Evaluation of Vertical Ozone Profiles Simulated by WRF/Chem Using Lidar-Observed Data

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Abstract

We evaluated the ability of a regional air quality (AQ) model, the Weather Research and Forecast chemistry (WRF/Chem), to reproduce vertical profiles of summertime ozone (O_3) pollution over the Kanto region of Japan using lidar-observed O_3 data collected over Tsukuba on 27–29 July and 16–21 August 2005. WRF/Chem well reproduced the high O_3 concentrations $> 2.5 \times 10^{18}$ molec. m^{-3} (~ 100 ppbv below 1.5 km above sea level ASL) observed on 28 July and 20 August, and the low concentrations on the other days below 1.5 km ASL, as well as day-to-day variations above 1.5 km ASL. The formation mechanism of the observed O_3 layer in the typical northward transport case on 28 July was also examined by the model. The O_3 plumes entered the free troposphere through vertical transport and mixing near the sea breeze front and southward transport by the return flow in the afternoon. In the evening, the O_3 layer formed at 0.7–1.5 km ASL possibly because of less transport between the sea breeze and return flow and longer photochemical lifetime than at the surface.

1. Introduction

Air quality (AQ) models have been developed to understand and forecast air pollution at regional scale (e.g., Uno et al. 2003) and global scale (Lawrence et al. 2003; Takigawa et al. 2005). To reproduce the ozone (O_3) and precursor distribution over the Kanto region of Japan (Fig. 1, inset) using an AQ model, fine-scale meteorology is important (Uno et al. 1996). Kimura (1985) showed that summertime O_3 pollution episodes over the Kanto region occur in the rural inland portion through horizontal transport by the “extended” land-sea breeze coupled with mountain-valley winds as well as synoptic winds. Vertical transport and mixing by thermal convection and sea-breeze front updraft are also key factors in the surface O_3 concentration. Dufour et al. (2005) clarified the multi-layered structure of O_3 over southern France using lidar observation and used a model to examine the effect of the O_3 layers on the surface concentration. Over Japan, some studies have conducted aircraft-based observations and have reported a multi-layered structure of O_3 (e.g., Wakamatsu et al. 1983). However, no previous studies have conducted quasi-continuous observations of vertical O_3 profiles and evaluated models against the observations.

An AQ forecasting system has been developed based on the Weather Research and Forecast chemistry model (WRF/Chem) and the use of chemical boundary data from the CHASER model (Takigawa et al. 2007). We evaluated a hindcast experiment by WRF/Chem against vertical O_3 profiles measured in summer 2005 by the recently developed lidar method (Nakazato et al. 2007).

Fig. 1. Model domains (D1 and D2) and terrain height in m (coloring) for the regional CTM. Inset: Detailed map of the Kanto plain including D2 and the observation site, TSU: Tsukuba (black circle).

2. Model and observation data

The regional AQ model was the regional chemical transport model (CTM) WRF/Chem version 2.1.2 (Grell et al. 2005) and obtained lateral boundary data of chemical species from the global CTM CHASER (Sudo et al. 2002). The CHASER model was developed in the framework of the Center for Climate System Research/National Institute for Environmental Studies/Frontier Research Center for Global Change (CCSR/NIES/FRCGC) atmospheric general circulation model (AGCM) version 5.7b (Hasumi et al. 2004). Both WRF/Chem and CHASER calculate meteorology and chemistry “on-line,” meaning that both the meteorological and chemical modules use the same time step, grid, and physics schemes. Takigawa et al. (2007) provided details of the model system.

The regional model consisted of two domains. The outer domain (D1) had 84×84 grids at 27-km resolution, and the inner domain (D2) had 39×39 grids at 9-km resolution (Fig. 1). Both domains had 30 layers from the surface to about 100 hPa and approximately 11 layers below 2 km. The planetary boundary layer (PBL) height and the vertical diffusion were calculated by the Mellor-Yamada Level 2.5 model based on prognostic turbulent kinetic energy (Janjić et al. 2002). A hindcast experiment was conducted from 25 July to 23 August 2005, when lidar observations of O_3 were conducted over Tsukuba (Fig. 1, black circle). For D1, boundary data for the meteorological field were from the three-hourly mesoscale model (MSM) analysis of the Japan Meteorological Agency (JMA). Simulated meteorological fields were updated by the MSM analysis at 09 Japanese Standard Time (JST = Universal Time + 9 h). The lateral

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Fig. 2. Surface temperature in K (a), wind direction in degrees (b), and O_3 mixing ratio in ppbv (c) at Tsukuba from the observation (black) and simulation (red) in July and August 2005.

boundary data of the chemical fields for D1 were obtained every 3 h from CHASER.

The simulated results were compared to vertical O_3 measured by differential-absorption lidar (DIAL) at the Meteorological Research Institute (MRI) in Tsukuba (140.10°E , 36.05°N) during the periods 27–29 July and 16–21 August 2005. The data used were hourly data integrated for 30 min temporally and for 120 m vertically. This integration was sufficient to measure O_3 concentration profiles from 400 m up to 10 km with estimated statistical errors $< 5\%$ at 1.5 km and 10% at 2.6 km. Details of the O_3 DIAL have been described by Nakazato et al. (2007). Data severely contaminated by clouds and aerosols were removed in advance using the following criteria: (1) if the estimated statistical errors were $> 0.8 \times 10^{17} \text{ molec. m}^{-3}$, (2) if the statistical errors were $> 10\%$, or (3) if the O_3 concentration was $> 3.5 \times 10^{18} \text{ molec. m}^{-3}$. The obtained data were then interpolated to the same altitude levels as those of the WRF/Chem output data.

3. Results and discussion

To confirm the reproducibility of local meteorology and O_3 concentration by WRF/Chem, observed and modeled time series at the surface level over Tsukuba were examined (Fig. 2). We focused on the periods of the DIAL measurement on 27–29 July and 16–21 August 2005. Tsukuba lies 60 km northeast of the Tokyo metropolitan region, and O_3 plumes are typically carried from the Tokyo region in the “northward transport” case, when the extended southeasterly sea breeze co-occurs with weak southerly to southwesterly synoptic wind. The simulation on 28–29 July and 18–20 August represented a typical summertime O_3 episode in the northward transport case in the Kanto area. The meteorological field at Tsukuba was representative of those at eastern Kanto region and showed less diurnal cycle of wind direction than in western Kanto region.

After the passing of Typhoon 200507 (“Banyan”), the surface temperature dropped (Fig. 2a), and easterly to southerly flow became dominant on 28 and 29 July (Fig. 2b) as a subtropical high over the Pacific Ocean covered Japan. On these two days, the modeled and observed O_3 mixing ratios increased to nearly 80 ppbv over Tsukuba and northern Kanto (Fig. 2c) in relation to the typical extended sea-breeze development. On 16 and 17 August, the surface temperature increased to at most 302 K, and winds were northerly to easterly with light rain on 16 August, after a frontal low passed over Japan on 15 August. On these two days, the O_3 mixing ratio was continuously low (~ 40 ppbv) for both the DIAL and model results. From 18 to 20 August, the tempera-

ture gradually increased and southerly wind became dominant over the Kanto region as a subtropical high over the Pacific Ocean covered the Kanto region and a cold front approached over northern Japan. The observed O_3 mixing ratio was at most 40 ppbv on 18 and 19 August and 60 ppbv on 20 August. The model well reproduced the O_3 concentration increases on these three days, but overestimated those observed between 19 and 21 August. This overestimate was likely caused by the underestimate of convective cloud development and of consequent development of strong southerly convergence flow over northern Kanto in the afternoon. The observed strong southerly flow could have led to less accumulation of O_3 in the southern Kanto region than in the model. During the whole period, the overall reproducibility of O_3 was in accordance with the surface statistics over the Kanto region (normalized mean bias [NMB] = 13.5% and correlation coefficient $r = 0.71$ based on 268 stations with a threshold of 40 ppbv).

We next compared the observed and modeled vertical profiles of O_3 concentration over Tsukuba. Plate 1 presents time-height sections of the O_3 concentration over Tsukuba on 27–30 July and 16–22 August 2005. Below 1.5 km above sea level (ASL), the O_3 concentration from the DIAL measurement (Plate 1a) showed day-to-day variation. On 28 July and 20 August, O_3 concentrations $> 2.5 \times 10^{18} \text{ molec. m}^{-3}$ (~ 100 ppbv in the PBL) appeared below 1.5 km ASL from around noon. The layered structure on 28 July at 0.5–1.5 km ASL remained until 03 JST on 29 July. From 16 JST on 19 August to 06 JST on 20 August, a layered structure of O_3 concentration of $\sim 1.25 \times 10^{18} \text{ molec. m}^{-3}$ was observed. On 17 and 21 August, there was little enhancement $> 2.0 \times 10^{18} \text{ molec. m}^{-3}$ in the DIAL data. The modeled O_3 (Plate 1b) compared well to the observed enhancement on 28 July and 20 August and to the low enhancement on 17 August. The model succeeded in well reproducing the observed nocturnal layered structure over the Kanto region on 28–29 July and 19–20 August for the first time. The statistical analysis of the time series of O_3 concentration from the DIAL and from the model during these periods indicated relatively good results (NMB = 7.82% and $r = 0.48$ at 1.0–1.5 km ASL). The relatively small r mainly arose from the overestimation of $\sim 0.5 \times 10^{18} \text{ molec. m}^{-3}$ for 16–21 August.

Above 1.5 km ASL, the DIAL O_3 concentration (Plate 1a) became $> 1.25 \times 10^{18} \text{ molec. m}^{-3}$ from 27 to 28 July and $> 1.0 \times 10^{18} \text{ molec. m}^{-3}$ on 17 August, whereas it remained $< 1 \times 10^{18} \text{ molec. m}^{-3}$ from 19 to 21 August. The model (Plate 1b) successfully captured the observed temporal variations with enhancement on 27–28 July and 17 August and without enhancement on 19–21 August over Tsukuba ($r = 0.61$ – 0.74 at 2.5–4.5 km ASL during these periods). In contrast, the model continuously underestimated the mean O_3 concentration at this altitude range (NMB = -21.9% at 2.5–3.5 km ASL and -30.9% at 3.5–4.5 km ASL during these periods). The combination of the overestimation below 1.5 km ASL and underestimation above 1.5 km ASL implies that vertical mixing in the PBL was underestimated. Kim et al. (2006) obtained similar results in a study involving WRF/Chem evaluation over the United States. They reported overestimates of O_3 , carbon monoxide, and nitrogenous species below 1–1.5 km and underestimates above 1–1.5 km. The underestimate of vertical mixing in the Mellor–Yamada level 2.5 closure model should be addressed, as noted by several previous studies (e.g., Nakanishi and Niino 2004). The improvement of the vertical mixing calculation would reduce the overestimate of the PBL O_3 concentration that was found during the periods that we described. However, the underestimate above 1.5 km ASL is considered to be more related to the underestimate of the time mean O_3 and precursor concentrations of the lateral boundary data from CHASER because the negative NMB increases with height. The inclusion of updated emission inventory and time-dependent hot-

Plate 1. Time-height sections of the O_3 concentration in 1×10^{18} molec. m^{-3} (~ 40 ppbv) over Tsukuba from the O_3 DIAL measurement (a) and the model (b). The PBL height (white line) is also plotted in (b). The DIAL data are based on hourly data integrated for 30 min, while the simulated data are hourly instantaneous values. Blanks in the DIAL data indicate that data were removed by quality controls or no observation occurred at that time.

spot data would help to reduce the underestimate above 1.5 km ASL during these periods. Model evaluations using longer-term DIAL data are needed to obtain more precise results.

To confirm the reproducibility of horizontal distributions by the model, we examined horizontal maps of the wind direction and O_3 mixing ratio at 16 JST on 28 July and 17 August from the observations and model (Fig. 3). The DIAL measured an O_3 plume on 28 July over Tsukuba, but not on 17 August (Table 1a). At 16 JST on 28 July, the wind direction was generally southerly at $139\text{--}140^\circ\text{E}$, $35\text{--}36.5^\circ\text{N}$ (Fig. 3a), and O_3 plumes were detected north of the Tokyo area by both the observations and the model (Fig. 3c). At 16 JST on 17 August, the observations and model indicated southeasterly flow over the Kanto area (Fig. 3b) and O_3 plumes southwest of the Tokyo area (Fig. 3d). For both days, WRF/Chem generally simulated the horizontal distributions of wind direction and O_3 , although the model missed the O_3 plumes north of 36.2°N on 28 July because of the delay of sea breeze onset.

The formation processes of the O_3 layer in the typical northward transport case of 28 July were also investigated using WRF/Chem. The O_3 concentration, meridional and vertical winds, and PBL height averaged over $139.7\text{--}140.2^\circ\text{E}$, an area that includes Tsukuba, are shown in Fig. 4. Northerly and southerly winds were dominant and the O_3 distributions were longitudinally homogeneous over this area on this day. At 12 JST (Fig. 4a), an O_3 mixing ratio of 70–80 ppbv and large vertical motions appeared at $35.5\text{--}36.8^\circ\text{E}$ (corresponding to the Tokyo Bay area) below 1 km ASL, indicating vertical transport of O_3 and its precursors and photochemical production of O_3 from its precursors. At 16 JST (Fig. 4b), high O_3 mixing ratios > 100 ppbv appeared over $35.4\text{--}36.1^\circ\text{N}$ up to 1.5 km ASL. Upward motion up to 1.7 km ASL and northerly return flow at 1.5–3 km occurred south of the sea breeze front at 36°N . PBL heights were 1.5–2.5 km ASL north of the sea breeze front and 0.5–1 km south of the front, with a large gap around the front. Consistently, the sea breeze layer was relatively cold and stable, whereas the northern regions and the front region were well heated and highly diffusive (not shown). These results suggest that the O_3 plumes were

Fig. 3. Horizontal maps of wind direction in degrees (a and b) and the O_3 mixing ratio in ppbv (c and d) near the surface level from the observation (circles) and the model (coloring) at 16 JST on 28 July (a and c) and 17 August (b and d). Stream lines from the model are indicated by white lines (a and b).

transported upward by the vertical advection and mixing around the sea breeze front and southward by the return flow into the free troposphere. Thus, the sea breeze likely plays a role in venting O_3 plumes into the free troposphere. At 20 JST (Fig. 4c), the extended sea breeze covered the whole region below 1 km ASL and northerly return flow occurred north of 36.3°N above 1.5 km ASL. Between the sea breeze and its return flow, O_3 plumes of > 100 ppbv were detected at 0.7–1.5 km ASL. At 00 JST on 29 July (Fig. 4d), the sea breeze below 1.3 km ASL and the O_3 plumes of > 80 ppbv at 0.7–1.5

Fig. 4. Latitude-altitude sections of the O_3 mixing ratio in ppbv (coloring), meridional and vertical wind fields in $m s^{-1}$ (horizontal and vertical arrows), and PBL heights (black lines) averaged over $139.7\text{--}140.2^\circ E$ from the model at 12 JST (a), 16 JST (b), and 20 JST on 28 July (c), and 00 JST on 29 July (d). Unit vector length of $12 m s^{-1}$ is shown at the bottom of the figures, and vertical winds are multiplied by fifty. Wind vectors are drawn for meridional wind speed $> 3 m s^{-1}$ or vertical wind speed $> 3 cm s^{-1}$. The location of Tsukuba is indicated by triangles.

km still remained. Using aircraft observations, Wakamatsu et al. (1983) found a similar O_3 layer around 0.5 km ASL in a land breeze area northwest of Tokyo Bay in the morning. They suggested that the O_3 layer was caused by old pollution that was trapped overnight by the back-and-force movement of the land-sea breeze. Our results indicate that the nocturnal O_3 layer between 0.7 and 1.5 km ASL was caused by less transport in the weak wind layer and longer lifetime of O_3 than at the surface. The extended sea breeze in the Kanto region has a spatial scale of 100×100 km, an amplitude of $3 m s^{-1}$, and a depth of > 1.5 km, making it a larger than normal coastal land-sea breeze, which usually has a spatial scale of 20–40 km, amplitude of $1 m s^{-1}$, and depth of 1 km. Similar extended land-sea breezes have been identified in California and Korea. We suggest that the venting effect of such extended sea breezes is at least 10 times greater than those of normal breezes based on the typical values noted above.

4. Conclusion

We evaluated the performance of an “on-line” regional model WRF/Chem for summertime episodes of O_3 pollution over the Kanto region of Japan using DIAL-observed O_3 data measured over Tsukuba on 27–29 July and 16–21 August 2005. Below 1.5 km ASL, the O_3 concentration determined using both the DIAL and model showed enhancement $> 2.5 \times 10^{18} \text{ molec. m}^{-3}$ (~ 100 ppbv) on 28 July and 20 August and little enhancement on the other days. Above 1.5 km ASL, day-to-day variations were well reproduced by the model, whereas the O_3 concentration was generally underestimated by 20–30% during this period. Model evaluations using DIAL data measured over longer periods should be conducted to obtain more precise results. The formation mechanism of the observed O_3 layer was examined for a typical northward transport case on 28 July using the model. The O_3 plumes were transported into the free troposphere by vertical advection and mixing near the sea breeze front and by return flow in the afternoon. In the evening, the O_3 layer formed at 0.7–1.5 km ASL in the weak wind layer between the sea breeze and return

flow, possibly because of less transport and longer photochemical lifetime than at the surface.

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