Ice Nucleation Properties of Coated and Uncoated mineral dust particles

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Motivation

- Mineral dust particles are abundant in the atmosphere (up to 1500 Tg/year) and effective IN. e.g., DeMott et al., 2003; Kulkarni et al., 2012; Hoose and Möhler, 2012

- Atmospheric dust particles frequently undergo aging by trace materials (sulfuric acid, nitric acid, ammonium sulfate, ammonia, ozone, organics). e.g., Russell et al., 2002; Sullivan et al., 2007

**Missing:** Effect of coating on different atmospherically relevant mineral dust particles on ice nucleation efficiency. Important from cloud modeling perspective as they now started treating effect of dust speciation on ice nucleation. e.g., Liu et al., 2013

\[
\text{Dust} + \text{H}_2\text{SO}_4 \text{ vapor} \rightarrow \text{IN properties??？}
\]

\[
\text{Dust} \rightarrow \text{IN properties??？}
\]
### Past studies

<table>
<thead>
<tr>
<th>Reference</th>
<th>IN investigation technique</th>
<th>Mode of ice nucleation or freezing</th>
<th>Temperature (°C)</th>
<th>Mineral dust Type</th>
<th>Diameter (μm)</th>
<th>Concerns</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cziczo et al. (2009)</td>
<td>Expansion type cloud chamber</td>
<td>Deposition, immersion, condensation</td>
<td>-20 to -45</td>
<td>Illite</td>
<td>&gt; 1</td>
<td></td>
</tr>
<tr>
<td>Neidermeier et al. (2010)</td>
<td>Continuous flow cooling chamber</td>
<td>Immersion</td>
<td>-34 to -40</td>
<td></td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td>Neidermeier et al. (2011)</td>
<td>Continuous flow cooling chamber</td>
<td>Immersion</td>
<td>-28 to -40</td>
<td></td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td>Wex et al. (2013)</td>
<td>Continuous flow cooling chamber</td>
<td>Immersion</td>
<td>-26, -30 and -34</td>
<td>Kaolinite</td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td>Sullivan et al. (2010a)</td>
<td>Continuous flow diffusion chamber</td>
<td>Deposition, immersion/condensation</td>
<td>-25 and -30</td>
<td></td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td>Tobo et al. (2012)</td>
<td>Continuous flow diffusion chamber</td>
<td>Deposition, immersion/condensation</td>
<td>-26, -30 and -34</td>
<td>Kaolinite</td>
<td>0.3 and 0.7</td>
<td>Yes</td>
</tr>
<tr>
<td>Knopf and Koop (2006)</td>
<td>Optical microscope with a flow cell</td>
<td>Deposition</td>
<td>-13 to -76</td>
<td></td>
<td>0.7 to 10</td>
<td>Yes</td>
</tr>
<tr>
<td>Eastwood et al. (2009)</td>
<td>Optical microscope with a flow cell</td>
<td>Deposition</td>
<td>-27 to -40</td>
<td></td>
<td>5 to 15</td>
<td>Yes</td>
</tr>
<tr>
<td>Chernoff and Bertram (2010)</td>
<td>Optical microscope with a flow cell</td>
<td>Deposition</td>
<td>-26 to -39</td>
<td>Illite, Quartz, Montmorillonite, Feldspar</td>
<td>~7 to 10</td>
<td>Yes</td>
</tr>
<tr>
<td>Present study</td>
<td>Continuous flow diffusion chamber</td>
<td>Deposition</td>
<td>-25, -30 and -35</td>
<td></td>
<td>0.2</td>
<td>Yes</td>
</tr>
</tbody>
</table>

**Concerns:** Use of ATD and Wet generation method. Murray et al., 2012; Hoose and Möhler, 2012; Herich et al., 2009; Sullivan et al., 2010;
Coated particles showed lower IN activation efficiency. Similar results for all other four dust species at three different temperatures: -25, -30 and -35.

Note: For very low gas concentration efficiency was similar. XRD showed similar patterns.
X-Ray Diffraction characterization patterns

Reveals why coated particles were poor IN

Acid treated substrate (interface) distort the atomic positions, decreasing the structural order. Active sites may have been destroyed (Sullivan 2010; Reitz 2011).

Large crystallographic differences or dislocations at the ice-substrate interface will raise the interface free energy, reducing the ability of the dust particle to induce ice formation (Pruppacher and Klett 1997).

Atoms are in a near-perfect periodic arrangement.

No periodic arrangement of atoms. Lacks the long-range crystalline order.

Acid treated substrate (interface) distort the atomic positions, decreasing the structural order. Active sites may have been destroyed (Sullivan 2010; Reitz 2011).

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Modeling Implications

Active site density parameterizations

Model sensitivity in simulated IWC and IC concentration using uncoated and coated dust particles.

Single-column version of the CAM5 (Neale et al., 2010; Liu et al., 2011) was used. Ice microphysical processes, including ice nucleation, vapor deposition, sublimation, melting, sedimentation, auto-conversion, and accretion, were considered in the simulations.

Optical cloud thickness increases as ice water path increases.
Ongoing work: Diesel soot characterization

Role of OC/BC ratio towards IN activity ???

Soot generation & dilution

- Diesel soot generator
- Load bank
- Pure Air supply
- Vacuum generator
- Filter system

Soot ageing, mixing & coagulation

- Excess diluted exhaust flow
- DMA

Soot measurement instrumentation

- HR-AMS (PNNL)
- SPLAT II (PNNL)
- CO₂, NO, O₃ (PNNL)
- PTRMS (PNNL)
- CO, CO₂, CH₄ (LANL)
- PSAP, Nephelometer (PNNL)
- PASS-3 & UV (LANL)
- CAPS (LANL)
- Photo. Interferometer (BNL)
- PAX – 405 & 532 (CMU)
- PAX – 870 (LANL)
- SEM/TEM (MTU)
- SP2 (LANL, BNL)
- SMPS, CPC, APM (PNNL)
- CCN, IN (PNNL)

Soot properties

- Chemical composition, size, shape, density, fractal dimension
- Gases, volatile organic compounds
- Absorption, scattering, extinction
- Mixing state, elemental composition, imaging
- BC mass & number, mixing state, coating
- Size, number concentration
- Hygroscopicity, Ice nucleation
Summary

- **IN nucleation efficiency** of uncoated particles **was higher** than coated particles. Efficiency also depends upon the gas concentration.

- Coated particles were amorphous in nature and showed large degree of structural disorderliness. **Crystalline character or ordered structure** of a dust particle is important towards nucleating ice.

- Modeling studies showed ice cloud microphysical properties **were sensitive** to the coated particles.

**Needed:** More measurements that cover wide range of dust minerals and gas concentration. Laboratory studies provide key insights, however, to fully understand the influence of chemical mixing state of dust particles or other particles on ice nucleation, **continuous IN measurements at various DOE-ARM sites** should be performed. { a topic for new measurements needs in the CAPI session} Long-term measurements would help to constrain/evaluate the cloud modeling output and remote-sensing retrieval algorithms, particularly for mixed-phase clouds.
Thank You
The simplest repeating unit in a crystal is called a **unit cell**. Each unit cell is defined in terms of **lattice points** the points in space about which the particles are free to vibrate in a crystal.
If you remove crystalline structure or some unit cells, the peak intensity drops as strength of reflected (diffracted) X ray reduces.