Metastable cloud droplets near cloud top? A study of cloud properties using lidar and polarimeter measurements

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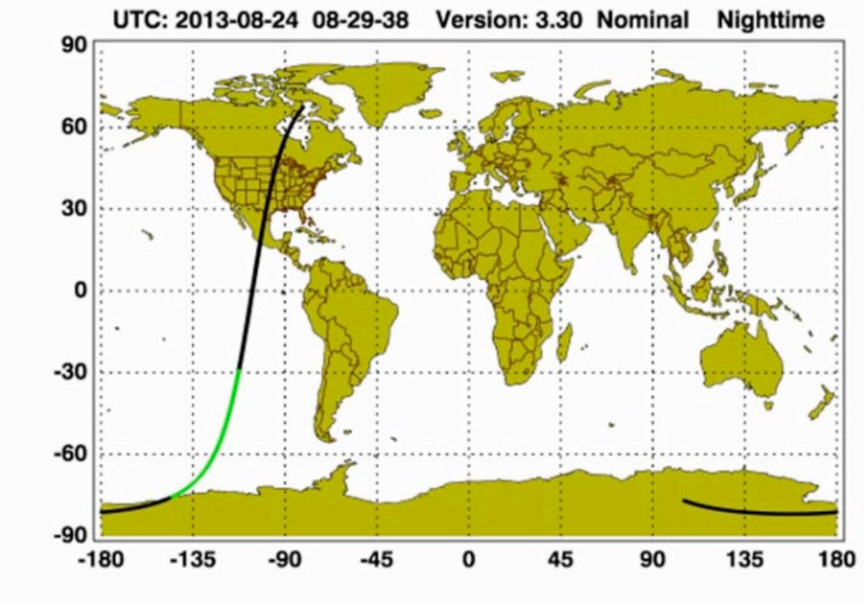
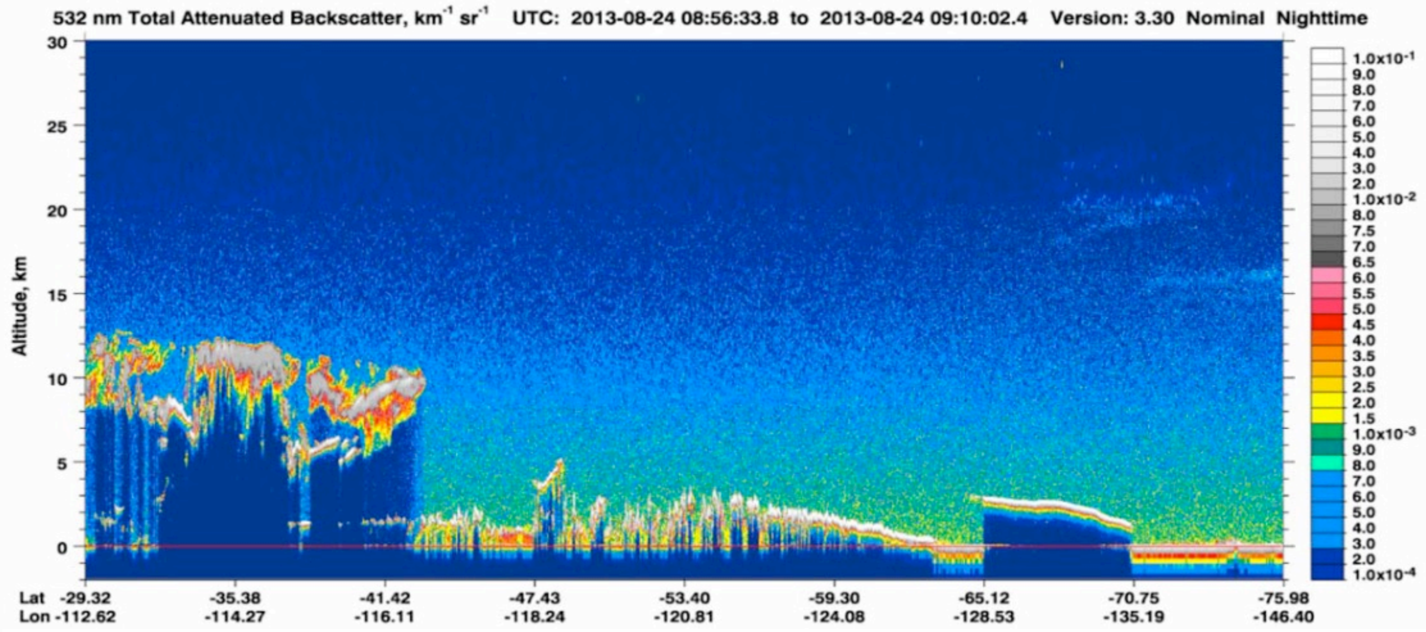
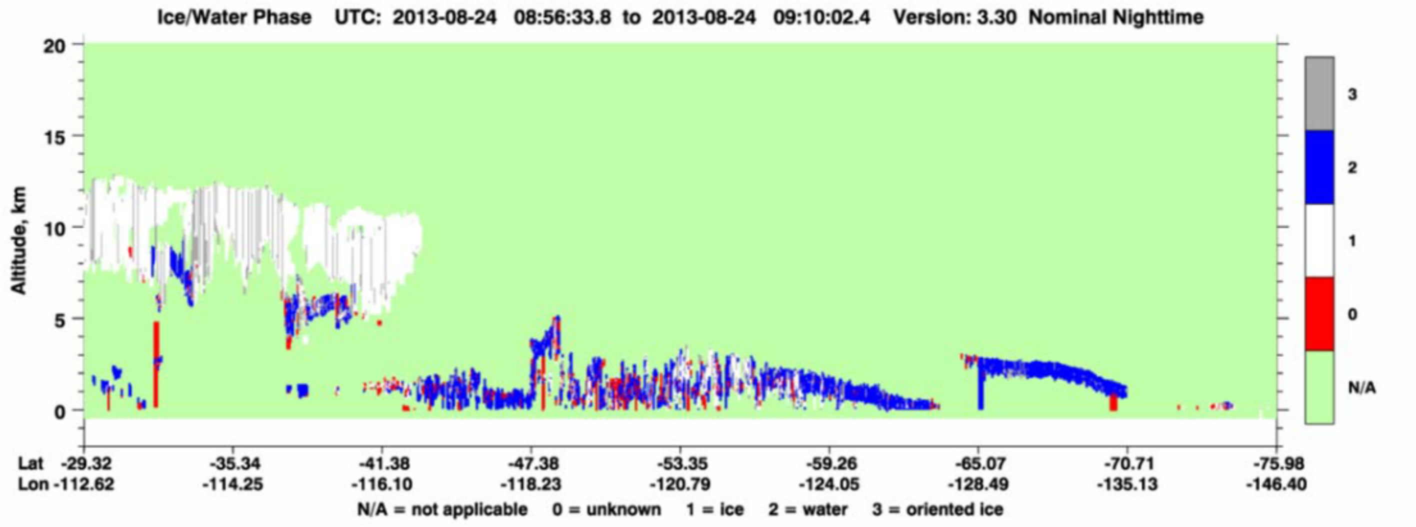
Outline

- Problem in understanding lidar measurements of water cloud droplets
  1. Measured lidar ratio of water clouds are smaller comparing with theory: backscatter too strong
  2. Problem is worse at nighttime comparing with day-time
  3. Potential explanation: enhancement from coherent backscatter? Refractive index different from lab measurements?
- Hypothesis: some of the supercooled water cloud droplets at cloud top are in meta-stable states
  1. Phase change of water droplets: meta-stable state may happen at cold temperature
  2. Meta-stable state of water has much lower super-saturation vapor pressure than water and ice
  3. Some studies suggested that amorphous water state is common in cold rain clouds
  4. Longwave radiation cools cloud droplets at cloud top at the rate of about 2^0C/second, which is very difficult to be balanced by condensation growth and heat diffusion and thus glassy water and can form, grow and crystalize – a potential ice nucleation mechanism in cold rain clouds ?
- Initial results from parasol measurements
  - Difference in P_{12} between water cloud droplet and amorphous water droplet
  - Glory observations from POLDER
  - If confirmed, the meta-stable cloud droplets can be key to modeling ice nucleation in cold rain process
  - Discussion, What’s next and POLCUBE-1

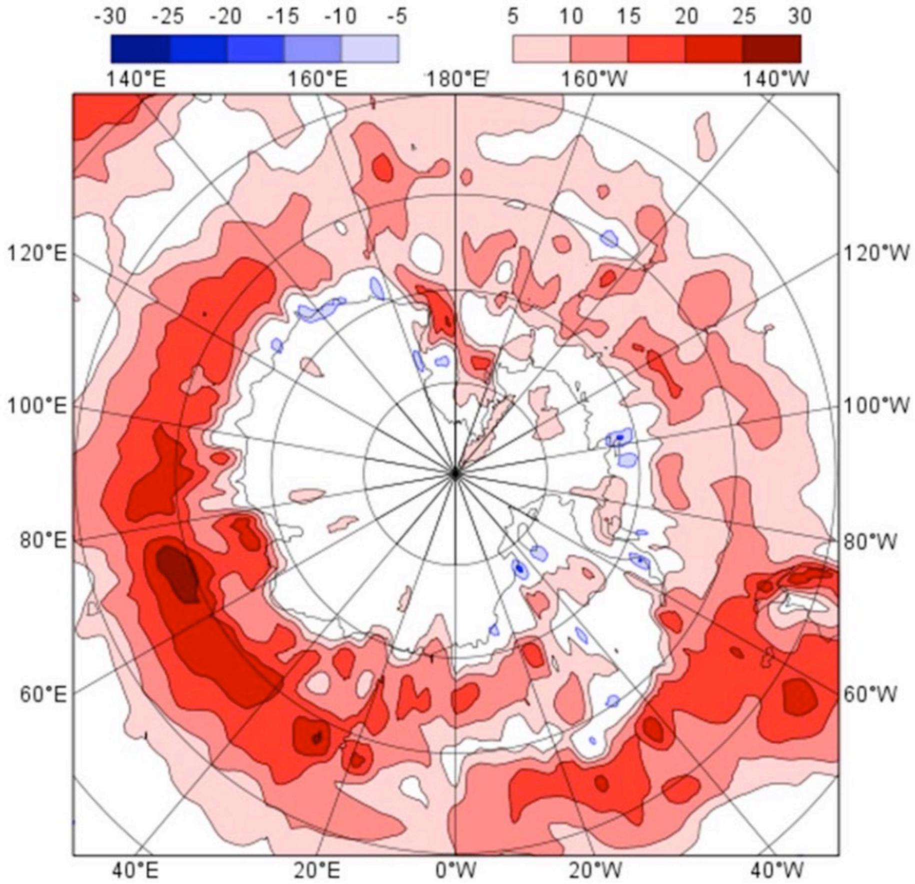
Southern oceans: frequent occurrence of supercooled liquid water cloud droplets



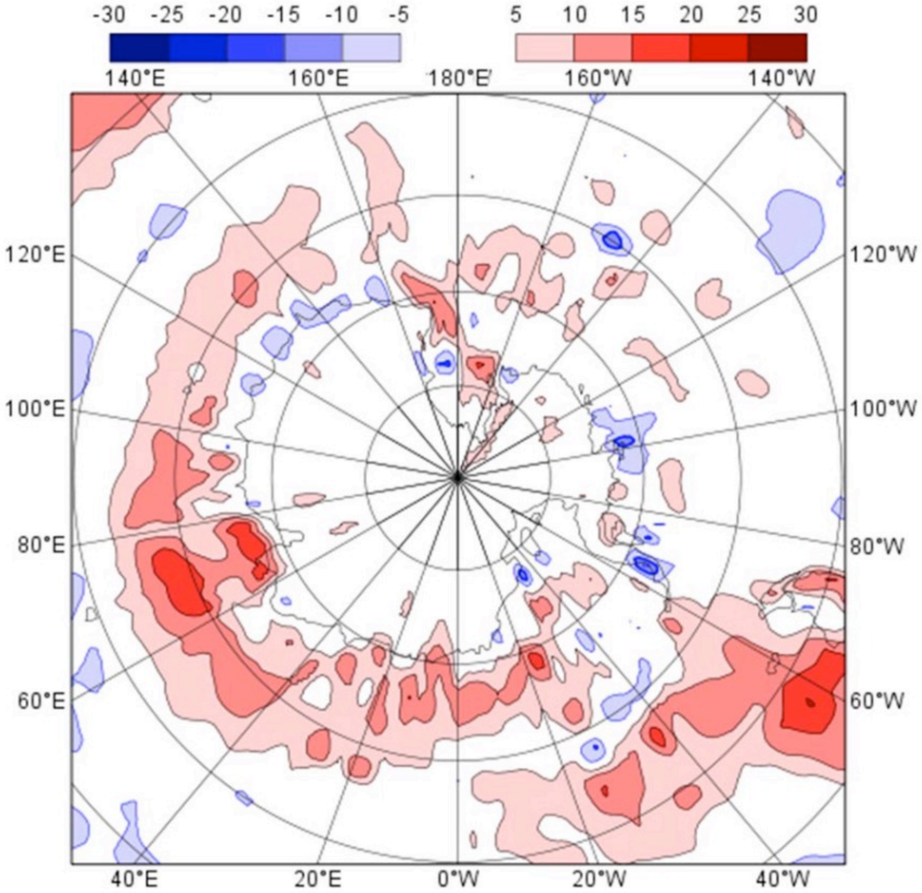




Importance of supercooled liquid water clouds: reduces shortwave radiative flux errors in models

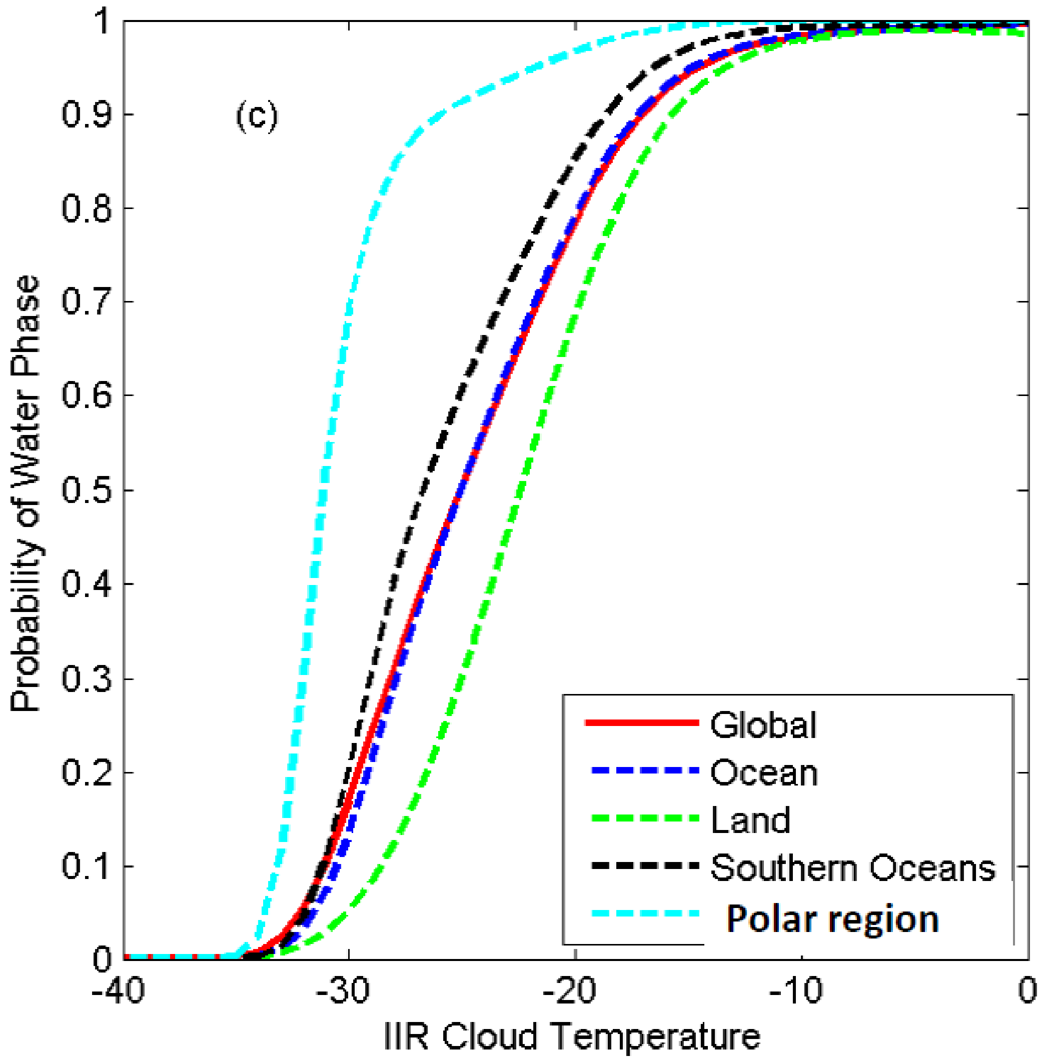


Without constraint of CALIPSO supercooled liquid water clouds



With CALIPSO supercooled liquid water clouds

Why no liquid droplet below -34 ^{0}C (instead of -40 ^{0}C) in clean (near ice-nuclei-free) southern oceans ?



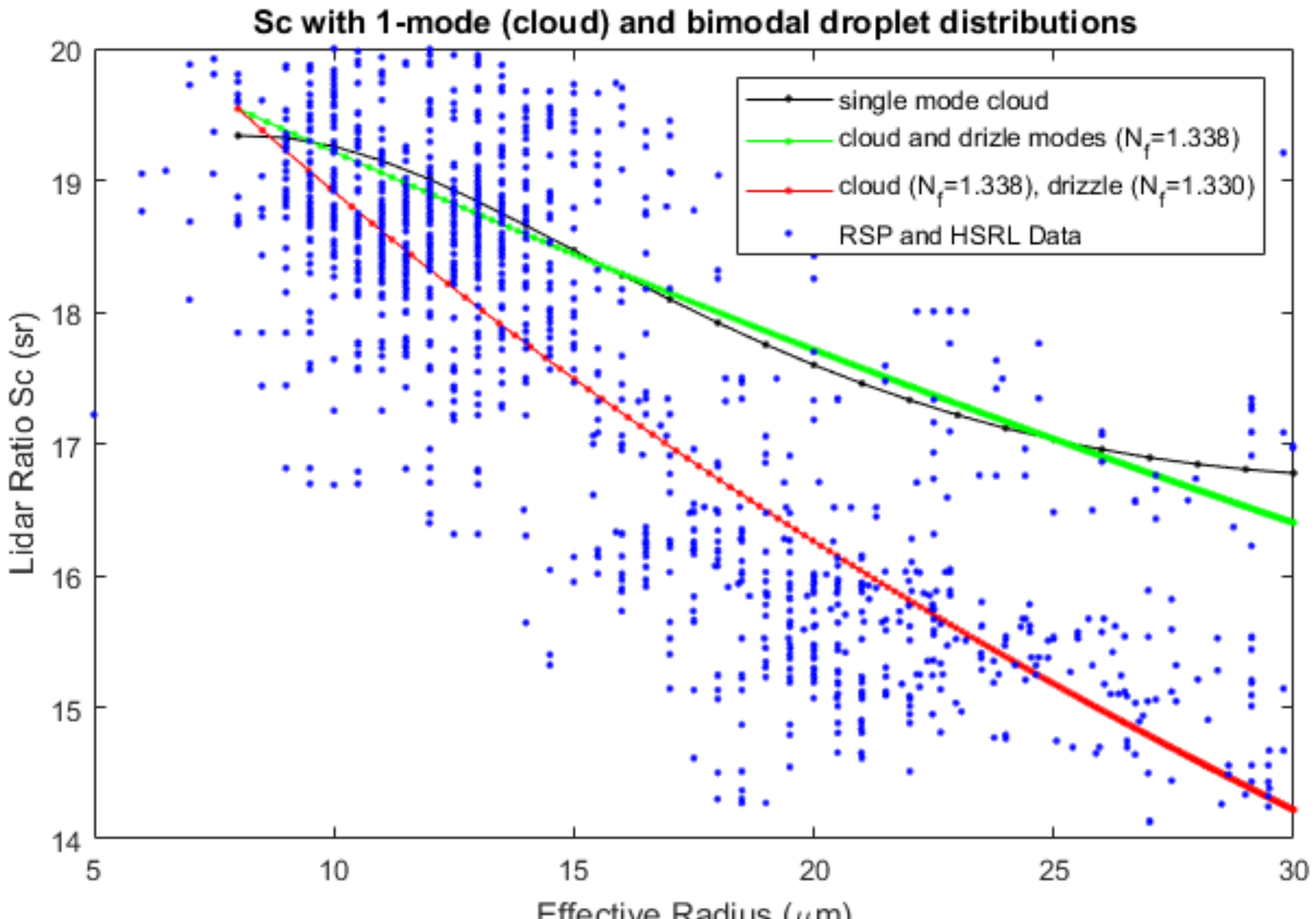
Another problem from lidar measurements of the water clouds:

lidar ratios (S_C) of water clouds from lidar measurements are lower than Mie scattering calculations

$$S_c = \frac{extinction\ cross\ section}{backscatter\ cross\ sction} = \frac{[(1 + depolarization)(1 - depolarization)]^2}{2 * layer\ integrated\ attenuated\ backscatter}$$

$S_C - R_e$ relation: theory vs aircraft measurements

1. *S_C is hyper sensitive to changes in refractive index (black and green line: Mie calculation with refractive index of water)*
2. *read line: Mie calculations with unrealistic refractive index (1.330)*

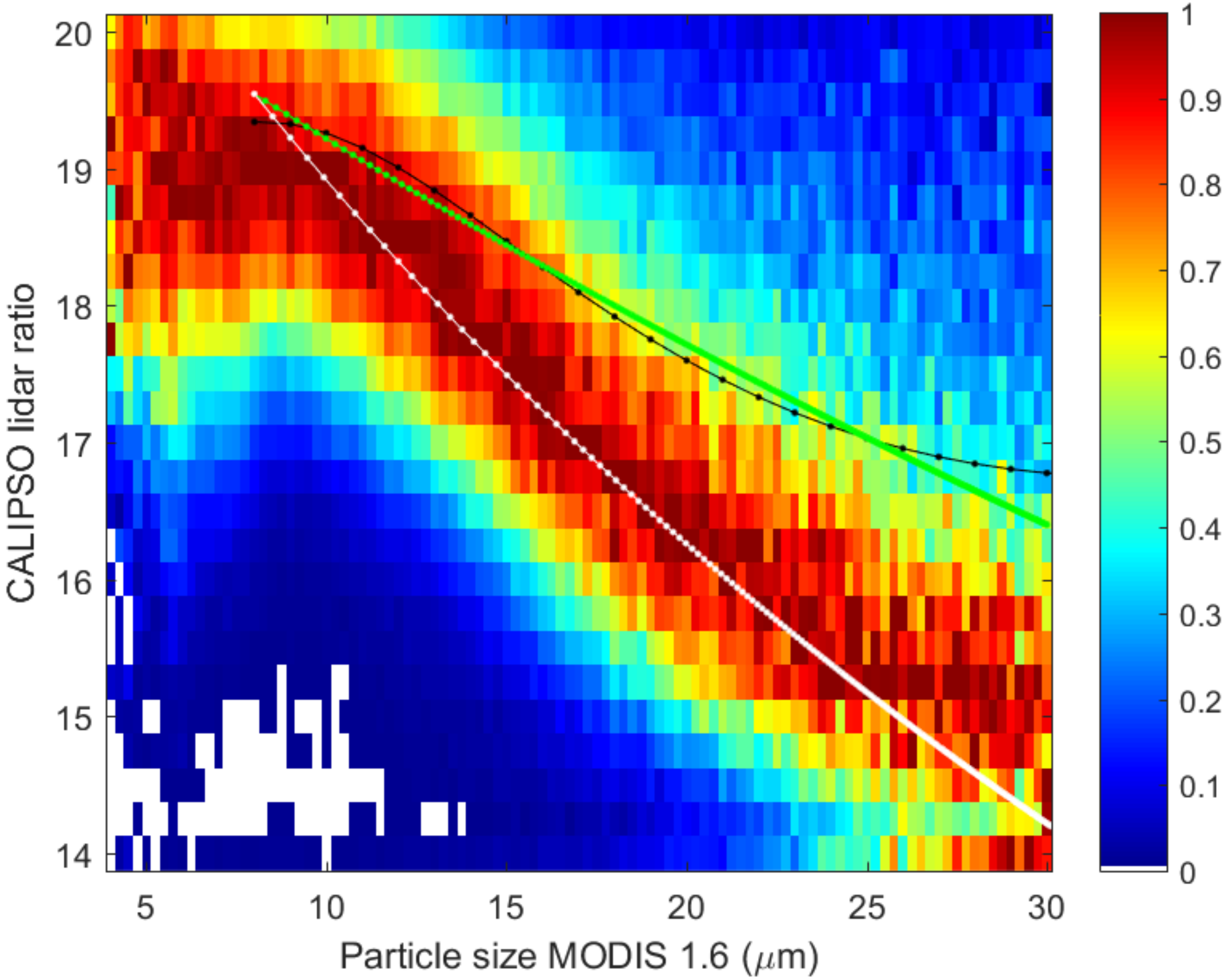


(Pengwang Zhai,2018)

lab measurements suggest refractive indice (N) of water at 532 nm somewhere between 1.334-1.340 (not 1.330 or less, which is needed to explain the lidar measurement of water clouds)

As refractive indice of water are between 1.334-1.340, theoretical lidar backscatter of water clouds are supposed to be weaker (lidar ratios larger) comparing with aircraft/satellite lidar observations

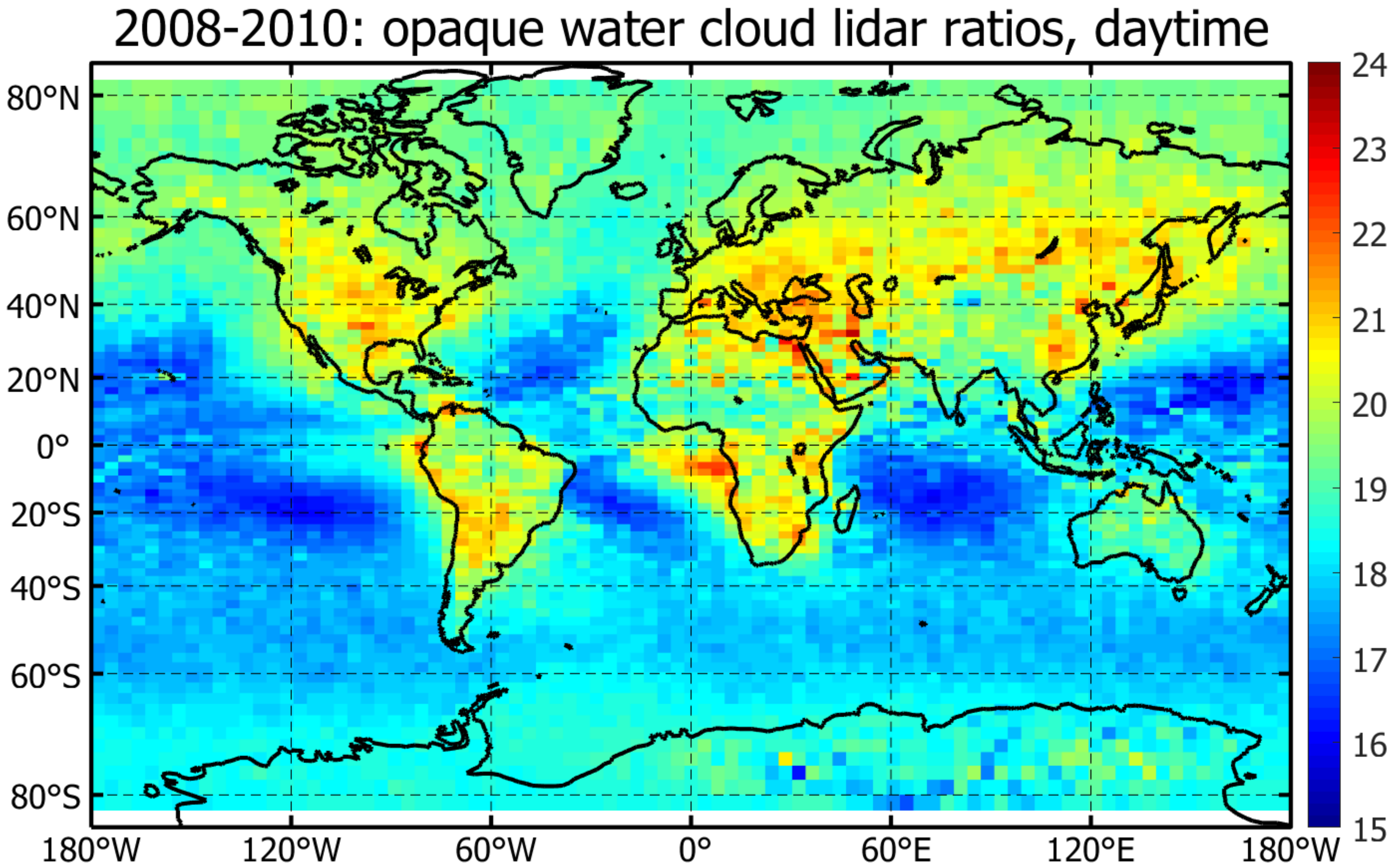
S_C – R_e relation: satellite measurements similar to aircraft measurements

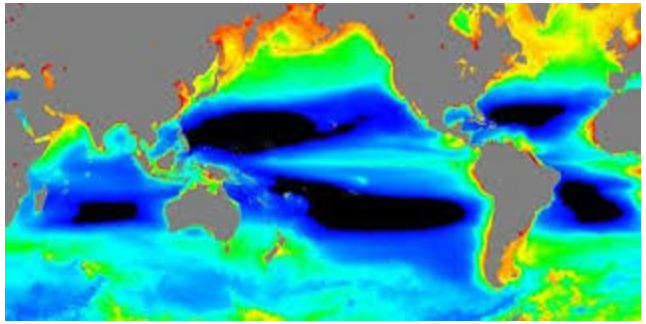


Green and black lines: Mie calculations with realistic refractive index

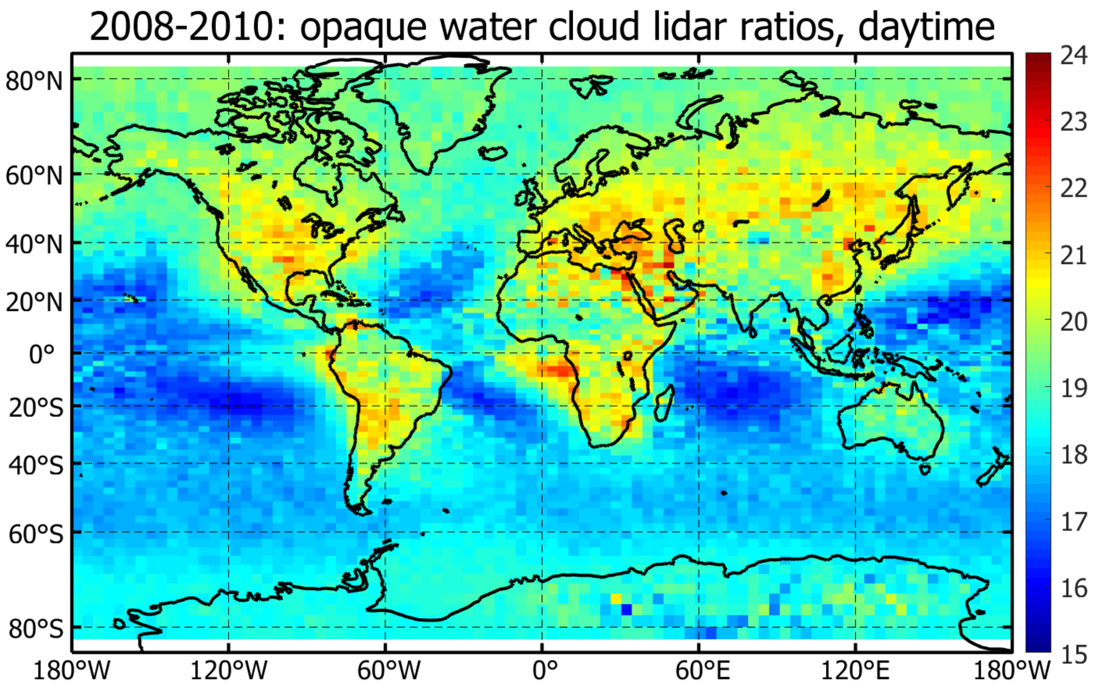
White line: drizzle mode with unrealistic refractive index

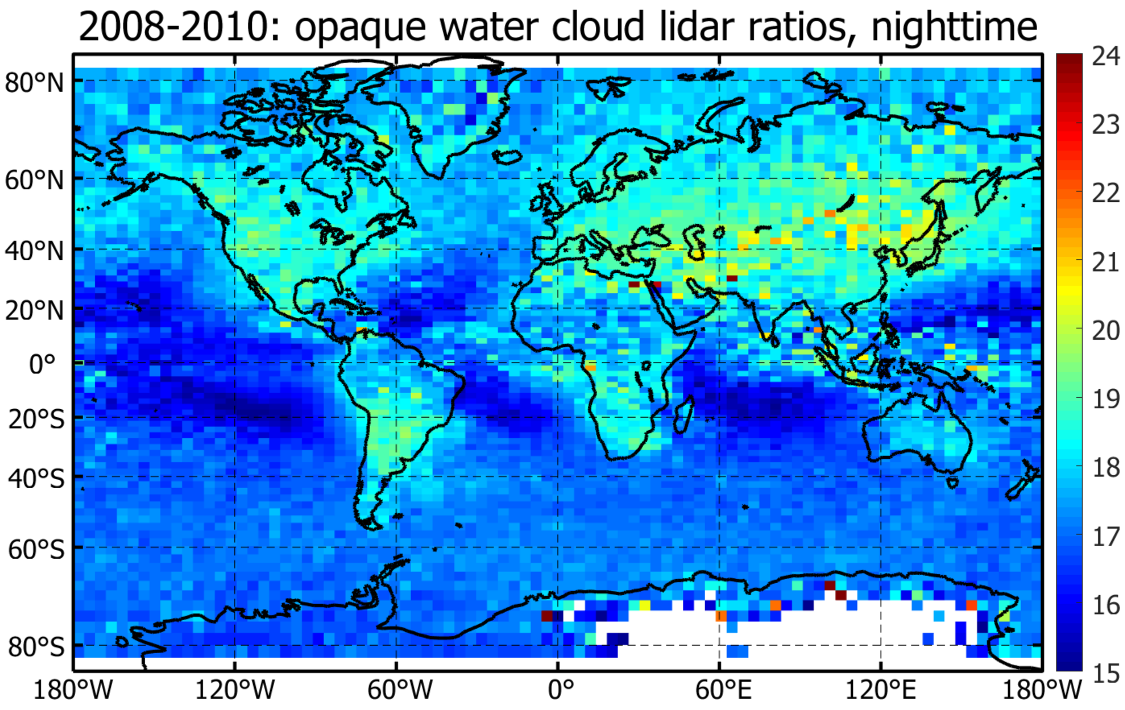
lidar ratios of water clouds derived from CALIPSO (small lidar ratio means stronger backscatter, or larger particles)





lidar ratios of water clouds at night is even smaller
(*stronger backscatter by water clouds at night*)





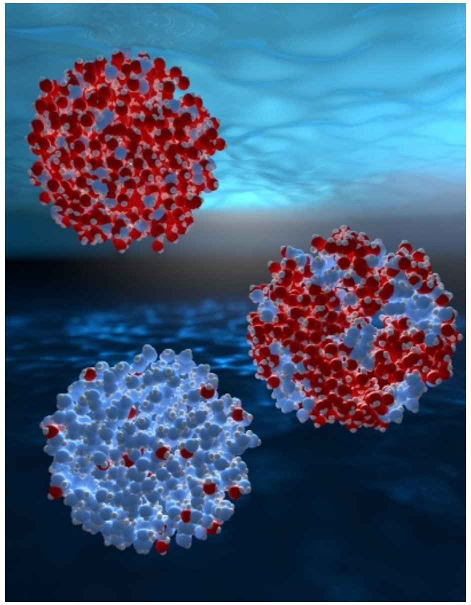
Hypothesis: meta-stable cloud droplets near cloud top with different refractive index (high density state with high backscatter)

States of water:

- liquid (warmer than 0 ^{0}C)
- Ice (colder than 0 ^{0}C)
- Meta-stable states:

  Supercool liquid water (between -40C??? and 0C)

  amorphous water (transient, below -45 ^{0}C and can stay for long time ‘ (more stable) at T < -80 0 ^{0}C)



WATER THERMODYNAMICS

Maxima in the thermodynamic response and correlation functions of deeply supercooled water

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Femtosecond x-ray laser pulses were used to probe micrometer-sized water droplets that were cooled down to 227 kelvin in vacuum. Isothermal compressibility and correlation length were extracted from x-ray scattering at the low–momentum transfer region. The temperature dependence of these thermodynamic response and correlation functions shows maxima at 229 kelvin for water and 233 kelvin for heavy water. In addition, we observed that the liquids undergo the fastest growth of tetrahedral structures at similar temperatures. These observations point to the existence of a Widom line, defined as the locus of maximum correlation length emanating from a critical point at positive pressures in the deeply supercooled regime. The difference in the maximum value of the isothermal compressibility between the two isotopes shows the importance of nuclear quantum effects.

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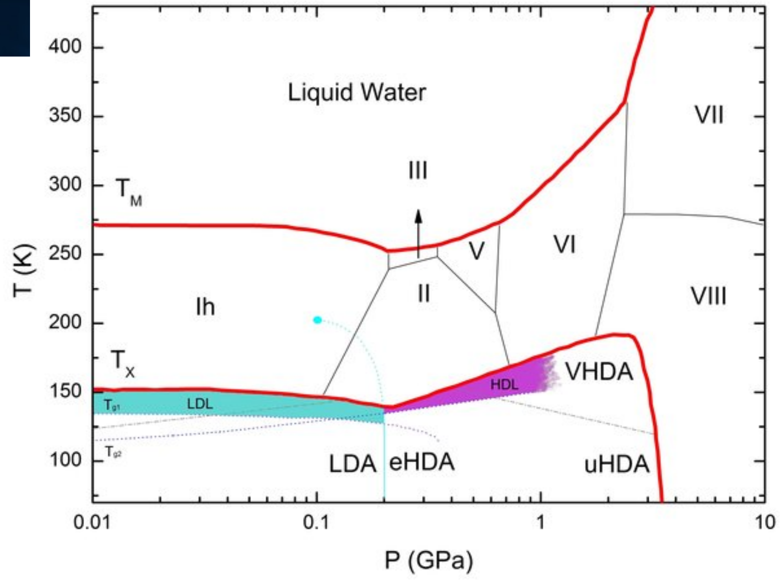

Physics behind formation of meta-stable state: rapid radiative cooling of water cloud droplets at cloud top

- Outgoing longwave radiation at **window region (8 μm to 12 μm)** cools cold cloud droplets at cloud top (the first 1-2 optical depth from cloud top) at a huge cooling rate of about 1 ^{0}C/second
  1. at **atmospheric window region** (8 μm to 12 μm) at which **molecular atmosphere do not absorb/emit**, net longwave window radiative flux divergence of cold water clouds: ~80 W/m^2
  2. this **cooling happens to water droplets** at cloud top with liquid water path around 10 g/m^2
  3. **radiative cooling rate of the droplets: ~ 2 K/second** (80/10/4.18, heat capacity 4.18 J/K/g)
- cooling rate will have to be balanced by latent heat releasing due to droplet condensational growth and (to a small degree) heat diffusion between the droplet and the air surrounding it (which requires **cloud water content doubles every 5 minutes** (2260/4.18/2/60)
- unfortunately, condensation growth rate is much slower (**latent heat heating due to condensation is less than 5% of the radiative cooling in longwave window**) than what is required to balance (Barkstrom, 1978, JAS) radiative cooling of the droplets,
- Latent heat of condensation cannot keep up with the longwave window cooling of the droplets, **water droplets at cloud top keep going colder** (and thus may **get cold enough to transition to meta-stable condition**)

Some Effects of 8–12 μm Radiant Energy Transfer on the Mass and Heat Budgets of Cloud Droplets

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ABSTRACT

In standard treatments of the mass and energy budget of cloud droplets, radiant energy transfer is neglected on the grounds that the temperature difference between the droplet and its surroundings is small. This paper includes the effect of radiant heating and cooling of droplets by using the Eddington approximation for the solution of the radiative transfer equation. Although the calculation assumes that the cloud is isothermal and has a constant size spectrum with altitude, the heating or cooling of droplets by radiation changes the growth rate of the droplets very significantly. At the top of a cloud with a base at 2500 m and a top at 3000 m, a droplet will grow from 9.5 to 10.5 μm in about 4 min, assuming a supersaturation ratio of 1.0013. Such a growth rate is more than 20 times the growth rate for condensation alone, and may be expected to have a significant impact on estimates of precipitation formation, as well as droplet spectrum calculations.

Previous studies found amorphous water droplets in mesosphere

Homogeneous nucleation of amorphous solid water particles in the upper mesosphere

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A B S T R A C T

Condensed water particles are known to exist in the high latitude upper mesosphere during the summer months. However, the mechanism or mechanisms through which they nucleate remains uncertain. It is postulated here that particles of amorphous solid water (ASW, condensed water with a non-crystalline structure) may nucleate homogeneously in the summer mesosphere. Using classical nucleation theory and a one-dimensional model, it is shown that more than $10^5\ cm^{-3}$ amorphous solid water particles can nucleate homogeneously under mesopause conditions. Furthermore, it is shown that homogeneous nucleation competes with heterogeneous nucleation on meteoric smoke particles when the cooling rate is >0.5 K/h. The homogeneous nucleation of amorphous solid water could provide an explanation for the high density of ice particles (many thousands per cm^3) thought to be required for electron depletions in the upper mesosphere. A parameterisation for homogeneous nucleation is presented which can be used in other mesospheric cloud models.

Previous studies with in situ measurements found amorphous water droplets in troposphere atmosphere

SOME PROPERTIES OF METASTABLE STATES OF WATER

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A number of new conclusions on properties of H_2O modifications metastable with respect to the transition into crystalline ice at $T < 0°C$ were obtained. Such modifications are supercooled ordinary water (water-1) and amorphous water (A-water). This study was initiated by new data on the microphysical structure of atmospheric cold clouds (CCc). Based on the new and previously known experimental data, the concepts on the nature and properties of water amorphous condensate were corrected and complemented. It was substantiated that the optical glory phenomenon on CCs is formed as a bow of sunlight scattering by A-water droplets with a refractive index of ~1.8. The molecular mechanism of frontal crystallization of the metastable form of water, which explains the observed effects of water freezing, was considered.

Not all agree with Nevzorov

Comment on "Glory phenomenon informs of presence and phase state of liquid water in cold clouds" by Anatoly N. Nevzorov

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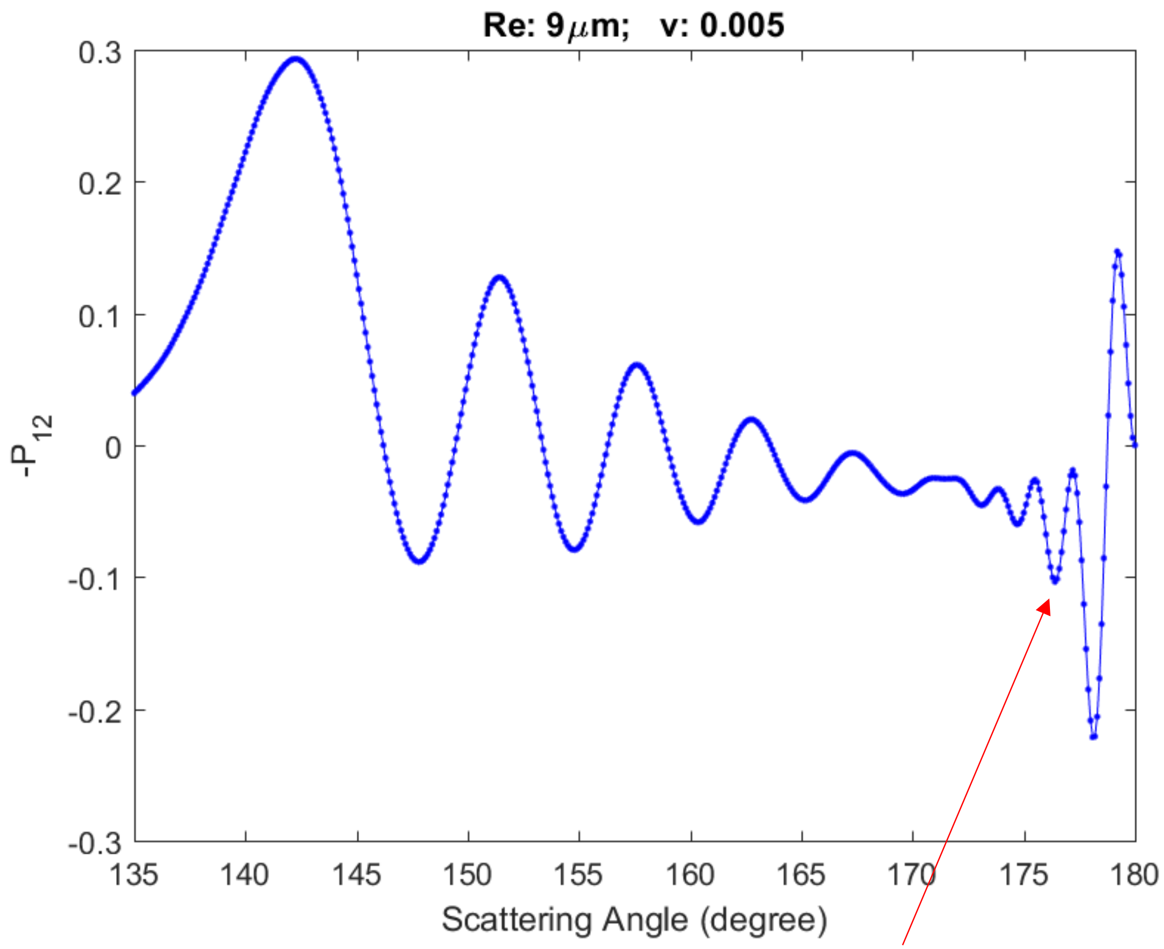
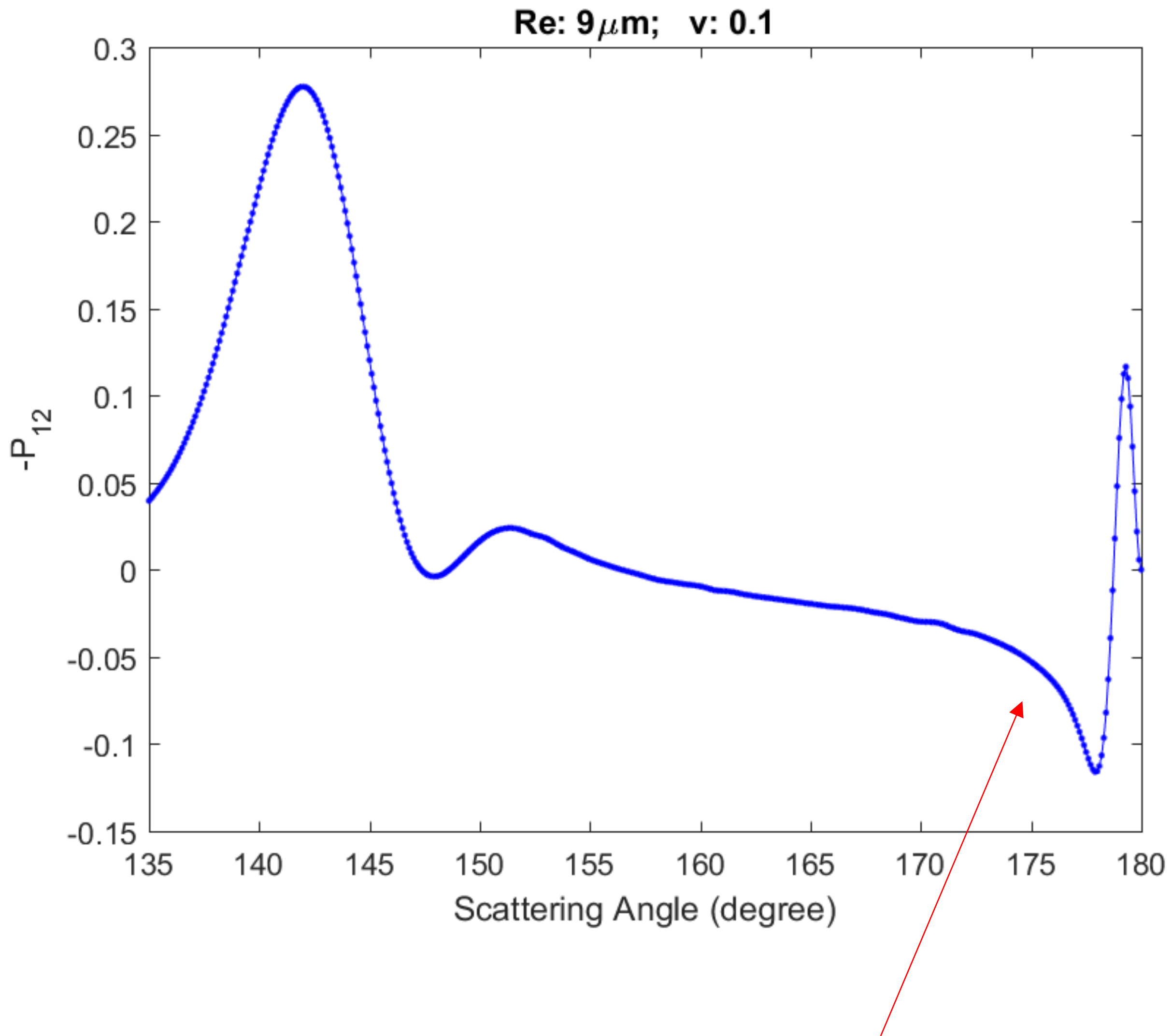
Abstract

In a recent publication "Glory phenomenon informs of presence and phase state of liquid water in cold clouds" Nevzorov [Nevzorov, A., 2006. Glory phenomenon informs of presence and phase state of liquid water in cold clouds. Atmospheric Research 82, 367–378] claims that "the convincing evidence has been provided that this sort of glory forms as a first-order bow from spherical particles with a refractive index of 1.81–1.82 and diameter over 20 μm". This is a highly unusual finding because the refractive index of liquid water and ice is between 1.30 and 1.35 in the visible spectral range. The author concludes that "once more corroboration is gained […] of droplets of liquid water in specific phase state referred to amorphous water, or A-water". Here we show that the phenomena described by the author are easily explained assuming liquid water with a refractive index of 1.33 and a realistic droplet size distribution with an effective radius of around 10 μm. We conclude that this type of observations does not corroborate the existence of amorphous water in the atmosphere. In a recent publication we showed how to quantitatively derive cloud optical thickness, effective droplet radius, and even the width of the size distribution from observations of the glory [Mayer, B., Schröder, M., Preusker, R., Schüller, L., 2004. Remote sensing of water cloud droplet size distributions using the backscatter glory: a case study. Atmospheric Chemistry and Physics 4, 1255–1263].

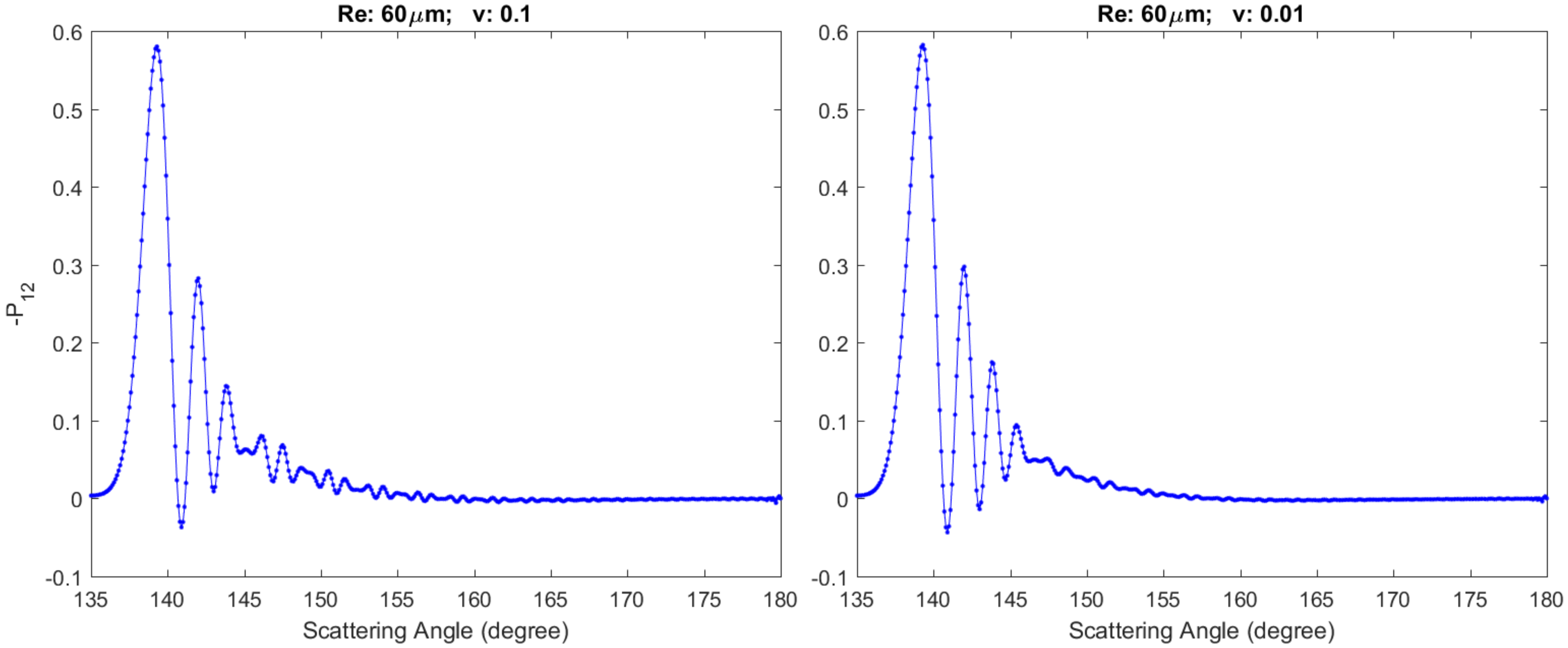
POLDER measurements of polarized radiances of glory from cold clouds are studied here in order to investigate the existence of meta-stable water droplets

Droplets in cloud mode: $-P_{12}$ of glory < 0

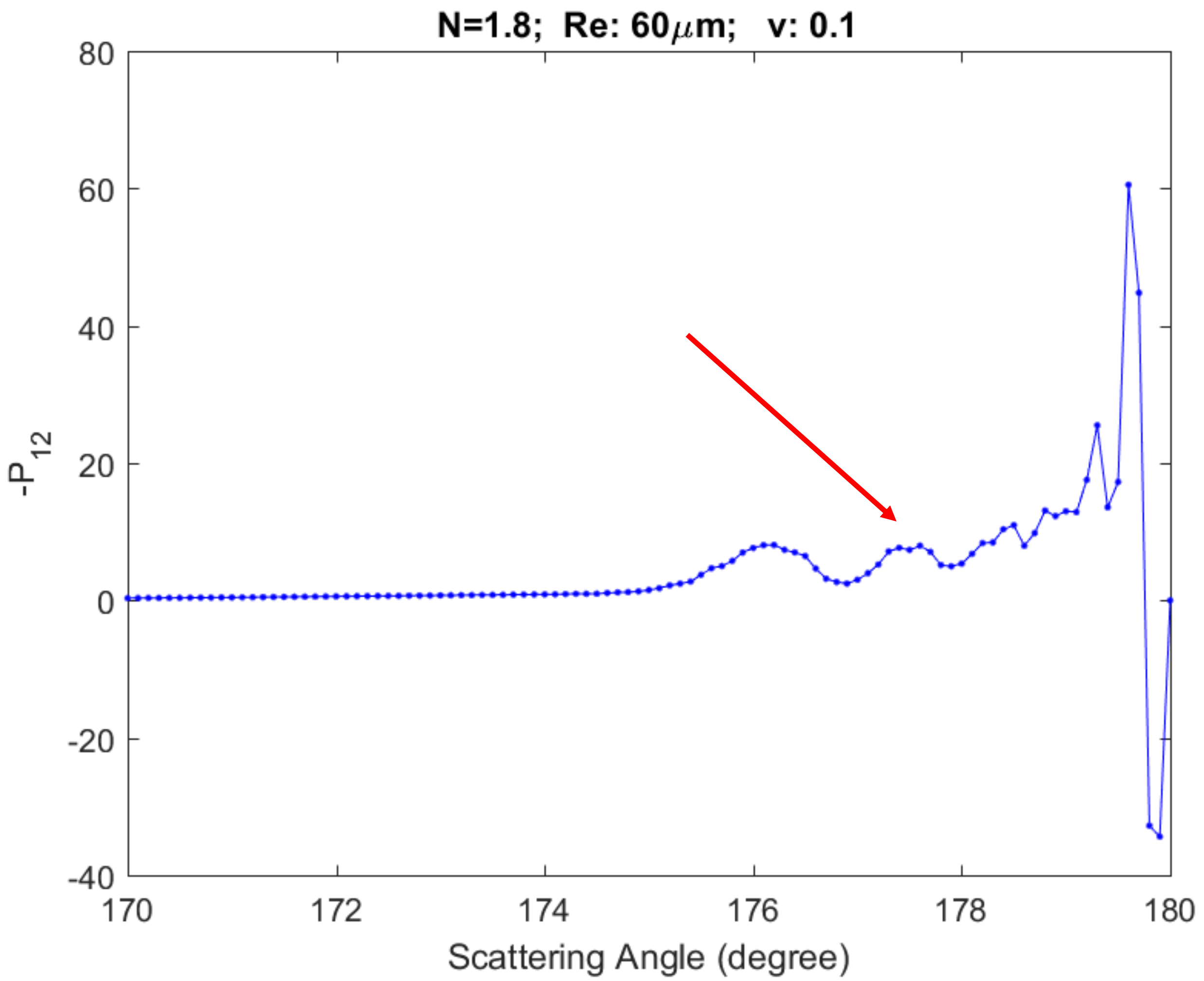


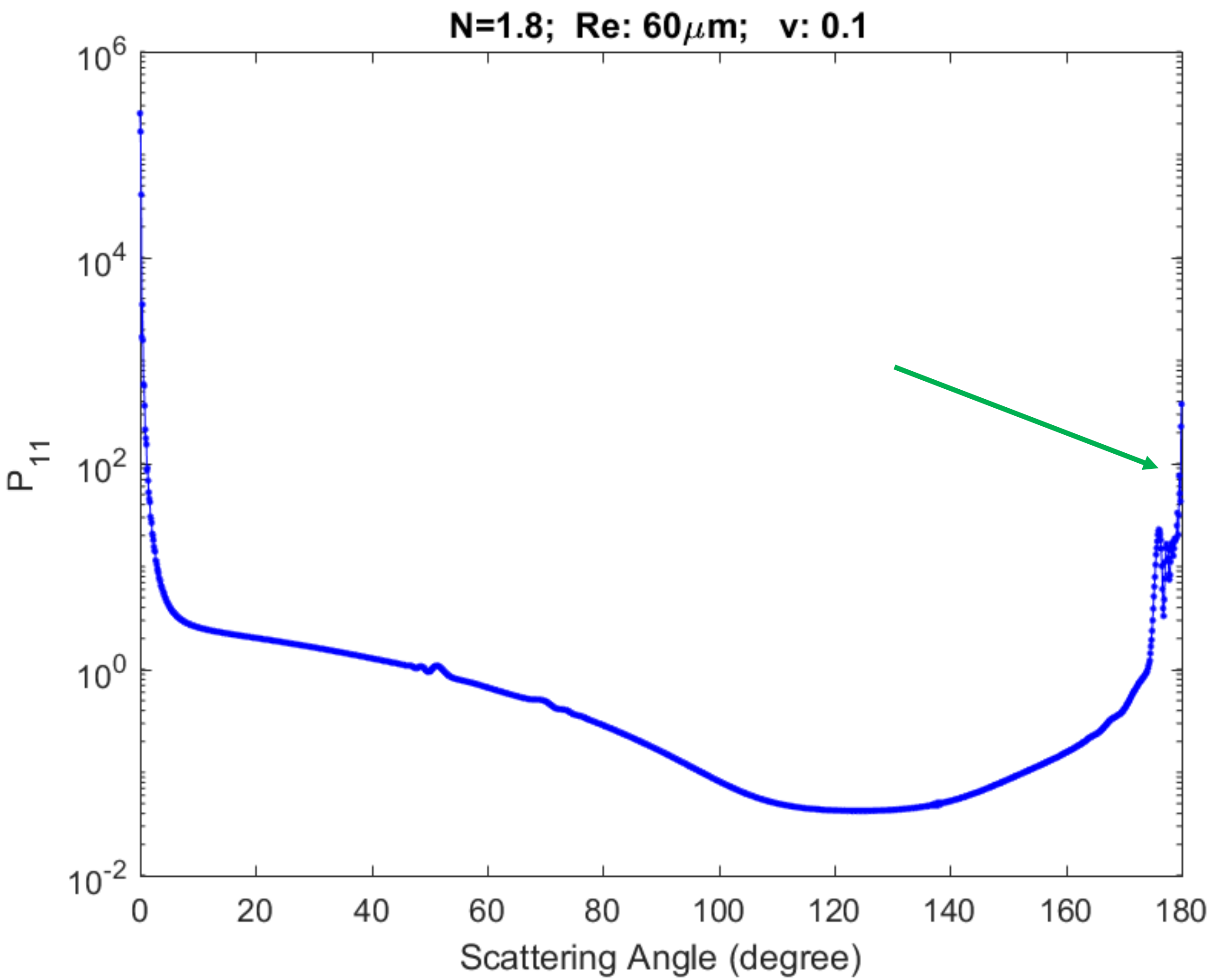


No Glory for drizzle mode droplets

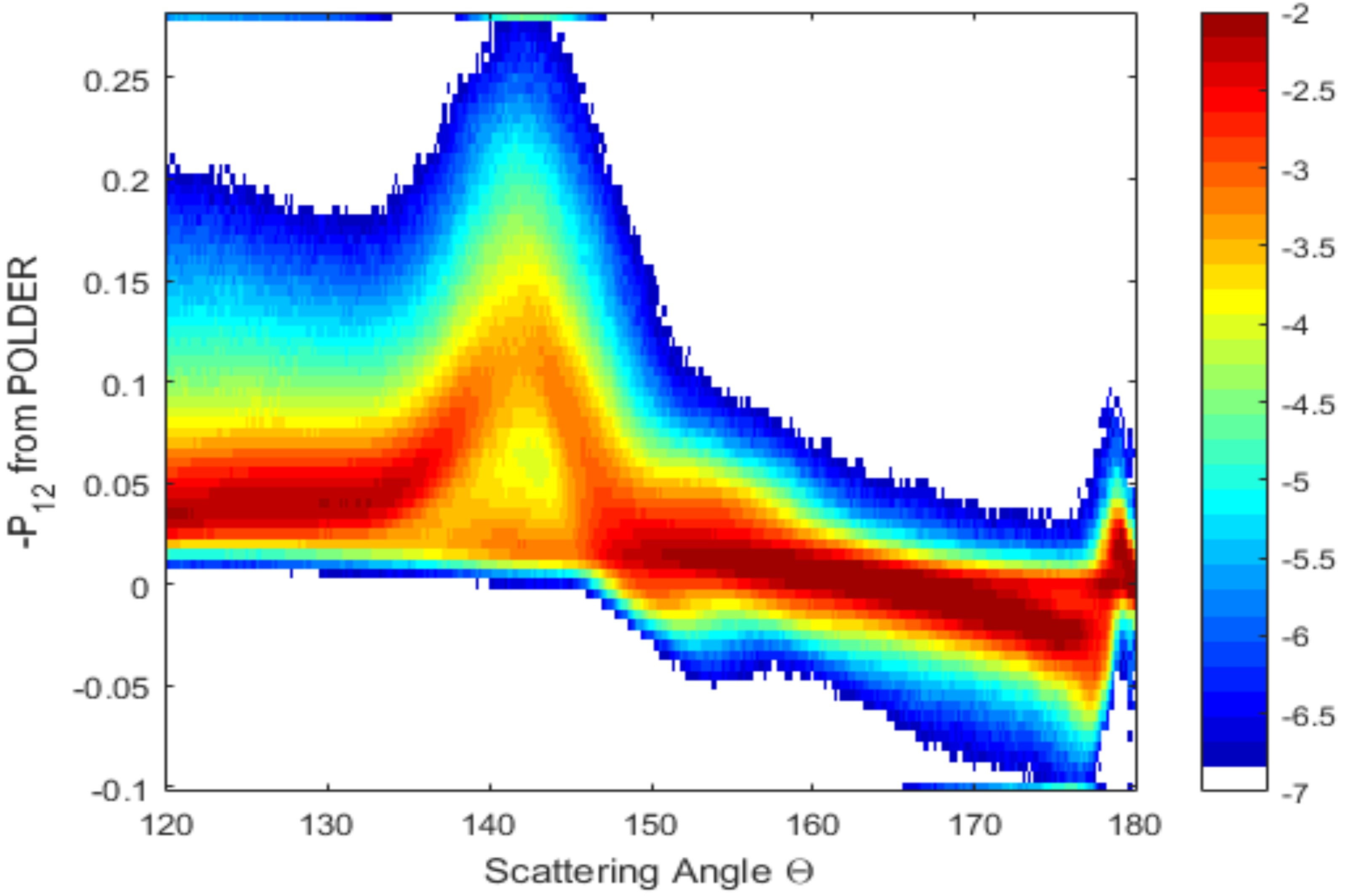


Strong glory for drizzle in high density metastable water droplets: $-P_{12}$ of glory > 0, very strong backscatter $P_{11}(180^0)$

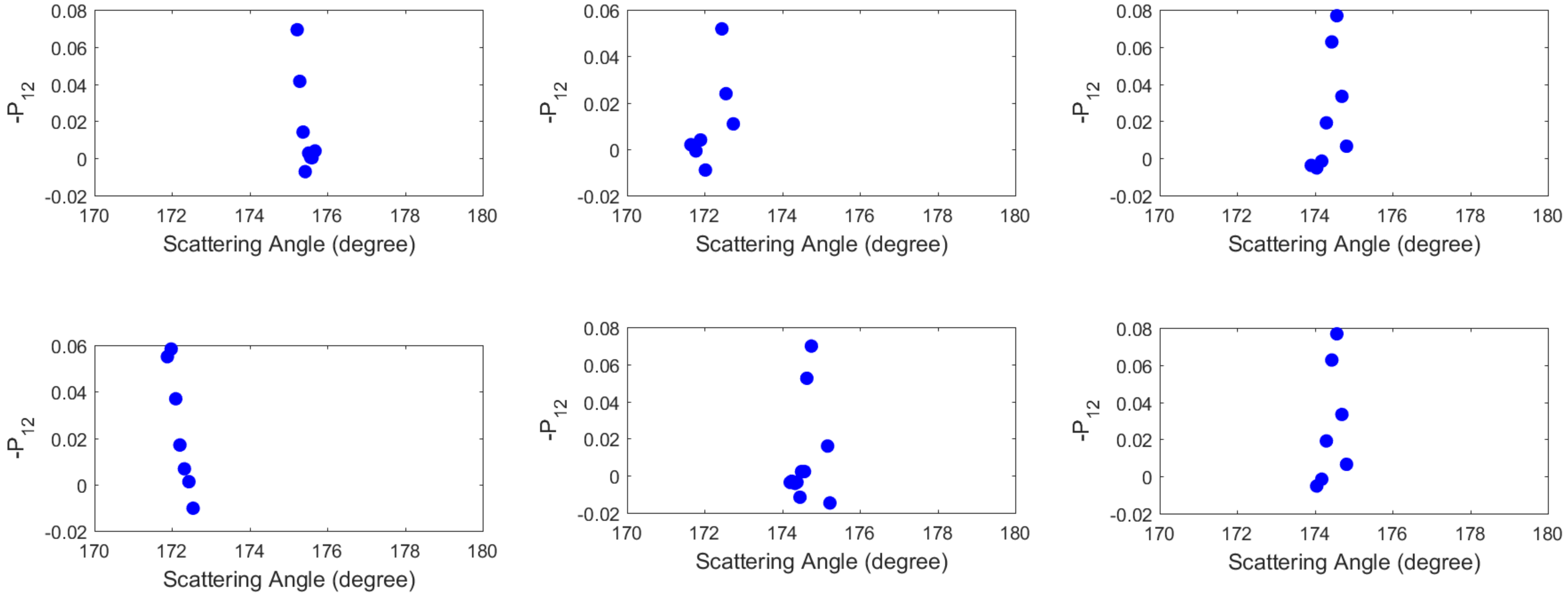




$-P_{12}(\Theta)$ estimated from cold clouds derived from POLDER measurements



There are a VERY small fraction of cloud observations with P_{12} features similar to amorphous clouds ($-P_{12} > 0$)



Possible reasons why so few data points: (1) the fraction of particles in meta-stable state is very small; (2) the particles do not exists long before crystalizing and falling; (3) measurement uncertainties ?

Summary

- Lidar measurements of water clouds cannot be explained well by theory
- Hypothesis: cloud droplets at cloud top can be in a meta-stable state with high density due to radiative cooling, which enhances lidar backscatter
- In general, polarized radiance measurements of cold clouds agree with Mie scattering calculations of typical liquid water cloud droplets (meta-stable high density cloud droplet type of feature not seen very often)
- There are a small fraction of cases where polarized radiance measurements of cold cloud glory differ from Mie scattering calculations of typical liquid water cloud droplets. It is likely that there is only a small fraction of the particles are in high density state because these particles exist in very short time period before it crystalizes/grows/falls from cloud top
- If confirmed, the meta-stable cloud droplets can play a role to ice nucleation for cold rain process
- Combined lidar (cloud identification, cloud phase and cloud backscatter) and POLCUBE-1 (optimized for polarization measurements of Glory angles) measurements can help