

Present Day Forcing by Methane (CH₄)

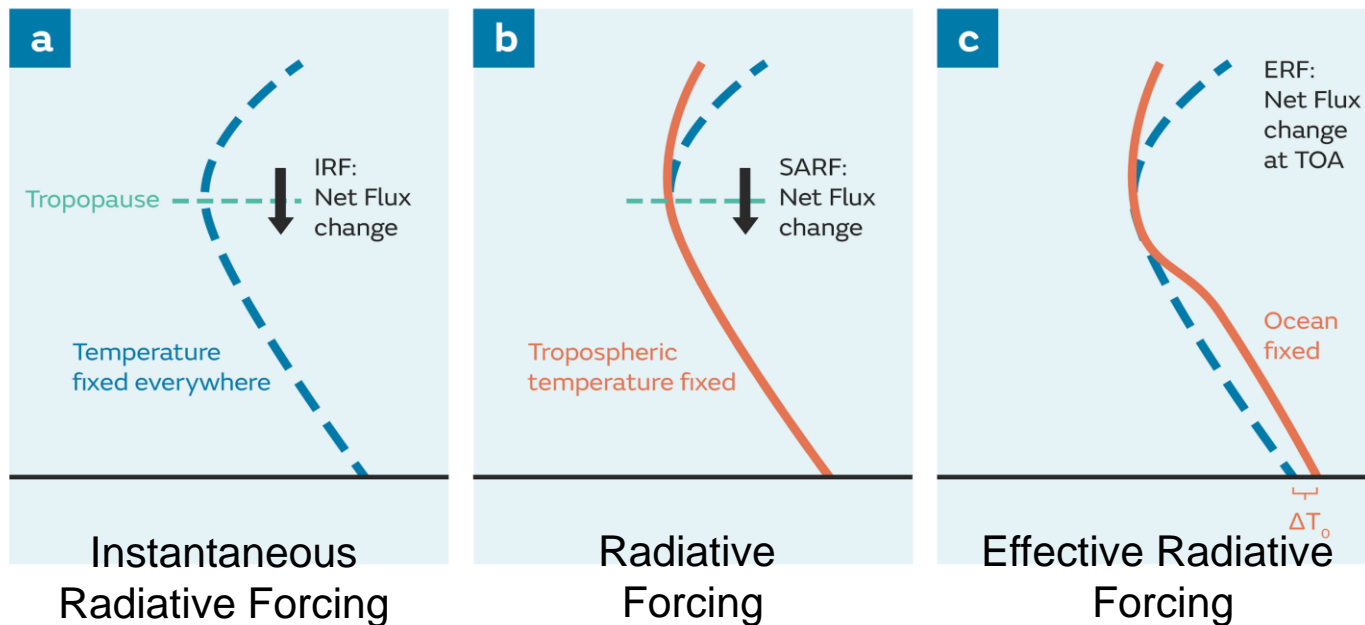
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CRESCENDO Final General Assembly, 15 March 2021

Motivation (1)



- Better characterization of changes to the Earth's radiative budget since the pre-industrial through co-ordinated experiments within RFMIP and AerChemMIP
- Use of the effective radiative forcing as the forcing metric of choice



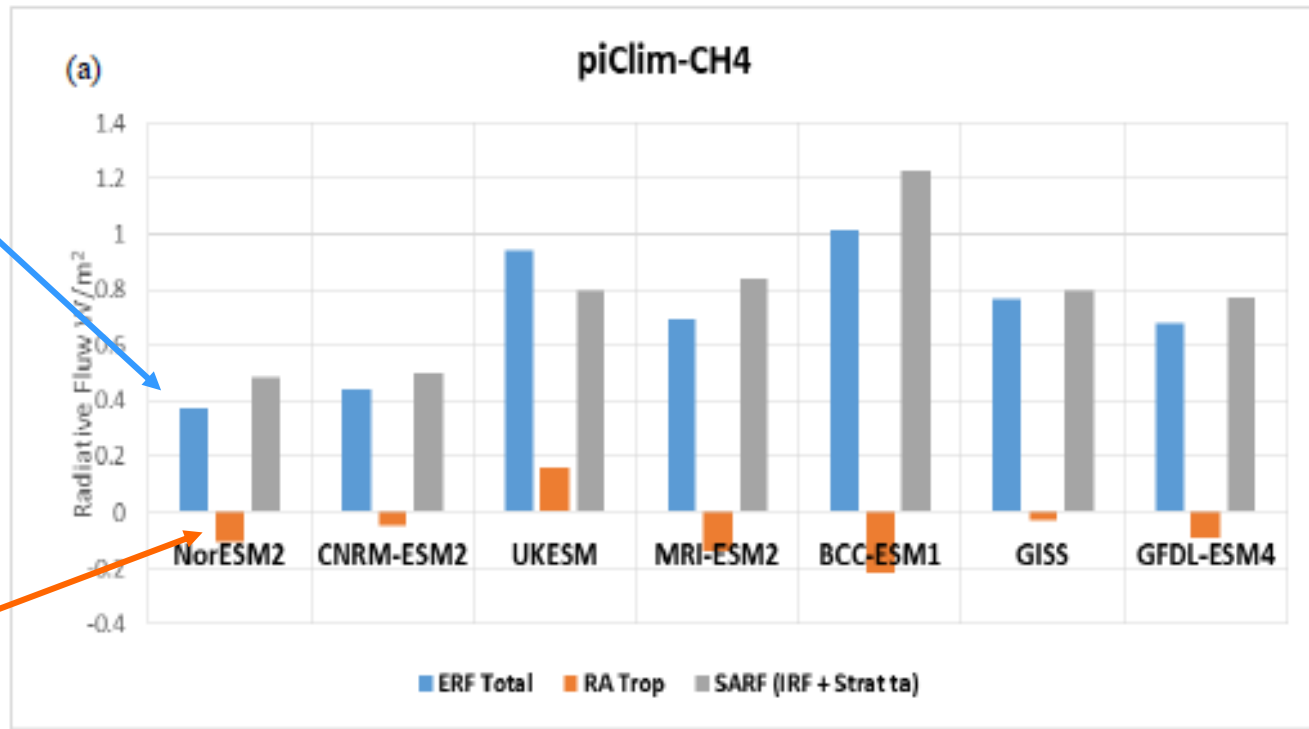
ERF is easier to diagnose in global climate models and is more representative of the eventual temperature response

$$ERF = IRF + \sum_i A_i$$

Motivation (2)

ERF

$\sum_{i, Trop} A_i$



Thornhill et al., Atmos. Chem. Phys. (2021)

- Sum of the tropospheric adjustments in all models are negative except UKESM1 – most likely due to the positive cloud adjustment in UKESM1
- To provide a process-based understanding of the positive cloud adjustment in UKESM1

Methane ERF Breakdown



Perturbation	NET	LW' _{CS}	SW' _{CS}	LW CRE'	SW CRE'	NET' _{CS}	NET CRE'
ΔCH_4	0.97 ± 0.04	0.74 ± 0.02	0.11 ± 0.02	-0.39 ± 0.02	0.50 ± 0.02	0.85 ± 0.03	0.12 ± 0.02

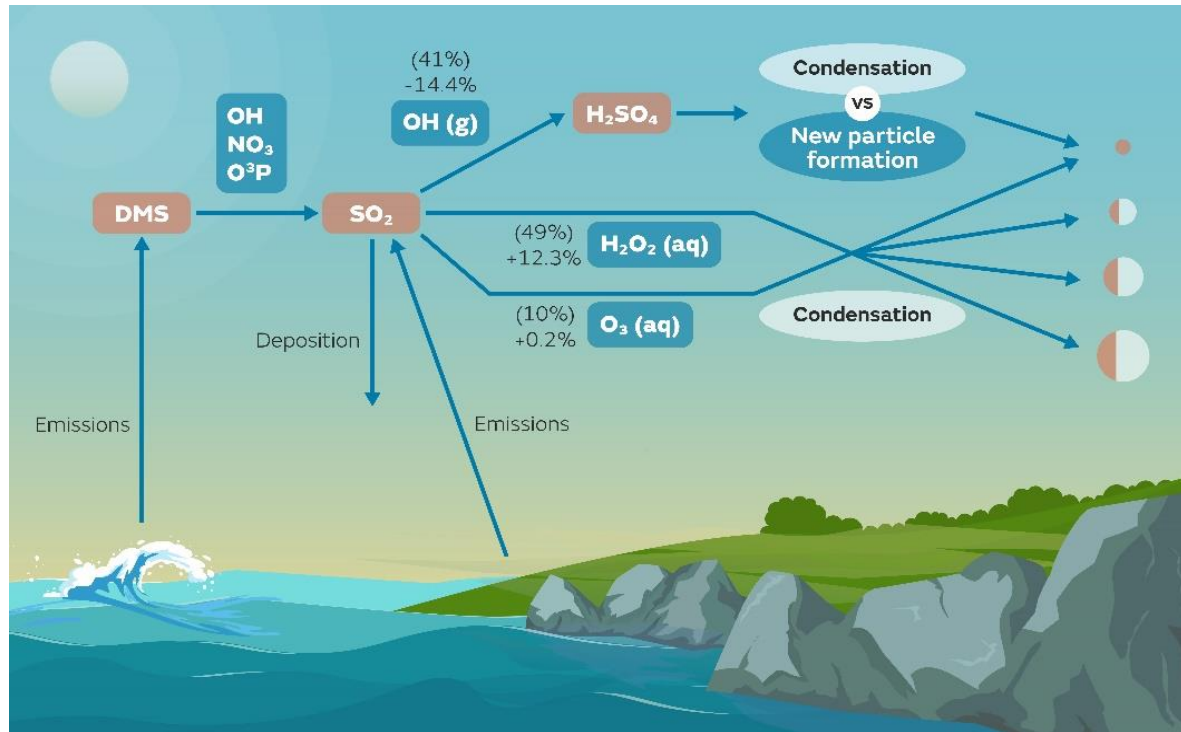
Change in CRE diagnosed as recommended in Ghan (2013)

Units: W m^{-2}

Active Forcing Agents and/or Interactions	LW' _{CS}	SW' _{CS}	LW CRE'	SW CRE'	NET CRE'
CH ₄ , O ₃ , H ₂ O, ARI, ACI	0.74 ± 0.02	0.11 ± 0.02	-0.39 ± 0.02	0.50 ± 0.02	0.12 ± 0.02
CH ₄ , O ₃ , H ₂ O, ARI	0.72 ± 0.03	0.11 ± 0.02	-0.38 ± 0.02	0.25 ± 0.03	-0.14 ± 0.03

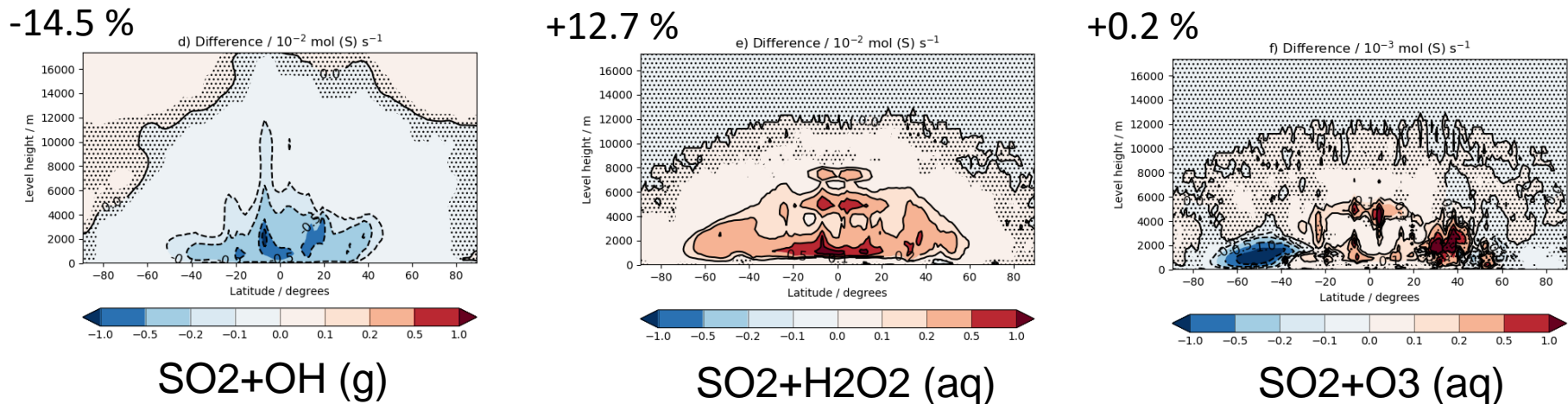
- Using additional paired simulations, the **change in the CRE in UKESM1 is only positive when aerosol-cloud interactions are active**

Sulphate Aerosol Formation



Schematic of sulphate aerosol formation in pre-industrial atmosphere

Stippling here indicates changes that are **not** significant at the 95% confidence level

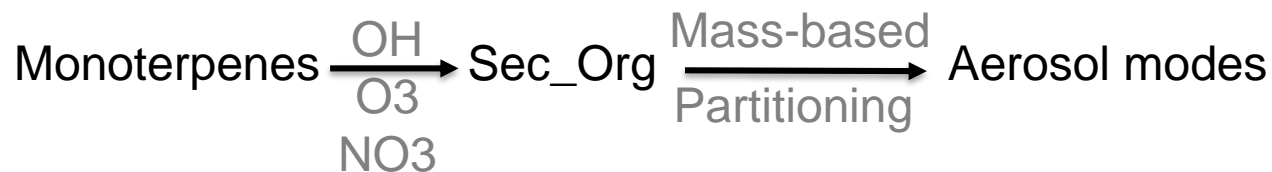
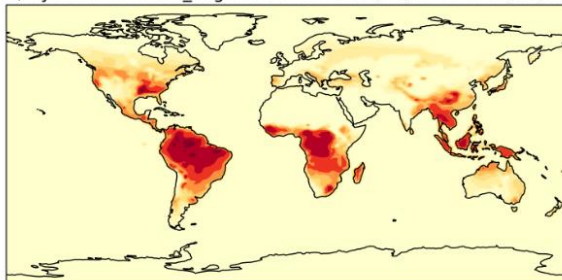


Biogenic SOA Formation

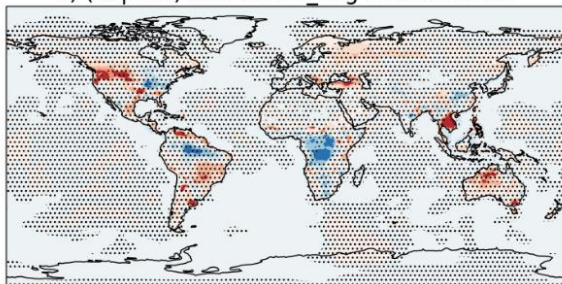


Species	Simulation	Production (Tg/yr)		Loss (Tg/yr)		Burden (Tg)
		Primary	Secondary	Dry	Wet	
OM	<i>piClim-control</i>	49.25 ± 0.01	38.24 ± 0.37	17.79 ± 0.14	69.35 ± 0.32	1.28 ± 0.02
	<i>piClim-CH4</i>	49.25 ± 0.01	38.35 ± 0.30	17.78 ± 0.12	69.46 ± 0.24	1.28 ± 0.02

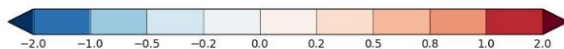
a) by903: Mean Sec_Org cond. from 1865/01/01 to 1894/12/30



c) (Exp-Ctl) diff. in Sec_Org condensation



Diff. in Sec_Org condensation rate (mol OM/s)



Global mean SOA production unchanged despite less condensation onto nucleation & Aitken modes

Aerosol Response

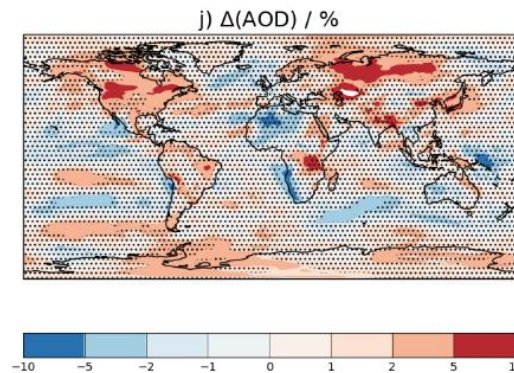
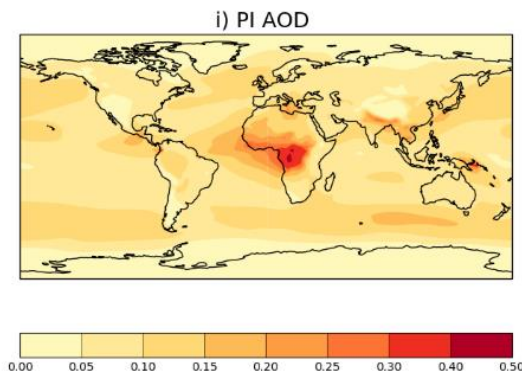
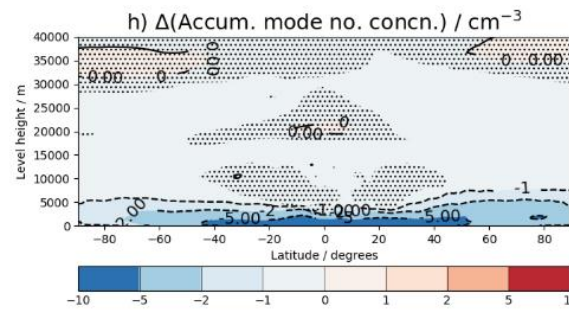
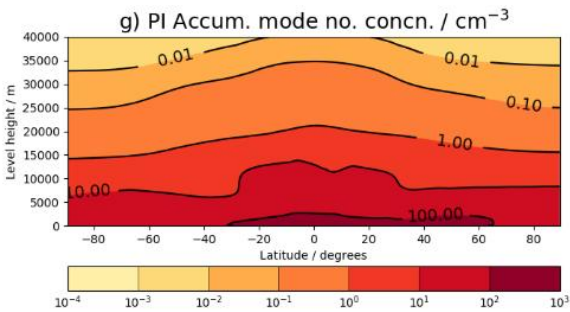
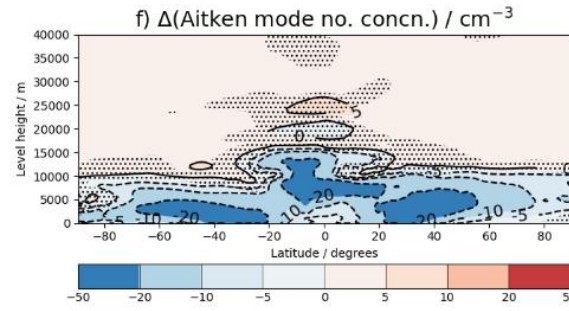
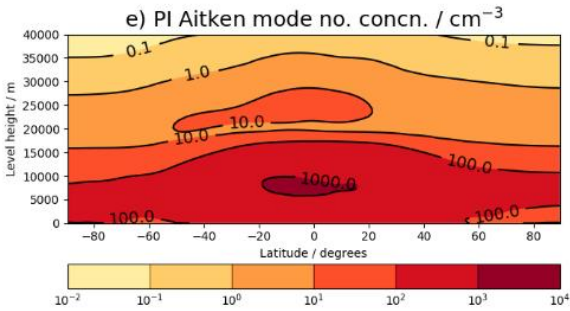


Aitken mode no. concn.

Accum. mode no. concn.

Aerosol optical depth (AOD)

Stippling here indicates changes that are *not* significant at the 95% confidence level

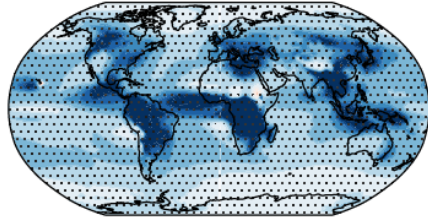


Global mean changes in aerosol mass & AOD are near-zero, but aerosol size distribution changes

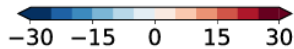
Aerosol & Cloud Response



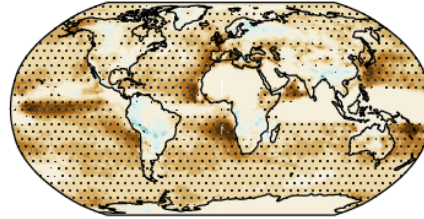
(a) ΔN_{50} (ish) (cm^{-3})



$\bar{\Delta} = -13.7$



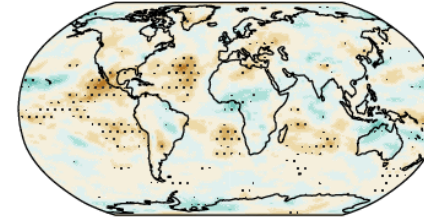
(b) Δ Cloud-top CDNC (cm^{-3})



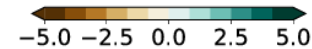
$\bar{\Delta} = -2.49$



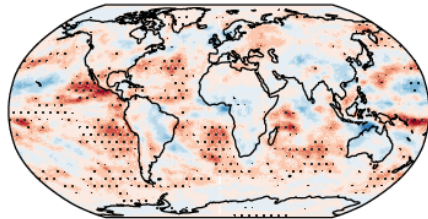
(c) Δ Total cloud fraction (%)



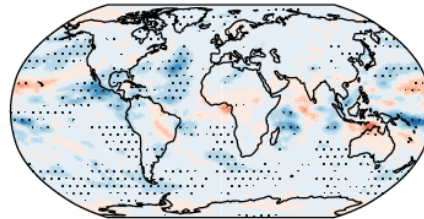
$\bar{\Delta} = -0.203$



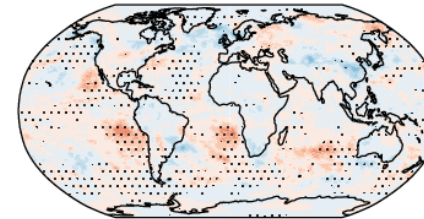
(d) Δ SW cloud rad effect (W/m^2)



(e) Δ LW cloud rad effect (W/m^2)



(f) Δ NET cloud rad effect (W/m^2)



Stippling indicates changes that are significant at the 95% confidence level

- Little evidence that the positive cloud adjustment is dynamically-driven
- Local to regional changes in CRE correspond mainly with changes in cloud fraction
- The global mean CRE is $\sim 0.12 \text{ Wm}^{-2}$ more positive: +ve SW outweighs -ve LW
- Cloud Droplet Number Concentration (CDNC) decline is making the SW CRE generally more positive

Conclusions



- Methane ERF from UKESM1 is one of the highest of the AerChemMIP models
- Partly due to the inclusion of tropospheric chemistry (e.g., O₃)
- Partly due to the tropospheric adjustments being positive (Cloud adjustment)

We find:

- Little evidence that the cloud adjustment in UKESM1 is dynamically-driven
- Positive cloud adjustment is aerosol-mediated
- Overall reduction in cloud fraction & the positive LW CRE outweighs the negative SW CRE
- Changes in SO₂ oxidation pathways lead to a reduction in nucleation, a shift in aerosol size distribution and a reduction in CDNC