

## Contribution of mixed-phase boundary layer clouds to the termination of ozone depletion events in the Arctic

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[1] During the springtime, ozone depletion events (ODEs) are frequently observed in the Arctic boundary layer. While the chemical reactions associated with the ODEs are understood, the processes responsible for their termination remain unclear. Previous studies proposed that wind shear above the Arctic boundary layer promotes enough vertical mixing to transport ozone-richer air from aloft to the nearly ozone-devoid surface and thus terminates the ODEs. In addition, ozone-richer air masses from mid-latitude regions can migrate to the high Arctic and replenish the Arctic boundary layer with ozone. In the present study, a new mechanism related to mixed-phase boundary layer clouds is proposed as a key contributor to the termination of the ODEs. A single-layer stratocumulus cloud observed over Barrow, Alaska (AK) on April 8, 2008 and its effect on the ODEs is simulated using high-resolution WRF/Chem model. One key finding of this investigation is that the cloud-top radiative cooling can induce strong downdrafts and updrafts. These downdrafts associated with mixed-phase boundary layer clouds can transport ozone-richer air from aloft to the surface, heralding the termination of ODEs. **Citation:** Hu, X.-M., F. Zhang, G. Yu, J. D. Fuentes, and L. Wu (2011), Contribution of mixed-phase boundary layer clouds to the termination of ozone depletion events in the Arctic, *Geophys. Res. Lett.*, *38*, L21801, doi:10.1029/2011GL049229.

### 1. Introduction

[2] Tropospheric ozone ( $O_3$ ) is one of the most important atmospheric constituents due to its role as a greenhouse gas, a secondary pollutant, and a contributor of the oxidation capacity of the atmosphere. Springtime anomalously low  $O_3$  mixing ratios in the Arctic atmospheric boundary layer (ABL) were first reported in the 1980s [Oltmans, 1981; Bottenheim *et al.*, 1986]. Such yearly  $O_3$  depletion events (ODEs) might have dramatic and unpredictable consequences and impact the global tropospheric  $O_3$  budget [Simpson *et al.*, 2007]. The Arctic ODEs have therefore received extensive attention by the research community [Hopper *et al.*, 1998; Bottenheim *et al.*, 2009; Jacobi *et al.*, 2010]. It is now generally accepted that the halogen (especially bromine) activation and the subsequent reactions can

cause the onset of ODEs [Simpson *et al.*, 2007; Piot and von Glasow, 2008]. While the basic features of the ODEs seem understood, several questions remain unclear [Jacobi *et al.*, 2010]. First, it is still not known why the ODEs are only observed in spring, but not in other seasons [Lehrer *et al.*, 2004]. Second, the spatial extent of the ODEs is not well established [Jacobi *et al.*, 2010]. Third, the processes responsible for the termination of the ODEs are poorly understood to separate the influences of transport (vertical or horizontal) and chemical reactions [Gong *et al.*, 1997; Hopper *et al.*, 1998; Strong *et al.*, 2002; Bottenheim *et al.*, 2009; Jacobi *et al.*, 2010]. The goal of the present manuscript is to investigate the processes related to the third question.

[3] Previous studies [e.g., Gong *et al.*, 1997; Strong *et al.*, 2002] indicated that wind shear just above the Arctic ABL can contribute to the transport of  $O_3$ -richer air from aloft to the surface, thus contributing to the termination of the ODEs. Jacobi *et al.* [2010] reported that northward moving lows may bring  $O_3$ -rich air from mid-latitude regions to the Arctic, leading to the termination of the ODEs. In this study, we propose another mechanism for the termination of the ODEs. We investigated the ODEs in Barrow, AK in spring of 2009 (coinciding with the field campaign of Ocean-Atmosphere-Sea Ice-Snowpack Interactions in Polar Regions (OASIS) 2009). During the recovery periods of some ODEs (e.g., March 16, 2009), surface wind remained calm, wind shear in the boundary layer stayed low, and air mass came from the Arctic Ocean. Therefore, horizontal transport was less likely the reason for the termination of ODEs. Additionally, vertical mixing was unlikely induced solely by wind shear as the cases reported by Strong *et al.* [2002]. Instead, the boundary layer clouds (observed on March 16, 2009) likely generated vertical mixing during the recovery period of the ODE. Arctic boundary layer clouds strongly impact the vertical structure of the ABL through cloud-top radiative cooling and generation of negative buoyancy [Pinto, 1998; Morrison and Pinto, 2006]. Thus, we hypothesize that the circulation patterns associated with Arctic boundary layer clouds contribute to the termination of the ODEs. Heretofore, the role of Arctic boundary layer clouds in the ODEs has not been investigated. Using high-resolution three-dimensional model simulations, the present study provides evidence about the contribution of clouds on the replenishment of  $O_3$  in the Arctic ABL following a major ODE.

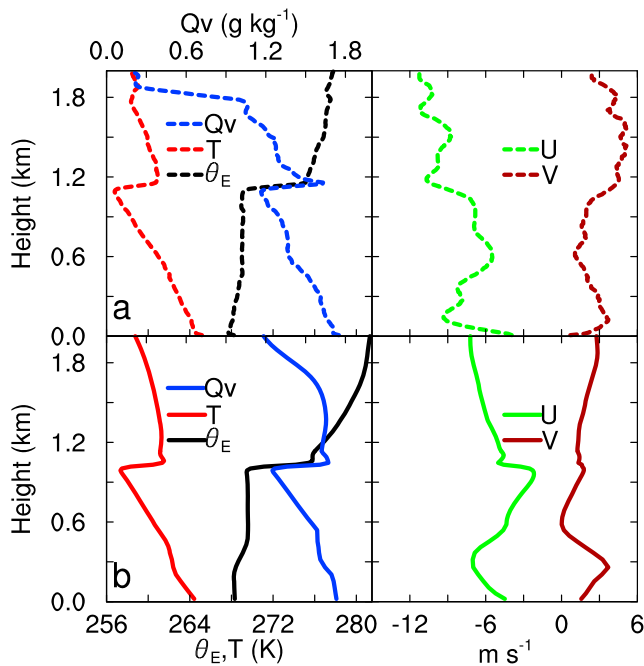
### 2. Model Setup and Experimental Design

[4] The Arctic ABL clouds present challenges to modelers because of their unique types and characteristics compared to clouds in lower-latitude regions. Previous numerical

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**Figure 1.** Profiles of water vapor mixing ratio ( $Q_v$ ), temperature ( $T$ ), equivalent potential temperature ( $\theta_E$ ),  $U$ , and  $V$  (a) measured at 17.6 UTC and (b) simulated at 18:00 UTC on April 8, 2008 at (71.33°N, 156.61°W).

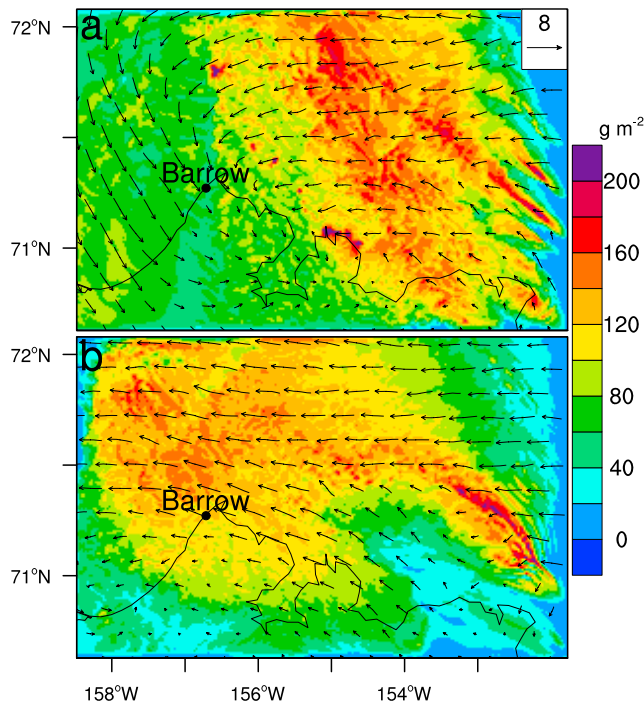
modeling studies of boundary layer stratocumulus clouds have indicated considerable spread in model solutions [Klein *et al.*, 2009; Morrison *et al.*, 2011]. To successfully simulate Arctic boundary layer clouds, a realistic treatment of ice microphysics and proper model resolution are critical [Klein *et al.*, 2009]. In this study, a single-layer stratocumulus cloud observed over the North Slope of Alaska on April 8, 2008 is simulated using the Weather Research and Forecasting model with Chemistry (WRF/Chem) version 3.2.1. Most model configurations follow those used by Solomon *et al.* [2011], in which the same cloud case was successfully simulated. We describe here only the configurations different from those described by Solomon *et al.* [2011] and those that are essential for the simulation of  $O_3$  distribution. Three model domains with horizontal grid spacings of 25 km, 5 km, and 1 km used in this study are the same as the first three nested domains employed by Solomon *et al.* [2011]. We do not nest the finer resolution domains since the simulation in the 1-km resolution domain captures some important characteristics (e.g., vertical extent and magnitudes of eddies associated with cloud) of the Arctic mixed-phase cloud [Solomon *et al.*, 2009, 2011]. Such characteristics play dominant roles in transporting  $O_3$ -richer air downward to terminate the ODEs. Thus the 1-km resolution simulation appears to be adequate to investigate the impact of mixed-phase cloud on vertical transport of  $O_3$ . Eighty-five pressure levels below 800 hPa are adopted as by Solomon *et al.* [2011] to better resolve the mixing and entrainment in the mixed-layer and entrainment zone. The National Centers for Environmental Prediction (NCEP) global forecast system (GFS) final (FNL) operational global analyses are used for the initial conditions and boundary conditions of all meteorological variables. The model spin-up time is 12 hours. The simulation starting from 12:00

Coordinated Universal Time (UTC), April 8 is used in the analysis. Since the  $O_3$  change due to chemical reactions in the Arctic is much slower than that due to transport [Gong *et al.*, 1997],  $O_3$  is represented by a tracer in WRF/Chem to investigate its transport in the presence of boundary layer stratocumulus. Initial and boundary conditions for  $O_3$  are set using  $O_3$  profiles obtained during typical ODEs, in which  $O_3$  is almost depleted in the lower 300 m near the surface while  $O_3$  is around 45 ppbv in the free troposphere [Gong *et al.*, 1997; Jacobi *et al.*, 2010]. Such setting of boundary conditions of  $O_3$  eliminates the possible contribution of advection to the replenishment of  $O_3$  following the ODE.

### 3. Results

[5] In Figure 1, the simulated environmental conditions at Barrow, AK at 18:00 UTC are compared with the nearest-in-time sounding taken at Barrow at 17.6 UTC, April 8, 2008. The model simulation realistically reproduced a well-mixed boundary layer as indicated by the profile of equivalent potential temperature. However, the simulated boundary layer was slightly lower than the observation by around 100 m. The simulation also captured the temperature and humidity inversion at the top of the boundary layer. Such meteorological conditions were consistent with the observations made for typical Arctic cloud-topped mixed layers [Curry *et al.*, 2000]. The humidity and temperature inversion above the cloud-topped mixed layer contributed to the persistence of the cloud deck by inhibiting evaporation associated with entrainment mixing at the cloud top [Curry *et al.*, 2000]. Such humidity inversion was a unique feature for Arctic boundary layer clouds. The simulated wind profile was qualitatively similar to the observation: strong shear in the surface layer and at cloud top, weaker winds within the cloud and sub-cloud layer. The turbulent kinetic energy budget was analyzed for the same case in Solomon *et al.* [2011]. Results indicated that wind shear generated turbulence within the entrainment zone (at cloud top) while wind shear played a minor role in turbulence production within the mixed layer [Solomon *et al.*, 2011]. Instead, radiative cooling played a dominant role in turbulence production in the mixed layer.

[6] The simulated spatial distributions of condensed water path in Figure 2 implied that the simulated cloud passed the Barrow site after 12:00 UTC. After the cloud moved to Barrow, the simulated surface  $O_3$  increased by around 15 ppbv during the period from 12:00 to 16:00 UTC (Figure 3a). By 23:00 UTC, the surface  $O_3$  increased to 28 ppbv (Figures 3a and 3b). Thus, it appears the presence of the cloud plays an important role in replenishing boundary layer  $O_3$  and terminating the ODE. Note that the observed termination of the ODE on April 8, 2008 occurred around 10:00 UTC. Simulated later termination of the ODE is largely due to the later formation of the mixed-phase cloud. This idealized modeling study intends to isolate the contribution of mixed-phase clouds to the termination of ODEs, discrepancy between the observed  $O_3$  variation and simulated  $O_3$  variation is expected, especially given the use of idealized initial and boundary conditions for the  $O_3$ . The discrepancy may be also partially due to the cold-start of the model and the cloud microphysics, which is key to the evolution of the mixed-phase clouds.

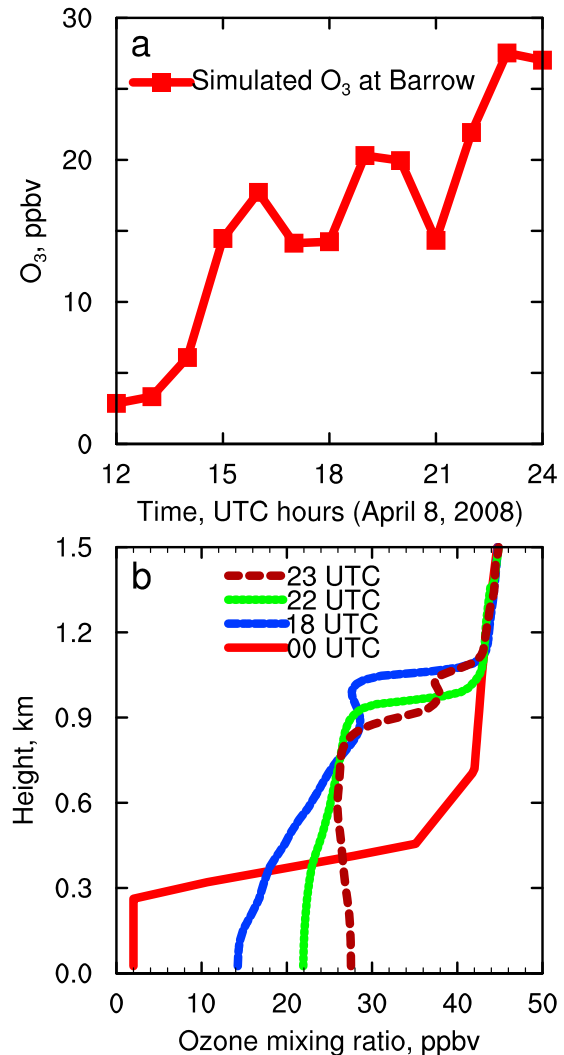


**Figure 2.** Column integrated condensed water (cloud water plus cloud ice) path at (a) 12:00 UTC (before cloud passing Barrow) and (b) 18:00 UTC (with presence of cloud at Barrow) on April 8, 2008. Wind vectors show the wind field at 10 m above ground.

[7] The vertical variation of cloud ice content, vertical velocity, and  $O_3$  are shown in Figure 4. A single-layer stratocumulus cloud was successfully simulated in the boundary layer. Liquid water resided at the cloud top, which was located between 1.15 km and 0.9 km along the west-east cross section through Barrow (figure not shown). The cloud top was at the base of the temperature inversion and its temperature ranged from  $-17$  to  $-13^\circ\text{C}$  at Barrow. Cloud ice formed within the liquid cloud layer. Thus, the cloud was in mixed phase. The resolved downdrafts and updrafts (Figure 4b) were highly correlated with the vertical distribution of cloud ice (Figure 4a). In the updrafts more cloud ice formed while in the downdraft cloud ice was reduced due to sublimation. The cloud liquid water path dominated cloud ice water path. The total column integrated liquid water accounted for over 90% of the total cloud water (liquid plus ice). The model simulated the mixed-phase stratocumulus starting at 11:00 UTC with a cloud top at 1.2 km at Barrow. Thereafter, the cloud top slowly descended, which was consistent with observation (figure not shown). Strong infrared cooling occurred at the cloud top. Because solar heating was low due to low Sun elevation angle in the Arctic, the cloud produced a net cooling effect [Curry, 1986]. The cloud top temperature at Barrow dropped by  $\sim 5$  K during the period from 7:00 to 20:00 UTC. The strong infrared cooling near the liquid cloud top generated enough turbulence to promote downdrafts and compensating updrafts (Figure 4b). Cloud top radiative cooling was shown to dominantly trigger the turbulence structure of Arctic ABL stratus clouds [Finger and Wendling, 1990]. Such radiative-cooling triggered downdrafts were strong enough to reach the surface. Such results were consistent

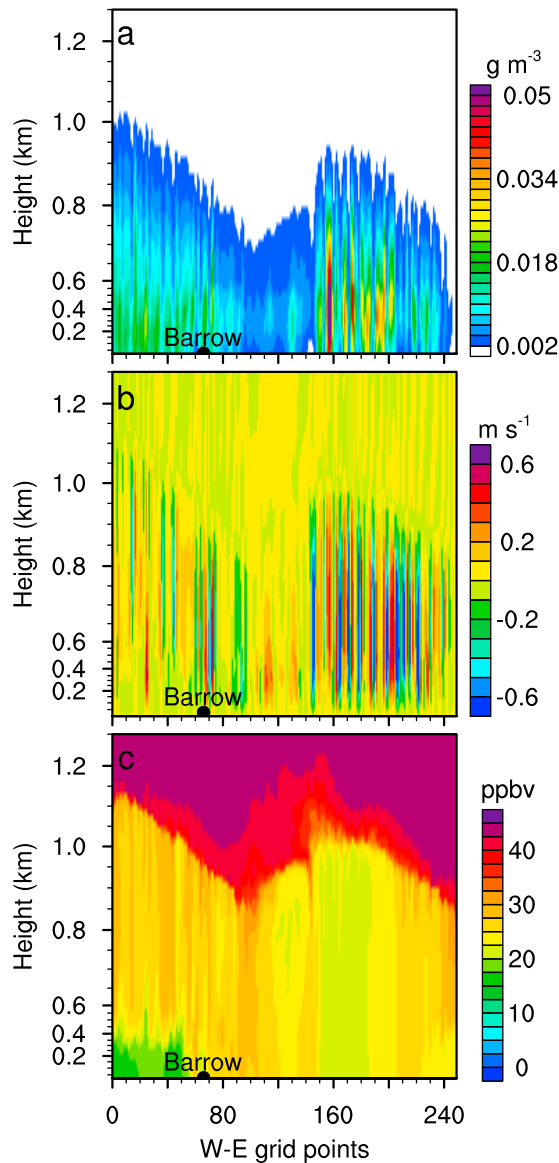
with the previous findings [e.g., Curry, 1986; Wang *et al.*, 2001; Zuidema *et al.*, 2005]. Before the cloud moved to Barrow, there was a strong temperature inversion near the surface at Barrow (figure not shown). The inversion disappeared after the cloud moved into the region (see the equivalent potential temperature profile in Figure 1) due to the fact that the downdrafts transported warmer air from the upper boundary layer to the surface. Thus, a cloud-top-cooled mixed-layer that extends from the cloud top to the surface forms. Different from a surface-heated convective boundary layer, where downdrafts are much weaker than updrafts, the compensating updrafts have about the same strength ( $\sim 0.6 \text{ m s}^{-1}$ ) as the downdrafts in this Arctic stratocumulus case (Figure 4b). The simulated composite structure was consistent with idealized stratus-topped boundary layer clouds reported by Moeng and Schumann [1991], in which the turbulence was maintained solely by cloud-top radiative cooling. It was also consistent with mixed-phase stratus topped boundary layer reported by Morrison and Pinto [2006].

[8] In addition to modifying the boundary layer structure, cloud downdrafts and updrafts significantly modified the



**Figure 3.** (a) Simulated surface  $O_3$  mixing ratio and (b) initial vertical  $O_3$  profile at 00:00 UTC and simulated  $O_3$  profiles at 18:00, 22:00, and 23:00 UTC at Barrow, AK.





**Figure 4.** Simulated vertical structure of (a) cloud ice content, (b) vertical velocity, and (c)  $\text{O}_3$  for the west-east cross section passing through Barrow, AK at 22:00 UTC on April 8, 2008.

vertical variability of atmospheric constituents. Since the chemical lifetime of  $\text{O}_3$  was much longer than the time scale of vertical mixing due to downdrafts and updrafts,  $\text{O}_3$  vertical distribution was dominated by the transport due to downdrafts and updrafts. The initial  $\text{O}_3$  profile was set to an observed profile during a typical ODE, in which  $\text{O}_3$  had higher mixing ratios above 300 m than at the  $\text{O}_3$ -depleted boundary layer. The vertical mixing due to downdrafts and updrafts extended to 0.9–1.1 km above the surface. Thus, downdrafts likely transported  $\text{O}_3$ -richer air to replenish  $\text{O}_3$  near the surface. The vertical  $\text{O}_3$  profiles at Barrow during the ODEs and in the presence of cloud are shown in Figure 3b. The  $\text{O}_3$  near the surface was replenished to around 28 ppbv at 23:00 UTC while the  $\text{O}_3$  mixing ratio between 0.4 and 1 km decreased. The change of vertically integrated  $\text{O}_3$  amount in the lower 1.2 km at Barrow from 00:00 to 23:00 UTC was less than 5%, which confirmed that the  $\text{O}_3$ -richer air

replenishing the surface  $\text{O}_3$  came from the upper layers due to downward transport induced by cloud top radiative cooling. This mechanism for the termination of ODEs is consistent with the one-dimensional (1D) model simulation reported by *Piot and von Glasow* [2008]. The inherent limitations of the 1D simulation and its results by *Piot and von Glasow* [2008] (e.g., a few assumptions regarding the initial, boundary, and forcing conditions were made for an ideal case) hindered a wide acceptance of this new mechanism. The current study confirms the ODE termination mechanism proposed by *Piot and von Glasow* [2008] with details of the cloud phases and dynamics of a real cloud case revealed from a full three-dimensional WRF/Chem simulation.

#### 4. Discussion and Conclusions

[9] The present investigation illustrates that the mixed-phase clouds commonly occurring in the Arctic may contribute to the vertical redistribution of  $\text{O}_3$  and the termination of ODEs. The mixed-phase clouds can impact the structure of the boundary layer through the influence of cloud-top radiative cooling. Downdrafts and compensating updrafts induced by the cloud-top radiative cooling can be sufficiently strong to reach the surface. The averaged vertical velocity in the presence of clouds may be as large as  $0.6 \text{ m s}^{-1}$  in the mixing layer. The vertical mixing associated with cloud updrafts and downdrafts triggered by the clouds can mix the free tropospheric  $\text{O}_3$ -richer air downward to replenish the  $\text{O}_3$  near the surface, thereby contributing to the termination of ODEs.

[10] Note that not all the downdrafts of mixed-phase clouds could reach the surface. The vertical extent of cloud induced mixing depends on cloud top radiative cooling and cloud-base stabilization [*Komurcu*, 2011]. Cloud top radiative cooling of the Arctic mixed-phase clouds is dominated by liquid water [*Pinto*, 1998]. Several factors (e.g., ice nuclei concentration, ice formation mechanism, and crystal habits) could affect the liquid water content in clouds [*Avramov and Harrington*, 2010; *Komurcu*, 2011]. Cloud-base stabilization is modulated by the degree of the ice growth and precipitation [*Harrington and Olsson*, 2001]. Whether mixed-phase clouds could generate strong enough downdrafts to terminate ODEs near the surface depends on the complex cloud microphysics and dynamic processes as demonstrated by the sensitivity of strength of cloud circulation (downdrafts and updrafts) reported by *Komurcu* [2011].

[11] The turbulent mixing in the boundary layer due to cloud top radiative cooling was reported many years ago [*Pinto*, 1998]. However, its implication for Arctic  $\text{O}_3$  redistribution is not widely realized yet. The new mechanism for the termination of ODEs reported in this study may have important implications for understanding the duration and the frequency of ODEs in the presence of the Arctic mixed-phased clouds. Observations indicate the Arctic region is cloudy about 85% of the year and mixed-phase clouds dominate the low-level types during the colder three-quarters of the year [*Intrieri et al.*, 2002; *Verlinde et al.*, 2007]. This study also emphasizes that it is necessary for models to adequately simulate mixed-phase boundary layer clouds to reproduce the Arctic ODEs.

[12] The Arctic low-level clouds occurred more frequently and have wider coverage in summer and fall than in spring [*Finger and Wendling*, 1990; *Intrieri et al.*, 2002;

Wang and Key, 2005]. Such seasonal variation of the Arctic low-level cloud amounts appears to be related to the temperature dependence of ice-phase microphysical processes [Beesley and Moritz, 1999]. A monthly averaged cloud occurrence could be as high as 95% in summer and fall [Intrieri et al., 2002]. The low-level clouds play an important role in shaping the temperature profile of the Arctic lower troposphere through vertical mixing induced by cloud-top radiative cooling and complex long-wave and short-wave radiation. The preponderance of low-level clouds in summer and fall can weaken or eliminate the strong surface temperature inversion [Beesley and Moritz, 1999], which is thought to be one of the prerequisites for the occurrence of ODEs [Lehrer et al., 2004; Bottenheim et al., 2009].

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