



## Lidar detection of high concentrations of ozone and aerosol transported from Northeast Asia over Saga, Japan

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**Abstract.** To validate products of the Greenhouse gases Observing SATellite (GOSAT), we observed vertical profiles of  
aerosols, thin cirrus clouds, and tropospheric ozone with a mobile lidar system that consisted of a two-wavelength (532 and  
1064 nm) polarization lidar and a tropospheric ozone Differential Absorption Lidar (DIAL). We used these lidars to make  
continuous measurements over Saga (33.24°N, 130.29°E) during 20–31 March 2015. High ozone and high aerosol  
20 concentrations were observed almost simultaneously in the altitude range 0.5–1.5 km from 03:00 to 20:00 Japan Standard  
Time on 22 March 2015. The maximum ozone volume mixing ratio was ~110 ppbv. The maxima of the aerosol extinction  
coefficient and optical depth at 532 nm were 1.2 km<sup>-1</sup> and 2.1, respectively. Backward trajectory analysis indicated that an  
air mass with high ozone and aerosol concentrations could have been transported from Northeast Asia. Based on the lidar  
data and the ground-based in-situ measurements at Saga, this air mass could have been transported to the surface by vertical  
25 mixing when the planetary boundary layer developed in the daytime. This plume, which contained high ozone and aerosol  
pollutant concentrations, impacted surface air quality substantially. After some modifications of its physical and chemical  
parameters, the Meteorological Research Institute Chemistry-Climate Model, version 2 (MRI-CCM2) approximately  
reproduced the high-ozone volume-mixing ratio. The Model of Aerosol Species IN the Global Atmosphere (MASINGAR)  
mk-2 successfully predicted high aerosol concentrations, but the predicted peak aerosol optical thickness was about one-third  
30 of the observed value.



## 1 Introduction

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Tropospheric ozone is a major air pollutant and impacts human health and vegetation (HTAP, 2010; Yue and Unger, 2014). It is also an important greenhouse gas (IPCC, 2013). Tropospheric aerosols are also air pollutants and aggravate respiratory conditions (HTAP, 2010). Tropospheric aerosols also enhance radiative forcing in a negative (sulfuric acid particles) or positive (black carbon) way (IPCC, 2013), and they affect remote sensing such as the measurement of greenhouse gases from space (Houweling et al., 2005; Uchino et al., 2012a). It is therefore very important to monitor tropospheric ozone and aerosols and to understand their temporal and spatial variations.

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To validate products of the Greenhouse gases Observing SATellite (GOSAT), we developed a two-wavelength (532 and 1064 nm) polarization lidar (hereafter abbreviated as Mie lidar) to observe vertical profiles of tropospheric and stratospheric aerosols and thin cirrus clouds at the National Institute for Environmental Studies (NIES), Tsukuba (36.05°N, 140.13°E), Japan in 2009. In 2010 we also developed a Differential Absorption Lidar (DIAL) to measure tropospheric ozone profiles (hereafter abbreviated as ozone DIAL). The ozone DIAL was installed in a container with the Mie lidar. In March 2011, we moved the lidar container to Saga (33.24°N, 130.29°E) in the Kyushu district of western Japan at a location 2.6 m above sea level. The ozone DIAL was modified in September 2012 (Uchino et al., 2014).

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Mie lidar has been used to demonstrate the influence of high-altitude aerosols and cirrus clouds on the GOSAT product of the column-averaged dry air mole fraction of carbon dioxide ( $X_{CO_2}$ ) retrieved from the Thermal And Near infrared Sensor for carbon Observation-Fourier Transform Spectrometer (TANSO-FTS) Short-Wavelength InfraRed (SWIR) spectral data onboard GOSAT. The  $X_{CO_2}$  data were improved by taking the vertical profiles of aerosols and cirrus clouds measured by Mie lidar into account (Uchino et al., 2012a). The increases of stratospheric aerosols caused by the 2009 Sarychev eruption and the 2011 Nabro eruption were observed by Mie lidar (Uchino et al., 2012b).

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Ozone DIAL has been used to validate the GOSAT ozone product retrieved from TANSO-FTS Thermal InfraRed (TIR) spectral data (Ohyama et al., 2012), to observe ozone concentrations in the lower troposphere, and to compare the observed concentrations with those predicted by the Meteorological Research Institute Chemistry-Climate Model, version 2 (MRI-CCM2) (Deushi and Shibata, 2011). Use of Mie lidar and ozone DIAL will facilitate satellite product validation not only for GOSAT but also for upcoming satellites such as the Tropospheric Monitoring Instrument (TROPOMI, Veefkind et al., 2012) and the Geostationary Environment Monitoring Spectroscopy (GEMS, Bak et al., 2013).

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In this paper we report an event during which high concentrations of ozone and aerosols were observed almost simultaneously below an altitude of 1.5 km over Saga on 22 March 2015, which substantially impacted surface air quality. We also compared the observational results with those simulated by the models.



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## 2 Lidar system and observations

Mie lidar and ozone DIAL were installed in a container with dimensions of about 228 cm (width), 683 cm (length), and 255 cm (height), as shown in Fig. 1. Mie lidar is a two-wavelength (532 and 1064 nm) polarization lidar based on a neodymium-doped yttrium-aluminum-garnet (Nd:YAG) laser; the characteristics are summarized in Table 1. The output energy at 532 and 1064 nm was 130 mJ, with a pulse repetition rate of 10 Hz. The diameter of the receiving telescope was 30.5 cm. The output signals from the photomultiplier tubes (PMT) and a silicon avalanche photodiode (APD) were processed by transient recorders with a 12-bit analog/digital converter and a photon counter.

The ozone DIAL consisted of a Nd:YAG laser and a 2-m-long Raman cell filled with CO<sub>2</sub> gas that generated four Stokes lines from stimulated Raman scattering by CO<sub>2</sub>; the characteristics are summarized in Table 2. In this study, we used three Stokes lines (276, 287, and 299 nm). The output energies of these Stokes lines were about 8–9 mJ per pulse, with a pulse repetition rate of 10 Hz. The receiving telescope diameters were 10 cm for boundary layer ozone measurements and 49 cm for free tropospheric ozone measurements. The output signals from the PMTs were also processed by transient recorders with a 12-bit analog/digital converter and a photon counter. The data analysis methods of Mie lidar and ozone DIAL have been described by Uchino et al. (2012b) and Uchino et al. (2014), respectively. The Mie lidar and ozone DIAL were synchronized by two pulse-delay generators. Next, we report the continuous lidar observational results made at Saga from 20 March to 31 March 2015.

## 3 Ozone DIAL data

Figure 2a shows a time-altitude cross-section of ozone volume mixing ratios observed by DIAL at Saga from 11:10 JST on 20 March to 14:33 JST on 31 March 2015. Lidar observations were not obtained from 15:56 JST on 27 March to 21:58 JST on 29 March 2015, mainly because of bad weather conditions. The vertical resolution was 270 m between altitudes of 450 m and 2 km and 540 m between altitudes of 2 and 6 km. The time resolution was set to 1 h to facilitate comparison with the MRI-CCM2. We made quality checks of the DIAL data. The gray regions in Fig. 2a correspond to areas where there were no observational data or the errors were larger than 10%. The errors were equated to 100% times the lidar signal-to-noise ratios. Regions surrounded by a black rectangle are areas where the data were affected by aerosols and/or clouds. In the lowest row of Fig. 2a, we show hourly data of surface oxidant volume mixing ratios (Ox) at Takagimachi in Saga city measured by the Saga Prefectural Environmental Research Center ([https://www.pref.saga.lg.jp/web/at-contents/kankyo1/shisetsu/\\_40810/\\_41304/\\_67819.html](https://www.pref.saga.lg.jp/web/at-contents/kankyo1/shisetsu/_40810/_41304/_67819.html)). Takagimachi is located about 2.8 km northeast from the ozone DIAL site. Because the contribution of other components such as peroxyacetylene nitrate (PAN) to oxidant concentrations was



extremely low, the oxidant volume mixing ratio was considered to be that of ozone.

Figure 2a indicates that the ozone volume mixing ratios measured by DIAL were usually about 50–70 ppbv during the study period. Comparatively high ozone concentrations, >75 ppbv, were detected at altitudes of 0.5–3 and 0.5–2 km on 20–23 March and 30–31 March, respectively. Notably high ozone volume mixing ratios of 90–110 ppbv at altitudes of 0.5–1.5 km were observed from 03:00 to 20:00 JST on 22 March. These high ozone concentrations were also seen in the surface photochemical oxidants data, i.e., the Ox equaled 92–101 ppbv from 15:00 to 21:00 JST on 22 March, as shown in the lowest row in Fig. 2a. The maximum concentration of Ox was 101 ppbv at 16:00 JST. This maximum value was far above the environmental quality standard of 60 ppbv for hourly photochemical oxidants in Japan (<https://www.env.go.jp/en/air/aq/aq.html>). These observational results show the descent of regions of high ozone concentrations and suggest that air with high ozone concentrations was transported to the surface by vertical mixing when the planetary boundary layer developed during the daytime.

### 3.1 Comparison of DIAL data with MRI CCM-2

The MRI-CCM2 is a global model that simulates chemical and physical processes that affect the distribution and evolution of ozone and other trace gases from the surface to the stratosphere (Deushi and Shibata, 2011). Uchino et al. (2014) have provided an outline of MRI-CCM2. The vertical resolution of the model increases from about 100 to 600 m from the surface to 6 km. The time step of the transport (chemistry) scheme is 30 (15) min. We used hourly model output data. The horizontal resolution is about 110 km. We examined whether or not the model could simulate DIAL observational results. The MRI-CCM2 simulated the DIAL observations reasonably well. However, the MRI-CCM2 could not reproduce the high ozone concentrations of 90–110 ppbv observed below an altitude of 1.5 km during 03:00–20:00 JST on 22 March 2015.

We therefore performed some simulations in which we changed the emission inventory data and the term that forced the reanalysis wind field. The most reasonable results were obtained when the following changes were made (Fig. 2b). The e-folding time of the nudging term was changed from 18 hours to 12 hours to more strongly force the simulated wind fields toward the reanalysis data. In addition, we changed the emission inventory of Regional Emission inventory in Asia version 1.1 (REAS 1.1) (Ohara et al., 2007) to REAS 2.1 (Kurokawa et al., 2013) and the NO<sub>2</sub>/NO<sub>x</sub> emissions ratio from 5% to 15% by volume, which is within the range of uncertainty (Carslaw, 2005). The emission inventory of NO<sub>x</sub> increased about 50% from REAS 1.1 to REAS 2.1. Figure 2c shows the differences between the observed and simulated ozone mixing ratios. Simulated ozone volume mixing ratios were about 60–70 ppbv below an altitude of 1.5 km from 14:00 JST on 21 March to 21:00 JST on 22 March 2015, lower by about 20–50 ppbv compared with the DIAL results. A model with higher horizontal resolution might be necessary to more realistically simulate high surface ozone concentration events in the planetary boundary layer.



#### 4 Mie lidar data

First, we summarize the physical parameters obtained by Mie lidar. The backscattering ratio  $R$  is defined as

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$$R = (BR + BA)/BR, \quad (1)$$

where  $BR$  and  $BA$  are the Rayleigh and Mie backscattering coefficients, respectively. Backscattering ratio profiles were derived by the inversion method (Fernald, 1984). We assumed the lidar ratio  $S$  (extinction-to-backscatter ratio) for aerosols to be 50 sr at 532 nm and 45 sr at 1064 nm (Sakai et al., 2003; Cattrall et al., 2005). To calculate  $BR$ , we used the atmospheric molecular density profiles obtained by operational radiosondes at the Fukuoka District Meteorological Observatory (33.58°N, 130.38°E). The aerosol extinction coefficient was calculated by multiplying  $BA$  by  $S$ .

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The total volume depolarization ratio  $D$  was defined as

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$$D = S / (P + S) \cdot 100 (\%), \quad (2)$$

where  $P$  and  $S$  are the parallel and perpendicular components of the backscattered signals, respectively. The particle depolarization ratio  $D_p$  was obtained from the equation

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$$D_p = (D \cdot R - D_m) / (R - 1), \quad (3)$$

where  $D_m$  is the atmospheric molecular depolarization ratio. We used a  $D_m$  value of 0.37% for this lidar system; we calculated  $D_m$  from the spectral transmission data of the interference filter at 532 nm and the Rayleigh backscattering cross sections (Sakai et al., 2003). The value of  $D_p$  indicates whether the particles are spherical or non-spherical; large values indicate the presence of non-spherical particles. The wavelength exponent,  $Alp$ , shows whether small or large particles account for most of the Mie particles and is defined by

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$$BA(\lambda) \propto \lambda^{-Alp}, \quad (4)$$

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where  $\lambda$  is the wavelength. Larger values of  $Alp$  indicate the predominance of smaller (i.e., submicrometer-sized) particles.

Figures 3a and 3b show time-altitude cross-sections of the backscattering ratio ( $R$ ) and the total volume depolarization ratio ( $D$ ), respectively, observed by Mie lidar at Saga from 11:10 JST on 20 March to 14:33 JST on 31 March 2015. Mie lidar observation were not obtained from 15:56 JST on 27 March to 21:58 JST on 29 March 2015, mainly because of bad



165 weather conditions. The vertical resolution was 150 m, and the time resolution was set to be 1 h for comparison with the Model of Aerosol Species in the Global Atmosphere (MASINGAR)-mk2 (Yukimoto et al., 2012). We made quality checks of Mie lidar data. Gray regions are areas where there were no observational data or the data were affected by clouds.

170 Aerosol layers with  $R$  in the range 2–4 almost always existed below an altitude of 2.5 km. An event of high aerosol loading with large values of  $R$  ( $\sim 4$ –19) was observed below altitudes of 1.5 km on 22 March, when the values of  $D$  were small (1–6%) compared with those before and after the event, when the values of  $D$  were larger than  $\sim 8\%$ . The main aerosol component during the event may have been submicrometer-sized spherical sulfate particles, because  $D_p$  was small (1–6%), and the wavelength exponent  $Alp$  was large ( $\sim 1.4$ ). In contrast, the main aerosol particles before and after the event were assumed to be supermicrometer-sized, nonspherical mineral dust particles because  $D_p$  was comparatively large ( $\sim 13\%$ ) and  $Alp$  was approximately 1.0.

175 Hourly values of the mass concentrations of particulate matter with a diameter of 2.5  $\mu\text{m}$  or less ( $\text{PM}_{2.5}$ ) at Takagimachi measured by the Saga Prefectural Environmental Research Center were 23  $\mu\text{g m}^{-3}$  at 10:00 JST and increased up to a maximum value of 110  $\mu\text{g m}^{-3}$  at 15:00 JST on 22 March; the concentrations were greater than 82  $\mu\text{g m}^{-3}$  during 13:00–16:00 JST and decreased to 17  $\mu\text{g m}^{-3}$  at 01:00 JST on 23 March. The daily mean value of  $\text{PM}_{2.5}$  was 50.6  $\mu\text{g m}^{-3}$  for 24 hours on 22 March at Takagimachi, larger than the environmental quality standard of 35  $\mu\text{g m}^{-3}$  in Japan (<https://www.env.go.jp/en/air/aq/aq.html>). Consideration of the lidar data and the temporal variation of  $\text{PM}_{2.5}$  at Takagimachi suggests that air with high concentrations of both aerosols and ozone was transported to the surface by vertical mixing when the planetary boundary layer developed in the daytime.

#### 4.1 Comparison of Mie lidar data with MASINGAR mk-2

185 The MASINGAR-mk2 is an improved version of the MASINGAR aerosol model (Tanaka et al., 2003); it treats five aerosol species: sulfate, black and organic carbon, sea salt, and soil dust. We used emission data for sulfur dioxide and for black and organic carbon from MACCity (Granier et al., 2011). Soil dust and sea salt were represented by 10 bins with particle diameters of 0.2–20  $\mu\text{m}$ . The model was coupled online with the atmospheric general circulation model MRI-AGCM3 (Yukimoto et al., 2012). The horizontal resolution of the MASINGAR-mk2 was about 60 km, and the number of vertical layers was 40 from the surface to 0.1 hPa. The vertical resolutions were 100, 300, and 600 m at the lowest level and altitudes of 1 and 6 km, respectively. The time step of the transport (chemistry) scheme was 450 seconds, and we used hourly model output data.

195 Figures 4a and 4b show the time-height cross sections of aerosol extinction coefficients observed by Mie lidar and simulated by MASINGAR-mk2, respectively. Figure 4c represents the difference between the observed and simulated extinction coefficients. The model was able to capture the general characteristics of the observational results rather well. A close look at Fig. 4c reveals that the model underestimated the aerosol extinction coefficients of the anthropogenic pollutant



event on 22 March but slightly overestimated the extinction coefficients associated with particles having larger total volume depolarization ratios on 30 and 31 March (i.e., dust-dominant case).

## 200 4.2 Comparison of aerosol optical depths

Figure 5 shows temporal variations of the aerosol optical depths (AOD) measured by Mie lidar at 532 nm and sky radiometer at 500 nm (Kobayashi et al., 2006, Uchino et al., 2012a) and simulated at 550 nm by MASINGAR-mk2 from 20 to 31 March. To estimate AODs from the lidar data, the extinction coefficient at 225 m was extrapolated to the ground, the extinction coefficient from 15 to 35 km was observed at night on the same day, and  $S$  was assumed to be 50 sr for all altitudes. When clouds and thick aerosols were present, AODs were not obtained. The sky radiometer was positioned on the roof of the building, which is four stories high and located to the west of the container (brown building in Fig. 1). Although it must be noted that the measured and simulated wavelengths differed slightly, the AODs were almost the same, except for the high aerosol and ozone event on 22 March. The maximum values of the AODs were 2.1 at 12:00 JST by lidar, 1.92 at 13:00 JST by sky radiometer, and 0.53 at 13:00 JST by MASINGAR-mk2. The model underestimated the AODs by factors of about 3.6–4 compared to the sky radiometer and lidar observations. One plausible explanation is that the model resolution (about 60 km) was insufficient to reproduce the observed prominent peak in which the observed AOD increased from 1.0 to 2.0 in 6 hours. In addition, inadequacies of the input anthropogenic emissions may partly explain the underestimate. To solve this problem, for example, it might be better to use the near real-time satellite data of sulfur dioxide and nitrogen dioxide provided by the Ozone Monitoring Instrument (OMI) onboard NASA's Aura satellite (Krotkov et al., 2016).

## 215 5 Discussion and concluding remarks

220 By using ozone DIAL and a two-wavelength polarization (Mie) lidar, we made continuous measurements of ozone and aerosol concentrations over Saga during 20–31 March 2015, with the exception of a period with bad weather conditions. High ozone and high aerosol concentrations that occurred nearly simultaneously were observed in the altitude range 0.5–1.5 km from 03:00 to 20:00 JST on 22 March 2015. The ozone volume mixing ratio was larger than 100 ppbv. The aerosol extinction coefficient and AOD at 532 nm were larger than  $0.5 \text{ km}^{-1}$  and 1.5, respectively.

225 To identify the origin of the ozone and aerosols and related transport processes, three-dimensional backward trajectories of air parcels were calculated with the NOAA Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPRIT) model (Draxler and Hess, 1998; Stein et al., 2015), as shown in Fig. 6. Twenty-seven air parcels were initially left at altitudes of 600–1000 m over the lidar site at Saga. The trajectories were calculated for three days from 00:00 UTC (09:00 JST) on 22 March 2015. Based on the results of the backward trajectories, the high ozone and aerosol concentrations on 22 and 31





230 March could have been transported from Northeast Asia. According to the ozonesonde measurements made by Wang et al.  
(2012), ozone concentrations  $\geq 90$  ppbv were observed over Beijing, China in late March. Ma et al. (2016) reported a  
significant increase of surface ozone from 2003 to 2015 at Shangdianzi (40.65°N, 117.10°E), which is located about 100 km  
northeast of suburban Beijing, and the maximum daily average 8-h concentrations of ozone appear to have been  $>100$  ppbv  
235 in March 2015 based on Fig. 2 in their paper. High  $PM_{2.5}$  and submicron aerosol concentrations have been observed in  
Beijing (Zhang et al., 2013; Sun et al., 2015). Ozone and aerosol concentrations may therefore have been high in March  
2015 over Northeast Asia.

Based on these lidar data and the in-situ measurement data at Takagimachi in Saga city, an air mass with high ozone and  
aerosol concentrations could have been transported from the free troposphere to the surface by vertical mixing when the  
planetary boundary layer developed in the daytime. A similar high-surface-ozone event was observed by eight ozonesonde  
240 measurements during 6–9 June 2003 over the Seoul metropolitan region (Oh et al., 2010). The combination of ozone DIAL  
measurements with surface in-situ ozone measurements is very useful for studying the process of descent of high ozone  
concentrations in the free troposphere to the surface and the impacts on surface air quality. Such measurements of pollution  
plumes that descend from the free troposphere to the surface are highly recommended (HTAP, 2010).

The MRI-CCM2 could approximately reproduce the high-ozone volume-mixing ratios after some modifications of  
245 physical and chemical parameters. MASINGAR mk-2 successfully predicted high aerosol concentration events, but the  
predicted peak AOD was about one-third of the observed AOD. For further improvement of these models, it will be  
important to continue comparing these models with ozone DIAL, Mie lidar, and surface in-situ ozone and particle  
measurements.

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surface oxidant and  $PM_{2.5}$  measured by the Saga Prefectural Environmental Research Center. The NOAA Hybrid Single  
Particle Lagrangian Integrated Trajectory (HYSPRIT) model was used to calculate backward trajectories of air parcels.

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360



**Table 1.** Characteristics of Mie lidar

Transmitter				
370	Laser		Nd:YAG	
	Wavelength	532 nm		1064 nm
	Pulse energy	130 mJ		130 mJ
	Pulse repetition rate		10 Hz	
	Pulse width		8 ns	
375	Beam divergence	0.2 mrad		0.2 mrad
Receiver				
	Telescope type	Schmidt Cassegrain		
	Telescope diameter	30.5 cm		
380	Focal length	3048 mm		
	Field of view	1 mrad		
		Polarization	P and S	None
	Number of channels	3	1	
Interference filter				
385	Center wavelength	532.0 nm		1064.1 nm
	Bandwidth (FWHM)	0.29 nm		0.38 nm
	Transmission	0.66		0.58
	Detectors	PMT		APD
		(Hamamatsu R3234-01)		(EG&G C30956EH)
390	Signal processing	12bit A/D + Photon counting		
	Time resolution	1 min		
	Vertical resolution	7.5 m		

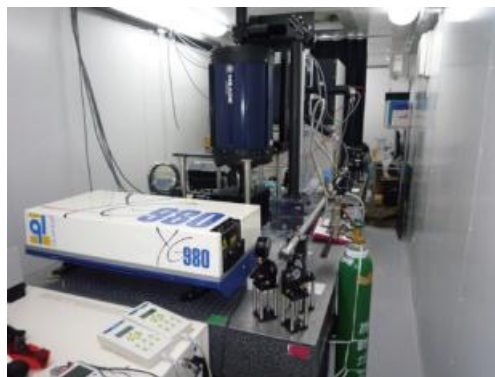
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400 **Table 2.** Characteristics of tropospheric ozone DIAL system

Transmitter					
	Pump laser	Nd:YAG			
	Wavelength	266 nm			
405	Pulse energy	107 mJ			
	Pulse repetition rate	10 Hz			
	Pulse width	8 ns			
	Raman active gas	CO <sub>2</sub>			
	Stokes lines	276 nm	287 nm	299 nm	312 nm
410	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)	
415	Telescope diameter	49 cm		10 cm	
	Focal length	1750 mm		320 mm	
	Field of view	1 mrad		3 mrad	
Interference filter					
	Center wavelength	287.2 nm	299.0 nm	312.0 nm	276.1 nm 287.2 nm
420	Bandwidth (FWHM)	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
Detectors					
	Signal processing	PMT (Hamamatsu R3235-01)			
	Time resolution	12bit A/D + Photon counting			
	Time resolution	1 min			
425	Vertical resolution	7.5 m			

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435 **Figure 1.** Mie lidar and ozone DIAL (right) were installed in the container at the left on the ground (left).

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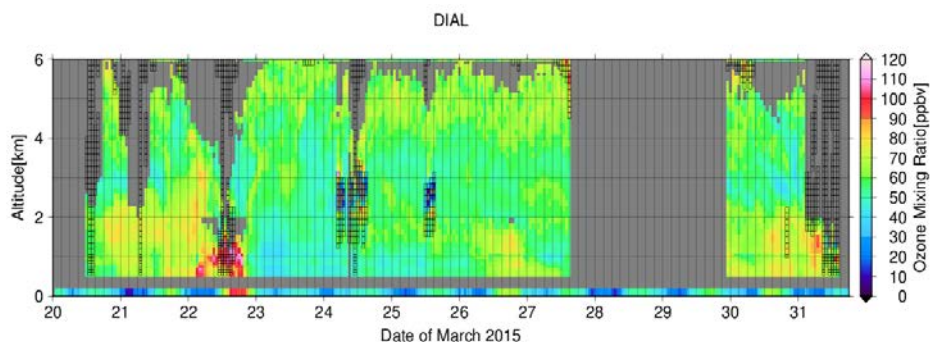
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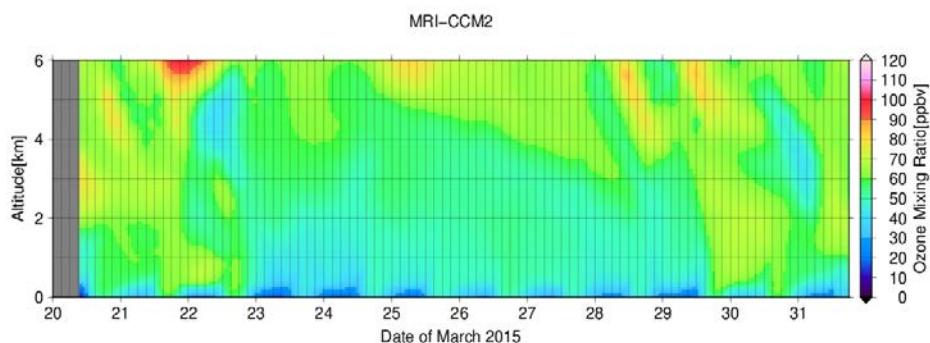
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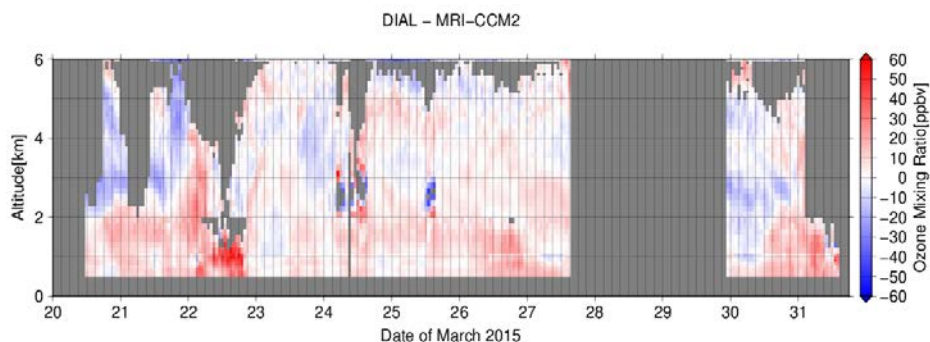
(a)



(b)



(c)



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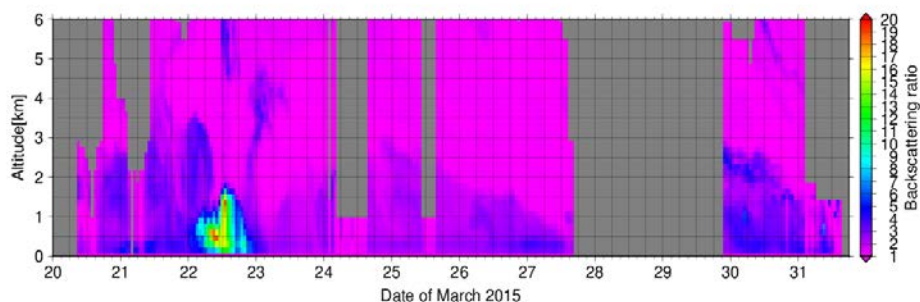
**Figure 2.** Time-altitude cross-sections of (a) ozone volume mixing ratios observed by DIAL over Saga from 11:10 JST on 20 March to 14:33 JST on 31 March 2015, (b) the ratios simulated by a modified MRI-CCM2 for 20–31 March 2015, and (c) the difference between the observed and simulated ozone volume mixing ratios (a–b). Gray regions indicate areas where there were no observational data or the statistical errors were larger than 10%. Regions enclosed with black rectangles are areas where the data were affected by aerosols and/or clouds. The lowest row in Fig. 2a shows photochemical oxidant (ozone) volume mixing ratios at Takagimachi in Saga city as measured by the Saga Prefectural Environmental Research



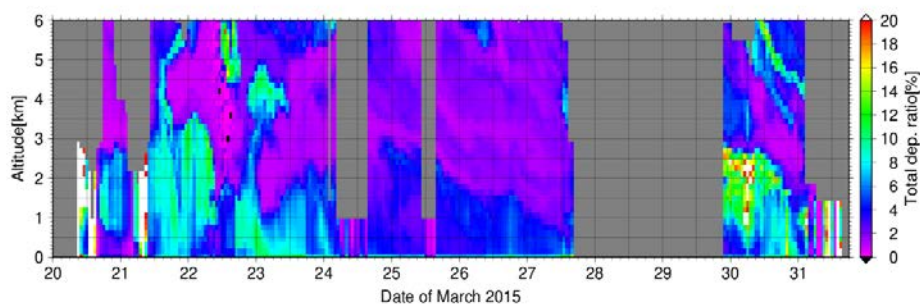


Center.

(a)



(b)



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**Figure 3.** Time-altitude cross-sections of (a) backscattering ratios and (b) total volume depolarization ratios at 532 nm observed by Mie lidar at Saga from 11:10 JST on 20 March to 14:33 JST on 31 March 2015. Lidar observations were not available from 15:56 JST on 27 March to 21:58 JST on 29 March 2015 mainly because of bad weather. Gray regions are areas where there were no observational data or where the observations were affected by clouds.

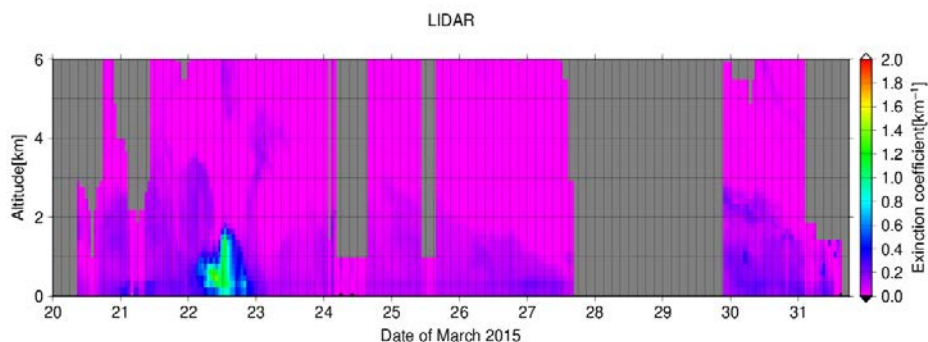
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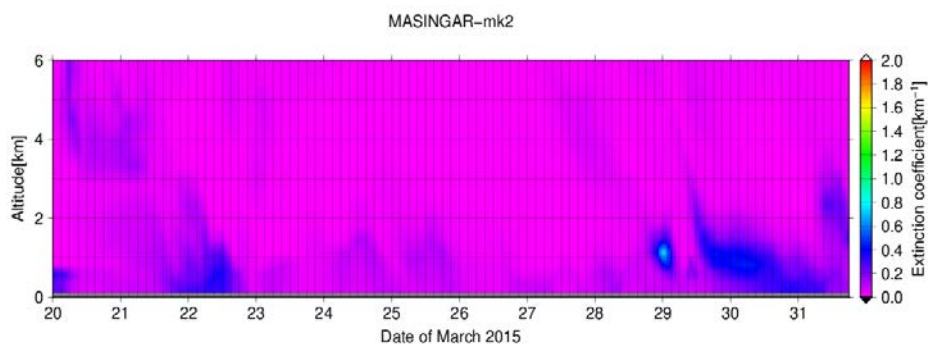


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(a)

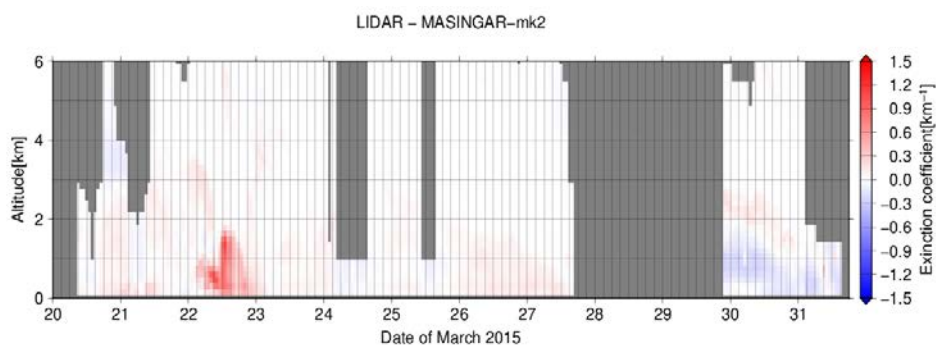


(b)



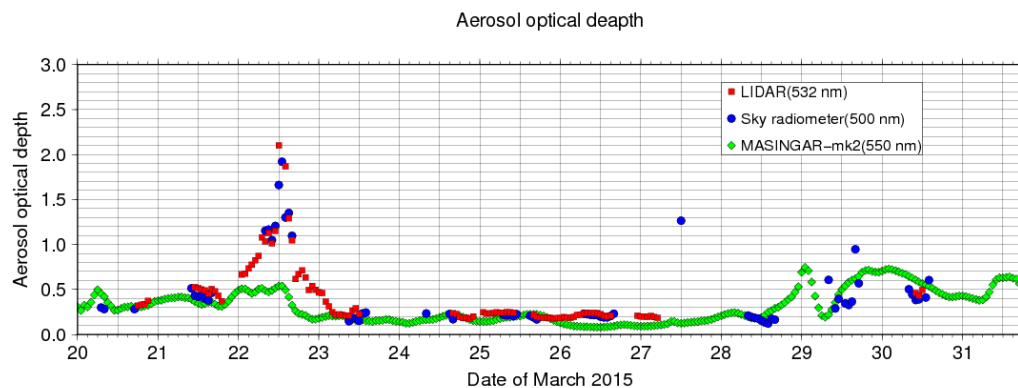
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(c)



**Figure 4.** Time-altitude cross-sections of (a) aerosol extinction coefficients observed by Mie lidar at 532 nm over Saga from 11:10 JST on 20 March to 14:33 JST on 31 March 2015, (b) the coefficients simulated by MASINGAR-mk2 at 550 nm for 20–31 March 2015, and (c) the difference between the Mie lidar observations and the simulation (a–b). Gray regions represent areas where there were no observational data.

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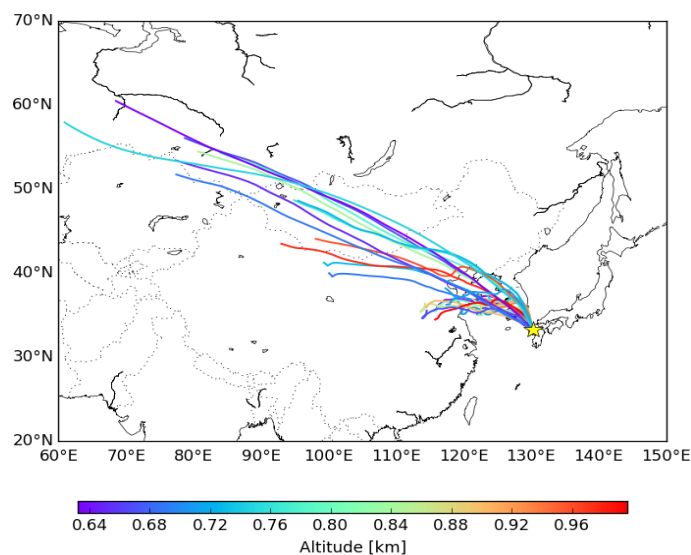
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**Figure 5.** Temporal variation of the aerosol optical depth (AOD) measured by Mie lidar at 532 nm (red circles), by sky radiometer at 500 nm (blues circles), and simulated at 550 nm by MASINGAR-mk2 (green circles).

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**Figure 6.** Horizontal projections of three-dimensional backward trajectories of 27 air parcels initially at altitudes of 600–1000 m over the lidar site at Saga. The colors show the initial positions of the air parcels. The trajectories were calculated for three days from 00:00 UTC (09:00 JST) on 22 March 2015 by the NOAA HYSPLIT trajectory ensemble option using the  $1^\circ \times 1^\circ$  Global Data Assimilation System (<http://ready.arl.noaa.gov/hypub-bin/trajtype.pl?runtype=archive>).

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