Author comments

Atmos. Meas. Tech. Discuss., 7, 171–194, 2014

DIAL measurement of lower tropospheric ozone over Saga (33.24°N, 130.29°E), Japan, and comparison with a chemical climate model,

by O. Uchino, T. Sakai, T. Nagai, I. Morino, T. Maki, M. Deushi, K. Shibata, M. Kajino, T. Kawasaki, T. Akaho, S. Takubo, H. Okumura, K. Arai, M. Nakazato, T. Matsunaga, T. Yokota, S. Kawakami, K. Kita, and Y. Sasano

The authors wish to thank two referees and Prof. M. J. Newchurch for helpful and thoughtful comments. Each comment is addressed individually below. The referee comments are written in black, and our responses are described in red.

The main changes of the paper since the APCD version are:

- We add a new figure after Fig.5.

  Fig. 6. Horizotal maps of ozone volume mixing ratio in ppbv and horizontal wind in m/s predicted by MRI-CCM2 for 500, 700, 800, 850 hPa pressure levels at 06 h UTC on 7 July 2012.

  Therefore, figure number for Fig. 6 is changed to Fig.7.

Review of manuscript by Anonymous Referee #2

General Remarks

The manuscript describes an upgraded version of an existing ozone lidar system. The performance of the system seems to be good. The value of lidar vertical sounding for model validation and synergetic use is demonstrated. However, some more literature on the benefit of model validation with lidar systems should be included. If a longer effort is planned this should be mentioned since in the past just single case studies have been made. I recommend publication after a few minor adjustments.

We included next three papers in References and our plan after line 19 in Sec. 5 in P. 183:

Roelofs, G. J., Kentarchos, A. S., Trickl, T., Stohl, A., Collins, W. J., Crowther, R. A., Hauglustaine, D., Klonecki, A., Law, K. S., Lawrence, M. G., von Kuhlmann, R., and van Weele, M.: Intercomparison of tropospheric ozone models: ozone transport in a complex tropopause folding event, J. Geophys. Res., 108, D12, 8529, doi:10.1029/2003JD003462, 2003.

Zanis, P., Trickl, T., Stohl, A., Wernli, H., Cooper, O., Zerefos, C., Gaeggeler, H., Schnabel, C., Tobler, L., Kubik, P. K., Priller, A., Scheel, H. E., Kanter, H. J., Cristofanelli, P., Forster, C., James, P., Gerasopoulos, E., Delcloo, A., Papayannis, A., and Claude, H.: Forecast, observation and modeling of a deep stratospheric intrusion event over Europe, Atmos. Chem. Phys., 3, 763-777, 2003.

Niwano, M., Takigawa, M., Takahashi, M., Akimoto, H., Nakazato, M., Nagai, T., Sakai, T., and Mano, Y.: Evaluation of vertical ozone profiles simulated by WRF/Chem using lidar-observed data, SOLA, 3, 133-136, doi:10.2151/sola.2007-034, 2007.

A major benefit of ozone DIAL is that it can measure continuously vertical ozone profiles with high vertical and temporal resolution. The process of stratospheric ozone intrusion into the troposphere was clearly observed on 20-21 June 2001 by continuous DIAL measurement at Garmisch-partenkichen, Germany and the excellent data set was used for model intercomparisons and validation (Roelofs et al., 2003; Zanis et al., 2003; Stohl et al., 2003). On the other hand, the ability of a regional air quality model was evaluated to reproduce summertime ozone pollution over the Kanto region in Japan using quasi-continuous ozone DIAL measurement data (Niwano et al, 2007). We plan to study the process that air mass of high ozone concentration in the free troposphere is mixed into the planetary boundary layer by continuous DIAL measurement. Further, we plan to utilize ozone DIAL data for validation of a new version of an MRI oxidant prediction system over an extended period of time. In the prediction system, a regional chemistry-transport model, known as MRI-PM/c (Kajino et al., 2012), with higher spatial resolution (20 km and/or 5 km) is nested in MRI-CCM2. The prediction system also adopts an up-to-date inventory and higher temporal resolution of biomass burning emissions. The regional model can reproduce meso-scale weather phenomena and advection of air mass. However, if boundary data for the regional model produced by the global model doesn't include adequate information on fine-scale structures in trace gas distributions, the performance of the prediction system will be degraded. Therefore, in order to improve total performance of the prediction system, it is necessary to validate not only the regional model but also the global model with observation data such as ozone DIAL data and surface ozone monitoring data extensively, which enable us to estimate error coming from outside of the boundary.

Details

(1) P. 173, line 22: Replace "pollution" by "pollutant".

Done

(2) P. 175 line 21: I suggest writing: "we modified the coaxial receiving system by adding a small telescope in biaxial configuration".

We changed to "we modified the coaxial receiving system by adding a mirror in biaxial configuration"

(3) P. 178, lines 3-4: Please, replace "systematic error" by "systematic uncertainty". A systematic error is a known quantity and must be corrected!

Done

(4) P. 181, lines 21-22: "A regional model....might solve these discrepancies": Such a statement requires more information on the air masses. For instance, it could be the case that air from different sources arrives side by side. This can be clarified by trajectory analyses or by looking at the model output for adjacent grid boxes. A smaller-scale model is helpful, if air filaments from outside the model domain of the fine-scale model are captured to some extent by the larger-scale model. Otherwise they are missed! I suggest improving also the corresponding part of the "Concluding remarks".

Instead of the sentence "A regional model with a higher spatial resolution of 20 km might solve these discrepancies" in P.181, line 21, we added following sentences with Fig.6:

Figure 6 shows horizontal maps of ozone volume mixing ratio and horizontal wind predicted by MRI-CCM2 for 500 hPa (about 5.5 km altitude), 700 hPa (3 km), 800 hPa (2 km), and 850 hPa (1.5 km) pressure levels at 06 UTC on 7 July 2012. According to the weather chart on the same day provided by JMA (http://www.data.jma.go.jp/fcd/yoho/hibiten/index.html), the Meiyu-Baiu-front extended from west to east through the center of a low-pressure system over Japan. In this prediction, the synoptic-scale pattern of horizontal-wind field is reproduced reasonably. A large north-south gradient of ozone volume mixing ratios is located near the stationary front in the lower free-troposphere. The ozone concentration is low over the south part of the front because of the chemical destruction in the summertime marine air mass, whereas high ozone level area is present over the north of the front. During the DIAL observing period the large gradient of ozone concentration was located near the Saga site in the lower free-troposphere (Fig.6). This suggests that one of the possible reasons for the significant discrepancy between the DIAL ozone profile and the

predicted one (Fig. 5) is that the meso-scale front structure and its position were not fully captured by the model due to its coarse spatial resolution. In addition, it is plausible that model transport with the coarse grid doesn't reproduce well fine-scale structures of air (trace gases) filaments.

(5) P. 182, line 8: Replace "exchanges" by "exchange" (or perhaps "exchange events").
We replaced to "exchange events".
(6) Sec. 5: There is no clear statement about future plans or work. The use of the regional model is sold as an option, not as a plan. Given the uncertainties of regional models with air parcels coming from remote sources mentioned above it is also advisable to announce further comparisons and optimization (see general remarks).
We included future plans in Sec. 5.

Review of manuscript by Anonymous Referee #1

We corrected many sentences in intelligible terms which are recommended by Referee #1. We understood the main comments by Referee are as follows:

1) Add some numerical values in DU in Abstract and Concluding remarks
We added some numerical values in DU in Abstract and Concluding remarks.
2) Problem in page 176, line 22: 1 single lens is not enough for such a wide spectral range (chromatic aberration) provide comment please.
It might be better to use an off-axis beam expander for a wide spectral range of 276–312 nm. We would like to study this matter further.

3) Normally Raman energies are S1>S2>S3, so explain why S1<S3
At a pumping energy of 107 mJ at 266 nm,
S1:S2:S3=10.0:10.1: 4.7 mJ (S1+S2+S3=24.8 mJ) for 8.1×10^5 Pa of CO_2 gas (8 atm.), and
S1:S2:S3=7.5:9.1: 8.4 mJ (S1+S2+S3=25.0 mJ) for 3.0×10^5 Pa of CO_2 gas (3 atm.).
As the total energies were almost the same (about 25 mJ), our experimental result was correct. The reason why S1<S3 is beyond the scope of this paper. We would like to study further the reason.
4) Why didn't you make measurement of the output energy at S4:312 nm?
It was difficult for us to measure the output energy at 312 nm in September 2012 due to the limited space in the container and the maximum output energy of 107 mJ at 266 nm

could not be obtained.

5) Add references

We added next three papers in References:

Papayannis, A., Ancellet, G., Pelon, J., and Mégie, G.: Multiwavelength lidar for ozone measurements in the troposphere and the lower stratosphere, Appl. Opt., 29, 467-476, 1990.

Papayannis, A., Balis, D., Zanis, P., Galani, E., Wernli, H., Zerefos, C., Stohl, A., Eckhardt, S., and Amiridis, V.: Sampling of an STT event over the Eastern Mediterranean region by lidar and electrochemical sonde, Annales Geophys., 23, 2039-2050, 2005.

Stohl, A., Bonasoni, P., Cristofanelli, P., Collins, W., Feichter, J., Frank, A., Forster, C., Gerasopoulos, E., Ga¨ggeler, H., James, P., Kentarchos, T., Kromp-Kolb, H., Kru¨ger, B., Land, C., Meloen, J., Papayannis, A., Priller, A., Seibert, P., Sprenger, M., Roelofs, G. J., Scheel, H. E., Schnabel, C., Siegmund, P., Tobler, L., Trickl, T., Wernli, H., Wirth, V., Zanis, P., and Zerefos, P.: Stratosphere-troposphere exchange: a review, and what we have learned from STACCATO, J. Geophys. Res., 108, D12, 8516, doi:10.1029/2002JD002490, 2003.

Interactive comment on Atmos. Meas. Tech. Discuss., 7, 171-194, 2014.

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We are very pleased to see this Japanese tropospheric ozone lidar obtaining data. As one of a very small number of tropospheric ozone lidars (e.g., TOLNet: http://wwwair.larc.nasa.gov/missions/TOLNet/) its contribution will have a large impact on the global available ozone lidar data available for scientific studies.

I have two suggestions for the authors to strengthen this paper:

1) Provide quantitative estimates of the accuracy of the lidar retrievals relative to the correlative observations (means, s.d., function of altitude, etc.).

We added following sentence after Page.178, line 21:

The average and one standard deviation of the difference between DIAL and ozonesonde data are 14±10 % below 2 km, 6±4 % for 2−7 km, and 20±13 % above 7 km.

2) Provide some example(s) of time/altitude plots of the temporal progression of tropospheric ozone over your sight (e.g.,

http://nsstc.uah.edu/atmchem/lidar/data/120606/120606.html).
We would encourage you to continue to strive for data at lower altitudes (e.g., <= 100m AGL) and during interesting geophysical conditions (diurnal PBL development, STE, land/ocean interactions, etc.). We look forward to future publications of these lidar observations.

As denoted in Sec. 5, we plan to study the process that air mass of high ozone concentration in the free troposphere is mixed into the planetary boundary layer by continuous DIAL measurement. In the process study, we would like to provide some examples of time/altitude plots of the temporal progression of tropospheric ozone.
We thank you for your encouragement of our ozone DIAL measurement in Japan.

Revised manuscript

DIAL measurement of lower tropospheric ozone over Saga (33.24°N, 130.29°E), Japan, and comparison with a chemistry climate model

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Abstract

We have improved an ozone DIfferential Absorption Lidar (DIAL) system, originally developed in March 2010. The improved DIAL system consists of a Nd:YAG laser and a 2 m Raman cell filled with 8.1×10^5 Pa of CO_2 gas which generate four Stokes lines (276, 287, 299, and 312 nm) of stimulated Raman scattering, and two receiving telescopes with diameters of 49 and 10 cm. Using this system, 44 ozone profiles were observed in the 1–6 km altitude range over Saga (33.24°N, 130.29°E) in 2012. High ozone layers were observed at around 2 km altitude during April and May. Ozone column amounts within the 1–6 km altitude range were almost constant (19.1 DU on average) from January to March, and increased to 26.7 DU from late April to July. From mid-July through August, ozone column amounts decreased greatly to 14.3 DU because of exchanges of continental and maritime air masses. Then in mid-September they increased again to 22.1 DU within 1–6 km, and subsequently decreased slowly to 17.3 DU, becoming almost constant by December.

The Meteorological Research Institute's Chemistry-Climate Model version 2 (MRI-CCM2) successfully predicted most of these ozone variations with the following exceptions. MRI-CCM2 could not predict the high ozone volume mixing ratios measured at around 2 km altitude on 5 May and 11 May, possibly in part because emissions were assumed in the model to be constant (climatological data were used). Ozone volume mixing ratios predicted by MRI-CCM2 were low in the 2–6 km range on 7 July and high in the 1–4 km range on 19 July compared with those measured by DIAL.

1 Introduction

Ozone is an important air pollutant that at high concentrations impacts on human health and ecosystems including crops (Parrish et al., 2012). Tropospheric ozone has two main sources: photochemical production in the troposphere and downward transport from the stratosphere. Tropospheric ozone is mainly produced from nitrogen oxides (NO_x), carbon monoxide (CO), and volatile organic compounds (VOCs) by photochemical reactions. More specifically in Asia, emissions of these ozone precursors increased between 1980 and 2003 (Ohara et al., 2007). In China in particular, NO_x increased from 1996 through 2004 (Zhang et al., 2007). Summertime ozone concentrations in the 0–3 km altitude range over Beijing increased at the rate of 3 % yr^{-1} from 2002 to 2010 (Wang et al., 2012). Ozone is also a greenhouse gas that plays an important role in climate

change. The radiative forcing due to tropospheric ozone is the third strongest after carbon dioxide and methane (IPCC, 2007).

In recent years, high concentrations of surface ozone have been observed from April through June in the Kyushu district of western Japan (http://www.env.go.jp/air/osen/). In 2011, we deployed an ozone DIfferential Absorption Lidar (ozone DIAL) system, originally developed by the National Institute for Environmental Studies (NIES) at Tsukuba, at Saga (33.24°N, 130.29°E) in the Kyushu district, with three aims: (1) to validate GOSAT (Greenhouse gases Observing SATellite) ozone products retrieved from thermal infrared spectral radiances by the Thermal And Near infrared Sensor for carbon Observation-Fourier Transform Spectrometer (TANSO-FTS) onboard GOSAT (Ohyama et al., 2012); (2) to detect high ozone concentrations in the lower troposphere; and (3) to compare the observed concentrations with concentration data predicted by the Meteorological Research Institute's (MRI) Chemistry-Climate Model, version 2 (MRI-CCM2) (Deushi and Shibata, 2011). Except in summer, Saga is downwind of the Asian continent most of the year. Thus, if DIAL can be used to detect high ozone concentrations, then inputting DIAL data into MRI-CCM2 will allow us to make useful predictions for photochemical oxidant advisories. In Sect. 2 we describe the ozone DIAL system, including some improvements made since 2011 and comparisons with ozonesonde data, and we describe MRI-CCM2 in Sect. 3. In Sect. 4, we present observational results obtained by the ozone DIAL system and compare them with MRI-CCM2 predictions. Section 5 is a summary and our conclusions.

2 Ozone DIAL system, data analysis and comparison with ozonesonde results

Tropospheric ozone DIAL systems have been first developed using tunable dye lasers (Gibson and Thomas, 1975; Pelon and Mégie, 1982; Browell et al., 1983; Profitt and Langford, 1997; Kuang et al., 2013). Later Stokes lines from stimulated Raman scattering (SRS) of pressurized gas pumped by excimer lasers (Uchino et al., 1983; Kempfer et al., 1994; Eisele et al., 1999), and by Nd:YAG lasers (Ancellet et al., 1989; Sunesson et al., 1994; McDermid et al., 2002; Papayannis et al., 2005; Nakazato et al., 2007; Apituley et al., 2010) were used. In 2010, we developed a tropospheric ozone DIAL system at NIES (Uchino et al., 2011) that was installed in a container with dimensions of about 228 cm × 683 cm × 255 cm. The system's transmitter transmits laser beams at three Stokes lines (276.2 nm, 287.2 nm, and 299.1 nm) of the SRS of carbon dioxide (CO_2) with vibrational shift of 1385 cm^{-1} pumped by the fourth harmonic (266 nm) of a

Nd:YAG laser (Nakazato et al., 2007; Seabrook et al., 2011).

We used this transmitter with a coaxial receiving system with a 49 cm-diameter Newtonian reflecting telescope to measure ozone profiles from a few hundred meters to about 10 km altitude. However, the signal-induced bias (SIB) from the photomultiplier tubes (PMTs) limited the measurement altitude to below about 6 km (Sunesson et al., 1994; Proffitt and Langford, 1997). A mechanical chopper with a fast rising time could suppress the SIB (Uchino and Tabata, 1991; McDermid et al., 2002; Godin-Beekmann et al., 2003; Apituley et al., 2010), but there was not enough space in the container to add a mechanical chopper to the DIAL receiving system. Therefore, in January 2012 we modified the coaxial receiving system by adding a mirror in biaxial configuration. In the biaxial system, the Stokes lines were transmitted upward by a 10 cm-diameter mirror with 90 % reflectivity that was set at a distance of 25 cm from the edge of the 49 cm telescope, while about 10 % of the laser energy was used to measure low-altitude ozone by the original coaxial system. Thus adding the biaxial system, we could measure ozone profiles up to about 10 km; However, it was time-consuming to measure ozone profiles from 1 to 10 km by alternating between biaxial and coaxial measurements.

To be able to measure tropospheric ozone profiles simultaneously in the altitude range of 0.3–10 km, we introduced an additional new 10 cm-diameter Newtonian receiving telescope in September 2012, using a 1.8 mm-diameter quartz optical fiber to transmit the receiving light from the position of the iris of the 10 cm telescope to the entrance of a spectrometer. A dichroic beamsplitter with a 15° incidence angle efficiently separated the signal into two wavelengths, 276 and 287 nm. Furthermore, we tested the use of a fourth Stokes line (312.0 nm) of SRS from CO_2 for ozone measurement up to 15–20 km on a clear night by two wavelengths of 299 and 312 nm.

A block diagram of the improved DIAL system is shown in Fig. 1. The maximum output pulse energy of the Nd:YAG laser (Quantel YG981C) is 107 mJ per pulse at 266 nm; However the normal averages of the output energies are 70–85 mJ at 266 nm. The laser beam is collimated into the center of a 2 m-long Raman cell, which consists of a stainless-steel tube with a diameter of 35 mm and 10 mm-thick UV-grade quartz windows with an anti-reflective (AR) coating. The input window is a lens with a focal length f_1=1100 mm. The output energy at 312.0 nm is not measured, but CO_2 pressure is set to be 8.1×10^5 Pa because the largest receiving signal is obtained at 312.0 nm. At 8.1×10^5 Pa of CO_2 pressure and a pumping energy of 107 mJ at 266 nm, the output energies of the three Stokes lines at S1: 276.2, S2: 287.2, and S3: 299.1 nm are 7.5, 9.1, and 8.4 mJ, respectively. A pumping energy of more than 90 mJ at 266 nm is necessary for DIAL measurement using the Stokes lines at 299 nm and 312 nm.

The four Stokes lines are expanded by a factor of about 3.9 by a 50 mm-diameter quartz lens with a focal length f_2=4290 mm. The beam divergence of the four Stokes lines transmitted into the atmosphere is about 0.1 mrad. The full field of views of the 49 cm and 10 cm telescope are 1.0 and 3.0 mrad, respectively. The system uses PMTs (Hamamatsu R3235) to detect the backscattered light from the atmosphere. For signal processing, a 12-bit A/D converter and photon counter (Licel TR20-160) are used. The timing of the DIAL system is controlled by a delay/pulse generator (Stanford Research Systems DG645). The specifications of this ozone DIAL system are summarized in Table 1.

The ozone number density is derived from the DIAL signals at the two wavelengths (Uchino and Tabata, 1991). The raw data were obtained with 7.5 m range resolution and 1 min integration time (i.e., 600 shots). The analog and photon counting signals for each receiving wavelength channel in Fig. 1 were connected to get high dynamic range. To increase the signal-to-noise ratio, the integrated spatial range interval (Δz) was varied from 75 to 375 m with increasing altitude. Ozone number concentration profiles were computed from the differential of the logarithm of the ratio of the signals by fitting a third-order polynomial to nine adjacent signals by the least-mean-squares method (Fujimoto and Uchino, 1994). The effective vertical resolution was about $9 \cdot \Delta z \cdot 0.4 = 3.6 \cdot \Delta z$, which was estimated from the full-width half-maximum of the ozone profiles retrieved when an impulse function of ozone density was input (Beyerle and McDermid, 1999). To calculate atmospheric molecular extinction, we used the atmospheric molecular extinction cross section (Bucholtz, 1995) and the atmospheric molecular density obtained from radiosonde data at the Fukuoka District Meteorological Observatory (33.58°N, 130.38°E). The ozone absorption cross sections were calculated by taking temperature dependence into account (Bass, 1984). The ozone volume mixing ratio was also calculated from the molecular density, including water vapor.

An example of DIAL measurements using four wavelengths, including 312 nm, is shown in Fig. 2. In this case, vertical profiles of the ozone number density and volume mixing ratio are plotted together with their statistical errors (precision) calculated from the lidar signal-to-noise ratios (Uchino and Tabata, 1991). A different wavelength pair was used for each of three altitude ranges to adapt to different ozone optical depths: 276/288 nm for 1.0–3.0 km (Δz = 75 m), 287/299 nm for 1.6–8.0 km (Δz = 150 m) and 8.0–18.6 km (Δz = 375 m), and 299/312 nm for 17.4–20.8 km (Δz = 375 m). The integration time was about 6 h to observe ozone vertical profiles up to 20 km (Fig. 2), but ozone vertical profiles up to 6 km were usually obtained within 30 min. From January through September 2012, we generally used an integration time of about 1–3 h

for biaxial and coaxial DIAL measurements, thus the resulting measurement errors were larger than those shown in Fig. 2. In this paper, we present the 2012 ozone data for altitudes lower than 6 km, measured by the DIAL system using three wavelengths (276, 287, and 299 nm). In this DIAL data analysis, the systematic uncertainty (accuracy) due to particulate backscatter and extinction was not taken into account. However, systematic uncertainty is probably on the order of 10–15 % in the planetary boundary layer and smaller than that in the free troposphere (Nakazato et al., 2007), although it can reach high values due to the presence of high aerosol loadings and the large aerosol concentration gradient at the top of the PBL (Papayannis et al., 1990).

We compared our DIAL measurement with ozonesonde data (Thompson et al., 2011) (Fig. 3). The ozonesondes used were composed of an ECC ozone sensor (ENSCI-Z) and a GPS radiosonde (Meisei RS-06G). The precision and accuracy of the ozone sensor were ±4 % and ±5 % at 1000 hPa and ±12 % and ±12 % at 200 hPa, respectively, and the vertical resolution of the sensor was 300 m (http://www.dropletmeasurement.com/products/airborne/ECC_Ozonesonde). Two ozonesondes were launched by the Japan Weather Association under contract with NIES, and the measurements used in our comparison were made on 9 and 15 January 2013. The ozonesonde and DIAL measurement times were from 12:31 to 13:53 LT and from 12:46 to 13:30 LT, respectively, on 9 January. On 15 January, the ozonesonde measurement time was from 12:31 to 14:04 LT under cloudy conditions, and we used DIAL data obtained from 14:00 to 17:04 LT under comparatively clear weather conditions for the comparison. The vertical resolution of the DIAL measurement was 110 m in the lower altitude range and 830 m in the upper altitude range. Within their measurement errors, the ozonesonde and DIAL data were consistent below an altitude of 6 km (Fig. 3). The average and one standard deviation of the difference between DIAL and ozonesonde data are 14±10 % below 2 km, 6±4 % for 2–7 km, and 20±13 % above 7 km.

3 MRI-CCM2

The MRI-CCM2 model uses chemical and physical processes from the surface to the stratosphere to simulate the global distribution and evolution of ozone and other trace gases (Deushi and Shibata, 2011). The previous version (MRI-CCM1) was a stratospheric chemistry-climate model (Shibata et al., 2005). Version 2, however, incorporates an elaborated mechanism for HO_x-NO_x-CH_4-CO photochemistry and

mechanisms for the degradation of non-methane hydrocarbons and heterogeneous tropospheric reactions of aerosols, so it can simulate tropospheric ozone chemistry as well as stratospheric chemistry. Deushi and Shibata (2011) showed that MRI-CCM2 can reproduce reasonably well the observed seasonal variations of the monthly means of ozone and carbon monoxide in the troposphere. This model is currently used to predict the distribution of photochemical oxidants near the surface in support of operational air-quality forecasts of the Japan Meteorological Agency (JMA). As first step toward better air quality prediction using the model, it is necessary to evaluate MRI-CCM2 by various observational data. It is important to compare ozone data predicted by the model with not only surface ozone data but also vertical ozone data.

The chemistry module of MRI-CCM2 considers 90 chemical species, 231 homogeneous gas-phase chemical reactions (172 chemical kinetic reactions and 59 photolysis reactions), and 16 heterogeneous reactions, and it incorporates grid-scale transport with a semi-Lagrangian scheme, sub-grid-scale convective transport and turbulent diffusion, dry and wet deposition, and emissions of trace gases from various sources. The chemistry module is coupled with an MRI atmospheric general circulation model (MRI-AGCM3; Yukimoto et al., 2011) via a simple coupler (Yoshimura and Yukimoto, 2008). The chemistry module receives meteorological and radiation fields and surface conditions from MRI-AGCM3. The horizontal coordinate system of MRI-CCM2 is a Gaussian grid with a resolution of about 110 km. In the vertical, a hybrid p-σ coordinate system is used, with 48 layers from the surface to the top of the atmosphere (0.01 hPa ≈ 80 km). In the hybrid p-σ coordinate system, the vertical coordinate is a terrain-following σ-coordinate ($\sigma = p/p_s$) near the surface, and it gradually changes to a pressure coordinate (p-coordinate) near the top. The vertical resolution increases from about 100 to 600 m from the surface to 6 km. The time step of the transport (chemistry) scheme is 30 (15) minutes.

In this study, the horizontal wind field in AGCM3 was forced toward the observed field by using a nudging assimilation with an 18 h e-folding time. JMA operational analysis data were used as the observed wind field for the nudging assimilation. In the chemistry module, trace gas emissions from the burning of fossil fuels by industrial activities and aircraft (anthropogenic sources), from biomass burning (anthropogenic and natural sources), and from vegetation, soils, and the ocean (natural sources) were prescribed. Global anthropogenic emission data, except those from East Asia, were obtained from the inventory in the Emission Database for Global Atmospheric Research (EDGAR) v2.0 (Olivier et al., 1996) modified by the seasonal adjustments of Müller (1992). For East Asia, the prescribed data were obtained from an inventory of Asian

anthropogenic emissions, the Regional Emission inventory in ASia (REAS), version 1.1 (Ohara et al., 2007). Biogenic and oceanic emissions were taken from Horowitz et al. (2003) and references therein. Emission of NO_X from lightning was diagnosed at 6 h intervals in the chemistry module by using the meteorological fields from AGCM3 (for details, see Deushi and Shibata, 2011). Emission of trace gases from biomass burning, which depends on the height above ground, is based on the Description of EDGAR 32FT2000(v8) (http://www.rivm.nl/edgar/Images/Description_of_EDGAR_32FT2000(v8)_tcm32-22222.pdf). Present-day concentrations of N_2O, CH_4, chlorofluorocarbons, and halons were prescribed at the surface. In this paper we used the data output by the model every hour for the comparison with the DIAL-observed data in this study.

4 DIAL observational results and comparison with MRI-CCM2 output

Hourly grid-point ozone data predicted by MRI-CCM2 were spatially interpolated for the Saga area, averaged over the DIAL measurement time (about 1–7 hrs), and then compared to our DIAL ozone data.

4.1 Vertical profiles

At first the vertical profiles of lower tropospheric ozone in the altitude range of 1−6 km measured by DIAL between January and May 2012 are compared with MRI-CCM2 predictions (Fig. 4). The DIAL-measured ozone volume mixing ratios on 30 January, being of about 40–50 ppbv were successfully predicted by MRI-CCM2, except in the 4−5 km altitude range. High ozone values were measured at around 2 km altitude in the daytime on 23 April, 5 May, and 11 May. A backward trajectory analysis suggested that the air masses linked with these high ozone volume mixing ratios at 2 km were transported to Saga across the Yellow Sea (23 April and 5 May) or over the Korean Peninsula (11 May) from polluted areas in East China (Parrish and Zhu, 2009) within a few days. MRI-CCM2 predicted well these high ozone densities on 23 April, but not those on 5 May or 11 May. This is possibly in part because emissions were assumed in the model to be constant (climatological data were used).

Figure 5 shows ozone profiles in the 1−6 km altitude range for July and September 2012. MRI-CCM2 was unable to predict the high ozone values of more than 70 ppbv above 2.3 km that were measured by DIAL on 7 July. On 19 July, the measured ozone

volume mixing ratios were 10–30 ppbv below about 4 km, whereas MRI-CCM2 predicted ratios of 30–40 ppbv below 4 km. On 28 July, however, MRI-CCM2 predicted well the ozone volume mixing ratios measured by DIAL of about 30–40 ppbv below 4 km. On 26 September, MRI-CCM2 predicted very well the ozone volume mixing ratios measured by DIAL of about 35–70 ppbv from 500 m to 6 km. According to the backward trajectory analysis, the air masses below 4 km with low ozone volume mixing ratios of about 20–40 ppbv on 19 and 28 July came from the Pacific Ocean south of Saga. The ozone volume mixing ratios predicted by MRI-CCM2 for 2−6 km on 7 July were lower, and those for 1−4 km on 19 July were higher, compared with the DIAL measurements.

Figure 6 shows horizontal maps of ozone volume mixing ratio and horizontal wind predicted by MRI-CCM2 for 500 hPa (about 5.5 km altitude), 700 hPa (3 km), 800 hPa (2 km), and 850 hPa (1.5 km) pressure levels at 06 UTC on 7 July 2012. According to the weather chart on the same day provided by JMA (http://www.data.jma.go.jp/fcd/yoho/hibiten/index.html), the Meiyu-Baiu-front extended from west to east through the center of a low-pressure system over Japan. In this prediction, the synoptic-scale pattern of horizontal-wind field is reproduced reasonably. A large north-south gradient of ozone volume mixing ratios is located near the stationary front in the lower free-troposphere. The ozone concentration is low over the south part of the front because of the chemical destruction in the summertime marine air mass, whereas high ozone level area is present over the north of the front. During the DIAL observing period the large gradient of ozone concentration was located near the Saga site in the lower free-troposphere (Fig.6). This suggests that one of the possible reasons for the significant discrepancy between the DIAL ozone profile and the predicted one (Fig. 5) is that the meso-scale front structure and its position are not fully captured by the model due to its coarse spatial resolution. In addition, it is plausible that model transport with the coarse grid doesn't reproduce well fine-scale structures of air (trace gases) filaments.

4.2 Lower tropospheric ozone column

Figure 7 shows time variations of the lower tropospheric ozone column amounts within the 1−6 km and 1−2 km altitude ranges over Saga in 2012 as measured by DIAL and predicted by MRI-CCM2. Because of restricting weather conditions, DIAL measurements were obtained only twice in May and twice in June, but at least three measurements were obtained in each of the other months. In 2012, the ozone column amounts showed an approximately seasonal variation except for a dip from early July

through mid-September. Column amounts within 1–6 km were almost constant from January to early March, and then increased until late June. The maximum value in the 1–6 km range was 7.82×10^{21} molecules m^{-2}, corresponding to 29.1 DU (1 DU = 2.688×10^{20} molecules m^{-2}), on 22 June 2012. Thereafter, ozone column amounts within the 1–6 km range decreased to early August, but with large variations. The minimum value within the 1–6 km range of 2.28×10^{21} molecules m^{-2} (8.5 DU) was observed on 15 August 2012. These variations are due to possibly exchange events between the ozone-rich continental and the ozone-poor maritime air masses. Subsequently, ozone column amounts increased again to mid-September, and then decreased slowly, becoming almost constant by December, when the ozone column amount within the 1–6 km range was 4.65×10^{21} molecules m^{-2} (17.3 DU). The results predicted by MRI-CCM2 were mainly consistent with our DIAL observational results.

The temporal variation of the ozone column values within the 1–2 km range in 2012 was similar to that in the 1–6 km range. The mean value was about 1.03×10^{21} molecules m^{-2} (3.8 DU) from 20 January to 11 February. Then, the ozone column amount increased to a maximum value of 2.02×10^{21} molecules m^{-2} (7.5 DU) on 5 May, followed by a decreased value down to 0.32×10^{21} molecules m^{-2} (1.2 DU) on 19 July. Moreover the mean ozone column amount within the 1–2 km range was about 0.55×10^{21} molecules m^{-2} (2.0 DU) from 28 July to 18 August. Subsequently, the column ozone value increased to more than 1.0×10^{21} molecules m^{-2} (3.7 DU) by 19 December. MRI-CCM2 predicted similar variations of the ozone column amounts within the 1–2 km range, but the predicted values were lower than the DIAL measurements on 5 and 11 May and 22 June.

5 Concluding remarks

With our improved DIAL system, 44 ozone profiles were observed in the 1–6 km altitude range over Saga in 2012. High ozone layers were observed at around 2 km altitude during April and May. Ozone column amounts within the 1–6 km altitude range were almost constant (19.1 DU) from January to March, and increased to 26.7 DU from late April to July. From mid-July through August, ozone column amounts showed large variations (8.5–22.0 DU), attributed to exchanges between continental and maritime air masses. Ozone column amounts within the 1–6 km range increased again to mid-September, then decreased slowly to 17.3 DU and became almost constant through December.

MRI-CCM2 successfully predicted these ozone variations with the following exceptions. MRI-CCM2 could not predict the high ozone volume mixing ratios measured at around 2 km altitude on 5 May and 11 May, possibly in part because emissions (especially from open biomass burning) were assumed in the model to be constant (climatological data were used). Ozone volume mixing ratios predicted by MRI-CCM2 were low in the 2–6 km range on 7 July and high in the 1–4 km range on 19 July compared with those measured by DIAL.

A major benefit of ozone DIAL is that it can measure continuously vertical ozone profiles with high vertical and temporal resolution. The process of stratospheric ozone intrusion into the troposphere was clearly observed on 20-21 June 2001 by continuous DIAL measurement at Garmisch-partenkichen, Germany and the excellent data set was used for model intercomparisons and validation (Roelofs et al., 2003; Zanis et al., 2003; Stohl et al., 2003). On the other hand, the ability of a regional air quality model was evaluated to reproduce summertime ozone pollution over the Kanto region in Japan using quasi-continuous ozone DIAL measurement data (Niwano et al, 2007). We plan to study the process that air mass of high ozone concentration in the free troposphere is mixed into the planetary boundary layer by continuous DIAL measurement. Further, we plan to utilize ozone DIAL data for validation of a new version of an MRI oxidant prediction system over an extended period of time. In the prediction system, a regional chemistry-transport model, known as MRI-PM/c (Kajino et al., 2012), with higher spatial resolution (20 km and/or 5 km) is nested in MRI-CCM2. The prediction system also adopts an up-to-date inventory and higher temporal resolution of biomass burning emissions. The regional model can reproduce meso-scale weather phenomena and advection of air mass. However, if boundary data for the regional model produced by the global model doesn't include adequate information on fine-scale structures in trace gas distributions, the performance of the prediction system will be degraded. Therefore, in order to improve total performance of the prediction system, it is necessary to validate not only the regional model but also the global model with observation data such as ozone DIAL data and surface ozone monitoring data extensively, which enable us to estimate error coming from outside of the boundary.

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Table 1. Characteristics of tropospheric ozone DIAL system

Transmitter					
Pump laser	Nd:YAG				
Wavelength	266 nm				
Pulse energy	107 mJ				
Pulse repetition rate	10 Hz				
Pulse width	8 ns				
Raman active gas	CO_2				
Stokes lines	276 nm	287 nm	299 nm	312 nm	
Pulse energy	7.5 mJ	9.1 mJ	8.4 mJ	No. meas.	
Beam divergence	0.1 mrad				
Receiver					
Telescope type	Newtonian			Prime focus (fiber coupled)	
Telescope diameter	49 cm			10 cm	
Focal length	1750 mm			320 mm	
Field of view	1 mrad			3 mrad	
Wavelength	287 nm	299 nm	312 nm	276 nm	287 nm
Bandwidth	1.02 nm	1.15 nm	0.82 nm	1.07 nm	1.05 nm
Transmission	0.18	0.32	0.36	0.17	0.21
Detector	PMT (Hamamatsu R3235-01)				
Signal processing	12bit A/D + Photon counting				
Time resolution	1 min				
Altitude resolution	7.5 m				

Figure captions

Fig. 1. Block diagram of tropospheric ozone DIAL system.

Fig. 2. Vertical profiles of ozone number density (left panel) and ozone volume mixing ratio (right panel) over Saga observed by DIAL on 20 October 2012. The error bars show the statistical errors calculated from the lidar signal to noise ratios.

Fig. 3. Comparison of ozone DIAL (black) and ECC ozonesonde (orange) measurements on 9 (left panel) and 15 January (right panel) in 2013. The error bars of ozone DIAL data show the statistical errors calculated from the lidar signal to noise ratios.

Fig. 4. Vertical profiles of ozone volume mixing ratios over Saga measured by DIAL (black lines) and predicted by MRI-CCM2 (orange lines) on 30 January, 23 April, 5 and 11 May in 2012. The error bars shows the statistical errors calculated from the signal to noise ratios for the lidar and the standard deviation for the period predicted by MRI-CCM2.

Fig. 5. Same as Fig. 4 but for 7, 19, and 28 July, and 26 September in 2012.

Fig. 6. Horizotal maps of ozone volume mixing ratio in ppbv and horizontal wind in m/s predicted by MRI-CCM2 for 500, 700, 800, 850 hPa pressure levels at 06 h UTC on 7 July 2012.

Fig. 7. Ozone columns for 1–6 km and 1–2 km altitude ranges over Saga in 2012 measured by DIAL and predicted by MRI-CCM2.

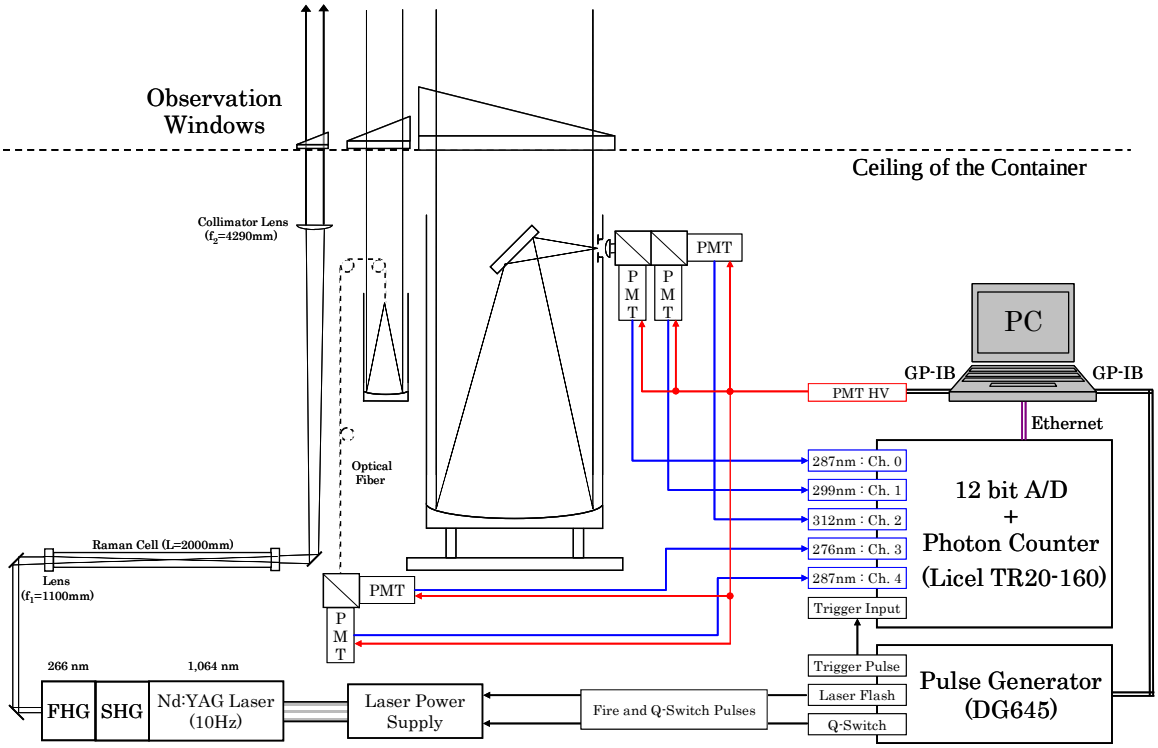
Observation Windows
Ceiling of the Container
Collimator Lens
(f_2=4290mm)
PMT
P M T
P M T
PC
GP-IB
GP-IB
PMT HV
Ethernet
Optical Fiber
287nm : Ch. 0
299nm : Ch. 1
312nm : Ch. 2
276nm : Ch. 3
287nm : Ch. 4
Trigger Input
12 bit A/D
+
Photon Counter
(Licel TR20-160)
Raman Cell (L=2000mm)
Lens
(f_1=1100mm)
PMT
P M T
266 nm
1,064 nm
FHG
SHG
Nd:YAG Laser
(10Hz)
Laser Power Supply
Fire and Q-Switch Pulses
Trigger Pulse
Laser Flash
Q-Switch
Pulse Generator
(DG645)

Fig. 1.

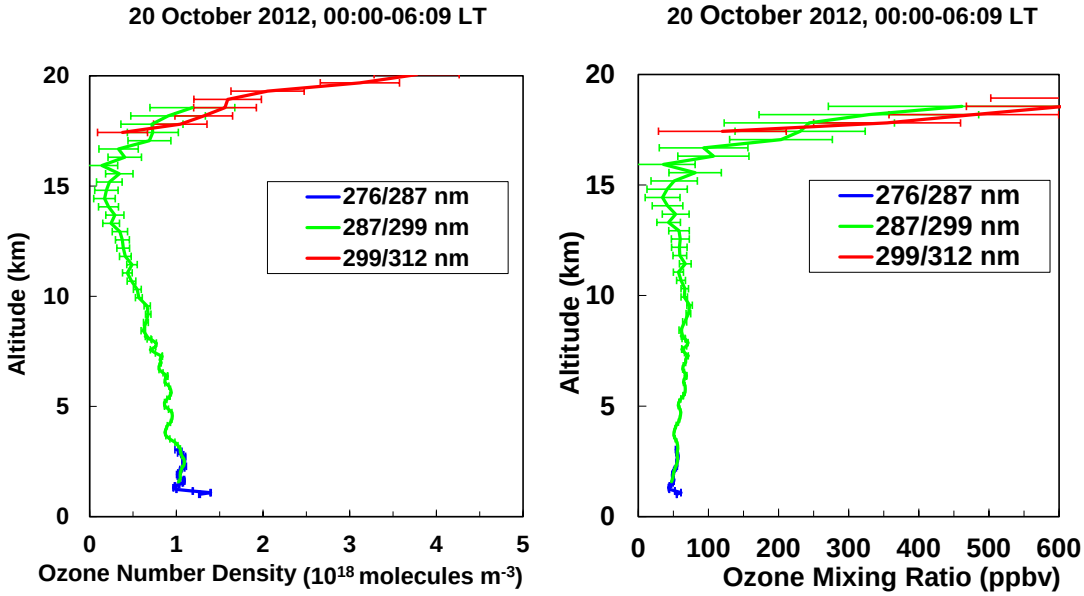
20 October 2012, 00:00-06:09 LT
Altitude (km)
Ozone Number Density (10^18 molecules m^-3)
276/287 nm
287/299 nm
299/312 nm
20 October 2012, 00:00-06:09 LT
Altitude (km)
Ozone Mixing Ratio (ppbv)
276/287 nm
287/299 nm
299/312 nm

Fig.2

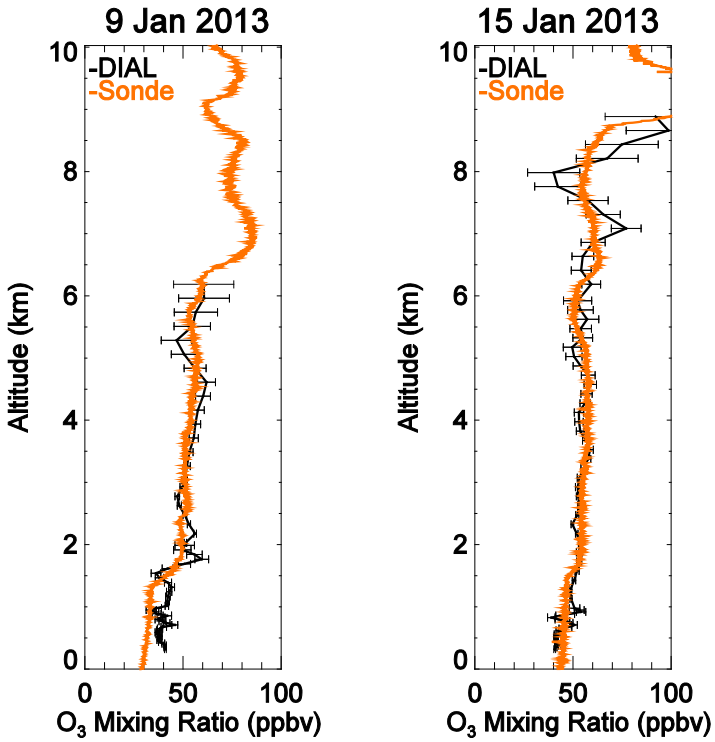
9 Jan 2013
15 Jan 2013
-DIAL
-Sonde
Altitude (km)
O3 Mixing Ratio (ppbv)
0
2
4
6
8
10
0
50
100

Fig. 3.

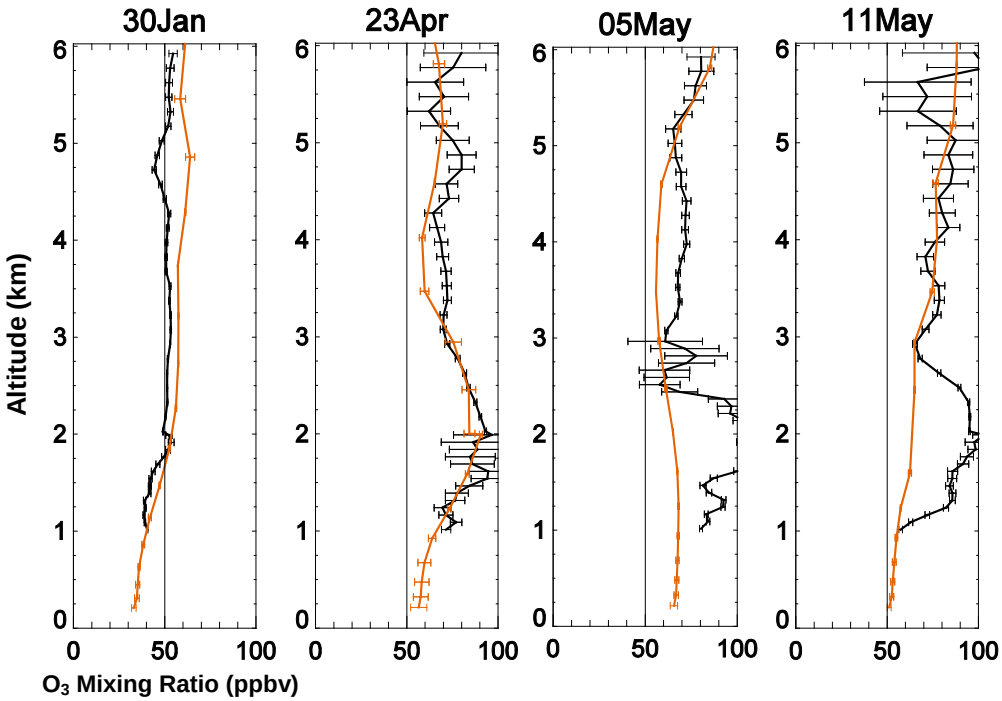
30Jan
23Apr
05May
11May
Altitude (km)
O3 Mixing Ratio (ppbv)
0
1
2
3
4
5
6
0
50
100

Fig. 4.

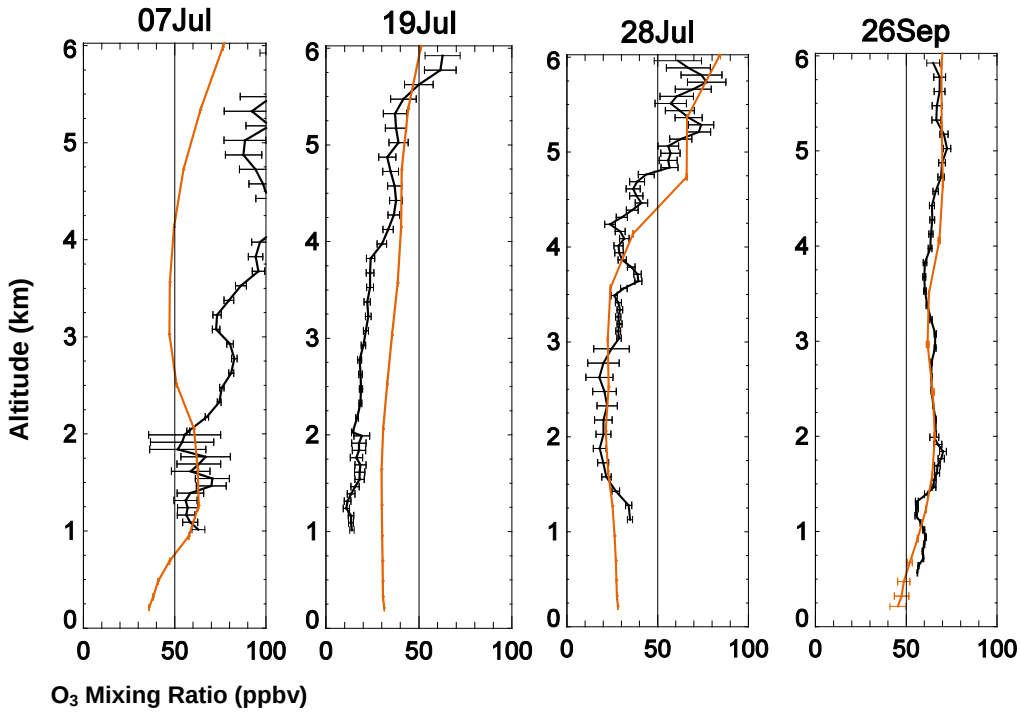
07Jul
19Jul
28Jul
26Sep
Altitude (km)
O3 Mixing Ratio (ppbv)
0
1
2
3
4
5
6
0
50
100

Fig. 5.

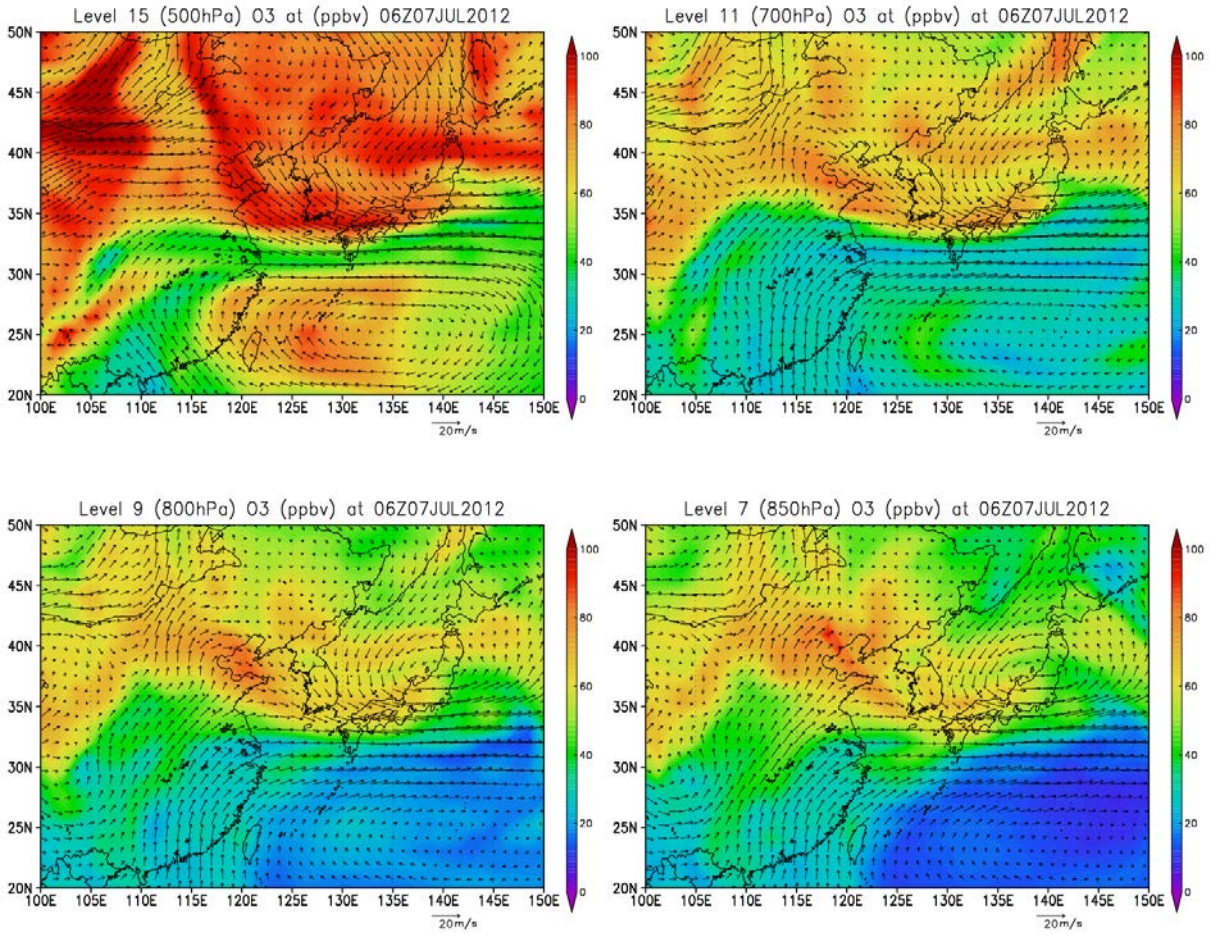
Level 15 (500hPa) O3 at (ppbv) 06Z07JUL2012
Level 11 (700hPa) O3 at (ppbv) at 06Z07JUL2012
Level 9 (800hPa) O3 (ppbv) at 06Z07JUL2012
Level 7 (850hPa) O3 (ppbv) at 06Z07JUL2012
50N
45N
40N
35N
30N
25N
20N
100E
105E
110E
115E
120E
125E
130E
135E
140E
145E
150E
20m/s
100
80
60
40
20
0

Fig. 6.

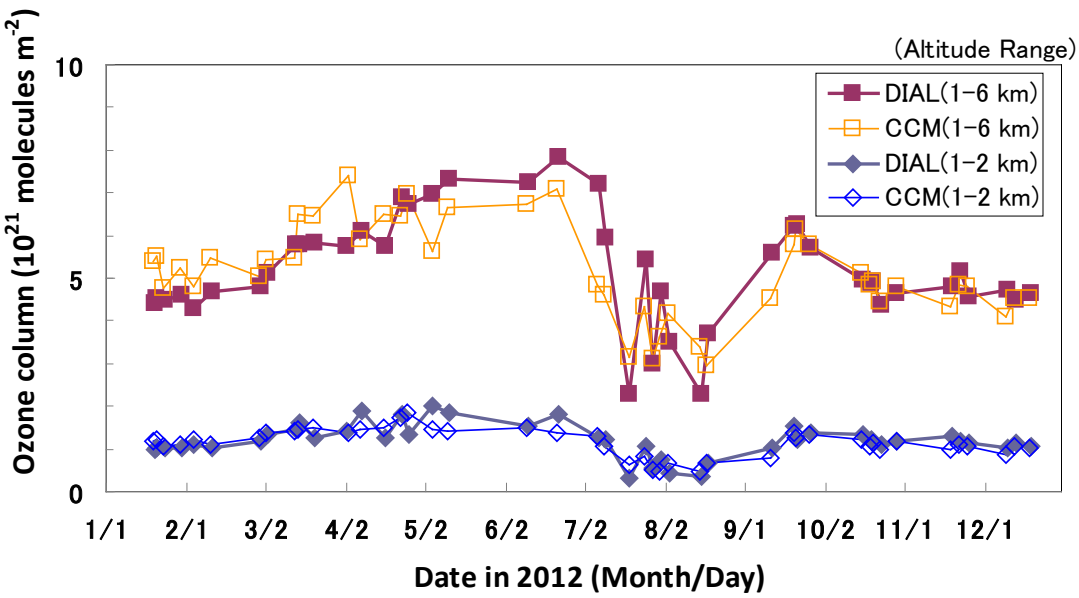
(Altitude Range)
DIAL(1-6 km)
CCM(1-6 km)
DIAL(1-2 km)
CCM(1-2 km)
Ozone column (10^{21} molecules m^{-2})
10
5
0
1/1
2/1
3/2
4/2
5/2
6/2
7/2
8/2
9/1
10/2
11/1
12/1
Date in 2012 (Month/Day)

Fig. 7.