Susceptibility of TTL cirrus to heterogeneous nuclei

Eric Jensen, Rei Ueyama, and Leonhard Pfister

• How do TTL water vapor concentration, relative humidity, cirrus microphysical properties, and cirrus occurrence frequencies change if the abundance of heterogeneous ice nuclei changes?

• Ueyama et al., “Dynamical, convective, and microphysical control on wintertime distributions of water vapor and clouds in the tropical tropopause layer”, JGR, in review. See poster.

• Motivation: Heterogeneous IN changes ➔ climate change

Data sources:
– ATTREX clouds (S. Woods, P. Lawson)
– ATTREX water, temp. (G. Diskin, J. DiGangi, T. Thornberry, A. Rollins, P. Bui)
– CALIOP clouds (M. Avery)
Homogeneous vs heterogeneous ice nucleation

• **Homogeneous freezing of aqueous aerosols**
  – General assumption is that most aerosols are aqueous sulfates (with other stuff)
  – Freezing occurs at $S_{ice} \approx 1.6–1.7$ ($RHI = 160–170\%$)
  – Freezing threshold is apparently composition independent, but small variations matter
  – Always plenty of aerosols ($N_{aer} = 10–10,000$ cm$^{-3}$, $N_{ice} < 1$ cm$^{-3}$)

• **Heterogeneous nucleation on solid particles**
  – Heterogeneous freezing on insoluble components in aqueous aerosols
  – Deposition nucleation on dry particles
  – Occurs at lower supersaturations ($S_{ice} \approx 1.1–1.6$)
  – Limited population of heterogeneous ice nuclei (IN, $N_{IN} < 100$ L$^{-1}$ (typically, no direct measurements in TTL))

• **Potential TTL IN**
  – Mineral dust
  – Metallic particles
  – Effloresced ammonium sulfate
  – Glassy organic-containing aerosols

**IN potentially changing as a result of invasive humans**
Hawkeye TTL cirrus measurements: ice concentration

- Reasonable consistency between different measurements of ice concentration

**Graph:**
- Ice number concentration
  - 2DS: optical array probe (10–4000 μm)
  - FCDP: spectrometer (1–50 μm)

**Data:**
- ATTREX-2014 2DS: $N_{i,\text{mean}} = 121.8 \text{ L}^{-1}$
- ATTREX-2014 FCDP: $N_{i,\text{mean}} = 93.1 \text{ L}^{-1}$
Hawkeye/CALIOP TTL cirrus measurements: extinction

- 2D-S (direct extinction measurement) has larger values than CALIOP (sampling bias?)

- Relatively small regional and interannual differences
ATTREX relative humidity measurements

- Diode Laser Hygrometer (DLH): Open-path TDL [G. Diskin]
- NOAA Water (NW): Internal-path TDL, vapor and total $H_2O$ [T. Thornberry, A. Rollins]
- Excellent agreement between measurements
- Peak near $RHI = 100\%$ physically expected
**TTL cirrus modeling approach**

1. Calculate 60-day backward diabatic trajectories from every 2° lat x 2° lon grid points in the tropics (20°S - 20°N) at 372 K (~100 hPa) level ending at 1 Feb 2007 using ERA-Interim temperatures and winds with enhanced wave-driven variability [Kim and Alexander 2013] and high-frequency waves added

   a sample of the trajectories and their temperatures

   • ERAi tropopause temperature bias < 0.3 K
TTL cirrus modeling approach

2. Use 1D (height) time-dependent microphysical model to simulate clouds along each parcel trajectory temperature curtain.

- **H\textsubscript{2}O curtain** (initialized with MLS measurements at time = 0)

- **Ice nucleation**

- **Convection** (air is saturated up to the cloud top height)

**Sedimentation is important for realistic simulation of ice concentrations**
Trajectories are traced through geostationary satellite convective cloud-top height fields

The model vertical column is saturated up to cloud top at intersections with convective clouds.
• High-frequency waves (< 2 cycles per day) produce rapid cooling events that result in large ice concentrations

• Need to evaluate with measurements (aircraft, balloon, etc.)
Sensitivity of microphysics to waves

ATTREX 2DS, \( N_{i,\text{mean}} = 121.5 \text{ L}^{-1} \)

\( T < 205 \text{ K} \)
Sensitivity of microphysics to waves

- Homogeneous freezing with nominal wave specification produces excessive ice concentrations
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- Strong sensitivity to high-frequency waves
Sensitivity of microphysics to waves

- Homogeneous freezing with nominal wave specification produces excessive ice concentrations
- Strong sensitivity to high-frequency waves
- Extinctions are too low without waves
Sensitivity of supersaturation to waves

- Homfrez only with waves ➞ too many ice crystals ➞ too little supersaturation
- No waves ➞ too few ice crystals ➞ too much supersaturation
Impact of waves on cloud frequencies

- Model with high-frequency waves does a reasonable job of simulating cloud frequencies

- Without waves, too few clouds
  - Primarily due to added temperature variability with waves [Kim and Alexander, *GRL*, 2015]
Impact of waves on cloud distributions

- Model (with waves) produces reasonable distribution of upper TTL cirrus, except for the south Pacific (ERA-Interim problem?)
Sensitivity of microphysics to heterogeneous nuclei

Ice number concentration

Relative frequency

T < 205 K

ATTREX 2DS, $N_{\text{mean}} = 121.5 \text{ L}^{-1}$

Homfrz only, $N_{\text{mean}} = 290.4 \text{ L}^{-1}$
Sensitivity of microphysics to heterogeneous nuclei

Ice number concentration

Relative frequency

T < 205 K

ATTREX 2DS, $N_{\text{mean}} = 121.5 \text{ L}^{-1}$

+ Homfrz only, $N_{\text{mean}} = 290.4 \text{ L}^{-1}$

$N_{\text{hi}} = 30 \text{ L}^{-1}, S_{\text{nuc}} = 1.3, N_{\text{mean}} = 197.5 \text{ L}^{-1}$
Sensitivity of microphysics to heterogeneous nuclei

Ice number concentration

Relative frequency

T < 205 K

ATTREX 2DS,  $N_{\text{mean}} = 121.5 \text{ L}^{-1}$

+ Homfrz only,  $N_{\text{mean}} = 290.4 \text{ L}^{-1}$

* $N_{n_h} = 30 \text{ L}^{-1}$, $S_{\text{nuc}} = 1.3$,  $N_{\text{mean}} = 197.5 \text{ L}^{-1}$

diamond $N_{n_h} = 100 \text{ L}^{-1}$, $S_{\text{nuc}} = 1.3$,  $N_{\text{mean}} = 145.3 \text{ L}^{-1}$
• Abundant IN improve model agreement with measured ice concentrations
Sensitivity of microphysics to heterogeneous nuclei

- Abundant IN improve model agreement with measured ice concentrations

- Extinctions are less sensitive to heterogeneous nuclei (10–30% changes with increasing IN)

### Ice number concentration

<table>
<thead>
<tr>
<th>Ice number concentration (L(^{-1}))</th>
<th>Relative frequency</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>0.001</td>
</tr>
<tr>
<td>100</td>
<td>0.010</td>
</tr>
<tr>
<td>1000</td>
<td>0.100</td>
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</tbody>
</table>

### Extinction

<table>
<thead>
<tr>
<th>Extinction (km(^{-1}))</th>
<th>Relative frequency</th>
</tr>
</thead>
<tbody>
<tr>
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<td>0.0001</td>
</tr>
<tr>
<td>0.0010</td>
<td>0.010</td>
</tr>
<tr>
<td>0.0100</td>
<td>0.100</td>
</tr>
</tbody>
</table>

### ATTREX 2DS

- Homfrz only, \(N_{\text{mean}} = 290.4\) L\(^{-1}\)
- \(N_{\text{IN}} = 30\) L\(^{-1}\), \(S_{\text{nuc}} = 1.3\), \(N_{\text{mean}} = 197.5\) L\(^{-1}\)
- \(N_{\text{IN}} = 100\) L\(^{-1}\), \(S_{\text{nuc}} = 1.3\), \(N_{\text{mean}} = 145.3\) L\(^{-1}\)
- \(N_{\text{IN}} = 1000\) L\(^{-1}\), \(S_{\text{nuc}} = 1.2-1.6\), \(N_{\text{mean}} = 123.3\) L\(^{-1}\)
Sensitivity of supersaturation to heterogeneous nuclei

- With abundant IN, large supersaturations rarely occur
Impact of heterogeneous nuclei on cloud frequencies

- Cloud frequencies (and regional distributions) are insensitive to heterogeneous nuclei

**Graph:**
- CALIOP
- Homfrz only
- $N_{IN} = 30 \text{ L}^{-1}$, $S_{i,nuc} = 1.3$
- $N_{IN} = 100 \text{ L}^{-1}$, $S_{i,nuc} = 1.3$
- $N_{IN} = 1000 \text{ L}^{-1}$, $S_{i,nuc} = 1.2-1.6$
Summary

- Dynamics, particularly high-frequency waves, have a large impact on TTL cirrus microphysical properties (via nucleation sensitivity to cooling rate) and occurrence frequency (via impact on minimum temperatures [Kim and Alexander, 2015]).
  - High-freq waves are required to explain extinctions and occurrence frequencies
  - TTL supersaturation sensitive to waves and cirrus ice concentrations

- With homogeneous freezing alone, model produces excessive ice concentrations
  - Composition dependence and high-frequency waves may reduce ice concentrations produced by homogeneous freezing

- Inclusion of heterogeneous ice nuclei improves agreement with measured ice concentrations and extinctions
  - Observed frequencies of large ice supersaturations and very high ice concentrations imply that effective IN do not dominate ice nucleation all of the time

- TTL cirrus extinctions and ice water contents are relatively insensitive to heterogeneous nuclei abundance

- Heterogeneous nuclei have very little impact on TTL cirrus occurrence frequencies
Geoengineering by seeding cirrus with effective ice nuclei

  - Introduction of heterogeneous nuclei into upper troposphere will solve climate problem

  - Cirrus modification side effects of stratospheric sulfate aerosol geo-engineering
  - Global model with parameterized cirrus used to estimate radiative forcings

  - Cirrus modification side effects of stratospheric sulfate aerosol geo-engineering
  - Trajectory aerosol simulations and cirrus box model used to provide radiative forcing estimates (?!?)

  - GCM with parameterized cirrus used to predict radiative forcing
  - Potential negative effects noted
Importance of sedimentation

- Parcel models will overestimate ice concentrations
• Ice concentration (just after nucleation) increases rapidly with cooling rate
• Ice crystals nucleated on IN quench rising supersaturation, prevent homogeneous freezing, and limit ice concentration
Hawkeye/NOAA-WV TTL cirrus measurements: ice water content

- Ice mass from 2D-S requires area-mass relationships
Uncertainties

- Trajectory curtain approach: parcel pathway (and temperature variability) only correct for parcel along single trajectory (run from 372 K); no wind shear effects on cloud evolution

- Monthly-mean heating rates used despite large variations in actual heating rates associated with clouds

- High-frequency waves are specified with a statistical parameterization independent of latitude, longitude, and time

- No horizontal mixing included

- No interaction between cloud radiative heating, dynamics, and cloud processes

- Data sampling biases (how representative is the 30+ hour ATTREX-Guam Hawkeye dataset?)