- Arctic low-level boundary layer clouds: In-situ measurements and simulations of mono- and 1
- 2 bimodal supercooled droplet size distributions at the top layer of liquid phase clouds
- 3
- 4
- M. Klingebiel<sup>1</sup>, A. de Lozar<sup>2</sup>, S. Molleker<sup>3</sup>, R. Weigel<sup>1</sup>, A. Roth<sup>3</sup>, L. Schmidt<sup>4</sup>, J. Meyer<sup>5,6</sup>, A. 5
- Ehrlich<sup>7</sup>, R. Neuber<sup>4</sup>, M. Wendisch<sup>7</sup>, S. Borrmann<sup>1,3</sup> 6
- 7 [1] Institute for Physics of the Atmosphere, University of Mainz, Germany
- [2] Max Plank Institute for Meteorology, Hamburg, Germany 8
- 9 [3] Max Planck Institute for Chemistry, Mainz, Germany
- 10 [4] Alfred Wegener Institute for Polar and Marine Research, Potsdam, Germany
- 11 [5] Institut für Energie- und Klimaforschung (IEK-7), Forschungszentrum Jülich, Germany
- 12 [6] Institute of Energy & Environmental Technology e.V. (IUTA), Duisburg, Germany
- 13 [7] Leipzig Institute for Meteorology, University of Leipzig, Germany
- 14

#### 15 Abstract

- 16 Aircraft borne optical in-situ size distribution measurements were performed within Arctic boundary layer
- 17 clouds with a special emphasis on the cloud top layer during the **VER** tical **D** is tribution of **I** ce in Arctic
- Clouds (VERDI) campaign in April and May 2012. An instrumented Basler BT-67 research aircraft 18
- 19 operated out of Inuvik over the Mackenzie River delta and the Beaufort Sea in the Northwest Territories
- of Canada. Besides the cloud particle and hydrometeor size spectrometers the aircraft was equipped with 20
- instrumentation for aerosol, radiation and other parameters. Inside the cloud, droplet size distributions 21
- with monomodal shapes were observed for predominantly liquid-phase Arctic stratocumulus. With 22
- increasing altitude inside the cloud the droplet mean diameters grew from 10 µm to 20 µm. In the upper 23
- 24 transition zone (i.e. adjacent to the cloud-free air aloft) changes from monomodal to bimodal droplet size
- 25 distributions (Mode 1 with 20 µm and Mode 2 with 10 µm diameter) were observed. It is shown that
- 26 droplets of both modes co-exist in the same (small) air volume and the bimodal shape of the measured size
- 27 distributions cannot be explained as an observational artifact caused by accumulating data point
- 28 populations from different air volumes. The formation of the second size mode can be explained by (a)
- 29 entrainment and activation/condensation of fresh aerosol particles, or (b) by differential evaporation
- 30 processes occurring with cloud droplets engulfed in different eddies. Activation of entrained particles
- seemed a viable possibility as a layer of dry Arctic enhanced background aerosol (which was detected 31 directly above the stratus cloud) might form a second mode of small cloud droplets. However, theoretical
- 32
- 33 considerations and model simulations (adopting Direct Numerical Simulation, DNS) revealed that,
- 34 instead, turbulent mixing and evaporation of larger droplets are the most likely reasons for the formation
- 35 of the second droplet size mode in the uppermost region of the clouds.

### 1 **1 Introduction**

2 The Arctic sea ice plays an important role in the global climate system as it partly insulates the Arctic 3 ocean especially in the Winter season and diminishes the exchange of heat, momentum and gases between 4 ocean and atmosphere (Lemke et al., 2007). Surface temperature analyses show the strongest temperature 5 increase in the Arctic region in the past decades between 1961-1971 and 2003-2013 as resulting from the 6 GISS data base (Hansen et al., 2010). This temperature rise drives a melting process of Arctic sea ice 7 which reduces the albedo and, thus, increases absorption of solar radiation at the sea surface, with the 8 consequence of even more enhanced ice melting. This so-called "direct albedo feedback" occurs in the 9 Arctic summer and is a self-amplifying melting effect. As a result the temperature of the upper mixing 10 layer of the Arctic Ocean is increasing. Most of the heat stored during summer in the mixing layer is released in winter leading to an indirect albedo feedback causing Arctic amplification of near-air surface 11 temperature (Wendisch et al., 2013). In the Arctic satellite measurements (CALIPSO and ICESat) 12 between 2003 and 2008 it is shown that cloud cover and optical depth reach a maximum over ice-free 13 14 waters (Palm et al., 2010). The authors also conclude that the higher optical depths are caused by geometrically thicker low level clouds over the water. In addition an increasing trend of cloud coverage 15 16 between 7% and 10% over a period of five years was observed from the satellite data, which is most likely as result of decreasing sea ice coverage (Palm et al., 2010). Several publications indicate that Arctic 17 18 clouds might have been an important factor in explaining the recent dramatic Arctic sea ice losses (Kay et 19 al., 2008; Perovich et al., 2008; Schweiger and Lindsay, 2008). The microphysical characteristics of clouds, e.g., particle phase, size, number concentration, and shape determine the radiative properties 20 21 which influence the atmospheric radiation budget (Curry et al., 1996; Ehrlich et al., 2008a). At the same 22 time, climate simulations indicate that the sea salt emissions may increase with receding ice coverage leading to increasing values of optical depths of the natural background aerosol particles. This implies a 23 negative direct climate forcing of -0.2 to -0.4 Wm<sup>-2</sup> (Struthers et al., 2011). Such estimates are still highly 24 25 uncertain because of the partly crude representation of Arctic clouds and aerosol-cloud interactions in the 26 many models (Struthers et al., 2011). Especially mixed-phase low level clouds are of major concern here. 27 Phase-dependent microphysical processes such as collision, coalescence, aggregation, riming and 28 secondary ice formation (e.g., Hallett-Mossop ice multiplication) determine the growth and shape of the 29 ice particles in mixed-phase Arctic clouds (Pruppacher and Klett, 2010). Nucleation, growth, and sedimentation processes of ice crystals are still not comprehensively understood which leads to 30 31 discrepancies between observed and modeled ice number concentrations (Fan et al., 2009; Morrison et al., 32 2008; Avramov et al., 2011). For the development process of clouds aerosol particles may play a significant role as they provide Cloud 33 34 Condensation Nuclei (CCN) and influence the direct and indirect cloud radiative effects (Lemke et al., 35 2007). Several previous measurements indicate a correlation between the number concentrations of aerosol and of cloud particles which implies an effect of aerosol particles on the cloud particle size 36 37 distribution (SD) (Pruppacher and Klett, 2010). Aerosol particles may also influence the lifetime of clouds

- 38 which form at the bottom of stable boundary layers (Stevens, 2005). These boundary layer clouds act to
- 39 cool the atmosphere (annually and globally averaged) which is explained by the dominance of the solar
- 40 albedo effect or the emission of thermal-infrared radiation. However Arctic boundary layer clouds lead to
- 41 a net warming effect of the below-cloud atmosphere depending on the season. This is due to the generally
- 42 low sun elevation, the long-lasting polar night, and the high solar surface albedo of land/sea ice and snow
- 43 (Wendisch et al., 2013; Shupe and Intieri, 2004). Moreover, the magnitude of the radiative forcing of
- 44 boundary-layer, mixed-phase Arctic clouds critically depends on the clouds' macroscopic and microscopic
- 45 properties. Therefore, in-situ measurements are necessary to study aerosol-cloud interactions and their

- 1 radiative effects (Lawson et al., 2001; Lihavainen et al., 2010).
- 2 With regard to the cloud radiative effects, the upper top layer of boundary layer clouds is very important
- 3 because it dominates the overall cloud radiative properties (Ehrlich et al., 2009) and thus is a key for the
- 4 Arctic radiation balance (Stevens, 2005). Platnick (2000) showed that for inhomogeneous liquid water
- 5 clouds the vertical weighting function of the reflected radiation has a maximum close below the cloud top.
- 6 For mixed-phase clouds Ehrlich et al. (2009) showed that a small fraction of ice crystals in the cloud top
- 7 layer may change the cloud top reflectivity significantly with possible consequences for the accuracy of
- 8 cloud remote sensing. Therefore, microphysical processes at cloud top, such as the cloud top entrainment,
- 9 are another critical issue which still needs proper representation in the numerical models. Cloud top
- 10 entrainment is driven by the evaporation inducing downward directed turbulent motion which then
- 11 entrains the cloud free air from above. And this is crucial for the lifetime of the entire cloud as well as the
- 12 radiative budget because the entrainment and its rates influence directly droplet size and thus reflectivity
- in the visible. The main sources that produce turbulent energy at the cloud top are radiative cooling and
- 14 evaporative cooling (Pettersen 1938; Siems and Bretherton 1992).
- 15 In the past 20 years several aircraft campaigns were conducted to study the properties of arctic clouds:
- 16 \* In April 2008, during the International Polar Year, the NASA Arctic Research of the Composition of the
- 17 Troposphere from Aircraft and Satellites (ARCTAS) mission was conducted to better understand the
- 18 changes in Arctic atmospheric chemical composition and climate (Jacob et al., 2010).
- 19 \* At the same time, the Indirect and Semi-Direct Aerosol Campaign (ISDAC) was performed by using 41
- 20 instruments on the NRC Convair-580 aircraft. With this measurement platform the most complete dataset
- on arctic boundary layer and cirrus microphysical properties was collected over the North Slope of Alaska
- 22 (McFarquhar et al., 2011).
- \* One year earlier, the Arctic Study of Aerosol, Clouds and Radiation (ASTAR 2007) campaign was
- 24 performed to characterize microphysical and optical properties of arctic clouds. The thermodynamic phase
- 25 of the detected clouds was estimated by measurements of the reflected solar radiation. Three different
- 26 methods were used to identify the ice phase in Arctic boundary layer clouds (Ehrlich et al., 2008).
- \* In 2004, the Mixed-Phase Arctic Cloud Experiment (M-PACE) was conducted to study the
- 28 microphysical structure of Arctic mixed-phase clouds (Verlinde, 2007).
- \* Two earlier campaigns were the Surface Heat Budget of the Arctic (SHEBA) (Perovich et al., 2000),
  and the
- 31 \* First ISCCP (International Satellite Cloud Climatology Project) Regional Experiment Arctic Clouds
- 32 Experiment (FIRE ACE) (Curry et al., 1999). The SHEBA campaign was conducted 1997-1998 to study
- the heat transfer processes between the ice covered Arctic Ocean and the atmosphere. At the same time, in
- 34 April July 1998, the FIRE ACE campaign was performed to improve (a) the satellite retrieval of cloud
- 35 and surface characteristics in the Arctic, and (b) the representation of Arctic clouds and radiation in
- 36 general circulation models. For this purpose, four aircraft were equipped with instrumentation as described
- 37 in Curry et al. (1999).
- 38 During FIRE ACE, cloud particle SDs of Arctic boundary layer clouds were studied. Monomodal SDs
- 39 were mostly observed under adiabatic and homogeneous liquid water cloud conditions whereas bimodal
- 40 SDs appeared near the cloud top, when the temperature and liquid water content profiles indicated non-
- 41 adiabatic and inhomogeneous cloud conditions (Lawson et al., 2001). Mixed-phase clouds, which consist
- 42 of liquid and ice phase particles, appear mainly in the autumn and springtime. Boundary layer clouds
- 43 containing ice and liquid water were typically detected in May in an altitude range from 150 to 1200 m
- 44 (Lawson et al., 2001). The detected ice particles have diameters between 8 μm and 1 mm while liquid

1 cloud droplets mostly are in a size range of tens of micrometers with maximum droplet number

concentrations of 500 cm<sup>-3</sup> (Curry et al., 1996). 2

3 The purpose of this study is to analyze in further detail the microphysical properties inside and at the top

- 4 of the boundary layer stratus clouds in the Beaufort Sea area with emphasis on the occurrence of
- 5 monomodal and bimodal distributions and the connection to turbulent mixing processes at the cloud top levels.
- 6
- 7
- 8
- 9
- 10

#### 11 2 Field campaign and instrumentation

12 Airborne microphysical measurements within Arctic boundary layer's mixed-phase clouds were

13 performed during the **VER**tical **D**istribution of **I**ce in Arctic Clouds (VERDI) field campaign. The

14 POLAR 5 research aircraft (Basler BT-67, modification of a DC-3), operated by the Alfred Wegener

- 15 Institute (AWI, Helmholtz Centre for Polar and Marine Research, Bremerhaven, Germany), was equipped
- with instruments to carry out in-situ measurements aimed in particular at mixed-phase clouds, during the 16

17 Arctic spring season. For the detection of cloud particles, a Cloud Combination Probe (CCP), a Small Ice

18 Detector (SID), a Novel Ice Experiment – Cloud and Aerosol Particle Spectrometer (NIXE-CAPS), a

19 Cloud and Aerosol Spectrometer with Depolarization (CAS-DPOL), and a Precipitation Imaging Probe

20 (PIP) were used. With these instruments it is possible to cover a diameter size range from  $0.6 \,\mu m$  up to

21 6.4 mm. Four of the cloud particle spectrometers with partially overlapping detection size ranges were

22 simultaneously operated side by side to provide independent SD measurements. Solar (spectral and

23 broadband) as well as broadband thermal-infrared radiation sensors and an aerosol Optical Particle

24 Counter (Sky-OPC) were deployed on the aircraft. Also, a cloud-aerosol LiDAR, and meteorological

25 instruments were part of the instrumentation. With this equipment, different situations of ice-, liquid-phase

26 and mixed phase clouds were probed. VERDI was performed from 25 April to 17 May 2012 out of Inuvik, 27 Canadian Northwest Territories, with the main operation area over the Mackenzie River delta and the

28 Beaufort Sea.

29 The measurements in this study are mainly based on the Cloud Combination Probe (CCP), the Sky-

- 30 OPCand the LiDAR instrument. The CCP integrates two instruments to measure parameters of individual
- 31 cloud hydrometeors (Wendisch and Brenquier, 2013) and was a modified version of the instrument
- initially manufactured by Droplet Measurement Technologies (DMT, Boulder, CO, USA, see 32

http://dropletmeasurement.com/). It consists of a Cloud Droplet Probe (CDP) and a Cloud Imaging Probe 33

34 (CIP grey scale – denoted as CIPgs in the following) which in combination cover a size diameter range

35 from 2 µm to 960 µm, including large aerosol particles, liquid cloud droplets and small frozen

hydrometeors. The technique of the CDP is based on forward light-scattering with a light collection angle 36

from 4° up to 12°, similar to the Forward Scattering Spectrometer Probe (FSSP). A laser diode wavelength 37

38 of 658 nm is used as light source. The CDP instrument measures particles in a diameter size region

39 between 2 µm and 50 µm with a size resolution provided by 30 detector channels. Its general performance

- 40 and the sample area were tested and measured using a piezoelectric droplet generator laboratory setup
- similar to the design of Lance et al. (2010). As a result a sampling area of  $0.27 \pm 0.025$  mm<sup>2</sup> was 41
- obtained. Measurements of cloud probes are often being affected by shattering (Jensen et al., 2009; 42
- Korolev et al., 2011). However, this CDP is an open path design and is equipped with tips of particular 43

- 1 shape to minimize the impact of shattering on the measurement (Korolev et al., 2013). Furthermore, the
- 2 instrument records the particle-by-particle (PbP) data which allows for excluding collection periods
- 3 potentially affected by shattering (Field et al., 2003; Field et al., 2006). The CIPgs instrument, the second
- 4 sub-instrument of the CCP, records two-dimensional shadow images of cloud particles in a size range
- 5 from 15  $\mu$ m up to 960  $\mu$ m with an optical resolution of 15  $\mu$ m. Specialized algorithms are used to process
- 6 and analyze the captured images in order to estimate particle number concentrations, particle SDs, and to
- 7 differentiate particle shapes (Korolev, 2007).
- 8 The Sky-OPC is a Series 1.129 aerosol spectrometer manufactured by Grimm Aerosol Technik GmbH
- 9 (see http://www.grimm-aerosol.com/en/), which determines the aerosol particle number concentrations
- 10 within 31 size channels and the particle sizes from 250 nm up to 32  $\mu$ m. However, due to the sampling
- 11 inlet efficiency of the applied (solid diffuser) aerosol inlet (Huebert et al., 2004) for the POLAR 5, only
- aerosol particles of up to 1 µm in diameter are representatively sampled from the ambient air. Individual
- 13 particles are detected by their light scattering signal within an angular range from 60 to 120 degrees of a
- 14 diode-laser with a wavelength of 655 nm. The geometry of the instrument is less sensitive to the refractive
- index than a narrow-cone forward scattering instrument. Nevertheless, because an inlet with a tube systeminside the aircraft is used, the detected aerosol at the instrument is much drier than outside the aircraft. The
- 17 sampling was such that particles were heated to temperatures more than 20 °C above ambient before the
- 18 size measurement of the OPC. The standard integration time of the Sky-OPC for one SD has been set to 6
- 19 seconds for the VERDI flights. Particle SDs recorded by several in-situ instruments show a good
- agreement in the overlapping size ranges between 250 nm and 6.4 mm.
- 21 The Airborne Mobile Aerosol LiDAR (AMALi) emits laser pulses into the atmosphere and measures
- backscatter from air molecules and aerosols at the wavelengths 532 nm and 355 nm, as well as
- depolarization at 532 nm. Here the measurement at 532 nm is used to characterize the spatial structure of
- 24 aerosol and cloud layers. During VERDI AMALi was operated in nadir pointing mode with a vertical
- 25 resolution of up to 7.5 m and a maximum temporal resolution of 1 s.
- 26 To identify the liquid phases during the campaign we used the NIXE-CAPS instrument which is able to
- 27 discriminate between spherical and aspherical particles based on measurements of the polarization of laser
- 28 light scattered by single particles. With this instrument it is possible to identify glaciated hydrometeors
- 29 within the size range from  $0.6 \,\mu\text{m}$  to  $950 \,\mu\text{m}$ .
- 30

## 31 **3 Observations**

## 32 **3.1** Features inside the Arctic boundary layer clouds

- 33 During VERDI, thirteen measurement flights were performed around Inuvik and above the Beaufort Sea
- 34 which was partly covered with ice. The probed super-cooled liquid water clouds which are the subject of
- this study were primarily observed after May 8, 2012. Before that time only mixed phase clouds,
- 36 occasionally including with glaciated hydrometeors as large as 6 mm, were encountered during the
- 37 observations.
- 38 The flight path of Flight 11 on 15 May 2012, is shown in Figure 1, where a cloud layer was observed
- between 69.8° and 70.5° latitude (marked by the red line) in an altitude interval between 630 and 1060 m
- 40 above sea level. Figure 2 shows profiles of different meteorological and microphysical parameters of this
- 41 cloud. The cloud top as marked by the dashed line was at an altitude of 990 m. The profile was measured
- 42 during a vertical ascent though the cloud layer. The layer directly above the dashed line (from 990 to
- 43 1060 m) is denoted as the transition zone in the following. Here the temperature *T* increased rapidly from

1 268 K to 271 K. The relative humidity *RH* inside the cloud was around 100 % and above the cloud top *RH* 

2 declined to values around 80 %. Thus, the cloud is capped by a layer of warmer and drier air which is

- 3 typical for the Arctic stratocumulus. The droplet number concentration, *N*, measured by the CDP, reveals
- 4 constant values around 70 cm<sup>-3</sup> almost independently of height. The liquid water content LWC (as derived
- 5 from the measured droplet SDs), increased with altitude inside the cloud. Both, constant *N* and *LWC*
- 6 increasing concurrently as function of height have been found to be typical for liquid water clouds
- 7 generated by adiabatic lifting/cooling (McFarquhar et al., 2007). The calculated adiabatic *LWC* is
- 8 indicated by the dash-dotted line in Panel d) of Figure 2. While the measured and calculated *LWCs* are

9 fairly close within the lowest 90 meters of the cloud the influence of the entrainment processes becomes

10 increasingly evident from the deviations between the two in the upper regions of the cloud. McFarquhar et

- al. (2011) showed similar trends in vertical profiles and in combination with radar measurements. Their
   results indicate vertical mixing driven by dynamics or turbulence.
- The fact has a base of the stars and basis of the line in the line interview of the stars and t
- 13 To further characterize the atmospheric stability inside the cloud, the liquid water potential temperature  $\theta_l$
- 14 (according to Stevens, 2005) is calculated.  $\theta_l$  represents the potential temperature attained by evaporating
- all the liquid water in an air parcel through reversible wet adiabatic descent (Betts, 1973) and is defined
- 16

as:

- 17
- 18

$$\theta_l \approx \theta \exp\left(\frac{-LWC \, L_v}{c_p \, T}\right).$$
(1)

- 19 Here  $\theta$  represents the potential temperature for dry air in Kelvin, *LWC* [in kg m<sup>-3</sup>] is the mass of liquid
- 20 water, while the latent heat of vaporization is denoted as  $L_v$  [in J kg<sup>-1</sup>K<sup>-1</sup>], the heat capacity as  $c_p$
- [in J kg<sup>-1</sup>K<sup>-1</sup>] and the temperature as T. In Panel e) of Figure 2 the liquid water potential temperature  $\theta_l$
- exhibits a continuous increase with altitude, which clearly indicates stable atmospheric conditions insidethe cloud.

24 In the following, the microphysical measurements from Flight 11 on 15 May are described in detail for the

- layer inside the cloud (from 630 to 990 m) and in the transition zone (990 1060 m) directly above.
- 26 The cloud consisted of super-cooled liquid water droplets in a size range up to 50 µm. Only a few ice
- 27 particles, mostly needles, with a number concentration of  $0.75 l^{-1}$ , were occasionally detected by the CIPgs
- 28 instrument between altitudes of 630 to 990 m. Figure 3 shows a time series of the aerosol number
- concentration (Figure 3a) and the droplet geometric mean diameter (Figure 3b). The altitude profile shown
- in Figure 3a and b was flown with a heading to the north which is also marked by the red line in Figure 1.
- 31 Inside the cloud and during ascent between 20:08 and 20:22 UTC the droplet geometric mean diameter
- 32 increased with altitude while the aerosol number concentration decreased.
- 33 The increasing size of the cloud droplets is also obvious in Figure 4a-4c where the narrow and monomodal
- 34 SDs from different altitudes are displayed. The modal droplet diameter grows continuously from 12 to
- $20 \ \mu m$  with altitude. Calculations for diffusional droplet growth, which are based on the set of four
- 36 differential equations given in Pruppacher and Klett (2010) including released latent heat conduction to
- 37 the air and back to the droplets, provide the necessary time for the droplets to grow from 12  $\mu$ m to 20  $\mu$ m
- under the given ambient conditions. Assuming values for the relative humidity of 100.15%, 100.10%, and
- 39 100.08% the calculations indicate growth times of 7, 15 and 37 minutes, respectively, which is well within
- 40 the range of lifetimes for such clouds or cloud fragments. Under the assumption that the vertical updraft is
- 41 1 m/s the cloud particles would ascend 420 m, 600 m and 2220 m, respectively. However, the cloud depth
- 42 that was studied is 360 m (altitude range: 630 990 m), which is smaller than the calculated ascents. For

1 the studied case, this implies that the vertical updrafts have to be smaller than 1 m/s or circulations effects

2 like those described in Korolev and Field (2008) must occur to maintain the cloud depth.

The monomodal shape of the SDs and the monotonic increase of their modal peak is comparable with observations by Lawson et al. (2001), who explained their appearance by adiabatic ascent of in-cloud air. As shown in Figure 2d, deviations between the measurements and the expected adiabatic *LWC* due to an adiabatic ascent are obvious and indicate that the in-cloud air does not experience fully adiabatic conditions in our case. Continuous ascent throughout a certain altitude range implies averaging over an extended section of the linear flight path inside the cloud. This may have contributed to the deviations as it differs from a real vertical profile above one specific location.

9 10

3

4 5

6

7

8

11 Inside the transition zone at the cloud top, bimodal SDs with a second mode around 10  $\mu$ m are observed, 12 as shown in Figure 5c. This phenomenon was encountered only in the transition zone of the cloud during

- 13 three VERDI flights (Flight 9 on 10 May, Flight 11 on 15 May and Flight 13 on 17 May). For the other
- 14 flights in liquid water clouds only an increase of the droplet size diameter with altitude was found. The
- reason why during these flights no bimodal SDs were detected in the transition zone can be explained by
- 16 the actual flight profile, which the aircraft conducted. The bimodal SDs only were observable (1) when the
- 17 aircraft spent at least 1 second (~70 m) in the transition zone, or (2) when the plane slowly enough
- 18 approached the top either from below inside the cloud or by entering the cloud descending from above.
- 19 Often the aircraft crossed the relevant highest cloud level too fast for the acquisition of a SD with good
- 20 counting statistics in each mode.
- 21 The SD shown in Figure 5b was detected inside the cloud at an altitude of 1004 m and shows a

22 monomodal shape similar to Figure 4. To distinguish both modes, in the following the mode at larger sizes

- $(20 \ \mu m)$  will be labeled as Mode 1 and the one at smaller sizes is denoted as Mode 2. Figure 5a indicates
- that during Flight 11 several flight sequences with bimodal SDs were detected between 20:22:00 and
- 25 20:27:00 UTC when the aircraft flew through the transition zone of the cloud.
- 26

27 Besides the cloud droplets, also aerosol particles were measured during the VERDI campaign by using the Sky-OPC and AMALi. Figure 6 provides two examples of AMALi data (attenuated backscatter) from two 28 29 different flights and for times when the aircraft emerged from the cloud top and turned around to fly back 30 over longer stretches above and in the vicinity of the cloud parts which were in-situ sampled directly 31 before. The total attenuated backscatter signal is shown, which consists of light scattered from the cloud 32 particles as well as the background contribution due to the air molecules. The coarse color scale is 33 normalized with respect to the values between 2100 m and 2400 m, assuming a low and homogeneous aerosol concentration at that altitude. The upper panel shows the backscatter signal from the penetrated 34 35 boundary layer cloud during Flight 11. During VERDI the encountered clouds mostly consisted of several stacked layers with clear air in between, and we did not often find such a "classic" boundary layer cloud 36 37 deck. The backscatter signal does not show a distinct pattern above the cloud, only a very slight decrease 38 with altitude as time progresses. The values of the attenuated backscatter due to the aerosol are indicative 39 of very low levels of the encountered Arctic aerosol. The penetrated cloud layer was overpassed at 20:41 40 UTC. In this region, the cloud top height shows no significant changes. As McFarquhar et al. (2011) pointed out substantial changes of cloud top height can occur during a flight. This is the case here too (see 41 42 Figure 6a) because at the end of the displayed flight segment the cloud top height is approximately 300 m lower than at the beginning. In order to highlight the differences compared to other atmospheric 43 conditions, the lower panel shows the data of Flight 7 on 5 May. During this flight an optically thick 44 45 aerosol layer (OD=0.02 + 0.005 assuming a lidar ratio of 40sr) is discernible above the cloud. Since this 46 layer is optically thicker it probably cannot be classified as clean Arctic background aerosol. HYSPLIT

- 47 reanalysis trajectories over 72 hours (see http://ready.arl.noaa.gov/HYSPLIT.php) indicate that the aerosol
- 48 originates from Yukon Territories and Alaska in an altitude range between 500 and 1500 m. By contrast
- reanalysis trajectories for the other days indicate that between 05.05.2012 and the 10.05.2012 air masses
- 50 containing aerosol were advected mostly from the Canadian and American mainlands.
- 51 Every time during Flight 11, when the ascending aircraft penetrated the cloud top , the number

- 1 concentration of the aerosol particles in a size region between 250 nm  $< D_p < 1 \mu$ m increased from
- 2 0.7 ( $\pm$  0.08) cm<sup>-3</sup> up to 7.5 ( $\pm$  0.02) cm<sup>-3</sup>. Figure 3a shows this increase in the aerosol number
- 3 concentration at 20:25:00 and 20:31:30 UTC. Aerosol particle SDs from the OPC measurements are
- 4 shown in Figure 7. In order to provide a comparison with a typical clean atmosphere encountered during
- 5 VERDI, a Sky-OPC SD from Flight 10 on 14 May is added and from the error bars (based on counting
- 6 statistics) the range of variability can be seen. The aerosol number concentration above the cloud,
- 7 particularly for smaller sizes between 250 nm  $< D_p < 500$  nm, is by a factor of about seven higher than
- 8 inside the cloud. Unfortunately, for detecting particles smaller than 250 nm no instrument was available
- 9 during the VERDI campaign. For this reason, the averaged dry Arctic background aerosol SD, which was
- 10 measured and reported as regional average by Lathem et al. (2012) with an Scanning Mobility Particle
- 11 Sizer (SMPS), is adapted and also included in Figure 7.
- 12 (Note: In addition to the SMPS system Lathem et al. (2012) used a Cloud Condensation Nuclei Counter
- 13 (CCNC) to determine the fraction of aerosol that can be activated to cloud droplets. They found an
- 14 activation ratio of 0.52 (at 0.55 % internal supersaturation inside the instrument) which shows that
- approximately half of the measured Cloud Nuclei (CN) were activated.)
- 16 Despite the uncertainties inherent in a comparison of measurements which were four years apart, this
- 17 juxtaposition shows that the aerosol concentrations from the single measurements during VERDI match
- 18 quite well with or at least do not strongly deviate from the previously mapped regional average. In
- 19 particular the background aerosol particles measured above the cloud during VERDI do not exhibit largely
- 20 different number densities than those reported by Lathem et al. (2012). Therefore, the detected aerosol
- above the cloud on 15 May 2012, might well be considered as dry Arctic background aerosol which seems
- to be continuously present in this region. The aerosol number concentrations above the cloud from May 5,
- however, are significantly higher than the background typically found during VERDI.
- 24 25

#### **1 3.2** Spatial distribution of the droplets from the bimodal size distributions

2 To exclude the possibility that the bimodal SDs are result of a sampling artifact caused by time-averaging 3 over spatially separated particle populations with different size modes, the PbP data from the CDP were 4 examined. In this way the sizes and the exact detection times for consecutive single detected particles 5 become available. Figure 8a shows a plot based on PbP analysis for data collected during a 3.5 second 6 period (i.e. a flight segment of roughly 254 m length) from Flight 11, which is the particular period where 7 the bimodal SD shown in Figure 5c was observed. Figure 8a displays a time series of the individual 8 particle detections with the two size modes separated by the dashed line with Mode 1 covering particle 9 diameters around 20 µm and Mode 2 those around 10 µm. Overall, 1023 particles were recorded in this 10 period. The time gaps, where no consecutive particles are displayed, result from the limited buffer capacity and transmission speed (i. e. a maximum of 256 particles in one second) of the PbP recording of 11 the CDP. The contiguous PbP-record segments in Figure 8a are roughly 0.3 seconds long and correspond 12 to about 20 m of flight distance within the cloud. To illustrate the mixing state of both modes, the detected 13 14 particles are labeled with two colors. The red color dots represent particles where the previously detected 15 particle was from the same size mode. The black dots mark particles where the previously detected particle originated from the other mode. If the aircraft had moved through one patch of air containing only 16 Mode 1 particles (like in the SDs of Figure 4) and afterwards through a different air parcel with mostly 17 droplets from Mode 2 - for example because the two parcels were from different entrainment filaments-, 18 19 then a bimodal SD might result from data averaging over a sufficiently long flight path stretch. In this case 20 the PbP "arrival time analysis", however, would have revealed such effects. To investigate the mixing of 21 the two modes on such scales a statistical approach is used by comparing the probabilities of black and red 22 dots to those of an ideal mixing state where particles from any of the two modes are detected at random. 23 The probability of detecting two successive particles from two different size modes in a fully random

24 process is:

$$P_{12+21} = p_1 p_2 + p_2 p_1 = 2(p_1(1-p_1)) = 2p_1 - 2p_1^2, \qquad (1)$$

26

with  $p_1$  and  $p_2$  designating the occurrence rates (i.e. the probabilities) of particles from Mode 1 and

- 28 Mode 2 and with  $p_1 + p_2 = 1$ . Figure 9 displays  $P_{12+21}$  from Equation 1 as black line.
- 29 Consider as example that 100 particles were successively detected with 60 (i.e. a value of 0.6 on the
- 30 abscissa) originating from Mode 2 and 40 from Mode 1. In this case the probability of having two random
- 31 consecutive particles and finding them in different modes is in theory 48 % as taken from the ordinate in
- 32 Figure 9. For the measured example (i.e. the bimodal SD detected between 20:22:46 and 20:22:50 UTC on

Flight 11), however, it was 44.7 % and in this case, the bimodal SD shown in Figure 8a is close to a

- 34 uniform distribution. The mean interarrival time between the individual particles is 1.45 milliseconds.
- 35 Therefore, with an aircraft speed of 70 m s<sup>-1</sup>, the mean distance between two detected particles is roughly
- 36 10 cm. It can be concluded that within this detection rate, limited by the nearly one-dimensional sample
- 37 volume of the instrument, both size modes are well mixed on the decimeter scale. Figure 8b provides
- 38 similar data based on PBP analysis from the same flight at an earlier time. Apparently here the black and
- red dots are mostly clustered either in Mode 1 or in Mode 2. Thus the two modes were sampled as disjoint
- 40 droplet populations. Averaging over the corresponding flight stretch would deliver a bimodal distribution
- 41 although both modes are in separate cloud droplet populations. This demonstrates how potential sampling
- 42 artefacts resulting from averaging can be identified by PbP analyses.
- 43 Further bimodal SDs from Flight 9 and Flight 13 were examined this way and also integrated in Figure 9.
- 44 In summary, four bimodal SDs (marked by circles) seem to result from well mixed air parcels. For the

- 1 other three bimodal SDs, the agreement with the uniform distribution is less pronounced which suggests
- 2 clustering of particles in each single mode (on sub-meter scale). In these mostly clustered cases, the cloud
- 3 droplets essentially were sampled from two disjoint particle ensembles.
- 4
- 5

## 6 4. Theoretical considerations and modeling

7 We first consider the possibility that the entrainment of dry aerosols from the free atmosphere might lead 8 to cloud droplet nucleation, and the formation of the second mode in the SD (around 10  $\mu$ m). This process 9 was invoked by Lawson et al (2001) to explain the bimodal SDs in their observations. Further aerosol 10 effects on ice nucleation, entrained in arctic clouds, were observed and analyzed by Jackson et al. (2012) 11 and Earle et al. (2011).

12 Isobaric mixing can lead to the necessary supersaturation for condensation if the saturation-vapor-pressure

13 curve is sufficiently nonlinear over the temperature range relevant for the mixing. Figure 10 shows the

- saturation-vapor-pressure curve in comparison with the studied case from the flight of 15 May 2012. The
- 15 saturation-vapor-pressure curve is almost linear in the range covered by the measurements, indicating that
- 16 the condensation of new droplets in the mixing is not possible. This argument can be substantiated by
- 17 discussing the total droplet number concentration. For the second Mode (around 10  $\mu$ m) in Figure 5c, in a
- 18 size range between 2.8  $\mu$ m and 13  $\mu$ m, the total droplet number concentration is 15.3 cm<sup>-3</sup>. In comparison,
- 19 this is twice as much as the aerosol number concentration (see Figure 3a), which is around 7.5  $\text{cm}^{-3}$ . This
- 20 means that more droplets develop in the second mode than there are available aerosol particles.
- 21 Nevertheless, we cannot exclude the possibility that single aerosol particles are activated, but evaporation
- processes seem to be the major effect. A "closure experiment" according to McFarquhar et al. (2011)
- 23 where the particle numbers from inside the cloud match those of the aerosol particles from above the
- cloud could not be performed here because our instruments did not cover a brought enough size range.
- 25 In this section we describe Direct Numerical Simulation (DNS) model calculations and results in order to
- try to explain the observed bimodal SDs in the cloud top entrainment zone. The main assumption is that
- the bimodal SDs result from the turbulent mixing dynamics at the stratocumulus top, which is supported
- by the fact that all bimodal SDs were observed close to the cloud top, in contrast to the monomodal SDsencountered inside the cloud. Direct Numerical Simulations were adopted (rather than e.g. Large Eddy
- 30 Simulations) to demonstrate how the turbulent mixing dynamics at the stratocumulus top can generate
- bimodal SDs similar to the observed ones. The two main mechanisms for turbulent mixing in
- 32 stratocumulus are infrared radiative cooling and evaporative cooling. By means of Doppler X-band radar
- 33 measurements McFarquhar et al. (2011) showed that such turbulent effects occur in the Arctic boundary
- 34 layer cloud systems. Close to the cloud top infrared radiation cools parcels that "fall back down into" the
- 35 cloud, initiating turbulence and mixing. Turbulent mixing entrains dry air from the free atmosphere above
- the cloud, causing the evaporation of cloud droplets. The evaporative cooling involved in this process can
- in turn considerably enhance the turbulent mixing at the cloud top, depending on the thermodynamic
- 38 properties of the cloud and adjacent free atmosphere. In some cases a horizontal wind shear can be strong
- enough to contribute or even dominate the mixing process.
- 40 As next step, three-dimensional direct numerical simulations (DNS) of a mixing layer were used in order
- 41 to test the hypothesis that turbulent mixing and evaporation can explain the bimodal SDs.
- 42 Mellado (2010) and de Lozar and Mellado (2014) showed that simulations of such a simplified
- 43 configuration are a valid approach for the investigations of cloud-top mixing. DNS provide a faithful

- 1 representation of the mixing, even at the smallest resolved scales, which is necessary to investigate how
- 2 the small-scale turbulent mixing modifies the SD. In our calculations we simulate scales from a
- 3 Kolmogorov length  $\eta = 15$  cm to an integral length scale of 200 m, thus capturing the relevant scales for
- 4 mixing. The main limitation of DNS is that current studies are constrained to small and simplified setups,
- 5 due to their great computational cost. For this reason our simulations are driven only by infrared radiative
- 6 cooling and evaporative cooling, and we neglect other cloud processes, like microphysics, surface fluxes,
- 7 shear, rain or short-wave radiation. Although those processes might have an influence on the cloud
- 8 dynamics, estimates show that their contribution to the turbulence and droplet growth is relatively small
- 9 when compared to radiative and evaporative cooling (for example see Stevens et al. (2005) and
- 10 Bretherton et al. (2007)). For this reason those processes are probably not strong enough to change
- 11 qualitatively the small-scale mixing behavior.
- 12
- 13 The set of governing equations is based on the one-moment bulk scheme described in de Lozar and
- 14 Mellado (2013b). The equations are further simplified by linearizing the buoyancy and the saturation-
- 15 vapor-pressure functions around a reference state. The resulting formulation is equivalent to the one
- 16 presented by Bretherton (1987), and is subject of a forthcoming manuscript. In this approximation the
- 17 saturation-vapor-pressure function is linear, and therefore the isobaric condensation mixing is neglected.
- 18 The long-wave radiation model is a one dimensional model, that assumes that radiation only propagates in
- 19 the vertical direction (Larson et al., 2007).
- 20 The thermodynamic parameters used in the simulation are based on the measurements from the flight of
- 21 15 May 2012: LWC = 0.25 g/kg, the sum of the vapor and liquid water content  $q_t = 3.15$  g/kg,
- 22 T = 268.15 K and  $\Theta_l = 275.2$  K in the cloud layer; and  $q_t = 2.4$  g/kg, T = 272.65 K, RH = 59% and
- 23  $\Theta_l = 280.5$  K in the dry air layer above the cloud. The atmospheric pressure is 905 hPa. The cloud-top
- radiative cooling,  $F_0 = 60$  W/m<sup>2</sup>, matches the data from the instruments on board the POLAR 5, and an
- extinction length was chosen to  $\lambda = 15$  m (see Larson (2007) and de Lozar and Mellado (2014) for an
- 26 explanation of the radiation parameters). The simulation runs for 12 minutes and covers for the turbulence
- 27 length scales from 15 cm to 200 m. The grid employed in the numerical simulations contains  $1024^3$  points,
- the width of the simulation domain is 270 m and the resulting grid spacing in the region of interest is
- 29 26 cm.
- 30 Figure 11 shows a vertical cross section from our simulations, in which the liquid water density ranges
- from 0.0 g/kg (blue) to 0.325 g/kg (white). Turbulence and mixing are evident in the transition zone
- 32 between the cloudy air and the free dry atmosphere, as it is typical for stratocumulus clouds. In the
- transition zone the entrained air concentrates on some regions that are indicated by red squares in
- Figure 11. We specify these regions as holes, in analogy to the liquid free regions observed by Gerber et
- al. (2005) in some tropical clouds. In comparison to the previously observed holes (Gerber et al., 2005),
- 36 the holes in our simulations contain more liquid water and stay closer to the cloud top. These differences
- are explained by the relatively high humidity of the free atmosphere measured in the flight of 15 May
- 2012 (RH = 59%) above the cloud. The potential for evaporation is lower and therefore it is not surprising
- that some liquid water droplets still are present in the holes after the mixing process. Given the
- 40 combination of low cloud water concentration and still enough water vapor in the free atmosphere, the
- 41 evaporative cooling is expected to be low. As a consequence the air in the holes is mostly positively
- 42 buoyant, and the holes have a tendency to remain close to the cloud top.
- 43 Our hypothesis is that the bimodal SDs shown in Figure 5c are a result of the mixing within the holes. For
- 44 testing we simulated the SD that an aircraft would measure in our model, when flying close to the cloud
- 45 top (black line in Figure 11). The mean diameter of the droplets in each grid box was calculated using the

1 liquid water from the simulations and a droplet number concentration N = 70 cm<sup>-3</sup> as measured e.g. in

2 Figure 2. For the last 70 m (equivalent to 1 second of flight time for the POLAR 5) of the simulated plane

- 3 trajectory (including the big hole highlighted by the bounding box) the SD shows a bimodal shape (see
- 4 Figure 13) which at least qualitatively agrees with the measured ones in Figure 5c. Even though the
- 5 minima between the two modes in the SDs are 4  $\mu$ m apart and the number concentrations of the large
- 6 droplet mode are quite different the SD are similar enough to suggest that the evaporation of droplets in7 the holes causes the bimodality.
- 8 The second mode in the SDs in the simulations can be explained if we assume that the large-scale
- 9 dynamics of each hole are dominated by a single large eddy, while the rapid mixing inside the holes is due
- 10 to much smaller turbulent eddies. The time scale of the large eddy determines the mixing and composition
- 11 inside a hole, defining a typical droplet diameter: Large eddies are able to capture large quantities of dry
- 12 air resulting in a small typical droplet diameter, while smaller eddies are characterized by larger droplet
- 13 diameters. This can be seen in Figures 11 and 12 where large and small holes occur. For example the
- 14 rightmost red box in Figure 11 contains such a large hole (i.e. a large parcel of drier air) yielding in the
- 15 simulation many small "Mode 2" droplets. Most likely the second mode with the smaller droplets in the
- SD represents the typical droplet diameter of the hole in a bounding box, whereas the first, larger droplet mode corresponds to cloud droplets that have not yet been mixed. A second simulation for a different
- flight sequence from Flight 13 also yielded to a good agreement between measured and calculated SD.
- Further, more extensive simulations with detailed sensitivity studies concerning the various governing
- 20 parameters are necessary here together with similar comparisons to field data, which should also cover
- 21 different thermodynamic situations (e.g. moistures, temperatures above the clouds). Since such DNS are
- 22 very expensive in terms of computational time, juxtaposition of the results with those from other
- approaches for numerical simulations, like Large Eddy Simulation (LES), are also of advantage. To
- 24 analyze this phenomenon in a higher resolution a holographic instrument capable of detecting particles in
- 25 a three-dimensional sample volume of a few cm<sup>3</sup> in a single snapshot (Spuler and Fugal, 2011;
- 26 Schlenczek et al. 2014) allows investigation of still smaller mixing scales for both size modes. Also new
- 27 imaging remote sensing techniques (e.g. as presented by Bierwirth et al., 2013) could help to identify the
- horizontal variability of the cloud top regions on different scales and thus to quantify the interplay
- 29 between eddy driven mixing and cloud microphysical processes.
- 30

# 31 5. Conclusions

32 The VERDI campaign was performed to study microphysical and radiative properties of Arctic boundary 33 layer clouds in the Beaufort Sea area. Previous microphysical measurements have shown the occurrence 34 of monomodal and bimodal size distributions caused by adiabatic ascent and activation of new CCN, 35 which were entrained. In the VERDI campaign similar size distributions were observed in boundary layer 36 stratus and stratocumulus clouds during three flights including monomodal size distributions with the 37 modal peak size varying between 12 and 22 µm dependent on altitude within the cloud. Measurements of 38 the LWC indicate deviations from adiabatic conditions and, therefore, entrainment processes at the top of 39 the penetrated cloud deck play a role. LiDAR and aerosol instruments above the cloud layers showed a 40 well mixed aerosol particle population which can be classified as dry Arctic background aerosol. In the 41 cloud transition zone at altitudes between 990 and 1060 m a change from monomodal towards bimodal 42 cloud droplet size distributions was observed. A method adopting particle-inter-arrival-times was applied 43 to study the mixing progress between both droplet modes ultimately demonstrating that for four of seven

44 cases with bimodal droplet size distributions a nearly uniform mixing process prevailed. This means that

- 1 an artifact due to (flight) time averaging over two spatially separated particle population can be excluded
- 2 down to scales of a decimeter. The other cases indicate a sampling from two disjoint particle ensembles.
- 3 The development of the second mode in the transition zone could in principle be explained (a) by
- 4 activation/condensation of newly entrained CCN or (b) by evaporation processes. Condensation from
- 5 isobaric mixing is only possible for configurations with large deviations in temperature and water vapor
- 6 concentration. In the measurements the temperature variations between cloud and free atmosphere are so
- 7 small that the saturation-vapor-pressure curve is almost linear in this range, making isobaric condensation
- 8 of new droplets impossible. For this reason evaporation processes were examined in more detail by using
- 9 a model simulation study adopting Direct Numerical Simulations of turbulent eddy ensembles covering
  10 length scales from 15 cm (i.e. the Kolmogorov range) to eddies as large as 200 meters. This model
- 11 considered evaporative and radiative cooling and produces regions with concentrated entrained air. The
- resulting simulated bimodal cloud droplet size distributions in the cloud top entrainment zone are in
- 13 qualitative agreement with the size distributions obtained from the in-situ measurements. Turbulent
- 14 entrainment of clear-air-eddies seem to be the cause for the development of the second droplet size mode
- 15 (at 10 µm diameter) whereas the first size mode (near 20 µm) represents droplets that have not yet been
- 16 mixed with the drier air from above.
- 17 In conclusion, inside the transition zone of the observed boundary layer clouds we detected bimodal size

18 distributions where the smaller size mode seems to be a result from eddy driven in-mixing of drier air with

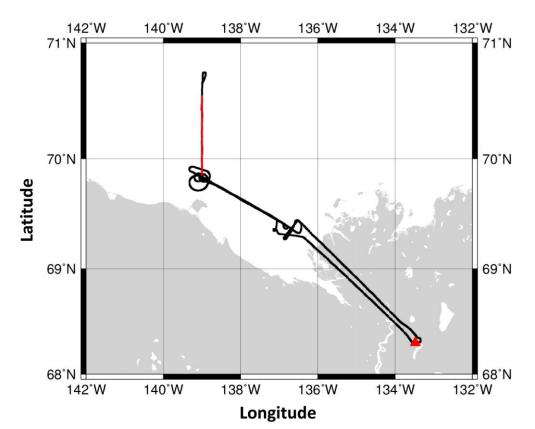
- 19 subsequent evaporation. More extensive experimental studies in real atmospheric clouds and simulations
- 20 with different numerical methods -focused on the cloud top entrainment zone of stratus and
- 21 stratocumulus- are necessary. These should cover different thermodynamic and cloud microphysical
- 22 parameter ranges to ultimately extend towards other cloud types as well.
- 23

## 24 Acknowledgements

25 This work was supported by internal sources of the Max Planck Society (Mainz and Hamburg) with

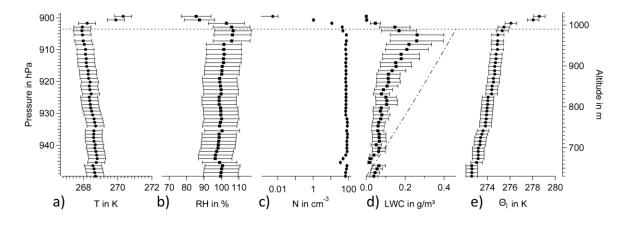
- 26 additional funds for the instrument integration and the research flights by the Alfred Wegener Institute for
- 27 Polar and Marine Research (Bremerhaven, Germany). We thank the staff of the Aurora Research Institute
- 28 (here the Western Arctic Research Center, WARC, in Inuvik, NWT, Canada) for their commendable
- 29 support and hospitality during the VERDI campaign. The support by the electrical engineers Wilhelm
- 30 Schneider and Christian von Glahn (University of Mainz) is very much appreciated. We particularly thank
- 31 the scientists, the pilots and the crew of the POLAR 5, as well as all other VERDI participants and
- 32 contributors. Computational time was provided by the Jülich Supercomputing Centre. We also would like
- to thank the two anonymous reviewers for their diligence and thoughtful suggestions.
- 34

- Figures



6 Figure 1. Track of Flight 11 as performed on 15 May 2012 in the Northwest Territories of Canada and the

Beaufort Sea. The triangle marks Inuvik Mike Zubko Airport and the red line shows the region where the boundary layer cloud was penetrated.



1

2 Figure 2. Altitude profiles inside the cloud encountered during Flight 11 (20:05 – 20:25 UTC) on 15 May

3 2012. Gradients of temperature *T* and the relative humidity *RH* above 990 m indicate that the cloud is

4 capped by a warmer and drier atmosphere. The droplet number concentration *N* is constant inside the

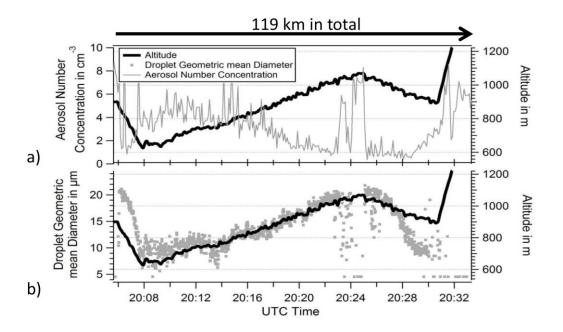
5 cloud. *LWC* and the liquid water potential temperature  $\Theta_l$  show an increase with altitude. The adiabatic

6 *LWC* is indicated by the dash-dotted line. The horizontal dashed line marks the beginning of the cloud top

7 transition zone in an altitude of 990 m. The error bars for *T* and *RH* result from the uncertainty of the

8 instruments and the errors for N are based on counting statistics. For LWC and  $\Theta_l$  the error is calculated by

9 error propagation from the measured parameters.



is indicated in both panels by a solid black line.

Figure 3. Time series of cloud in situ measurements, Aerosol number concentration (Panel a, grey line)

and mean cloud droplet diameter (Panel b, grey dots) during Flight 11 on 15 May 2012. The flight altitude

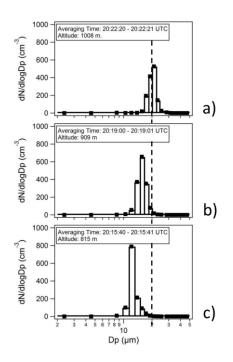
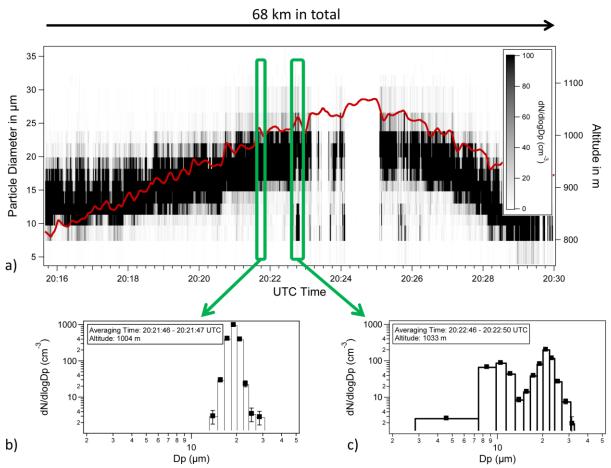




Figure 4. Monomodal cloud droplet size distributions from Flight 11 on 15 May 2012. Note the linear 3 scale of the ordinates with the dashed line marking the 20 µm diameter. A continuous increase of the

droplet particle diameter with increasing altitude from Panel c) to Panel a) is evident. 4



1

Figure 5. Upper panel: Time series of measured cloud particle data from Flight 11 on 15 May 2012. The
contour plot displays particle diameters over time, the greyscale encodes the number concentration and the
red line represents the flight altitude. Lower panel: Two different droplet size distributions: Panel b) as
detected inside the cloud and Panel c) in the cloud top transition zone. Bimodal size distributions only

6 occur in the cloud top transition zone.

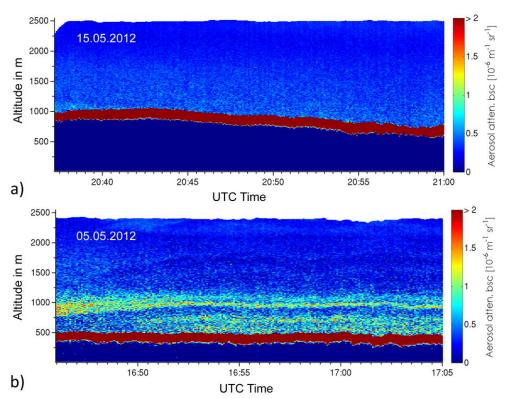
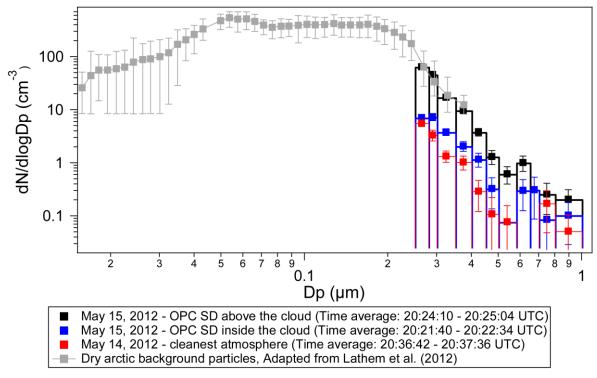


Figure 6. Panel a) LiDAR (AMALi instrument) backscatter signal from the studied boundary cloud layer in comparison with measurements including a thick aerosol layer above a cloud layer (Panel b).



2 3



- 4 measurements during a cloud free, clear air situation from Flight 10 on May 14, 2012. The error bars
- 5 denote the uncertainties due to the counting statistics while the width of the columns indicates the size bin
- 6 limits. The data shown for sizes below 0.4 μm (grey line and symbols) are SMPS measurements from
- 7 Lathem et al. (2012) reflecting the regional background aerosol levels from spring time 2008.

8

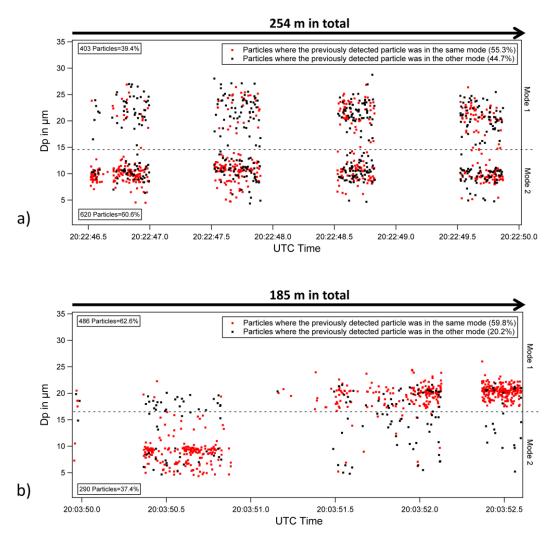


Figure 8. Time slice of Flight 11 on15 May 2012 with particle-by-particle data: The detected particles are
separated in Mode 1 and Mode 2. The red points mark particles where the previous detected particle was
in the same mode. Black points mark particles where the previous detected particle was in the other size
mode. Panel a) represents a good mixing behavior and more particles (60.6%) were detected in Mode 2.
Panel b) indicates a clustered case where most of the particles are in Mode 1.

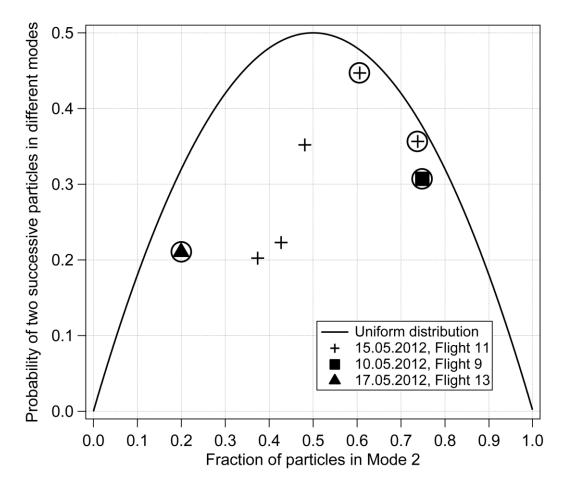




Figure 9. Statistical "proximity" analysis for the measured bimodal droplet size distributions. The black
line shows a theoretical, uniform distribution between both modes. The results from Figure 8a (i.e.
abscissa = 0.606 = 60.6% and ordinate = 0.447 = 44.7%) indicate that the measured bimodal size

distribution is close to uniform distribution. Four of the seven bimodal size distributions (marked by circles) show a good agreement with the theoretical, uniform distribution.

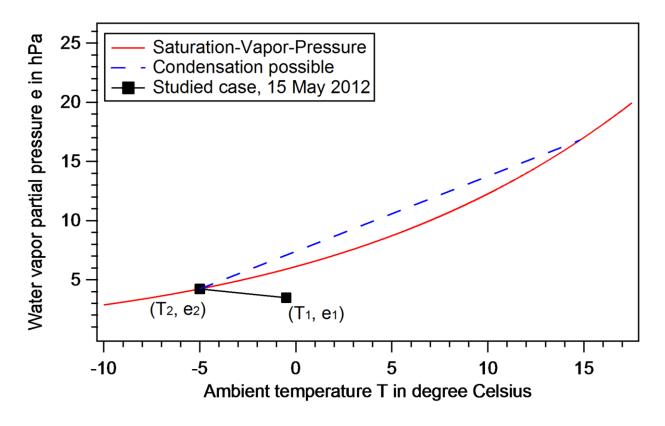
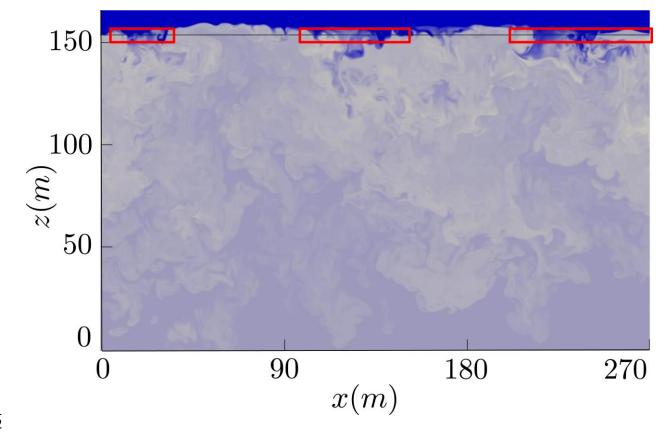


Figure 10. Isobaric mixing diagram for parcels of cloudy and a dry air: The red line denotes the water
vapor partial pressure for saturation, and the blue dashed line exemplifies a mixing line along which the
cloudy air parcel (T<sub>2</sub>, e<sub>2</sub>) could have been mixed with another parcel such that supersaturation results.
However, for the studied case from Flight 11 on 15 May 2012 (as represented by the short, straight, black

7 line) no conditions for supersaturation and for aerosol particle activation arise. (The thermodynamic

8 parameters of  $T_1 = -0.5^{\circ}C$ ,  $RH_1 = 59\%$ ,  $e_1 = 3.5$  hPa for dry air and  $T_2 = -5^{\circ}C$ ,  $RH_2 = 100\%$ ,  $e_2 = 4.2$  hPa

9 inside the cloud are based on the measurements.)



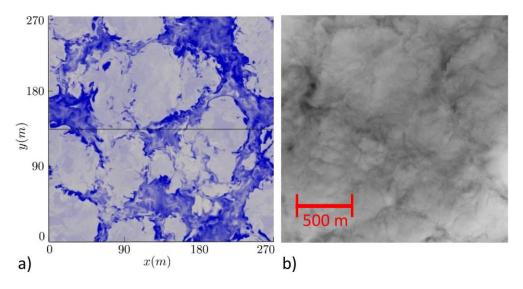
### 1 2

3 Figure 11. Results from Direct Numerical Simulation of the turbulence in the cloud top entrainment zone:

4 Vertical cross section of the calculated cloud liquid water with the color scale extending from 0.0 g/kg

5 (blue) to 0.325 g/kg (white). The black line indicates the simulated aircraft trajectory through the scenario

6 and the red rectangles mark the "cloud holes", i.e. air parcels mostly of cloud free air.





2 Figure 12. Panel a) DNS simulation of the horizontal cross section for cloud liquid water content. The

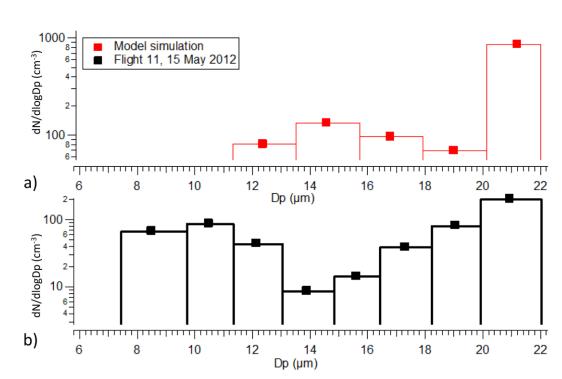
simulated aircraft trajectory is given by the black line and the scaling of the color code is the same as inFigure 11.

5 Panel b) Photograph, taken by a camera mounted at the fuselage of the POLAR 5 aircraft showing the top

6 of the boundary layer cloud deck which was observed during Flight 11. The scales of Panel a) and Panel

7 b) are different but the self-similarity of the structures is demonstrated qualitatively for the cloud

8 encountered in the real atmosphere and the simulated cloud liquid water patterns.



1
2 Figure 13. Panel a) A bimodal size distribution of the cloud droplets as obtained from the Direct

Numerical Simulation for the last 70 m of the simulated aircraft trajectory. Panel b) For comparisons the
measured bimodal size distribution from Flight 11 as also shown in Figure 5c.

- 5
- 6

### 1 References

- 2 Avramov, A., Ackerman, A. S., Fridlind, A. M., van Diedenhoven, B., Botta, G., Aydin, K., Verlinde, J.,
- 3 Korolev, A. V., Strapp, J. W., McFarquhar, G. M., Jackson, R., Brooks, S. D., Glen, A., and Wolde, M.:
- 4 Toward ice formation closure in Arctic mixed-phase boundary layer clouds during ISDAC, Journal of
- 5 Geophysical Research: Atmospheres, 116, D00T08, 10.1029/2011jd015910, 2011.
- Betts, A. K.: Non-precipitating cumulus convection and its parameterization, Quart. J. R. Met. Soc., 99,
  178-196, 1973.
- 8 Bierwirth, E., Ehrlich, A., Wendisch, M., Gayet, J. F., Gourbeyre, C., Dupuy, R., Herber, A., Neuber, R.,
- 9 and Lampert, A.: Optical thickness and effective radius of Arctic boundary-layer clouds retrieved from
- 10 airborne nadir and imaging spectrometry, Atmos. Meas. Tech., 6, 1189-1200, 10.5194/amt-6-1189-2013,
- **11** 2013.
- 12 Bretherton, C. S.: A theory for nonprecipitating moist convection between 2 parallel plates .1.
- 13 Thermodynamics and linear solutions, J. Atmos. Sci., 44 (14), 1809-1827, 1987.
- 14 Bretherton, C.S., Blossey, P. N. and Uchida, J.: Cloud droplet sedimentation, entrainment efficiency, and
- subtropical stratocumulus albedo. Geophys. Res. Lett., 34, L03813, doi:10.1029/2006GL027648, 2007.
- Curry, J. A., Rossow, W. B., Randall, D., and Schramm, J. M.: Overview of Arctic cloud and radiation
   characteristics, J. Climate, 9, 1731-1764, 1996.
- 18 Curry, J. A., Hobbs, P. V., King, M. D., Randall, D. A., Minnis, P., Isaac, G. A., Pinto, J. O., Uttal, T.,
- 19 Bucholtz, A., Cripe, D. G., Gerber, H., Fairall, C. W., Garrett, T. J., Hudson, J., Intrieri, J. M., Jakob, C.,
- 20 Jensen, T., Lawson, P., Marcotte, D., Nguyen, L., Pilewskie, P., Rangno, A., Rogers, D. C., Strawbridge,
- K. B., Valero, F. P. J., Williams, A. G., and Wylie, D.: FIRE Arctic Clouds Experiment, Bull. Amer.
- 22 Meteor. Soc., 81, 5-29, 1999.
- 23 de Lozar, A., and Mellado, J. P.: Cloud droplets in a bulk formulation and its application to buoyancy
- reversal instability, Quarterly Journal of the Royal Meteorological Society, 140, 1493-1504,
- **25** 10.1002/qj.2234, 2014.
- 26 de Lozar, A., and Mellado, J. P.: Direct Numerical Simulations of a Smoke Cloud–Top Mixing Layer as a
- Model for Stratocumuli, Journal of the Atmospheric Sciences, 70, 2356-2375, 10.1175/jas-d-12-0333.1,
  2013b.
- 29 Earle, M. E., Liu, P. S. K., Strapp, J. W., Zelenyuk, A., Imre, D., McFarquhar, G. M., Shantz, N. C. and
- Leaitch, W.R.: Factors influencing the microphysics and radiative properties of liquid-dominated Arctic
   clouds: Insight from observations of aerosol and clouds during ISDAC, J. Geophys. Res., 116, 16, 2011.
- 32 Ehrlich, A., Bierwirth, E., Wendisch, M., Gayet, J.-F., Mioche, G., Lampert, A., and Heintzenberg, J.:
- Cloud phase identification of Arctic boundary-layer clouds from airborne spectral reflection
- 34 measurements: test of three approaches, Atmos. Chem. Phys., 8, 7493-7505, 2008.

- 1 Ehrlich, A., Wendisch, M., Bierwirth, E., Herber, A., and Schwarzenböck, A.: Ice crystal shape effects on
- solar radiative properties of Arctic mixed-phase clouds Dependence on microphysical properties, Atmos.
- 3 Res., 88, 266-276, 2008a.
- Ehrlich, A., Wendisch, M., Bierwirth, E., Gayet, J.-F., Mioche, G., Lampert, A., Mayer, B.:Evidence of
  ice crystals at cloud top of Arctic boundary-layer mixed-phase clouds derived from airborne remote
- 6 sensing, Atmos. Chem. Phys., 9, 9401-9416, 2009.
- 7 Fan, J., Ovtchinnikov, M., Comstock, J. M., McFarlane, S. A., and Khain, A.: Ice formation in Arctic
- 8 mixed-phase clouds: Insights from a 3-D cloud-resolving model with size-resolved aerosol and cloud
- 9 microphysics, J. Geophys. Res., 114, D04205, 2009.
- Field, P. R., Wood, R. and Brown, P. R. A.: Ice Particle Interarrival Times Measured with a Fast FSSP, J.
  Atmos. Oceanic Technol., 20, 249-261, 2003.
- Field, P. R., Heymsfield, A. J. and Bansemer, A.: Shattering and Particle Interarrival Times Measured by
   Optical Array Probes in Ice Clouds, Amer. Meteor. Soc., 23, 1357-1371, 2006.
- Gerber, H., Frick, G., Malinowski, S. P., Brenguier, J. L., and Burnet, F.: Holes and entrainment in
   stratocumulus, J. Atmos. Sci., 62(2), 2005.
- Hansen, J., Ruedy, R., Sato, M., and Lo, K.: Global surface temperature change, Rev. Geophys., 48, 29,
  2010.
- 18 Huebert, B. J., Howell, S. G., Covert, D., Bertram, T., Clarke, A., Anderson, J. R., Lafleur, B. G.,
- 19 Seebaugh, W. R., Wilson, J. C., Gesler, D., Blomquist, B., and Fox, J.: PELTI: Measuring the Passing
- 20 Efficiency of an Airborne Low Turbulence Aerosol Inlet, Aerosol Science and Technology, 38, 803-826,
- **21** 2004.
- 22 Jackson, R. C., McFarquhar G. M., Korolev, A. V., Earle, M. E., Liu, P. S. K., Lawson, R. P., Brooks, S.,
- 23 Wolde, M., Laskin, A., Freer, M.: The dependence of ice microphysics on aerosol concentration in arctic
- 24 mixed-phase stratus clouds during ISDAC and M-PACE, J. Geophys. Res., 117, 20, 2012.
- 25 Jacob, D. J., Crawford, J. H., Maring, H., Clarke, A. D., Dibb, J. E., Emmons, L. K., Ferrare, R. A.,
- 26 Hostetler, C. A., Russel, P. B., Singh, H. B., Thompson, A. M., Shaw, G. E., McCauley, E., Pederson, J.
- 27 R., and Fisher, J. A.: The Arctic Research of the Composition of the Troposhere from Aircraft and
- 28 Satellites (ARCTAS) mission: design, execution, and first results, Atmos. Chem. Phys., 10, 5191-5212,
- 29 2010.
- Jensen, E. J., Lawson, P., Baker, B., Pilson, B., Mo, Q., Heymsfield, A. J., Bansemer, A., Bui, T. P.,
- 31 McGill, M., Hlavka, D., Heymsfield, G., Platnick, S., Arnold, G. T., and Tanelli, S.: On the importance of
- 32 small ice crystals in tropical anvil cirrus, Atmos. Chem. Phys., 9, 5519-5537, 2009.
- Kay, J. E., L'Ecuyer, T., Gettelman, A., Stephens, G., and O'Dell, C.: The contribution of cloud and radiation anomalies to the 2007 Arctic sea ice extent minimum, Geophys. Res. Lett., 35, 5, 2008.

- 1 Korolev, A. V.: Reconstruction of the Sizes of Spherical Particles from Their Shadow Images. Part I:
- 2 Theoretical Considerations., J. Atmos. Oceanic Technol., 24, 376-389, 2007.
- Korolev, A. V., Emergy, E., Creelman, K.: Modification and Tests of Particle Probe Tips to Mitigate
  Effects of Ice Shattering, J. Atmos. Oceanic Technol., 30, 690-708, 2013.
- 5 Korolev, A. V., Emery, E. F., Strapp, J. W., Cober, S. G., Isaac, G. A., Wasey, M., and Marcotte, D.:
- 6 Small ice particles in tropospheric clouds: fact or artifact? Airborne Icing Instrumentation Evaluation
- 7 Experiment Bull. Amer. Meteor. Soc. , 92, 967-973, 2011.
- Korolev, A. V., Field, P. R.: The Effect of Dynamics on Mixed-Phase Clouds: Theoretical Considerations,
  J. Atmos. Sci., 65, 66-86, 2008.
- 10 Lance, S., Brock, C. A., Rogers, D., and Gordon, J. A.: Water droplet calibration of the Cloud Droplet
- Probe (CDP) and in-flight performance in liquid, ice and mixed-phase clouds during ARCPAC, Atmos.
   Meas. Tech., 3, 1683-1706, 2010.
- Larson, V. E., Kotenberg, K. E., and Wood, N. B.: An analytic longwave radiation formula for liquid layer
   clouds, Mon. Wea. Rev., 135, 689-699, 2007.
- 15 Lathem, T. L., Beyersdorf, A. J., Thornill, K. L., Winstead, E. L., Cubison, M. J., Hecobian, A., Jimenez,
- 16 J. L., Weber, R. J., Anderson, B. E., and Nenes, A.: Analysis of CCN activity of Arctic aerosol and
- 17 Canadian biomass burning during summer 2008, Atmos. Chem. Phys. Discuss., 12, 24677-24733, 2012.
- Lawson, R. P., Baker, B. A., and Schmitt, C. G.: An overview of microphysical properties of Arctic
  clouds observed in May and July 1998 during FIRE ACE, J. Geophys. Res., 106, 14,989 915,014, 2001.
- 20 Lemke, P., J. Ren, R.B. Alley, I. Allison, J. Carrasco, G. Flato, Y. Fujii, G. Kaser, P. Mote, R.H. Thomas,
- and Zhang, T.: Observations: Changes in Snow, Ice and Frozen Ground. In: Climate Change 2007: The
- 22 Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the
- Intergovernmental Panel on Climate Change [Solomon, S., D. Qin, M. Manning, Z. Chen, M. Marquis,
   K.B. Averyt, M. Tignor and H.L. Miller (eds.)]. , Cambridge University Press, Cambridge, United
- 24 Kingdom and New York, NY, USA., 2007.

- 28 McFarquhar, G. M., Ghan, S., Verlinde, J., Korolev, A., Strapp, W., Schmid, B., Tomlinson, J. M., Wolde,
- 29 M., Brooks, S. D., Cziczo, D., Dubey, M. K., Fan, J., Flynn, C., Gultepe, I., Hubbe, J., Gilles, M. K.,
- 30 Laskin, A., Lawson, P., Leaitch, W. R., Liu, P., Liu, X., Lubin, Dan., Mazzoleni, C., MacDonald, A.-M.,
- 31 Moffet, R. C., Morrison, H., Ovchinnikov, M., Shupe, M. D., Turener, D. D., Shaocheng, X., Zelenyuk,
- 32 A., Bae, K., Freer, M. and Glen, A.:Indirect and Semi-Direct Aerosol Campaign, Bull. Amer. Meteor.
- **33** Soc.,92,183-201, 2011.
- 34 McFarquhar, G. M., Zhang, G., Poellot, M. R., Kok, G. L., McCoy, R., Tooman, T., Fridlind, A., and
- 35 Heymsfield, A. J.:Ice properties of single-layer stratocumulus during the Mixed-Phase Arctic Cloud
- 36 Experiment, J. Geophys. Res., 112, 19, 2007.

<sup>Lihavainen, H., Kerminen, V.-M., and Remer, L. A.: Aerosol-cloud interaction determined by both in situ
and satellite data over a northern high-altitude site, Atmos. Chem. Phys., 10, 10987-10995, 2010.</sup> 

- 1 Mellado, J. P.: The evaporatively driven cloud-top mixing layer, J. Fluid Mech., 660, 5-36, 2010.
- 2 Morrison, H., Pinto, J. O., Curry, J. A., and McFarquhar, G. M.: Sensitivity of modelled arctic mixed-
- phase stratocumulus to cloud condensation and ice nuclei over regionally varying surface conditions, J.
  Geophys. Res., 113, D05203, 2008.
- Palm, S. P., Strey, S. T., Spinhirne, J., and Markus, T.: Influence of Arctic sea ice extent on polar cloud
  fraction and vertical structure and implications for regional climate, J. Geophys. Res., 115, 2010.
- The first of the f
- 7 Perovich, D. K., Andreas, E. L., Curry, J. A., Eiken, H., Fairall, C. W., Grenfell, T. C., Guest, P. S.,
- 8 Intrieri, J., Kadki, D., Lindsay, R. W., McPhee, M. G., Morison, J., Moritz, R. E., Paulson, C. A., Pegau,
- 9 W. S., Persson, P. O. G., Pinkel, R., Richter-Menge, J. A., Stanton, T., Stern, H., Sturm, M., Tucker, W.
- 10 B., and Uttal, T.: Year in Ice Gives Climate Insights, EOS, 80, 481-492, 2000.
- Perovich, D. K., J. A. Richter-Menge, K. F. Jones, and Light, B.: Sunlight, water, and ice: Extreme Arctic
  sea ice melt during the summer of 2007, Geophys. Res. Lett., 35, 2008.
- Pettersen, S.: On the causes and the forecasting of the California fog. Bull. Amer. Meteor. Soc. 19, 49-55,14 1938.
- Platnick, S.: Vertical photon transport in cloud remote sensing problems, J. Geophys. Res., 105, 2291922935, 2000.
- Pruppacher, H. R., and Klett, J. D.: Microphysics of Clouds and Precipitation, Springer Dordrecht
   Heidelberg London New York, 954, 2010.
- 19 Schlenczek, O., Fugal, J. P., Bower, K. N., Crosier, J., Flynn, M. J., Lloyd, G. and Borrmann, S.:
- Properties of pure ice clouds in an alpine environment, Extended abstract, Amer. Meteor. Soc. 14th
  Conference on Cloud Physics, July 07-11, Boston, USA, 2014.
- Schweiger, A. J., and Lindsay, R. W.: Relationships between Arctic Sea Ice and Clouds during Autumn, J.
  Climate, 21, 4799-4810, 2008.
- Shupe, M. D., and Intieri, J. M.: Cloud radiative forcing of the Arctic surface: The influence of cloud
   properties, surface albedo, and solar angle, J. Climate, 17, 616-628, 2004.
- Siems, S. T. and Bretherton C. S. : A numerical investigation of cloud-top entrainment instability and
   related experiments, Quart. J. Roy. Meteor. Soc., 118, 787-818, 1992.
- Spuler, S. M., and Fugal, J.: Design of an in-line, digital holographic imaging system for airborne
   measurement of clouds, Appl. Opt., 50, 1405-1412, 2011.
- 30 Stevens, B.: Atmospheric Moist Convection, Annu. Rev. Earth Planet. Sci., 33, 605-643, 2005.
- 31 Stevens, B., Moeng, C.-H., Ackerman, A. S., Bretherton, C. S., Chlond, A., de Roode, S., Edwards, J.,
- 32 Golaz, J.-C., Jiang, H., Khairoutdinov, M., Kirkpatrick, M. P., Lewellen, D. C., Lock, A., Müller, F.,

- 1 Stevens, D. E., Whelan, E. and Zhu, P.: Evaluation of Large-Eddy Simulations via Observations of
- 2 Nocturnal Marine Stratocumulus, Mon. Wea. Rev., 133, 1443-1462, 2005.

Struthers, H., Ekman, A. M. L., Glantz, P., Inversen, T., Kirkevåg, A., Mårtensson, E. M., Seland, Ø., and
Nilsson, E. D.: The effect of sea ice loss on sea salt aerosol concentrations and the radiative balance in the

- 5 Arctic, Atmos. Chem. Phys., 11, 3459-3477, 2011.
- 6 Verlinde, J., Herrington, Y., McFarquhar, G. M., Yannuzzi, V. T., Avramov, A., Greenberg, S., Johnson,
- 7 N., Zhang, G., Poellot, M. R., Mather, J. H., Turner, D. D., Eloranta, E. W., Zak, B. D., Prenni, A. J.,
- 8 Daniel, J. S., Kok, G. L., Tobin, D. C., Holz, R., Sassen, K., Spangenberg, D., Minnis, P., Tooman, T. P.,
- 9 Ivey, M. D., Richardson, S. J., Bahrmann, C. P., Shupe, M., DeMott, P. J., Heymsfield, A. J., and
- 10 Schofield, R.: The Mixed-Phase Arctic Cloud Experiment, Bull. Amer. Meteor. Soc, 88, 205-221, 2007.
- 11 Wendisch, M., and Brenquier, J.-L.: Airborne Measurements for Environmental Research: Methods and
- 12 Instruments, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim, Germany, 655, 2013.
- 13 Wendisch, M., P. Yang, and A. Ehrlich: Amplified climate changes in the Arctic: Role of clouds and
- 14 atmospheric radiation. Sitzungsberichte der Sächsischen Akademie der Wissenschaften zu Leipzig.
- 15 Mathematisch-Naturwissenschaftliche Klasse, S. Hirzel Verlag, Stuttgart/Leipzig, 132, 1-34, 2013.
- 16