GMI simulations of:
Cirrus ice crystal number
Dust impacts on droplet number

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Part 1:
Sensitivity of the global distribution of cirrus ice crystal concentration to heterogeneous freezing
Multiple mechanisms for ice formation can be active.

- **Homogeneous Freezing**
  - Mainly depends on $RH_i$ and $T$

- **Heterogeneous Freezing**
  - (Immersion, deposition, contact, …)
  - Also depends on the material and surface area

Wet aerosol particles + Insoluble Material ("Ice Nuclei")

http://www.alanbauer.com
Conceptual Model of Ice Formation in Cirrus

- **Height (km):**
  - 10
  - 9.5
  - 9
  - 8.5
  - 8

- **Homogeneous freezing of droplets**
  - Crystal growth, fresh IN continue to freeze and deplete vapor

- **Heterogeneous IN freezing**
  - Begin forming ice

- **Expansion cooling and ice supersaturation development**

- **Soluble and insoluble aerosol initial distribution**

- **Liquid droplets + Insoluble material**

- **Ice Crystals**

- **RH$_i$ (%)**
  - 100
  - 110
  - 120
  - 130
  - 140
  - 150
  - 160
The Effect of IN on Ice Crystal Number Concentration

- Homogeneous
- Homogeneous and Heterogeneous
- Heterogeneous

“Limiting” IN concentration

Barahona and Nenes, ACP, 2009a.
## Cirrus Formation in Global Climate Models: Current State of the Art

<table>
<thead>
<tr>
<th></th>
<th>Advantages</th>
<th>Disadvantages</th>
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<tbody>
<tr>
<td><strong>Empirical</strong></td>
<td>Very fast</td>
<td>Limited Coverage</td>
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<tr>
<td></td>
<td>Give reasonable values</td>
<td>Cannot be used to assess aerosol effects</td>
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<tr>
<td>Off-line solutions</td>
<td>Fast</td>
<td>Can’t consider all the variability in formation conditions</td>
</tr>
<tr>
<td>(e.g., Liu and Penner, 2005)</td>
<td>Physically-based</td>
<td></td>
</tr>
<tr>
<td><strong>Analytical-Numerical</strong> (e.g., Kärcher, et al., 2006)</td>
<td>Most of the physics included</td>
<td>Not fast enough</td>
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</table>

**Analytical models based on cloud formation physics are needed!**
Parameterizing ice formation: Solve the Mass and Energy Balances for cirrus

\[
\frac{dS_i}{dt} = M_a p \left[ (1 + S_i) \left( gM_a V - \Delta H_s M_w \frac{dT}{dt} \right) \right] + (1 + S_i) \left[ gM_a V - \Delta H_s M_w \frac{dT}{dt} \right]
\]

Condensation-type equations with dependencies on:
- Temperature, Updraft Velocity
- Aerosol composition, size, and freezing properties
- Crystal growth rate

\[
\frac{dw_i}{dt} = \rho_i \left( \frac{\partial n_c(D_c)}{\partial \ln(D_c)} \right)
\]

\[
\frac{dD_c}{dt} = \left( S_i - S_{i,eq} \right) \Gamma_1 D_c + \Gamma_2
\]

Ice Crystal Growth

IC: Initial droplet size distribution, relative humidity, and temperature.

Barahona and Nenes, JGR, 2008; ACP, 2009
Final expression: A Unified Description of Particle Formation and Growth

The solution of the PDE system reduces to:

\[
\frac{N_{het}(s_{\text{max}})}{N^* \sqrt{\Delta s_{\text{char}}}} = \frac{1}{\sqrt{\Delta s_{\text{char}}}} \left(1 + s_{\text{max}}\right) e^{\lambda s_{\text{max}}}^{2}
\]

- Simple and physically based. Completely theoretical and analytical (i.e., robust). Very fast!
- Accounts for homogeneous and heterogeneous freezing
- Works with a general definition of heterogeneous freezing:
  - Can take into account the contribution from several freezing modes and aerosol species.
  - Allows direct incorporation of theoretical and empirical data into large scale models.

Barahona and Nenes, ACP, 2009b.
Parameterization evaluation: Compare Against Numerical Solution

Evaluation over a broad range of cloud formation conditions, aerosol concentrations, and freezing parameters.

Average error: $5 \pm 12 \%$.

Orders of magnitude faster than the numerical solution

Including the ice formation parameterization in GMI


- Implementation:
  - Wind fields derived from GISS II’ GCM
  - Dust and black carbon as IN precursors
  - Cirrus allowed for T<235 K. Time step 1h, resolution 4°×°5

- Dynamical forcing: Integrate over a Gaussian distribution of updraft velocities

\[ \sigma_u = 25 \text{ cm s}^{-1} \]

Gayet et al. (2006)
Description of Aerosol Freezing

- Homogeneous (sulfate): Koop et al. (2000)

- Heterogeneous (dust and black carbon): Nucleation Spectrum:

  \[ N_{IN} (\text{Supersaturation}) \mid T, N_{aerosol} \]

  Dust and Black carbon freezing fraction as a function of supersaturation

Aerosol size, chemical composition, surface properties

There is still uncertainty in the form of \( N_{IN} (S_i) \); we tested several definitions currently used in cloud studies.
IN Spectra used in this Study

<table>
<thead>
<tr>
<th>IN Spectra</th>
<th>Description</th>
<th>Type</th>
</tr>
</thead>
<tbody>
<tr>
<td>All IN</td>
<td>All dust and black carbon freeze at Si = 130% Theoretical</td>
<td>CNT-BN</td>
</tr>
<tr>
<td></td>
<td>Freezing fraction varies as predicted by CNT. Barahona and Nenes [2008].</td>
<td>Semi-Empirical</td>
</tr>
<tr>
<td>BKG</td>
<td>No explicit dependency in aerosol concentration, Phillips et al. [2007]</td>
<td>Empirical</td>
</tr>
<tr>
<td>PDA</td>
<td>Freezing fraction is scaled using total aerosol surface area, Phillips et al. [2008]</td>
<td>Empirical</td>
</tr>
</tbody>
</table>

Ice Nuclei Number Concentration (cm⁻³) vs. Ice Supersaturation
Heterogeneous IN Concentrations

\[ P = 281 \text{ hPa} \]

About two orders of magnitude difference in IN concentration

Barahona, Rodriguez, and Nenes, JGR, in press.
Comparison against Homogeneous Freezing

At least a factor of 10 variation in global mean ice crystal concentration. Most significant in Northern Hemisphere

Barahona, Rodriguez, and Nenes, JGR, in press.
A simple, analytical and fast parameterization of cirrus formation is presented which accurately reproduces numerical results.

Considers the dependency of the ice crystal number on cloud dynamics, supercooled liquid aerosol size and number, deposition coefficient, and ice nuclei concentration.

Accounts for the competition between homogeneous and heterogeneous nucleation and between different nucleation modes in pure heterogeneous freezing.

Implemented in GMI and used to test the sensitivity of ice number to IN parameterization spectra available in literature.

2 orders of magnitude IN variability $\rightarrow$ 10-fold change in crystal number. This sensitivity is preserved across met fields.

BC does not really contribute to IN; predominant ice production mechanism however depends on the IN parameterization. Potentially important for predicted IWP.
Part 2: Effect of insoluble (fresh) dust particles on global CCN and droplet number
Mineral Dust as CCN

- Dust contributes to ~ 10% global CCN ‘burden’ (regionally larger).
- Dust acts as giant CCN, and its wet size controls its efficiency as a collector drop.
- Limited number of studies include dust-cloud interactions
- CCN activity of dust (especially freshly-emitted) is not well understood.

Hoose and Lohmann (2008)
Adsorption Activation:
A new theory of droplet formation

Current CCN theory makes very important assumptions about insoluble particles (dust).

- Insoluble substances (like dust) are not good CCN.
- The CCN activity of solubles depends *solely* on soluble “coatings” from atmospheric processing.
- Theory works well when there is a fair amount of soluble solute in the particles (~ >10% by mass).

Does this mean fresh dust (no solute) are not CCN?

- Dust particles are wettable, and can swell. This means they do interact with water, but only on their surface.
- Look at the process of *adsorption* of water on dust, and its potential impact on droplet formation.
Example: adsorption/desorption curves of Illite

Adsorbed water increases with RH

Semi-reversible (hysteresis)

Substantial amounts of water can adsorb at equilibrium

Same for other clays.

Schuttlefield et al., JGR, (2007)
Describing water adsorption on Dust

Use the Frenkel-Halsey-Hill (FHH) multilayer adsorption isotherm can be used to describe the interaction of water vapor with the dust surface.

“Bulk” Water Phase

“Adsorbed” Phase

- Long-range Van-der-Walls interactions between surface and upper layers are considered by $A_{FHH}$ & $B_{FHH}$.
- $A_{FHH}$ accounts for interactions between the first monolayer and substrate.
- $B_{FHH}$ quantitatively accounts for the adsorption strength of each additional H$_2$O monolayer.
Can water adsorption turn particles into CCN?

Plot the equilibrium vapor pressure of the dust particle as more and more water adsorbs on it...

The combined curvature and adsorption effects determine the wet diameter at equilibrium.

Looks like a “classical” CCN.

Here is why...
When does an aerosol particle act as a CCN?

Dynamical behavior of a dust particle in a variable RH environment.

When ambient saturation ratio exceeds $S_c$, particles act as CCN.

$CCN = f\ (\text{Size, Adsorption parameters})$
CCN activity of dry-generated dust (in the lab)

Collect soil samples from active dust source emission regions

North American Soils
East Asian Soils
African Soil Niger Dust

Dryland Systems
- Hyper-arid
- Arid
- Semiarid
- Dry subhumid

Source: Millennium Ecosystem Assessment

Kumar, P., et al, ACP, submitted
CCN Activity of Regional Dusts

CCN activity is region dependent (Asian>Niger>ATD)

... but not terribly variable

- Fine mode dust is CCN active – equivalent to having up to 10% vol. fraction of ammonium sulfate (@ 100 nm). Smaller particles even higher.

- “Average” dust: $A_{FHH} \sim 2.50 \pm 0.50 \quad B_{FHH} \sim 1.20 \pm 0.10$
**Including Adsorption Activation in droplet formation parameterizations**

**Basic Idea:** Solve conservation laws for energy and mass balances for water vapor condensing on aerosol particles in the ascending parcel.

The final result is the number of activated droplets $N_d$ that is calculated at the parcel maximum supersaturation, $s_{max}$.

This is a two step process:

1. Obtain parcel $s_{max}$
2. Determine droplet number, $N_d$ by counting particles with $s_c < s_{max}$

Kumar, P., Sokolik, I. N. and Nenes, A., *ACP*, 2009
Including Adsorption Activation in droplet formation parameterizations

**Supersaturation Balance Equation: solved for \( s_{\text{max}} \)**

\[
\alpha V + e V \left[ \frac{\Delta H_T M_w}{R T^2} \left( T - T' \right) - (1 - RH) \right] - I_e (0, s_{\text{max}}) = 0
\]

\[I_e (0, s_{\text{max}})\] is the ‘rate’ of condensation of water vapor on the activated CCN and contains contributions from soluble and insoluble particles.

\[I_e (0, s_{\text{max}}) = I_{K,1} (0, s_{\text{part}}) + I_{K,2} (s_{\text{part}}, s_{\text{max}}) + I_{FHH} (0, s_{\text{max}})\]

**Kumar, P., Sokolik, I. N. and Nenes, A., ACP, 2009**
Including droplet formation from adsorption activation in GMI

**Emissions**
- University of Michigan current day (Liu et al., 2007)

**Aerosol-CDNC link**
- Sulfate, OC, Seasalt (soluble), Dust (insoluble) using experimentally-defined activation properties

Globally: Dust not as important
Regionally: can dominate CDNC.
Dust CCN take home points

- Application of KT and FHH-AT leads to very different predictions in CCN and cloud droplet number for the same aerosol size distribution. FHH-AT seems to be a better description of CCN activity.

- **CCN measurements** for dust generated from soils give on average:

  \[ A_{FHH} \sim 2.50 \pm 0.50 \quad B_{FHH} \sim 1.20 \pm 0.10 \]

- Fresh dust \( \sim 100\text{nm} \) acts like a particle with \( \kappa \sim 0.025-0.05 \). Strong size-dependence of the “k” though. This is not from solute, but from the surface-water interactions alone.

- The new theory can directly be introduced in droplet formation parameterizations, especially since the parameters are to first order constant.

- Initial simulations with the GMI suggest that dust near their sources can have an important contribution to droplet number.
THANK YOU!!

Photo: A. Sorooshian