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Key Points:
- A mechanism of vertical transport of O3 within a cloud topped boundary layer is revealed using measurements and simulations.
- The contribution from vertical transport to surface O3 increases with cloud liquid water path, with maximum surface O3 enhancing by 30 ppb at nighttime.

Correspondence to:
X.-M. Hu and T. Zhao,
tlzho@nuist.edu.cn

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Abstract
Nighttime ozone (O3) reserved over the stable boundary layer (SBL) could contribute significantly to surface O3 levels. While many studies have revealed that daytime convective eddies and turbulence induced by nighttime low-level jets can mix O3-rich air over the SBL down to the surface, increasing nocturnal surface O3 within the cloud topped boundary layer has rarely been examined. In this study, we used measurements and a single-column photochemistry model to investigate O3 variations within the nighttime cloud topped boundary layer over the Sichuan Basin, which is the cloudiest region in southwest China. The Santa Barbara Distort Atmospheric Radiative Transfer radiation model was coupled into the single-column photochemistry model to investigate the nighttime radiative effects of clouds. The results showed that the nocturnal cloud top radiative cooling generated turbulence and enhanced vertical mixing below the cloud top layer. The cloud-driven turbulent eddies even penetrated through the SBL to the surface. Consequently, it weakened the decoupling of the SBL from the residual layer. O3-rich air over the SBL was entrained downwards to the surface, decreasing O3 aloft and increasing surface O3 at nighttime. The cloud top radiative cooling rates were enhanced with the increase in the cloud liquid water path. Higher cooling rates produced stronger turbulent transport of O3, leading to higher nocturnal surface O3 levels. The turbulent transport of O3 induced by cloud top radiative cooling is revealed as an important mechanism of vertical transport of O3 within a cloud topped boundary layer, contributing to the maximum nocturnal surface O3, which has great implications for understanding atmospheric O3 variation.

1. Introduction

Ozone (O3) reserved over the stable boundary layer (SBL) contributes to surface O3 buildup in the redeveloping planetary boundary layer (PBL) (Zhang & Rao, 1999, 1998). After sunset, due to formation of SBL, surface O3 concentrations typically reach minimum levels by nitrogen oxide (NO) titration and dry deposition (Sillman, 1999; Xie et al., 2016). In contrast, the residual layer (RL) above the SBL remained a high-level O3 due to the lack of O3 consumption (Neu et al., 1994). On the following day, the high-level O3 within the RL could be entrained downward by surface-driven turbulence in redeveloping PBL, also known as the mixing layer (ML), and consequently, contribute significantly to O3 buildup at ground level in the morning (Aneja et al., 2000; Hu et al., 2018; Klein et al., 2014; Tong et al., 2011). This process accounts for 60%–70% of the subsequent daytime O3 maximum (Morris et al., 2010).

The vigorous turbulence induced by the low-level jets (LLJs) could mix O3-rich air in RL down to the surface in the nighttime, which consumed RL O3 and reduced contribution to the surface O3 maximum on the following day. Hu et al. (2012) indicated that turbulent mixing generated by LLJs weakened the decoupling of SBL and RL. This facilitated the transport of O3 in RL down to the surface within nocturnal PBL, which reduced the increase in surface O3 in the following morning by approximately 8 ppb (Hu, Klein, & Xue, 2013; Hu, Klein, Xue, et al., 2013).

The cloud layer could also produce strong turbulence, but there are few concerns regarding its impact on nighttime O3 vertical variability in RL. Turbulence is generated by cloud top longwave radiative cooling and evaporative cooling, where the radiative cooling effect is the dominant force in the nocturnal cloud topped boundary layer (Vanzanten, 2002). Such turbulence may even penetrate down to the surface with negative
vertical velocity skewness, indicating that the downdrafts are typically narrower and more intense than the updrafts. This is called an “upside-down” version of surface-driven turbulence (Hogan et al., 2009; Mahrt & Vickers, 2002; Nicholls, 1984; Sorbjan & Uliasz, 1999). Thus, strong turbulence could exert an important impact on the vertical distribution of air pollutants at nighttime (Hu et al., 2011; Wang & Albrecht, 1994). Many studies have revealed that cloud top radiative cooling produces strong vertical turbulent mixing below the cloud top layer using large-eddy models (Moeng, 1986, 1987; Sorbjan & Uliasz, 1999). However, few concerns the impacts on SBL structures and vertical diffusion of air pollutants in the nocturnal cloud topped boundary layer, especially for turbulent transport of \( \text{O}_3 \)-rich air aloft.

The Sichuan Basin (SCB) is the cloudiest region in southwest China, where it is worth studying the cloud top radiative effect on the vertical distribution of \( \text{O}_3 \). Over SCB, the annual cloud fraction is over 80%, and higher cloud fractions occur frequently at night (Cai et al., 2017; Jia et al., 2013; Qian et al., 2007; Y. Zhang et al., 2019). Single-layer clouds are commonly observed in SCB, where the frequency is greater than 60% (Cai et al., 2017). Thus, the SCB is an ideal region for studying the cloud top radiative effect on the vertical distribution of \( \text{O}_3 \).

In this study, a cloudy case without precipitation over the SCB was selected to explore the underlying mechanism related to turbulent transport of \( \text{O}_3 \) in the nocturnal cloud topped boundary layer. In addition, a high-level \( \text{O}_3 \) event was chosen to verify the impacts of cloud variations on the SBL stability and \( \text{O}_3 \) vertical mixing based on sensitivity simulations. The rest of this paper is organized as follows: Section 2 describes the measurements and models. Section 3 presents the turbulent transport of \( \text{O}_3 \) below the nocturnal cloud topped boundary layer and the modeling results. The discussion and conclusions are summarized in Section 4.

### 2. Methods

The observations were performed in Chengdu (CD; 30.42°N, 103.50°E) and Deyang (DY; 31.09°N, 104.10°E) over the SCB. Both sites are surrounded by flat terrain and agricultural land. We used a surface flux observation system to measure the near-surface stability variations within the nocturnal cloud topped boundary layer. The system consists of a data collector (CR3000), a gradient measurement subsystem (HMP45C), a radiation measurement subsystem (CNR1, IRR-P), a three-dimensional supersonic anemometer (CSAT3), and an infrared gas analyzer of \( \text{CO}_2 \) (LI7500). Measurements made at these two surface energy balance sites also included near-surface meteorological parameters, including 2 m air temperature, 10 m wind speed (WS), and 10 m wind direction (WD). To obtain the PBL structures, a tethersonde system including a tethered meteorological radiosonde (TTS111, Vaisala) and an ozonesonde (TTO111, Vaisala) was used to measure the vertical distributions of air temperature, air pressure, relative humidity (RH), wind speed, wind direction, and \( \text{O}_3 \) mixing ratio in the atmosphere over CD at 14:00, 18:00, and 20:00 on January 20, 2017 (all times mentioned in this paper are local time). Information about the above-mentioned sensors is listed in Table 1. The air quality monitoring data used in this study, including hourly mass concentrations of \( \text{O}_3 \), \( \text{NO}_2 \), and \( \text{PM}_{2.5} \), were sourced from the Ministry of Ecology and Environment of China (http://www.mee.gov.cn).

| Index | Sensor name | Manufacturer                  | Observation element                                      |
|-------|-------------|-------------------------------|----------------------------------------------------------|
| 1     | CR3000      | Campbell Scientific Inc., USA | Control the sensors and collect data                     |
| 2     | HMP45C      | Vaisala, Finland              | Air temperature and relative humidity (RH)               |
| 3     | CNR1        | Kipp and Zonen, Netherlands   | Four-channel (upward and downward long/short waves) radiation |
| 4     | CSAT3       | Campbell Scientific Inc., USA | Three-dimensional wind speed                              |
| 5     | LI7500      | Li-Cor, USA                   | Carbon dioxide (\( \text{CO}_2 \)) and water vapor       |
| 6     | TTS111      | Vaisala, Finland              | Vertical profiles of air temperature, air pressure, RH, wind speed, wind direction |
| 7     | TTO111      | Vaisala, Finland              | Vertical profiles of \( \text{O}_3 \)                     |
A single-column (1D) photochemical model (Forkel et al., 2006) was employed in this study to address the impacts of nocturnal cloud top radiative cooling on PBL structures and atmospheric O\(_3\) variations in CD. There are 40 vertical layers extending from the surface to approximately 2 km above ground level with a vertical grid spacing of 1 m for the lowest layer and approximately 350 m for the uppermost layer, which aims to adequately capture the PBL structure. The initial conditions were determined from the surface and vertical profiles in CD. In addition, the microphysics scheme was not considered in the 1D model because it contributed less to cloud top cooling compared to radiative cooling (Vanzanten, 2002). More details regarding the 1D model parameterizations and setup can be found in Hu et al. (2015), Hu, Klein, and Xue (2013), and Hu, Klein, Xue, et al. (2013).

The plane-parallel radiative transfer model Santa Barbara Distort Atmospheric Radiative Transfer (SB-DART) was employed to calculate the nocturnal cloud top radiative cooling rate (Ricchiazzi et al., 1998). The standard midlatitude atmosphere was used in SBDART to investigate the climatological means cloud top radiative cooling rate. Furthermore, the radiative cooling rates from the SBDART model were coupled into a 1D photochemical model to calculate the impacts on turbulent diffusion coefficients and vertical distribution of potential temperature and O\(_3\) concentrations.

### 3. Results and Discussions

#### 3.1. Increase in Nocturnal Surface O\(_3\) by Cloud-Driven Vertical Mixing

Elevated nocturnal surface O\(_3\) was observed in western SCB including CD and DY within the cloud topped boundary layer, which were chosen to investigate underlying processes concerning cloud-driven vertical mixing of O\(_3\). The observed surface O\(_3\), NO\(_2\), PM\(_{2.5}\), CO\(_2\), wind speed (WS), and direction (WD) during January 20–21, 2017, in CD and DY are shown in Figure 1. The surface O\(_3\) concentration increased by 10 and 4 ppb in CD and DY from 00:00 to 02:00, respectively, while NO\(_2\) and PM\(_{2.5}\) concentrations declined by 20 and 7 ppb and 28 and 16 μg m\(^{-3}\), respectively (Table 2). The elevated nocturnal surface O\(_3\) concentrations are mainly attributed to meteorological processes due to a lack of photochemical reactions (Hu et al., 2012; Zhang & Rao, 1999; Zhang et al., 1998). The climate in SCB is characterized by low PBL height and weak winds due to the deep basin topography, resulting in a strong local accumulation of air pollutants in air pollution (Cao et al., 2020; Ning et al., 2018; L. Zhang et al., 2019). In our measured period, the near-surface wind speed was no more than 3 m s\(^{-1}\) at two sites (Figure 1e), and the sea level pressure was uniformly distributed over SCB, indicating weak horizontal transport. The near-surface northerly winds prevailed with mean wind speeds of approximately 2 m s\(^{-1}\) at two sites (Figure 1e). The surface CO\(_2\) concentrations in CD were about 385 ppm during the night, much lower than DY (Figure 1d), which is located 50 km north of CD. The surface CO\(_2\) concentrations in CD did not present an apparent rise with the condition of northerly winds and higher-level CO\(_2\) in the north, which meant that horizontal transport contributed less. Thus, horizontal transport contribution was less, and vertical transport contribution might be the main cause of the increase in nocturnal surface O\(_3\).

To investigate the near-surface atmospheric stability during this period, the time series of surface radiation flux, heat flux, turbulence kinetic energy (TKE), and vertical velocity skewness in CD and DY are presented in Figure 2. The TKE was calculated by the measured three-dimensional wind speed at a frequency of 10 Hz

\[
\text{TKE} = 0.5 \times \left( \overline{u^2} + \overline{v^2} + \overline{w^2} \right)
\]

The vertical velocity skewness was calculated in DY to identify the source of turbulence

\[
\text{s} = \left( \overline{w^3} / \overline{w^2} \right)^{3/2}
\]

The negative vertical velocity skewness indicates the narrower and more intense downdrafts, which means the top-down cloud top-driven convection and downward transport of turbulence eddies (Hogan et al., 2009; Moyer & Young, 1991). The surface average total net radiation (TNR), which is the difference between downward and upward radiation, were −13 and −18 W m\(^{-2}\), and downward longwave radiation (DLWR) were −328 and −325 W m\(^{-2}\) during the increasing O\(_3\) period in CD and DY, respectively. Such low TNR and high DLWR indicated cloudy conditions during this period, which was in accordance with observations from the FY2G satellite.
Additionally, during the surface O₃ increasing period, TKE was enhanced after 20:00 and reached a peak at approximately 0.5 and 0.3 m² s⁻² in CD and DY at 00:00. Then, it decreased to nearly zero at approximately 02:00. The vertical velocity skewness was negative and less than −0.5, which indicated that downward turbulent transport of TKE was dominant (Hogan et al., 2009). Meanwhile, the sensible heat flux (H) decreased to the minimum at −14 and −11 W m⁻² corresponded to the maximum in TKE, and the latent heat flux (LE) reached up to 8 and 10 W m⁻² in CD and DY. Such variations indicated that enhanced surface turbulence was transported from the upside. Consequently, the decoupled SBL was weakened substantially by downward transport of turbulence as surface O₃ increased and NO₂ and PM₂.5 decreased.

Boundary layer structures on January 20, 2017 were clearly illustrated to further investigate turbulence generation by the measured vertical profiles of potential temperature, virtual potential temperature, relative humidity (RH), O₃ mixing ratio, wind speed, and wind direction in CD (Figure 3). At 14:00, the PBL height was 600 m, and the virtual potential temperature was constant at 286.4 K below 600 m owing to strong convection within the ML. After sunset, the PBL height dropped to about 400 m because of weakened convection, and SBL was formed at 18:00 due to surface radiative cooling. However, the PBL height was elevated to 750 m at 20:00. The virtual potential temperature was uniformly distributed from 100 to 750 m and was constant at 285.5 K. Moreover, O₃ was mixed well and was kept at a low value below 750 m. Uniformly
Figure 2. Time series of (a) surface downward longwave (DLWR) and total net (TNR) radiations; (b) sensible (H) and latent (LE) heat fluxes; (c) turbulence kinetic energy (TKE) and vertical velocity skewness measured in the Chengdu (CD; solid line) and Deyang (DY; dashed line) during January 20–21, 2017.

Figure 3. Vertical profiles of potential temperature, virtual potential temperature, relative humidity (RH), O$_3$ mixing ratio, wind speed, and wind direction measured in Chengdu (CD) at 14:00, 18:00, and 20:00 on January 20, 2017.
distributed virtual potential temperature, well-mixed \( O_3 \) concentration, and elevated PBL height indicated enhanced vertical mixing within PBL at nighttime. However, LLJs played a minor role in turbulence generation within the PBL at 20:00, as shown in Figure 3. Additionally, RH was over 90% from 300 to 750 m, which was detected as a cloud layer according to the cloud retrieval method (Zhang et al., 2020). The cloud layer could produce buoyancy-generation of turbulence by emitting longwave radiation and cooling ambient air (Caughey & Kitchen, 1984; Duynkerke & Driedonks, 1987; Duynkerke et al., 1995), which enhances the downward transport of turbulence and induces strong vertical mixing below the cloud top layer. Hence, the increase in nocturnal surface \( O_3 \) was associated with enhanced vertical mixing, which might be induced by cloud radiative effects at nighttime.

Considering the evidence described above, we identified that the nocturnal \( O_3 \) peak was attributed to cloud-driven vertical mixing, which weakened decoupled SBL and entrained \( O_3 \) aloft downward.

### 3.2. Sensitivity Study of Cloud-Driven Vertical Mixing

To examine the roles of nocturnal cloud top radiative cooling in the turbulent transport of \( O_3 \), this study conducted control and sensitivity simulations using the 1D photochemistry model. The simulations were initialized by measured profiles over CD at 14:00 on January 20, 2017. The control simulation had a cloud top radiative cooling rate at approximately 700 m from 19:00 to 03:00 on the following day, while the sensitivity experiment did not apply the cooling rate. The cloud top radiative cooling rate was 28 K day\(^{-1}\), which is reasonable compared to previous studies (Hartung et al., 2013; Sieglaff et al., 2011; Zhang...
et al., 2020). The latter was otherwise the same as in the control experiment. The control simulation results remained within a reasonable range compared to the measurements (Figure 4), even though the modeled surface O$_3$ concentrations were a little lower. The reason for that O$_3$ increased from the ground surface to about 150 m height was attributed to one-level emission at surface used in the model, which consumed near-surface O$_3$ quickly and kept a low value of O$_3$ below the stable boundary. It successfully produced an abrupt increase in surface O$_3$ concentration at the onset of cloud top radiative cooling and uniform vertical distributions of potential temperature and O$_3$ below the cloud layer at night (Figure 4).

To compare the vertical distributions of O$_3$ and the potential temperature between the control and sensitivity simulations, the time-altitude cross sections of simulated O$_3$ and potential temperature are shown in Figure 5. In the control simulation, the O$_3$ aloft was leaked downward to the surface after 20:00, which reduced O$_3$ aloft over 400 m and increased O$_3$ concentration at low altitude. The potential temperature was uniformly distributed from 100 to 700 m at nighttime. After 05:00, the surface O$_3$ mixing ratio declined to a low value and the potential temperature inversion became strong again. In the sensitivity simulation, the PBL structure was more stable during the night, and SBL was decoupled after sunset. The potential temperature inversion was much stronger and the surface O$_3$ mixing ratio was nearly depleted due to NO titration and dry deposition (Figure 5b). Comparing the two simulations, the cloud top radiative cooling resulted in a uniformly distributed potential temperature and well-mixed O$_3$ below the cloud layer and weakened PBL stability. The induced vertical mixing entrained O$_3$ aloft down to the surface, causing O$_3$ aloft leaking and surface O$_3$ increasing (Figures 4c and 5). These results confirmed the hypothesis that cloud top radiative cooling was the main reason for the turbulent transport of O$_3$ during the night.

### 3.3. Impacts of Cloud Variations on Turbulent Transport of O$_3$

To further examine this mechanism concerning vertical transport of O$_3$ in the pollution condition, a high-level O$_3$ event during April 30 to May 1, 2017 was employed to investigate the factors affecting the cloud top cooling rate and the contribution to surface O$_3$ in the ideal simulations. The cloud top radiative cooling rate is determined by the cloud liquid water path (LWP) and the droplet effective radius (Rosenfeld et al., 2019). Cloud LWP is a measure of the total amount of liquid water in the atmosphere above a unit surface area, which is a key parameter regulating many cloud physical processes and largely determining cloud radiative forcing (Shupe & Intrieri, 2004). Previous studies have revealed that the cloud top radiative cooling rate is not sensitive to the cloud effective radius (Zhang et al., 2020; Zheng et al., 2016). Therefore, the SBDART model was employed to examine the role of LWP in nocturnal cloud top radiative cooling. The input LWP was 5, 10, 20, 50, and 90 g m$^{-2}$ and uniformly distributed over the altitude range from 1,000 to 1,300 m in the simulations. The cloud droplet effective radius was set as the default value of 15 μm in the simulations. The SBDART simulation results showed that a strong radiative cooling rate occurred within the upper portion of the cloud layer (Figure 7a). The radiative cooling rate increased with LWP, but the difference in rates between 50 and 90 g m$^{-2}$ of LWP was lower. This agrees well with previous studies showing that the cooling rate dependence was strong at low LWP but saturated when LWP > 50 g m$^{-2}$ (Kazil et al., 2016).

The SBDART modeling results were coupled in a 1D photochemistry model to investigate the impacts on turbulent transport of O$_3$. The vertical profiles measured at 14:00 during a high-level O$_3$ event initialized 1D model to examine the effects of related processes on atmospheric O$_3$ variations during a pollution episode. The maximum surface hourly O$_3$ concentration was about 110 ppb in the afternoon.
on April 30, 2017, during the O₃ pollution episode (Figure 6). The cloud top radiative cooling rates from the SBDART modeling results were added between 01:00 and 04:00 on the following day in five sensitivity experiments. Moreover, the control run without cloud top radiative cooling was evaluated and set for comparison (Figures 6 and 7). The control run results remained within a reasonable range compared to observations (Figure 6). Compared with the control experiment, a strong turbulent diffusion coefficient occurred at 800 m. The vertical mixing intensity increased with the enhanced cloud top radiative cooling rate in five sensitivity experiments. The potential temperature inversion layer intensity near the surface was weakened in five sensitivity experiments, which facilitated weakening of the decoupling between SBL and RL. The potential temperature between 300 and 1,500 m decreased as the radiative cooling rate increased, but all presented well-mixed conditions. The O₃ mixing ratio decreased at high altitudes but increased at low altitudes compared with the control experiment. The profiles of O₃ also presented well-mixed conditions in the sensitivity experiments. The O₃ aloft exhibited less differences among these five experiments, and all were uniformly distributed at 85 ppb between 300 and 1500 m (Figure 7d). However, the surface O₃ mixing ratio increased as the cloud top radiative cooling rate increased, and the maximum surface O₃ increased by over 30 ppb (Figure 7e). Thus, the contribution from turbulent transport to surface O₃ increased with the cloud top radiative cooling rate, which was associated with LWP.

4. Conclusions

The present investigation illustrated that the low-level clouds commonly occurring at night over the Sichuan Basin (SCB) in China contributed to the vertical redistribution of O₃ and nocturnal surface O₃ maximum. A case study was conducted for January 20–21, 2017, when a low-level cloud layer and elevated surface O₃ mixing ratio were measured at night. During this nighttime, the virtual potential temperature and O₃ were uniformly distributed below the cloud topped boundary layer, and PBL height was even elevated by 400 m. Moreover, the near-surface turbulence and downward sensible heat flux became typically stronger during the period. Thus, the maximum nocturnal surface O₃ appeared to be associated with cloud-driven vertical mixing. Simulation results from the 1D photochemistry model confirmed the impacts of clouds on boundary layer structures by cloud top radiative cooling. The cloud top radiative cooling induced strong vertical turbulent mixing below the cloud layer, which weakened the near-surface temperature inversion
layer intensity and entrained O\textsubscript{3}-rich air aloft down to the surface. Consequently, it led to an increase in the nocturnal surface O\textsubscript{3} within the cloud topped boundary layer. The vertical turbulent mixing intensity increased with the cloud top radiative cooling rate, which was associated with the cloud liquid water path. The stronger vertical mixing resulted in higher surface O\textsubscript{3} levels at nighttime, with maximum surface O\textsubscript{3} increasing by 30 ppb during an O\textsubscript{3} pollution episode.

This revealed that turbulent transport of O\textsubscript{3} induced by cloud top cooling is an important mechanism (Figure 8), which has substantial implications for understanding the consumption of nocturnal O\textsubscript{3}-rich aloft and the impacts on surface O\textsubscript{3} levels following day (Hu, Klein, Xue, et al., 2013). Furthermore, cloud-driven turbulence has an important impact on weakening PBL stability and facilitating vertical diffusion of near-surface air pollutants in the nighttime. Even though we used a low-level O\textsubscript{3} case measured in winter to examine this mechanism, the effects on nocturnal atmospheric O\textsubscript{3} during the pollution period were verified based on sensitivity simulations. The turbulent transport of O\textsubscript{3} within a cloud topped boundary layer in summer smog is to be further analyzed using a three-dimensional model considering the cloud microphysics process and more comprehensive measurements, including high-temporal-resolution atmospheric profiles and cloud properties.

Figure 7. Profiles of simulated (a) cooling rate, (b) turbulent diffusion coefficient, (c) potential temperature, (d) O\textsubscript{3} mixing ratio at 04:00, and (e) time series of surface O\textsubscript{3} mixing ratio in Chengdu (CD) during a high-level O\textsubscript{3} episode during April 30 to May 1, 2017.
Data Availability Statement

The data presented in this study were archived at https://figshare.com/projects/Impacts_of_nocturnal_cloud-top_radiative_cooling_on_surface_O3_in_Sichuan_Basin_southwestern_China/92705.

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