Ice Nucleation Properties of Coated and Uncoated mineral dust particles

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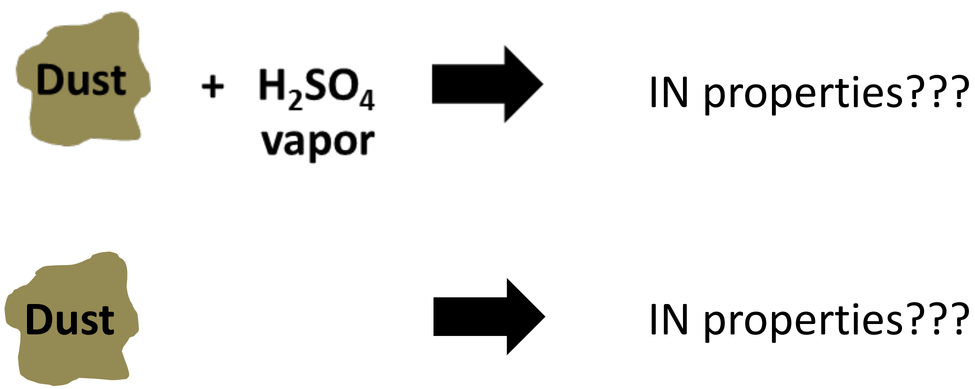
Motivation

- 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

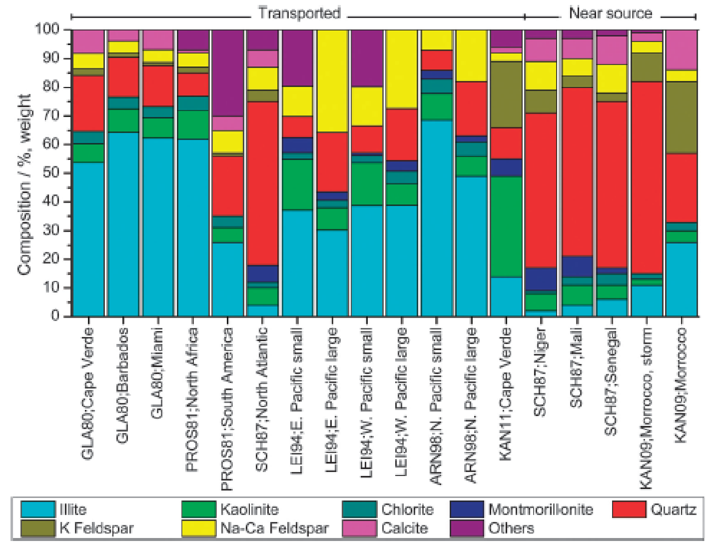
- Atmospheric dust particles frequently undergo aging by trace materials (sulfuric acid, nitric acid, ammonium sulfate, ammonia, ozone, organics). *e.g., Russell et al., 2002; Sullivan et al., 2007*

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*



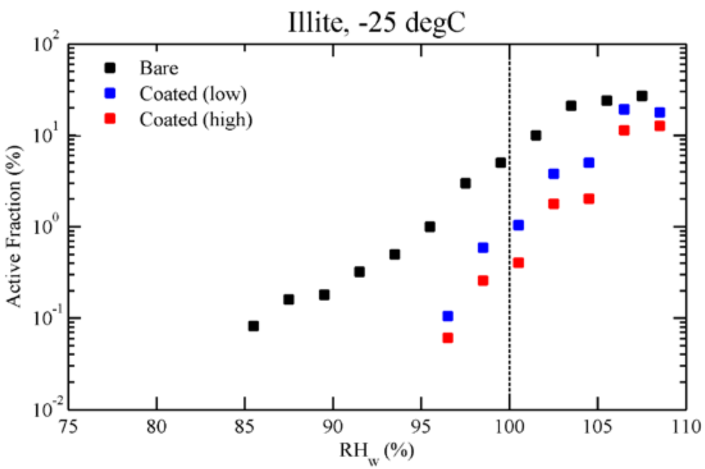
Past studies

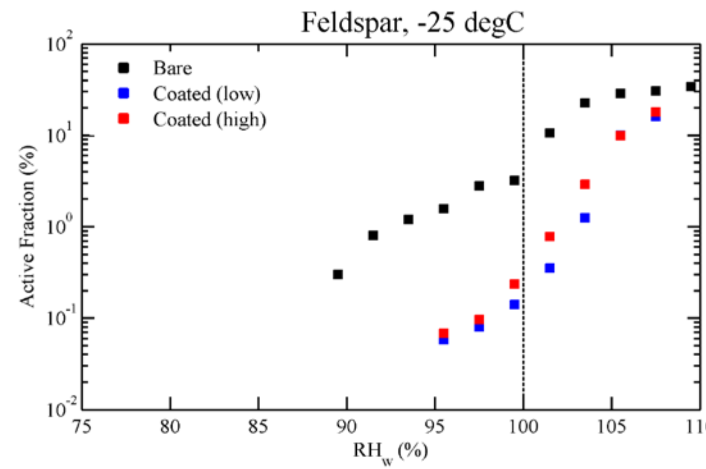
Reference	*IN investigation technique*	*Mode of ice nucleation or freezing*	*Temperat-ure (ºC)*	*Mineral dust*				
				Type	*Diam (μr*			
Cziczo et al. (2009)	Expansion type cloud chamber	Deposition, immersion, condensation	-20 to -45	Illite	< ~			
Neidermeier et al. (2010)	Continuous flow cooling chamber	Immersion	-34 to -40		0.			
Neidermeier et al. (2011)	Continuous flow cooling chamber	Immersion	-28 to -40		0.			
Wex et al. (2013)	Continuous flow cooling chamber	Immersion	-26, -30 and -34	Kaolinite	0.			
Sullivan et al. (2010a)	Continuous flow diffusion chamber	Deposition, immersion/ condensation	-25 and -30		0.			
Tobo et al. (2012)	Continuous flow diffusion chamber	Deposition, immersion/ Condensation	-26, -30 and -34	Kaolinite	.3 and 0.7	Dry particle suspension/ generation	CCN activation, aerosol mass spectrometry	Yes
Knopf and Koop (2006)	Optical microscope with a flow cell	Deposition	-13 to -76		0.7 to 10	Wet atomization	Assumption of spherical core shell model	No
Eastwood et al. (2009)	Optical microscope with a flow cell	Deposition	-27 to -40		5 to 15	Wet atomization	Assumption of spherical core shell model	Yes
Chernoff and Bertram (2010)	Optical microscope with a flow cell	Deposition	-26 to -39		~7 to 10	Wet atomization	Assumption of spherical core shell model	Yes
Present study	Continuous flow diffusion chamber	Deposition	-25, -30 and -35	Illite, Quartz, Kaolinite, Montmorillonite, Feldspar	0.2	Dry particle suspension/ generation	CCN activation, XRD	Yes. Depends on the coating thickness

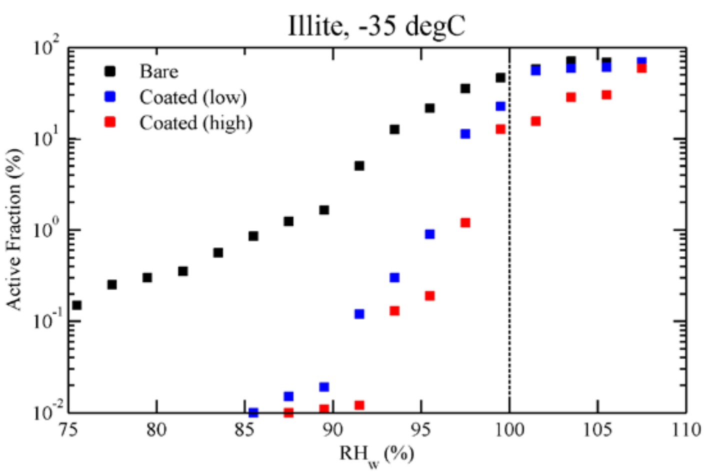


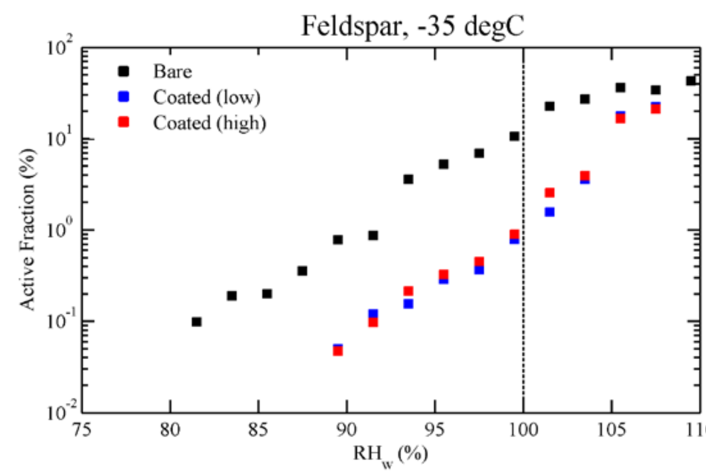
Concerns: Use of ATD and Wet generation method.
Murray et al., 2012; Hoose and Möhler, 2012; Herich et al., 2009; Sullivan et al., 2010;

Results







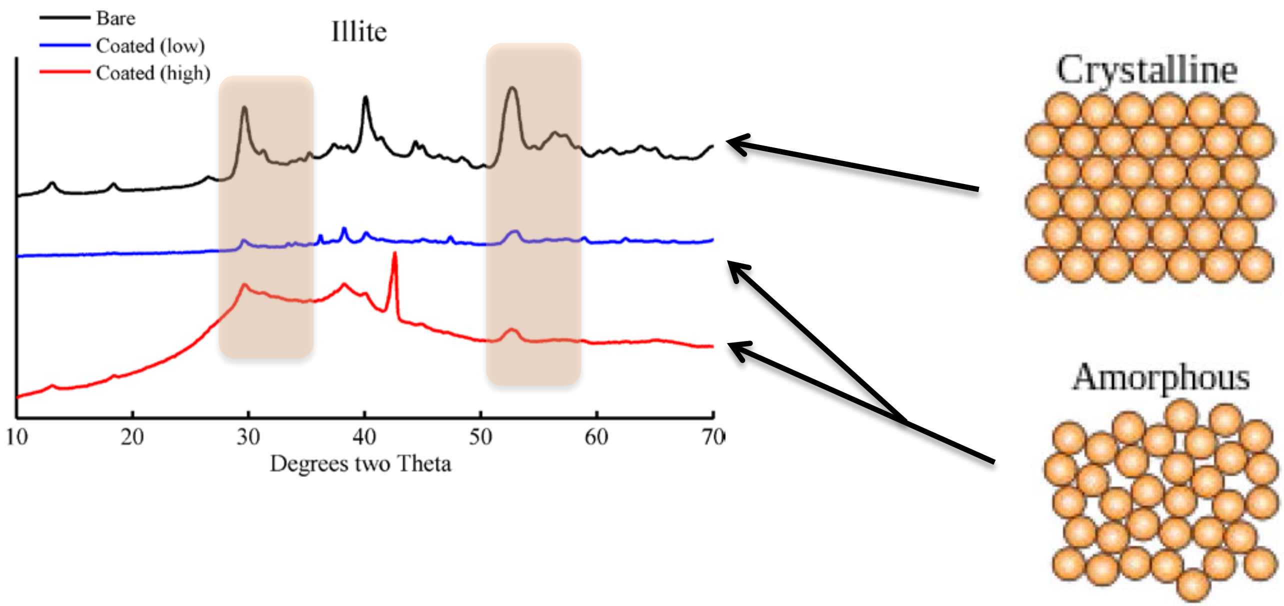


Coated particles showed lower IN activation efficiency. Similar results for all other four dust species at three different temperatures: -25, -30 and -35.

Note: For very low gas concentration efficiency was similar. XRD showed similar patterns.

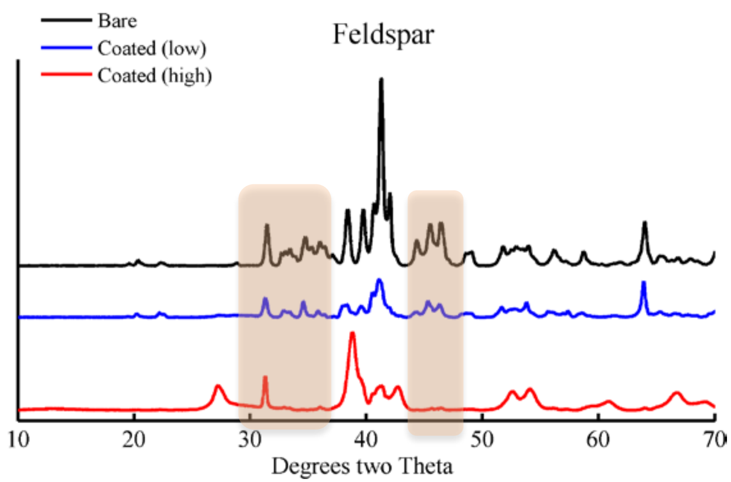
X-Ray Diffraction characterization patterns

Reveals why coated particles were poor IN



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).

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).

Modeling Implications

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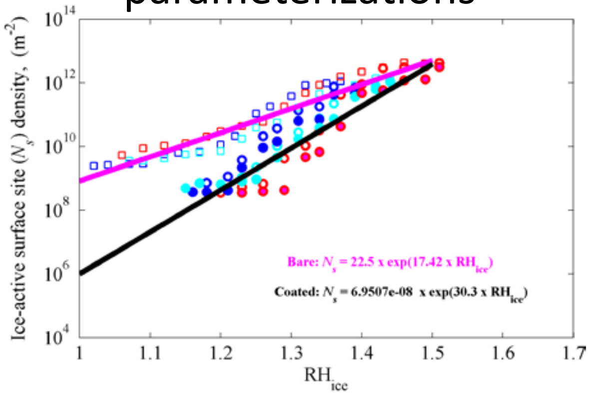
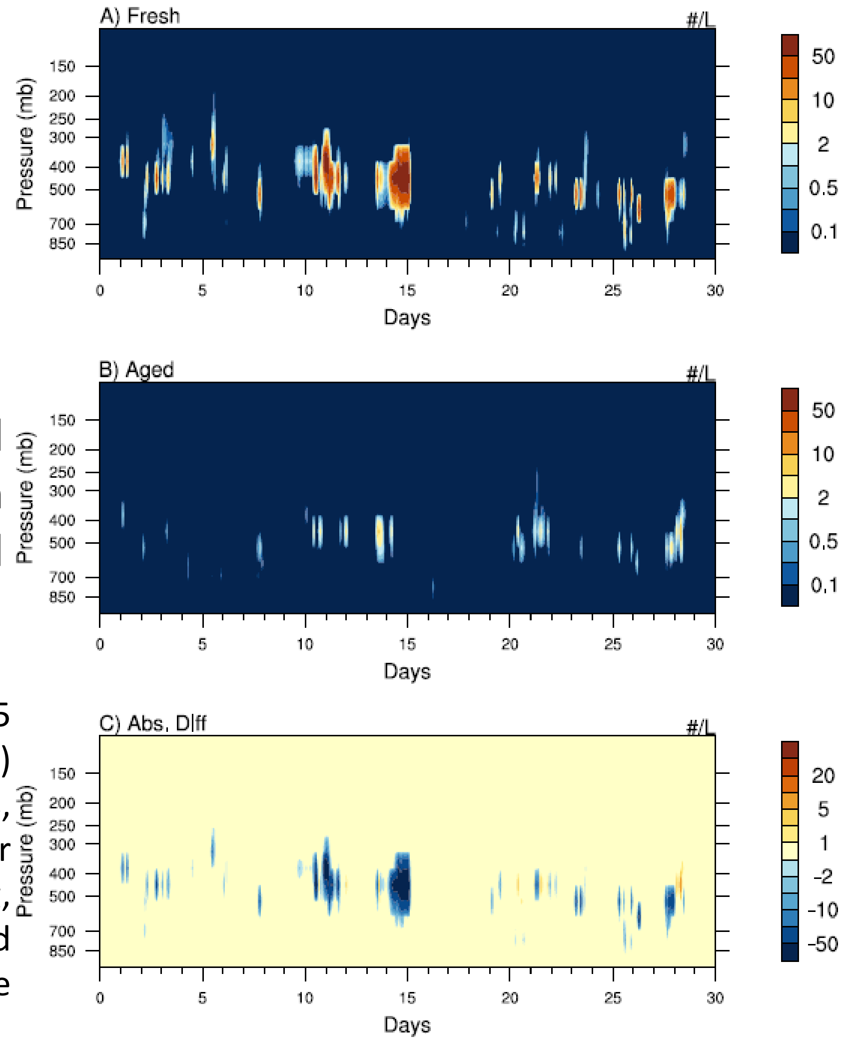
Active site density parameterizations

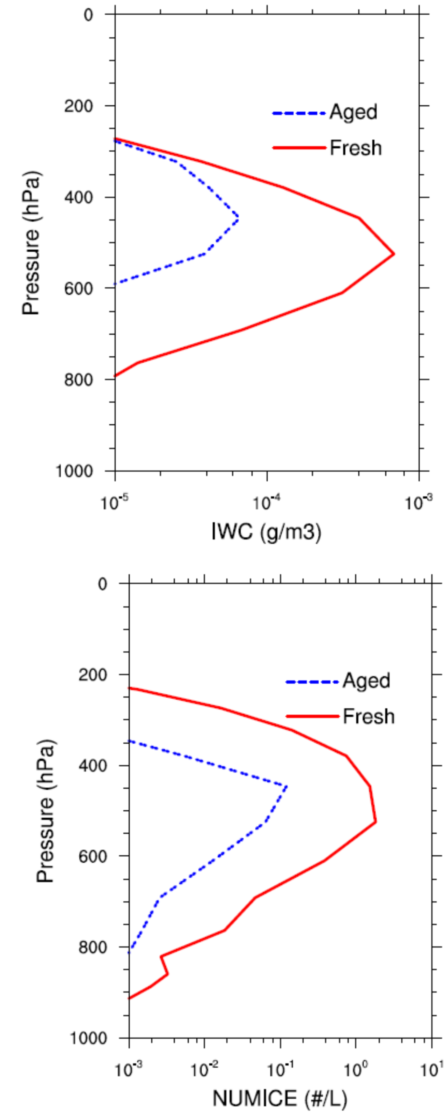


Model sensitivity in simulated IWC and IC concentration using uncoated and coated dust particles.

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.



Monthly mean profile



Optical cloud thickness increases as ice water path increases.

Ongoing work: Diesel soot characterization

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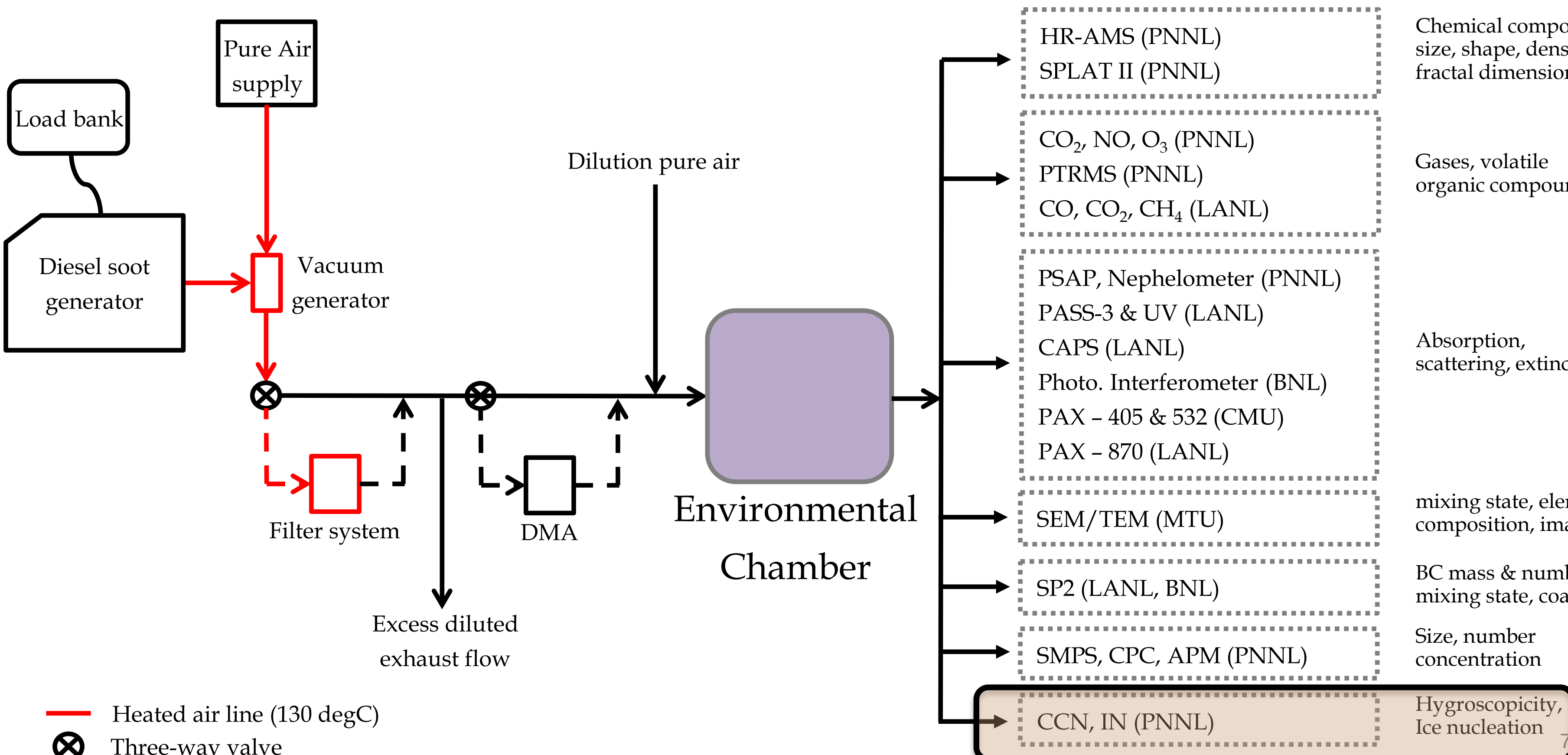
Role of OC/BC ratio towards IN activity ???

Soot generation & dilution → Soot ageing, mixing & coagulation → Soot measurement instrumentation → Soot properties



Summary

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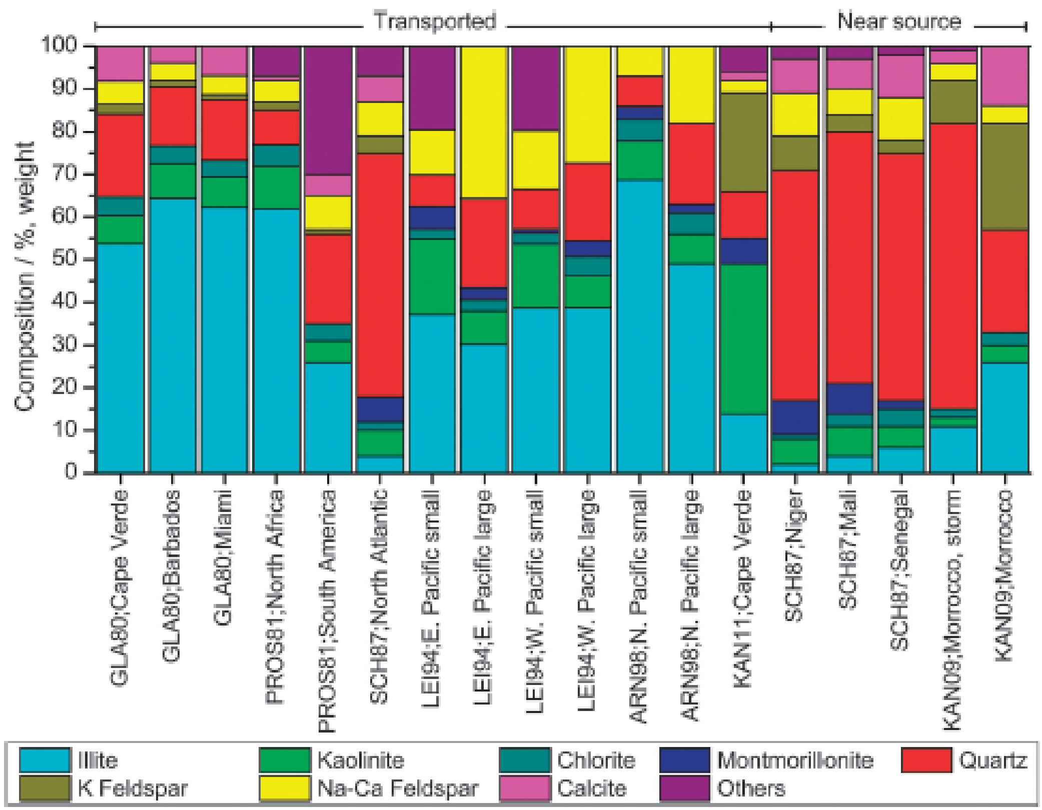
- ✓ IN nucleation efficiency of uncoated particles was higher than coated particles. Efficiency also depends upon the gas concentration.

- ✓ 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


Transported
Near source
Composition / %, weight
100
90
80
70
60
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40
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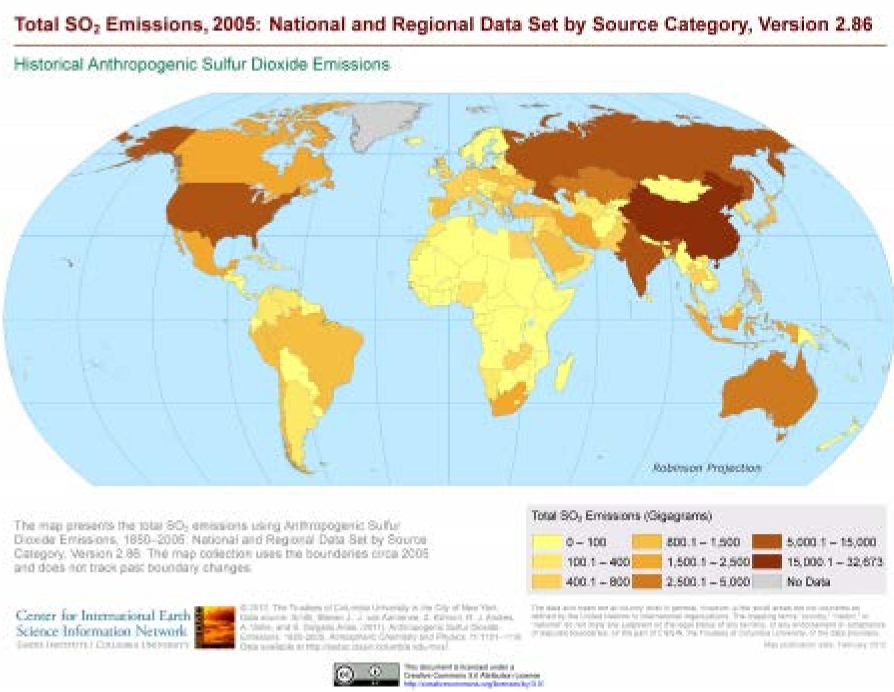
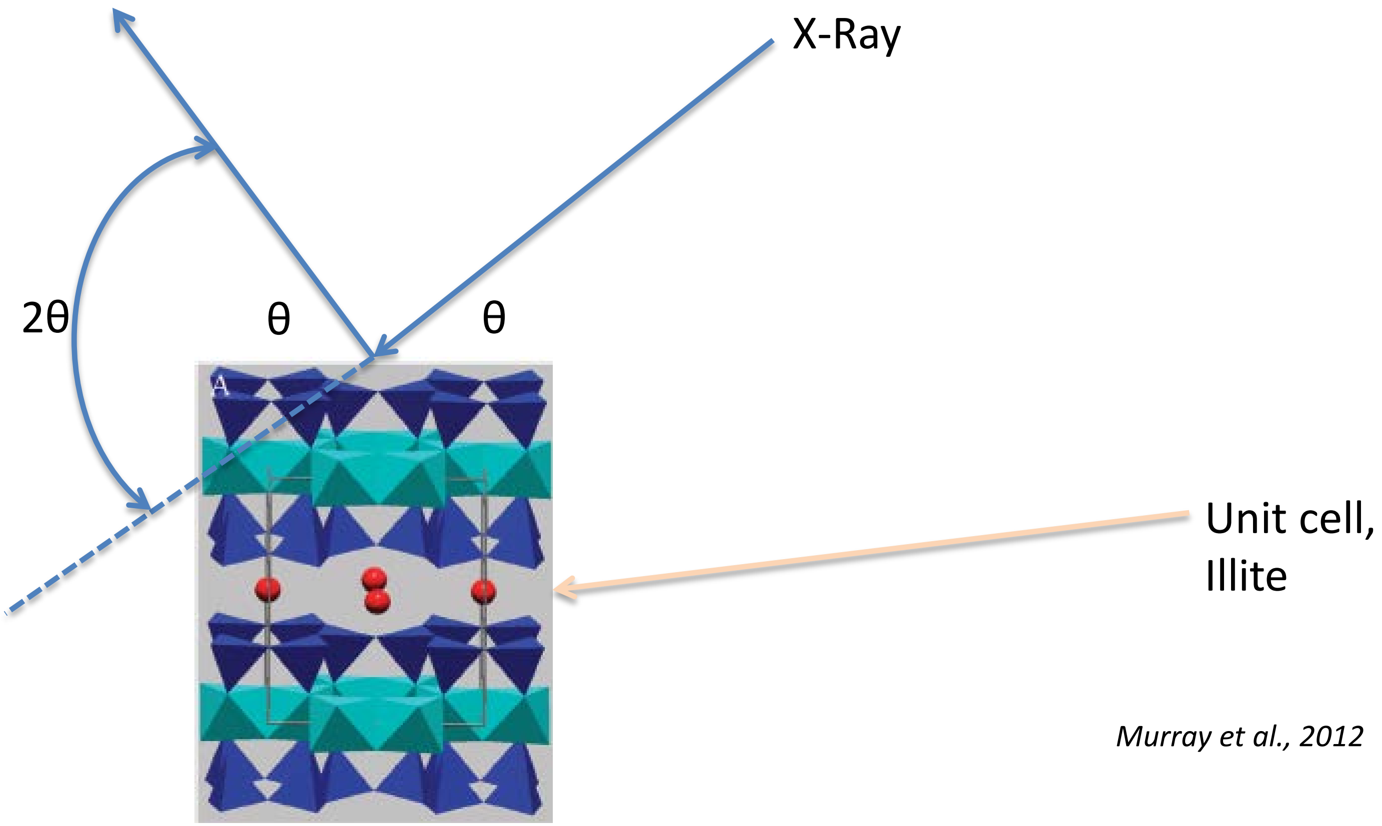
GLA80;Cape Verde
GLA80;Barbados
GLA80;Miami
PROS81;North Africa
PROS81;South America
SCH87;North Atlantic
LEI94;E. Pacific small
LEI94;E. Pacific large
LEI94;W. Pacific small
LEI94;W. Pacific large
ARN98;N. Pacific small
ARN98;N. Pacific large
KAN11;Cape Verde
SCH87;Niger
SCH87;Mali
SCH87;Senegal
KAN09;Morrocco, storm
KAN09;Morrocco
Illite
Kaolinite
Chlorite
Montmorillonite
Quartz
K Feldspar
Na-Ca Feldspar
Calcite
Others


Total SO_2 Emissions, 2005: National and Regional Data Set by Source Category, Version 2.86
Historical Anthropogenic Sulfur Dioxide Emissions
Robinson Projection
The map presents the total SO_2 emissions using Anthropogenic Sulfur Dioxide Emissions, 1850–2005: National and Regional Data Set by Source Category, Version 2.86. The map collection uses the boundaries circa 2005 and does not track past boundary changes.
Total SO_2 Emissions (Gigagrams)
0 – 100
100.1 – 400
400.1 – 800
800.1 – 1,500
1,500.1 – 2,500
2,500.1 – 5,000
5,000.1 – 15,000
15,000.1 – 32,673
No Data
Center for International Earth Science Information Network

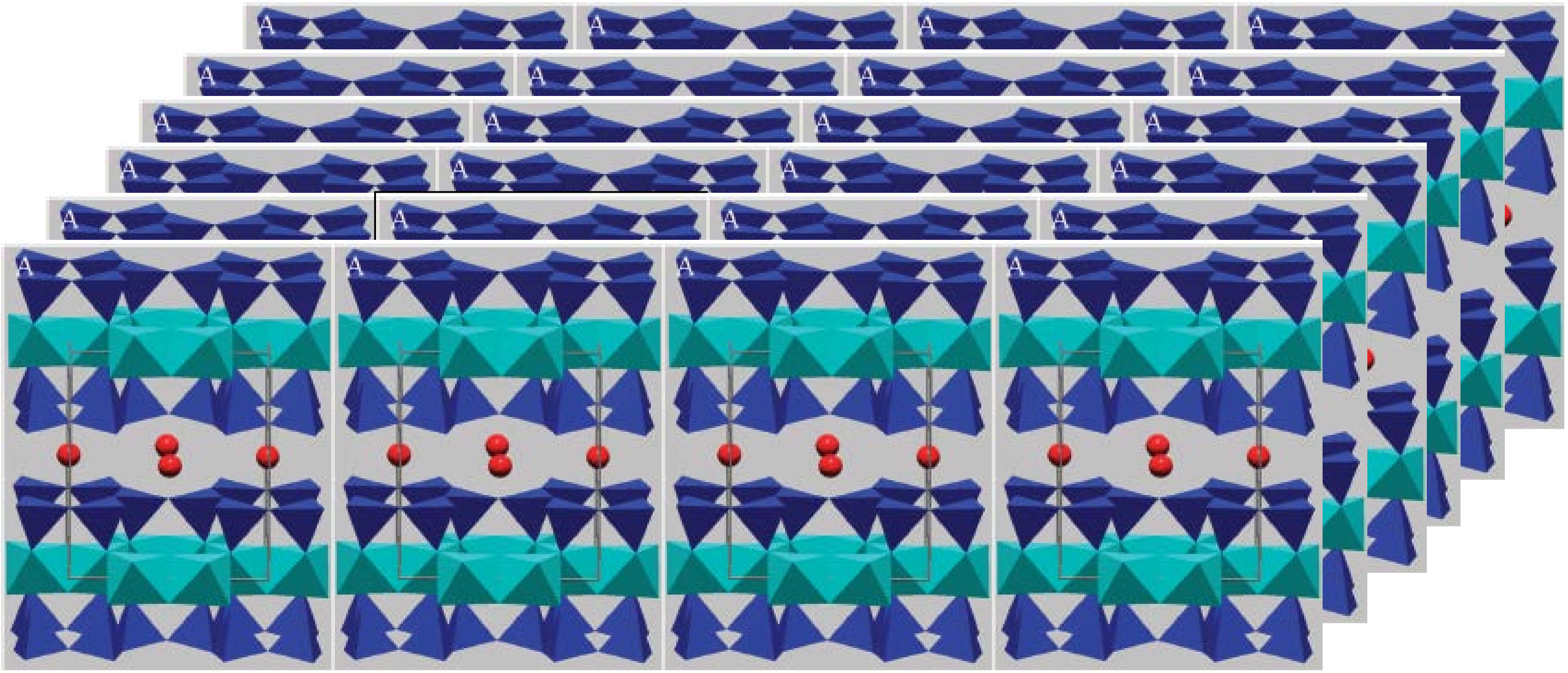
Supplementary Information



Murray et al., 2012

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.

Supplementary Information



If you remove crystalline structure or some unit cells, the peak intensity drops as strength of reflected (diffracted) X ray reduces.