Progress in determining the climate impact of aerosol particles emitted from aircraft on large-scale cirrus clouds

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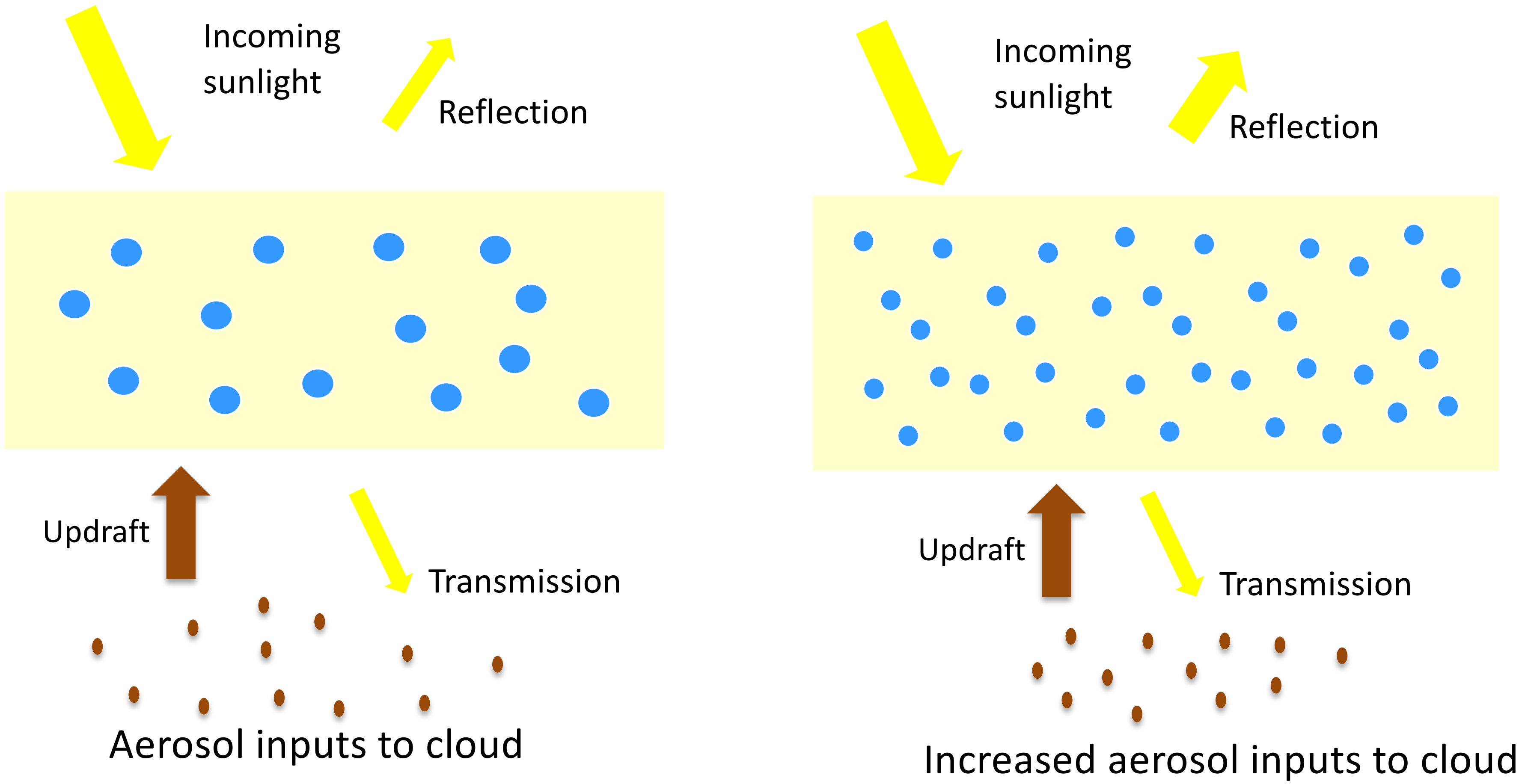
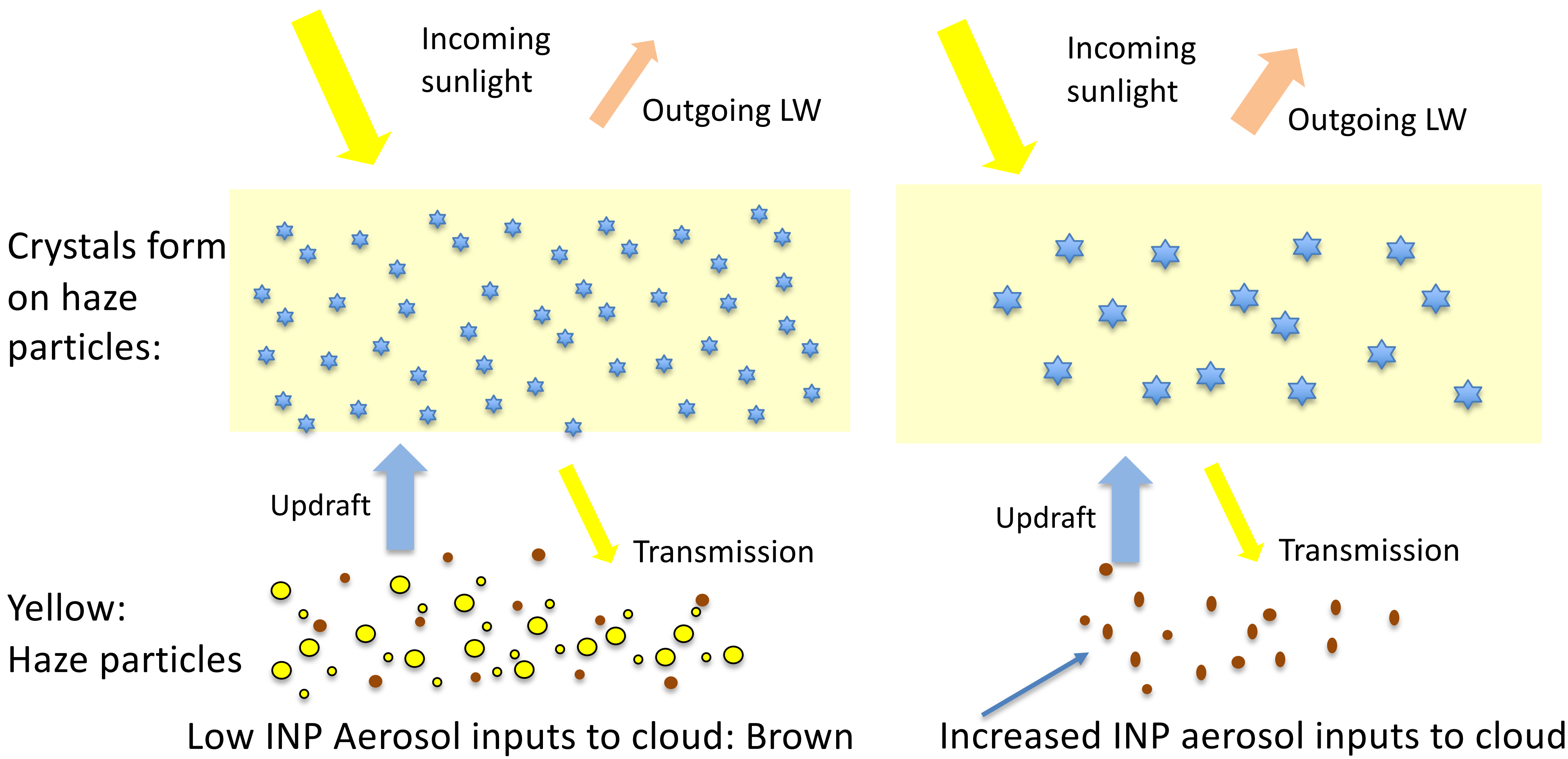
Outline

- Introduction: cirrus effects of aerosols vs warm-cloud effects
- Summary of controversies in aircraft soot effects
- Discussion of aspects contributing to differences in cirrus ice number concentrations in different models
- Laboratory observations relevant to the role of aircraft soot as INP
- Satellite and field observations of clouds with/without aircraft emissions
- Comparison with model results for aircraft-flight decreases in Ni
- Conclusions

Warm clouds: increasing aerosols increase SW reflection:



Cirrus clouds: increasing heterogeneous INPs decreases ice crystals and increases outgoing LW :

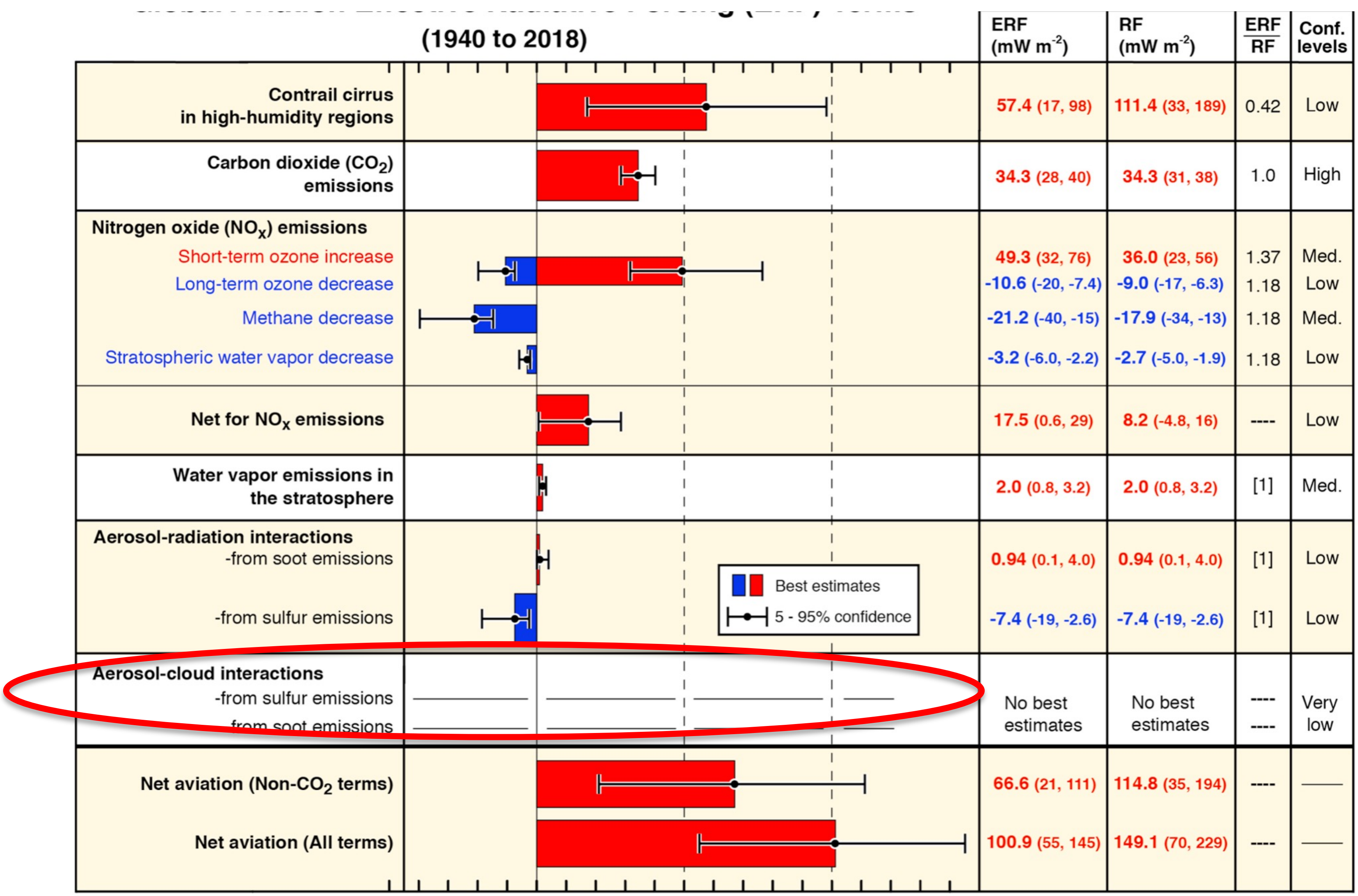


Homogeneous freezing & heterogeneous nucleation

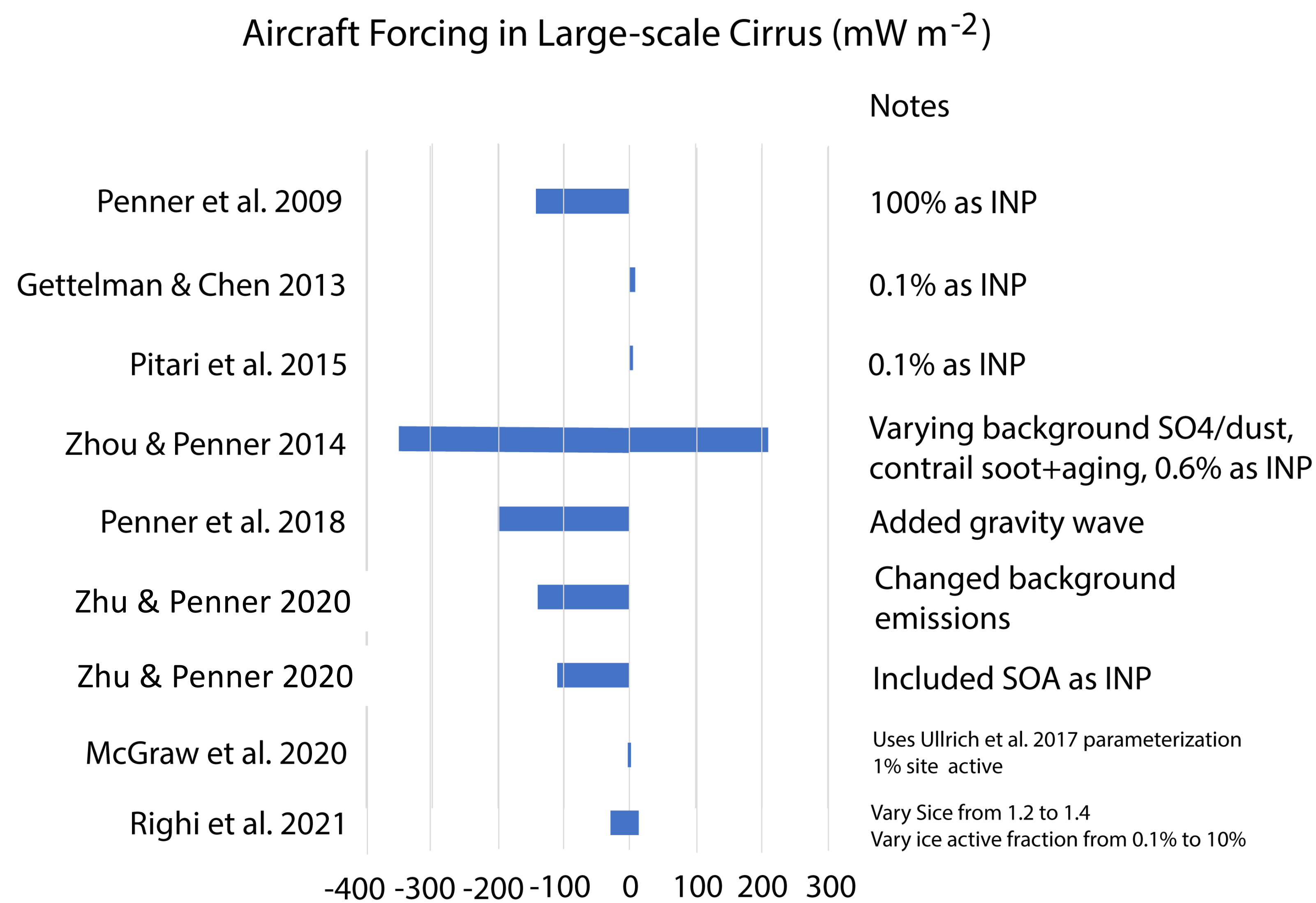
- Homogeneous freezing occurs mainly on sulfuric acid particles, which are high in number concentration at cirrus altitudes - occurs at RHi ≈ 150%
- Heterogeneous nucleation by INP occurs on dust particles and to some extent on soot/organic particles: number concentrations of these IN are much smaller than those of sulfate particles - occurs at RHi ≈ 135%

Adding INP to an area dominated by homogeneous freezing decreases ice number, (but increases ice number when added to regions where heterogeneous nucleation dominates)

Lee et al. (2020) estimates of aviation forcing:



Summary of estimates of aircraft forcing in large-scale cirrus



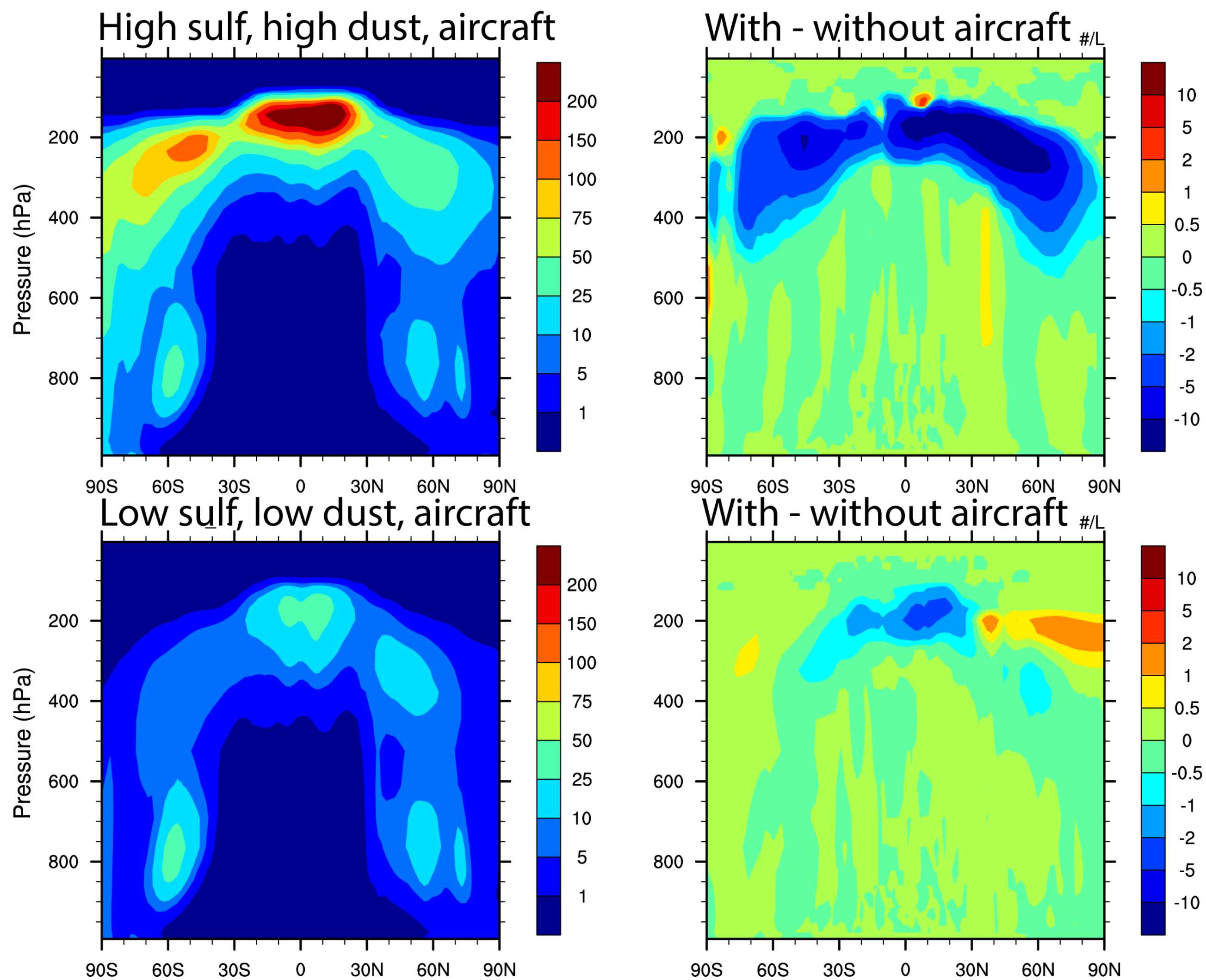
Modeled ice number concentrations (and thus indirect forcing) depends on:

- Predicted aerosol number as INP concentrations
- Treatment of updraft velocity
- Whether ice nucleation is assumed to take place within existing ice clouds or treated as a clear sky phenomena

Methodology

- Here, we use the coupled CAM5/IMPACT model. The IMPACT module simulates a total of 17 aerosol types and/or size bins with sulfate+organics coating associated with all types:
  - 3 sizes representing the number and mass of pure sulfate aerosols (i.e. nucleation, Aitken and accumulation modes)
  - 3 sizes of nucleated SOA aerosols read in from version of IMPACT that treats a mechanistic formation of SOA, formed from the oxidation of a-pinene
  - 3 types of fossil/bio-fuel soot that depend on its hygroscopicity or the amount of sulfate on the soot
  - 1 biomass soot mode
  - 4 dust sizes
  - 4 sea salt sizes
  - 2 aircraft soot modes (preactivated in contrails or not)

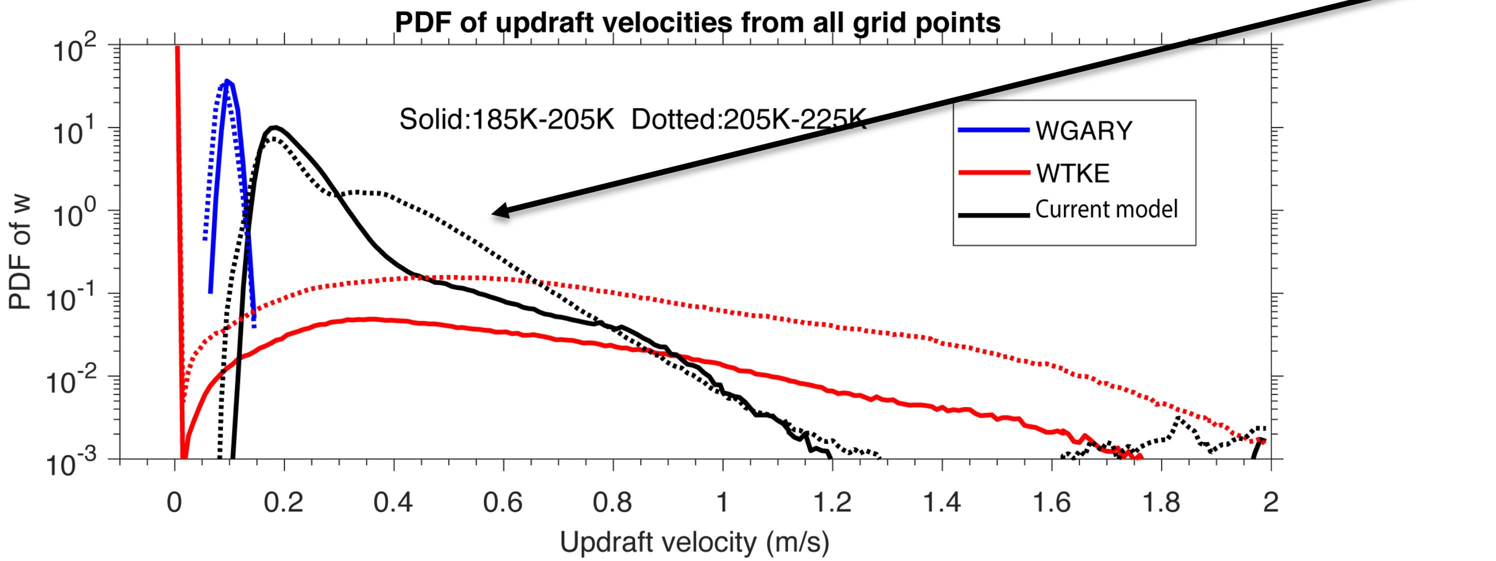
Effect of aircraft soot on ice number



Zhou & Penner, 2014

Treatment of updraft velocities differ in different models

New wave treatment based on Podglajen et al. 2016



Values based on Gary (2006, 2008) are average values based on mesoscale temperature variations from aircraft observations, but are too small, compared to new data from Podglajen et al. (2016)

Examined a number of sensitivies in model: best model includes SOA as an INP

Cases	Shortwave	Longwave	Net forcing
All aircraft - no aircraft	1.77	-1.41	0.36
Contrail-processed soot - no aircraft	0.36	-0.56	-0.20
D100, contrail-processed soot – no aircraft	0.06	-0.08	-0.03
Size-determined SOA, contrail-processed soot - no aircraft (Zhu & Penner, JGR, 2020)	0.32	-0.43	-0.11
Vary Δt, contrail-processed soot - no aircraft	0.26	-0.44	-0.18
Constant updraft: contrail-processed soot– no aircraft	0.51	-0.82	-0.31
Fossil+biomass soot aerosols- w/o (ff BC/OC+bb BC/OC)	0.09	-0.24	-0.15
Biomass: bb BC/OC - w/o bb BC/OC	0.08	-0.17	-0.09
Fossil fuel: ff BC/Oc - ff BC/OC	0.01	-0.07	-0.06

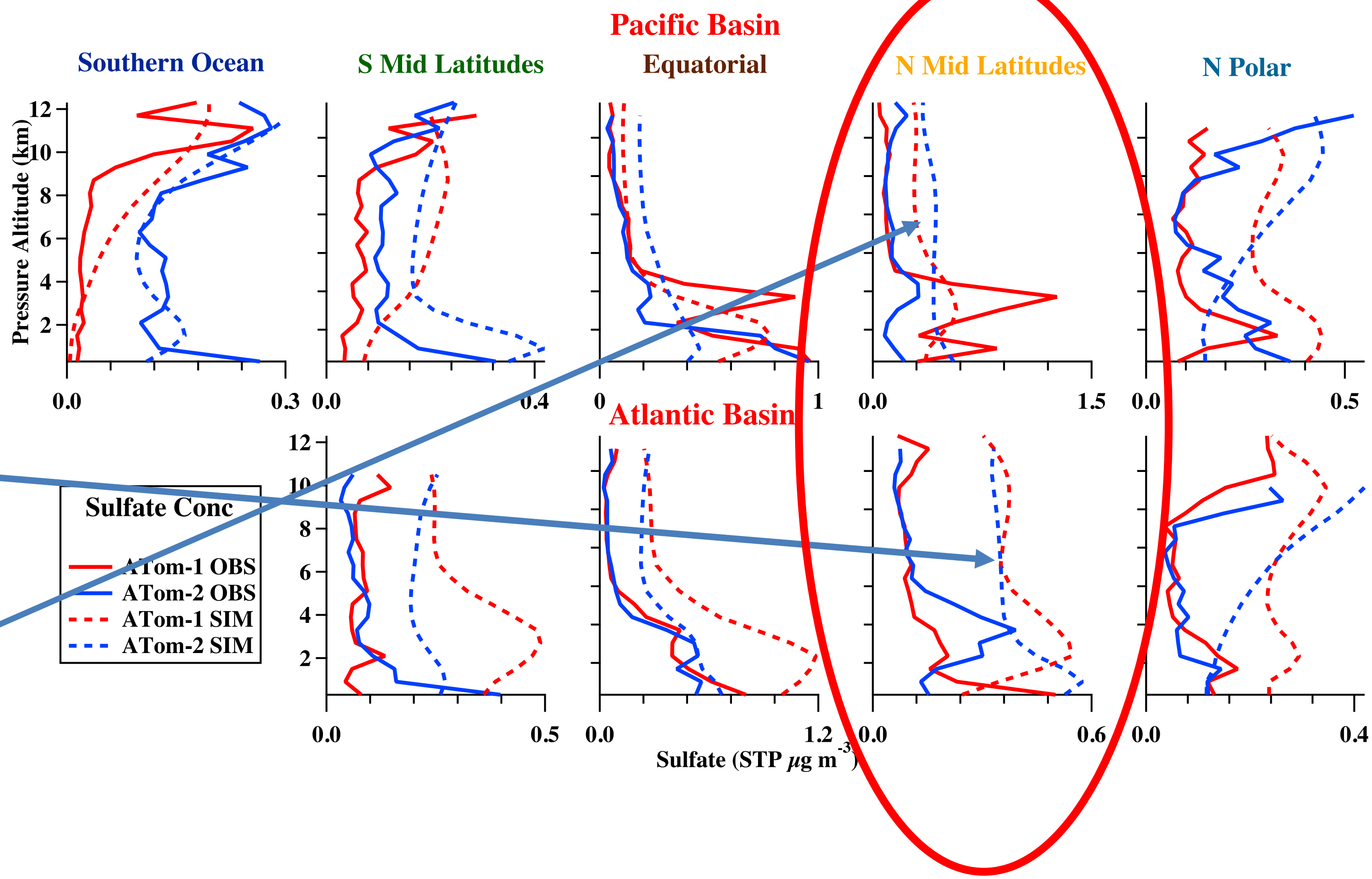
Penner et al. JGR 2018

Are model-predicted aerosol burdens appropriate?

Focus on Northern Hemisphere upper troposphere since that is where CALIPSO sees largest increases in Ni

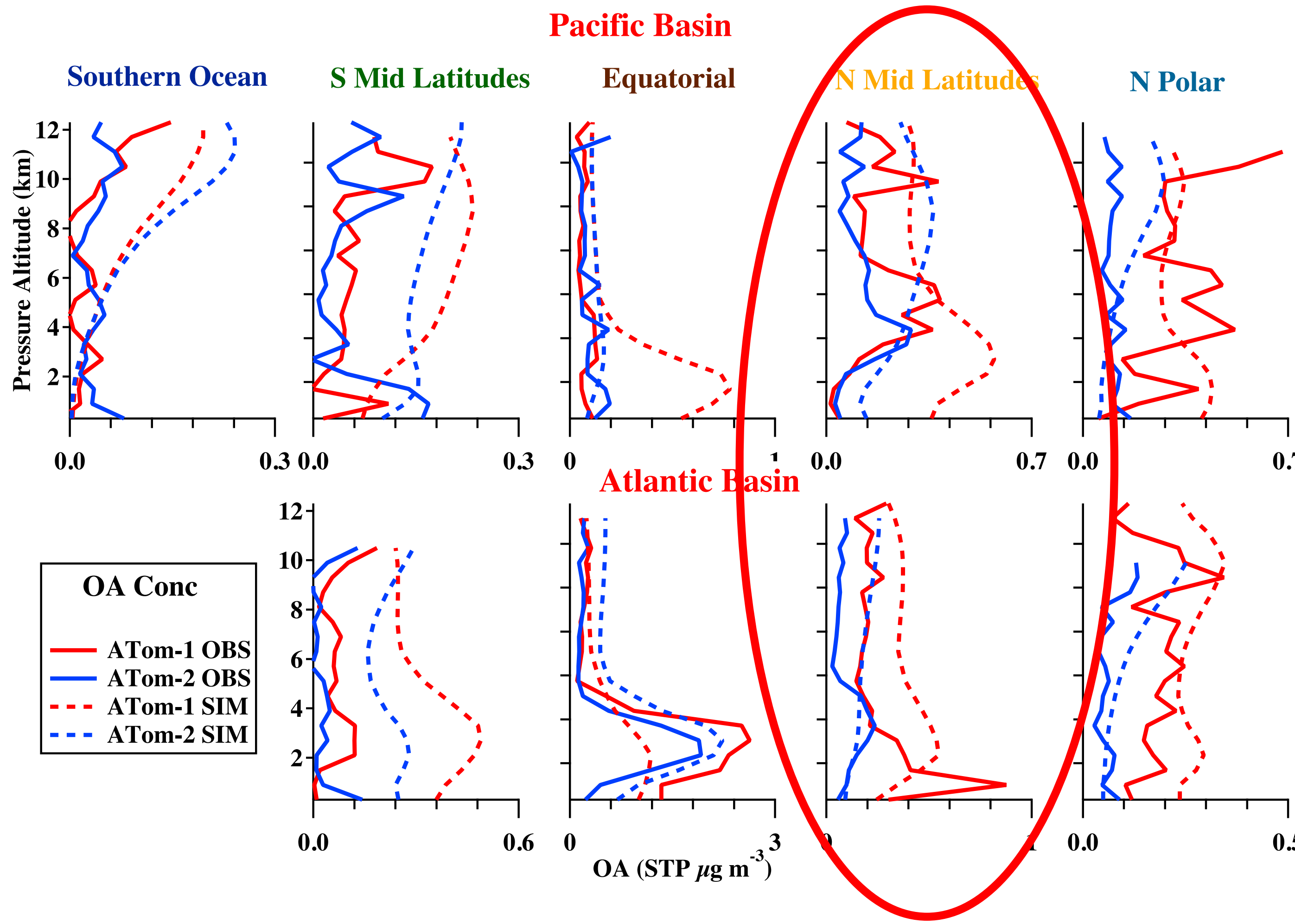
Comparison to AToM: Sulfate mass

For NH mid-latitudes: Sulfate mass in the model is too high in the Atlantic basin, but reasonable in Pacific



Comparison to AToM: Organic Matter

OM is reasonable in upper troposphere NH mid-latitudes of Pac. Basin, and Atlantic



Are model-predicted BC burdens appropriate?

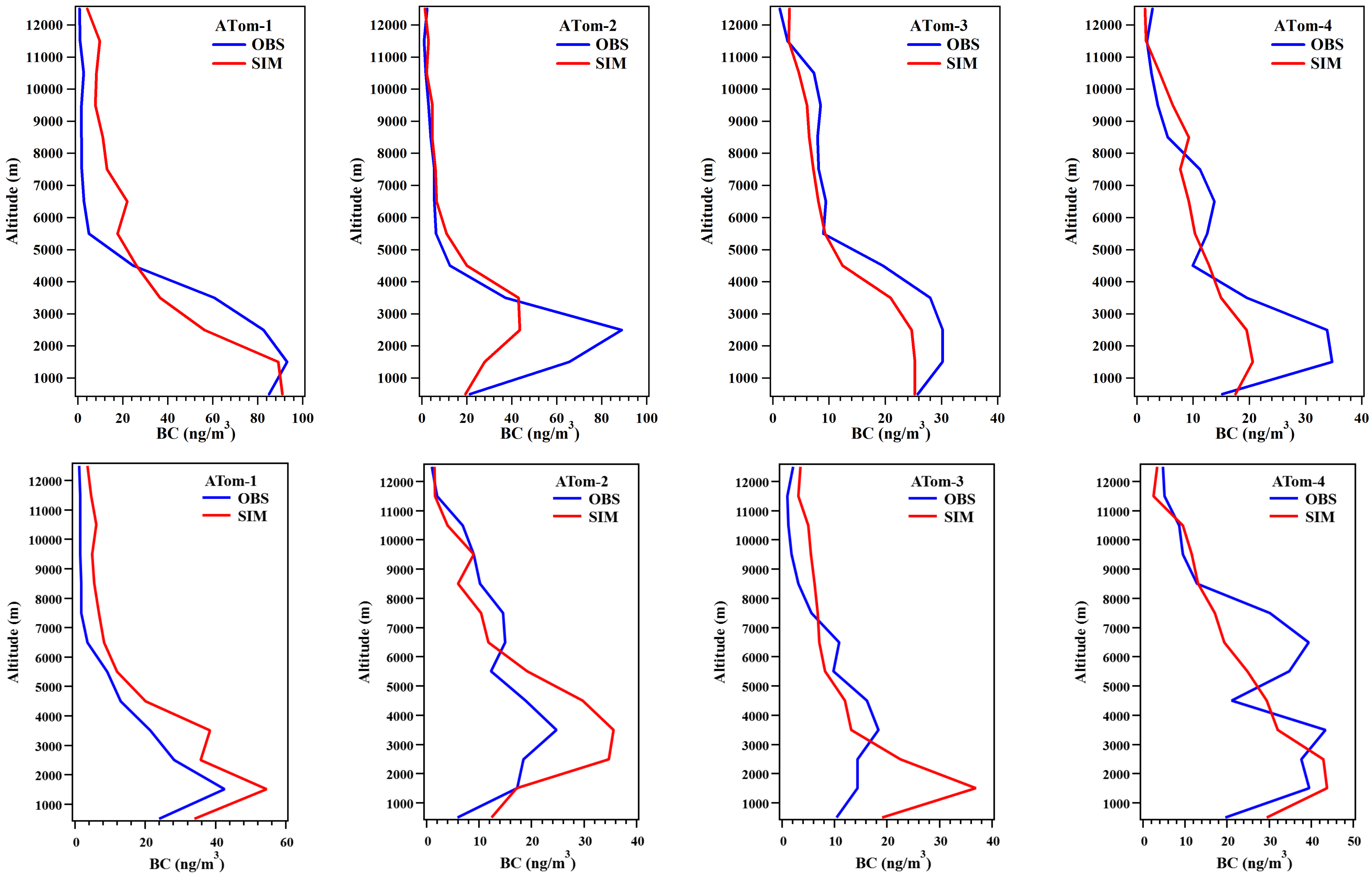
AToM-1 AToM-2 AToM-3 AToM-4

Average all latitudes AToM Observations

30-60N AToM Observations

Model

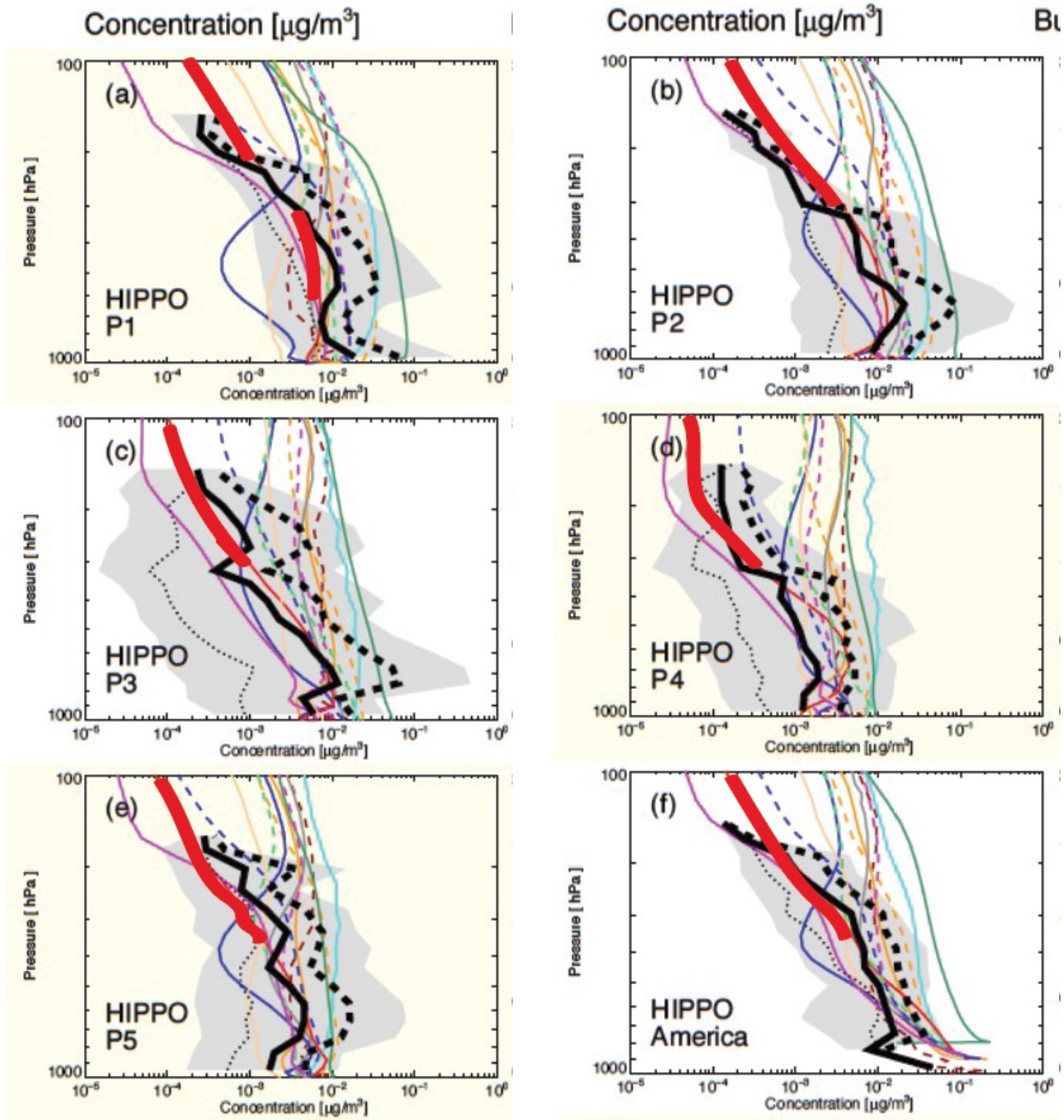
Obs.



Are model-predicted BC burdens appropriate?

P1-P5 are different months for BC concentrations averaged over Pacific; Lower right: over North American

From Samset et al., 2014

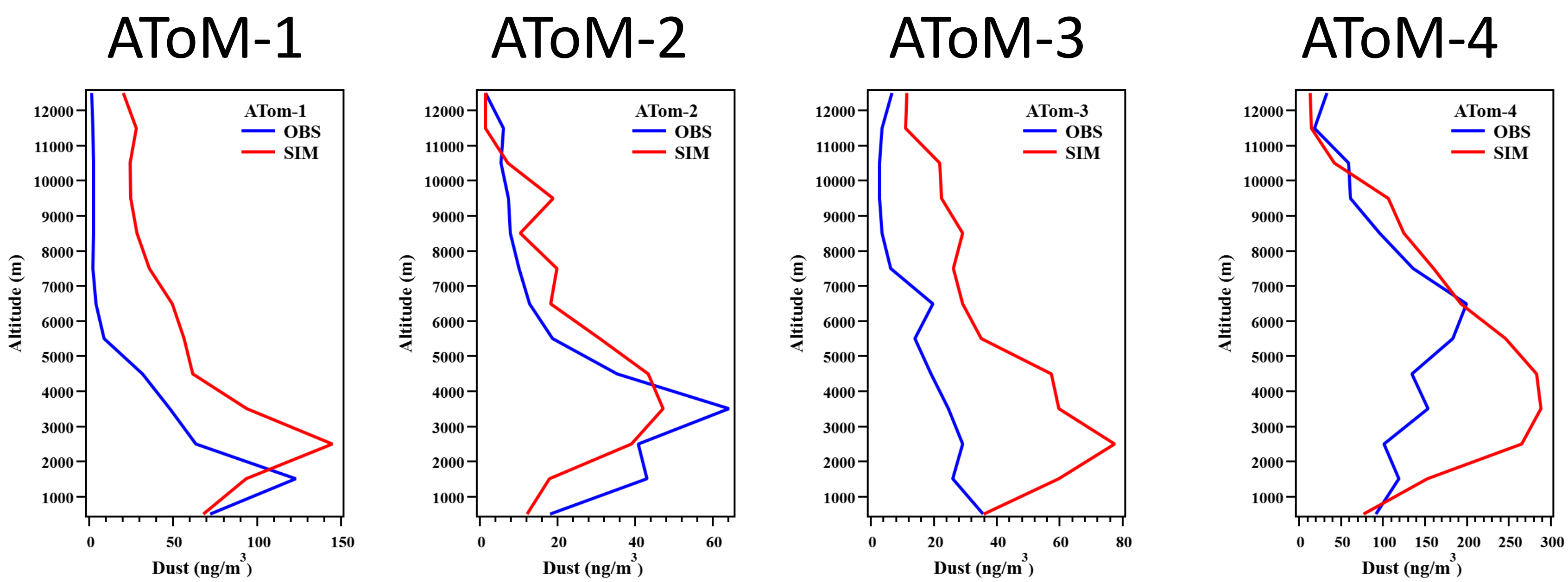


Are model-predicted dust burdens appropriate?

30-60N ATom Observations D < 1 **µm**

Model

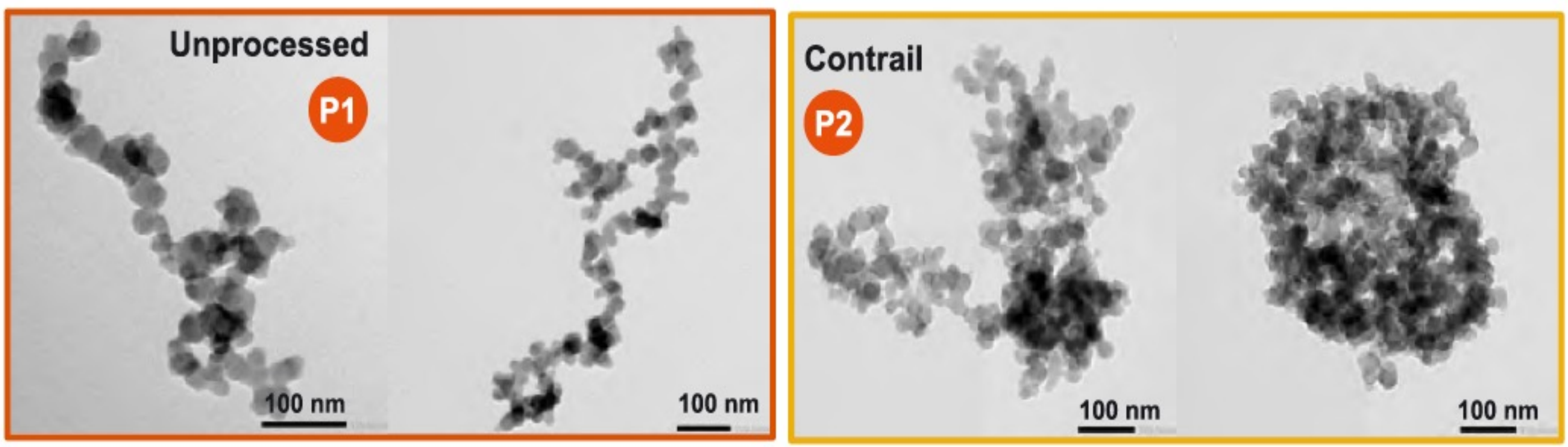
Obs.



Examine laboratory observations relevant to the role of aircraft soot as an INP

Laboratory observations show small INP efficiency for aircraft-like soot unless processed within contrails

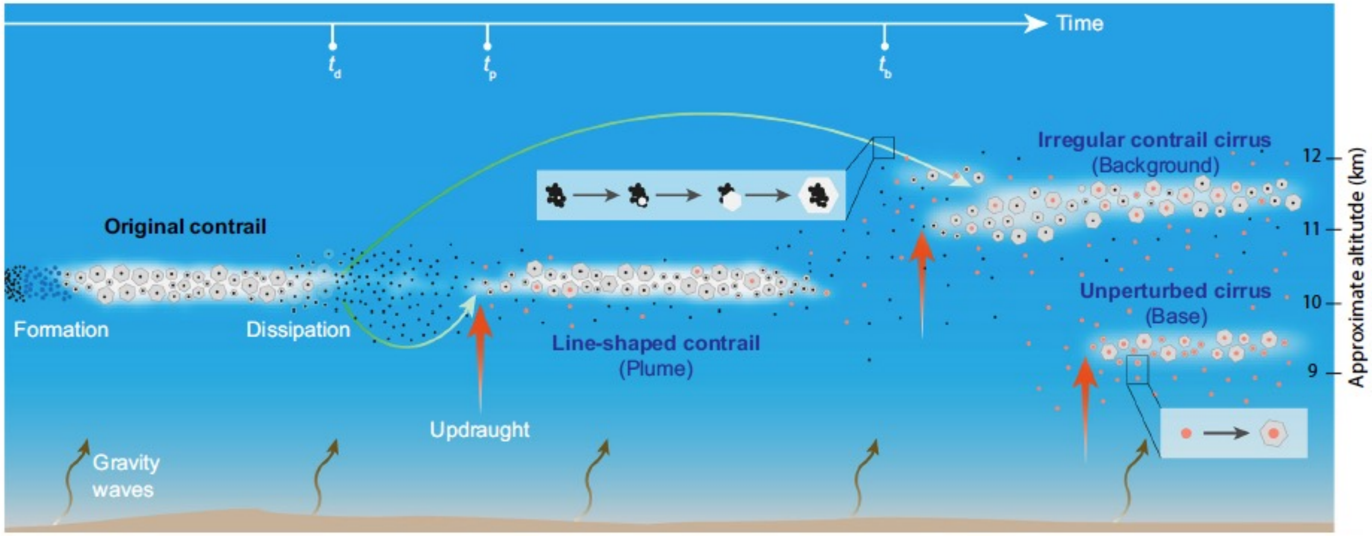
- Mahrt et al. (JGR, 2020) showed that incorporating aircraft-like soot in contrails would increase INP efficiency (due to enhanced pore condensation), but only treated for larger particles (primary diameter 30 nm, with overall D=400 nm,



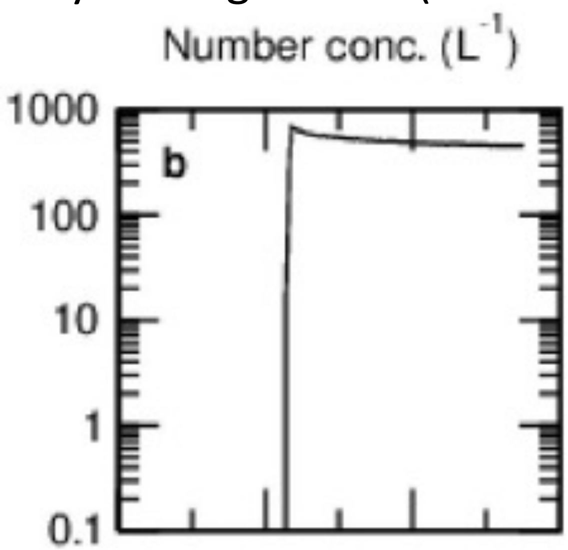
Marcolli et al. (ACP, 2021) used a theoretical treatment to extend to smaller sizes relevant to aircraft soot

- Develop theory that predicts ice formation on soot particles via pore condensation and freezing (PCF)
- During PCF, water is taken up into pores of the soot aggregates by capillary condensation and freezes homogeneously
- Karcher et al. (2021) applied this theory to aircraft soot using a 1-D model found <1% of soot particles form cirrus when homogeneous freezing particles also present

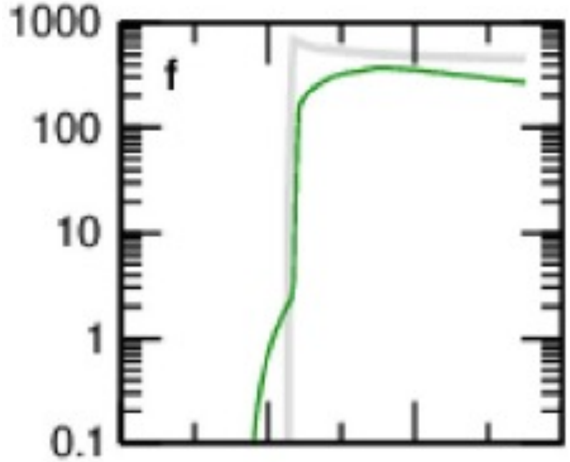
Karcher et al. (2021)



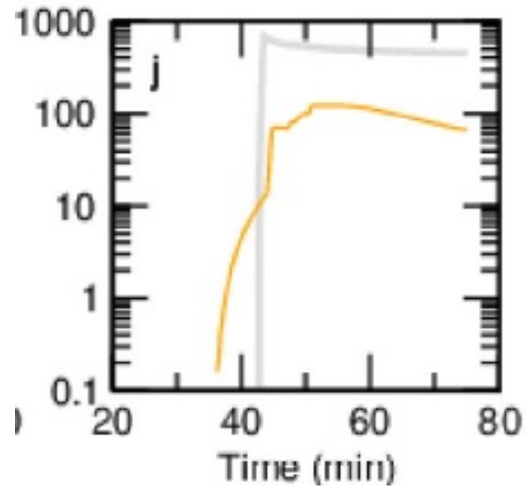
Only homogeneous (no soot)



Plume Ni (0.5 hr)

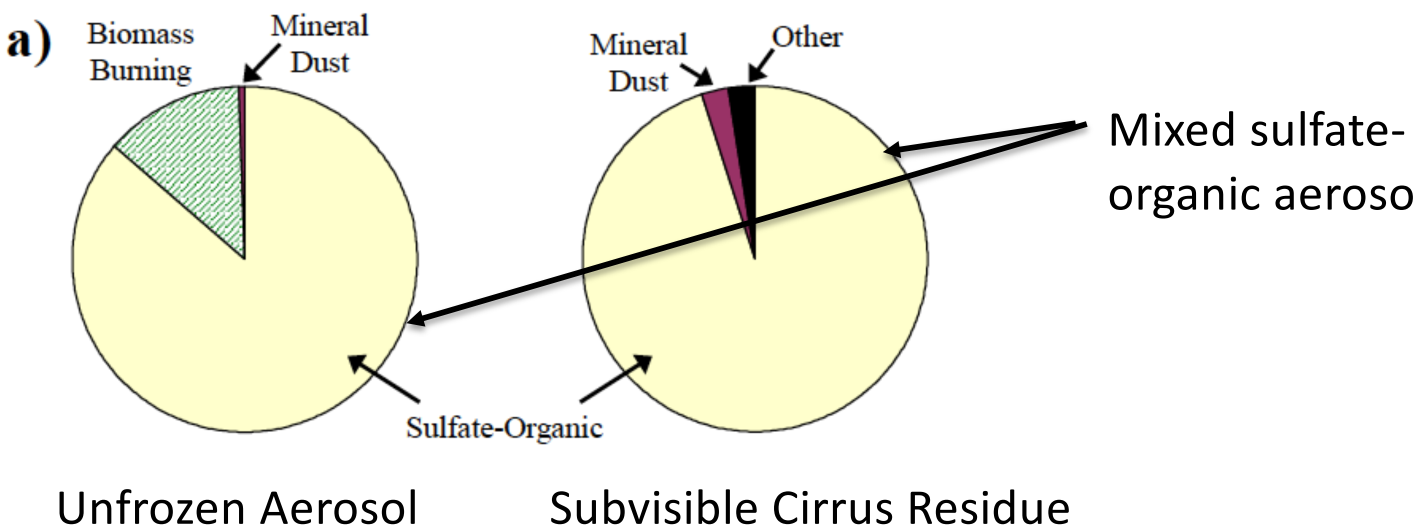


Background contrail cirrus (5 hr)



However, atmospheric 'soot' is likely covered by organics/sulfates, but these also form Ni

- 1 in 200 – 500 internally mixed org/sulfates form subvisible cirrus clouds: (Froyd et al., ACP, 2010; see also Murphy et al., ACP, 2021)



It is possible that the organic coating on aircraft soot becomes glassy and thus may act to form ice

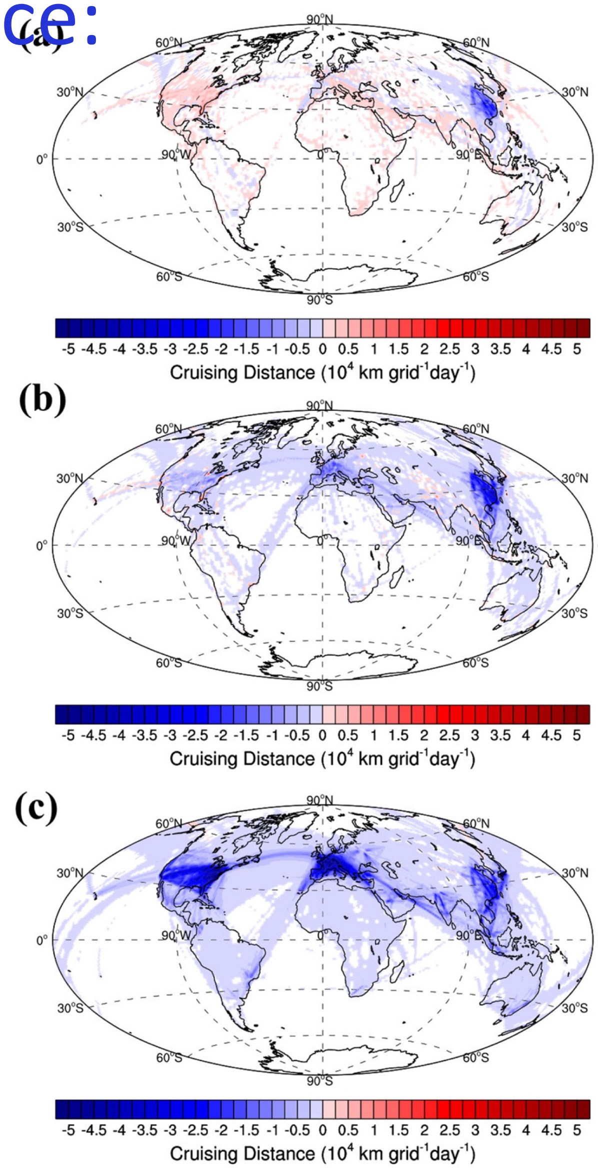
Model results for burden of sulfate/OM on contrail-processed aircraft soot (ng/m²)-April/May

		2018	2019	2020
Sulfate coating	30 - 60N	2.5	2.7	2.3
	0 - 30N	2.1	2.3	2.2
	30S - 0	1.9	2.0	2.0
	60 – 30S	1.4	1.4	2.4
Organic coating: Perhaps organics are good INP				
	30 - 60N	7.1	8.0	6.5
	0 - 30N	11.4	17.6	9.0
	30S - 0	14.4	20.8	9.4
	60 – 30S	8.7	9.2	6.8

Examine recent satellite and field observations that distinguish clouds with high aircraft emissions from those with low aircraft emissions

Difference in aircraft cruise distance:

- Jan/Feb 2020-2019 (-0.3%)
- Mar 2020-2019 (-25%)
- Apr/May 2020-2019 (-73%)

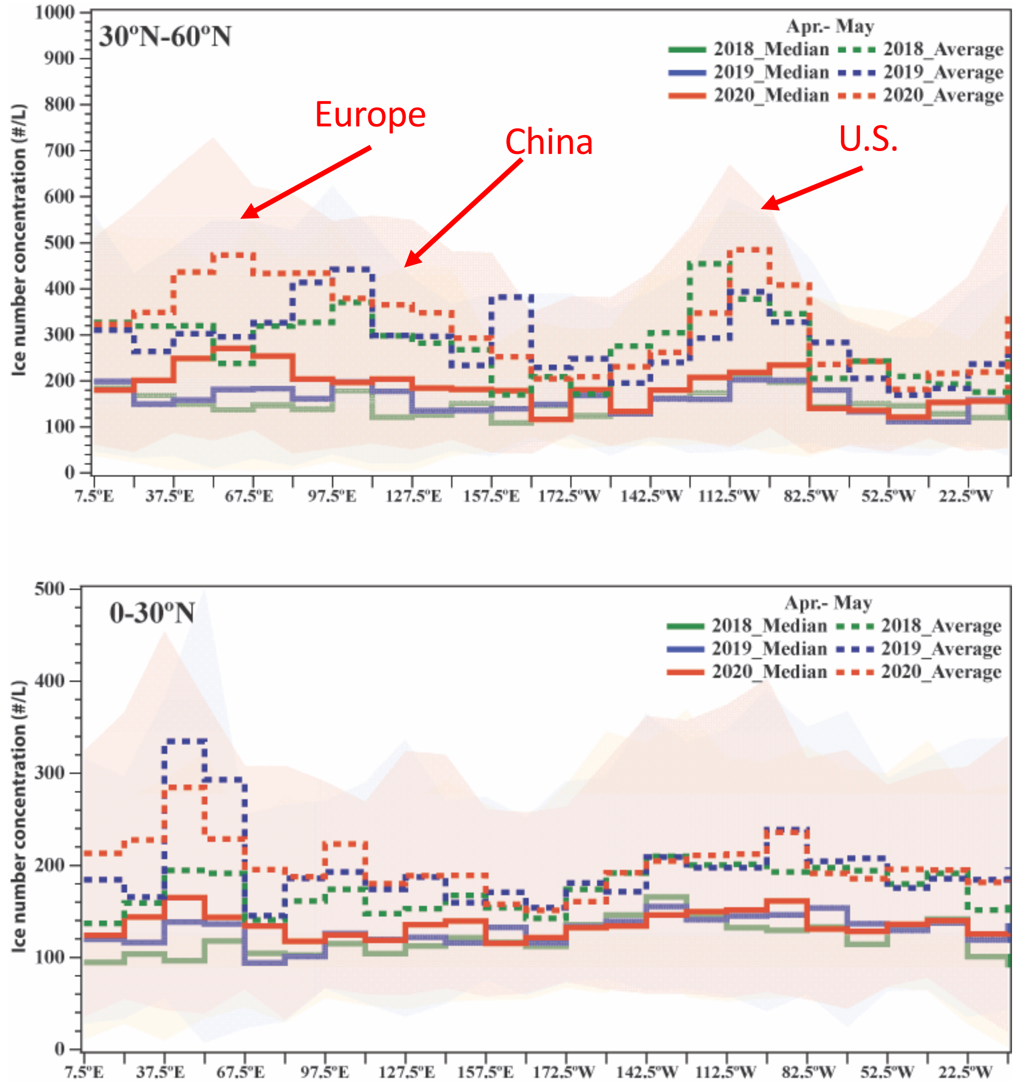


Zhu et al., AGU Advances, 2022

Change in Ni observed by CALIPSO:

- In April and May, median and average Ni values for 30-60N, especially from 8E to 70E (Europe) are higher in 2020 than either 2019 or 2018

- We examined whether the decrease in INP from aircraft allowed more homogeneous nucleation and caused the increased ice crystal number using the model



The difference in Ni measured by CALIPSO in April-May

Differences in Red are significant at a 95% confidence level

The difference in Ni values in April-May in 2020 are significantly larger than 2018-2019 and 2010-2019 for both the average and the median Ni for 30N-60N

AVE1 is 2018-2019

AVE2 is 2010-2019

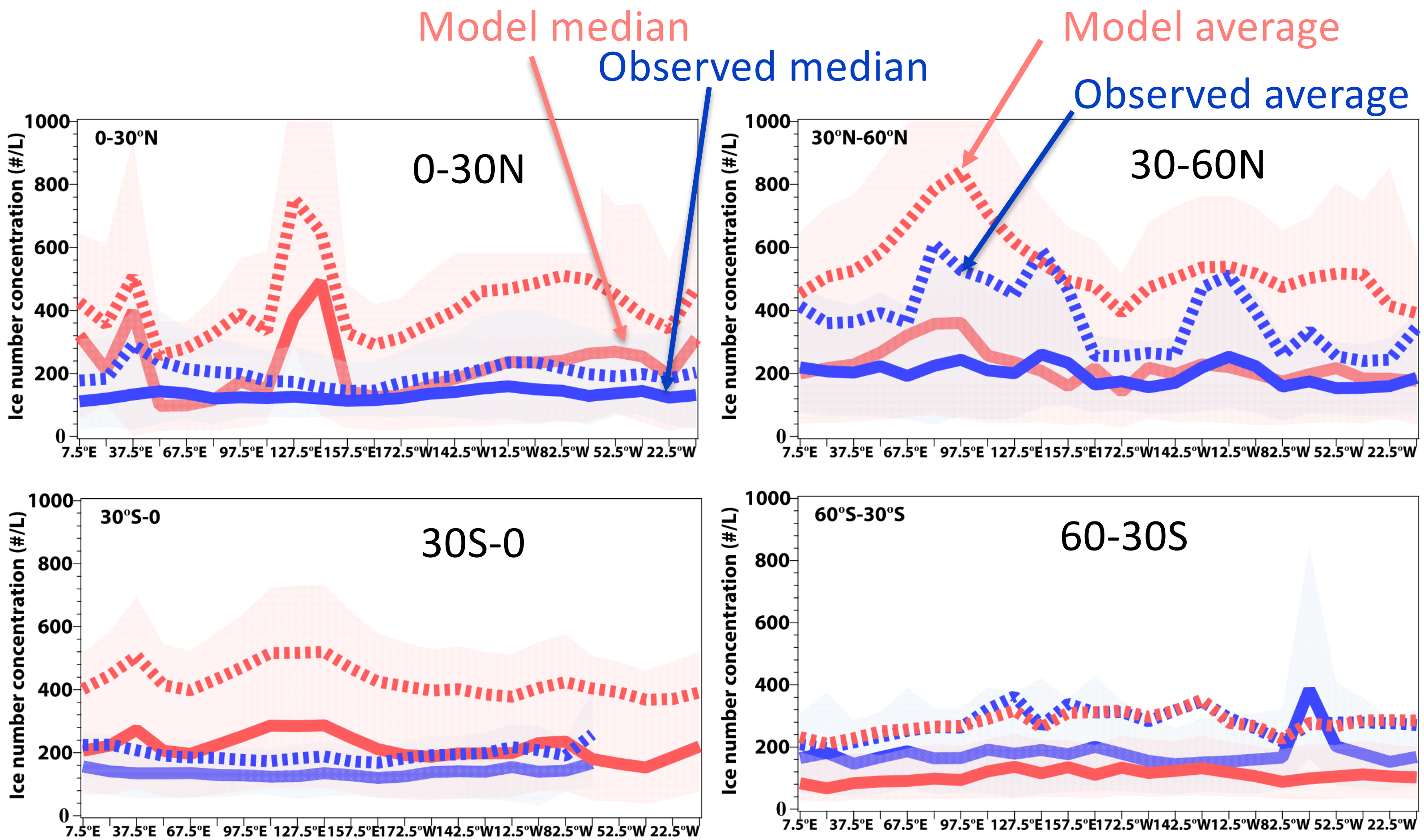
Median

	2019-2018	2020-2018	2020-2019	2020-AVE1*	2020-AVE2#
30°N-60°N	9.70	***36.24***	26.54	***31.39***	***24.21***
0°-30°N	9.93	15.55	5.61	10.58	***9.07***
30°S-0°	-7.03	-4.60	2.43	-1.09	4.69
60°S-30°S	-18.89	7.85	26.74	17.30	***11.63***

Average

	2019-2018	2020-2018	2020-2019	2020-AVE1*	2020-AVE2#
30°N-60°N	7.45	***42.15***	***34.71***	***38.43***	***29.68***
0°-30°N	***22.06***	***25.41***	3.35	***14.38***	***12.06***
30°S-0°	40.43	40.45	0.01	4.19	6.56
60°S-30°S	-24.45	5.53	29.98	17.76	7.75

Comparison with model: Average ice crystal numbers are higher than observations, but median values agree well:



- Comparison of CALIPSO and Model results for 2020 – 2018/2019 with observations
- Focusing on 30-60N: Median differences from model are smaller than observations

Median-CALIPSO

	2019-2018	2020-2018	2020-2019	2020-AVE1*	2020-AVE2#
30°N-60°N	9.70	***36.24***	26.54	***31.39***	***24.21***
0°-30°N	***9.93***	***15.55***	5.61	***10.58***	***9.07***
30°S-0°	-7.03	-4.60	2.43	-1.09	4.69
60°S-30°S	-18.89	7.85	26.74	17.30	***11.63***

Median-Model

	2019-2018	2020-2018	2020-2019	2020-AVE1*	2020-AVE2#
30°N-60°N	-0.50	18.34	18.84	18.59	--
0°-30°N	3.35	17.9	14.55	16.23	--
30°S-0°	-9.79	-9.06	0.73	-13.45	--
60°S-30°S	22.16	0.87	-21.29	***-10.21***	--

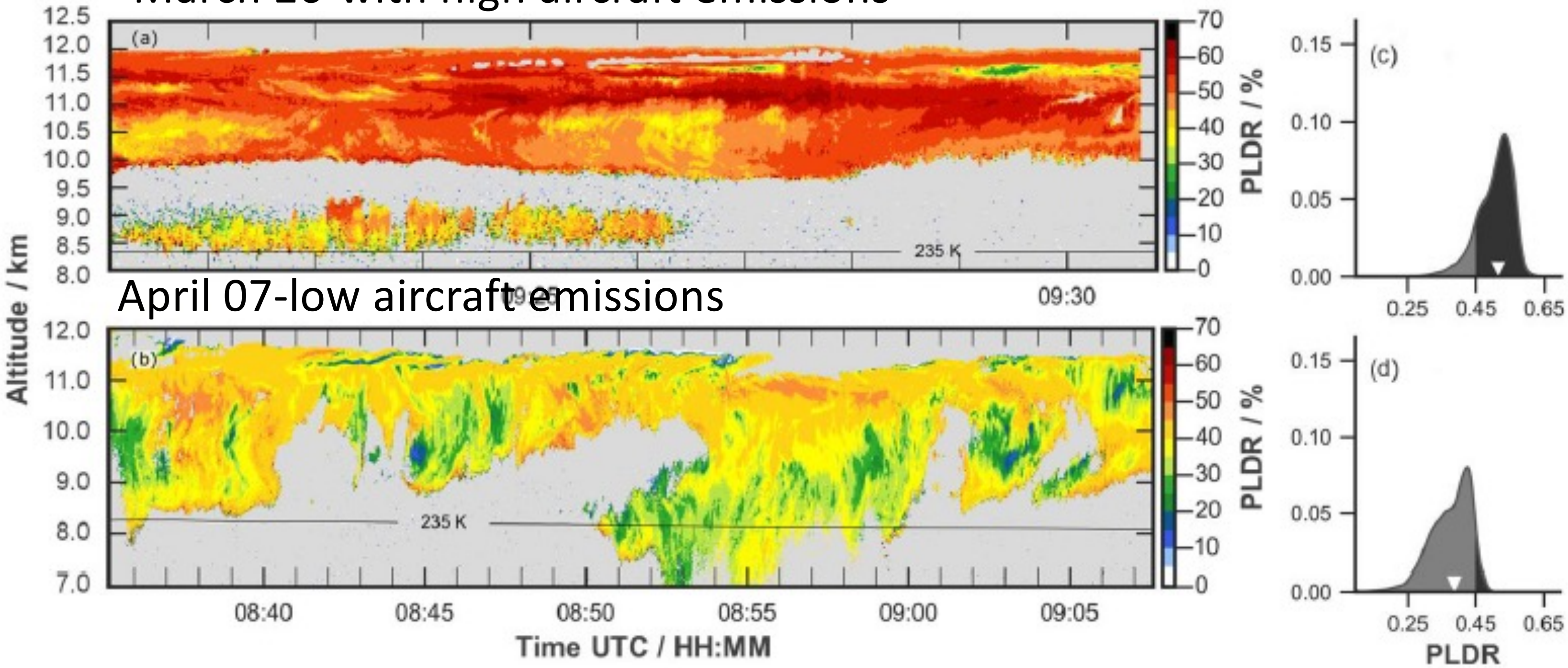
Recent observations of the effect of aircraft INP on Ni over Europe

- Gross, Kramer et al. (2022) used combined airborne lidar and in-situ ice cloud measurements to investigate naturally formed cirrus clouds, which either formed under influences of aerosol emissions from aviation or which formed under pristine conditions
- Clouds, which are more affected by aviation aerosol emissions exhibit larger mean effective ice particle diameters connected to decreased ice particle number concentrations
- The sign of the change in Ni is consistent with Zhu, Penner, et al. (2022) (lower Ni when clouds are impacted by aerosol)

Case study contrail-cirrus polarization ratio in 2014: region with high PLDR are regions with high aircraft

March 26-with high aircraft emissions

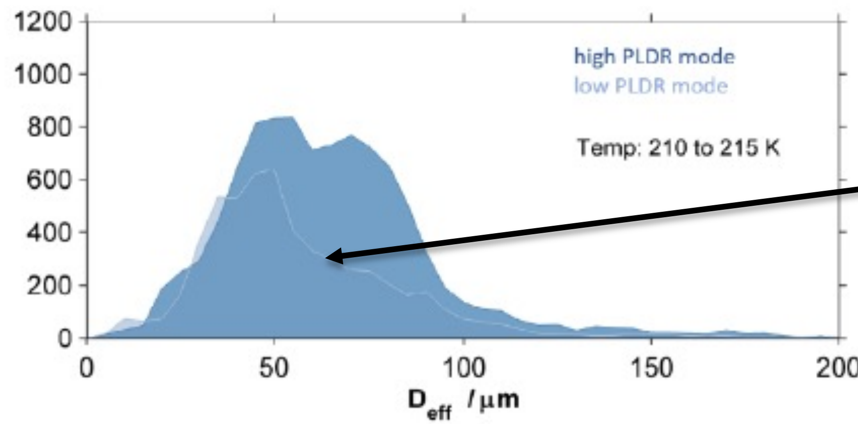
April 07-low aircraft emissions



PLDR = Prticle linear depolarization ratio

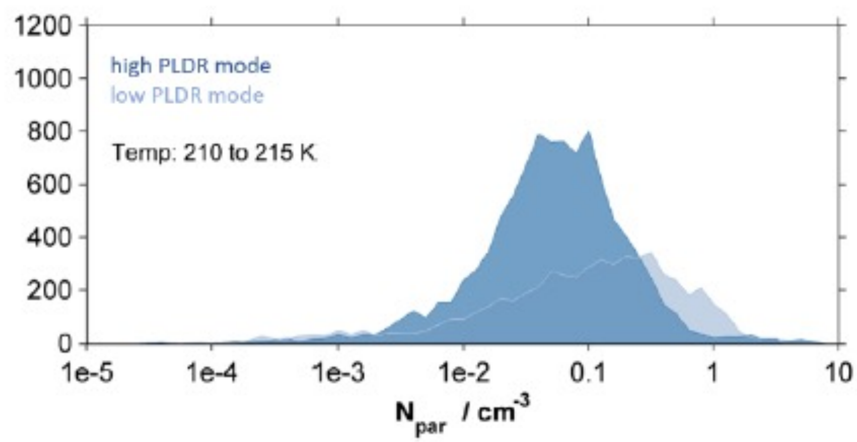
Since T also affects PLDR, and want same amount of data, examine D and Ni only 210-215K

D_{eff}



Hard to see, but median Deff = 50.7 µm low PDLR
61.4 µm high PDLR

Ni



Ni 0.11 for low PDLR
0.05 for high PDLR

Gross, Kramer et al. (2022) consistent with:

- Homogeneous freezing plays larger role in low PLDR clouds that are less impacted by aircraft
- This means that homogeneous freezing is expected to result in higher Ni and smaller crystal sizes as found in Zhu, Penner et al. (2022) during COVID-19

Conclusions

- Considerable progress in aerosol models showing that aging can mix sulfates and organics on aircraft soot (consistent with observations)
- Effects of aircraft soot can be positive or negative in models, depending on the amount of aircraft soot that act as INP and background aerosols, but new field and satellite data favor aircraft soot causing lower Ni (negative forcing)
- Our best estimate for aircraft forcing is -0.11 W/m^2 is consistent with the sign of Ni changes in 2020 in NH midlatitudes, but our predicted 2020 change is smaller than the satellite data
- Further work is needed to match all observations (especially in tropics and SH mid-latitudes)