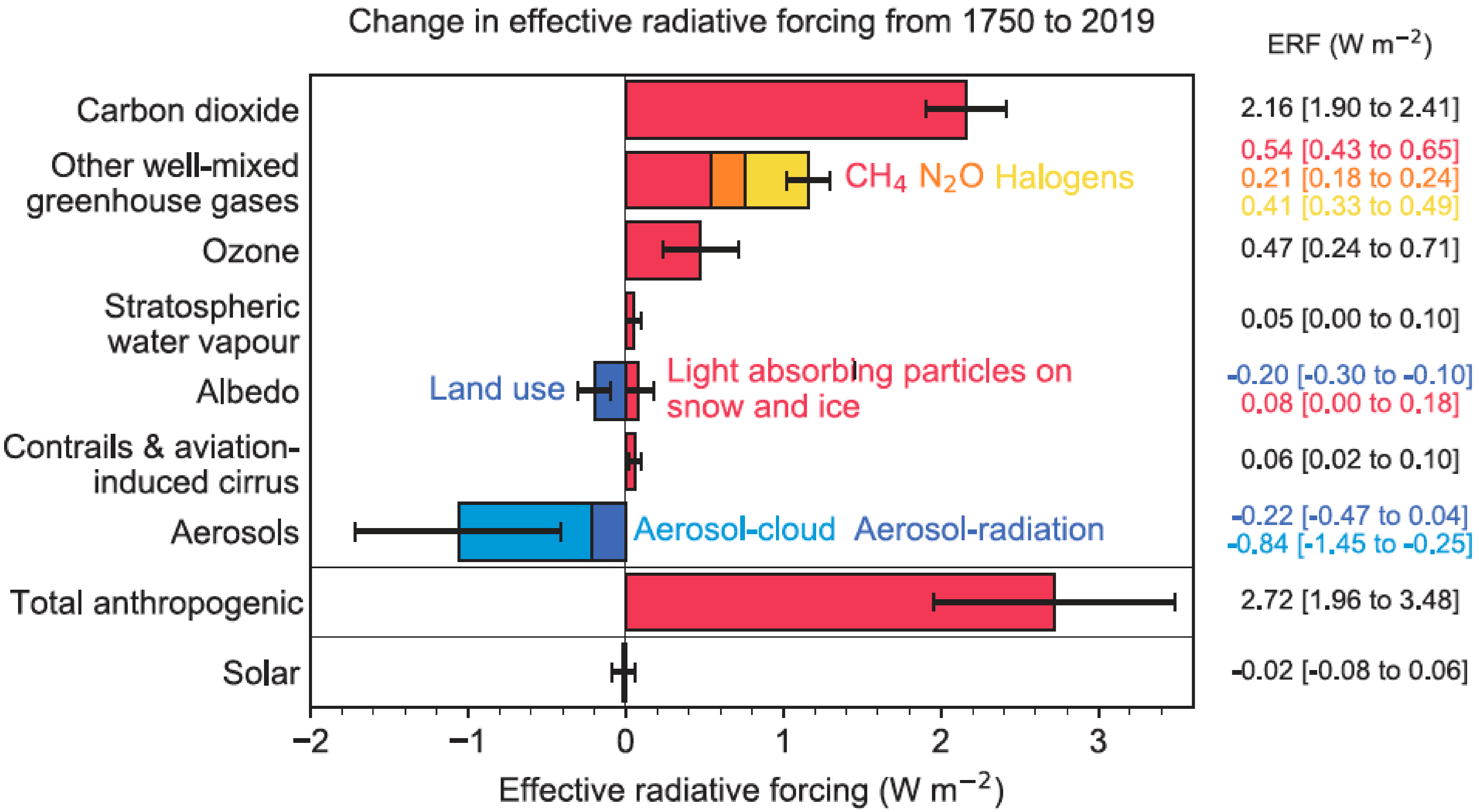
Which aspects of aerosol physics are currently most limiting for understanding aerosol impacts on climate?

October 23, 2022

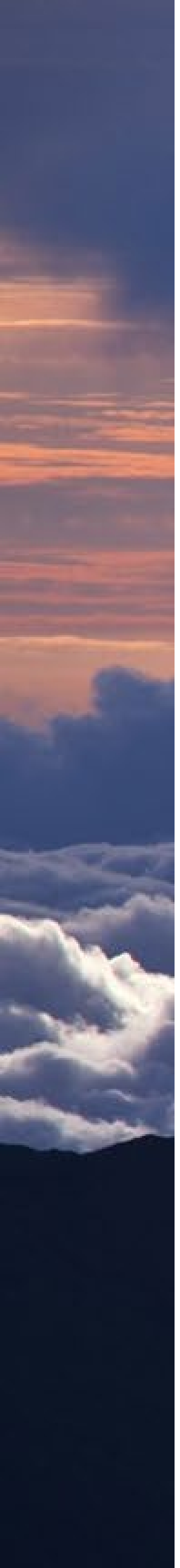
Susannah Burrows
Pacific Northwest National Laboratory

PNNL is operated by Battelle for the U.S. Department of Energy

Updated estimates of effective radiative forcings from the latest IPCC report (AR6; Aug 2022)



IPCC AR6, Figure 7.6



Changes since AR5:
Poor □ good agreement between models and observational evidence

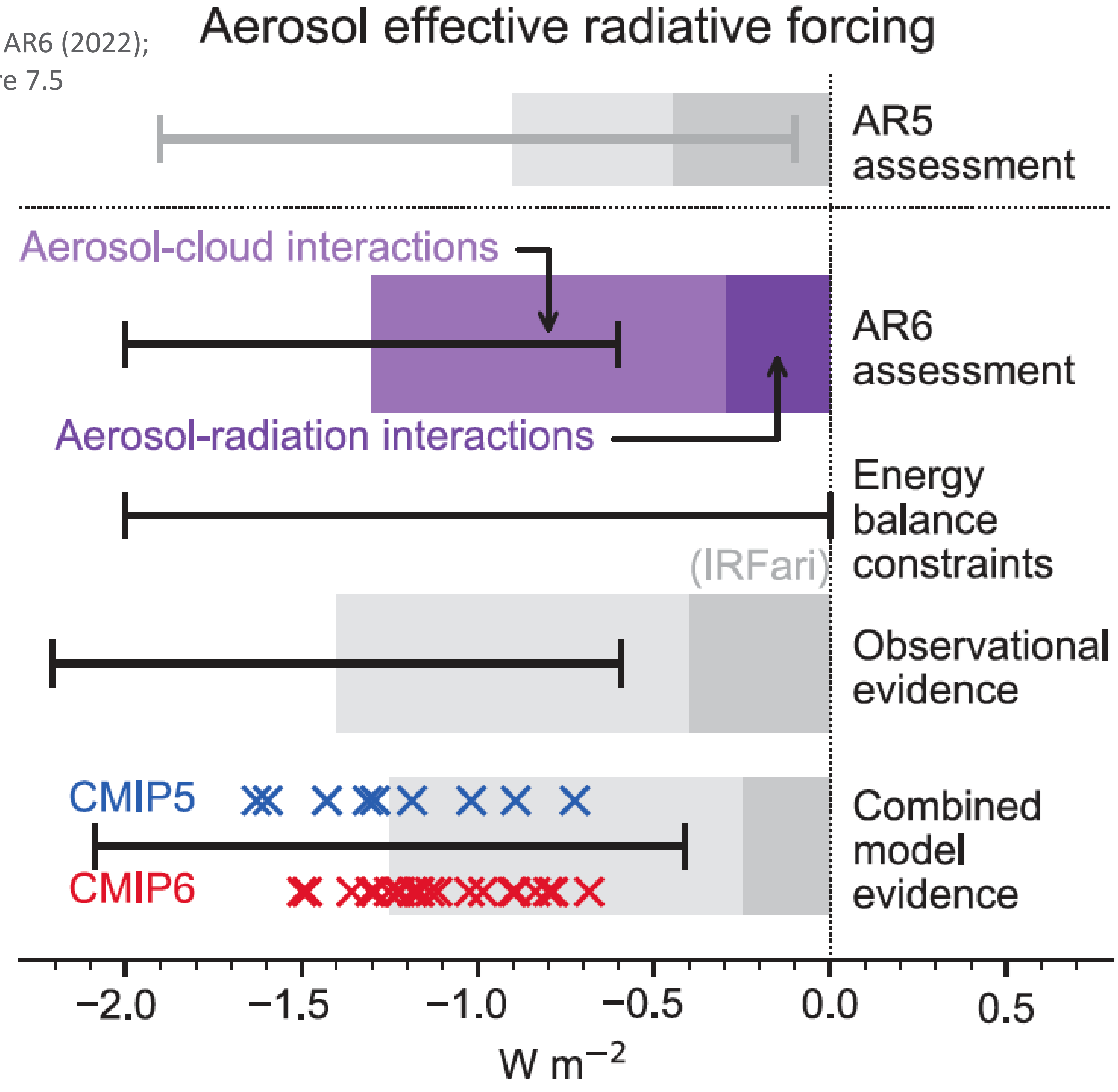
Impacts of aerosol-cloud interactions (ERF_{aci}) INCREASED by about 85%

Direct radiative effects of aerosol (ERF_{ari}) REDUCED by about 50%

Aerosol-cloud interactions (ACI) contribute **70-85% of the total aerosol effective radiative forcing**.

<u>This has implications for which aerosol processes are most limiting on our understanding of climate</u> (as opposed to air quality).

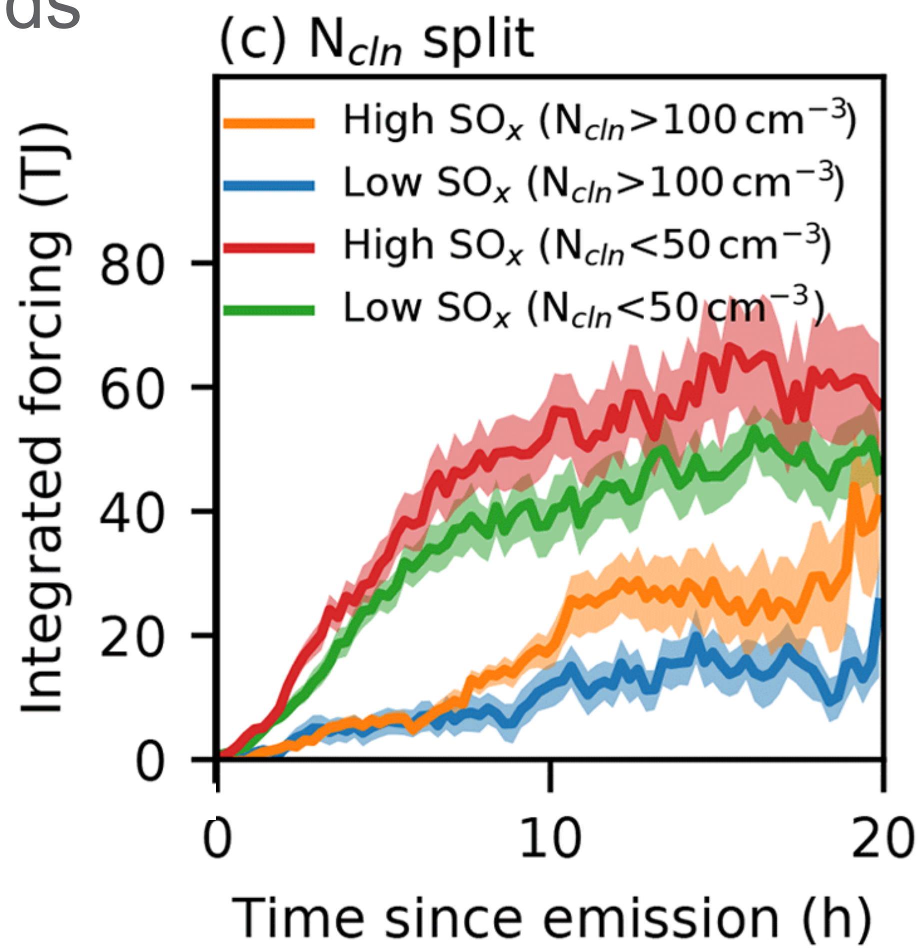
IPCC AR6 (2022); Figure 7.5



A provocative assertion: Natural aerosol processes are now more constraining on climate understanding than anthropogenic processes

1. The ***concentration of CCN under pristine conditions*** is the most important aerosol-related uncertainty for warm clouds

- Cloud forcing by aerosol is a strong function of the background aerosol state, e.g., Lagrangian analysis of cloud responses to ship tracks by Gryspeerdt et al. (2021)
- Difficult to observe lower bound with satellites due to detection limits and cloud masking (Ma et al., *Nat. Comm.*, 2018)
- Truly pristine aerosol conditions occur infrequently in North America, but more frequently occur in remote regions (e.g., Southern Ocean; Hamilton et al., *PNAS,* 2014)

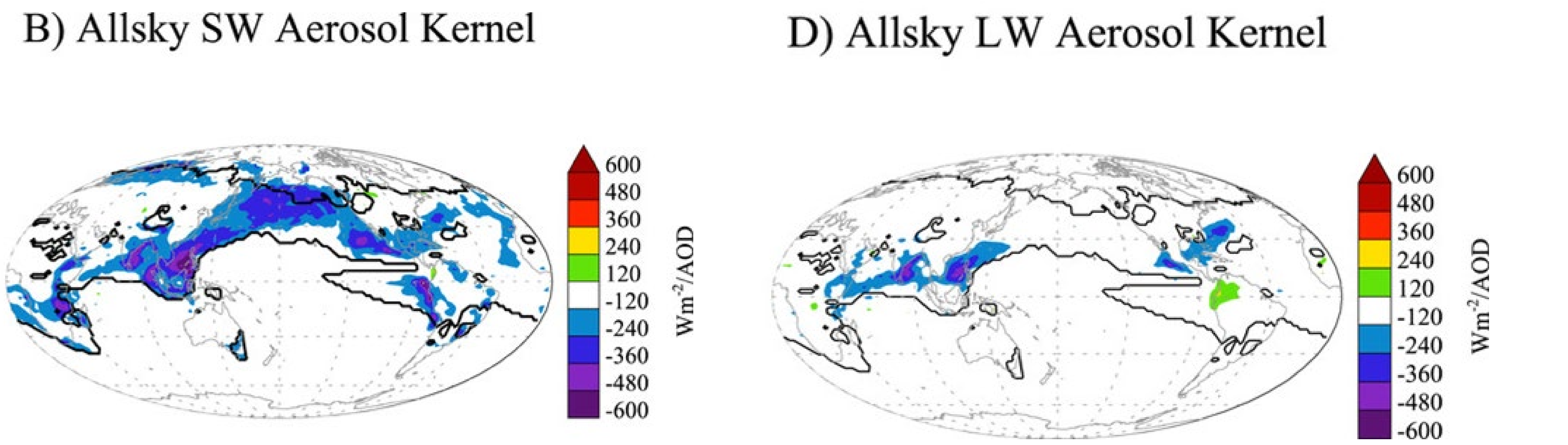


Gryspeerdt et al. (2021)

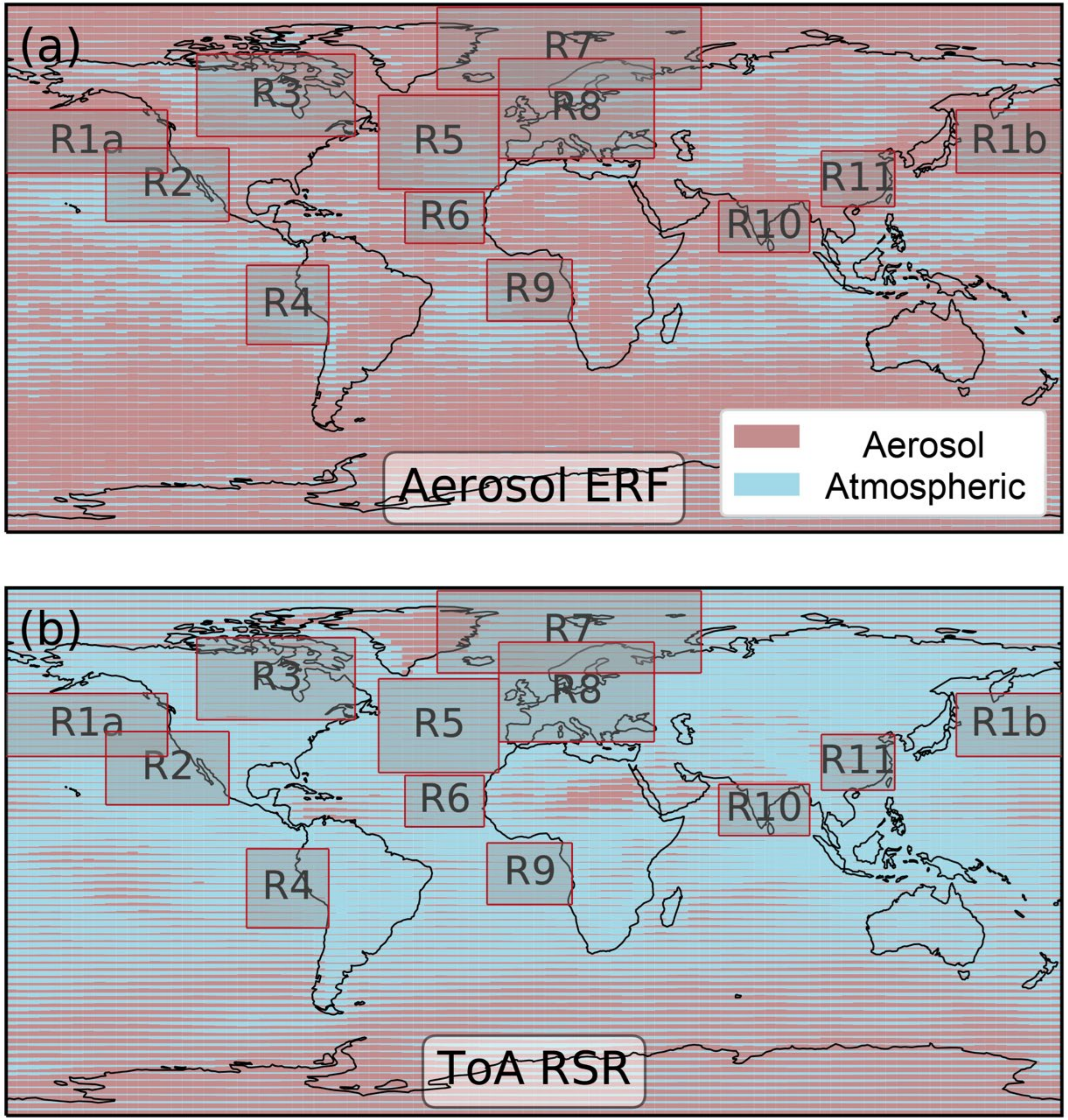
A provocative assertion: Natural aerosol processes are now more constraining on climate understanding than anthropogenic processes

1. The ***concentration of CCN under pristine conditions*** is the most important aerosol-related uncertainty for warm clouds

   - Unit changes in AOD caused larger radiative responses over ocean in CESM (Gettelman et al., 2016)

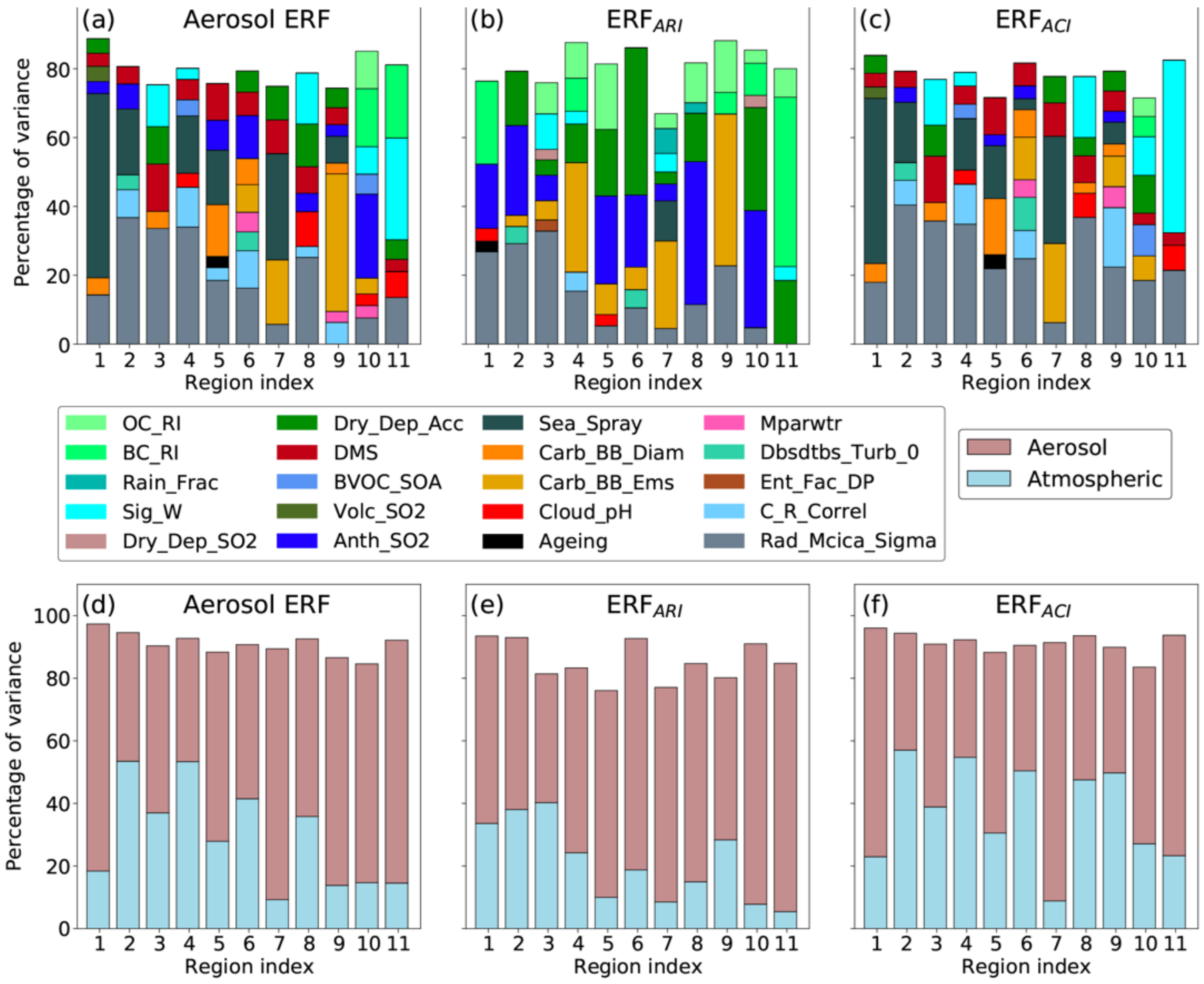


Gettelman et al. (2016)



Natural marine aerosol sources contribute most to parametric uncertainty in aerosol climate impacts

- Regional sources of variance in the 1850-2008 Effective Radiative Forcing (ERF) from aerosol and atmospheric parameters
- 80% of variance in global ERF was explained by four parameters: rad_mcica_sigma, **sea_spray**, **DMS**, and Sig_W.



GLOMAP model

Regayre et al. (2018)

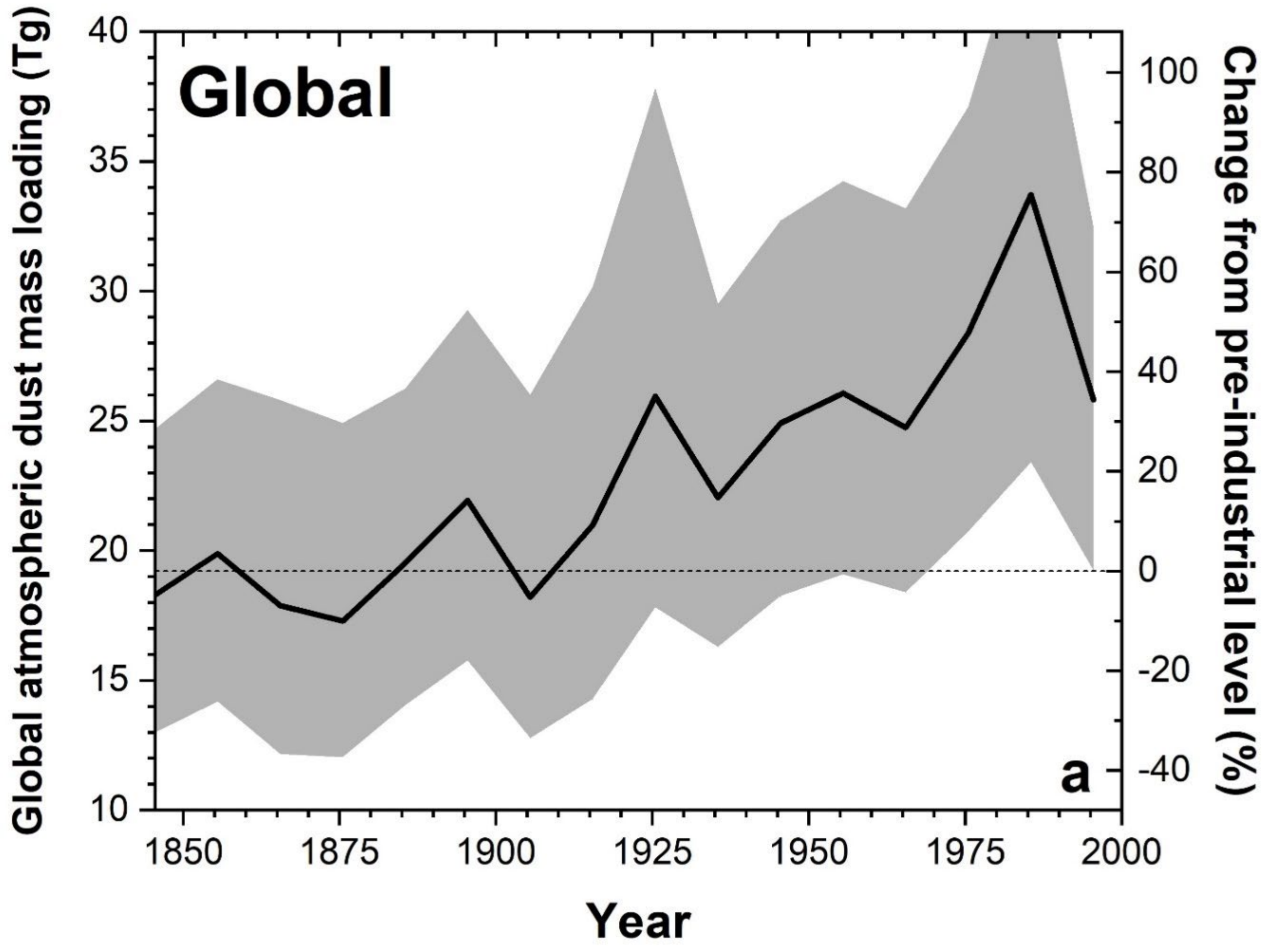
A provocative assertion: <u>Natural</u> aerosol processes are now more constraining on climate understanding than <u>anthropogenic</u> processes

1. The ***concentration of CCN under pristine conditions*** is the most important aerosol-related uncertainty for warm clouds
2. Natural aerosol has important climate ***feedbacks***

Natural aerosol-climate feedbacks are likely important but often missing from GCMs

- **Climate feedbacks to natural emissions due to changes in winds, sea ice extent, aridity, etc.:**
  - Sea spray aerosol
  - Marine precursor gases
  - Wildfires & biomass burning aerosol
  - Dust (Kok et al., 2018, *Nat. Comm.*), including changes in high-latitude emissions

Historical changes in dust emissions due to climate and land use change, reconstructed from observed deposition records



Kok et al. (2022, *in press*)
Preprint available on EarthArXiv

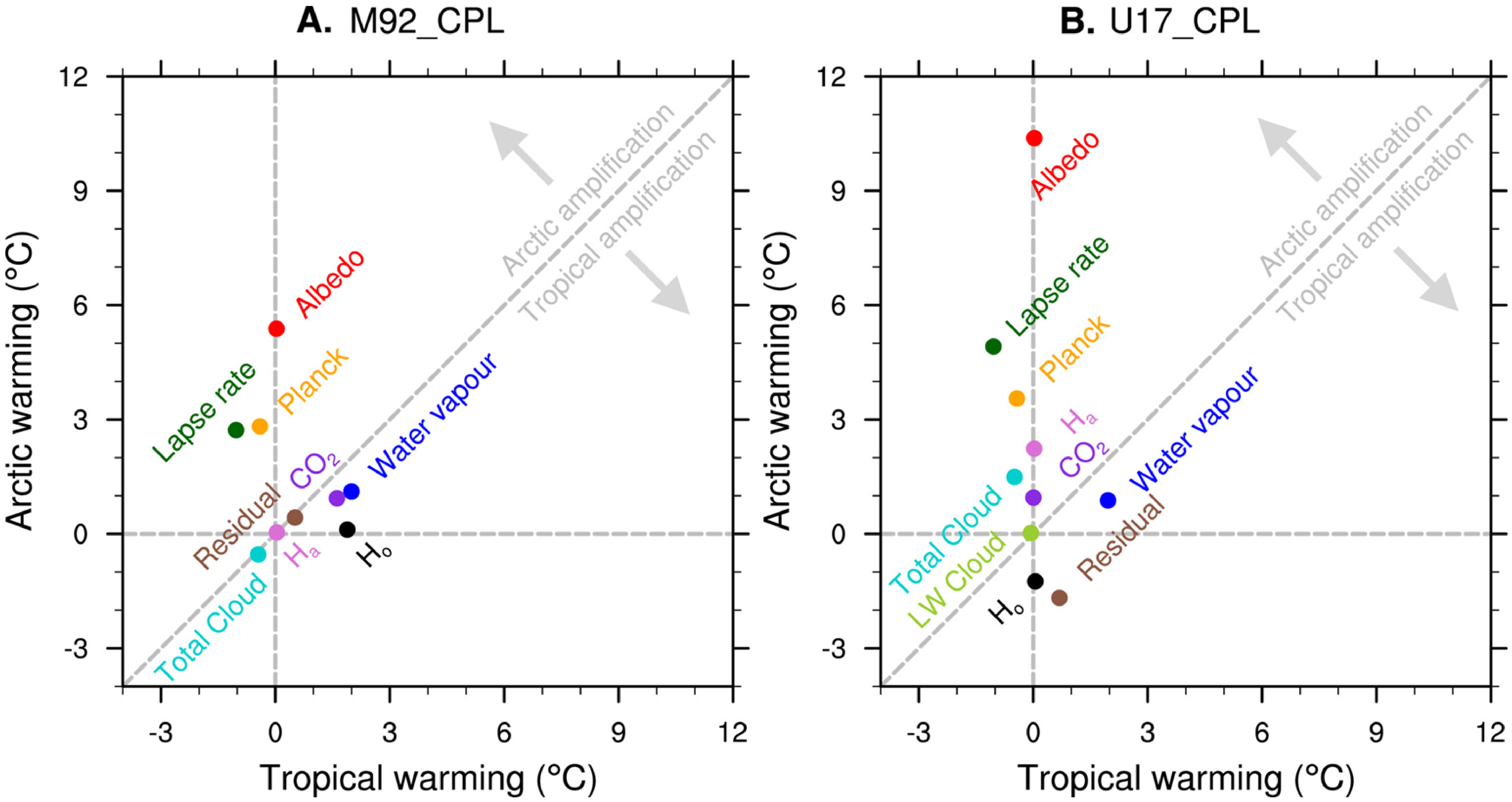
Pacific Northwest
NATIONAL LABORATORY

A provocative assertion: Natural aerosol processes are now more constraining on climate understanding than anthropogenic processes

1. The ***concentration of CCN under pristine conditions*** is the most important aerosol-related uncertainty for warm clouds
2. Natural aerosol has important climate ***feedbacks***
3. Anthropogenic aerosol uncertainties in the ***future*** are primarily controlled by ***human behavior*** – the key uncertainties are associated with emission scenarios, not atmospheric processes

Another major known unknown: aerosol impacts on mixed-phase clouds

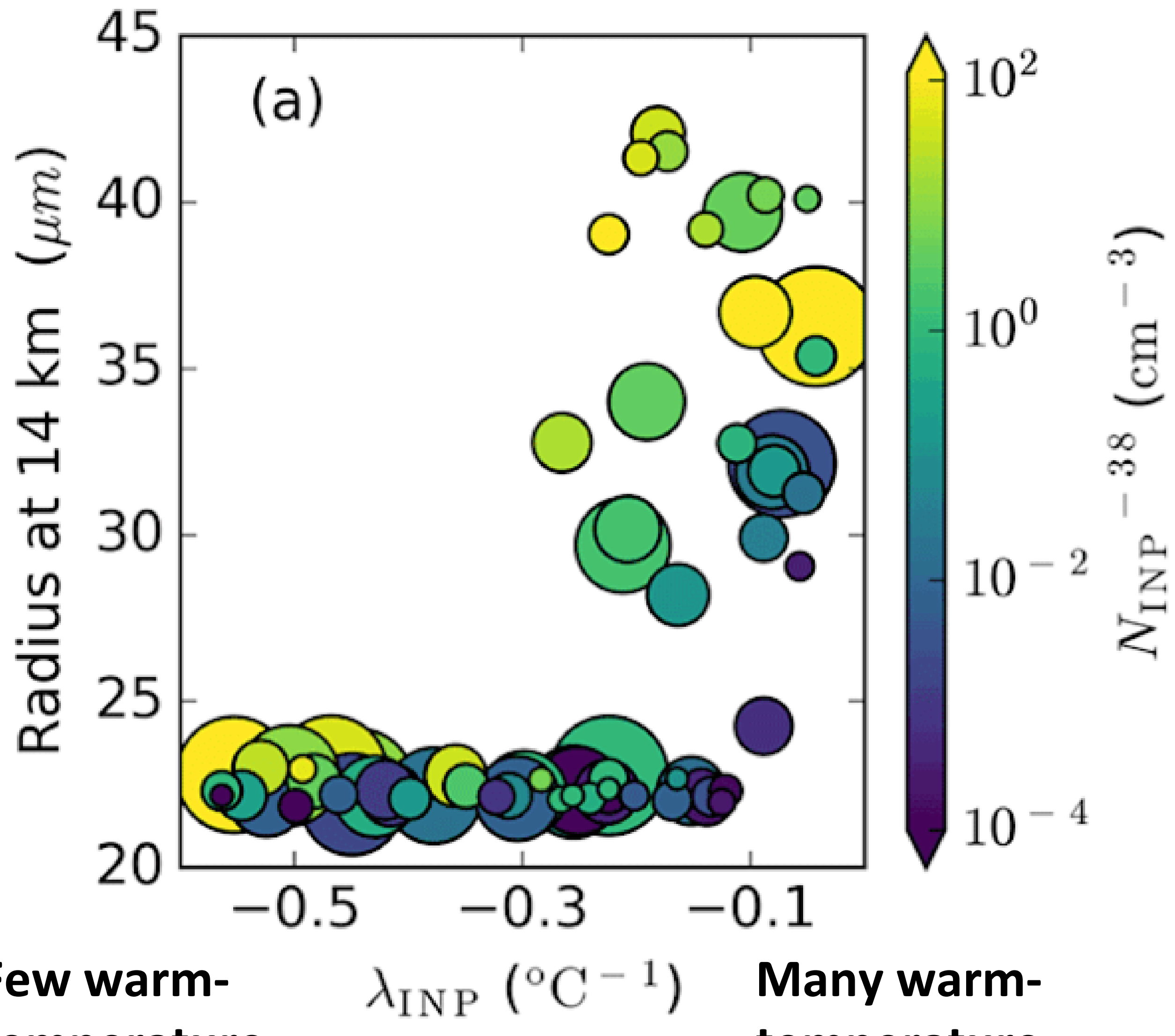
Likely important, but largely missing and/or poorly resolved in GCMs



Tan, Barahona, and Coopman (2022)

Temperature dependence of INPs is likely important: secondary ice production (SIP) and warm-temperature INPs could have joint effects

- Hawker et al. (2021) used a model emulator (of LES) to simulate the joint effects of INPs and SIP (H-M) on deep convective anvil cirrus
- Key findings:
  - Ice crystal *number concentrations* depended cold-temperature INPs
  - Ice crystal *size* depended on warm-temperature INPs
  - **High rates of SIP did not occur unless sufficient warm-temperature INPs are present, regardless of H-M efficiency**



Few warm-temperature INPs

Many warm-temperature INPs

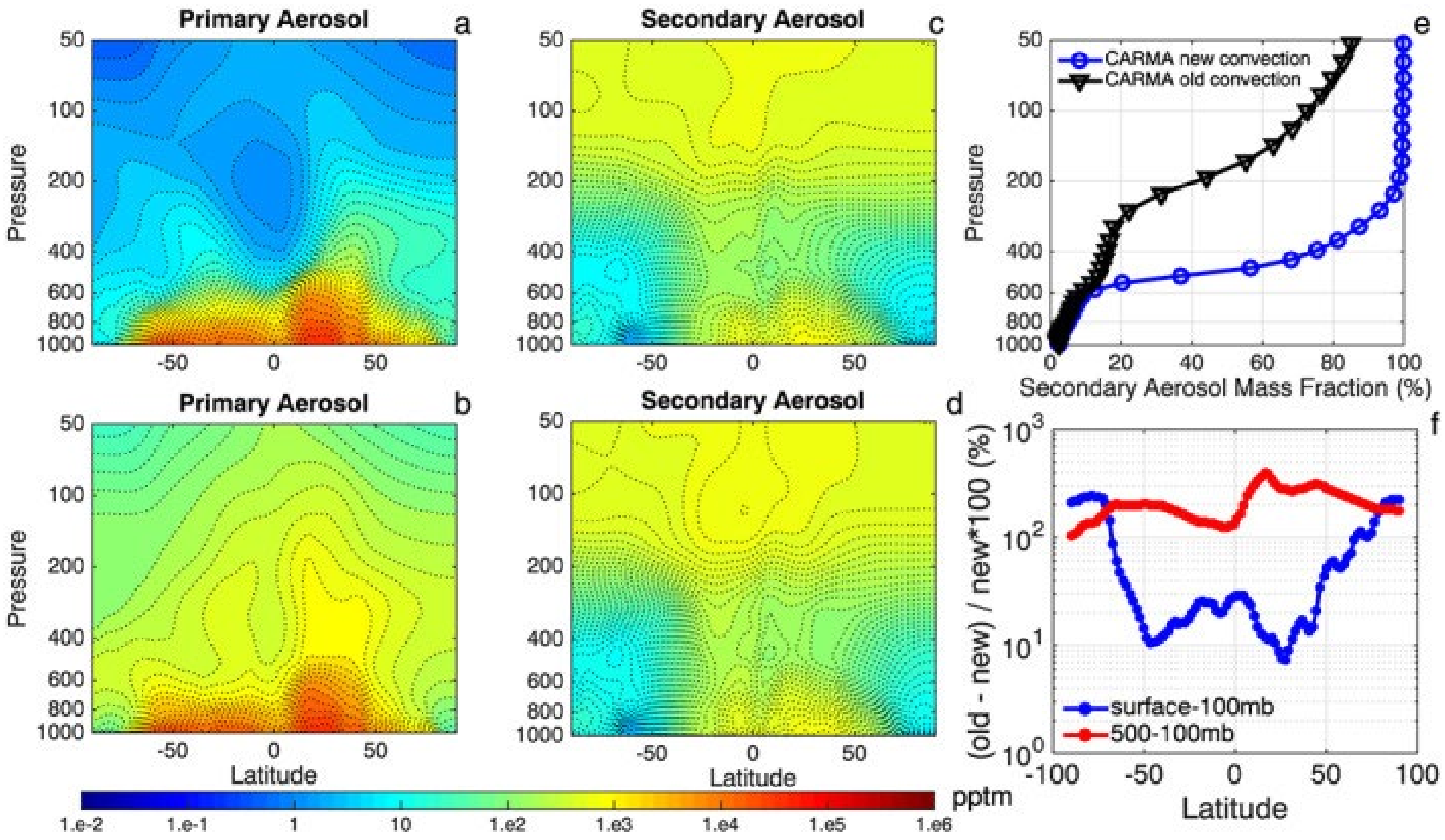
Sources and impacts of warm-temperature INPs

- Warm-temperature INPs appear to be ***mostly primary biological particles***, based on evidence from growing number of studies in a variety of locations and environments (Burrows et al., 2022, *Rev. Geophys.*)
- **Ambient INP measurements**
  - **Arctic** – Creamean et al. (2022); **Argentina / agricultural** – Testa et al. (2021); **Western Europe** – Conen et al. (2022), **Coastal marine boundary layer site, Canada** – Mason et al. (2015); **Agricultural harvesting-generated particles** – Suski et al. (2018); **Punta Arenas, Chile / Patagonia** – Gong et al. (2022); **Asian airborne dust, Beijing, China** – Chen et al. (2021)
- **Precipitation measurements** reviewed by Petters et al., 2015
  - **France, Antarctic, Yukon (Canada), Montana (USA), Louisiana (USA)** *Christner et al.*, 2008; **Wyoming (USA)**: *Hill et al.*, 2014; **Switzerland**: *Stopelli et al.*, 2014; **France (Puy de Dome)**: *Joly et al.*, 2014
- However, we still have significant gaps in our fundamental understanding of bio-INP identities, emissions, atmospheric transport, INP efficacy, and cloud impacts

Critical need for constraints on particle vertical transport processes, especially for large primary particles that are important sources of INPs

Convective mass transport + removal is a major source of intermodal differences in tracer transport

- especially in the upper troposphere and at high latitudes
- important for primary aerosol particles (including INPs)



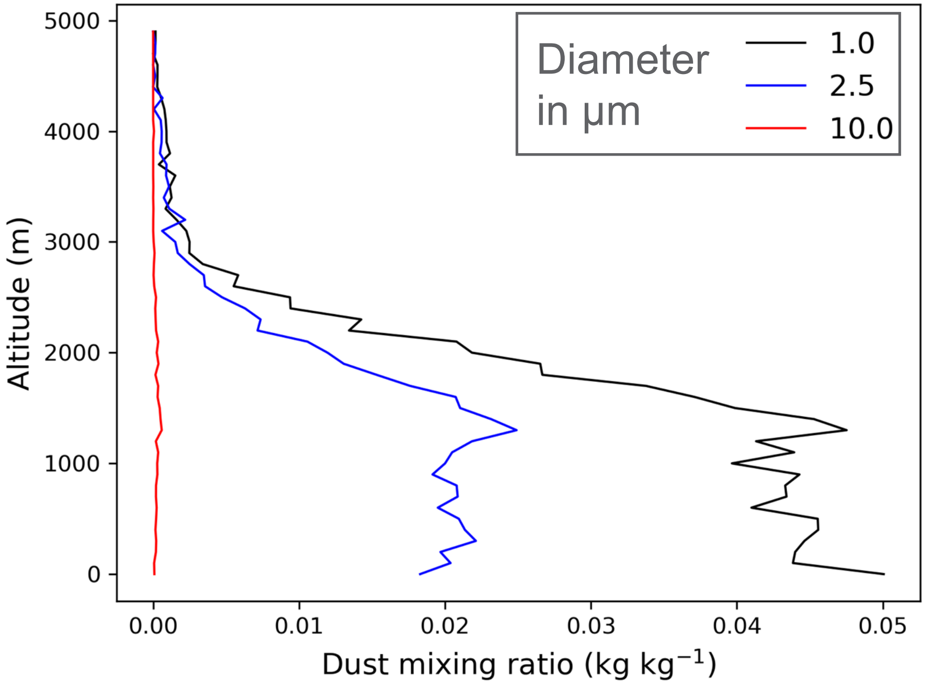
Yu et al. (2019) *Geophys. Res. Lett.*

Export of dust INPs from boundary layer is largely controlled by particle size, with smaller impact of meteorology

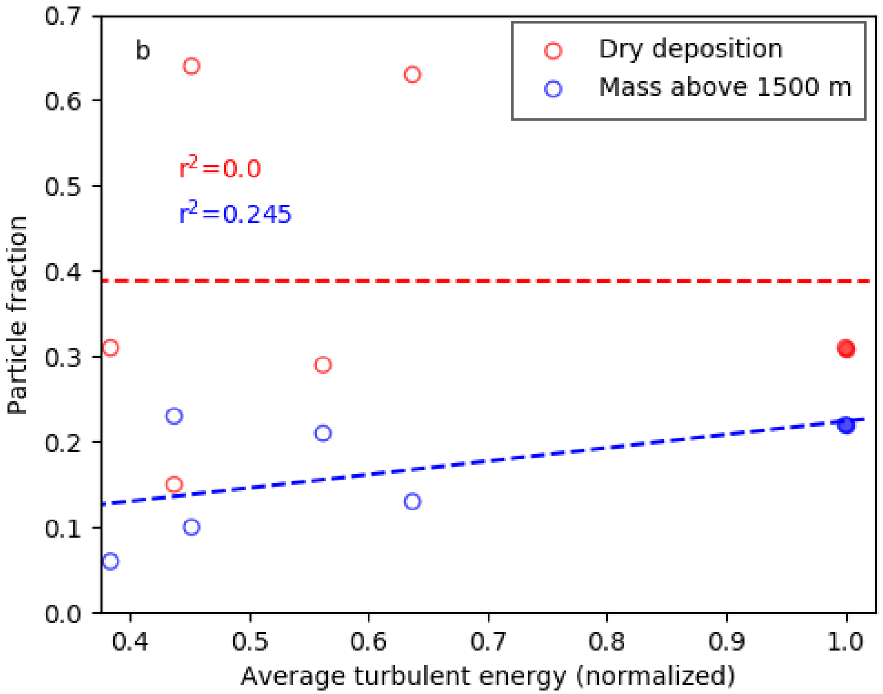
Export from boundary layer controls subsequent availability of INP to clouds

Across six simulated days

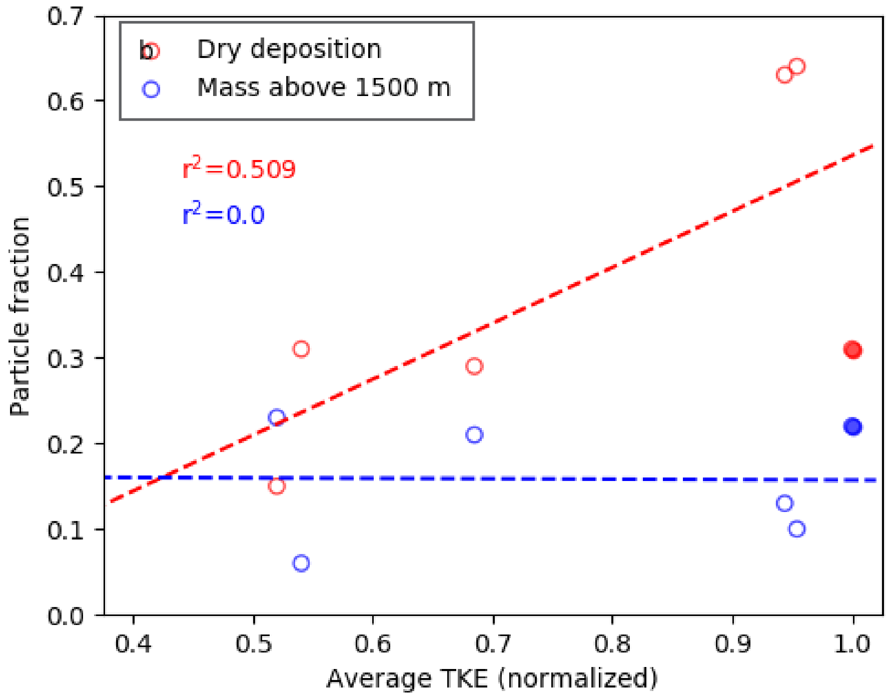
Vertical profile at end of simulated day by particle size



Export weakly correlates with total turbulent energy



Dry deposition moderately correlates with subgrid turbulent energy



Resolving boundary-layer turbulence leads to large differences in simulated vertical particle export, as compared with parameterizing turbulence at coarse resolution

Cornwell, Xiao, Berg, and Burrows, *Journal of Geophysical Research – Atmospheres* (2021)

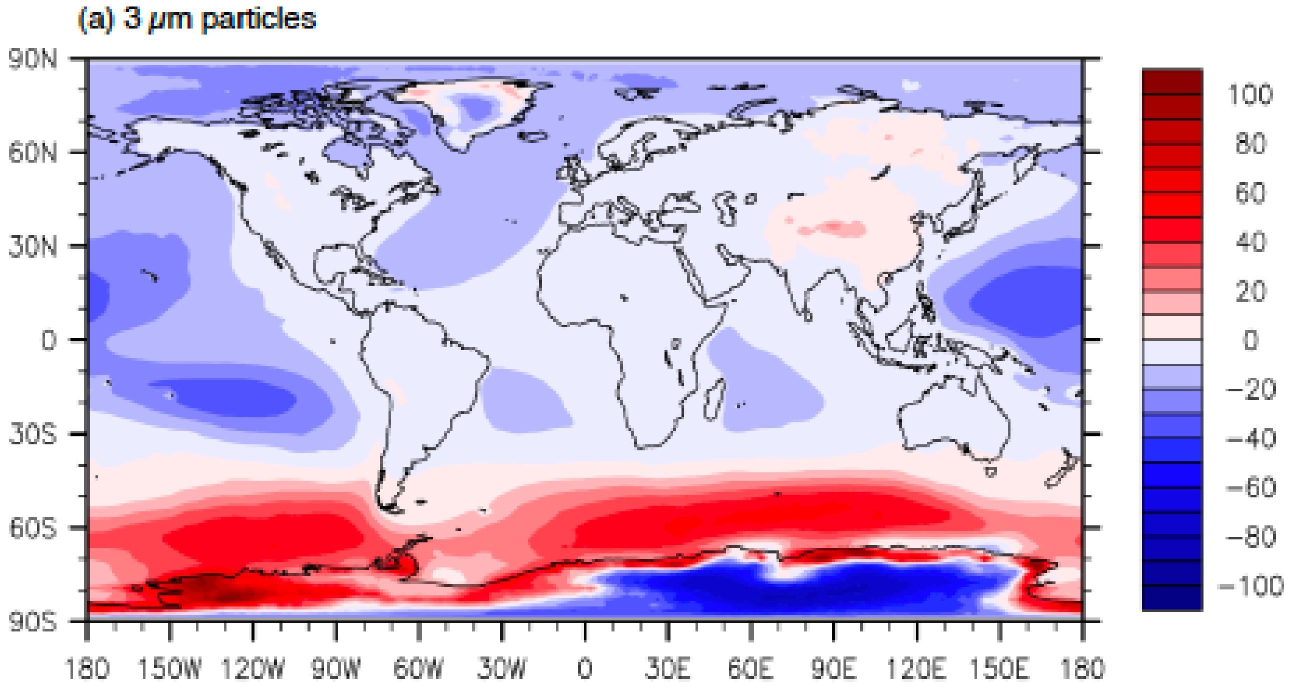
Wet removal is still highly uncertain; depletion of INPs by mixed-phase clouds is suspected but not easily quantified

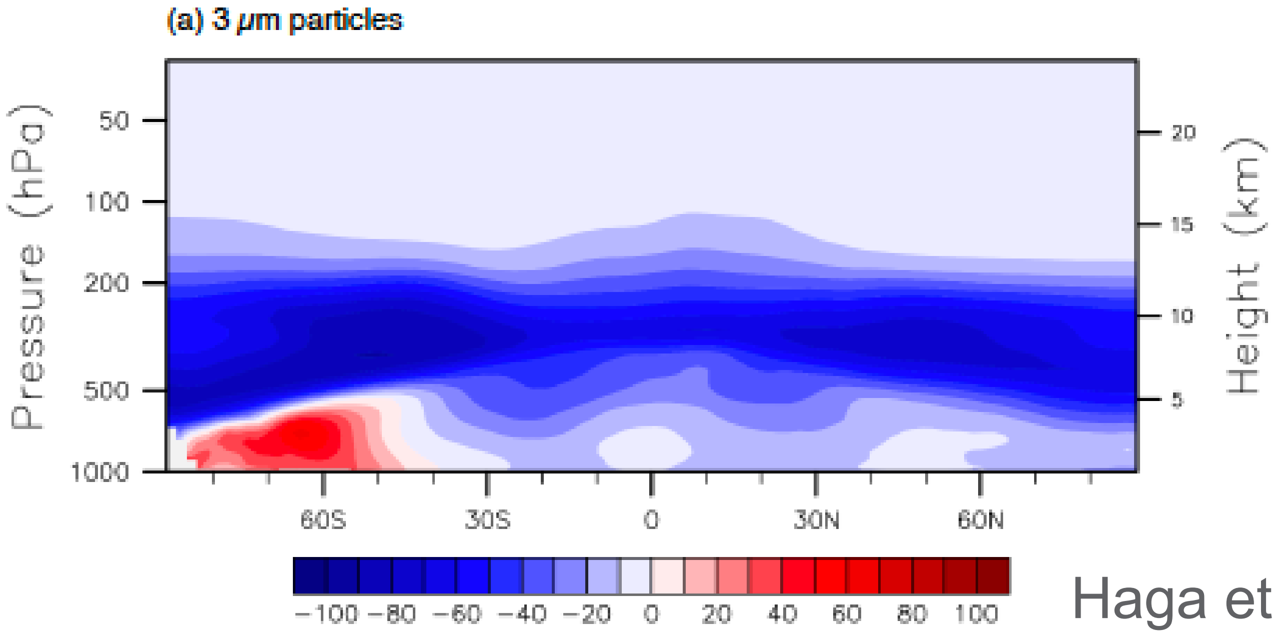
Pacific Northwest
NATIONAL LABORATORY

Simplified sensitivity study: scavenging in mixed-phase clouds ON or OFF

MPC scavenging of INPs has potentially large, but poorly-constrained impacts on availability of INPs at high latitudes and in UT: <u>right where INPs matter most</u>

- Can lab experiments help provide guidance on scavenging rates?
- Or, carefully designed field experiments to constrain scavenging simulated by LES?





Haga et al. (2014)

The ARM/ASR community is well-positioned to advance understanding of the processes controlling INPs relevant to mixed-phase clouds

Burrows et al. (2022) *Rev. Geophys.*

- **High-latitude INP sources**, including oceanic sources, high-latitude dust, INPs from melting permafrost
- **High-latitude aerosol-cloud interactions:** e.g., building on recent results from Arctic (MOSAiC and COMBLE) and Southern Ocean (MARCUS, MICRE)
- **Primary biological INPs active at warm temperatures:** better evaluate current source functions, and improve fundamental understanding of controlling processes, build and evaluate climatologies
- **Particle vertical transport/loss processes:**
  - Boundary-layer turbulence, dry deposition, particle transport within, and export from, the boundary layer
  - Deep convective mass transport and wet removal
  - Depletion of INPs via selective wet removal in mixed-phase clouds

Pacific Northwest
NATIONAL LABORATORY

Thank you