On Immersion Freezing as a Nucleation Mechanism in Mixed-Phase Stratus

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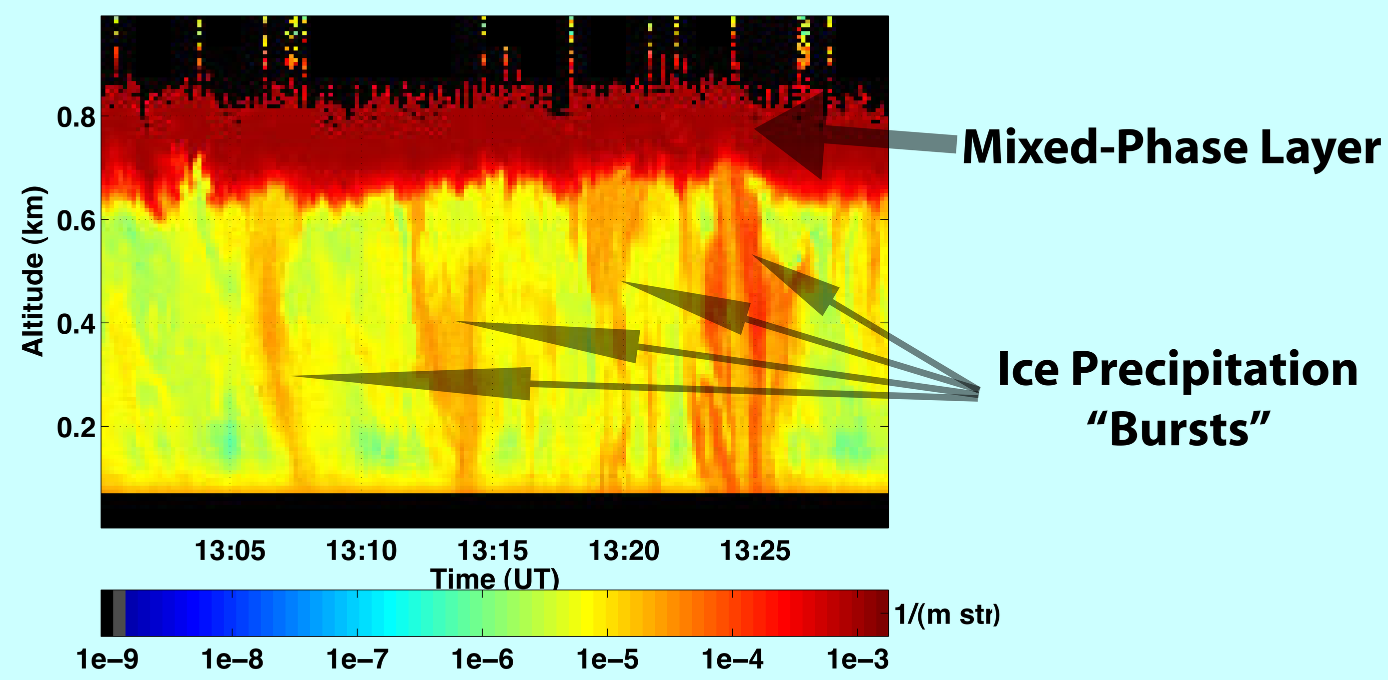
(1) Introduction

Ice formation appears to a dominant factor controlling the lifecycle of Arctic mixed-phase clouds. To date, our understanding of ice formation in these long-lasting cloud structures does not explain the formation of observed ice amounts. Particularly puzzling are observations taken from the 2004 Mixed-Phase Arctic Clouds Experiment (M-PACE) at the ARM North Slope of Alaska site (NSA) which show continuous mixed-phase clouds present with only minimal ice forming nuclei (IN) available. In-situ measurements of both ice particle and IN concentrations show IN concentrations multiple orders of magnitude lower than the ice particle concentrations. This discrepancy leads to the belief that certain classical nucleation mechanisms, such as contact, condensation and deposition freezing are not primarily responsible for ice production, as all require free IN for activation. Immersion freezing is not included with this grouping, however, as it is unclear whether IN immersed inside cloud droplets would be observed at all with instruments commonly used to measure IN concentrations.

Here, we investigate the potential role of immersion freezing in Arctic mixed-phase stratus. A theory on how immersion freezing fits into the lifecycle of these clouds, as well as a review of previous studies supporting this theory are presented.

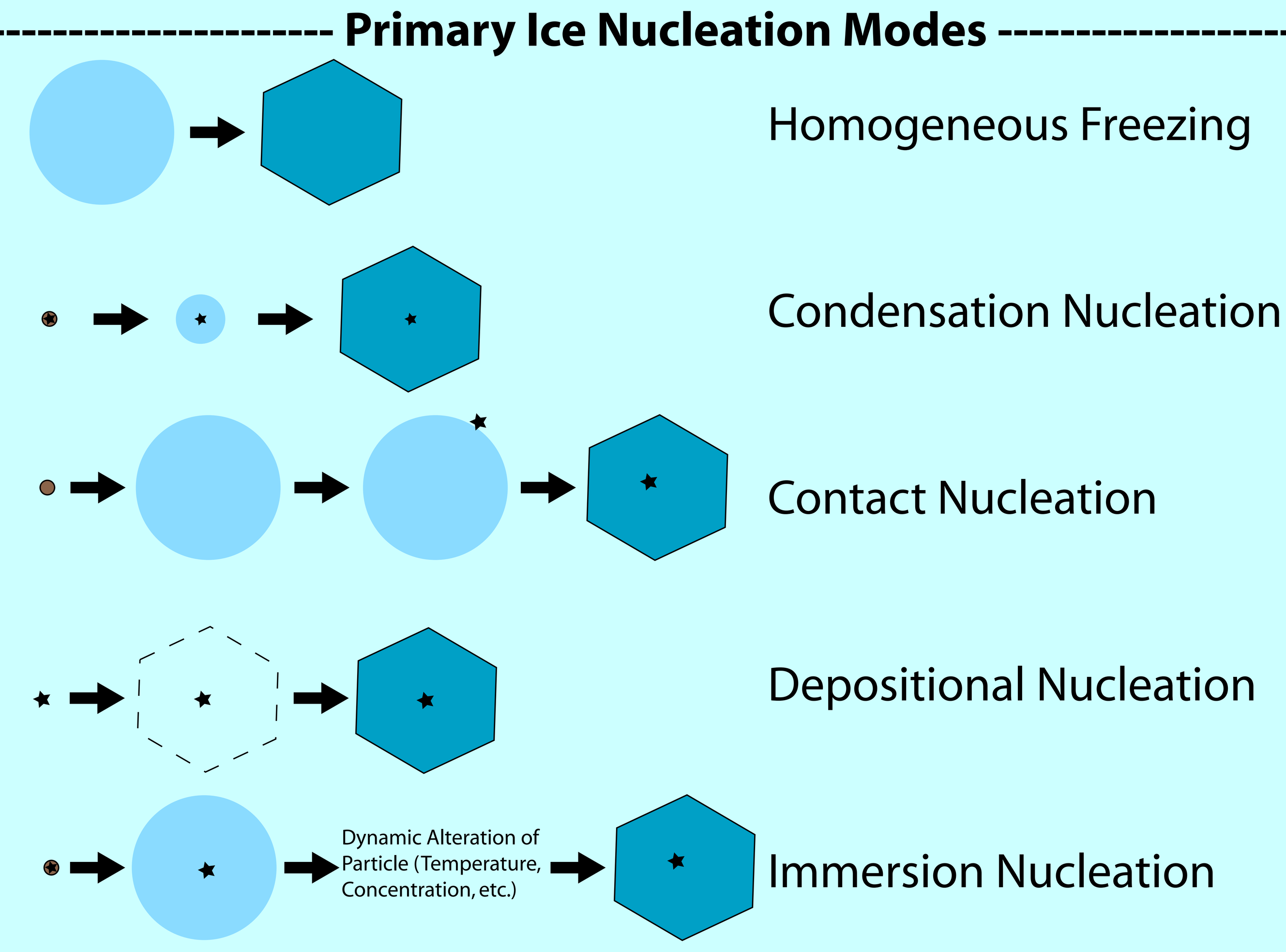
(2) Mixed-Phase Arctic Stratus from M-PACE

Lidar backscatter cross section (Masked values shown in black and white)



(3) Fundamentals

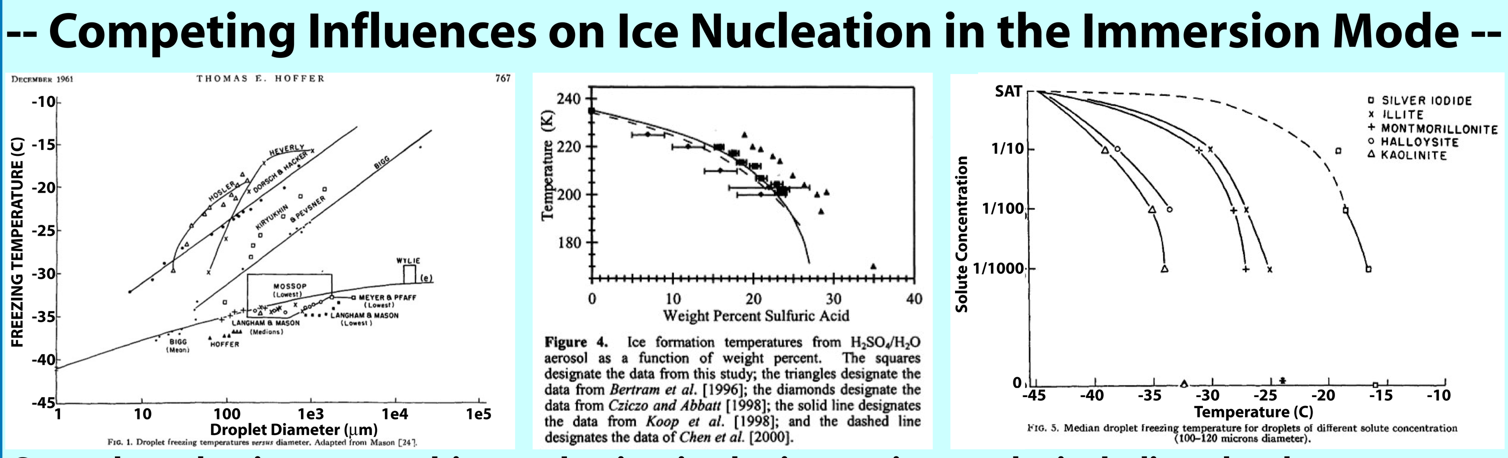
Primary Ice Nucleation Modes



So which one drives ice production in Arctic Stratus?

- Homogeneous freezing is insignificant > -35 C (Hagen et al., 1981; Sassen and Dodd, 1988; Jensen et al., 1998, others) Arctic stratus are observed at temperatures significantly above this (de Boer et al., 2008 in preparation).
- Ice crystal concentrations often significantly exceed measured IN concentrations (particularly for M-PACE!) (Mossop, 1970; Fridlind et al., 2007), meaning contact and depositional nucleation likely are unlikely the driving nucleation mechanism.
- Condensation/deposition nuclei would be detected with conventional instruments(such as a CFDC).

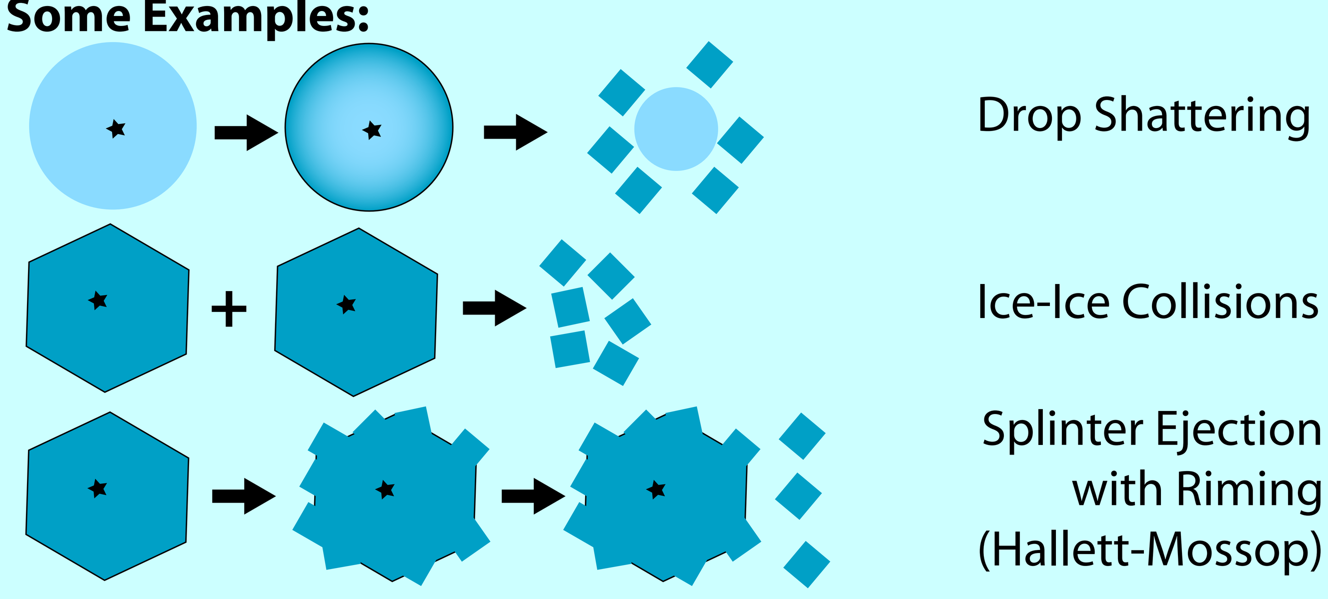
Competing Influences on Ice Nucleation in the Immersion Mode



Several mechanisms control ice nucleation in the immersion mode, including the drop curvature effect (left, from Hoffer (1961)), the solution effect (center, from Prenni (2001)), and the effect of the immersed insoluble particle (right, from Hoffer (1961)).

Secondary Ice Nucleation Modes

Some Examples:



How do these processes contribute?

- Drop shattering may result in 15 ice fragments/drop, but only in about 10% of drops larger than 50 µm, multiplying total ice by factor of 2, rarely (if ever) greater than factor of 10. (Pruppacher and Klett, 1997)
- Ice-ice collision multiplication (Rangno and Hobbs, 2001) requires significant ice to be present initially and would require a several order of magnitude multiplication factor.
- Splinter ejection during the riming process appears to be limited to air temperatures of -3° to -8° C. (Heymsfield and Mossop, 1984) Additionally, the production from this is estimated at 1 splinter per 250 larger than 12 µm drops rimed onto one crystal (Koenig, 1977; Beheng, 1982,1987; Cotton et al. 1986).

Although these and likely other (e.g. evaporation freezing, Fridlind et al., 2007) mechanisms may be active within mixed-phase stratus, it remains unproven that any one of these mechanisms would serve as a dominant nucleation mechanism covering the discrepancy in ice observed in these clouds and IN measurements.

(4) Theory

Why Immersion Freezing?

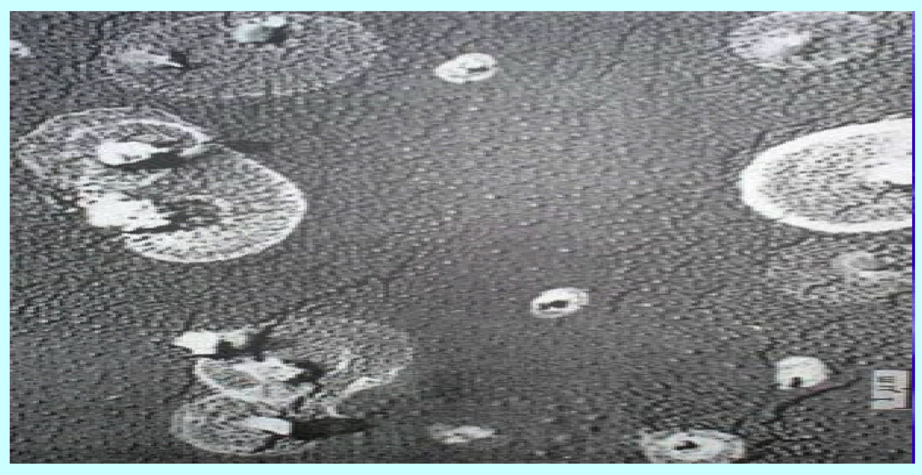


Image courtesy of J.P. Blanchet (From Bigg, 1980)

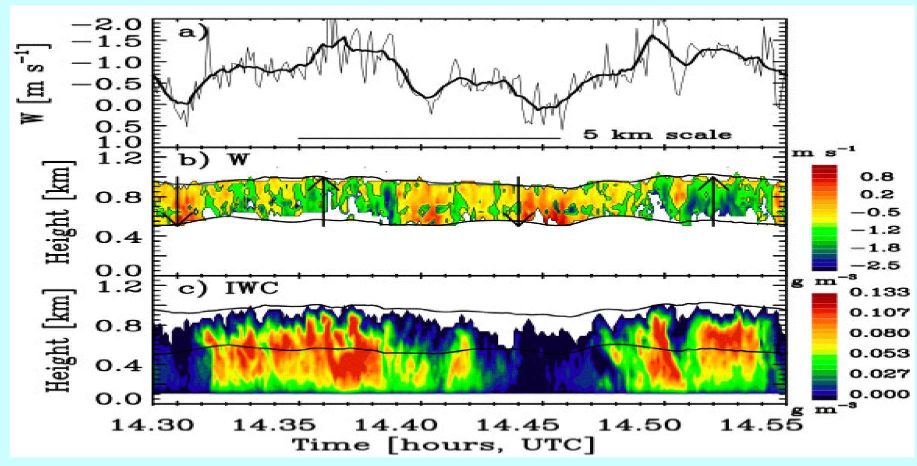
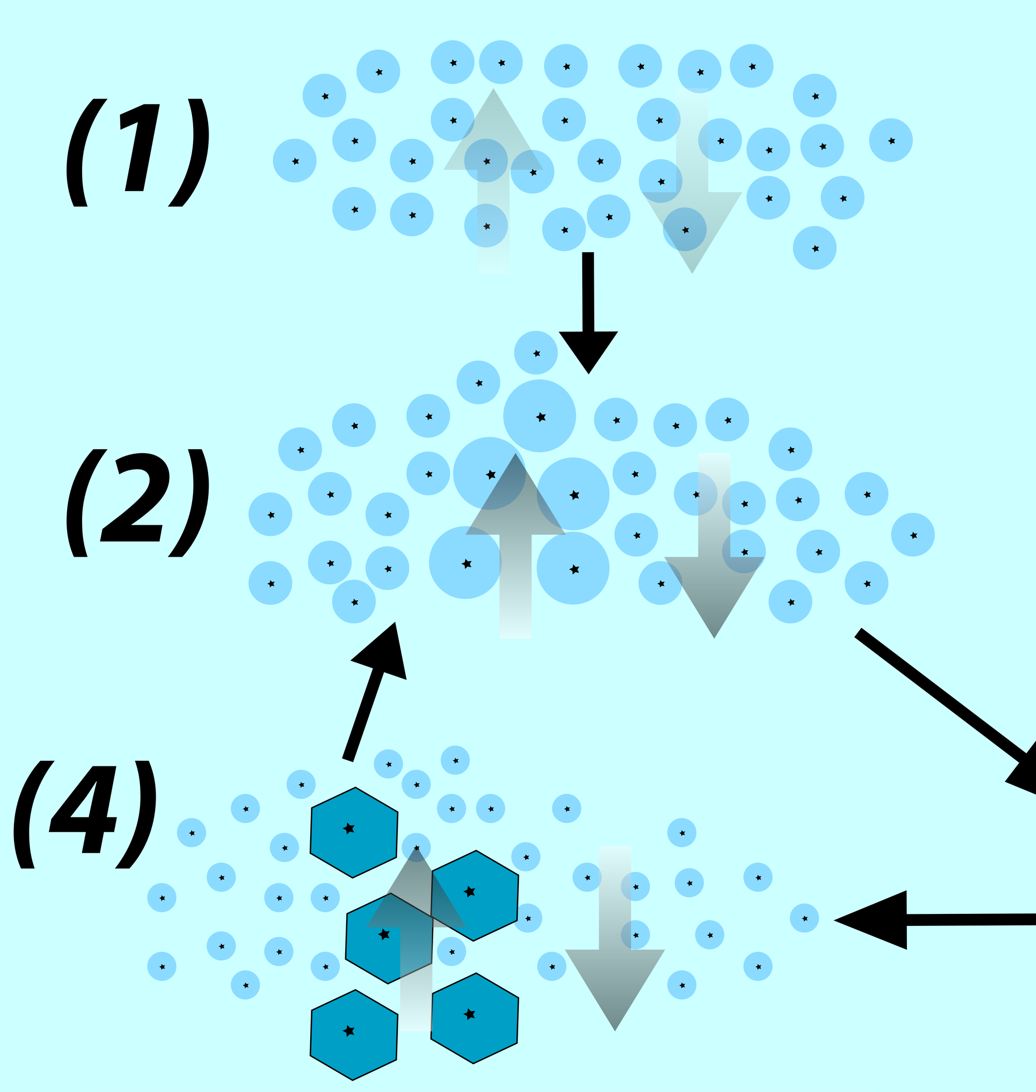


Image courtesy of Matthew Shupe

- Bigg (1980) observed sulfuric acid coating on aerosol particles during the winter.
- Blanchet (2007) hypothesizes that this sulfur coating is the result of anthropogenic emissions from Siberia, and are transported throughout the Arctic.
- This coating of soluble material inhibits ice formation on these particles, a process confirmed in the laboratory by Bertram and Girard, preventing uniform rapid ice formation.

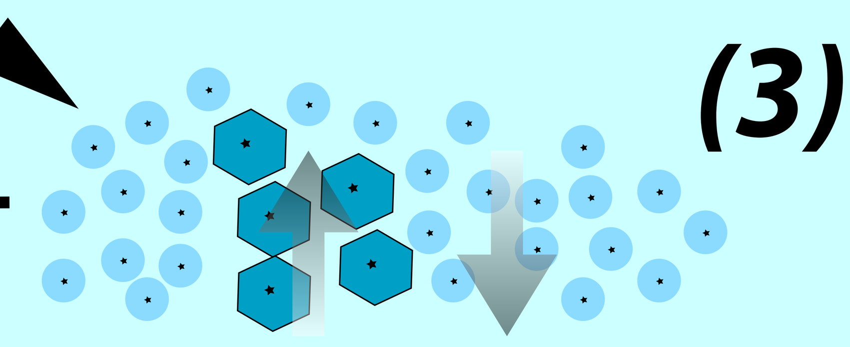
- Shupe (2006) illustrated that ice formation is seemingly linked to areas of upward vertical motion. This indicated that the formation of ice is tied into the internal dynamics of the cloud system, and likely an alteration of the aerosol or cloud particles involved in nucleation.
- Additionally, Shupe illustrated that ice water content and liquid water content seem to vary in phase with each other, hinting that liquid growth may lead to ice formation.
- In-situ measurements from Rangno and Hobbs (2001) reveal that ice crystal concentrations are highly proportional to the concentration of drops larger than 20 µm.

(5) Conceptual Model



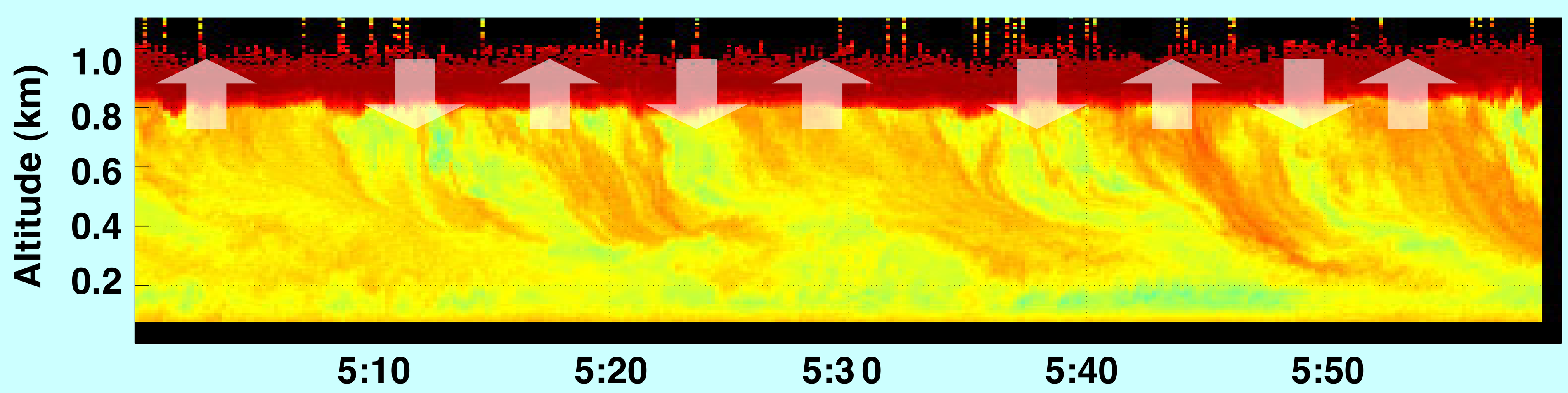
Radiative cooling from the surface leads to the saturation of a moist layer, and a liquid cloud forms. Some of these liquid droplets contain IN that had been coated in soluble material.

Radiative cooling from cloud top leads to vertical motion within the cloud layer. Droplets in the updrafts cool through expansion and accumulate liquid mass through condensational growth, decreasing the fraction of soluble mass within the droplet.

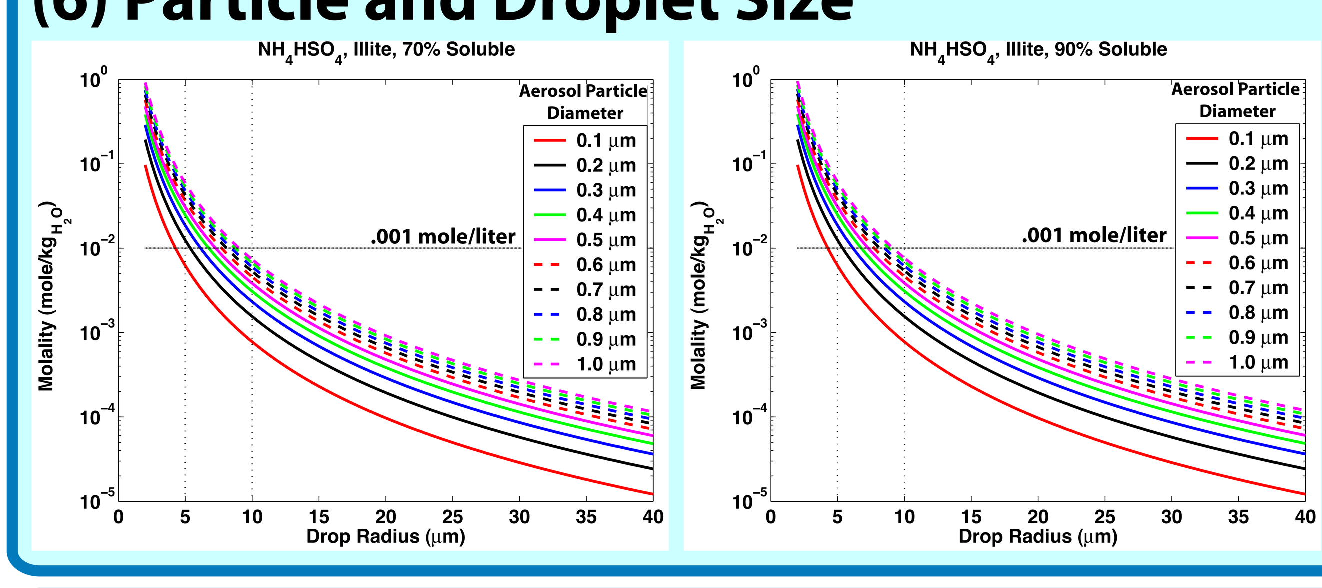


Growth of these ice particles uses up available moisture, and further droplet growth (and therefore ice formation) is halted. The ice particles rapidly grow to a size where they precipitate from the cloud layer, and the cycle starts over.

The fraction of soluble mass in the growing drops decreases to the point where freezing is no longer inhibited, and the larger nucleate into ice particles through the immersion freezing process.

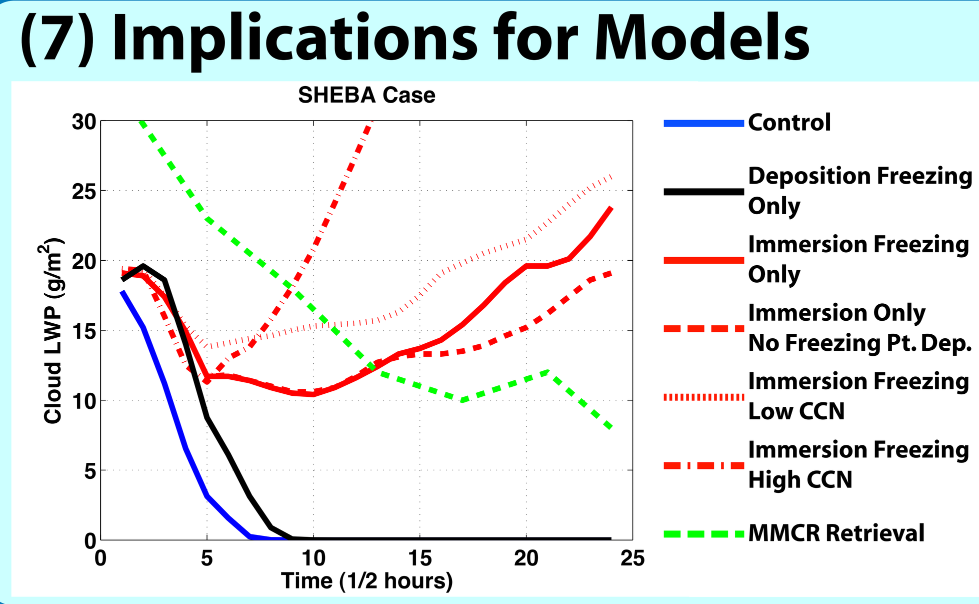


(6) Particle and Droplet Size

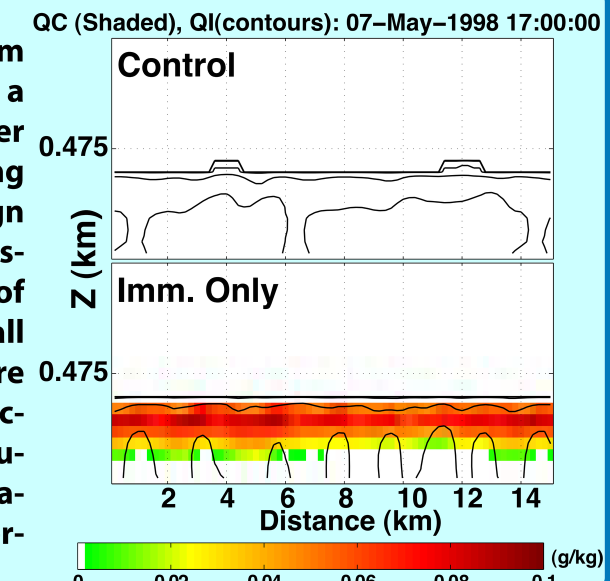


At left are plots of molality of the droplet solution for different drop radii. The different lines represent variation in initial mixed aerosol particle size (and thereby the amount of soluble material within the droplet). The right-hand plot is for aerosol particles that are 70% soluble material (by mass) and the left-hand plot is for 90% (by mass) soluble particles. Also plotted on both graphs is a horizontal line at 0.001 mole/liter. This threshold is mentioned in Prupacher and Klett as the point at which salts in a solution no longer significantly affect freezing temperature.

(7) Implications for Models



A comparison of LWP from several simulations of a mixed-phase single-layer cloud observed during the SHEBA campaign (left). Right, a figure illustrating the depletion of liquid cloud when all nucleation processes are active. The bottom section illustrates the simulated cloud for a simulation only using the immersion freezing process.



(8) Contact Information and Acknowledgements

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