Evaluating Impacts of Aitken Mode Aerosols on Marine Mixed Phase Clouds over the Arctic

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Low-level mixed phase clouds (those containing liquid and ice) are ubiquitous over the Arctic, and can persist for days to weeks in spite of being thermodynamically unstable (e.g., Morrison et al., 2012). Occurrence frequencies from 2007 to 2010 from CALISPO (Mioche et al., 2015)

- Liquid-topped and often precipitating ice.
- Supercooled liquid tops result in strong cooling rates, driving buoyant production of upward, turbulent motion.

Taken from Shupe (2007)
• Tendency of aerosols acting as CCN depends on size (amongst other factors), where larger CCN are more likely to activate at lower ambient supersaturations.

• However, likelihood of activation also depends on availability of CCN. Fewer CCN (e.g., remote marine regions, scavenging, etc.) can allow larger supersaturations to activate Aitken mode CCN (<100 nm) (e.g., Fan et al., 2018).

Background – Aitken mode CCN

SAM-LES: Cu rising into Sc layer from ACE-ENA RF16 (McCoy et al., submitted)
Background – Aitken mode CCN

- Evidence Aitken mode cloud condensation nuclei (CCN) can increase cloud brightness/lifetimes (*Aitken Buffering*)

- However, most studies have relied on modeling and focused on warm clouds

**Objective:** Use observations to perform climatological analysis of Aitken mode CCN impacts on mixed phase clouds in the Arctic
Instrumentation and Observation sites

1. ARM Cold-Air Outbreaks in the Marine Boundary Layer Experiment (COMBLE) Field Campaign
   • Andøya, Norway (69° latitude)
   • 12/2019-05/2020
   • CCN counter (critSS: 0.1–1%)
   • SMPS

2. ARM Ground site: Utqiaġvik (formerly Barrow), Alaska (69° latitude)
   • 03/2007-06/2011
   • 35 GHz Radar
   • Doppler Lidar
   • CCN counter (critSS: 0.1–1.5%)
   • Temporal resolution ~1 hr (time for CCN counter to scan all SScrit)

Task #1: Derive CCN data product reported as function of diameter
   • Use results from COMBLE to derive/verify product

Task #2: Use CCN product at location for multi-year analysis (Utqiaġvik, Alaska)
How do we get obtain Aitken mode CCN from ground site?

Convert critical supersaturation (critSS) to dry aerosol diameter ($D_d$)

$$D_d = \sqrt[3]{\frac{4A^3}{27\kappa n^2(\text{critSS})}}$$  

$$A = \frac{4\sigma_{s/a} M_w}{RT \rho_w}$$

Petters and Kreidenweis (2007) provide relationships between dry particle diameter and CCN activity using single hygroscopicity parameter ($\kappa$)

**Task #1:** Derive CCN concentrations as function of dry diameter for COMBLE

Fig. 1. Calculated critical supersaturation for $0 \leq \kappa \leq 1$ computed for $\sigma_{s/a}=0.072$ J m$^{-2}$ and $T=298.15$ K. The gray lines are linearly spaced intermediates.
Comparing SMPS ($N_{aer}$) with CCN counter ($N_{CCN}$) - COMBLE

\[ N_{aer,D1toD2} \approx N_{ccn,D2} - N_{ccn,D1} \]

CCN counter supersaturations

Derived dry CCN diameters
Comparing SMPS ($N_{aer}$) with CCN counter ($N_{CCN}$) - continued

Although relatively significant differences occur between CCN and SMPS for smaller particle sizes,

- Differences are robust between small and large particles
- Can compensate with sufficiently large sample size

Can compare and contrast Aitken mode ($N_{CCN\_Aitken}$; ~50-100 nm) and accumulation mode ($N_{CCN\_Accum}$; >~100 nm)

Percent difference ($\%$) = $\frac{|N_{aer} - N_{ccn}|}{N_{aer}^2} \times 100\%$
Initial CCN findings – Utqiagvik, Alaska | 2007-2011

- Aitken mode peaks in Spring & Summer consistent with previous studies (e.g., Freud et al., 2017)

Task #2: Use CCN product for multi-year, mixed phase cloud analysis
Initial CCN findings – Utqiaġvik, Alaska | 2007-2011

- Aitken mode peaks in Spring & Summer consistent with previous studies (e.g., Freud et al., 2017)

Aitken mode is more variable than Accumulation mode
Impacts of Aitken mode CCN on mixed phase cloud depth

**Conditions:**
- Synergistic method (radar and lidar) to obtain phase (Shupe, 2007)
- In-cloud samples using Clothiaux et al. (2000) cloud mask
- Mixed phase condition: Mixed phase / All phase range bins > 0.2
- Results only shown for high Aitken months (March-August)

Deeper clouds associated with high $N_{CCN\_Aitken}$
- Greatest differences at low $N_{CCN\_Accum}$

### Dependent Variables
- **CCN_Accum**
- **CCN_Aitken**

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<thead>
<tr>
<th>CCN_Accum</th>
<th>CCN_Aitken</th>
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<tr>
<td>Lower</td>
<td>Middle</td>
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### Samples Sizes
227-1533
Conclusions:
- Can derive a CCN product to compare and contrast Aitken and Accumulation mode CCN
  - Derive CCN size distributions using varying critSS from CCN counter
- Significant variability of Aitken mode in spring and summer
- Mixed phase cloud layers are deeper in high Aitken mode environments
  - Most notably in low accumulation mode environments

Future work:
- Continue evaluation of mixed phase cloud – Aitken mode CCN interactions
  - Reduce CCN product uncertainty (explore determination of \( \kappa \))
- Compare ground-based and airborne in-situ measurements
  - ACE-ENA, ACME

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References:


Extra slides
Aitken and Accumulation mode $N_{CCN}$ primarily have positive covariance at large $N_{CCN\_Ait}$ and ~0 covariance at low $N_{CCN\_Ait}$. 

Colored by Cov($N_{CCN\_Ait}$, $N_{CCN\_Accum}$) ($cm^{-6}$)
Non-mixed phase samples (mixed phase ratio = 0)
- March to August
Mixed phase samples)
- All year

<table>
<thead>
<tr>
<th>Cloud depth (m)</th>
<th>N CCN_Accum</th>
<th>Lower tercile</th>
<th>N CCN_Aitken</th>
<th>Middle tercile</th>
<th>N CCN_Accum</th>
<th>Upper tercile</th>
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- Lower tercile:
  - N: 33
  - Aitken: 33
- Middle tercile:
  - N: 67
  - Aitken: 67
- Upper tercile:
  - N: 67
  - Aitken: 67

-3 cm

-3 cm

-3 cm
Background – Arctic clouds

**Arctic amplification** is the result of multiple drivers and feedbacks, many of which are related to clouds.

Linear trends in temperature from 1960-2009. (Serreze & Barry, 2011)

**Climate modeling errors**

Significant errors and spread in Coupled Model Intercomparison Project phase 6 (CMIP6) cloud fractions as well as liquid and ice water paths.

Wei et al. (2021)
Background – Mixed phase clouds

Low-level mixed phase clouds
Liquid-topped and often precipitating ice

Aerosol-cloud interactions globally

Spread in radiative forcing $\sim 2.5-3$ W m$^{-2}$

Arctic clouds are especially susceptible to indirect effect (2 to 8 times more compared to other regions) due to relatively low cloud droplet concentrations. (Coopman et al. 2018)
Initial findings - Utqiagvik, Alaska | 2007-2011

Accumulation mode (>~100nm)
Aitken mode (~50-100nm)

N_{CCN} (cm^{-3})

# of samples

Month (Jan-Dec)

N_{CCN, Aitken}

Accumulation mode (>~100nm)
Aitken mode (~50-100nm)
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Spread in radiative forcing ~2.5-3 W m⁻²

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Spread in radiative forcing ~2.5-3 W m\(^2\)

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Taken from Lohmann (2017)
Derived kappa parameter: The Cloud Condensation Nuclei (CCN) Counter and Scanning Mobility Particle Sizer (SMPS) Derived Hygroscopicity Parameter Kappa (CCNSMPSKAPPA) value-added product uses measurements from CCN counter and SMPS instruments. The particle size distribution was integrated from the largest particle size toward smaller particle size until the total number of particles was equal to the CCN number. The particle size at which CCN and size distribution concentrations are equal is called the critical diameter, and for each critical diameter value, a single hygroscopicity parameter kappa using well-established kappa-Köhler theory was calculated (Petters and Kreidenweis 2007). The kappa values are calculated at each different water supersaturation value (0.1 to 1.0%).