SENSITIVITY TESTS OF DROPLET AND ICE CRYSTAL ACTIVATION WITH A PARCEL MODEL.

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Droplet activation

FUNCTION OF AT LEAST 12 PARAMETERS

Size distribution (diameter^1 (D), number concentration^2 (N_t) and standard deviation^3 (σ_g))

Thermodynamic state of environment (temperature^4 (T) and pressure^5 (P))

Cloud dynamics (updraft^6 (w))

Droplet growth kinetics (condensation coefficient^7 (α))

Aerosol chemical properties (solute molecular weight^8 (M_s), bulk solute density^9(ρ_s), surface tension^{10} ($\sigma_{s/a}$), number of dissociating species in solution^{11}(v) and practical osmotic coefficient^{12} (φ_s))

Sensitivity of droplet activation to uncertainties in activation parameters.

How accurate do we need to know the activation parameters to predict droplet concentrations?

Changes in number of activated droplets can lead to changes in cloud properties, such as droplet size distribution, radiative properties, precipitation, and cloud lifetime.

Relative sensitivity: $S(X_i) = \delta \ln N_d / \delta \ln X_i$

X_i represents the different activation parameters.

Relative sensitivity: How much does a change in X_i affect changes in number of predicted droplets, N_d?

Reduce number of parameters

Hygroscopisity parameter, κ
(Petters and Kreidenweis, 2007)

Hygroscopisity parameter describes the effective aerosol composition and include solute molecular weight, density, number of soluble ions, and osmotic coefficients.

$$\kappa=f(M_s,\rho_s,\nu,\Phi_s)$$

Now down to 9 variables:

κ, $\sigma_{s/a}$, D_g, N_t, σ_g, w, α, T and P

Reduce number of parameters

Diameter-surface tension-kappa relationship:

Number of CCN activation is dependent on the supersaturation (s). All particles having a critical supersaturation $s_c < s$ will activate.

$$s_c \approx \sqrt{\frac{4A^3}{27\kappa D_d^3}} \qquad A \approx \frac{4\sigma_{s/a} M_w}{RT\rho_w}$$

Petters and Kreidenweis (2007)

Activation of CCN: $\underbrace{f(s_c)}=f(\sigma_{s/a},\kappa,D_d)=f(\Psi)$

A new physicochemical activation parameter

$$\boxed{\Psi = \frac{\sigma_{s/a}}{\kappa^{1/3} D_g}}$$

Now down to 7 variables, Ψ, N_t, σ_g, w and α, T, and P

Sensitivity studies with a detailed microphysical Lagrangian parcel model

T =10°C and P = 800 hPa

$50 \leq N_t \leq 50{,}000$ cm^{-3}	n = 10	(Total aerosol concentration)
$1.25 \leq \sigma_g \leq 2$	n = 4	(Standard deviation of aerosol distribution)
$0.1 \leq w \leq 10$ m/s	n = 10	(Updraft velocity)
$0.04 \leq \alpha \leq 1$	n = 2	(Condensation accommodation coefficient)
$91 \leq \Psi \leq 58{,}000$ x10 J m^{-3}	n = 10	(Physicochemical activation parameter)
	($0.2<\kappa<1.3$, $0.024<D_g<0.5$ μm $0.05<\sigma_{s/a}<0.08$ Jm^{-2})	

For each input combination we also had inputs where the variables were ± 10% of original value

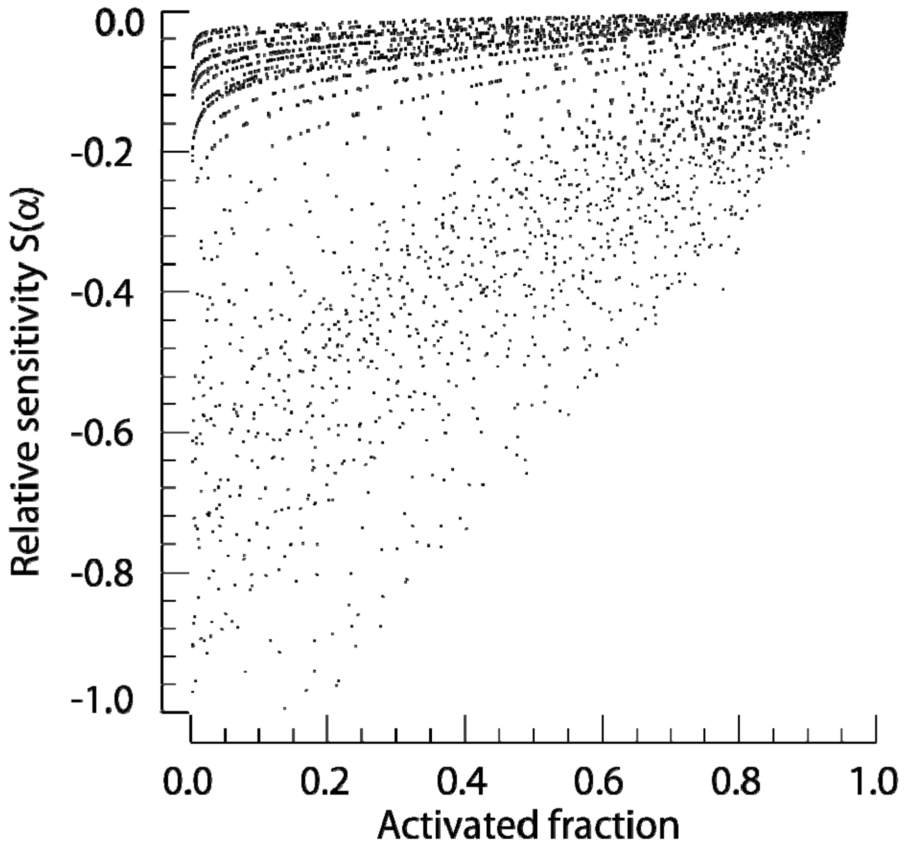
→ 80,000 simulations

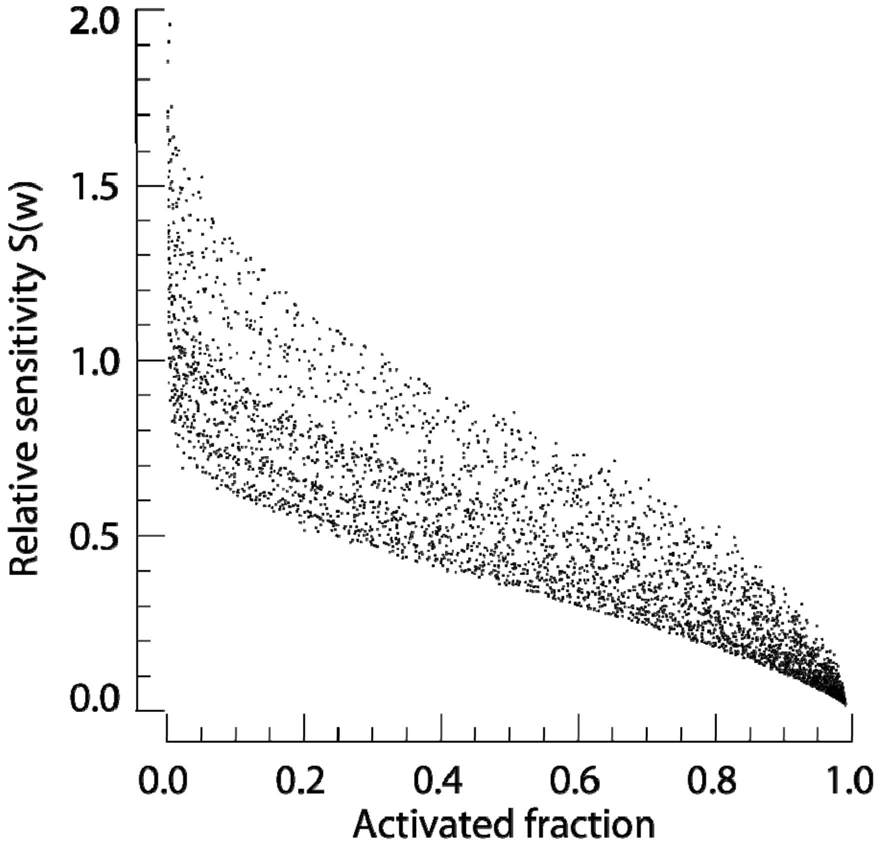
Sensitivity to Kinetics (α) and Updraft (w)

$$S(X_i) = \frac{\delta \ln N_d}{\delta \ln X_i}$$

Here, $\delta \ln X_i$ = 10 %.

Relative sensitivity, $S(X_i)$ indicates how sensitive the droplet number concentration is to a 10 % change in X_i

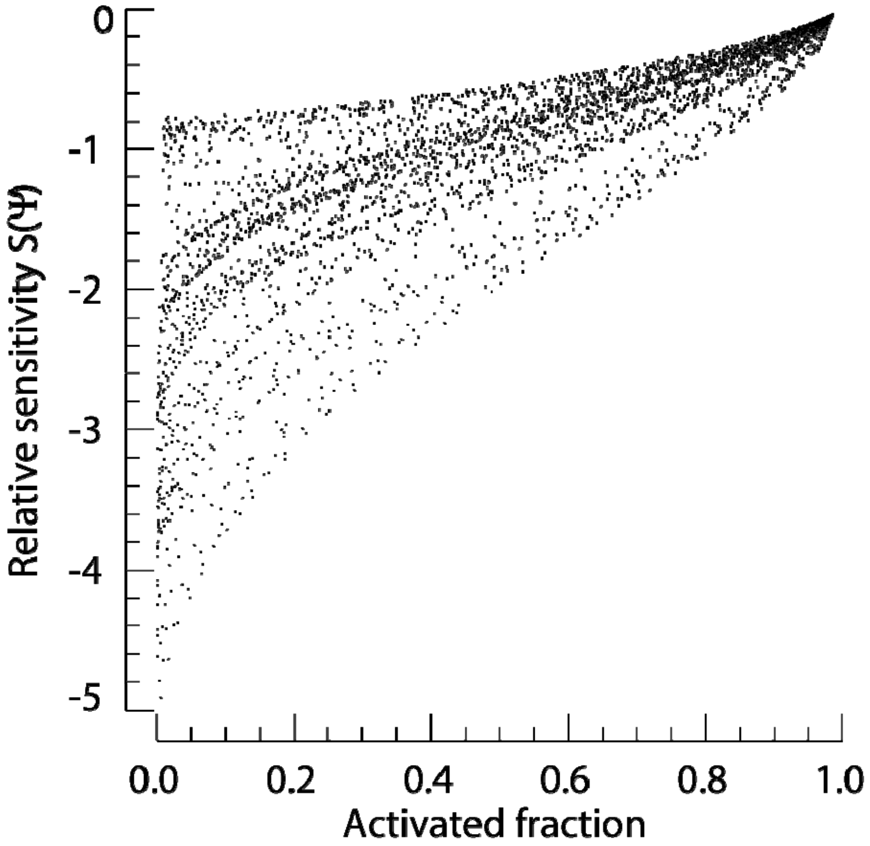


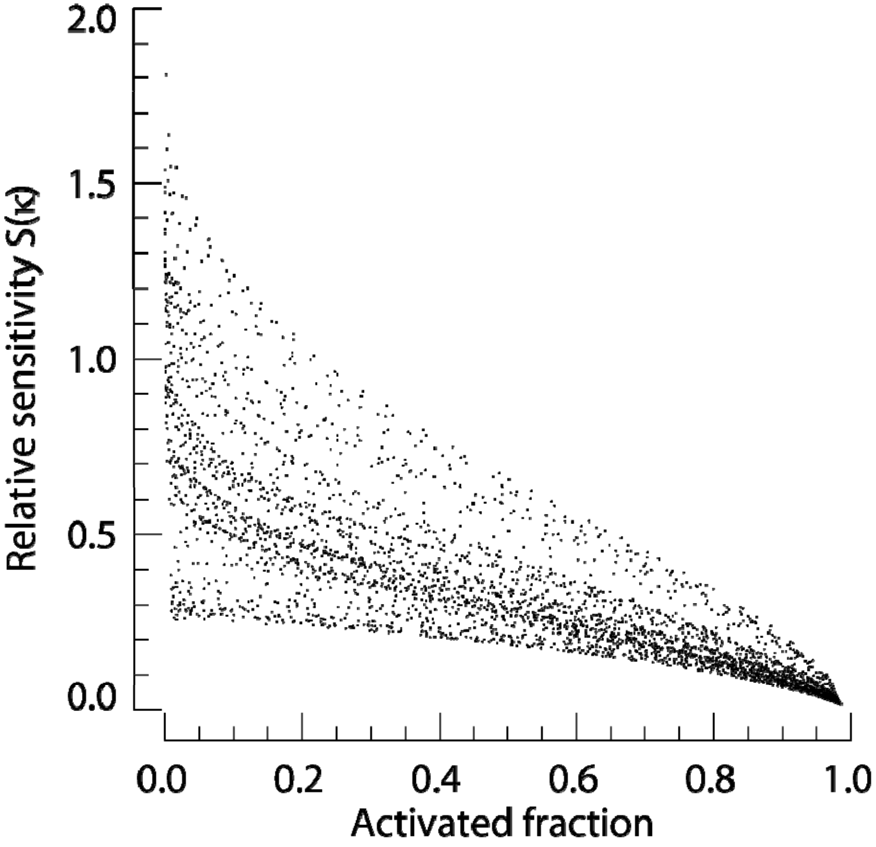


Sensitivity to Physicochemical parameter (Ψ)

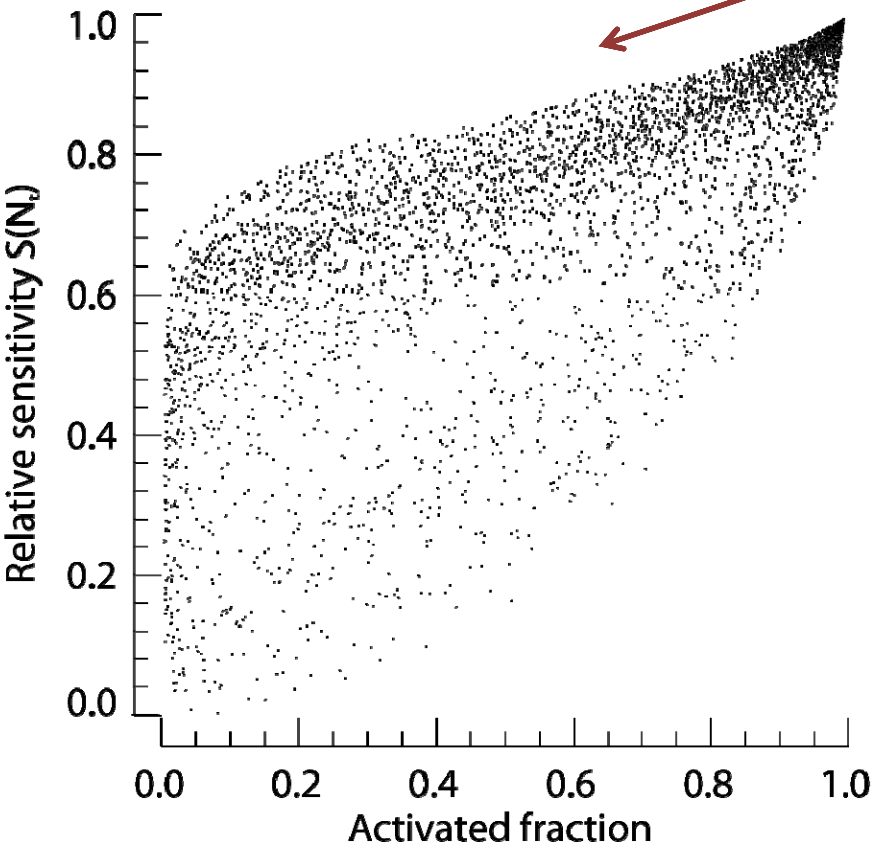
$$S(\kappa)=\frac{\partial \ln N_d}{\partial \ln \Psi}\frac{\partial \ln \Psi}{\partial \ln \kappa}=-\frac{1}{3}S(\Psi)$$

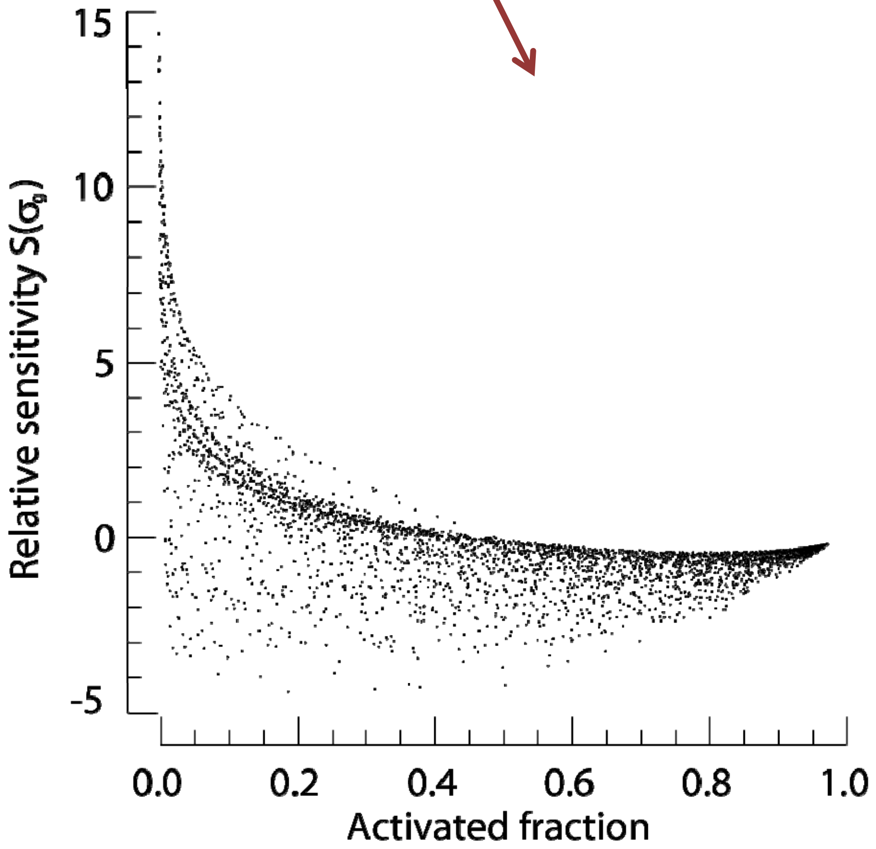
$$-S(D_g)=S(\sigma_{s/a})=S(\Psi)$$





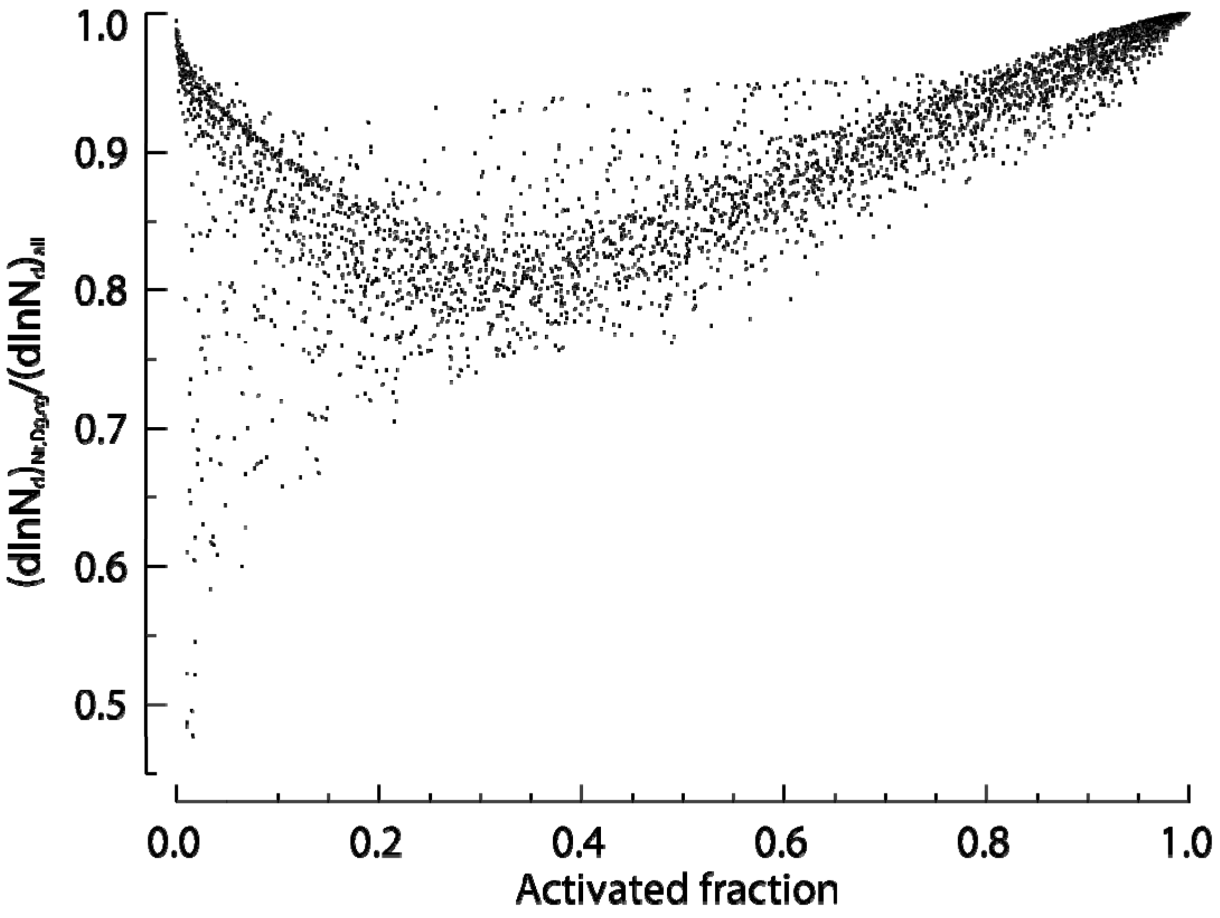
Sensitivity to Size distribution (N_t, σ_g)





Relative sensitivity of size distribution to all activation parameters

Total relative uncertainty = contribution from size distribution, updraft, kinetics and chemical parameters



Summary droplet activation

Uncertainties in size distribution has a larger effect than chemical, kinetics and updraft, **assuming 10 % uncertainties**

$S(D) = S(\sigma_{s/a})$: 0 to -3.5

$S(N_t)$: 1 to 0

$S(\sigma_g)$: -4 to 4

$S(\kappa)$: 0 to 1.2

$S(\alpha)$: 0 to -1

$S(w)$: 0 to 1.3

ICE-L
(Ice in Clouds Experiment – Layer clouds)

Goal was to conduct measurements in clouds where only primary heterogeneous nucleation occur, or can be separated (in time and space) from secondary processes.

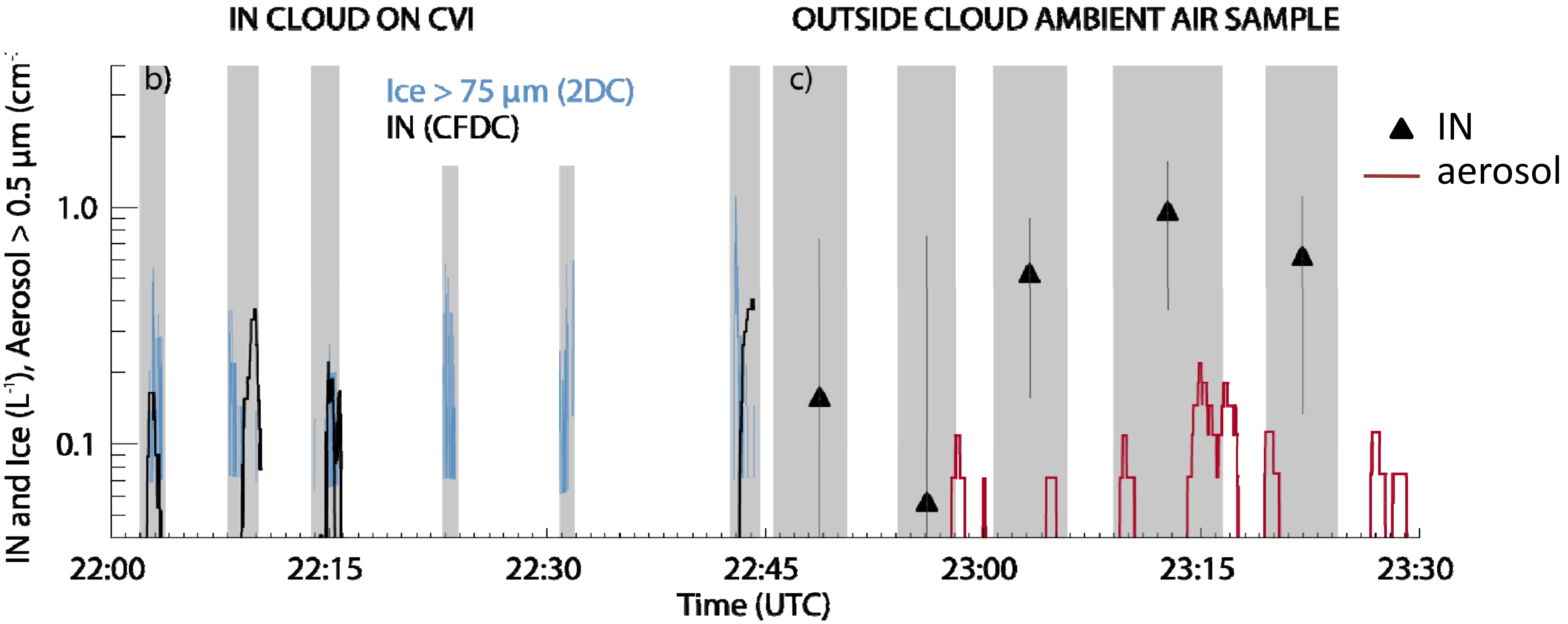
Mainly layer clouds, especially lenticular wave clouds.

In an idealized setting, air parcels will spend only a few hundred seconds in a layer cloud and mixing and precipitation is not expected.

Wave clouds are ideal for parcel model studies.

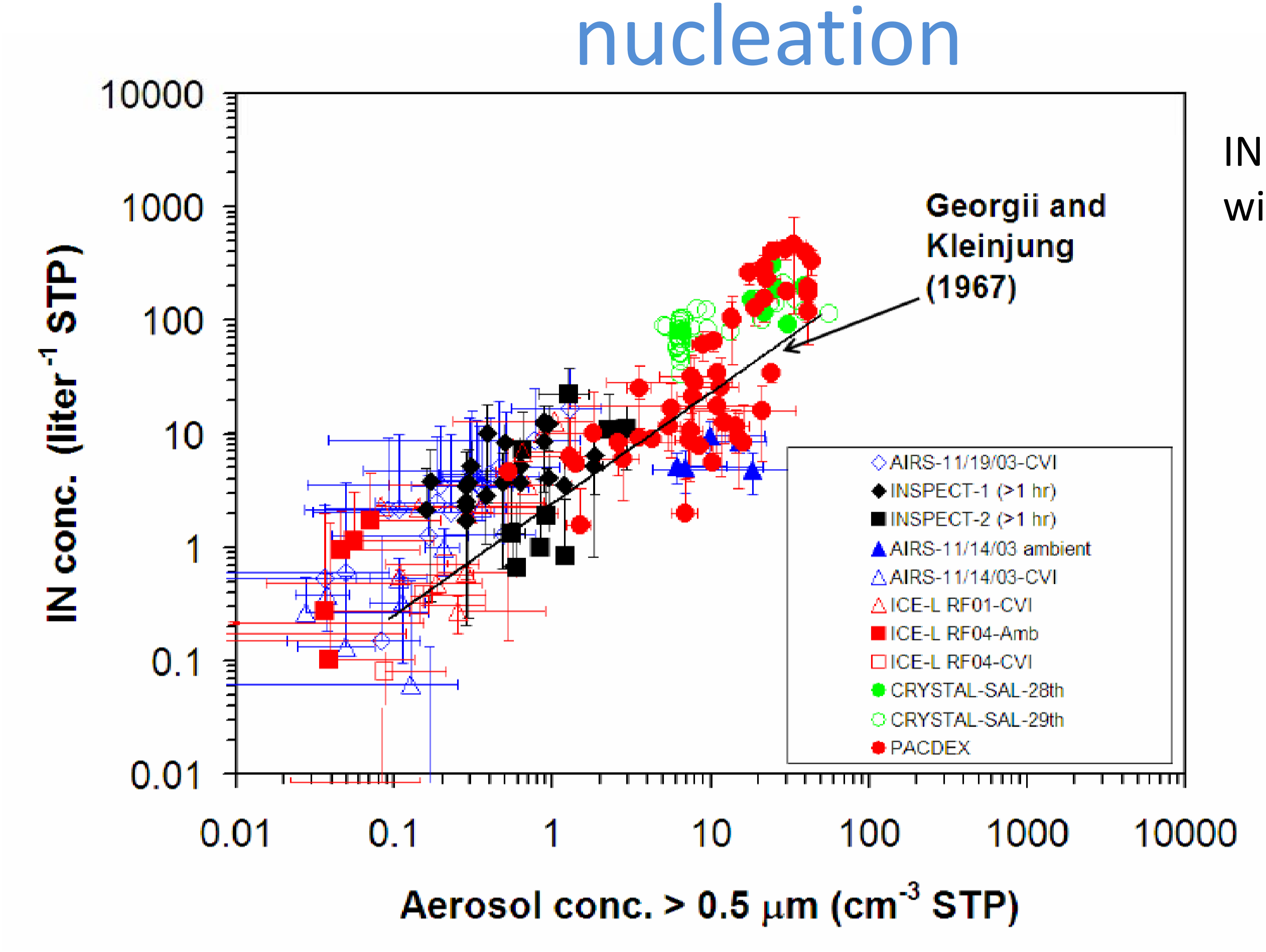
Ice crystal/ IN measurements in wave cloud

-30 < T < -25 °C
w ± 3 m/s



CFDC: Continuous flow diffusion chamber, measure IN in deposition, condensation and immersion mode
2DC: Cloud particles

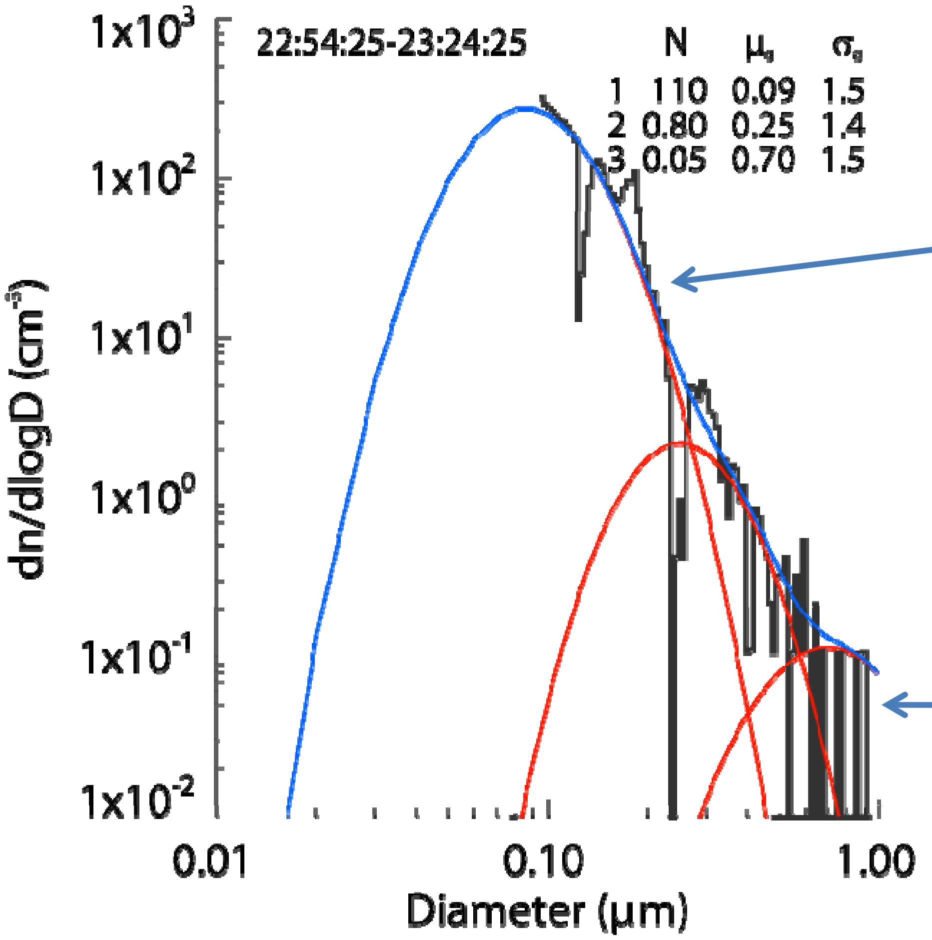
Aerosols and heterogeneous ice nucleation



IN measurements with the CSU CFDC

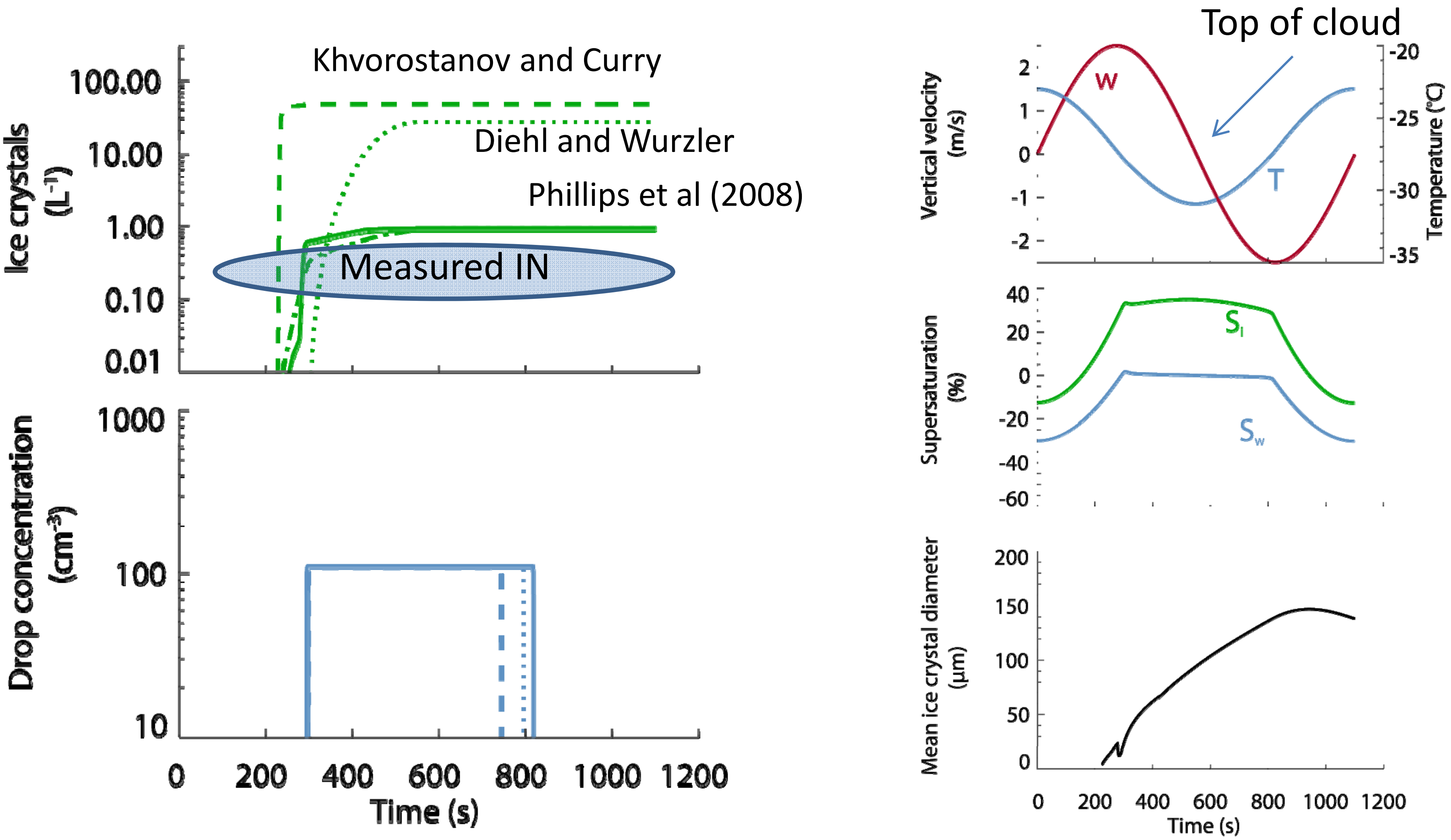
DeMott

Measured aerosols size distribution downwind of wave cloud with UHSAS



Ammonium sulfate

Internally mixed dust (90 %) and ammonium sulfate (10%)



Diehl and Wurzler, 2004

Immersion freezing, droplet volume based

Heterogeneous nucleation parameterization by Phillips, DeMott and Andronache, 2008, JAS

Based on IN measurements from INSPECT

Khvorostyanov and Curry, 2004

From classical heterogeneous nucleation theory

Ice crystal concentrations are scaled based on an aerosol surface area reference value.

Summary

Droplet activation

Uncertainties in size distribution determine sensitivities at high activation fractions and also dominate at lower activation fractions

Ice nucleation

There is a strong link between the concentration of insoluble particles and IN, and this should be accounted for in ice heterogeneous nucleation parameterizations.

The one parameterization that have a constraint on predicted ice crystal concentration relative to aerosol loading is doing the best in comparison with measurements.