Evidence of high ice supersaturation in cirrus clouds using ARM Raman lidar measurements

Jennifer M. Comstock, Thomas P. Ackerman, and David D. Turner
Pacific Northwest National Laboratory, USA

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[1] Water vapor amounts in the upper troposphere are crucial to understanding the radiative feedback of cirrus clouds on the Earth’s climate. We use a unique, year-long dataset of water vapor mixing ratio inferred from ground-based Raman lidar measurements to study the role of ice supersaturation in ice nucleation processes. We find that ice supersaturation occurs 31% of the time in over 300,000 data points. We also examine the distribution of ice supersaturation with height and find that in the uppermost portion of a cloud layer, the air is ice supersaturated 43% of the time. These measurements show that large ice supersaturation is common in cirrus clouds, which supports the theory of ice forming homogeneously. Given the continuous nature of these Raman lidar measurements, our results have important implications for studying ice nucleation processes using cloud microphysical models. INDEX TERMS: 0320 Atmospheric Composition and Structure: Cloud physics and chemistry; 3360 Meteorology and Atmospheric Dynamics: Remote sensing; 3399 Meteorology and Atmospheric Dynamics: General or miscellaneous. Citation: Comstock, J. M., T. P. Ackerman, and D. D. Turner (2004), Evidence of high ice supersaturation in cirrus clouds using ARM Raman lidar measurements, Geophys. Res. Lett., 31, L11106, doi:10.1029/2004GL019705.

1. Introduction

[2] Water vapor is a crucial component of the earth-atmosphere system because it is a greenhouse gas and plays an essential role in radiative feedback mechanisms and cloud formation. Accurate measurements of upper tropospheric (UT) water vapor are important for two reasons. First, small changes in UT moisture significantly impact the outgoing longwave radiation, which controls the planetary radiative energy loss. Second, water vapor measurements in the UT are critically important in determining the role of heterogeneous and homogeneous nucleation in cirrus cloud formation. Large values of ice supersaturation (SS) are associated with homogeneous nucleation, while heterogeneous nucleation of insoluble particles can occur at smaller values [DeMott et al., 1997].

[3] Upper tropospheric water vapor concentrations are difficult to measure, particularly on a continuous basis. Standard uncorrected radiosonde water vapor measurements are typically unreliable at cold temperatures (\(<\sim 40^\circ\text{C}\)) [Miloshevich et al., 2001]. Previous aircraft based studies have shown that large ice SS exists in the UT [Gierens et al., 2000; Jensen et al., 2001], but these experiments are typically limited in duration and spatial extent. The Measurement of Ozone and Water Vapor by Airbus in-service Aircraft (MOZAIC) project measured UT water vapor over a 3 year period and found that the air sampled was ice SS \(\sim 13.5\%\) of the time [Gierens et al., 1999]. During the MOZAIC study, aircraft were not equipped with cloud sensors, but commercial aircraft typically avoid clouds when possible. In contrast to aircraft and radiosonde observations, geostationary satellite measurements provide a unique method for tracking the global distribution of water vapor [Soden, 1998], but lack the resolution required to study cirrus nucleation processes. Here, we present for the first time an extensive dataset of continuous UT water vapor profile measurements at a single location, which can be used to study the link between water vapor, cirrus formation mechanisms, and their potential climatic impacts.

2. Observations

[4] We analyzed 1 year of Raman lidar (RL) measurements obtained in 2000 by the Department of Energy’s Atmospheric Radiation Measurement Program [ARM; Ackerman and Stokes, 2003] at the Southern Great Plains (SGP) facility near Lamont, OK (36°37’N, 97°30’W). The ARM RL [Goldsmith et al., 1998] transmits a laser pulse at 355 nm, which undergoes elastic scattering due to clouds and aerosols. The ARM RL also detects inelastic scattered photons produced by the rotational-vibrational Raman effect of nitrogen (387 nm) and water vapor (408 nm) molecules. The water vapor mixing ratio is proportional to the ratio of the water vapor and nitrogen signals, which are measured simultaneously. The RL mixing ratio data are calibrated to agree with co-located microwave radiometer column water vapor measurements [Turner and Goldsmith, 1999]. The RL also measures the co-polarized and cross-polarized returns at 355 nm. The ratio of these two signals, the linear depolarization ratio (LDR), is useful for distinguishing ice and water in optically thin clouds. The advantage of using the RL for measuring UT water vapor is that it provides continuous, high resolution water vapor profiles with an accuracy of 5% [Ferrare et al., 2004], and simultaneously detects the presence of clouds. This allows us to compile a climatologically significant sample of midlatitude UT water vapor concentrations.

[5] The solar background energy increases the noise in the water vapor channel during daytime hours, which limits the maximum profile height to \(\sim 3\text{km}\). Therefore, we limit our dataset to night observations when profiles can extend to 12 km. The horizontal resolution in time is 10 minutes and the vertical resolution varies between 78 m below 5 km to over 400 m in the UT. This averaging is required to...
increase the signal-to-noise ratio and reduce the random error in the water vapor measurement. We also degrade the depolarization resolution to match the water vapor measurements. If the random error is >20%, which often occurs near cloud top, the measurement is not included. This reduces the number of valid data points in the uppermost portion of the cloud where temperatures are coldest and ice nucleation processes occur. Since the RL is unable to penetrate through optically thick clouds, we are limited to times when the UT is unobstructed by low opaque clouds. In addition to these data quality issues, there are two months (January and July) for which lidar measurements are not available. Despite these limitations, our dataset is comprised of nearly 300,000 data points (~9500 profiles) over the 1 year period. To ensure that we include only ice clouds, we require that the LDR > 20% and only consider clouds that are located above 7 km.

[6] Using profiles of water vapor mixing ratio and temperature, we estimate relative humidity with respect to ice [RHI; Goff, 1965] over the ARM SGP site. We use continuous thermodynamic profiles (1 minute by 250 m resolution), which are a combination of radiosonde data and Rapid Update Cycle (RUC) model output [Benjamin et al., 1994]. We supplement RUC temperature profiles with data from ARM radiosonde launches that occur four times per day. Since UT temperature does not typically experience rapid changes over a 6 hour period, we estimate the accuracy to be approximately ±1°C, which gives a RHI variance of ~8.0% at −40°C and ~10.0% at −60°C. Since most of our water vapor measurements coincide with temperatures warmer than −50°C, we feel the temperature accuracy is reasonable for this analysis.

3. Results

[7] To examine the relationship between temperature and ice SS, we plot frequency of occurrence of RHI as a function of temperature (Figure 1). Our results indicate that

Figure 1. Frequency of occurrence of RHI for 10°C temperature divisions. Results are shown for cloudy (dark line) and cloud free sky conditions (light line). The number of points used in each figure is listed in the upper right corner.

SS (RHI > 100%) occurs in ~31% of cloudy data points. There are a small number of cases (~9%) where cloud free air is ice SS, which could occur if insufficient numbers of cloud condensation nuclei (CCN) or ice nuclei are present, or the airmass has not reached the threshold for homogeneous nucleation, which can exceed 140%. The frequency distributions in Figure 1 do not vary significantly with temperature; however the percentage of ice SS cases decreases from 43% between −60 to −70°C to 31% between −30 to −40°C.

[8] Previous studies [Jensen et al., 2001] report 49% of cloudy points were SS during the SUCCESS aircraft field campaign, which took place in the same geographic region as the ARM SGP site. Although a significant fraction of cloudy points in Figure 1 have high SS, the mode frequency is below ice saturation for all temperature regions. This result is consistent with RHI measurements from the INCA (INterhemispheric difference in Cirrus properties from Anthropogenic emissions) field campaign, which sampled cirrus clouds near Prestwick, Scotland (55°N latitude) and Punta Arenas, Chile (55°S latitude) [Ovarlez et al., 2002]. The INCA campaign found that 31% and 51% of in-cirrus measurements near Prestwick and Punta Arenas, respectively, were ice SS. These differences could be a function of meteorological conditions, cloud types sampled, and the preferred ice crystal nucleation process in cirrus formation.

[9] Aircraft in situ measurements have helped to quantitatively define the structure for typical ice generating cirrus clouds [Heymsfield, 1975; Heymsfield and Miloshevich, 1995]. Aircraft microphysical probes sampled an ice generating cirrus cloud that was up to 20% ice SS in the upper 2/3 of the cloud and below ice saturation in the region near cloud base. Under these conditions, ice crystal number concentrations were typically largest in the upper regions of the cloud and smallest near cloud base where aggregation of ice crystals was observed. Using these aircraft measurements, they formulated the structure of cirrus clouds that places an ice generating region near cloud top, an ice crystal growth or deposition region in the middle of the cloud, and sedimentation or sublimation region near the cloud base.

[10] We use the RL dataset to investigate the vertical variability of RHI within a cloud layer by dividing single layer clouds with a depth >1 km into 3 regions based on total cloud depth, where the regions are defined as the lower 25%, middle 50%, and upper 25% (Figure 2). By doing so, we can examine RHI in the 3 typical regions found in ice crystal
generating cirrus clouds. In the upper 75% of the cloud, the frequency distribution of RHI is relatively similar, with 43% of cloudy points SS in the uppermost layer where ice crystal nucleation is most likely to occur, and 34% in the middle layer or ice crystal growth region. The lowest cloud region (sublimation zone) has a noticeably different distribution and 84% of cases are below ice saturation. These results are consistent with explicit microphysical model simulations of cirrus clouds [Khvorostyanov and Sassen, 1998], which predict that maximum RHI values and increased ice SS production are located in the ice crystal growth region where updraft velocities are typically strongest.

[11] To illustrate the different phases of a typical cirrus lifecycle observed at the ARM SGP site, we examine RHI, LDR, and radar reflectivity on 15 November 2000. A 9 hour segment of RL measurements reveals UT RHI ranges from 60–140% (Figure 3c). The LDR (Figure 3b) clearly shows where the RL detects ice crystals. The cloud optical depth (not shown) varies between 0.01 and 1.5. This case demonstrates several different phases of cirrus cloud evolution. The first stages of ice crystal formation are evident between 0200 and 0400 UTC (Figure 3b). Since ice crystals are small during this phase, they are below the detection threshold of radar measurements (Figure 3a). Later between 0400 and 0500 UTC, RHI is still above ice saturation and a distinct region of ice crystal formation and growth exists between 10 and 12 km. There does not appear to be significant sedimentation occurring at this time. According to combined radar reflectivity–infrared radiometer retrievals of column integrated microphysical properties [Mace et al., 1998], the ice crystal number concentration (Figure 3d) is relatively high (>100 L⁻¹) and particle effective radius (Figure 3e) is small (≈10 µm), which is typical for the ice generation region. RHI is >120% during this time period near cloud top. A short time later (0600 UTC), ice crystal fall streaks are visible in radar reflectivity measurements, indicative of ice sedimentation. RHI decreases from 120% near cloud top to ~70% near cloud base. Between 0800 and 0830 UTC the cloud is primarily composed of larger particles (50–80 µm) and is dominated by the ice sedimentation. Due to the large RHI and cold temperatures (~60°C) observed near cloud top, it is likely that homogeneous nucleation initiated the formation of ice crystals.

[12] Bulk microphysical cirrus and general circulation models typically assume that ice SS regions quickly deplete excess vapor in 20–30 minutes [Khvorostyanov and Sassen, 1998]. The 15 November case contains an extensive ice SS layer in the middle region of the cloud (~0800 UTC), suggesting that relaxation times are longer than 1 hr. Khvorostyanov and Sassen [1998] indicate that deposition of vapor into condensed ice can take several hours using their explicit cloud model. This result does not appear to be the typical scenario (Figure 2), but does appear to be happening on 15 Nov (Figure 3).

4. Discussion

[13] Our results have unique implications for studies of ice cloud formation and evolution. The data set spans an extended time period encompassing seasonal variability, a variety of dynamical situations, and various stages of cirrus evolution. Microphysical cirrus models require a RHI ~120% to initiate homogeneous nucleation in regions devoid of ice crystals [Sassen and Dodd, 1989]. Our results, as well as other studies [Jensen et al., 2001; Ferrare et al., 2000; Ovarlez et al., 2002], indicate that RHI >120% frequently occurs at temperatures warmer than −70°C. A recent study explains how nitric acid increases RHI in cold clouds (<−70°C) [Gao et al., 2004]. We estimate that only 2.5% of all ice clouds detected during the observation period occur at temperatures colder than −70°C. Therefore, the Gao et al. [2004] mechanism does not explain the majority of high RHI occurrence in cirrus clouds. One method to verify that these high RHI values are plausible is to compare the frequency of occurrence of RH with respect to water (RHw) vs. temperature (Figure 4) with the threshold conditions for homogeneous nucleation [Sassen and Dodd, 1989]. Only 6% of the RHw points lie above the nucleation threshold of vapor into condensed ice can take several hours using their explicit cloud model. This result does not appear to be the typical scenario (Figure 2), but does appear to be happening on 15 Nov (Figure 3).

Figure 3. Height vs. time display of 35 GHz radar reflectivity (a), LDR (b), and RHI (c) derived from ARM measurements on 15 November 2000 at the SGP site. Also shown are time series of mean layer number concentration (d) and effective radius (e).

Figure 4. Frequency of occurrence of RHw vs. temperature for cloudy points during 2000 at the SGP site. Contours are frequency in %, the dashed line represents the homogeneous nucleation threshold [Sassen and Dodd, 1989], and the dotted line is ice saturation.
threshold, while 14% and 21% of RHI are greater than 120% and 110%, respectively. Taking into account wind speed and temporal and spatial resolutions, we estimate the sample volume of the Raman lidar is $\sim 3.6 \times 10^6$ at cirrus altitudes, which is much larger than the volume of a typical nucleating cell. Therefore, our findings are consistent with the conditions required for homogeneous nucleation and high ice SS appears to occur frequently and at warmer temperatures than models suggest.

[14] Although our results are consistent with homogeneous nucleation, heterogeneous nucleation may also have a role in cirrus formation. Smoke, pollution, and commercial aircraft contribute to aerosol loading at the ARM SGP site (depending on synoptic conditions or season), which could influence the role of heterogeneous nucleation in the UT. However, without knowledge of ice nuclei, aerosol concentrations, and vertical velocity in the UT during this time period, the role of heterogeneous nucleation remains uncertain.

[15] The relative frequency of homogeneous vs. heterogeneous nucleation is important for determining the radiative feedback and climatic impact of cirrus clouds. Ice crystals generated by heterogeneous nucleation occur at lower RHI, in weaker updrafts and have smaller concentrations of larger particles as compared with homogeneous nucleation. The heterogeneous nucleation may also produce more frequent, widespread cirrus with smaller optical depth and lower net radiative forcing [DeMott et al., 1997]. Cloud scale vertical velocity is a crucial component to understanding the role of each nucleation process, but is difficult to measure. By combining this RHI dataset with retrieved microphysical properties and a detailed microphysical model, further insight into nucleation processes is possible, as demonstrated in the 15 November 2000 case. In addition, this dataset can provide a distinct link between measurement and modeling communities by providing accurate water vapor profiles in cirrus clouds, which is needed by modeling groups who are currently testing state-of-the-art cirrus cloud models [Lin et al., 2002]. This knowledge can only lead to an improved understanding of the effects of cirrus clouds on the Earth’s global energy budget and climate.

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T. P. Ackerman, J. M. Comstock, and D. D. Turner, Pacific Northwest National Laboratory, USA. (jennifer.comstock@pnl.gov)