

## Long-term trends of stratospheric ozone concentration obtained by NIES ozone DIAL over Tsukuba, Japan

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**ABSTRACT:** Differential absorption lidar (DIAL) at the National Institute for Environmental Studies (NIES) in Tsukuba (36°N, 140°E), Japan has been making routine observation for almost 20 years. Since 1988, more than 600 vertical profiles of stratospheric ozone and temperature have been obtained. We compared the lidar data with satellite data from the Stratospheric Aerosol and Gas Experiment (SAGE II) and assimilation data from the National Center for Environmental Prediction (NCEP). The lidar and SAGE II ozone profiles agreed within 5% in altitude range from 18 km to 40 km and within 10% up to 45 km. The lidar and NCEP temperatures agreed within 7 K in the 35- to 50-km range. Ozone levels were highest in spring at altitudes below 20 km. Above 30 km, the ozone maximum occurred during summer. The annual cycle of temperature is observed with a spring maximum for all altitudes in the 35- to 50-km range. Ozone variations caused by the Quasi-Biennial Oscillation (QBO) and the 11-year solar cycle are discussed, along with ozone trends observed after subtraction of the natural variations.

### INTRODUCTION

Over the last several decades, increasing attention has been paid to stratospheric species and parameters related to ozone depletion and climate change. To understand the characteristics and their changes in the stratosphere, long-term measurements of ozone concentrations have been conducted by means of ground-based sensors. The Network for the Detection of Atmospheric Composition Change (NDACC) was established in 1991 and has been playing a key role in long-term international monitoring efforts. Lidars have been demonstrated to be reliable ozone-profiling instruments with relatively high altitude resolution [1]. In this paper, we present an overview of ozone data obtained with the lidar system at the National Institute for Environmental Studies (NIES) in Tsukuba, Japan, in the context of long-term observation of ozone concentration and temperature for period from 1988 to 2007.

### METHODS AND APPARATUS

The NIES ozone lidar system (located at 36°N, 140°E) is a typical UV differential absorption lidar (DIAL) system for low stratospheric ozone measurements. The lidar system has been in operation since 1988, and several replacements and improvements have been made since it was first installed [2,3]. The system parameters and improvements were presented in detail by Park *et al.* [4]. Currently, the NIES ozone lidar system uses wavelength channels of 308/355-nm (for the Mie/Rayleigh scattering DIAL mode) and 332/386-nm (for the Raman scattering DIAL mode). In this study, we applied an improved data processing and retrieval algorithm (version 2) for ozone [4]. The vertical profiles of ozone concentration were calculated by means of the DIAL equation [5]. The vertical profiles of temperature were obtained using the algorithm of Chanin and Hauchecorne [6]. The SAGE II is a satellite instrument [7], that measured ozone absorption at various altitudes by comparing the visible part of solar spectra obtained with different absorption cross-sections through the atmosphere. The SAGE II data were obtained using retrieval algorithm version 6.20. The vertical profiles were restricted to data obtained within  $\pm 3^\circ$  latitude and  $\pm 20^\circ$  longitude from the NIES lidar location. In this study, we used only values of ozone concentration with statistical errors smaller than 20% for both instruments.

## RESULTS AND DISCUSSIONS

Figure 1 plots point-to-point long-term temporal variations of ozone concentration measured at altitudes of 15, 20, 25, 30, 35, and 40 km. The solid circles present results of measurements by NIES ozone DIAL during period from 1988 to 2007. The open squares show results from SAGE II up to July 2005. The time series plotted on the Fig.1 are based on 549 vertical profiles of the NIES ozone DIAL and 374 profiles of SAGE II.

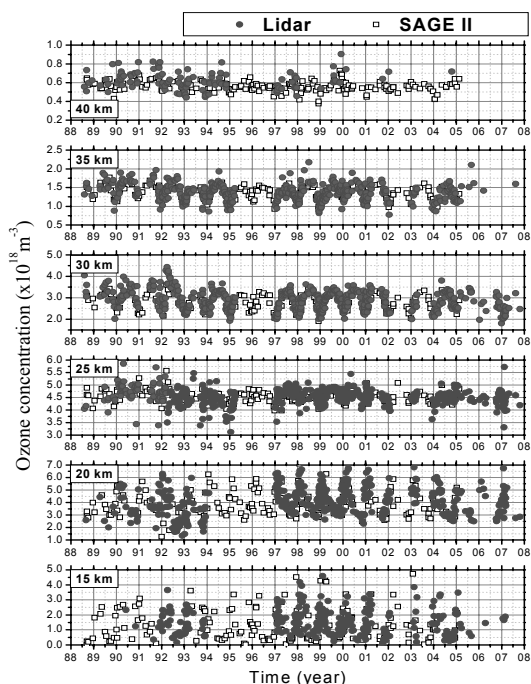


Figure 1. Time series of ozone concentrations obtained with the NIES ozone lidar (solid circles) and by the SAGE II (open squares)

all the altitude in Fig. 1. Therefore, to accurately analyze other variations and the long-term trends, subtraction of annual cycle is required.

The climatological mean vertical profiles obtained by lidar and SAGE II, and the relative differences between them, are shown in Figure 2. These profiles were obtained by averaging of all available monthly mean profiles from 1988 to 2005, separately for the two instruments.

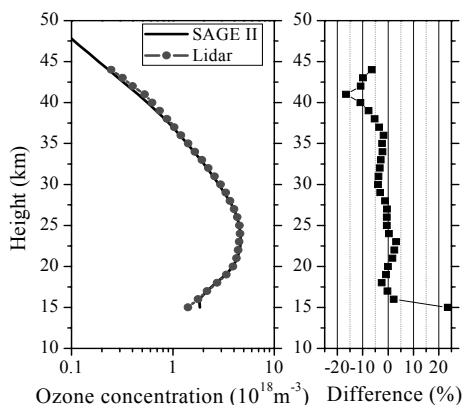


Figure 2. Climatological mean vertical ozone concentration profiles obtained with the NIES ozone lidar and the SAGE II, along with relative difference between them.

the altitude range where the difference between the lidar and SAGE II profiles was smaller than 5%, namely from 16 to 35 km.

In general, the variations in ozone observed with the NIES ozone lidar agree well with those measured with the SAGE II, at all altitudes and over the entire period. The two instruments did not obtain measurements at exactly the same place and or time, and therefore there were small differences between individual data points for the two instruments. Systematic differences due to diurnal variations of ozone concentration have to be considered because the SAGE II measurements were taken at sunrise and sunset, whereas the lidar measurements were taken only at night. The fact that most of the profiles were not obtained during the same day, might result in overestimation of the disagreement between the DIAL and SAGE II data. To minimize these effects, we used monthly mean values of ozone concentration for the following analysis.

The time series of ozone exhibited not only long-term trends but also shorter time scale variations, in which annual cycle, Quasi-Biennial Oscillation (QBO) and the 11-year solar cycle are significant. Among these, only annual variations of ozone can be clearly distinguished for all the altitude in Fig. 1. Therefore, to accurately analyze other variations and the long-term trends, subtraction of annual cycle is required.

The climatological mean vertical profiles obtained by lidar and SAGE II, and the relative differences between them, are shown in Figure 2. These profiles were obtained by averaging of all available monthly mean profiles from 1988 to 2005, separately for the two instruments. The climatology mean ozone profiles measured by lidar and SAGE II agreed within  $\pm 5\%$  in the altitude range from 16 to 37 km and within 20% up to 44 km. At 15 km, the climatological mean obtained by lidar was markedly smaller than the SAGE II mean (relative difference 23%; Figure 2). This large difference might have been caused by the presence of small-scale structures in the lower stratosphere and by meteorological inhomogeneties. In the altitude range from 37 to 45 km, the lidar climatological means were larger than the means obtained from the satellite data. Detailed analysis of the time series at this altitude range shows that around 75% of successful lidar data points were taken in the period from 1990 to 1993. Ozone concentrations were higher during this period because the 11-year solar cycle was at its maximum. In consequence, a systematic shift may have contribution to the lidar's climatological mean.

On the basis of the vertical profiles of the climatological means, we limited the experimental data to the

The average annual cycles of ozone concentration recorded by the two instruments are shown in Figure 3 for altitudes of 20, 25, 30, and 35 km. We obtained these average annual cycles by averaging all the monthly mean profiles separately for each month of the year. The annual cycles for the lidar data agreed well with the SAGE II data for the presented altitudes, as relative difference was less than 4%. All the features of annual cycles are typical for the Northern Hemisphere [8].

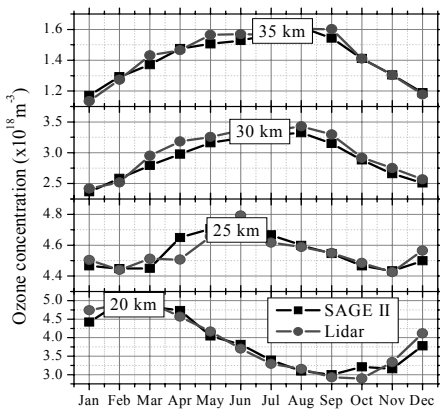


Figure 3. Annual cycles of ozone concentrations reported with the NIES ozone lidar and by SAGE II over the 1988 to 2005.

summer.

Figure 4 plots evolution of relative anomalies since 1988 at altitudes of 20, 25, 30, and 35 km. Anomalies are defined as the deviation of individual monthly means from climatological annual cycles. The individual annual cycles were subtracted for each instrument at each altitude, and data were smoothed by a 6-month running mean. The curve at the top shows the solar flux at 10.7 cm, a proxy for the solar activity.

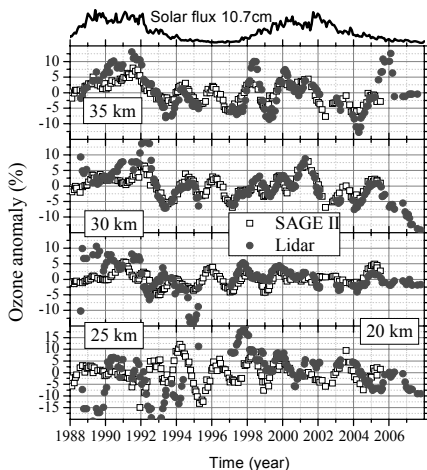


Figure 4. Temporal variation of ozone anomalies obtained with the NIES ozone lidar and the SAGE II. The monthly mean values are presented. Data are smoothed by a 6-month running mean. The curve at the top shows the solar flux at 10.7 cm.

In the lower stratosphere (20 km), the annual cycle of the ozone shows the highest ozone levels occurred in late winter and early spring – February to March. At 25 km the maximum of the ozone concentration is found to be in late spring – May to June. It is caused by the global Brewer-Dobson circulation that transports ozone to low altitudes in late winter in high altitude. In the middle stratosphere (30 km and 35 km heights), a maximum of the ozone concentration was observed in summer. The general form of the annual cycle in the middle stratosphere mainly reflects the photochemical production of ozone by solar radiation, which is low in winter and high in

summer.

In general, the ozone anomalies obtained by lidar are in good agreement with the anomalies obtained from the SAGE II at all altitudes. Both instruments showed variations due to the QBO reaching 5-10% at all the altitudes plotted in Figure 4. QBO oscillations were detected over all the years of observation. We observed ozone anomalies with a period of around 29 months, which were related to the QBO. For example, ozone anomaly maxima were detected in the winters of 1994, 1996, and 1998. The QBO-related peaks occurred during or shortly after the occurrence of the equatorial zonal winds at 40hPa. Modification of the ozone concentrations were observed at altitude 30 and 35 km in phase with the 11-year solar cycle. During the period of increase of solar flux from 1989 to 1993, the ozone levels were higher around 5 % and 7% at 30 km and 35 km, respectively. In the next period of solar maxima from 1999 to 2002, we observed relative increasing of approximately about 3 % at 30 and 35 km.

Figure 5 shows the ozone trend after subtraction of the QBO and solar cycle effects for altitude range from 30 to 40 km (upper stratosphere). Subtraction is done by multiple linear least-squares fitting of appropriate harmonic time series to the observed ozone variations [9,10]. Here we use harmonics with about 29 months to describe QBO, and harmonics with 127 months to describe the 11-years solar cycle. The long-term ozone trend obtained from the lidar data is in good agreement with that obtained by SAGE II. The relative differences between two trends were smaller than 20% for the entire observation period of observations 1988-2005. However, a residual oscillation with QBO like period still can be seen in figure 5. The plots in Figure 5 clearly show a difference in the ozone trend behaviours for period before and after 1998. The ozone concentration in the upper stratosphere decreased markedly during the period from 1988 to the end of 1997. The ozone trend during that period for the lidar data is  $-6.0 \pm 0.5\%/decade$ . The error represent  $2\sigma$  error level. The trend for the SAGE II data is  $-5.2 \pm 0.4\%/decade$ . The values of the ozone trend are nearly the same within the range standard deviations. After 1998, the decreases in ozone concentrations obtained with the two instruments were negligible:  $-0.8 \pm 1.1\%/decade$  for the lidar data, and

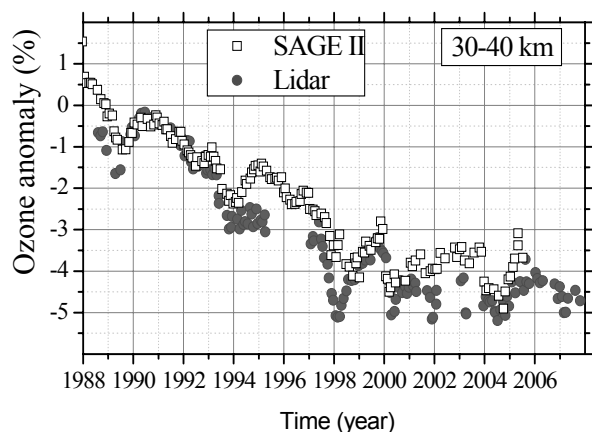


Figure 5. Temporal variation of ozone anomalies after subtraction of the effects of the annual cycle, the QBO, and the solar cycle at 30 -40 km. The monthly means values are presented. Data are smoothed by a 6-month running mean.

concentration was observed in summer. Ozone variations caused by the Quasi-Biennial Oscillation (QBO), the 11-year solar cycle, were also observed. We observed decreasing trend of ozone concentration of  $-6\%/decade$  by ozone DIAL and  $-5.2\%/decade$  by SAGE II over Tsukuba, Japan in the upper stratosphere over the period from 1988 to the end of 1997, and a negligible decrease in ozone concentration after 1998. The results from the both instruments show that ozone trend has changed from marked decrease to negligible around 1997. Over the lidar site, we observed the changes about 10 years after the Montreal Protocol to control of the harmful anthropogenic Chloro-Fluoro-Carbons emissions (1987). The change of the trend of the ozone after 1998 is one demonstration of the success of the Montreal Protocol in reducing the anthropogenic substances that damage the stratospheric ozone layer.

#### ACKNOWLEDGMENTS

This study was funded by the Monitoring Program of the Center for Global Environmental Research (CGER) at NIES. The SAGE II data were obtained from NASA Langley Research Center Atmospheric Science Data Center.

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$-0.9\pm 1.0\%/decade$  for the SAGE II data. However, we could not draw definitive conclusions about the decreasing ozone concentration after 1998, because of the high standard deviations of the data points; for both datasets, the standard deviations exceeded the values of the trends.

#### CONCLUSIONS

Long-term variations of stratospheric ozone concentration over Tsukuba, Japan were observed with the NIES ozone DIAL system. Comparison of the climatological mean ozone profiles measured by lidar and the SAGE II showed agreement within 4% in altitude range from 16 km to 37 km and within 15% up to 45 km. The annual cycles of the ozone concentration show highest ozone concentration in spring at altitude 20 km. In the middle stratosphere (30 and 35 km), a maximum of the ozone