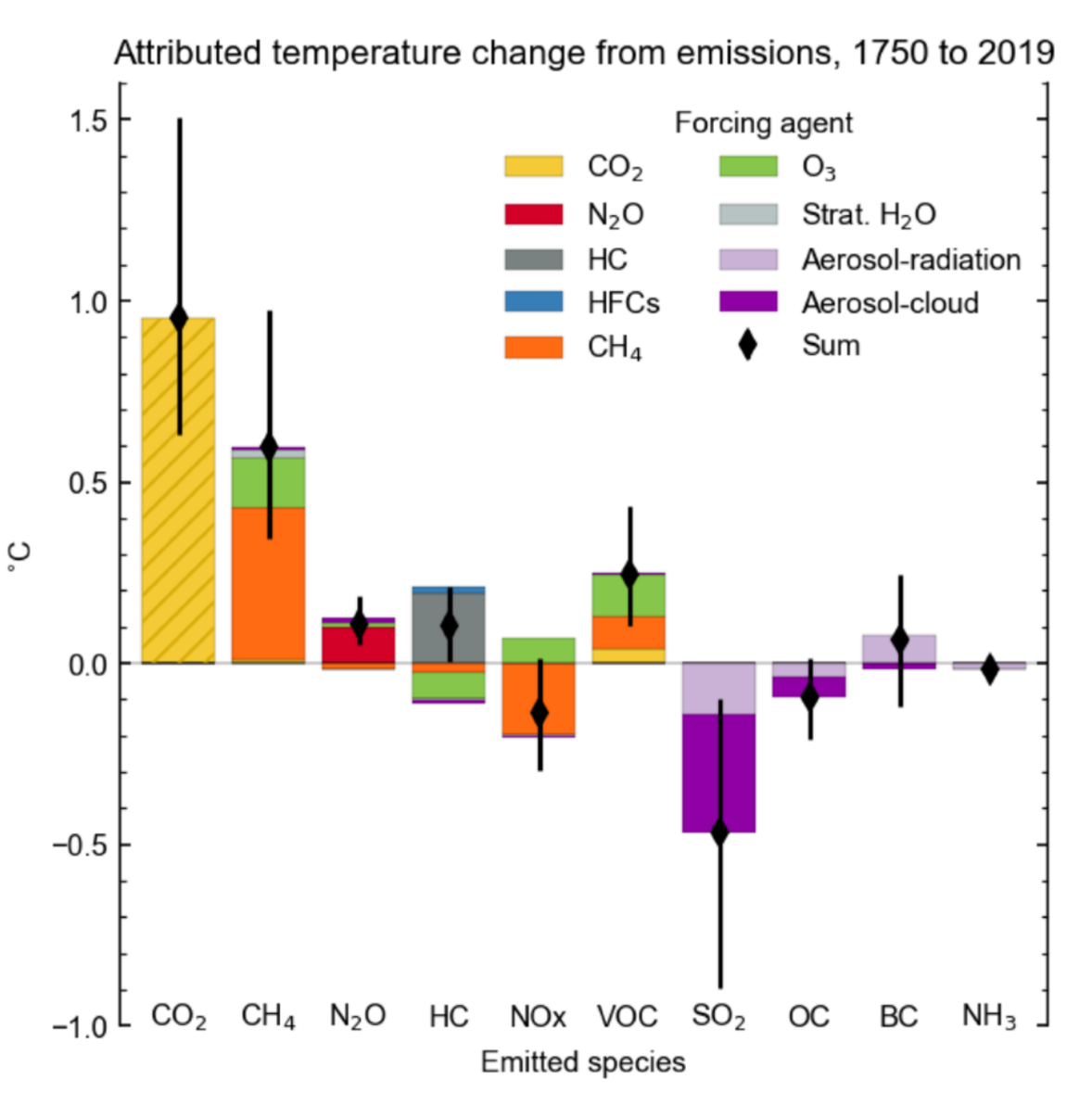
IPCC AR6: ozone as a short-lived climate forcer



- CMIP6 produced an attribution of temperature change by species
- Also breaks down anthropogenic emissions by impact
- Positive forcing (warming) and negative forcing (cooling) are found
- Some large error bars!

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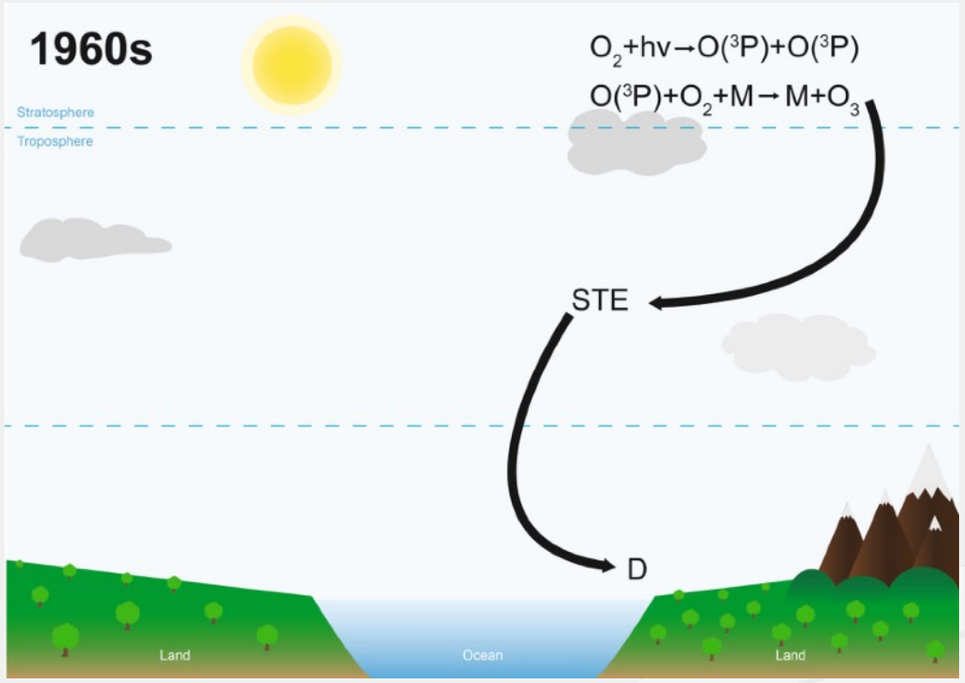
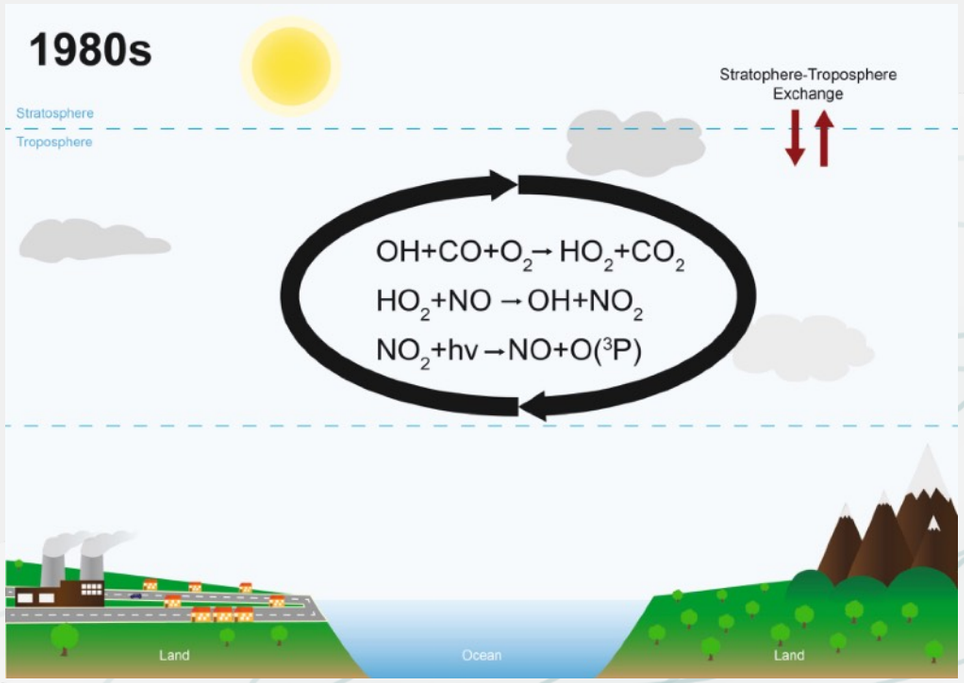
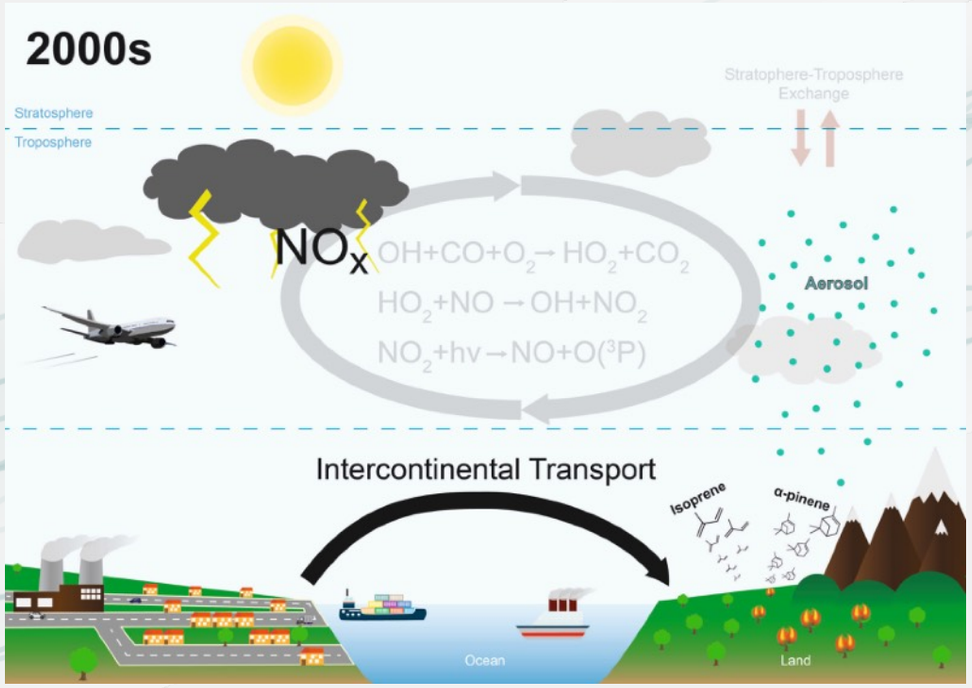
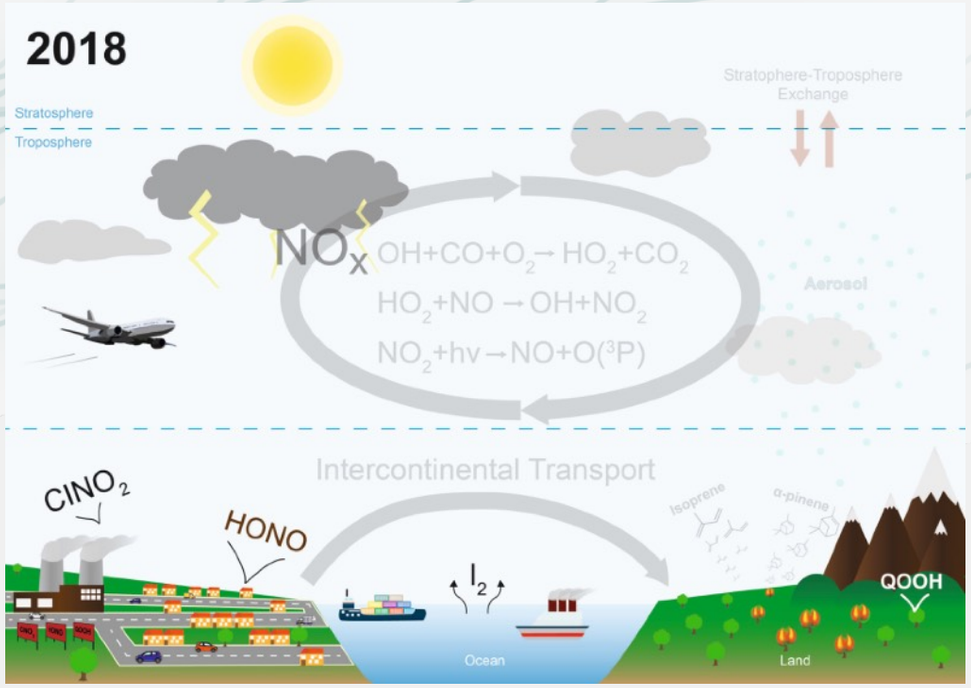
Ozone in CCMs – developing complexity









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Archibald et al., TOAR "Budget", Elementa 2021

How does tropospheric ozone evolve in CMIP6?

Questions for a chemistry-led assessment:

- How well do models simulate ozone across the historical period?
- Where do models agree consistently? Where is there uncertainty?
- What drives ozone changes across the historical period?
- How will ozone evolve into the future?

Not covered by our assessment - done elsewhere

- What are the radiative impacts?
- What about health, vegetation impacts?

Drew heavily on the Tropospheric Ozone Assessment Report (2018-2021) led by Owen Cooper at NOAA

Co-lead with Lee Murray, U. Rochester

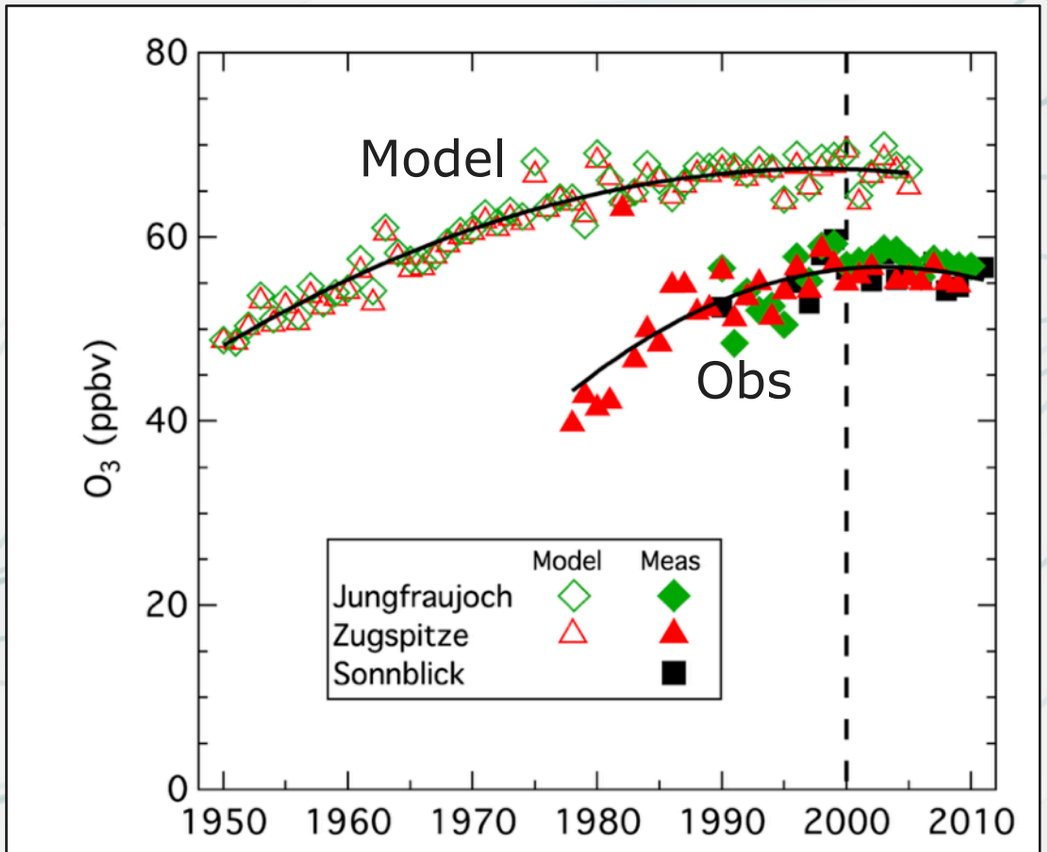

Figure 1. Seasonally averaged springtime (March, April, and May) O_3 concentrations at alpine sites in Europe. Closed and open symbols give measurements and GFDL CCM results, respectively. The solid lines give quadratic fits to respective results. The vertical dashed line indicates the year 2000 reference.

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How does tropospheric ozone evolve in CMIP6?

- 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 effect of stratospheric ozone depletion and recovery
- 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								

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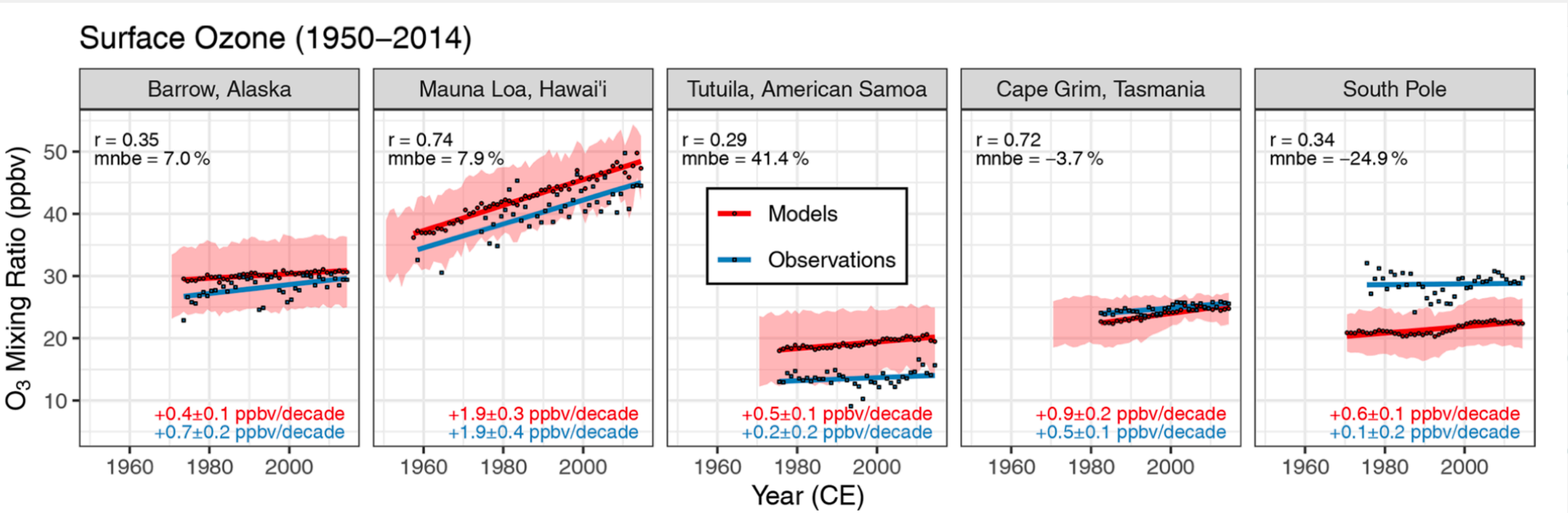
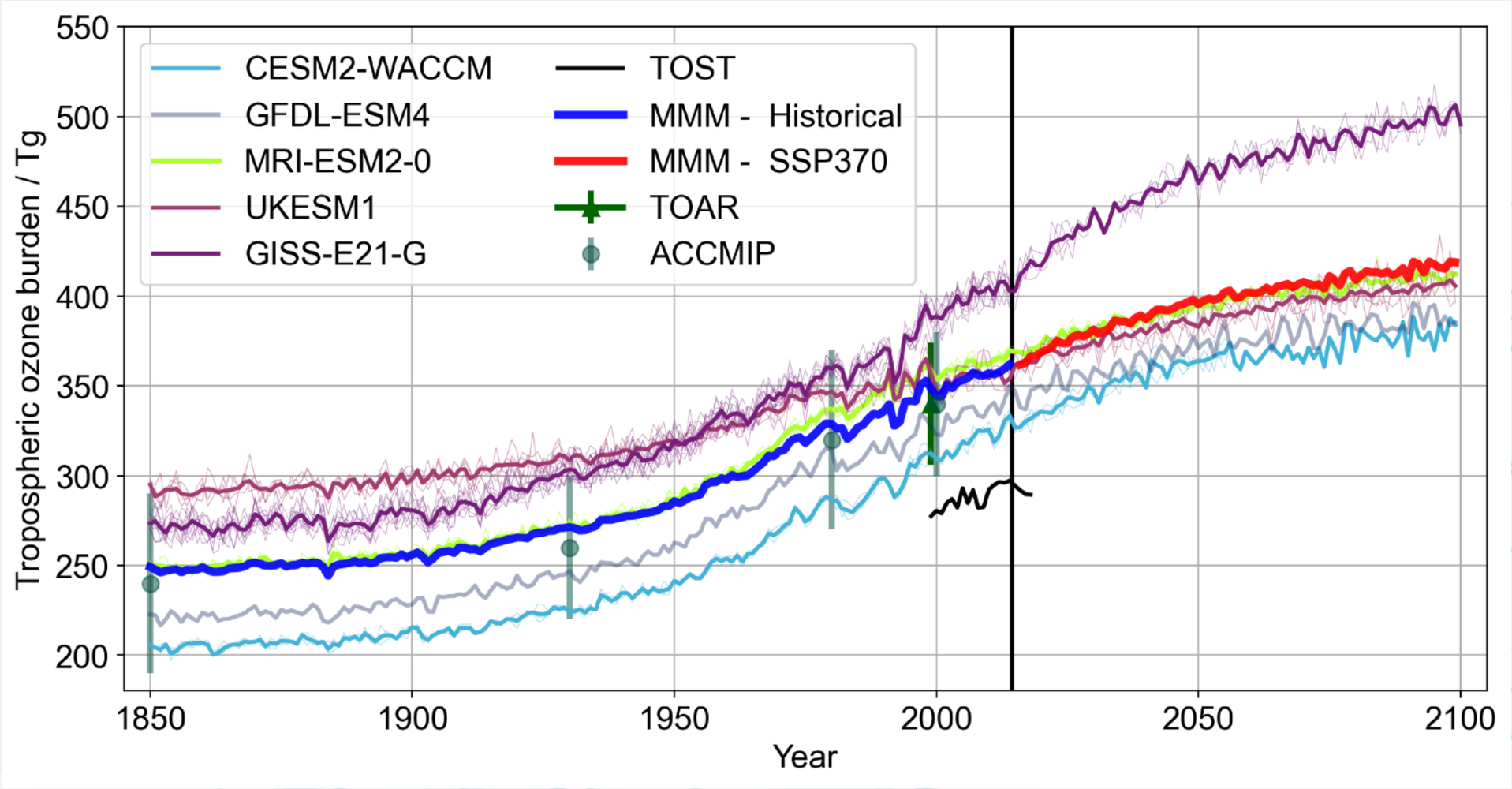
How does tropospheric ozone evolve in CMIP6? Comparison with obs



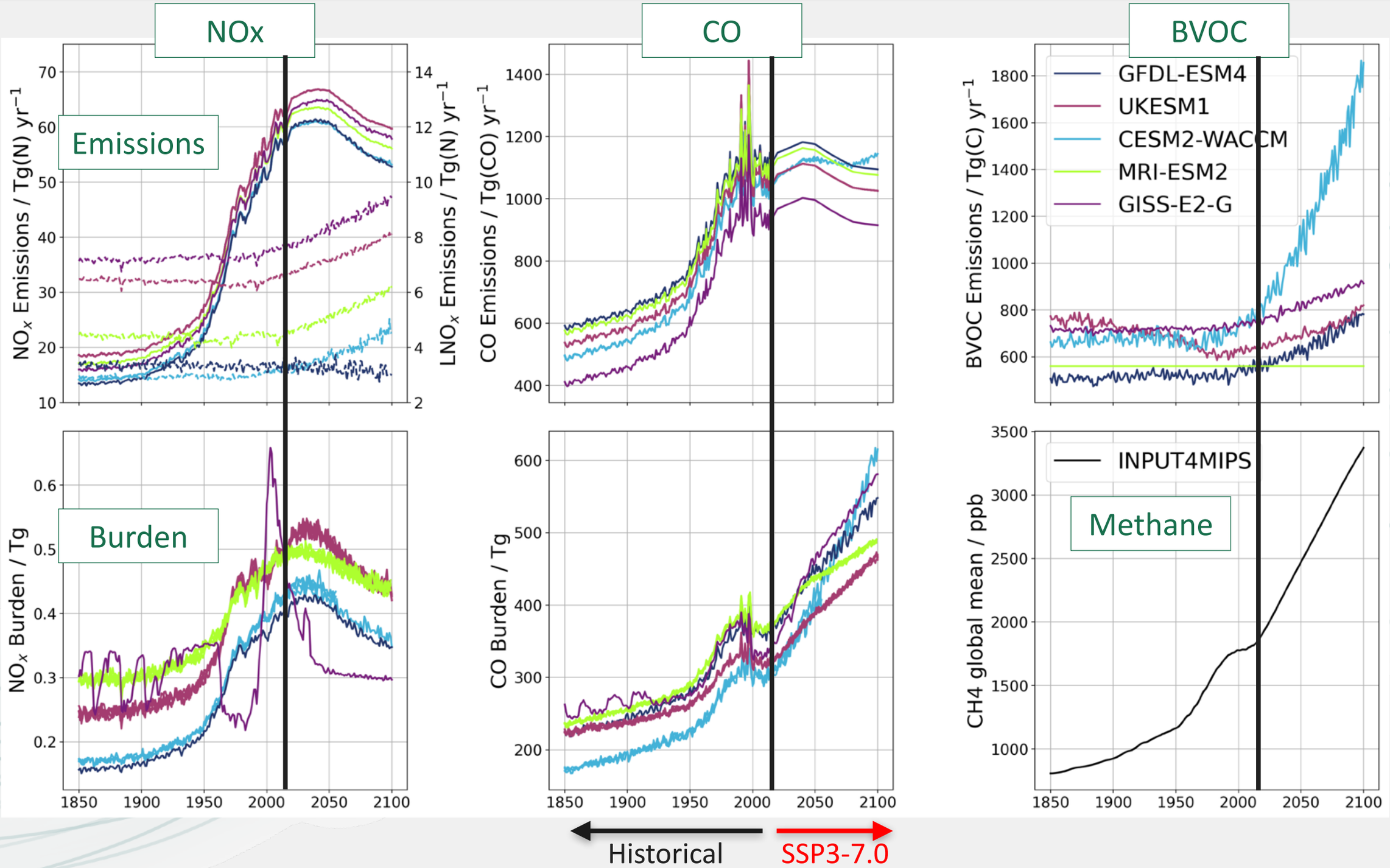
- CMIP6 featured coupled atmosphere-ocean models with online, whole-atmosphere chemistry.
- **Good agreement between models and observations for the remote sites studied here.**
- Also found nice agreement between in-situ ozone sonde measurements.
- **Assessment using EO products more of a challenge** - 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 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.

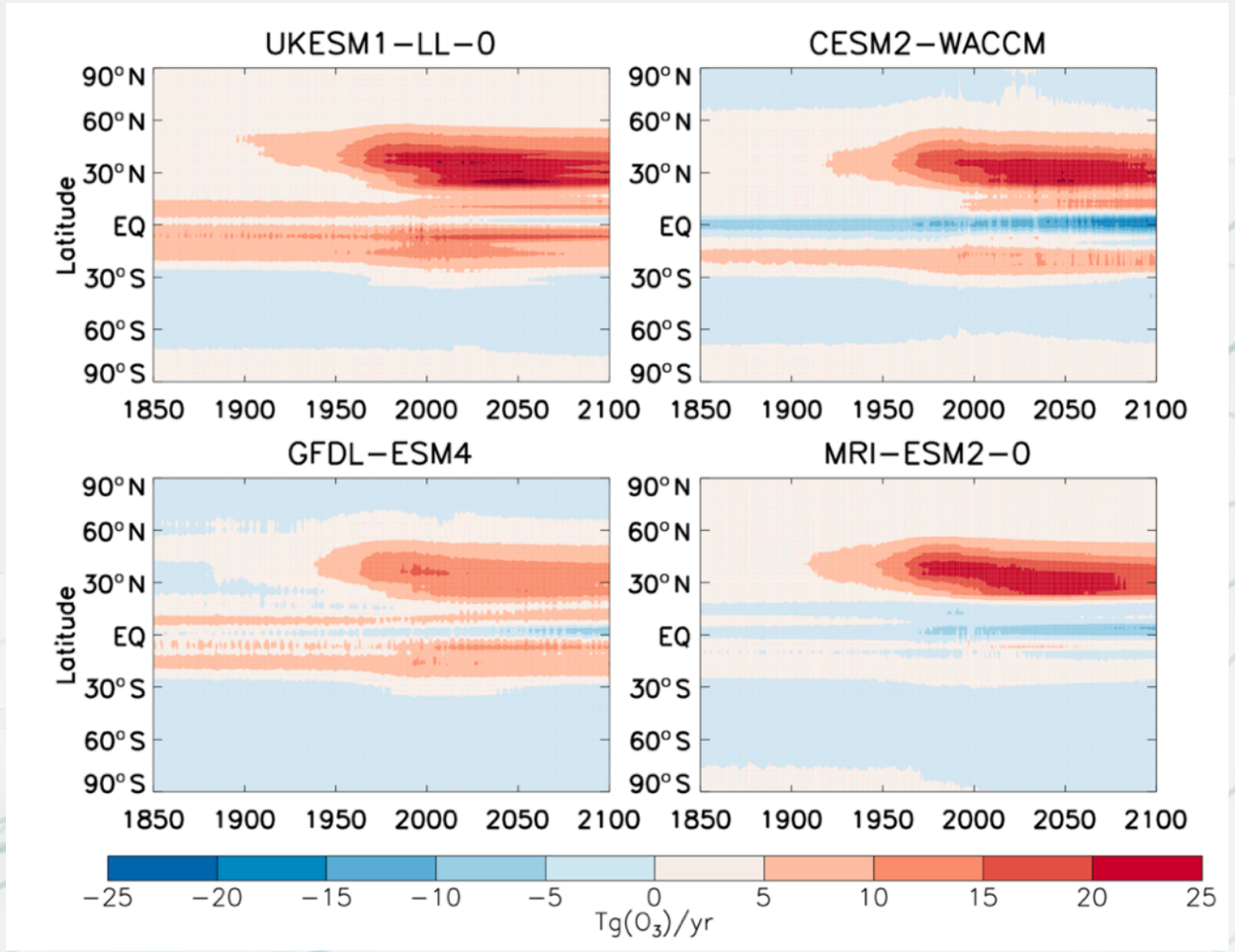
What drives tropospheric ozone in CMIP6?

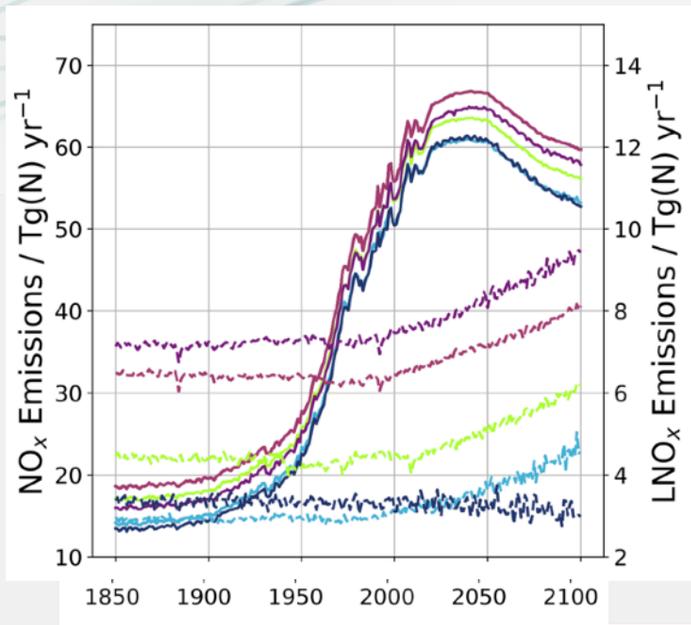


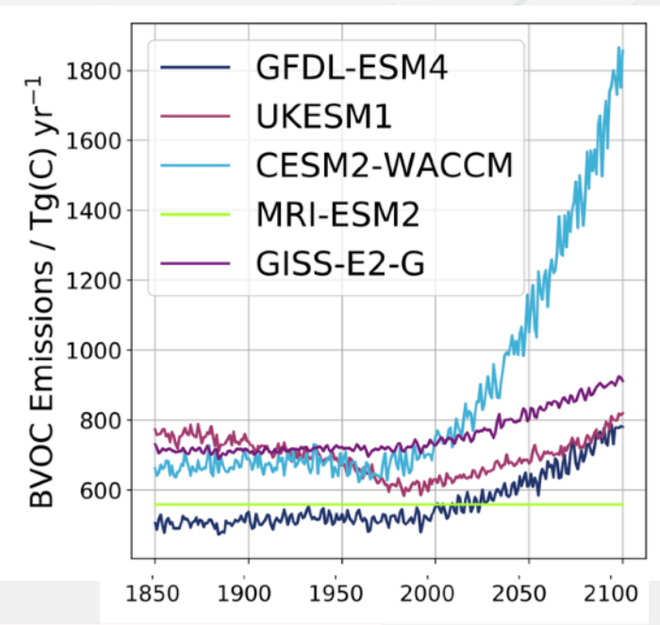
- Decline in precursor emissions in SSP3-7.0 experiments
- NB **steady increase in ozone burden** - Strat O3 recovery increasing role + LiNOx

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

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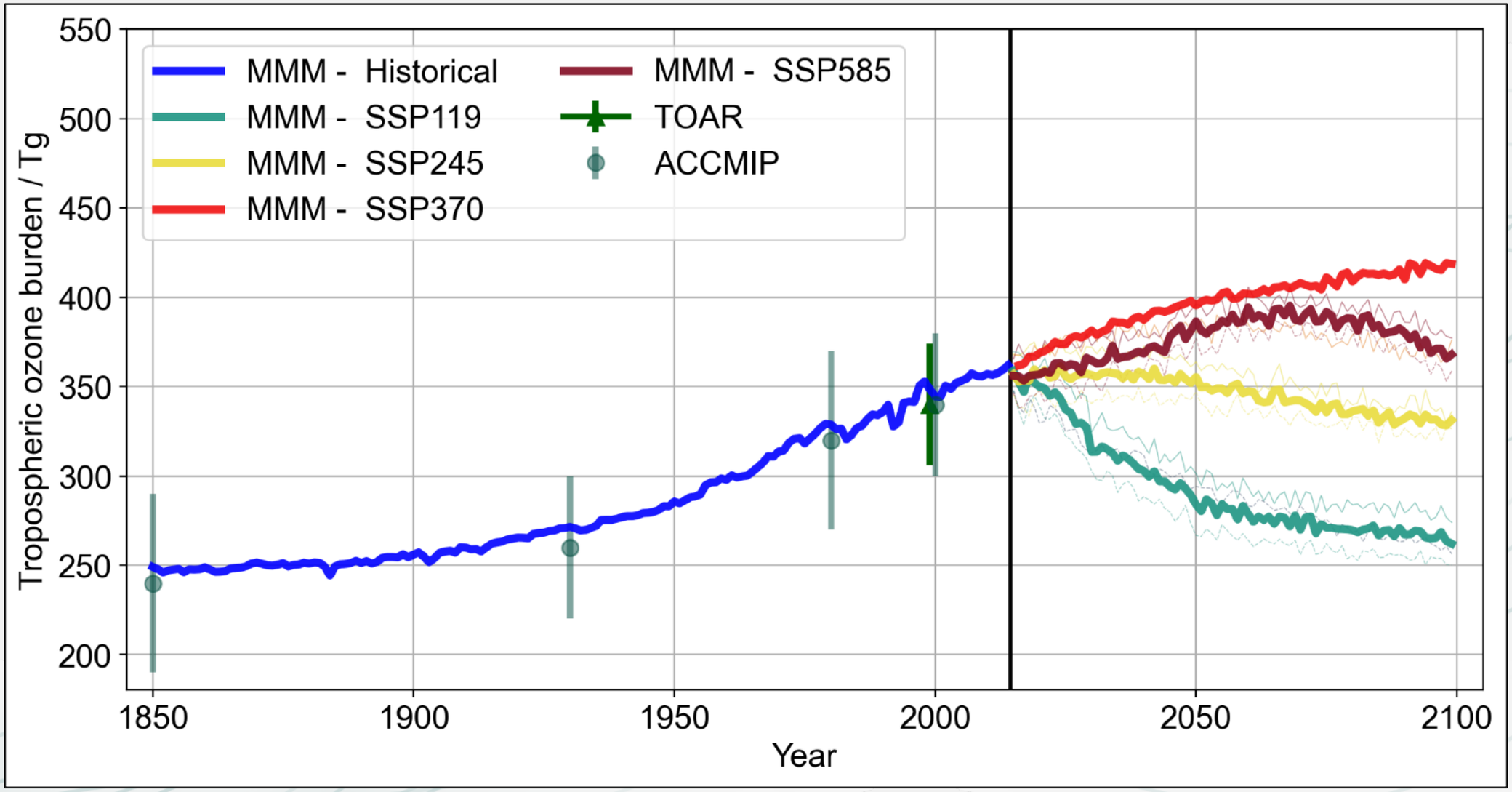
Database of of tropospheric ozone burden changes



- Initial results (dataset is rather incomplete)

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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

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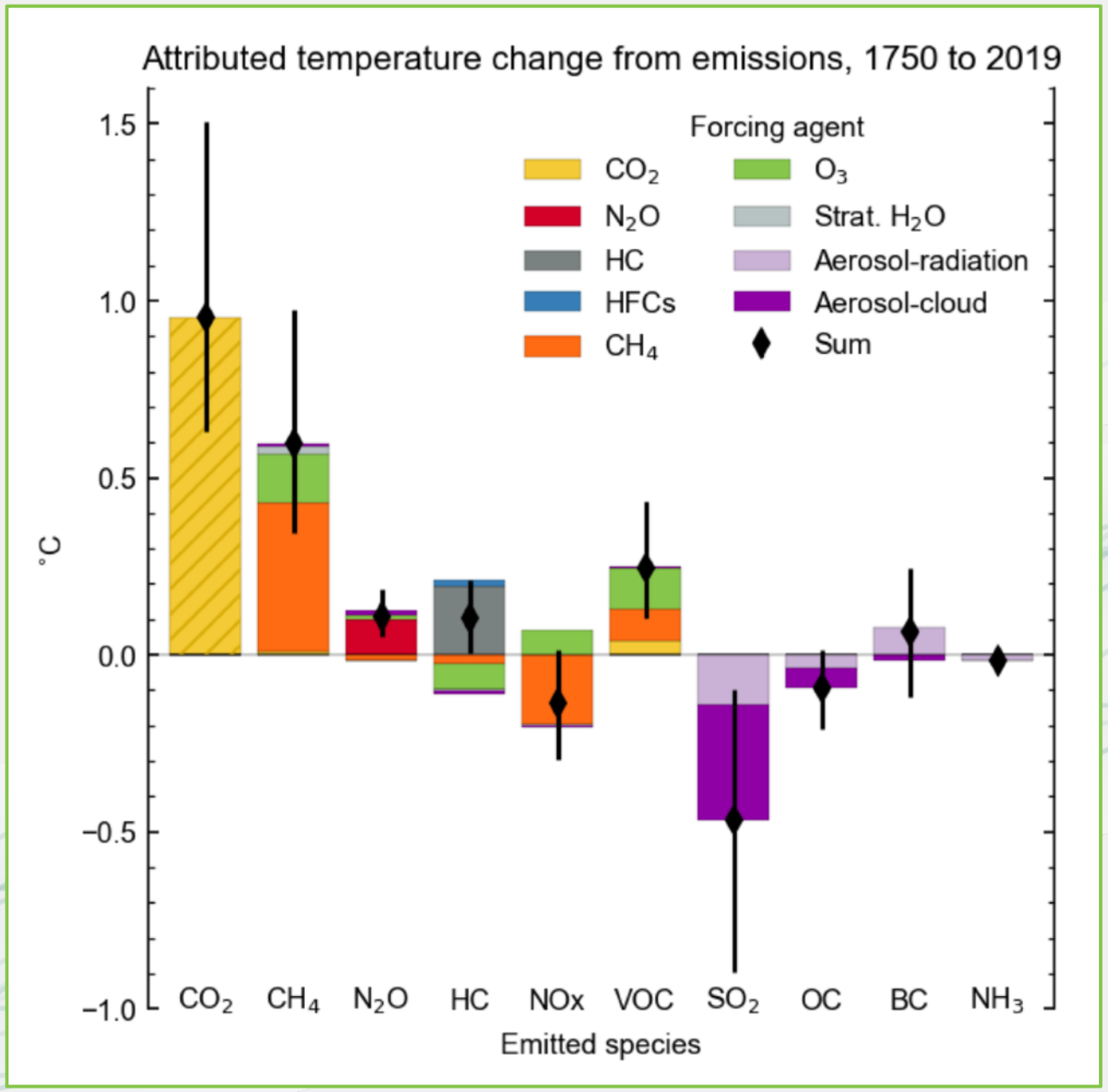
The role of methane and oxidants in climate



- CMIP6 attributed the radiative forcing and temperature change to various chemical species.
- Methane provides the second largest anthropogenic climate driver.

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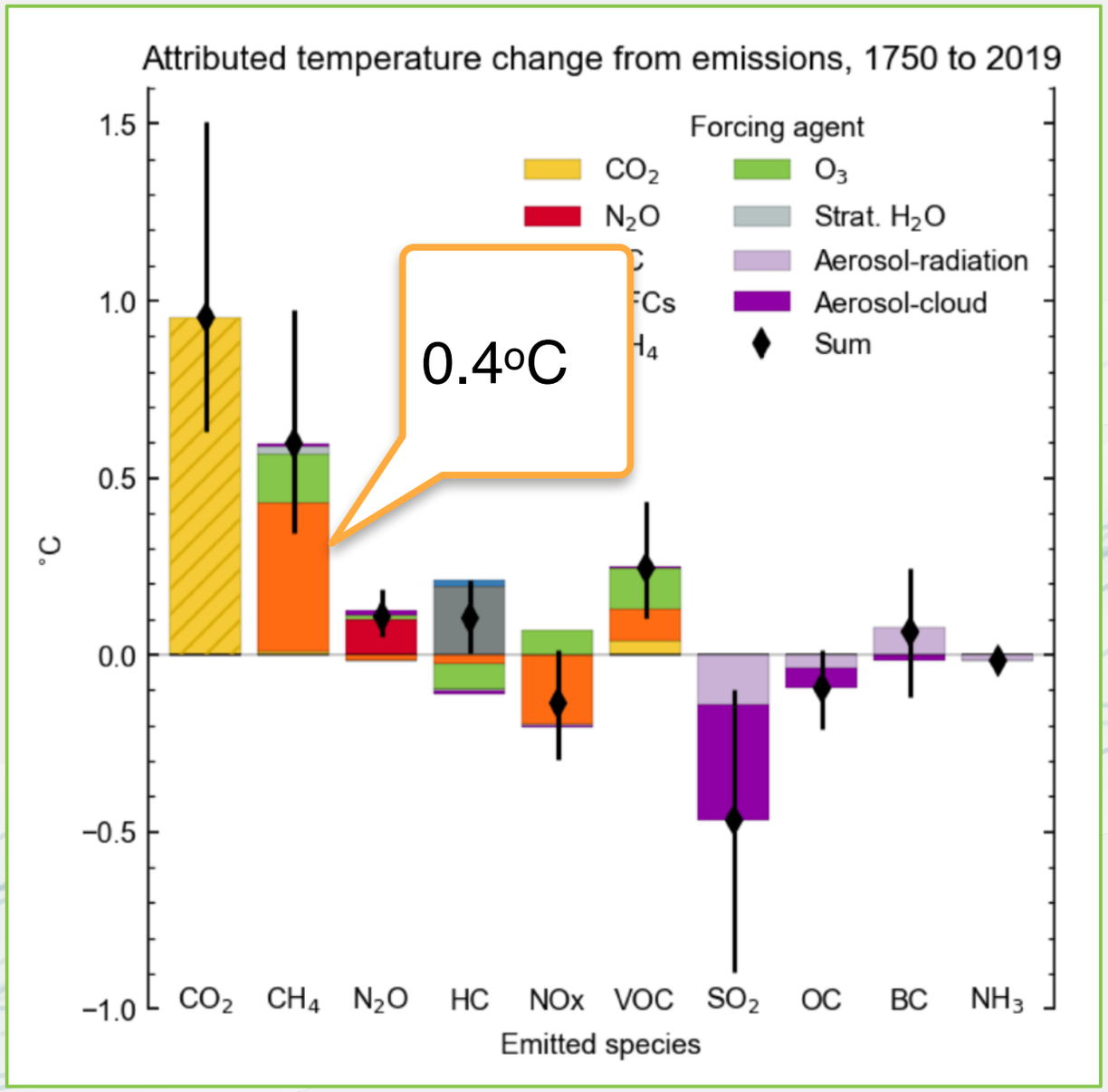

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- It's a greenhouse gas
-

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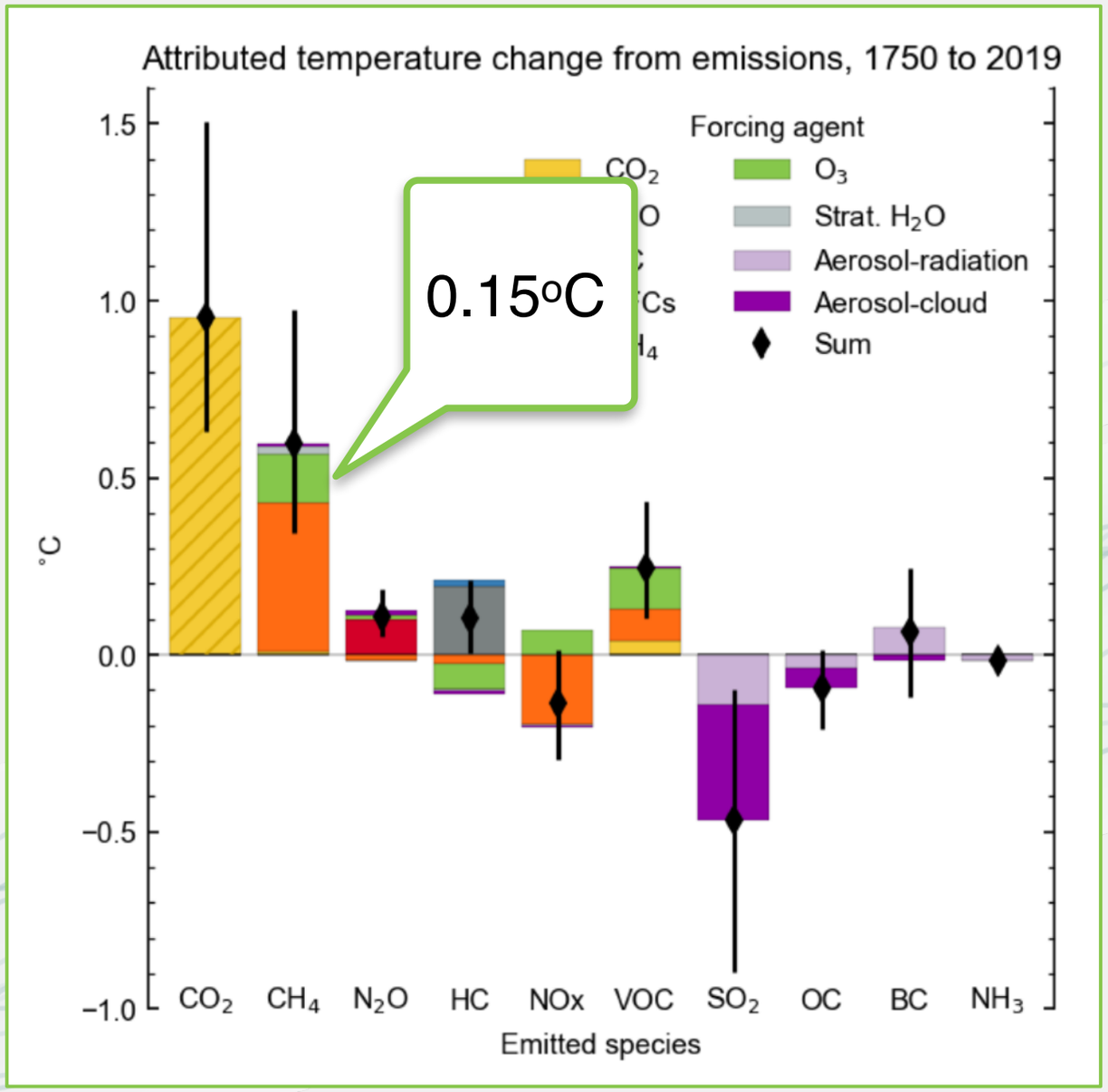

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- It's a greenhouse gas
- But is also an ozone precursor

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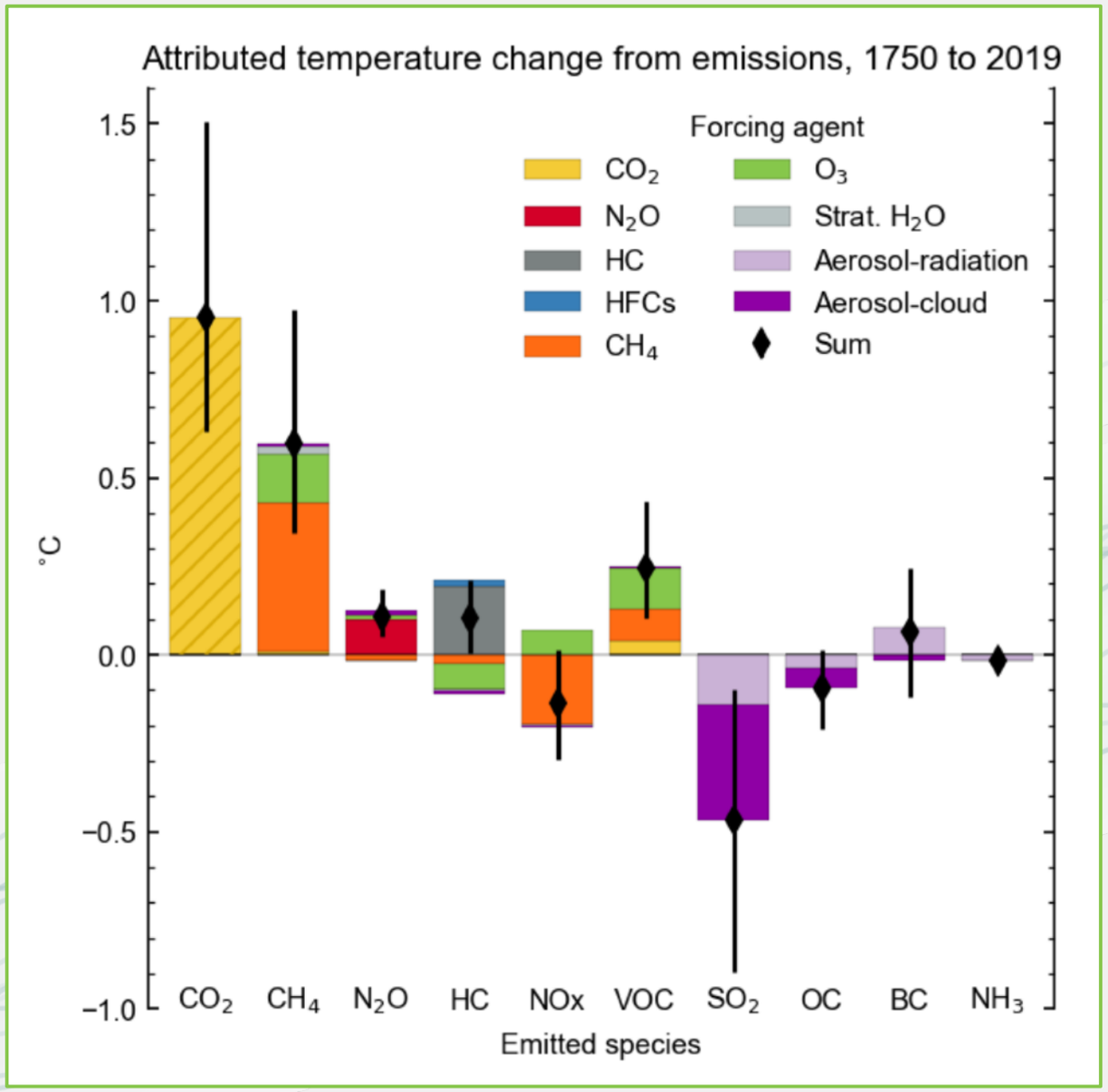

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- Modelling these gases and aerosols is a challenge:
  - Lifetimes variable:
    - CH4 τ~10 years
    - Ozone τ ~30 days
    - Aerosol τ ~ weeks
    - NOx τ ~ days
  - Heterogeneous sources and sinks

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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)

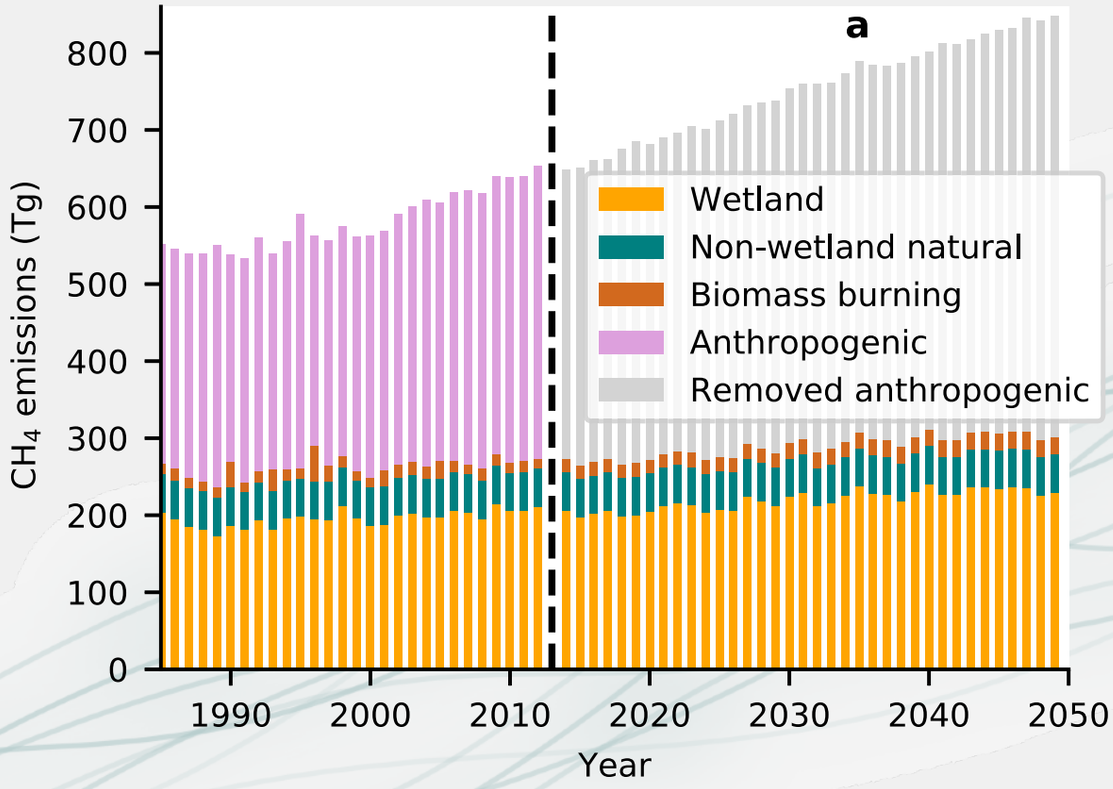
- A large Global Warming Potential – 28 on a 100-year horizon (per-molecule w.r.t. CO_2)
- Large sources – 585 Tg CH_4 per year, with strong chemical sinks. Lifetime of 10 years
- Methane oxidation leads to ozone and water vapour – both greenhouse gases – with methane an important source of stratospheric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

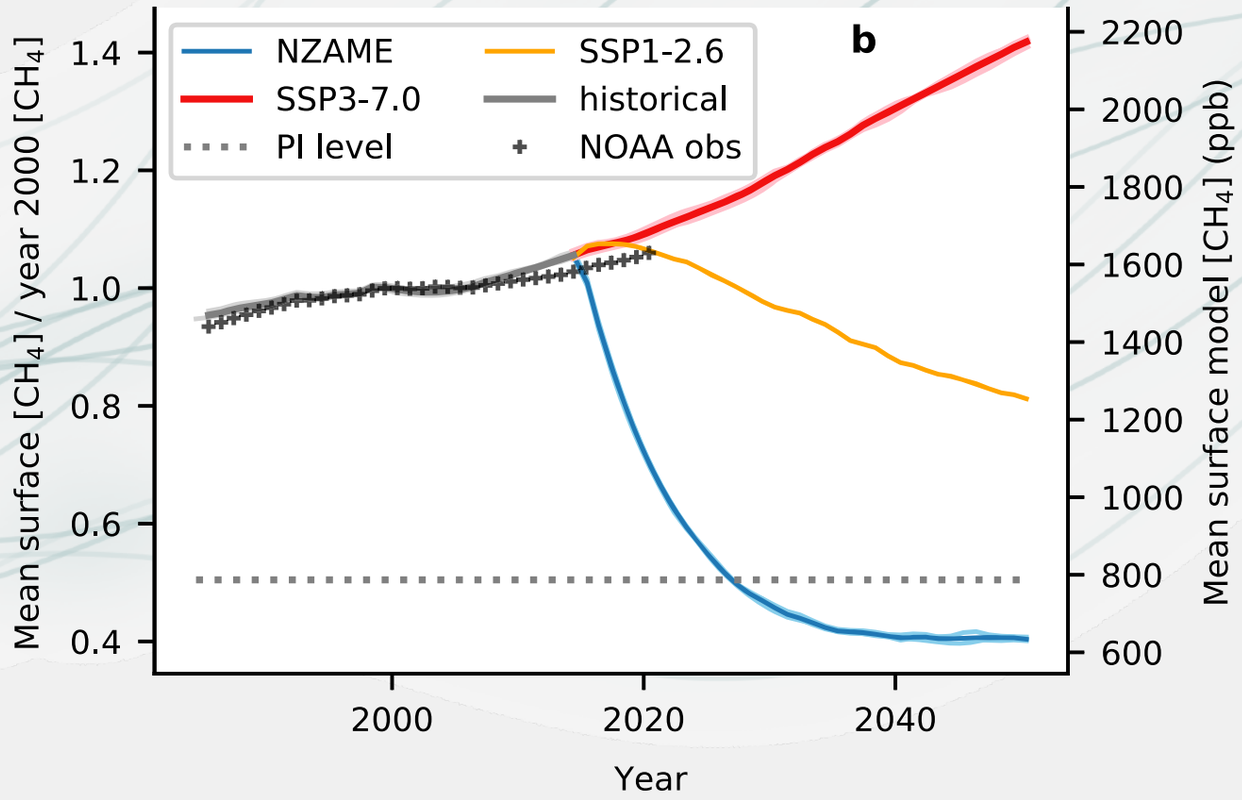
Sources	Wetlands	Fossile fuels gas and coal	Termites	Ruminants	Rice	Waste landfill	Biomass burning
Tg CH_4 per year	177-284	85-105	2-22	87-94	33-40	67-90	32-39

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 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



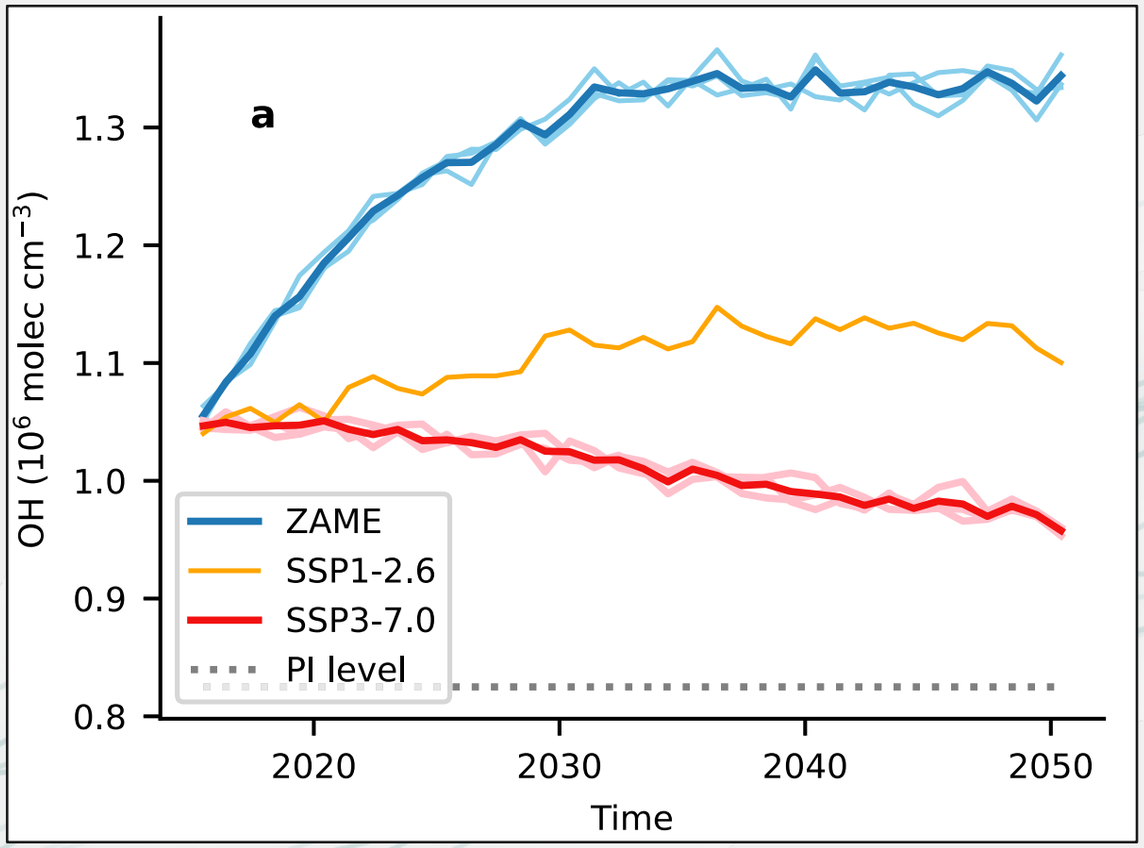


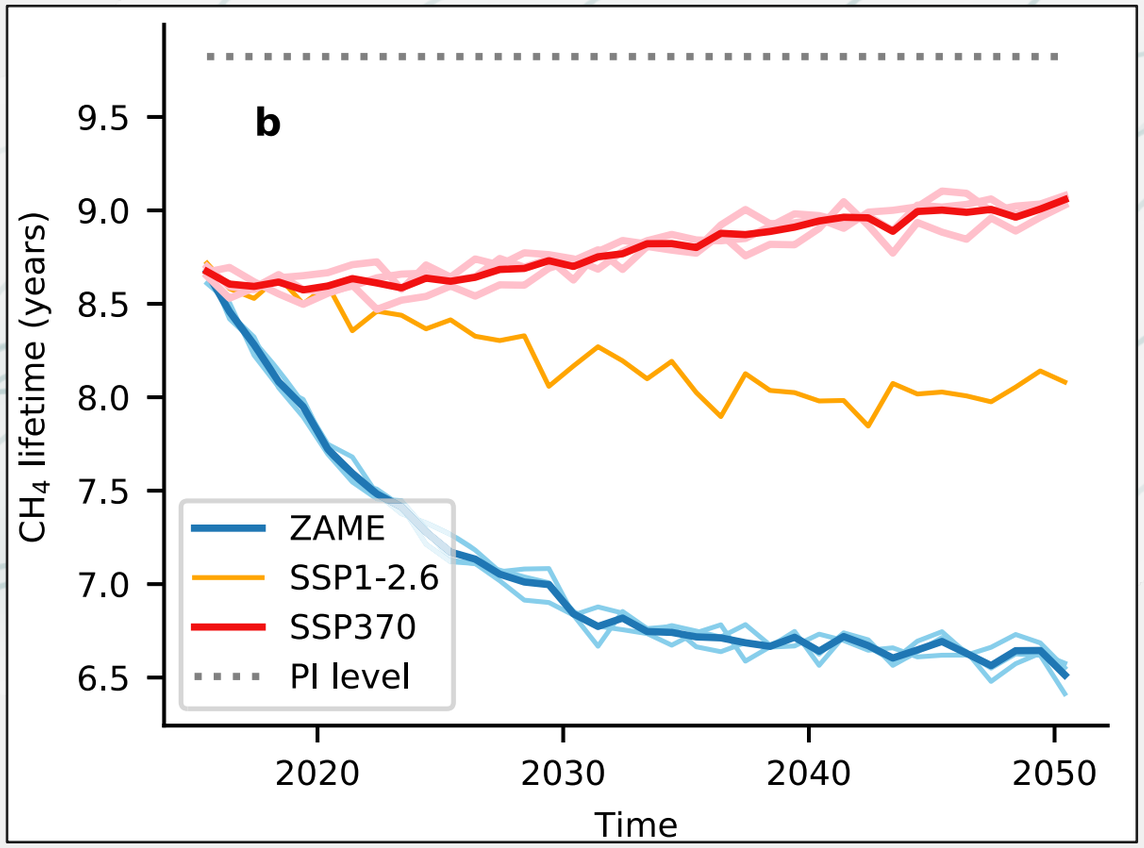
- Comparison with SSP3-7.0 (‘regional rivalry’) and SSP1-2.6 (‘sustainability’) as a counterfactual
  - What are the risks of methane emissions?
  - What are the benefits of constraining future methane emissions?

Figures by Zosia Staniaszek

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

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

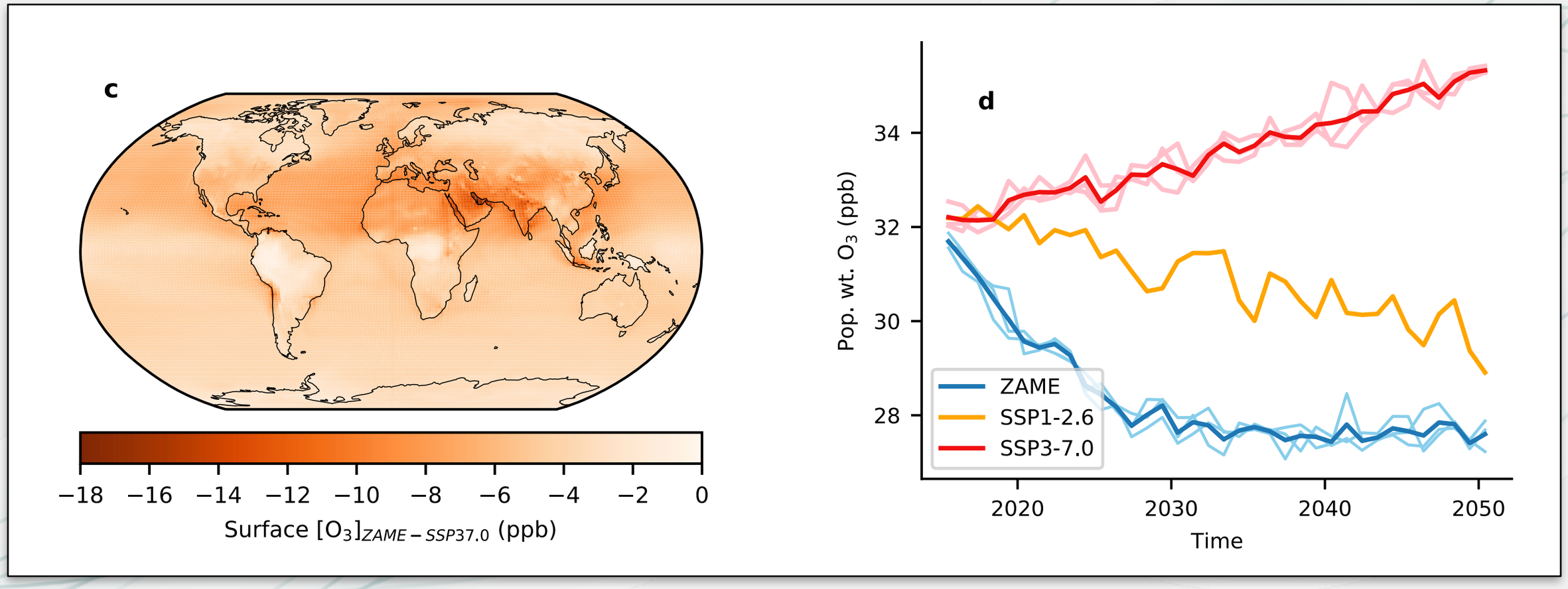




Figures by Zosia Staniaszek

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 in ozone at surface
- Projected decrease in AQ-related mortality of the order of 500k per year

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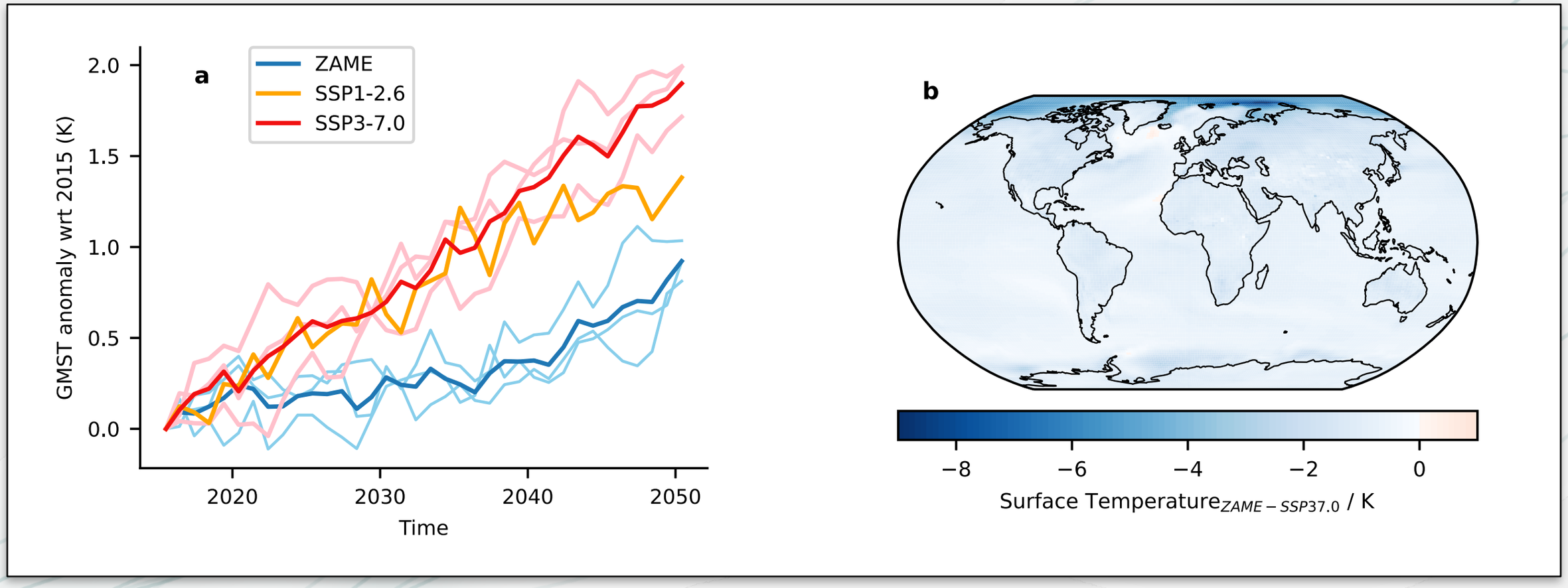
Figures by Zosia Staniaszek

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



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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 anthropogenic methane emissions
  - Produce approx. 0.5°C of global surface temperature rise, depending on SSP
  - Increase tropospheric ozone levels (any improvement in WHO 8hr levels?) with benefits to O3 RF, with consequences for health.
  - Supporess OH - increasing methane lifetime and GWP ('methane self-feedbacks')

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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]✉

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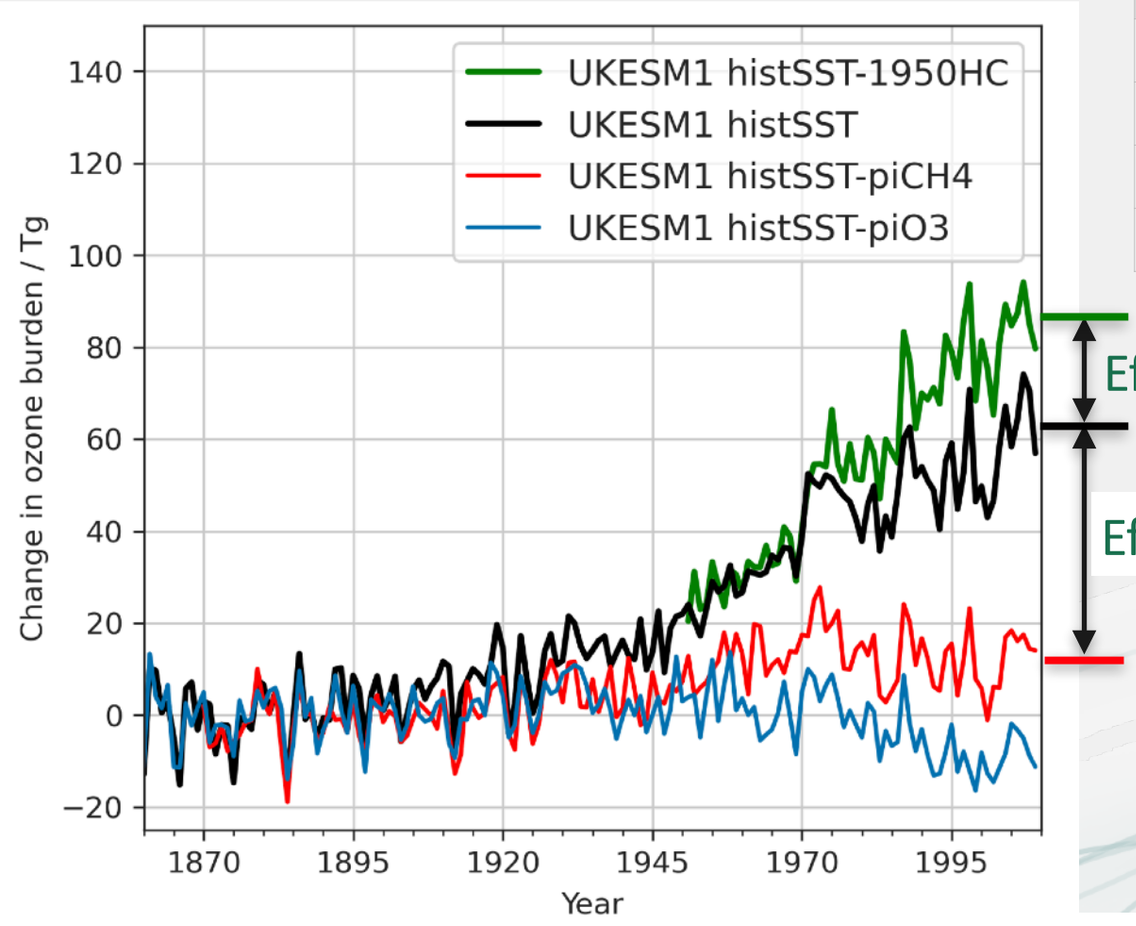
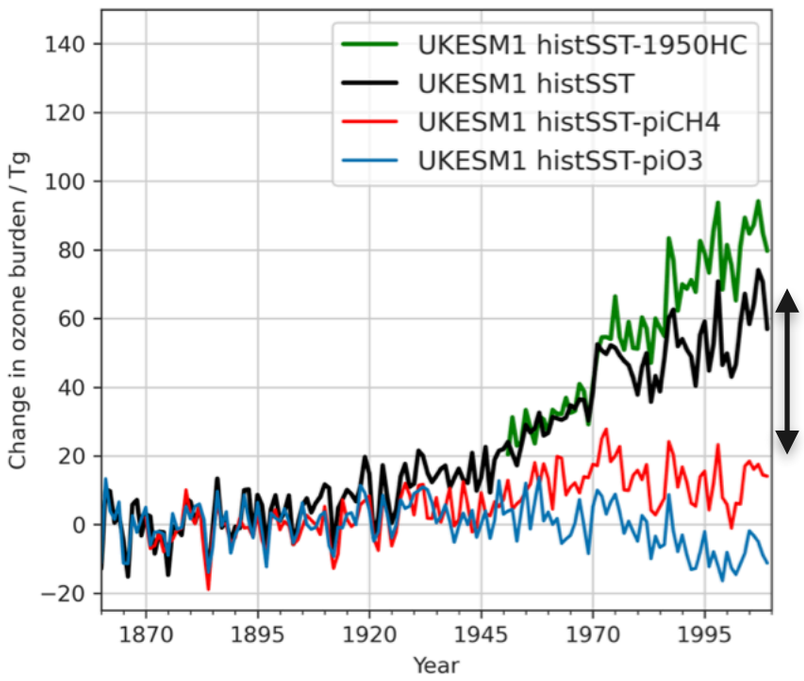
Methane is important to tropospheric ozone - AerChemMIP

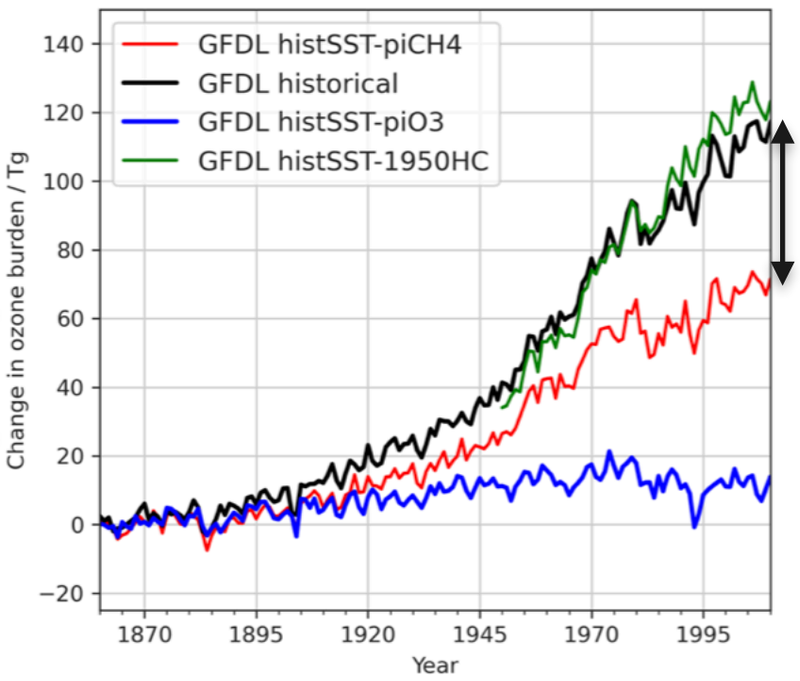
Experiment_ID	CH4	N2O	AERPRE	O3PRE	CFC/HCFC
histSST	Hist	Hist	Hist	Hist	Hist
histSST-piAer	Hist	Hist	1850	Hist	Hist
histSST-piO3	Hist	Hist	Hist	1850	Hist
histSST-piCH4	1850	Hist	Hist	Hist	Hist

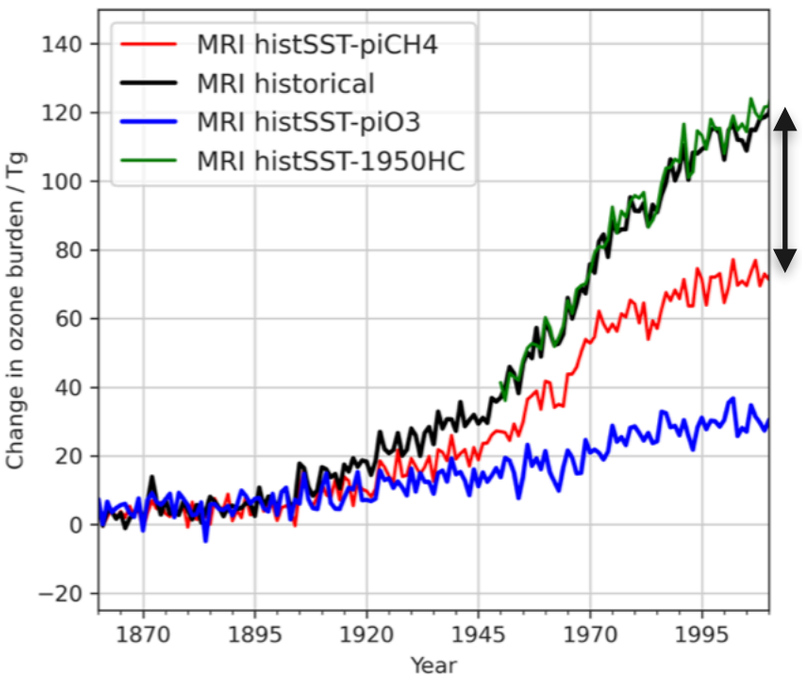


Effect of historical ODS emissions

Effect of historical methane emissions







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

Atmospheric composition and climate impacts of a future hydrogen economy

Nicola J. Warwick[1,2], Alex T. Archibald[1,2], Paul T. Griffiths[1,2], James Keeble[1,2], Fiona M. O'Connor[3,4], John A. Pyle[1,2], and Keith P. Shine[5]

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[2]National Centre for Atmospheric Science (NCAS), University of Cambridge, Cambridge, CB2 1EW, UK
[3]Met Office Hadley Centre, Exeter, EX1 3PB, UK
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- Replacing fossil fuels with H2 - no CO2 emissions during combustion, so cleaner
- More/less NOx (maybe) and leakage of H2 into the atmosphere may be important.
- Various scenarios:
  - Increased H2 usage, so less CH4 consumption
  - Increased H2 leakage, so H2 levels increase
  - Clean H2 - less NOx and CO from combustion - change in O3?
- Goals: Radiative effects, CH4 lifetime, stratospheric impacts

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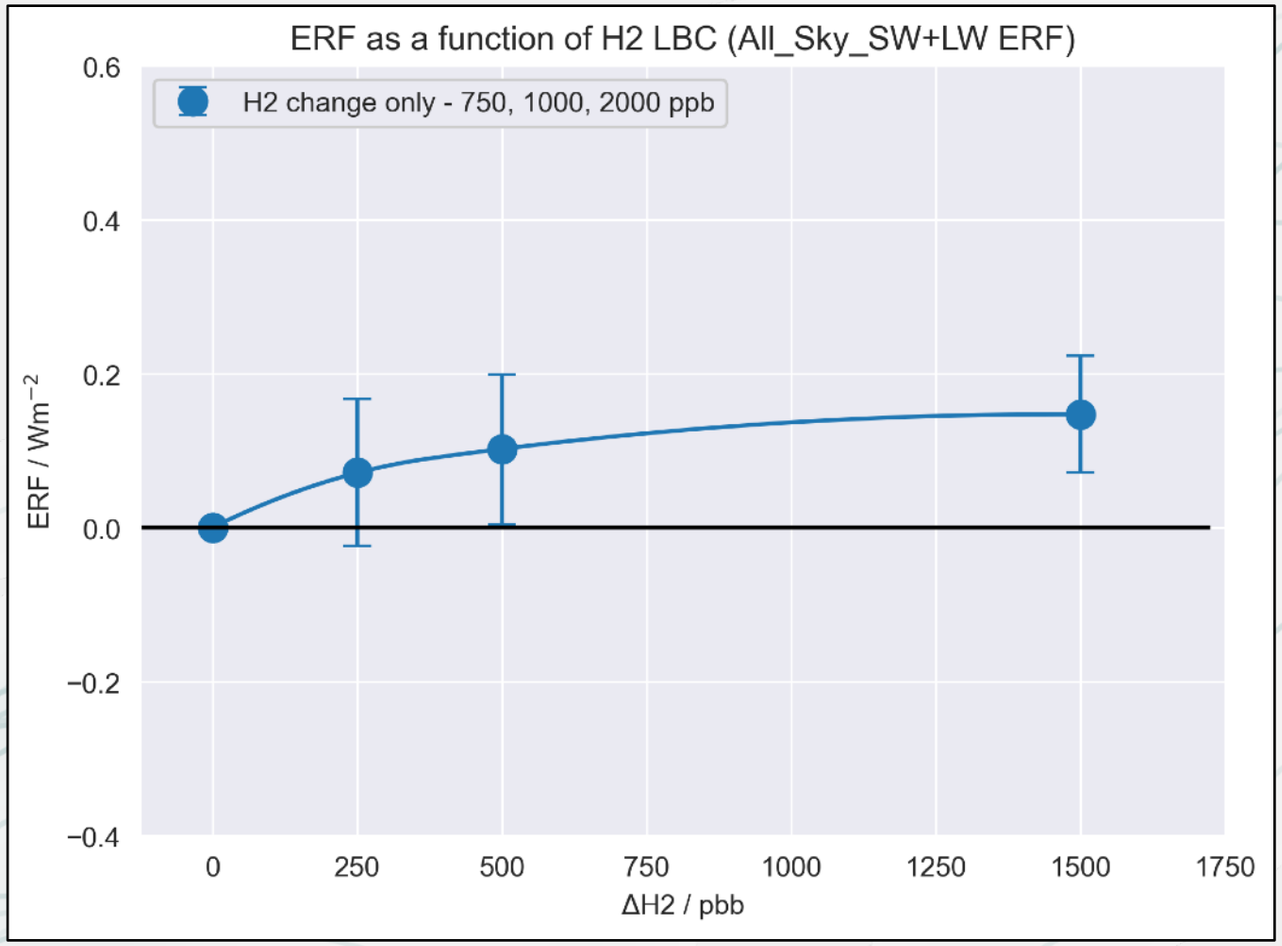
Climate effects of oxidant changes - what is the effect of H2 fugitive emissions?

- Experiments with varying H2 concentration in the atmosphere - various leakage rates.
- 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 - expect positive GG forcing.



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

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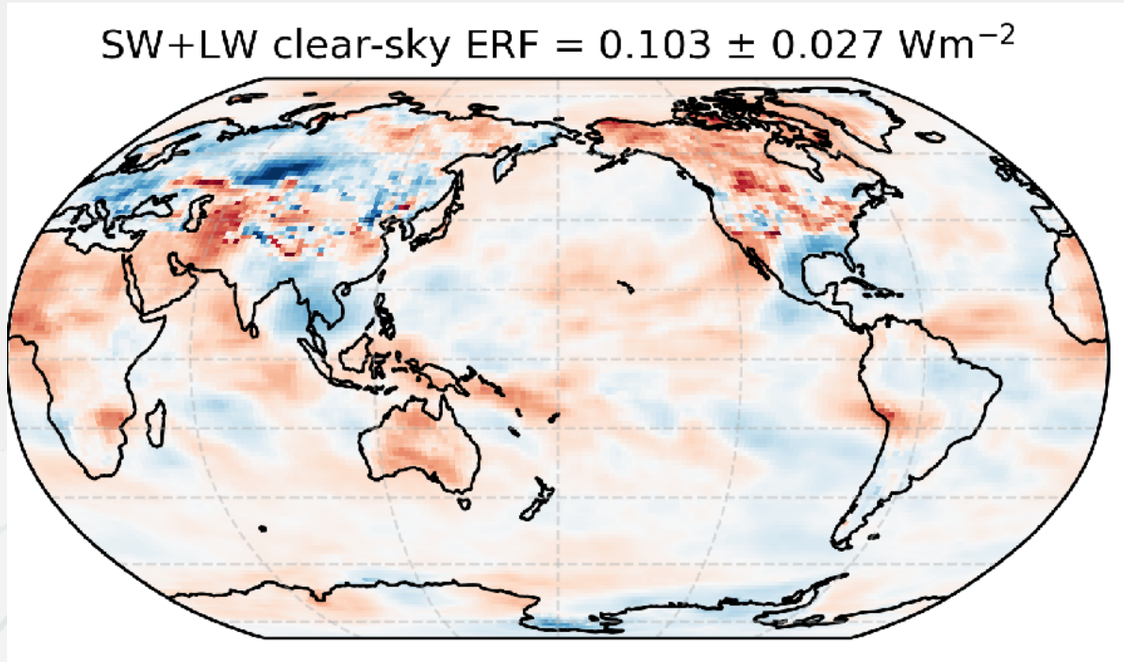
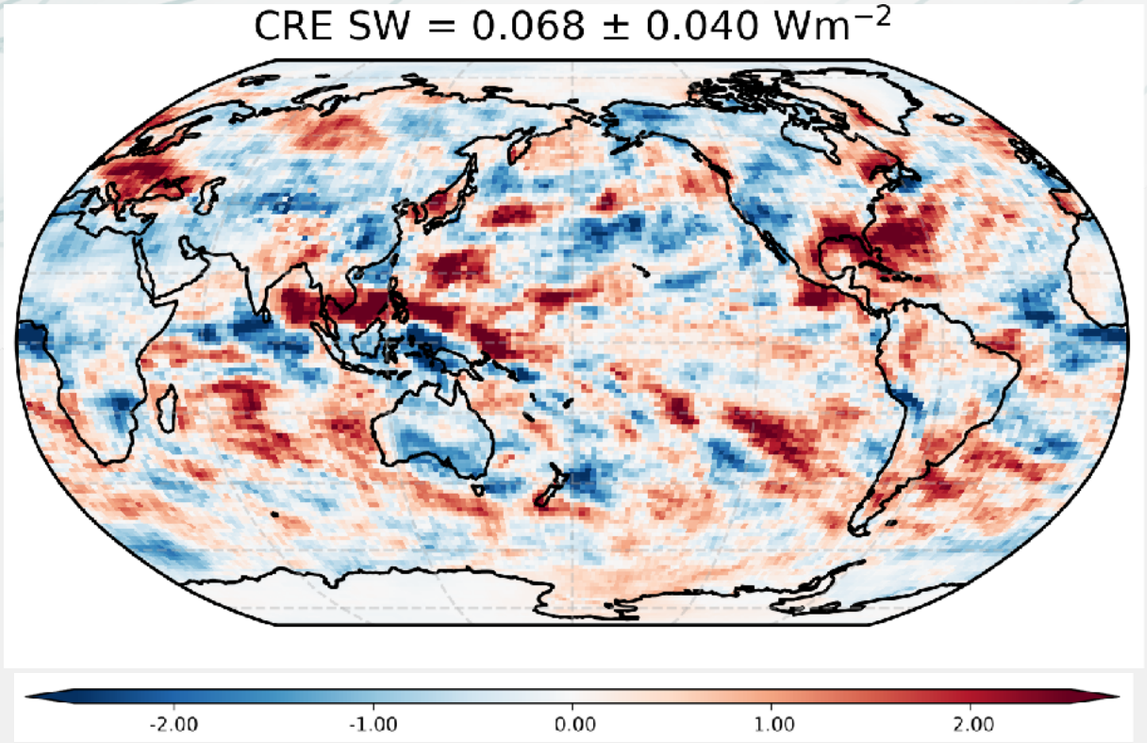
Breaking ERF down into clear-sky and cloud effects

- 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





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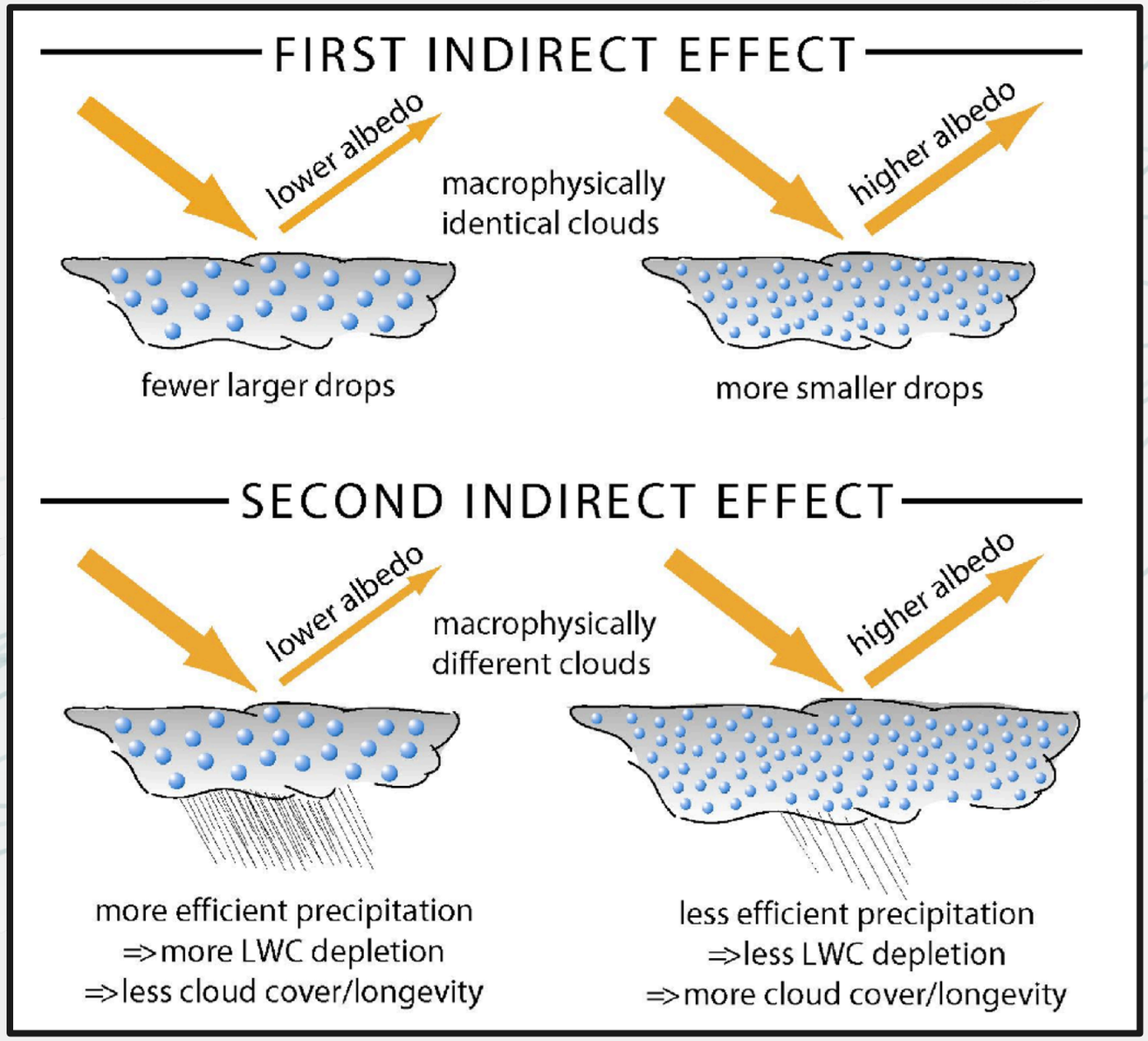
Cloud radiative properties respond to aerosol changes

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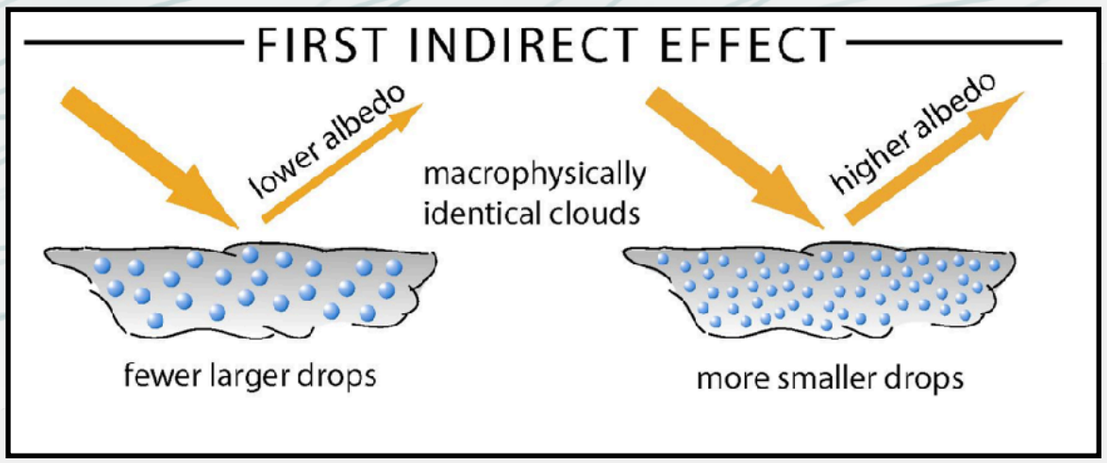
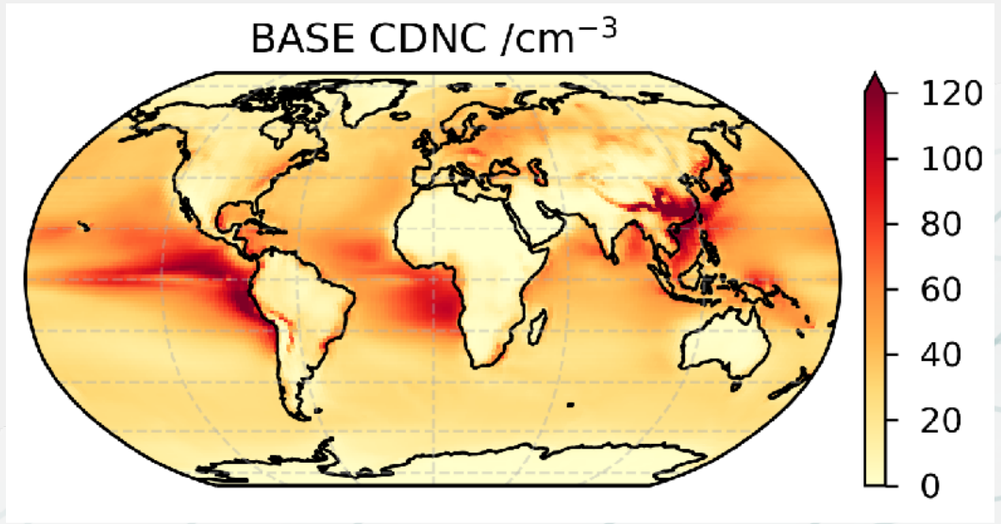
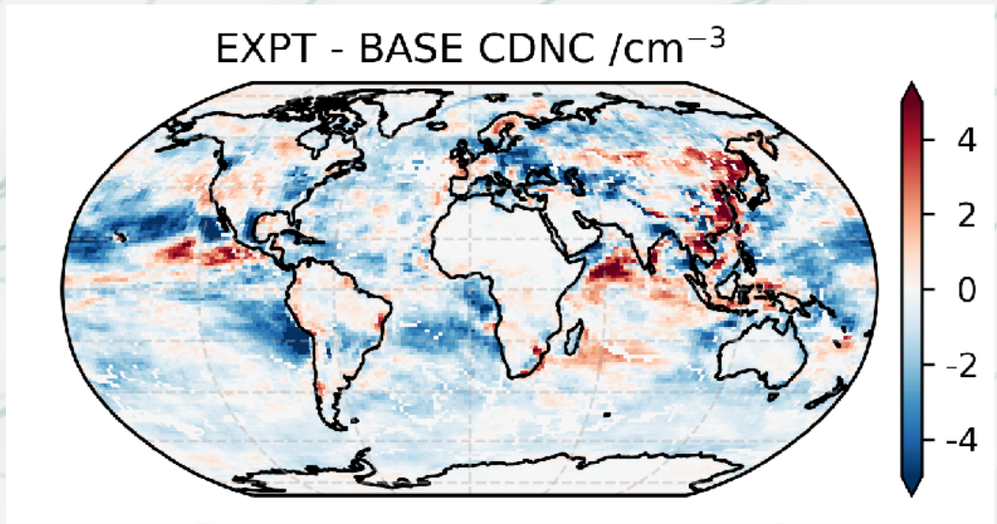
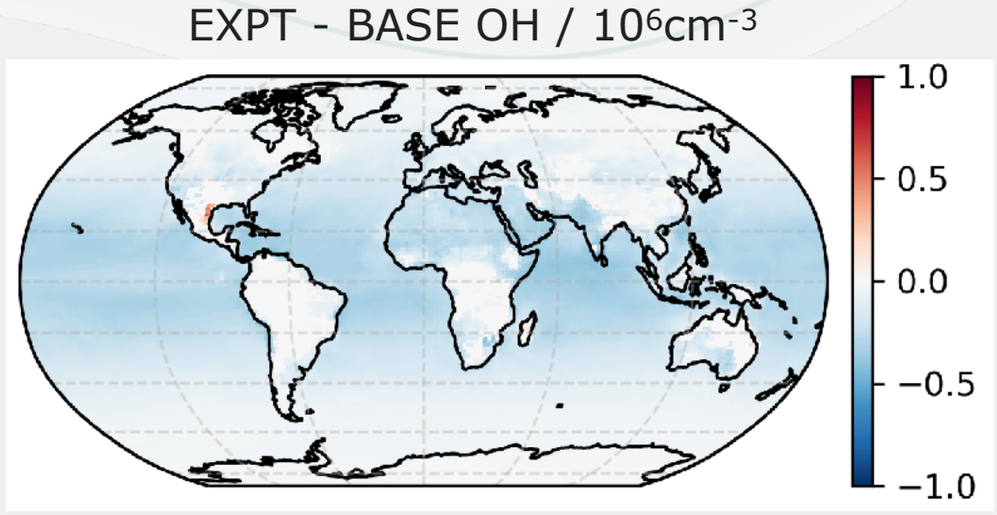
ERF - the coupling of gas phase oxidant to aerosol levels and cloud properties

- OH levels control sulfuric nucleation.
- More H2 → less OH → less nucleation.
- The **additional H2 causes a decrease in cloud droplet number concentration** (CDNC)
- Increased H2 suppresses OH, and this is having knock-on effects on aerosol and on other components (e.g. CH4 and O3).
- Fewer cloud droplets → less reflective cloud →decreased planetary albedo → positive forcing









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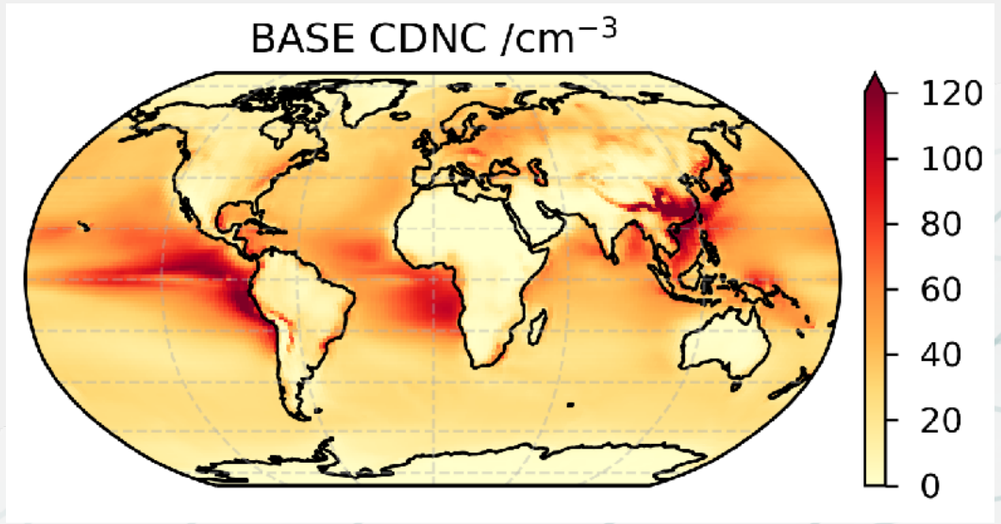

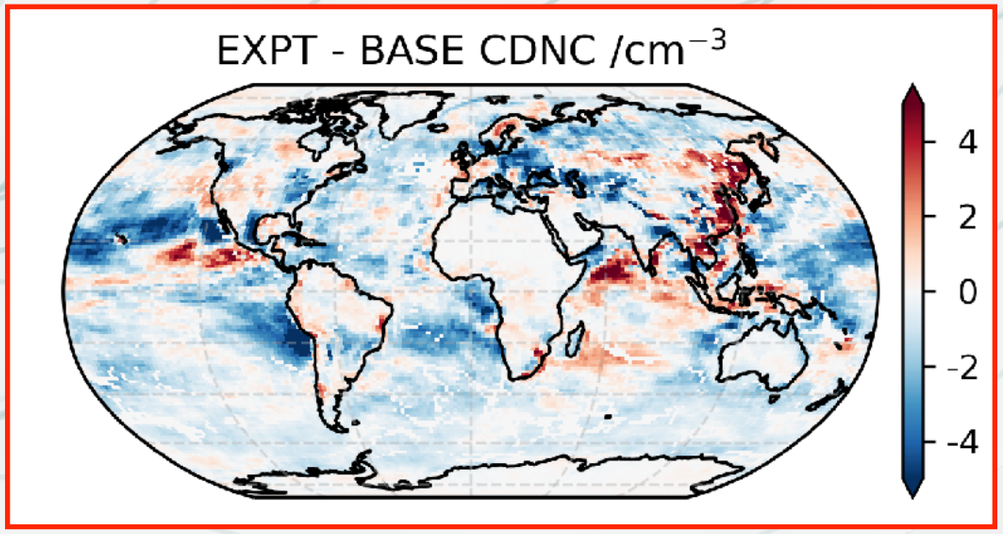

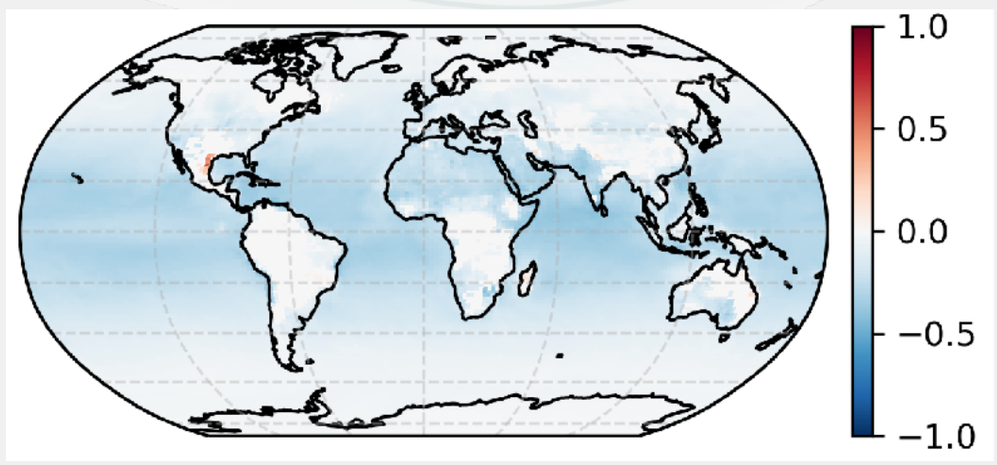

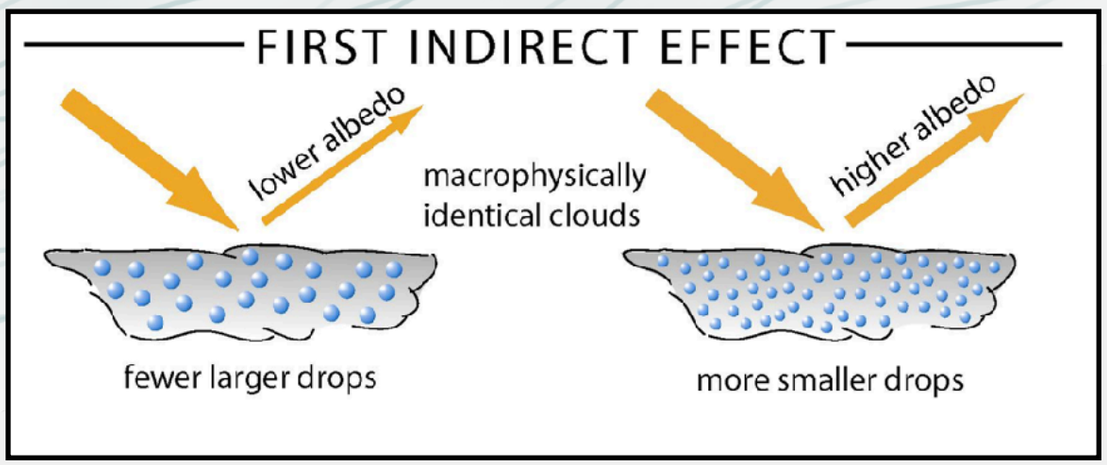

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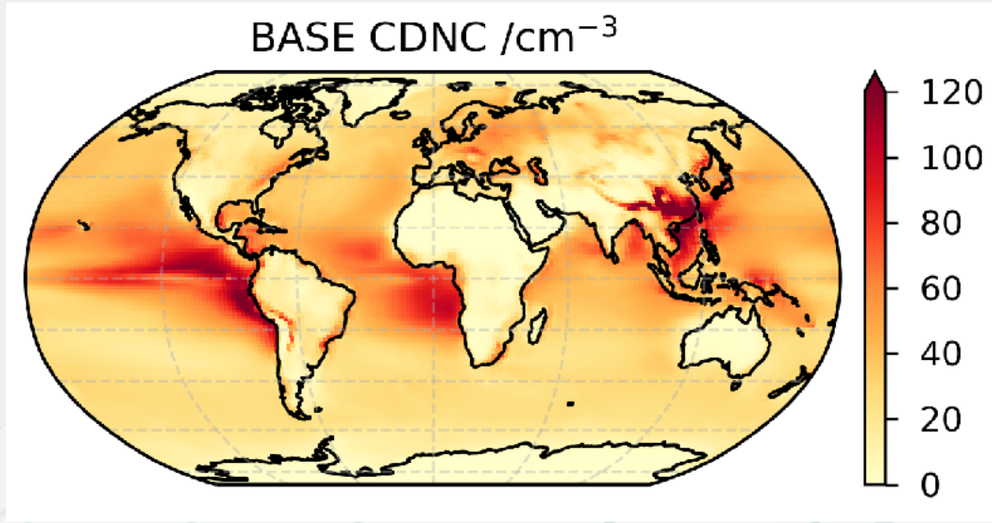
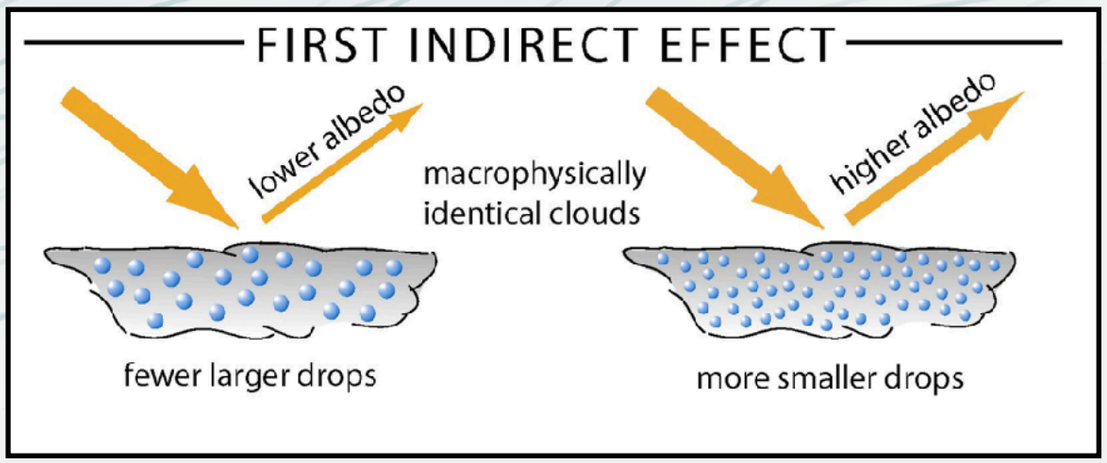




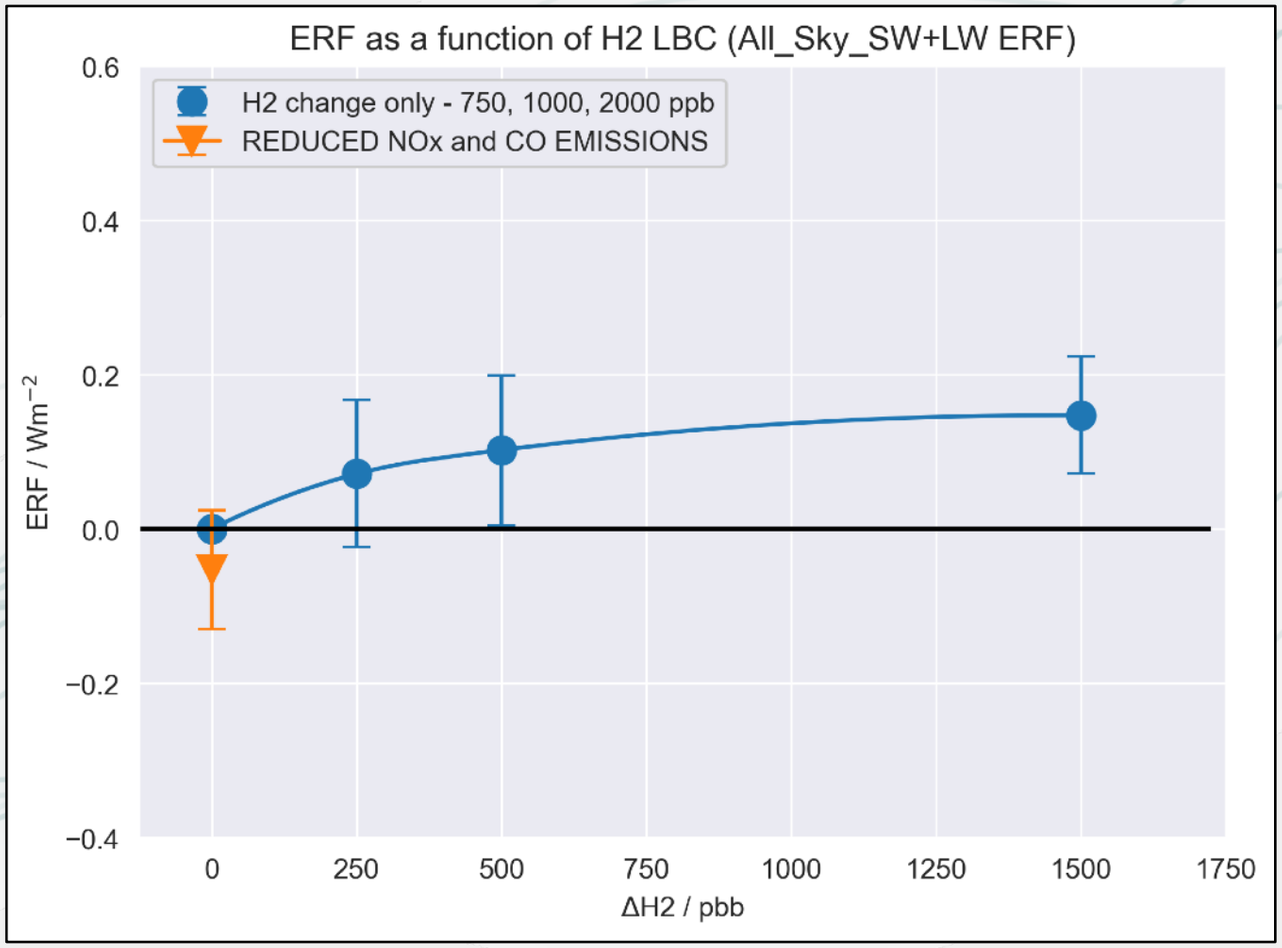
Climate effects of oxidant changes - what is the effect of H2 fugitive emissions?

Expt 2

- A move to H2, and cleaner fuel/combustion.
- No leakages - H2=500ppb

Conclusions

- Lower ozone precursor emissions → a slight negative ERF, since O3 lower.
- Positive climate benefit.



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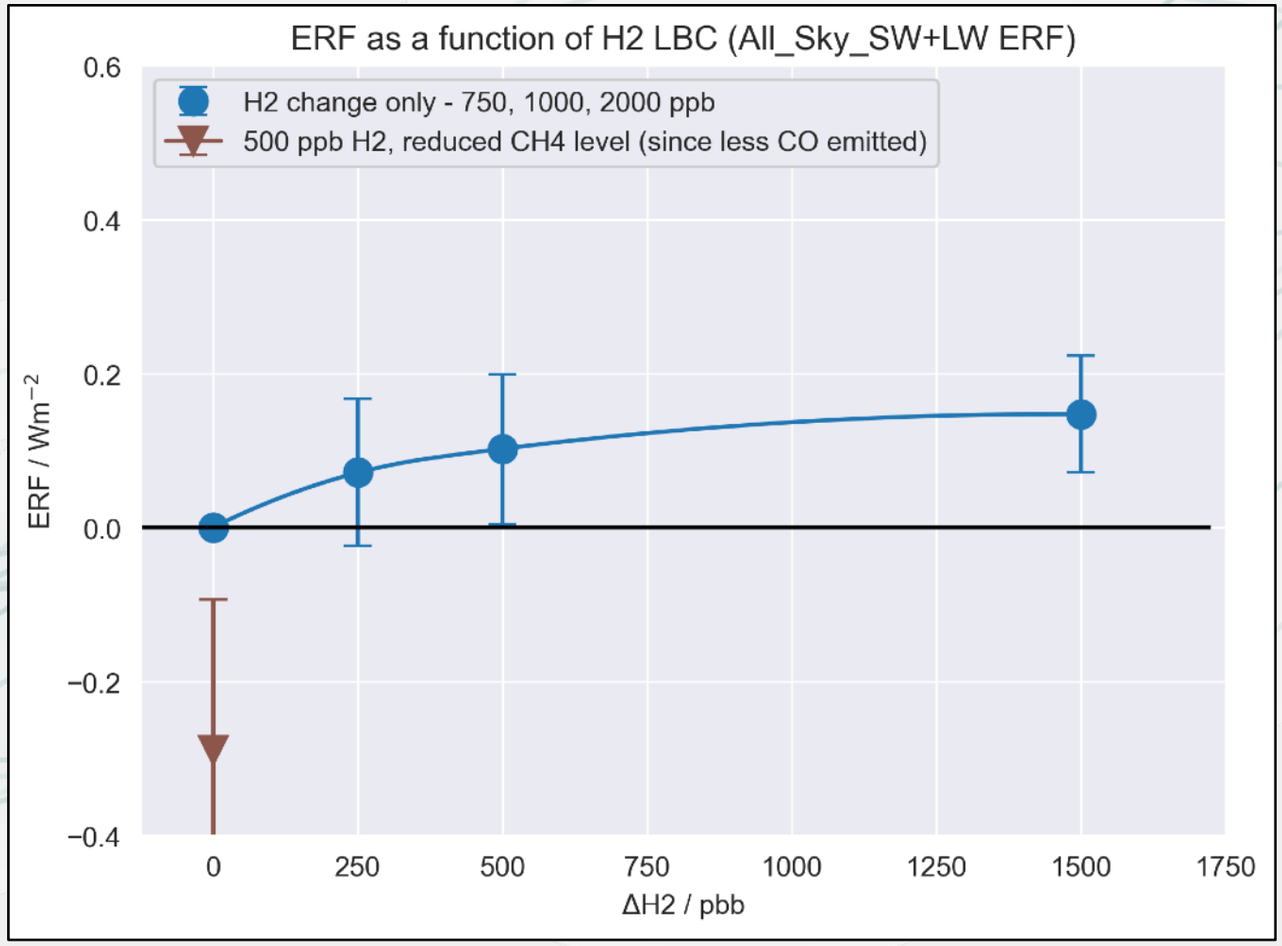
Climate effects of oxidant changes - what is the effect of H2 fugitive emissions?

Expt 3

- A move to H2, and cleaner fuel/combustion.
- No leakages - H2=500ppb
- Less CO emission



Conclusions

- OH levels increase as CO decreases
- Higher OH → lower levels of CH4
- Reduced forcing by CH4
- Positive climate benefit

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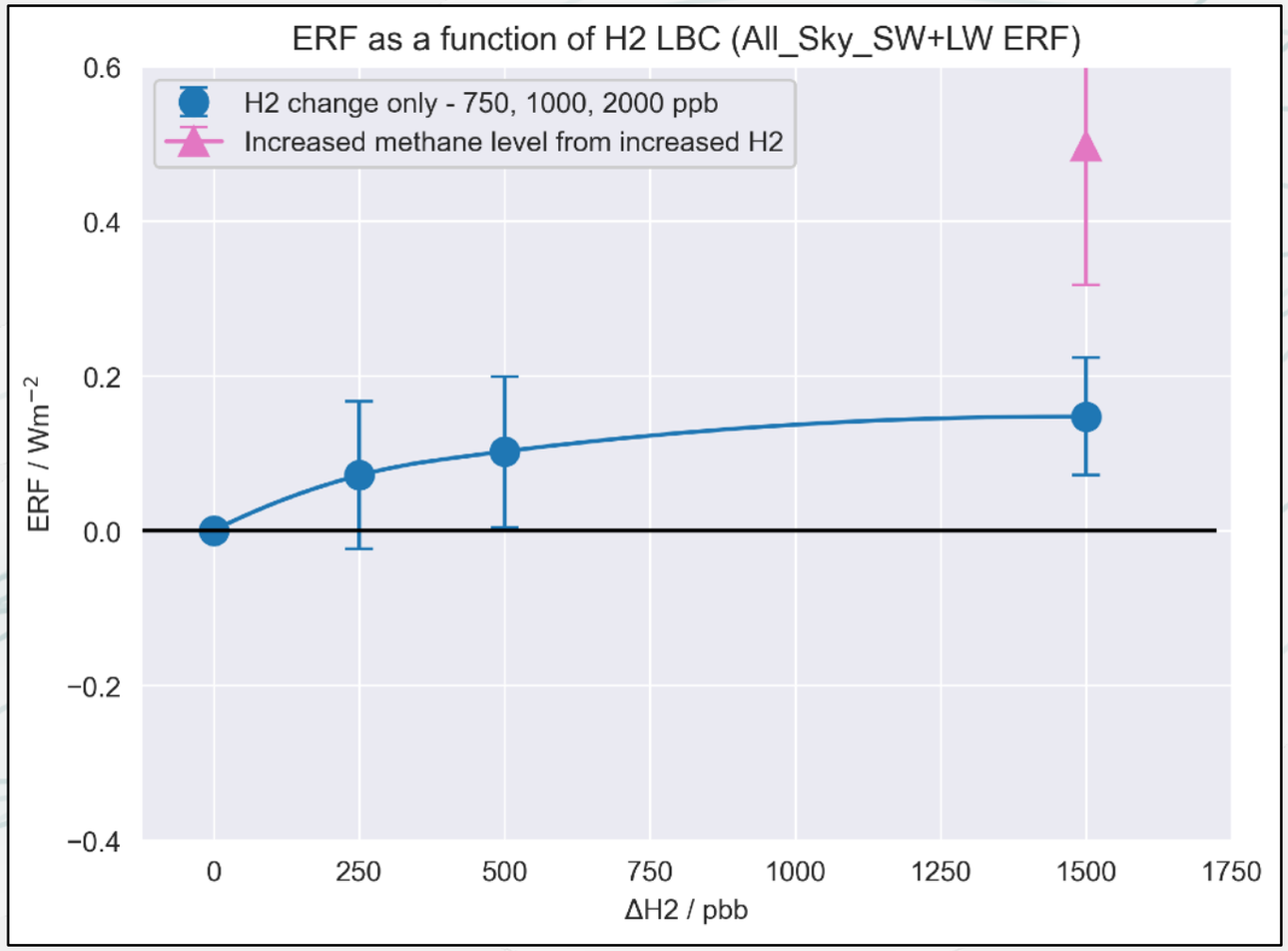
Climate effects of oxidant changes - what is the effect of H2 fugitive emissions?

Expt 4

- Significant leakages H2=2000ppb
- CH4 responds to OH suppression by H2

Conclusions

- OH levels decrease as H2 increases
- Lower OH → higher levels of CH4
- Increased forcing by CH4
- Negative climate benefit



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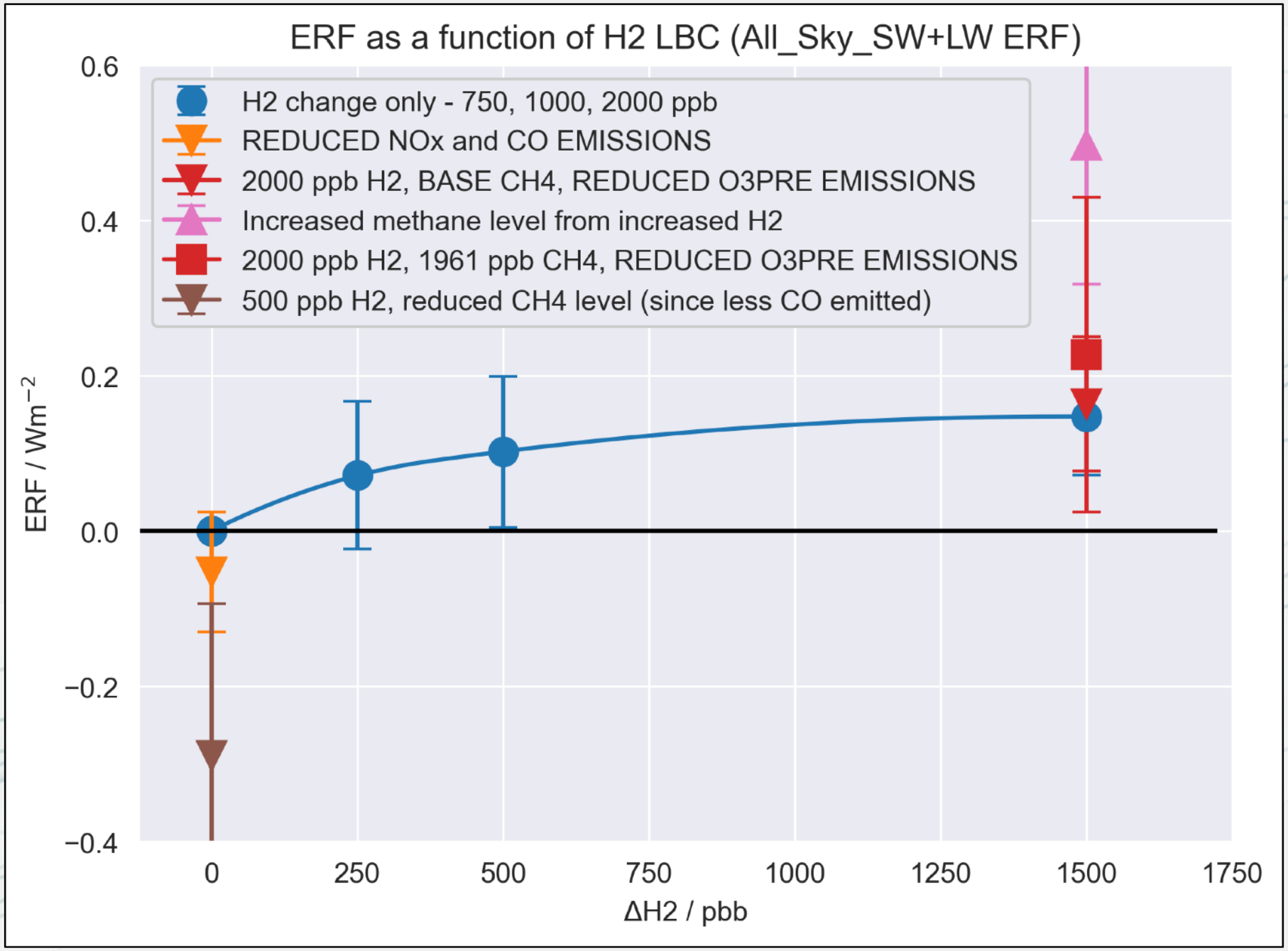
Climate effects of oxidant changes - what is the effect of H2 fugitive emissions?



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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 a number of 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
  - More H2 → less OH → more CH4
- All of these can 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).

Conclusions

- CMIP6 → CMIP7: more emissions-driven models; expect increase model diversity
- Pre-industrial atmosphere is important to PI-PD RF calculations - effort needed to intercompare between models?
- Whole-atmosphere chemistry shows that stratospheric ozone recovery is important to 21st century air quality - The TOAR2 ROSTEES project addresses this using CCMI-2022 and CMIP6 data (James Keeble and Paul Griffiths, leading)
- Other online components important to radiative forcing: LNOx, biogenic VOCs, online aerosol formation.
- Atmospheric chemistry important to the RF of methane,
-

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Takeaways

- Ozone is produced and destroyed in large amounts in the troposphere: responds similarly to emissions changes.
- Climate change drives significant changes in chemistry, ozone levels
- 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
- Atmospheric chemistry important for
  - GHG lifetime and GWP
  - Air pollution at the surface
  - Oxidant-aerosol coupling and cloud radiative impacts

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