The Relationship Between Cloud Droplet Number Concentrations and Anthropogenic Pollution: Observations and Climate Implications

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Background

- Works cited:
  - Albrecht, Charlson, Pruppacher/Klett, Twomey (Wigley, Slingo)
- What did *WE* know before?
  - Increased sulfate (in theory) should increase cloud reflectance for thin clouds, with absorption increasing less (Twomey)
  - A variety of physical mechanisms determine the shape of droplet distributions; collision, coalescence, adsorption, absorption (Leaitch and Isaac, Hoppel)
  - Over ocean, Fractional Cloudiness (consequently: LWC) is hypothesized to increase through suppression of precipitation, due to an increase droplet N (Albrecht)
  - Droplet energetics are dominated by surface tension and dilution effects for salts (so [C]), function of radius (Kohler)
  - Ocean DMS production influences the background, “clean-air” sulfate concentrations *(how do “background” continental*

Objectives…

- Establish (measure) relationship between pollution ($SO_4^=$ proxy – justified later…) and Droplet Number/LWC in Stratiform and Cumiliform clouds
- Albrecht study over marine layer (20<N<100), this study over urban land (Table 1: 110<N<510 cm^{-3})

TABLE 1. Seasonal and Combined Percentile Data for Cloud Sampling

Project	Percent	Altitude km	LWC $g\ m^{-3}$	CDNC cm^{-3}	Temperature °C	$nEq\ m^{-3}$ cw$SO_4^=$	$nEq\ m^{-3}$ cwNO_3^-
Summer	25	1.5	0.18	260	3.7	17	8.0
1982	50	2.1	0.30	360	7.2	34	15
(19)	75	2.8	0.61	400	12	63	99
Winter	25	1.0	0.08	110	−10	17	4.9
1984	50	1.3	0.11	190	−7.4	36	23
(27)	75	1.8	0.17	250	−3.8	57	46
Autumn	25	1.2	0.14	200	−1.7	15	8.1
1984	50	1.5	0.23	240	5.7	22	14
(17)	75	1.8	0.33	270	8.0	56	34
Summer	25	1.5	0.18	240	7.6	7.6	3.6
1988	50	2.0	0.26	350	10.	16	10
(29)	75	2.3	0.37	510	14.	50	36
All	10	1.0	0.09	110	−9.8	4.0	2.6
(92)	25	1.2	0.12	170	−5.1	15	7.4
	50	1.8	0.19	250	4.5	25	15
	75	2.1	0.34	380	11	57	48
	90	2.7	0.53	510	14	130	100

The values are strictly percentiles; no relationships among the different quantities should be inferred from this table. The values in parentheses indicate the effective number of samples used. LWC is liquid water content, and CDNC are cloud droplet number concentrations.

Approach…

- 4 intensive studies: 4 seasons, 1 location (Ontario, Canada), 6 years, 400 samples, averaged over 300m sections, compressed to 85 data points.
- Statiform (59), cumilform(26), supercooled and warm, each sample averaged over size ~2-40km
- Sulphate, Nitrate, LWC (~>10μm), cloud droplet number (FSSP 1 to 35μm) and aerosol (0.17μm to 2.4μm)
- $SO_4^=$ data normalized to divide out LWC, so that concentrations can be expressed per volume of air (*what about interstitial? …R_{drop}<10_m?)*
  - Final concentrations expressed at 0C, 1 atm *(adiabatic, saturated adiabatic compression assumed?)*,
  - …and ultimately treated as a measure of ground based dry aerosol concentration *(is this a valid assumption? Kohler…gas-phase oxidation, cloud processing…)*

Instruments

- ASASP – Active Scattering Spectrometer Probe
  - “intrusive”: Air is forced into a deceleration chamber, then pumped (1 cc/min) to a heated detection area (for drying and to prevent icing).
  - Problems in Cloud: heating is ~ineffective when chamber gets too wet
- FSSP – Forward Scattering Spectrometer Probe
  - “non-intrusive”: samples passing air at 20-40 cc/min
- Slotted-Rod – cloud water collections
  - Problem: Bias towards larger drops

Observations

- What they didn't see….
  - Strong temperature/cloud droplet number relationship.
    - At/Near cloud base: Expect an increased N (a decreased R) to cause a decrease in T, due to a heightened supersaturation threshold
    - Observations show the opposite or nothing, possibly because southerly winds (warmer) correlate with higher N (more pollution).
    - Logic: Anthropogenic aerosols are generally associated with

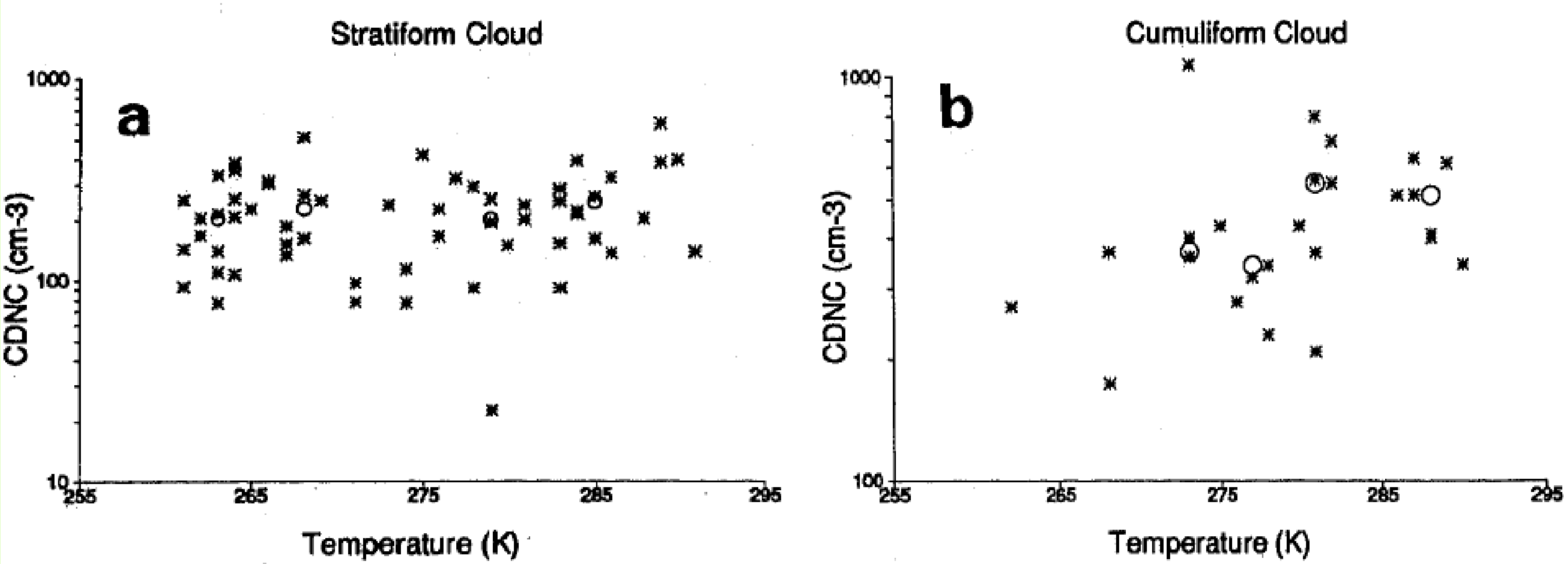


Fig. 5. The CDNC plotted as a function of the cloud temperature for the data split between stratiform and cumuliform cloud. The circles represent the median CDNC for each temperature quartile.

Observations

- What they didn't see….
  - Sulphate/LWC relationship (assume no precipitation effect…)
    - Expected dependence???
    - To be discussed later: T and LWC dependence btw clean and polluted cases

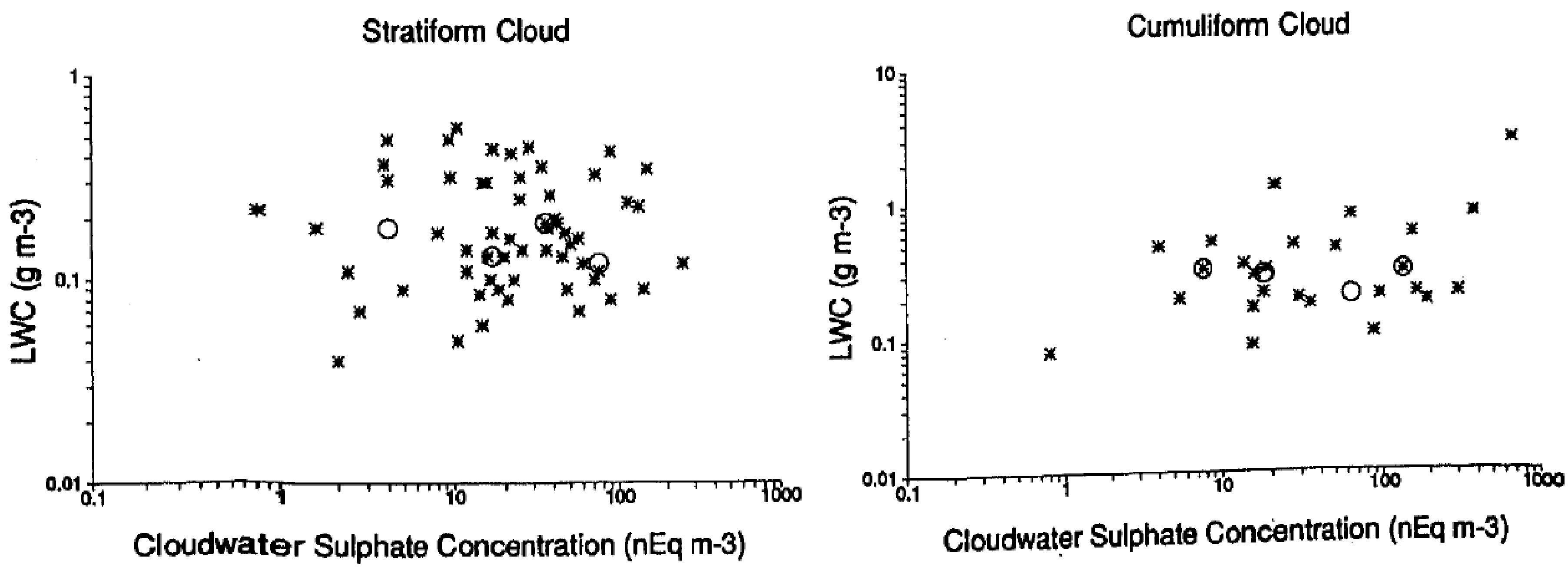


Fig. 7. The LWC plotted as a function of $cwSO_4^=$ for the data split between stratiform and cumuliform cloud. The circles represent represent the median LWC for each $cwSO_4^=$ quartile.

Observations

- What they did see….

$$\log(\mathrm{CDNC})_S = 0.257(\pm 0.052)\log(\mathrm{cwSO}_4^{=}) + 1.95(\pm 0.21) \quad (1)$$

$$\log(\mathrm{CDNC})_C = 0.186(\pm 0.038)\log(\mathrm{cwSO}_4^{=}) + 2.33(\pm 0.13) \quad (2)$$

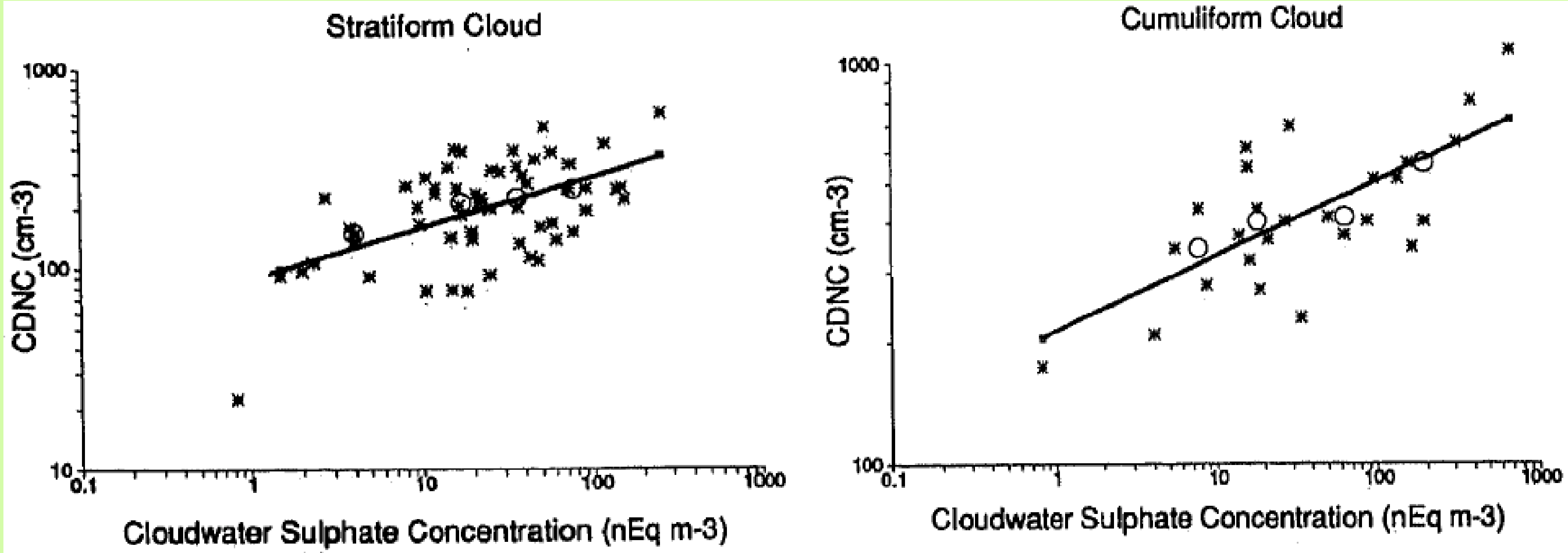


Fig. 4. The CDNC plotted as a function of the $\mathrm{cwSO}_4^{=}$ for the data split between stratiform and cumuliform cloud. The circles represent the median CDNC for each $\mathrm{cwSO}_4^{=}$ quartile and the solid lines represent regressions of the bulk data.

Observations

$$\log(\mathrm{CDNC})_S = 0.257(\pm 0.052)\log(\mathrm{cwSO_4^{=}}) + 1.95(\pm 0.21) \quad (1)$$

$$\log(\mathrm{CDNC})_C = 0.186(\pm 0.038)\log(\mathrm{cwSO_4^{=}}) + 2.33(\pm 0.13) \quad (2)$$

Paper assumes that low regression implies other physics than sulfate are important (updraft velocity, temperature, aerosol composition).

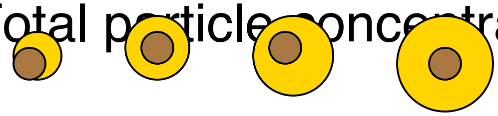
Not clear why other mechanisms aren't hypothesized: droplet size distribution, age of cloud, or particle/droplet interaction and evolution as described by Hoppel and Kohler ("value of greatest N"- limited water content)

A simplistic example:

- Each drop has its own "core": N α [$SO_4^{=}$] (assuming a monodisperse sulfate aerosol)
- Drops form without effect from sulfate, N independent of [$SO_4^{=}$]
- Combination of two or more effects? Nucleation, gas-phase oxidation, adsorption

Observations

- 6a: Fraction $SO_4^=$ increases with aerosol mass concentration ($SO_4^=$ adsorbs more readily than forms nuclei? Heterogeneous oxidation of SO_2?)
  - (also a moderating factor for fig. 4: *weaker relationship for overall pollutants?*)
- 6b: Total particle concentration increases with SO_4 concentration



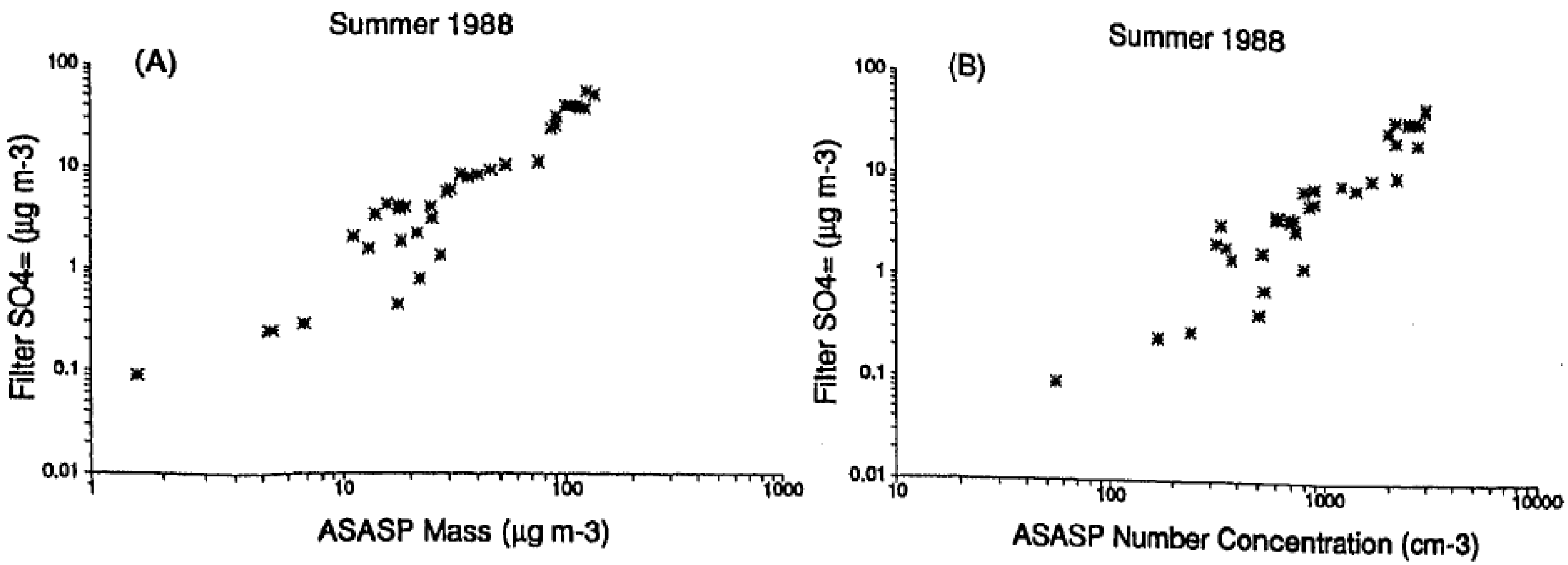


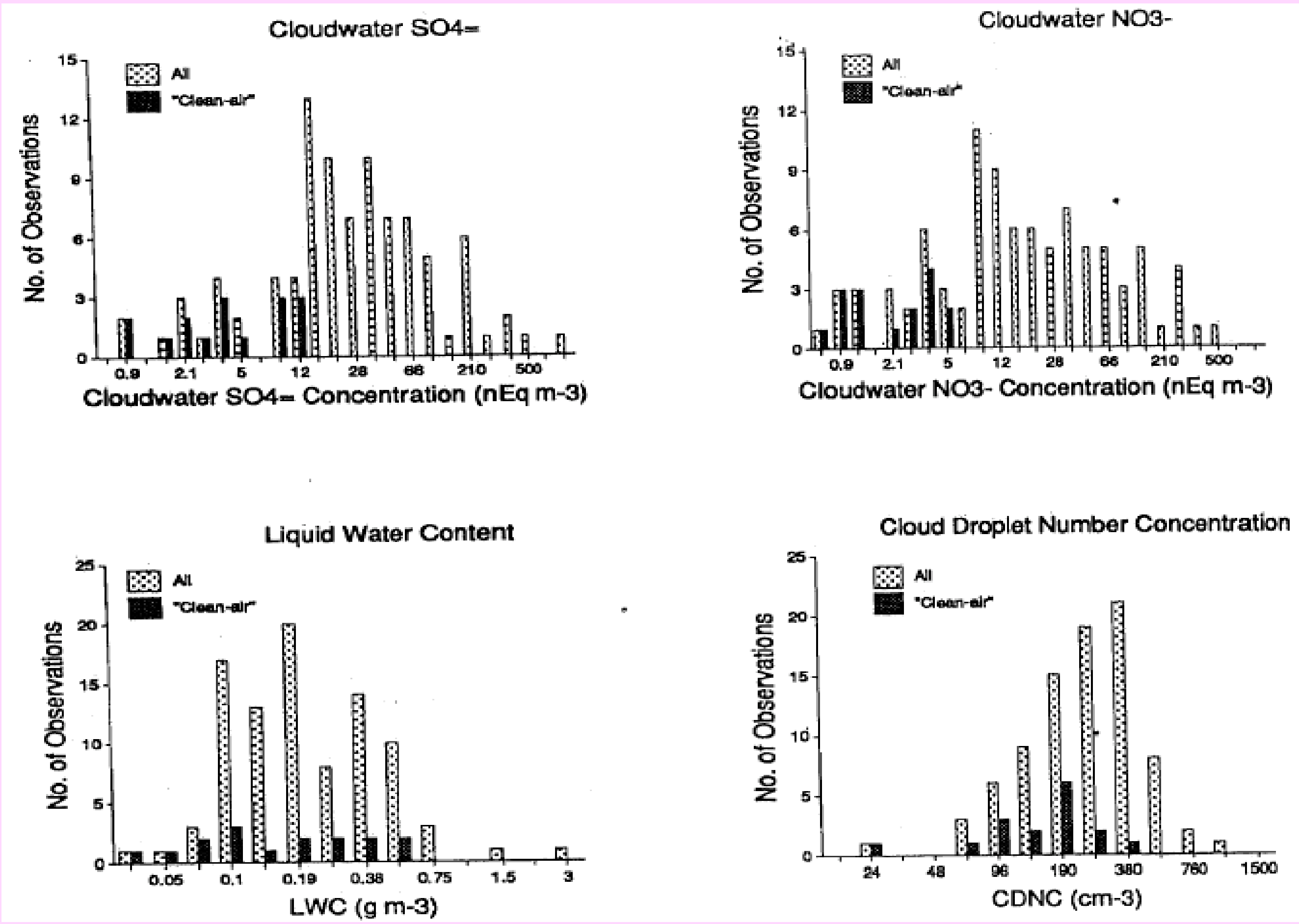
Fig. 6. (*a*) Airborne filter sulfate concentration plotted as a function of estimated aerosol mass in the 0.17–2 μm size range, assuming a mass density of 2 g cm^{-3}. (*b*) Airborne filter sulfate concentration plotted as a function of aerosol number concentration in the 0.17–2 μm size range.

Figure 6 cont'd…

- Conclude that similarity "indicates mean properties of the size distribution of the aerosol do not change dramatically with concentration".
- Range of aerosol number larger than for droplets, explained by reduced activation and collision efficiency of small drops. Contribution to total sulfate smaller for small drops *(aerosol relationship applies to droplet distribution?)*
- APOS: if this is particle not droplet, expect 10-60% error based on APOS in measurements between dry and aqueous phase – not well understood or relationship to $[cwSO_4^{=}]$

Cloud Albedo and Anthropogenic Emisions

- In attempt to get “background” non-anthro conditions, look at “clean air” range - <25% for both nitrate and sulphate.
- Variance of concentration distributions from lognormal/poission ascribed to meteorological variations.



Cloud Albedo and Anthropogenic Emisions

- Some justification of clean air classification by external measurements.
- Table 5, compare median (?) CDNC for clean air versus all, reject same at 99.5%: 290 and 170 respectively
- No note however of LWC, which also appears to significantly vary with [sulphate/nitrate] for clean air versus all for cumuliform: 0.47 and 0.27 g/m^3 and co-vary with CDNC…
- No note of strong temperature dependence on median/mean/geom mean

TABLE 5. Median and Mean Cloud Data for Stratiform Cloud, Cumuliform Cloud, and All Cloud

	No.*	Altitude km	LWC $g\ m^{-3}$	CDNC cm^{-3}	Temperature °C	nEq m^{-3} cw$SO_4^=$	nEq m^{-3} cwNO_3^-
Stratiform Cloud							
All median	(61)	1.5	0.16	210	1.8	22	12
All mean	(61)	1.6	0.20	220	1.4	38	28
All geometric mean	(61)	1.5	0.17	200	1.1	22	13
Clean-air median	(12)	1.9	0.16	120	5.4	3.9	2.8
Clean-air mean	(12)	2.0	0.20	140	3.4	5.3	2.7
Clean-air geometric mean	(12)	1.9	0.15	120	3.3	3.9	2.1
Cumuliform Cloud							
All median	(31)	2.1	0.32	400	7.6	35	34
All mean	(31)	2.3	0.47	450	6.4	89	67
All geometric mean	(31)	2.1	0.34	410	3.6	37	27
Clean-air median	(4)	1.9	0.24	240	5.5	6.3	2.2
Clean-air mean	(4)	2.0	0.27	260	3.7	6.8	2.2
Clean-air geometric mean	(4)	1.9	0.21	250	6.3	4.4	1.8
All Cloud							
All median	(92)	1.8	0.19	250	4.5	25	15
All mean	(92)	1.8	0.29	290	3.1	56	41
All geometric mean	(92)	1.8	0.21	250	2.8	26	16
Clean-air median	(16)	1.9	0.16	160	5.4	4.0	2.7
Clean-air mean	(16)	2.0	0.22	170	3.5	5.7	2.6
Clean-air geometric mean	(16)	1.9	0.16	140	3.4	4.0	2.0

*Indicates number of samples. For CDNC only, the number is 59 for stratiform cloud, 26 for cumuliform cloud, and 85 for all cloud.

Climatic Implications

- A_s~0.2, A_c~0.8
- Equilibrium, no feedback cooling: 2-3 W/m2

TABLE 6. Percentage Change in Planetary Albedo for an Increase in CDNC From 160 to 250 cm^{-3}

					Percent	
A_s	A_c	A_p	A'_c	A'_p	ΔA_p	$\Delta A_p/A_p$
1.0	0.9	1.0	0.913	1.0	0.0000	0
0.8	0.9	0.929	0.913	0.935	0.0067	0.72
0.6	0.9	0.913	0.913	0.923	0.0097	1.1
0.4	0.9	0.906	0.913	0.917	0.0112	1.2
0.2	0.9	0.902	0.913	0.914	0.0120	1.3
0.0	0.9	0.900	0.913	0.913	0.0126	1.4
1.0	0.8	1.0	0.823	1.0	0.0000	0
0.8	0.8	0.889	0.823	0.896	0.0074	0.83
0.6	0.8	0.846	0.823	0.860	0.0138	1.6
0.4	0.8	0.824	0.823	0.841	0.0179	2.2
0.2	0.8	0.810	0.823	0.830	0.0207	2.6
0.0	0.8	0.800	0.823	0.823	0.0227	2.8
1.0	0.6	1.0	0.635	1.0	0.0000	0
0.8	0.6	0.846	0.635	0.852	0.0055	0.65
0.6	0.6	0.750	0.635	0.764	0.0142	1.9
0.4	0.6	0.684	0.635	0.707	0.0223	3.3
0.2	0.6	0.636	0.635	0.665	0.0293	4.4
0.0	0.6	0.600	0.635	0.635	0.0351	5.9
1.0	0.4	1.0	0.436	1.0	0.0000	0
0.8	0.4	0.824	0.436	0.827	0.0033	0.40
0.6	0.4	0.684	0.436	0.695	0.0103	1.5
0.4	0.4	0.571	0.436	0.590	0.0188	3.3
0.2	0.4	0.478	0.436	0.506	0.0276	5.8
0.0	0.4	0.400	0.436	0.436	0.0362	9.0
1.0	0.2	1.0	0.225	1.0	0.0000	0
0.8	0.2	0.810	0.225	0.811	0.0014	0.18
0.6	0.2	0.636	0.225	0.642	0.0052	0.82
0.4	0.2	0.478	0.225	0.489	0.0107	2.2
0.2	0.2	0.333	0.225	0.351	0.0174	5.2
0.0	0.2	0.200	0.225	0.225	0.0249	12

$\Delta A_p/A_p$ is zero for all A_c = 0 and 1.

Open Questions

- *Role of ice in initiating precipitation and scavenging of liquid water (changing LWC-collected the same as droplets?) (Vong, 1993)?*
- *Temperature of cloud base vs. CCN population (chemistry, size, and concentration)*
- *Potential influences on LWC*
  - *Is it really only dependent on how efficiently water vapor gets advected upwards?*
- *how are "background" continental emissions different from background marine?*
- *Would these results change if we could measure interstitial and…R_{drop}<10_m sulphate?*
- *Bimodal Sulfate concentrations- cause?*
- *Would results change if used mean, geom mean instead of median?*
- *What's so great aboat Canadia, eh?*