Climate Impact of Anthropogenic Aerosols on Cirrus Clouds

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Outline

- Introduction: cirrus effects of aerosols vs warm-cloud effects
- Discussion of aspects contributing to differences in cirrus ice number concentrations in different models
- Introduction of wave formulation for cirrus formation
- Results for aircraft soot forcing with sensitivity tests
- New method for treating ice formation for forcing when haze particles change
- Examination of the effects of COVID-19 decreases in aircraft flights

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

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

- Predicted aerosol number concentrations
- Treatment of updraft velocity
- Water vapor deposition coefficient
- 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 associated with all types:
 - 3 sizes representing the number and mass of pure sulfate aerosols (i.e. nucleation, Aitken and accumulation modes),
 - 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)
 - SOA read in from version of IMPACT that treats a mechanistic formation of SOA, including the formation of newly nucleated SOA in 3 sizes formed from the oxidation of a-pinene
- Standard CAM5 aerosol scheme uses 3 internally mixed modes: Aitken, Accumulation, Coarse; BC is added to Accumulation mode;

Concentrations of pre-activated aircraft soot IN: 0.6% of total aircraft soot



Effect of aircraft soot on ice number



Treatment of updraft velocities differ in different models



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.

Previous attempts to treat nucleation of ice particles

- All efforts other than Penner et al. (2009) assumed a single fixed and constant updraft velocity at each grid point (Penner used 30 updrafts/grid point with a pdf as measured in the upper troposphere)
- Assuming a constant updraft is not a true representation of gravity waves that form cirrus:



Penner et al, JGR, 2018

Const. W vs. Waves in CAM5



Aerosol treatment in CAM/IMPACT model

- Homogeneous freezing nuclei: Pure sulfate aerosol (although likely has SOA in it; Zhu et al., PNAS 2017)
- Heterogeneous INP:
 - Dust particles with less than 3 monolayers of sulfate coating
 - Contrail-processed aircraft soot with less than 3 monolayers of sulfate coating
 - 0.1% of biomass burning aerosols
 - 0.1% hydrophilic/0.05% hydrophobic fossil fuel soot
 - Assume accumulation mode newly nucleated SOA are heterogeneous IN

Cloud fraction-weighted homogeneous freezing occurrence frequency



Summary of estimated forcing in cirrus clouds (6 yr averages) with Podglajen et al. (2016) waves

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	0.32	-0.43	-0.12	
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	

When all dust particles are INPs, forcing is small negative

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If assume newly formed SOA is an INP, the forcing by aircraft is halved

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Contrail-processed soot – no aircraft with SOA			
as an INP	0.36	-0.47	-0.10

One contentious issue is whether aircraft soot is actually a good INP

- We only calculate that 0.6% of aircraft soot may be an INP, yet, it is the largest INP forcing agent in cirrus clouds
- Thus, we examined cirrus clouds in April-May 2020, relative to earlier years to understand whether aircraft soot may be an effective INP
- We used ice crystal number concentrations measured by CALIPSO in 2018, 2019, 2020 and compared these with model simulated ice crystal numbers

Difference in aircraft cruise distance:

• Jan/Feb 2020-2019 (-0.3%)

• Mar 2020-2019 (-25%)

• Apr/May 2020-2019 (-73%)





- 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
- The decrease in INP from aircraft has allowed more homogeneous nucleation and, thus, increasing the ice crystal number





The 2020, 30N-60N, values of Ni are unusual in last 10 years:



Differences in Ni measured by CALIPSO

		The difference in Minicasured by CAER 50				
icant at a			2019-2018	2020-2018	2020-2019	2020-AVE
a student'	Ś	60°S-60°N	3.44	-1.59	-5.04	-3.32
are	Median	30°N-60°N	19.31	-5.37	-24.68	-15.02
	weulan	0°-30°N	-2.56	-2.93	-0.38	-1.66
		30°S-0°	0.52	2.72	2.19	2.45
	Jan-Feb —	60°S-30°S	-4.00	-0.08	3.92	1.92
8, 2019	Jan-reb	60°S-60°N	3.26	-2.61	-5.87	-4.24
nedian	A	30°N-60°N	22.68	6.19	-16.49	-5.15
	Average	0°-30°N	7.69	-6.86	-14.55	-10.7
		30°S-0°	-4.45	-9.95	-5.51	-7.73
		60°S-30°S	-12.88	0.19	13.08	6.64
		60°S-60°N	-1.28	13.95	15.23	14.59
	Median	30°N-60°N	9.70	36.24	26.54	31.39
	Wicdian	0°-30°N	<i>9.93</i>	15.55	5.61	10.58
		30°S-0°	-7.03	-4.60	2.43	-1.09
۸n	Apr-May —	60°S-30°S	-18.89	7.85	26.74	17.30
	Api-iviay	60°S-60°N	3.00	20.01	17.01	18.51
	Average	30°N-60°N	7.45	42.15	34.71	38.43
		0°-30°N	22.06	25.41	3.35	14.38
		30°S-0°	40.43	40.45	0.01	4.19
		60°S-30°S	-24.45	5.53	29.98	17.76

The difference in Ni measured by CALIPSO

Differences in **Red** are significant at a 95% confidence level using a student's T-test

Ni values in April-May 2020 are significantly larger than 2018, 2019 and their average for both median and average Ni for 30N-60N

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

Values are the average of Jan-May in 2018-2020



Modeled occurrence frequency of homogeneous nucleation (Jan – May 2018-2020)



The cloud weighted occurrence frequency of homogeneous nucleation is <15% with average of 2.2%



When restricted to CALIPSO measurement conditions (0.3< τ_c <3), the frequency is < 7% with largest values in polar regions.

CALIPSO may miss infrequent high concentrations associated with homogeneous nucleation due to its infrequent revisit times and restricted viewing conditions, which may explain why model average N_i is higher than CALIPSO average

This fits where Ni from CALIPSO is largest

 Median Ni (L⁻¹) for 1st 5 months of 2020



50 150 250 350 450 550

Conclusions

- Considerable progress in aerosol models now shows that assuming internal mixing for all aerosols (as in CAM5) will bias results
- Use of constant updraft velocity to treat ice formation in cirrus clouds biases results
- Effects of aircraft soot can be positive or negative, depending on the amount of aircraft soot that act as INP
- Our best estimate for aircraft forcing is -0.14 (HYBRID) to -0.2 (KL) W/m² when treating contrail-processed soot using the Podglajen parameterization (but this decreases to close to zero if 100% of dust or to -0.1 Wm⁻² if SOA is a heterogeneous IN).
- Effects of biomass burning soot INP and fossil fuel burning soot INP are both small (and negative) (-0.09 W/m2 and -0.06 W/m2)
- The total PD-PI effect of all aerosols on cirrus clouds is -0.2 W/m2 (HYBRID)
- Further work is needed to match all observations
- Results from our model are corroborated by 2020 CALIPSO observations