

Retrieval of Cloud Condensation Nuclei to Quantify Radiative Forcing due to ACI

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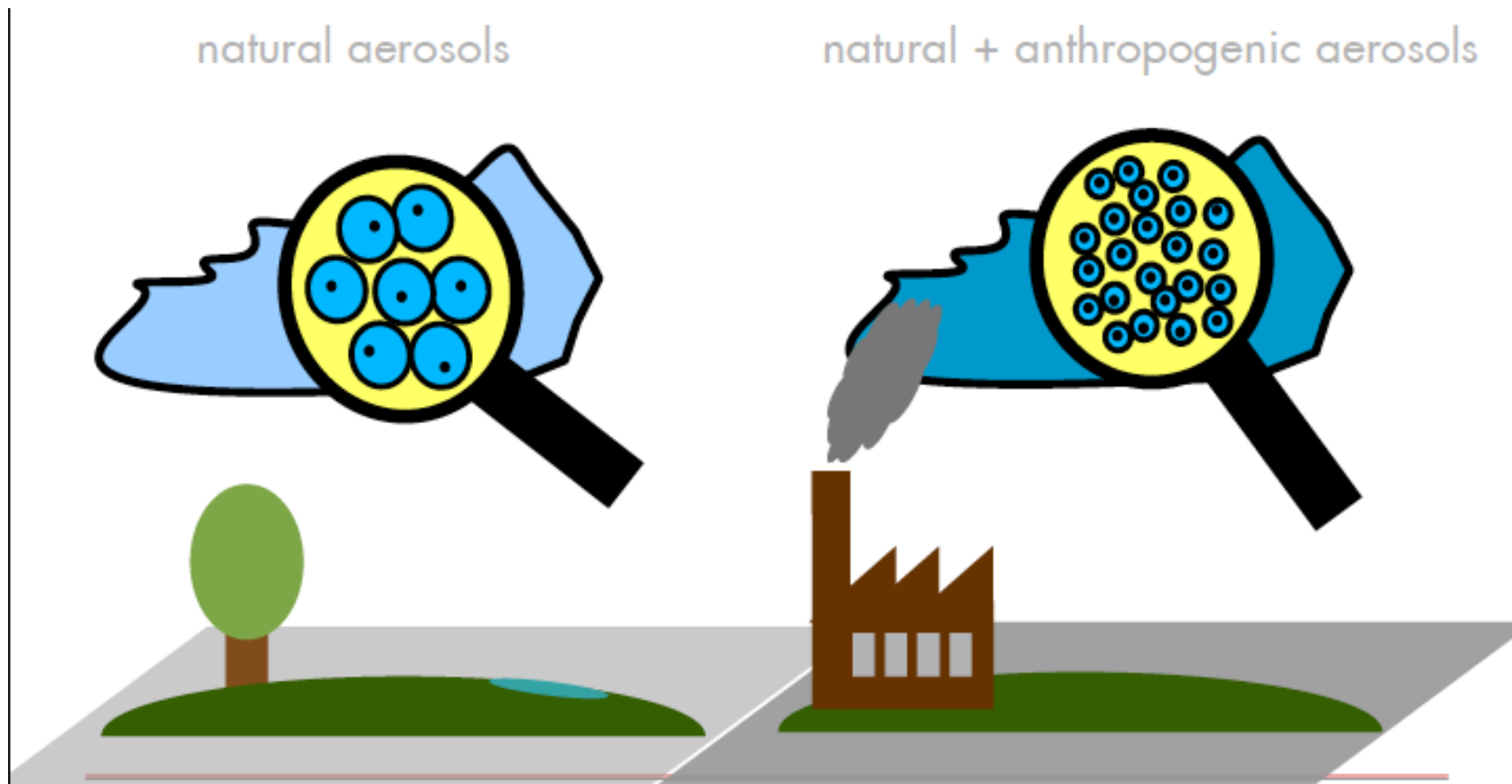
Johannes Quaas (University of Leipzig, Germany)

Hasekamp, O.P., Gryspeerdt, E. & Quaas, J. Analysis of polarimetric satellite measurements suggests stronger cooling due to aerosol-cloud interactions. *Nature Commun* **10**, 5405 (2019). <https://doi.org/10.1038/s41467-019-13372-2>

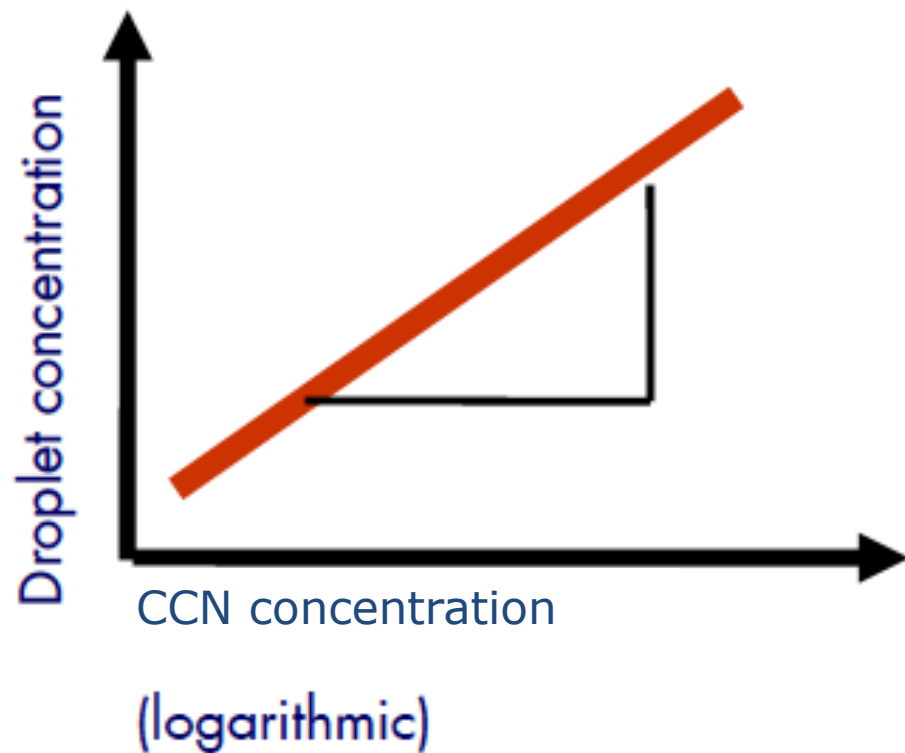


Radiative Forcing due to aerosol-cloud interactions (RF_{aci})

Instantaneous effect of aerosols on cloud droplet number concentration (N_d) and cloud albedo.



Susceptibility



$$S = \frac{d\log(Nd)}{d\log(n_{\text{ccn}})}$$

Satellite studies: 0.10-0.50
In situ / airborne: 0.25-0.90

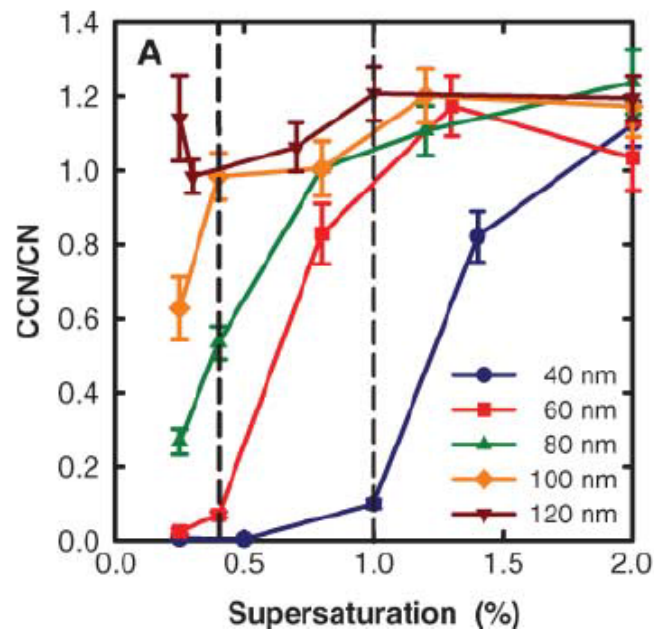
Feingold et al, GRL, 2001
McComiskey and Feingold, GRL, 2008

Caveats of Satellite studies

- Variation in AOD is not only determined by particle concentration but also by variation in extinction cross section ($AOD = N * \sigma$).
- Use of Aerosol Index (AI) - the product of AOD with Angstrom Exponent - partly remedies this, but to what extend? Based mainly on qualitative arguments.
- Not all aerosols are suited as CCN (e.g. very small particles, hydrophobic mineral Dust)

Size Matters More Than Chemistry for Cloud-Nucleating Ability of Aerosol Particles

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F. Drewnick,² S. Hings,² D. Jung,³ S. Borrmann,^{2,3} M. O. Andreae¹

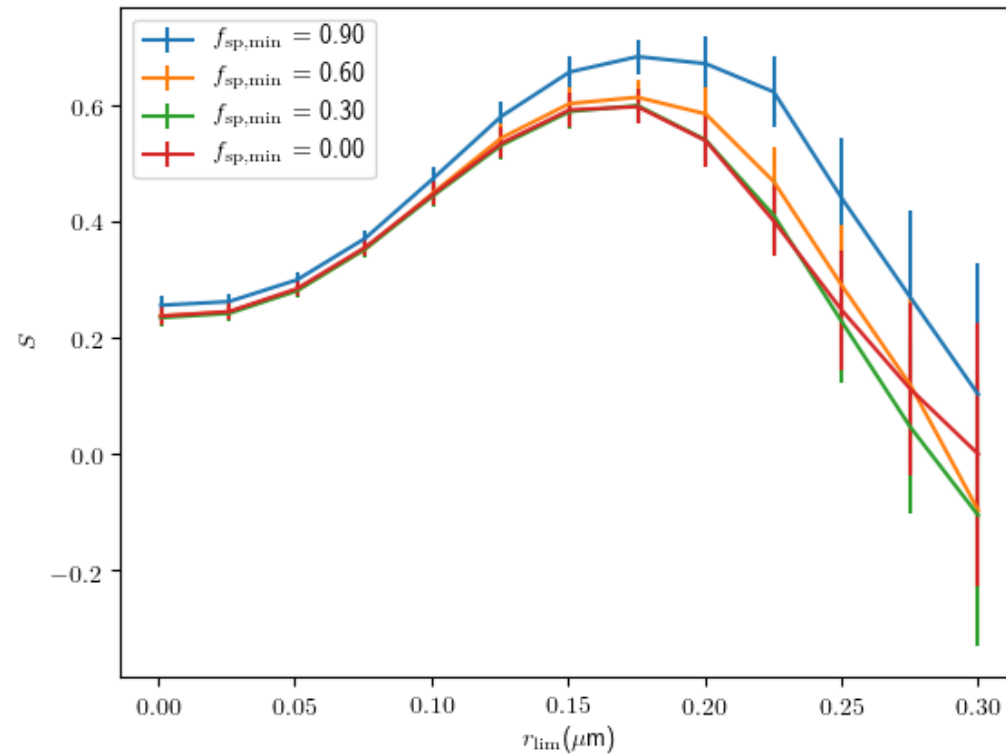


Only aerosols with $r > r_{lim}$ are suitable as CCN

Susceptibility from POLDER-3 and MODIS

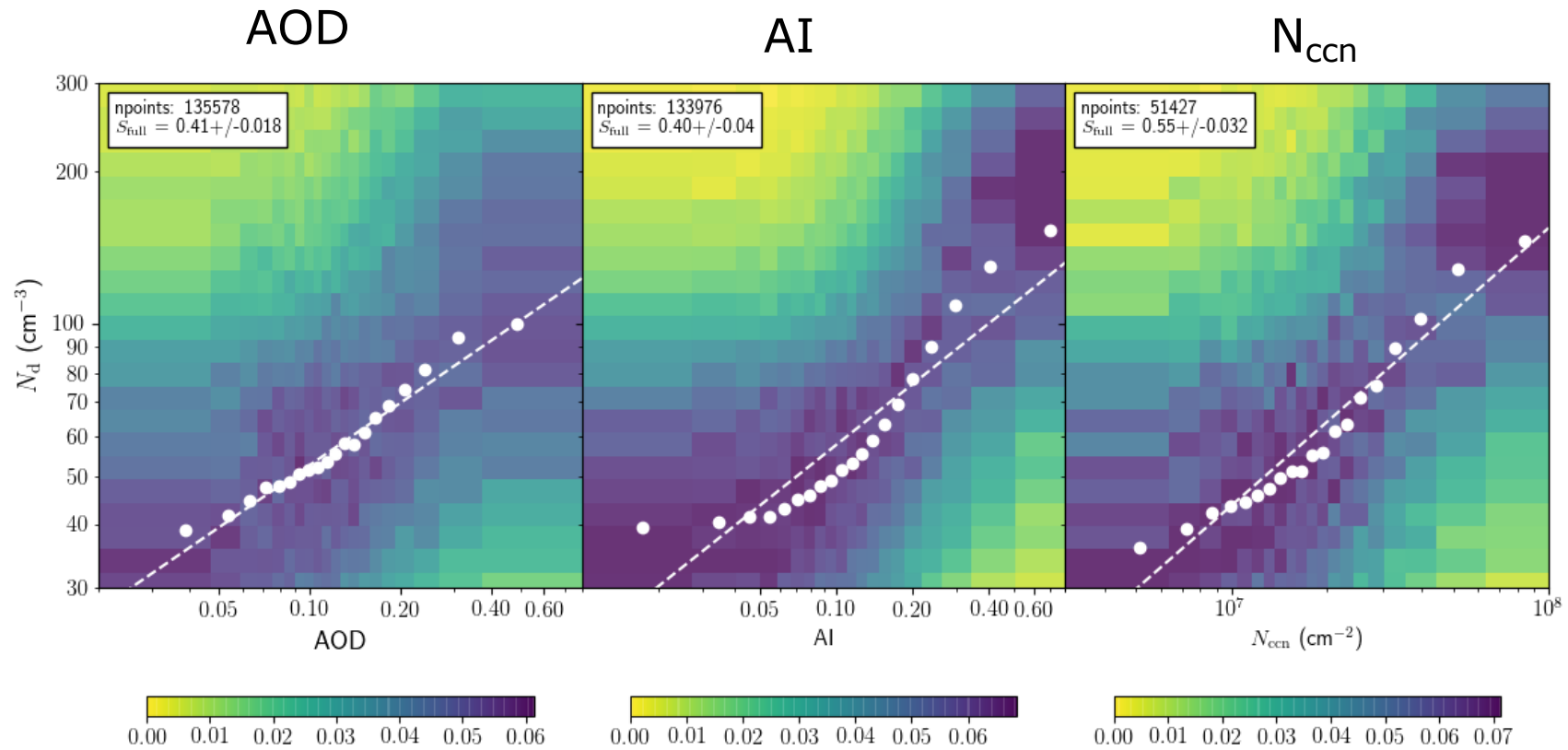
- POLDER-3/PARASOL provides retrievals of particle column and size distribution.
- From this the column of particles with $r > r_{lim}$ can be computed
→ proxy for CCN (N_{ccn})
- MODIS provides retrievals of cloud droplet number concentration (N_d).
- Aggregated to 1x1 degree. Use grid cells that contain both POLDER-3 aerosol retrievals and MODIS N_d retrievals.
- From binned N_{ccn} - N_d relationship compute susceptibility
$$S = \frac{d\log(N_d)}{d\log(N_{ccn})}$$
 using linear regression.

Dependence of Susceptibility on Size and Shape



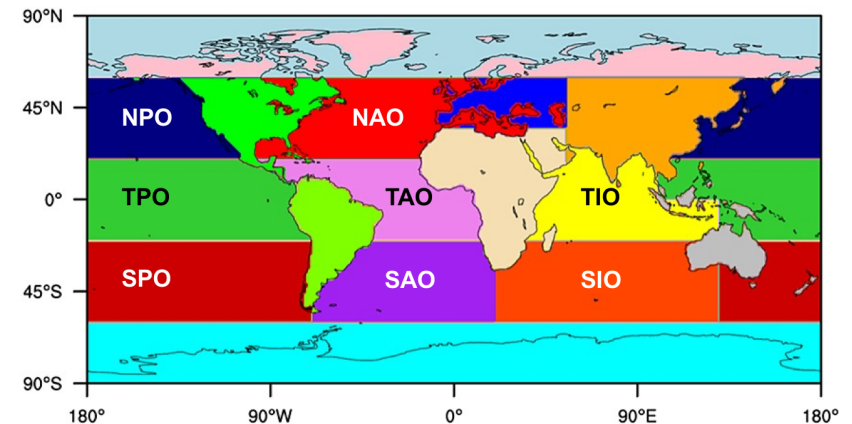
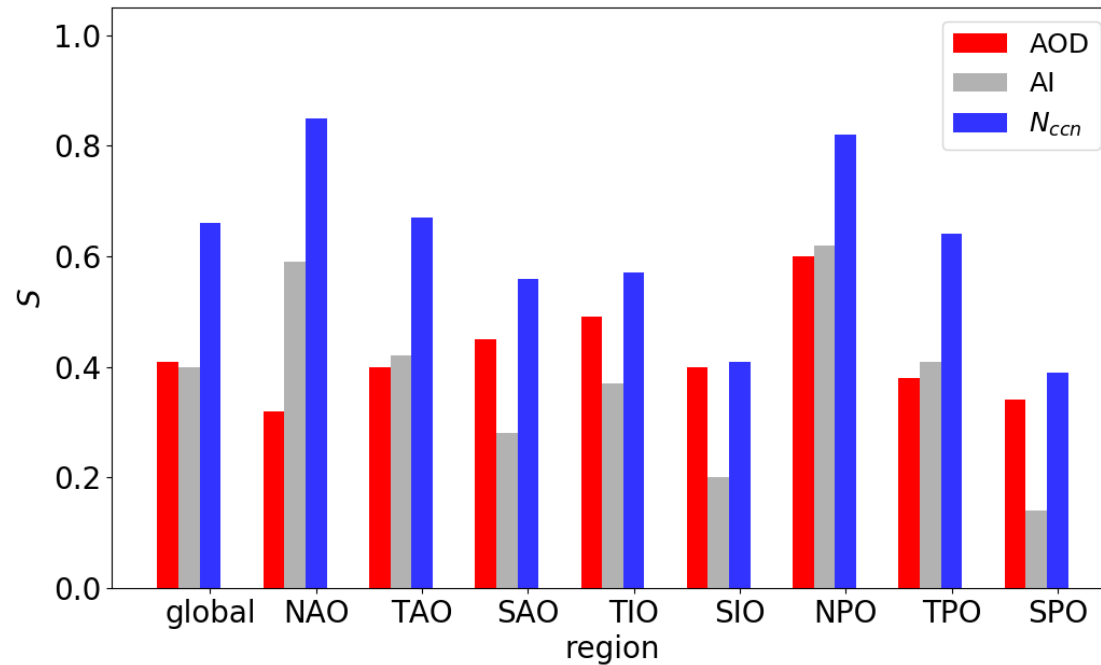
- Inclusion of non-CCN yields too small susceptibility as they are not related to Nd.
- Best CCN proxy N_{ccn} for $r > 0.15 \mu\text{m}$ and $f_{sphere} > 0.90$.
- Taking a factor ~ 2 growth due to humidification into account, our r_{lim} corresponds to $0.075 \mu\text{m}$ dry radius (Dusek et al suggest $\sim 0.06 \mu\text{m}$).

Aerosol - N_d relationships for AOD, AI and N_{ccn}



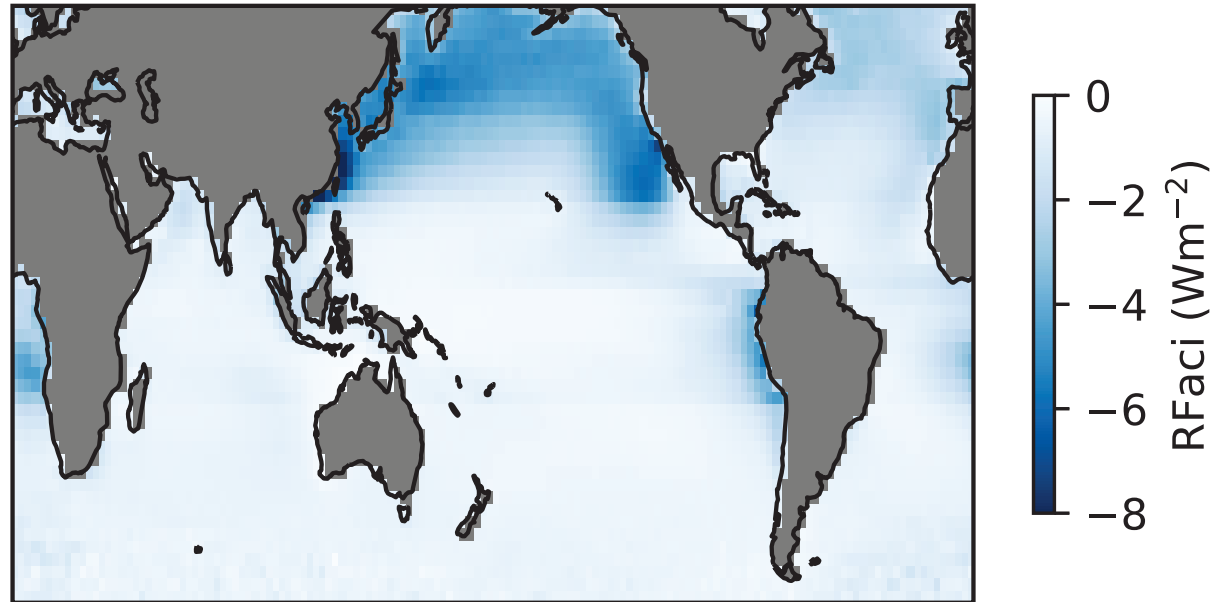
Susceptibilities for different regions

In the important regions, the new susceptibilities based on N_{ccn} are more than 50% higher than values based on AOD or AI ('old approach')



Radiative Forcing due to Aerosol Cloud Interactions (RFaci)

Use 5 different aerosol climate models to provide an estimate of the increase in N_{ccn} between Pre-Industrial (PI) and Present Day (PD).



Global ocean value: -0.76 Wm^{-2}

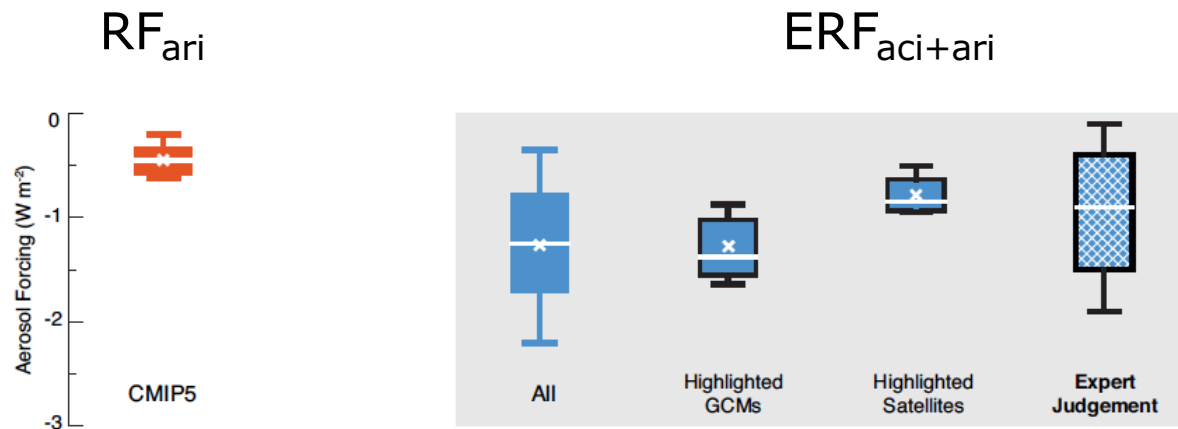
Use ratio $\text{RFaci}_{\text{global}} / \text{RFaci}_{\text{ocean}}$ from 13 different aerosol climate models to estimate contribution over land.

Global value: **-1.14 Wm^{-2}** (-0.84 to -1.72 Wm^{-2})

AOD based: -0.33 Wm^{-2} (-0.19 to -0.54 Wm^{-2})

AI based: -0.80 Wm^{-2} (-0.58 to -1.24 Wm^{-2})

What does the IPCC 5th assessment say?



GCMs give $ERF_{aci} \sim -0.90 W m^{-2}$

Satellites give $ERF_{aci} \sim -0.35 W m^{-2}$

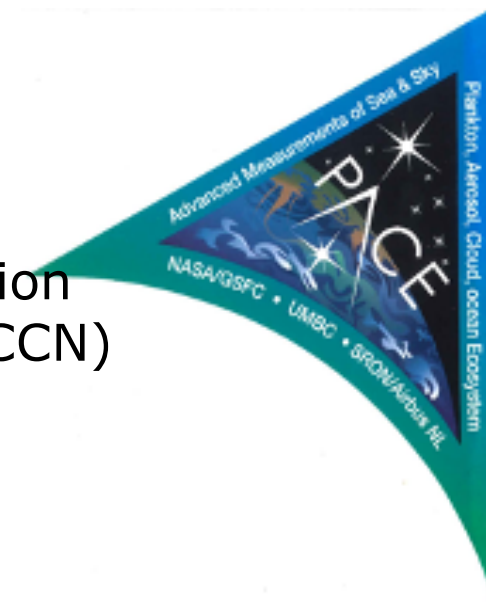
Expert judgement put more weight on satellite estimates:

$ERF_{aci} = -0.45 W m^{-2}$, $ERF_{aci+ari} = -0.90 W m^{-2}$

But most satellite studies in IPCC AR5 are based on AOD, some on AI
So, it seems that aerosol radiative forcing is underestimated.

What's next?

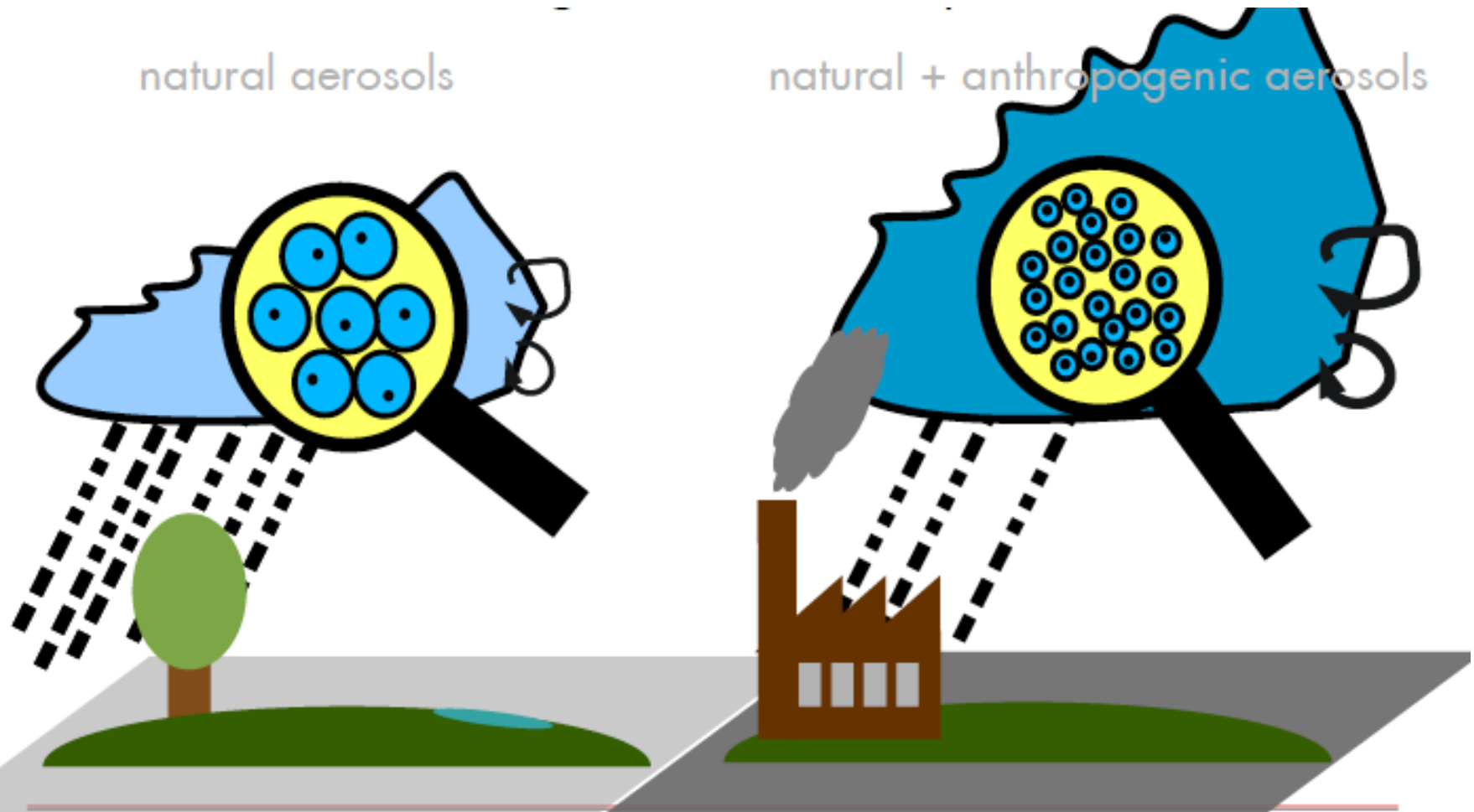
- Largest (known) uncertainty comes from the contribution over land to RFaci. Strong need for accurate aerosol (CCN) retrievals over land.
- SPEXone on PACE (launch 2023) will provide these accurate retrievals over land.
- HARP-2 on PACE will provide accurate retrievals of CDNC.
- To go from RFaci to ERFaci, the effect of aerosol on cloud fraction is the big unknown → difficult to estimate because of aerosol swelling.
- SPEXone on PACE will provide accurate retrievals of the aerosol refractive index → amount of aerosol water. This makes it possible to define CCN based on dry aerosol size → removes swelling effect.



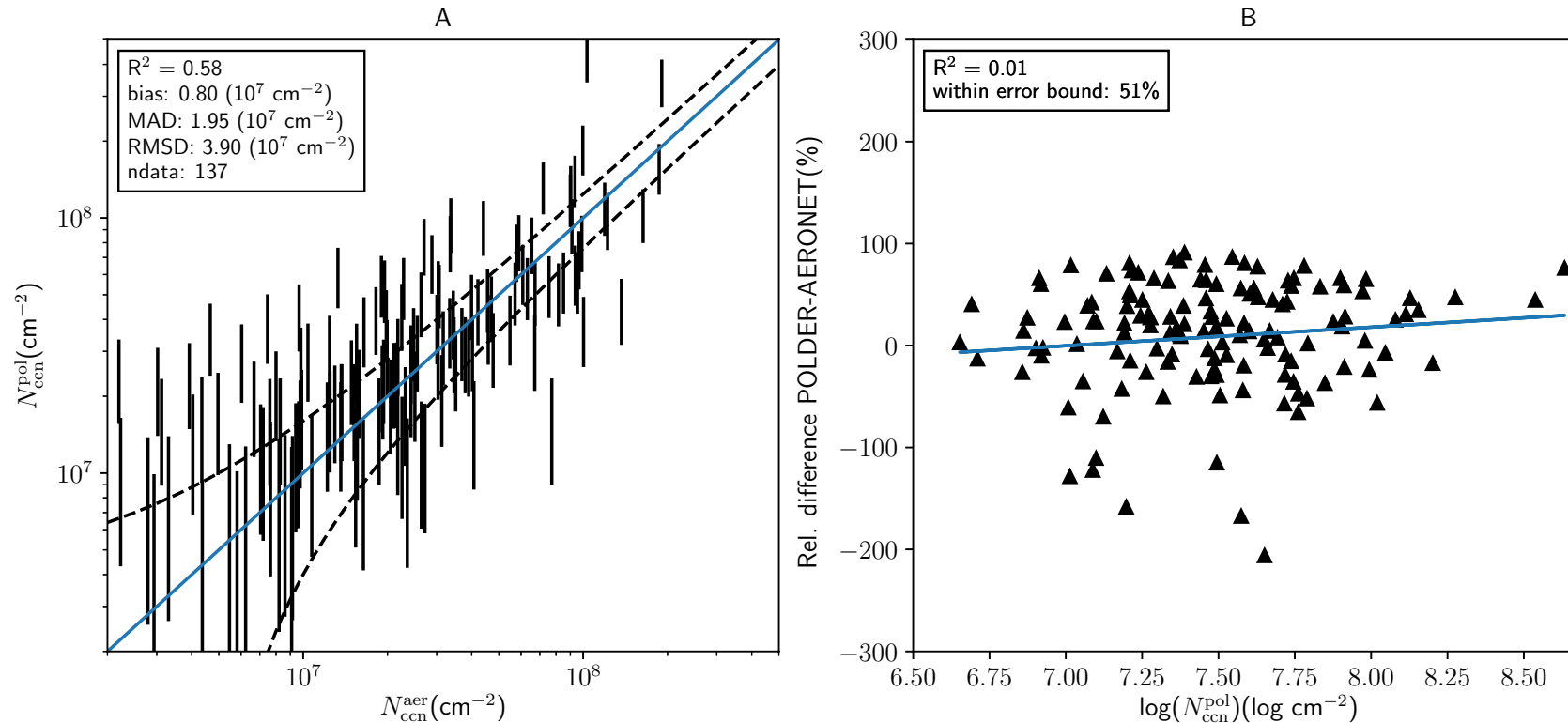
BACKUP SLIDES

Effective Radiative Forcing due to aerosol-cloud interactions (ERFaci)

Includes also cloud adjustments (Liquid Water Path, Cloud Fraction, etc.)



How well can we retrieve Nccn?



- Synthetic retrievals suggest an accuracy of $0.20 N_{ccn} + 4 \cdot 10^6 cm^{-2}$.
- 51% of the data have a difference with AERONET smaller than this value.
- This confirms that it is a reasonable error estimate, as errors in the AERONET retrievals also contribute to the differences.

How do retrieval errors affect susceptibility?







ARTICLE

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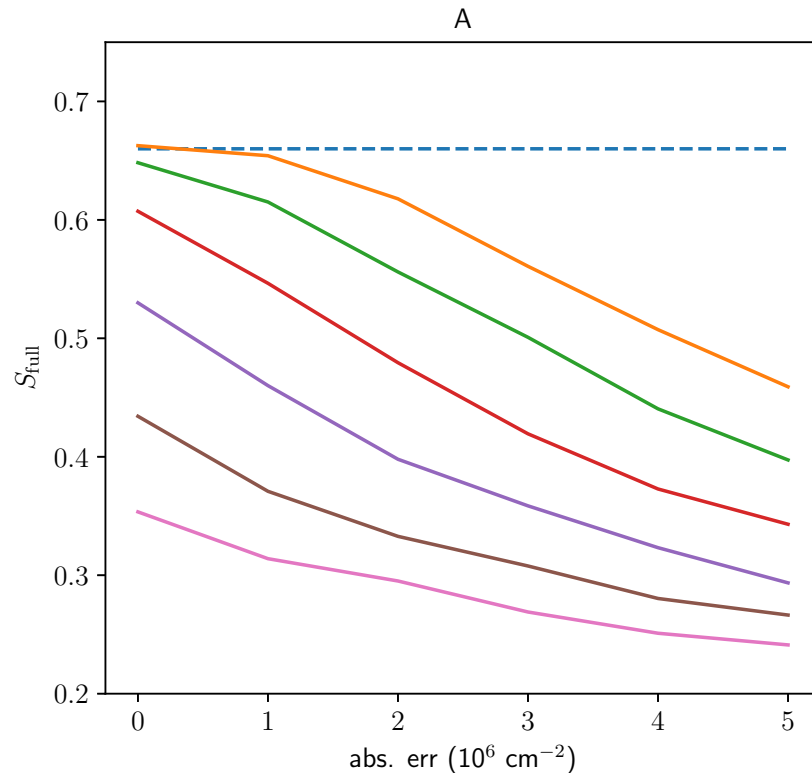
OPEN

Observational constraint on cloud susceptibility weakened by aerosol retrieval limitations

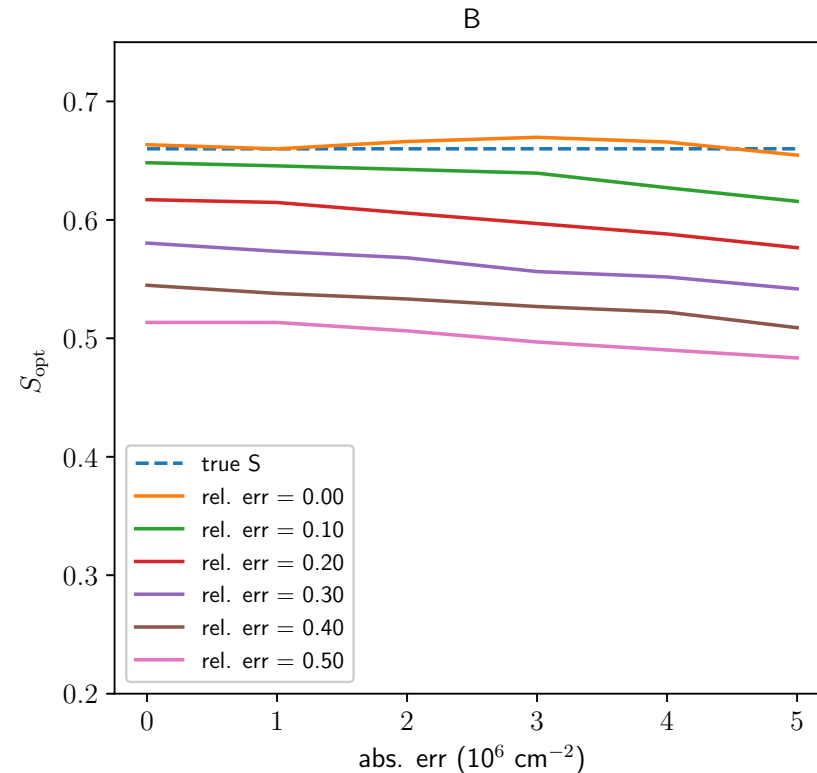
Po-Lun Ma ¹, Philip J. Rasch ¹, H el ene Chepfer^{2,3}, David M. Winker ⁴ & Steven J. Ghan ¹

How do retrieval errors affect susceptibility?

Using all retrievals



Using only $N_{\text{ccn}} > 10^7 \text{ cm}^{-2}$



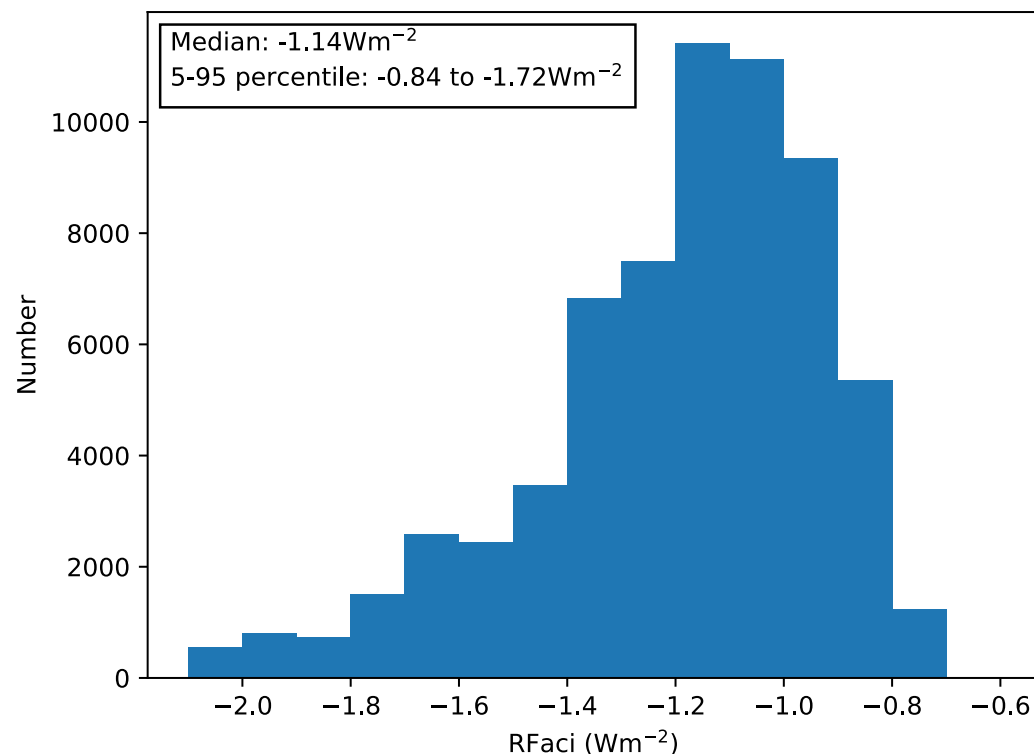
- Measurement errors at low N_{ccn} values lead to strong underestimation of S .
- Leaving out small values largely remedies this effect \rightarrow closer to true S .
- Same reasoning holds for AOD and AI, but previous RFac estimates did not exclude small values.

From Susceptibility to Forcing

- Use 5 different aerosol climate models to provide an estimate of the increase in N_{ccn} between Pre-Industrial (PI) and Present Day (PD).
- Compute the increase in N_d : $\Delta \log(N_d) = S * \Delta \log(N_{\text{ccn}})$.
- From ΔN_d compute the change in cloud albedo (Twomey) and the corresponding Radiative Forcing (RFaci).
- This gives 5 estimates of RFaci over the ocean (as POLDER only provides sufficiently accurate information over ocean).

Global RFaci estimate

- Use ratio $\text{RFaci}_{\text{global}} / \text{RFaci}_{\text{ocean}}$ from 13 different aerosol climate models.
- Combine with 5 estimates of $\text{RFaci}_{\text{ocean}}$ and the uncertainty estimate on S.



The uncertainty range is dominated by uncertainty in $\text{RFaci}_{\text{global}} / \text{RFaci}_{\text{ocean}}$. This uncertainty term alone results in a range -0.85 to -1.70 Wm⁻².