

1 **Sunset–Sunrise Difference in Solar Occultation Ozone**  
2 **Measurements (SAGE II, HALOE, and ACE–FTS) and its**  
3 **Relationship to Tidal Vertical Winds**

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5 **T. Sakazaki<sup>1</sup>, M. Shiotani<sup>1</sup>, M. Suzuki<sup>2</sup>, D. Kinnison<sup>3</sup>, J. M. Zawodny<sup>4</sup>, M. McHugh<sup>5</sup>,**  
6 **and K. A. Walker<sup>6</sup>,**

7  
8 [1]{Research Institute for Sustainable Humanosphere, Kyoto University, Uji, Japan}

9 [2]{Institute of Space and Astronautical Science, Japan Aerospace Exploration Agency,  
10 Sagamihara, Japan}

11 [3]{National Center for Atmospheric Research, Boulder, USA}

12 [4]{NASA Langley Research Center, Hampton, USA}

13 [5]{Global Atmospheric Technologies and Sciences, Newport News, USA}

14 [6]{Department of Physics, University of Toronto, Toronto, Canada}

15  
16 Correspondence to: T. Sakazaki (takatoshi\_sakazaki@rish.kyoto-u.ac.jp)

17  
18 **Abstract**

19 This paper contains a comprehensive investigation of the sunset–sunrise difference (SSD; i.e.,  
20 the sunset-minus-sunrise value) of the ozone mixing ratio in the latitude range of 10°S–10°N.  
21 SSD values were determined from solar occultation measurements based on data obtained  
22 from the Stratospheric Aerosol and Gas Experiment (SAGE) II, the Halogen Occultation  
23 Experiment (HALOE), and the Atmospheric Chemistry Experiment Fourier Transform  
24 Spectrometer (ACE-FTS). The SSD was negative at altitudes of 20–30 km (–0.1 ppmv at 25  
25 km) and positive at 30–50 km (+0.2 ppmv at 40–45 km) for HALOE and ACE–FTS data.  
26 SAGE II data also showed a qualitatively similar result, although the SSD in the upper  
27 stratosphere was two times larger than those derived from the other datasets. On the basis of

1 an analysis of data from the Superconducting Submillimeter Limb Emission Sounder  
2 (SMILES), and a nudged chemical-transport model (the Specified Dynamics version of the  
3 Whole Atmosphere Community Climate Model: SD–WACCM), we conclude that the SSD  
4 can be explained by diurnal variations in the ozone concentration, particularly those caused by  
5 vertical transport by the atmospheric tidal winds. All datasets showed significant seasonal  
6 variations in the SSD; the SSD in the upper stratosphere is greatest from December through  
7 February, while that in the lower stratosphere reaches a maximum twice: during the periods  
8 March–April and September–October. Based on an analysis of SD–WACCM results, we  
9 found that these seasonal variations follow those associated with the tidal vertical winds.

10

## 11 **1 Introduction**

12 Stratospheric ozone ( $O_3$ ) plays a critical role in the climate system through radiative processes,  
13 while simultaneously protecting the Earth’s surface from harmful ultraviolet radiation. Since  
14 the discovery of the ozone hole, long-term changes in ozone concentration have been studied  
15 extensively using various ground-based and satellite measurements (WMO, 2011). Useful  
16 datasets for long-term monitoring of vertical profiles of ozone levels can be obtained from  
17 solar occultation instruments, such as the Stratospheric Aerosol and Gas Experiment (SAGE)  
18 II (McCormick, 1987; McCormick et al., 1989), the Halogen Occultation Experiment  
19 (HALOE) (Russell et al., 1993), and the Atmospheric Chemistry Experiment Fourier  
20 Transform Spectrometer (ACE-FTS) (Bernath et al., 2005), as they have measurements that  
21 are self-calibrating and have high sensitivity. These measurements have been used to detect  
22 seasonal and interannual variability, as well as long-term trends, in the stratospheric ozone  
23 concentration (e.g., Shiotani and Hasebe, 1994; Randell and Wu, 1996; Newchurch et al.,  
24 2003; WMO 2011; Kyrölä et al., 2013).

25 These solar occultation instruments measure the atmosphere at sunrise and sunset (hereafter  
26 SR, SS). It has been reported that there is a difference in the observed values between the SR  
27 and SS profiles (hereafter, this is referred to as the sunset–minus-sunrise difference, SSD).  
28 For SAGE II, the SSD is reported to be up to 10% between altitudes of 35 and 55 km, with a  
29 maximum occurring in the tropics (cf. SAGE II version 5.9 data: Wang et al., 1996; version  
30 6.2 data: McLinden et al., 2009; version 7.0 data: Kyrölä et al., 2013). For HALOE (version  
31 17 data), Brühl et al. (1996) reported an SSD of approximately 5% in the tropical stratopause.

1 SSD values based on ACE–FTS (Fourier Transform Spectrometer) observations have yet to  
2 be reported, as far as we know.

3 Quantification of the SSD and a understanding the source of this difference are necessary for  
4 the construction of combined datasets from SR and SS profiles. As we will explore in this  
5 study, the SSD could be caused by any one of three factors, including: (1) systematic  
6 instrumental/retrieval bias between SR and SS observations; (2) natural diurnal variations;  
7 and (3) sampling issues. The factor (3) relates to the fact that if the times of SR and SS  
8 measurements are considerably different, the background ozone concentration changes  
9 (background changes occur on timescales longer than a day; e.g., subseasonal variations)  
10 during such periods are misinterpreted as an example of SSD (see Section 3 for details). In  
11 this sense, such spurious contributions, or contamination, should be assessed and removed  
12 before discussing the SSD.

13 In previous studies, the ozone SSD was mainly attributed to factor (1); in other words, factor  
14 (2) was thought to be negligible (Wang et al., 1996; Brühl et al., 1996). This was because  
15 diurnal variations in stratospheric ozone level were considered to be small and/or to be  
16 symmetric between SS and SR, if only photochemistry processes are taken into account.  
17 However, note that there are few observations of diurnal variations in the stratosphere and,  
18 thus, the above assumption had not been confirmed. In addition, factor (3) may not have been  
19 properly considered in some studies (e.g., in the context of interannual variations of the SSD,  
20 as shown by Kyrölä et al. 2013; see Section 3).

21 Recently, Sakazaki et al. (2013a; hereafter S13) revealed diurnal variations in ozone  
22 concentration from the lower to upper stratosphere using data from both the Superconducting  
23 Submillimeter Limb Emission Sounder (SMILES) and chemical-transport models (CTMs).  
24 They showed that the peak-to-peak difference in ozone levels reaches 8% during a day. An  
25 important point here is that, at some altitudes, the diurnal variations exhibit asymmetric ozone  
26 levels when comparing measurements at SS and SR, with the SSD reaching around 4% in the  
27 upper stratosphere. S13 showed that the diurnal variations ( $[\text{O}_3]'$ ) are largely controlled by the  
28 following equation,

$$29 \quad \frac{\partial[\text{O}_3]'}{\partial t} = -w' \frac{\partial \overline{[\text{O}_3]}}{\partial z} + S' \quad (1),$$

30 where  $t$  represents time,  $w'$  the diurnal variations in the vertical winds (tidal vertical winds),  $z$   
31 altitude,  $[\overline{\text{O}_3}]$  the daily, zonal mean ozone, and  $S'$  the diurnal variations in chemical

1 production or loss. The first term on the right-hand-side denotes the dynamical variation due  
2 to vertical transport by atmospheric tidal winds, while the second term denotes the  
3 photochemical variation. It should be noted that the dynamical variation results in a maximum  
4 at 1800–2000 local time (LT) in the upper stratosphere (and at 0000–0600 LT in the lower  
5 stratosphere; cf. figure 7d of S13), possibly causing a positive (negative) SSD. In contrast,  
6 although the photochemical variations are also important above an altitude of 30 km during  
7 the daytime, they are almost zero at SR/SS (see figure 7b of S13), and so do not contribute to  
8 the SSD. This has already been demonstrated and discussed in previous studies (e.g., Pallister  
9 and Tuck, 1984; Wang et al., 1996; Brühl et al., 1996). For the tropical mesosphere and lower  
10 thermosphere, Marsh and Russell (2000) used HALOE data to attribute the SSD of nitric  
11 oxide (NO) to diurnal variations caused by tidal vertical transport.

12 With regard to tides, the diurnal migrating tide (i.e., the diurnal harmonic component, which  
13 does not depend on longitude, but only on the LT of a particular latitude band) is basically  
14 dominant over other higher-order harmonics in the tropical stratosphere. Vertical winds  
15 associated with the diurnal migrating tide become maximal in the tropics, so that diurnal  
16 variations in ozone levels caused by vertical transport also reach a maximum in the tropics  
17 (see figure 8 of S13). In the tropical stratosphere, the diurnal migrating tide is known to  
18 exhibit a significant seasonal dependence, with its amplitude reaching a peak in January–  
19 February and July–September, as seen in satellite-based temperature measurements (e.g., Wu  
20 et al., 1998; Zeng et al., 2008; Huang et al., 2010; Sakazaki et al., 2012). These findings  
21 suggest that the tidal vertical winds and, consequently, the SSD, also show similar seasonal  
22 variations. Note that the tidal vertical wind is relatively difficult to observe directly because of  
23 its rather small amplitude. We suggest that in the presence of vertical gradient of ozone, the  
24 SSD pertaining to the ozone levels could be used to obtain quasi-observational evidence of  
25 tidal vertical winds in the tropical stratosphere.

26 As outlined above, SSDs have been noticed but they were considered independently for each  
27 occultation instrument. In addition, they have not been investigated fully in terms of diurnal  
28 ozone variations. The purpose of this study is, first, to quantify the SSD, including its  
29 seasonal dependence, by analyzing and comparing data from three independent solar  
30 occultation instruments: SAGE II, HALOE, and ACE–FTS. The differences among the  
31 datasets due to differences in measurement times are discussed with the aid of CTM data  
32 (from the Specified Dynamics version of the Whole Atmosphere Community Climate Model:

1 SD–WACCM), which provides a homogeneous dataset covering the full operational periods  
2 of each of the three satellite missions. Next, we interpret the SSD in terms of diurnal  
3 variations based on SMILES measurements and three-hourly, 3D-gridded SD–WACCM data,  
4 which provide data not only at SR/SS, but for the full diurnal cycle. The 3D-gridded SD–  
5 WACCM data are also used to discuss diurnal variations related to vertical transport and  
6 photochemistry. This study focuses on the tropical stratosphere at latitudes of 10°S–10°N  
7 between 20 and 60 km in altitude, as the SSD is greatest in the tropics. The remainder of this  
8 article is organized as follows. Sections 2 and 3 describe the datasets and analysis methods,  
9 respectively. Section 4 describes the SSD and its seasonal variations, and we also discuss the  
10 contribution from diurnal ozone variations. Section 5 summarizes our main results.

11

## 12 **2 Data Description**

13 In this study, the average SSD in the tropical stratosphere within the latitude range 10°S–  
14 10°N is analyzed and discussed using data from the three solar occultation instruments:  
15 SAGE II, HALOE, and ACE–FTS. To quantify the SSD caused by natural diurnal variations  
16 in ozone concentration, data from SMILES limb emission measurements are also analyzed. In  
17 addition, two datasets from SD–WACCM are used: the satellite coincidence data subset and  
18 the full three-hourly, 3D-gridded data. The former is used to discuss the difference in SSD  
19 among the different satellite datasets, while the latter is used to consider the SSD in terms of  
20 diurnal variations. All data are analyzed at geometric altitudes in the stratosphere between 20  
21 and 60 km.

22 Note that there is no systematic difference in the tracking procedure between SR and SS.  
23 SAGE II tracks the Sun by moving the field-of-view (FOV) across the solar disk. As the field-  
24 of-view went off the edge of the Sun, the scan-mirror reverses direction. HALOE tracks the  
25 top edge of the sun, with the science FOV held a fixed angle below this. Extensive SR/SS  
26 comparisons have been carried out over the 14-year measurement record and no significant  
27 bias in tracking is perceivable. The ACE-FTS is aligned by a tracking system that keeps it  
28 pointed at the radiometric center of the sun. While there might exist a very small difference  
29 between SR and SS tracking, this has been examined carefully and no significant SR-SS bias  
30 exists. Further details are provided in the following subsections.

## 1 2.1 SAGE II

2 The SAGE mission has deployed three instruments; i.e., SAGE I, II, and III. This study  
3 uses data from SAGE II because these measurements cover the longest period. SAGE II was  
4 launched in October 1984 on board the Earth Radiation Budget Satellite (ERBS) and  
5 remained operational until August 2005, providing data for over 21 years. The ERBS  
6 operated in a 610 km altitude, circular orbit with an inclination of 56°. SAGE II is a seven-  
7 channel spectrometer capable of obtaining ozone levels from measurements in the 600 nm  
8 Chappius band (McCormick et al., 1987, 1989).

9 Figure 1a shows the SAGE II measurement locations obtained in 1992. The latitude coverage  
10 ranges from 80°S to 80°N. SAGE II performed two occultation measurements per orbit,  
11 sampling two narrow latitude regions each day. As a result, for each day up to 15 SR and 15  
12 SS profiles were obtained near the same latitude regions, but spaced some 24° apart in  
13 longitude. Approximately 10 times per year, the tracks of the SR and SS measurements  
14 crossed over. The sampling pattern (i.e., the measurement day of the year at a particular  
15 latitude) changed gradually from year to year, and so cover the whole year when data are  
16 collected for several years.

17 We analyzed SAGE II version 7.0 data (Damaedo et al., 2013) obtained during the period  
18 1985–2005. Data were provided for geometric altitudes between 0 and 70 km at intervals of  
19 0.5 km. Geometric altitudes were determined by the viewing geometry with the aid of data  
20 from the Modern Era-Retrospective Analysis for Research Applications (MERRA) reanalysis  
21 (Rienecker et al., 2011). The uncertainty in altitude registration is less than 20 m (Damaedo et  
22 al., 2013). Ozone mixing ratio is calculated as the ratio of ozone number density to  
23 atmospheric density, both of which are provided in SAGE II data. The atmospheric density is  
24 not measured by SAGE II but derived from MERRA data; however, the results of SSD (%)  
25 (cf. Figure 5b) do not change even if the following analysis is done based on ozone number  
26 density. Prior to our detailed analysis, data were screened following the guidelines provided  
27 in the SAGE II version 7.0 readme file  
28 ([https://eosweb.larc.nasa.gov/project/sage2/sage2\\_release\\_v7\\_notes](https://eosweb.larc.nasa.gov/project/sage2/sage2_release_v7_notes)), as follows. Any profile  
29 with an error estimate larger than 10% at altitudes between 30 and 50 km was rejected. Data  
30 points with an uncertainty estimate of 300% or greater were rejected. Data points at the  
31 altitude of and below the occurrence of an aerosol extinction value greater than 0.006 km<sup>-1</sup>  
32 were rejected. Data points at the altitude of and below the occurrence of both the 525 nm

1 aerosol extinction value exceeding  $0.001 \text{ km}^{-1}$  and the 525/1020 extinction ratio of less than  
2 1.4 were rejected. Data points pertaining to altitudes below 35 km with an uncertainty  
3 estimate of 200% (or larger) were rejected. On the basis of this screening process, 3.9% and  
4 5.6% of all profiles were rejected for the times of SR and SS, respectively.

## 5 **2.2 HALOE**

6 HALOE was launched in September 1991 on board the Upper Atmosphere Research Satellite  
7 (UARS) and remained operational until November 2005, providing data over 15 years. UARS  
8 was placed in a 585 km circular orbit, with an inclination of  $57^\circ$  (Russell et al., 1993). Its  
9 ozone measurements were made at mid-infrared (mid-IR) wavelengths ( $9.6 \mu\text{m}$ ).

10 The HALOE measurement locations during 1992 are shown in Figure 1b. The latitude  
11 coverage was approximately  $80^\circ\text{S}$ – $80^\circ\text{N}$ . The instruments measured up to 15 SR and 15 SS  
12 profiles each day. Approximately 10 times per year, the tracks of the SR and SS  
13 measurements crossed over. As with SAGE II, the sampling pattern of HALOE changed  
14 gradually from year to year.

15 We analyzed HALOE version 19 data obtained over the period 1991–2005 (e.g., Randall et al.,  
16 2003). Data were provided at 271 pressure levels; i.e.,  $1000 \times 10^{-(i-1)/30}$  hPa ( $i = 1, 2, \dots, 271$ ),  
17 between 1000 and  $10^{-6}$  hPa, with a corresponding vertical spacing of approximately 0.5 km.  
18 Geometric altitudes were also provided in relation to the pressure levels for each profile, so  
19 that we mapped ozone mixing-ratio data onto geometric altitude levels between 0 and 70 km,  
20 with a spacing of 0.5 km. No screening was performed on the HALOE data.

21 HALOE profiles are registered in altitude in an iterative fashion, and the resulting registration  
22 is generally assessed to be better than 100 m (McHugh et al., 2003, 2005). Initially, the data  
23 are placed on an altitude grid based on the HALOE measurements versus zenith angle. A first  
24 pressure registration is performed using the measured transmittance profile from the  $2.8 \mu\text{m}$   
25 channel (dominated by  $\text{CO}_2$  absorption). In this process, the altitude grid is shifted to best  
26 match simulated transmittances in the 30 to 45 km altitude range, assuming the temperature  
27 profile from the National Center for Environmental Prediction (NCEP) reanalysis (Beaver et  
28 al., 1994; Remsberg et al., 2002; see Kalnay et al., 1996 for NCEP reanalysis). Temperatures  
29 and corresponding hydrostatic pressures are then retrieved in an iterative upward fashion from  
30 30 km at 1.5 km spacing. Finally the retrieved  $T(p)$  profile is merged into the NCEP profile  
31 from 34 km (pure NCEP) to 43 km (pure HALOE retrieval). Pressures are calculated

1 assuming hydrostatic balance. This entire process is repeated again after retrieval of water  
2 vapor, and finally after a retrieval of aerosols, to account for all significant interfering  
3 absorption in the 2.8  $\mu$  m CO<sub>2</sub> channel.

### 4 **2.3 ACE-FTS**

5 ACE-FTS was launched in August 2003 on board the SCISAT-1 satellite. SCISAT-1 is  
6 operating in a circular, low-Earth orbit, with a 74° inclination and an altitude of 650 km  
7 (Bernath et al., 2005). ACE-FTS measures ozone at IR wavelengths (9.6  $\mu$ m).

8 Figure 1c shows the ACE-FTS measurement locations during 2005. The latitude coverage is  
9 approximately 85°S–85°N. The instruments measure up to 15 SR and 15 SS profiles each day.  
10 Approximately six times per year, the tracks of the SR and SS measurements cross over.  
11 Unlike SAGE II and HALOE, however, the sampling pattern changes little from year to year.  
12 Thus, data for 10°S–10°N are obtained only in February, April, August, and October of each  
13 year (see Figure 1c).

14 We analyzed the most recent data, version 3.0 (Dupuy et al., 2009; Waymark et al., 2013),  
15 obtained between 2004 and 2011. Data were provided at geometric altitudes from 0 to 140 km,  
16 with a spacing of 1 km. Some of the data obtained during this period had measurement  
17 problems and were not used based on the recommendations from the ACE-FTS Data Issues  
18 List ([https://databace.scisat.ca/validation/data\\_issues.php](https://databace.scisat.ca/validation/data_issues.php)).

19 Altitudes in ACE-FTS measurements are defined as follows (see Boone et al., 2005 for  
20 details). At low altitudes (below ~42 km), pointing information from the satellite is relatively  
21 poor and the pointing is quite variable, with refraction effects plus clouds and aerosols  
22 affecting the instrument's pointing. Therefore, geometry is derived through analysis of the  
23 ACE-FTS spectra. CO<sub>2</sub> volume mixing ratio is fixed to its "known" profile, and then CO<sub>2</sub>  
24 lines are analyzed to determine pressure and temperature. Hydrostatic equilibrium is used to  
25 express the altitude separations between measurements in terms of pressure and temperature  
26 during the retrieval, so geometry is determined simultaneously. At high altitudes (above 60-65  
27 km), geometry is relatively well known from first principles (i.e., given the knowledge of the  
28 location of the satellite and the look direction to the sun at a given time, one can calculate the  
29 pointing information). In the middle (42-60 km), CO<sub>2</sub> VMR is still "known" and geometry is  
30 known. This region allows us to determine the pressure at the "crossover" point (near 42 km)  
31 to be used in the calculation of altitude at low altitudes (below ~43 km) via hydrostatic



1 equilibrium. A final altitude registration is performed using the Canadian Meteorological  
2 Centre model output at the lowest altitudes (~15-25 km). The uncertainty on altitude  
3 registration for the ACE-FTS is 150 m.

#### 4 **2.4 SMILES**

5 SMILES was attached to the Japanese Experiment Module (JEM) on board the International  
6 Space Station (ISS) through the cooperation of the Japan Aerospace Exploration Agency  
7 (JAXA) and the National Institute of Information and Communications Technology (NICT).  
8 The ISS is in a circular orbit at a typical altitude between 350 and 400 km, with an inclination  
9 of 51.6°. Its latitude coverage is 38°S–65°N. SMILES measured the Earth's limb and made  
10 global observations of the minor constituents in the middle atmosphere in the 600 GHz range  
11 between 12 October 2009 and 21 April 2010 (Kikuchi et al., 2010). Unlike the solar  
12 occultation measurements (SAGE II, HALOE, and ACE-FTS), limb emission measurements  
13 are made continuously following the orbit track, not only at SR and SS, but throughout the  
14 day and night. As a result, two datasets for ascending and descending nodes are obtained  
15 every day, in each latitude band. In addition, the ISS is not in a Sun-synchronous orbit, and its  
16 orbital plane rotates every 60 days; consequently, a full diurnal cycle in the tropics is covered  
17 in 30 days by the ascending/descending nodes.

18 For SMILES, we defined the SS and SR profiles as those characterized by solar zenith angles  
19 between 80° and 100°. Note, again, that SMILES measurements cover the full diurnal cycle,  
20 as well as during SS and SR. Thus, the SSD related to diurnal variations in ozone level can be  
21 calculated. With regard to the quality of ozone data from SMILES, Imai et al. (2013a)  
22 compared the ozone concentration profiles from the SMILES version 2.1 dataset with those  
23 from five satellite measurements and two CTMs (including SD-WACCM), and found that in  
24 the stratosphere the agreement was generally better than 10%. Imai et al. (2013b) compared  
25 the ozone concentrations profiles from the SMILES version 2.3 dataset with those from  
26 worldwide ozonesonde stations and found that the agreement was between 6% and 15% for  
27 20-26 km at low latitudes; part of this difference was attributed to issues related to the  
28 ozonesonde measurement system.

29 In this study, version 2.4 data were analyzed (JEM/SMILES L2 Products Guide for version  
30 2.4: [http://smiles.tksc.jaxa.jp/l2data/pdf/L2dataGuide\\_130703.pdf](http://smiles.tksc.jaxa.jp/l2data/pdf/L2dataGuide_130703.pdf)). Data were provided at  
31 geometric altitudes between 8 km and 118 km, with a vertical spacing of 2 km (below 58 km)

1 or 3 km (above 58 km). The number of retrieved profiles in version 2.4 was significantly  
2 larger than that in version 2.0, by updating the screening conditions. We did not find any  
3 significant differences in diurnal variation between versions 2.4 and 2.0 (which was used by  
4 S13). For stratospheric diurnal variations in the SMILES data, Parrish et al. (2013) found  
5 good agreement with the results from ground-based microwave measurements at Mauna Loa  
6 (19.5°N, 204.5°E), as well as those from the Microwave Limb Sounder onboard the UARS  
7 and Aura satellites.

8 SMILES does not measure geometric altitude, so that temperature (from SMILES below ~40  
9 km and from SD-WACCM (for ver.2.4) above ~40 km) and pressure (from SMILES) are  
10 used to calculate the "relative" altitudes from some pressure level, with the hydrostatic  
11 equilibrium assumption. For deriving the "absolute" altitude, pressure at 30 km from SD-  
12 WACCM output is used (for ver.2.4) (C. Mitsuda, personal communication). The uncertainty  
13 in altitude registration for SMILES is considered to be in the same order as that for other  
14 datasets. Note that there is no systematic difference in the altitude registration between SR  
15 and SS.

## 16 **2.5 SD-WACCM**

17 The NCAR Whole Atmosphere Community Climate Model, version 4 (WACCM4), is a  
18 comprehensive numerical model spanning the range of altitude from the Earth's surface to the  
19 thermosphere (Garcia et al., 2007; Kinnison et al., 2007; Marsh et al., 2013). WACCM4 is  
20 based on the framework of the NCAR Community Atmosphere Model, version 4 (CAM4),  
21 and includes all of the physical parameterizations of CAM4 (Neale et al., 2013) and a finite  
22 volume dynamical core (Lin, 2004) for the tracer advection. A version of WACCM4 has been  
23 developed that allows the model to be run with specified (external) meteorological fields  
24 (Kunz et al., 2011). Here specified dynamics (SD)-WACCM is a chemical transport model  
25 (CTM) that nudges WACCM4 temperature, horizontal winds, and surface pressure towards 6-  
26 hourly time series from MERRA reanalysis (Rienecker et al., 2011) below 50 km. The CTM  
27 data was constrained by meteorological conditions with an hourly timescale, so that they  
28 could be directly compared with our observational data. Note that the stratospheric ozone  
29 concentration and its diurnal variations in the SD-WACCM data have been analyzed and that  
30 the results agreed well with those based on the SMILES data (Imai et al., 2013a; S13).

1 The horizontal resolution used was 2.5° in longitude and 1.9° in latitude, and the model time  
 2 step was 30 min. We prepared and analyzed a satellite coincidence data subset, which was  
 3 sampled at the nearest time and grid point to each satellite measurement. The measurement  
 4 longitude, latitude, and time were used pertaining to an altitude of 30 km. In other words, we  
 5 did not consider any altitude dependence of the longitude, latitude, or time for a given profile.  
 6 This may cause estimation errors in the mesosphere, where the ozone concentration changes  
 7 rapidly at SR and SS (e.g., S13). Data were provided at 88 pressure levels and mapped onto  
 8 geometric altitude grids corresponding to those of the satellite datasets. The geometric altitude  
 9 ( $z$ ) is calculated from geopotential height ( $Z$ ) by the equation,

$$10 \quad z = \frac{rZ}{\frac{g}{g_0}r - Z}, \quad (2)$$

11 where  $g$  is the gravity at ( $z, \phi$ ),  $g_0 (= 9.80665 \text{ m s}^{-2})$  is the reference gravity,  $r$  is the radius of  
 12 the Earth (cf. Mahoney, 2008, available from the web site

13 [http://mtp.mjmahoney.net/www/notes/pt\\_accuracy/ptaccuracy.html](http://mtp.mjmahoney.net/www/notes/pt_accuracy/ptaccuracy.html))

14 As well as the satellite coincidence dataset (only at SR and SS), we analyzed three-hourly,  
 15 3D-gridded data obtained between 2008 and 2010 (hereafter referred to as the ‘full-grid’ data)  
 16 to extract the full diurnal cycle. We only used data for these three years because of constraints  
 17 on data storage. As well as ozone mixing ratio, vertical wind in geometric altitude ( $w$ ) and  
 18 chemical generation or loss ( $S$ ) in SD-WACCM data are also analyzed to assess the  
 19 dynamical and photochemical contributions to the SSD (cf. Equation (1)). On pressure levels,  
 20  $w$  is calculated on pressure levels from geometric altitude ( $z$ ) and vertical velocity ( $\omega$ ), by  
 21 using the fomula,

$$22 \quad w = \frac{Dz}{Dt} = \frac{\partial z}{\partial t} + \mathbf{u} \cdot \nabla_z + \rho g \omega, \quad (3)$$

23 where  $\mathbf{u}$  is horizontal wind,  $\rho$  is density, and  $g$  is gravity acceleration; then, the calculated  $w$  is  
 24 mapped onto geometric altitude grids.

25

### 26 **3 Analysis Methods**

27 When calculating the SSD, careful consideration should be given to the sampling issues.

28 Figure 2 shows the number of SR (red) and SS (blue) profiles in the latitude range of 10°S–

1 10°N obtained during the observation period. We see that the SR and SS profiles are not  
2 distributed evenly; e.g., there are no SR (SS) profiles from November to January (May to  
3 July) between 2001 and 2005 for SAGE II. This indicates that if the SSD is calculated by  
4 simply averaging all SR and SS profiles for a certain period (e.g., Kyrölä et al., 2013), it could  
5 be significantly contaminated by seasonal/interannual variations in ozone concentrations. In  
6 fact, Kyrölä et al. (2013) reported that the SSD profile for 2001–2005 was considerably  
7 different to those recorded in the other periods (1985-1990, 1991-1995, and 1996-2000); we  
8 attribute this to contamination caused by uneven sampling during this period. To resolve this  
9 issue, we calculated the SSD in SR–SS pairs which have both SR and SS profiles measured at  
10 almost the same time (see the open stars in Figure 1). A pair is defined such that it has more  
11 than one profiles for both SR and SS within 15 days (30 days) for SAGE II and HALOE  
12 (ACE-FTS). However, note that there is still a difference of 10–15 days in the SR and SS  
13 measurement times for each pair. We found that changes in ozone levels during these 10–15  
14 days also contaminate the SSD. Therefore, this effect was further assessed and removed (see  
15 below).

16 Approximately eight pairs for both SAGE II and HALOE, and four pairs for ACE–FTS were  
17 defined for each year (see Figure 1). The differences in measurement days between SR and  
18 SS profiles were less than about 10 days (15 days) for SAGEII and HALOE (ACE–FTS). The  
19 total number of pairs for the entire period was 128 for SAGEII, 86 for HALOE, and 25 for  
20 ACE–FTS. There were no SS–SR pairs for SAGE II data after 2001, although the  
21 measurements continued until 2005. Also, for ACE-FTS, SS-SR pairs are defined only in  
22 February, April, August and October. Between 10 and 50 profiles were obtained for SR and  
23 SS in each pair (see Figure 2).

24 Figure 3 shows the time series of the ozone mixing ratio at an altitude of 40 km (10°S–10°N)  
25 based on HALOE data for the period 1992–1993. The mean profile for each SR and SS for  
26 the  $i^{\text{th}}$  SR–SS pair ( $SR^i$  and  $SS^i$ ) was produced by averaging the SR (red open circles) and SS  
27 (blue open circles) data, respectively. Subsequently, for each pair, the original SSD ( $SSD_{\text{org}}^i$ )  
28 was calculated as the difference between the mean SS and SR profiles (i.e.,  $SSD_{\text{org}}^i = SS^i -$   
29  $SR^i$ ). Figure 4 shows individual  $SSD_{\text{org}}^i$  values averaged at altitudes of 40–45 km based on  
30 HALOE data, and sorted by time of year (black closed circles). We note again that, although  
31 the differences in measurement days between the SR and SS profiles are less than 10–15 days,  
32 seasonal variations still produced significant differences during this period and contaminate

1 the SSD. In fact, this effect is not negligible in the stratosphere, where semiannual variations  
 2 in ozone concentration associated with the stratospheric semiannual oscillation (SAO) exist  
 3 (e.g., Ray et al., 1994). Thus, we attempted to further remove the contamination caused by  
 4 seasonal variations, as follows.

5 For both SAGE II and HALOE, we assessed the background ozone concentration changes  
 6 ( $O_{3\_BG}$ ) at each altitude using linear multivariable regression:

$$7 \quad O_{3\_BG} = b + at + q_1 QBO_{10}(t) + q_2 QBO_{30}(t) + \sum_{n=1}^2 (c_n \cos(n\omega t) + s_n \sin(n\omega t)), \quad (4)$$

8 where  $t$  is expressed in days,  $QBO_{10}(t)$  and  $QBO_{30}(t)$  are the quasi-biennial oscillation (QBO)  
 9 indices defined by monthly zonal winds at 10 and 30 hPa, respectively, at Singapore  
 10 (Naujokat, 1986), and  $\omega = 2\pi/365.25$  (rad day<sup>-1</sup>). The QBO indices are used after  
 11 normalization by one standard deviation and application of a 12-month running mean (e.g.,  
 12 Gebhardt et al., 2013). For the regression, least-squares fitting was applied to the time series  
 13 of the average of the SR and SS in each pair (i.e.,  $(SR^i + SS^i)/2$ ; the time series of open stars  
 14 in Figure 3). The resulting  $O_{3\_BG}$  is also shown in Figure 3 (solid curve). We see that  $O_{3\_BG}$   
 15 changes by approximately 0.1 ppmv over 10 days, which is comparable with the SSD (see  
 16 below and Figure 4).

17 The SSD caused by seasonal variation for the  $i^{\text{th}}$  pair ( $SSD_{seas}^i$ ) was then evaluated as:

$$18 \quad SSD_{seas}^i = O_{3\_BG}(t_{SS}^i) - O_{3\_BG}(t_{SR}^i), \quad (5)$$

19 where  $t_{SS}^i$  and  $t_{SR}^i$  are the day of  $i^{\text{th}}$  SS and SR, respectively. Figure 4 also shows  $SSD_{seas}^i$   
 20 (blue closed circles).  $SSD_{seas}^i$  exhibits semiannual variations that follow the ozone SAO, with  
 21 an amplitude of up to 0.1 ppmv at this altitude. Finally, the corrected SSD ( $SSD_{cor}^i$ ; red open  
 22 circles in Figure 4) was calculated as

$$23 \quad SSD_{cor}^i = SSD_{org}^i - SSD_{seas}^i. \quad (6)$$

24 We expect that this SSD was only caused by diurnal variations and/or instrumental/retrieval  
 25 bias .

26 For ACE–FTS, only four pairs were obtained during the months of interest every year  
 27 because of the repeating orbit pattern. This leads to incorrect assessment of the seasonal  
 28 variations (particularly of the SAO) by the above fitting procedure (not shown). Thus, the  
 29 SSD for ACE ( $SSD_{cor}^i$ ) was calculated using  $SSD_{org}^i$  from ACE–FTS data and  $SSD_{seas}^i$  from

1 HALOE data. That is,  $SSD_{seas}^i$  is calculated for 2004-2011, using the fitting coefficients  
2 estimated from HALOE data during 1990-2005.

3 Having calculated the SSD for each SR-SS pair, the  $SSD_{cor}^i$  was binned and averaged using  
4 monthly bins. For example, if the day of a SR-SS pair was 13 April, the relevant  $SSD_{cor}^i$  was  
5 assigned to the April bin. Figure 4 shows the monthly average values of  $SSD_{cor}^i$ , as well as  
6 those of  $SSD_{org}^i$  and  $SSD_{seas}^i$ . If  $SSD_{org}$  was used for the analysis (see the black curve), it  
7 shows a semiannual variation with maxima in February and July/August/September; however,  
8 this is partially the result of contamination from  $SSD_{seas}$ , which shows a semiannual variation  
9 (ca. 0.1 ppmv) associated with the stratospheric SAO (see the blue curve; Ray et al., 1994).  
10 When this contamination is removed, as is done in the present study,  $SSD_{cor}$  instead shows an  
11 annual variation with a maximum between December and February (see the red curve). This  
12 seasonal variation is discussed in Section 4.2.

13 Finally, the annual mean SSD was calculated as the average of the monthly mean  $SSD_{cor}$ .  
14 Note that correction for  $SSD_{seas}$  is important in the context of the annual mean SSD as well.  
15 For SAGE II and HALOE, whose measurements cover the entire year, the annual mean of  
16  $SSD_{cor}$  is nearly the same as that of  $SSD_{org}$ , because the annual mean of  $SSD_{seas}$  should be  
17 zero. This is, however, not the case for ACE-FTS, which made tropical measurements only in  
18 particular months (i.e., February, April, August, and October). In this case, the annual mean  
19 of  $SSD_{seas}$  is not zero, but reaches a maximum of approximately 0.1 ppmv; thus, the annual  
20 mean of  $SSD_{org}$  is not equal to that of  $SSD_{cor}$  (not shown).

21 For SMILES, two kinds of SSDs are calculated with different procedures. The first kind of  
22 SSD is based on SR-SS pairs. SR-SS pairs that had more than 10 profiles for both SR and SS  
23 were analyzed, and four SR-SS pairs met this requirement (3 November 2009, 31 December  
24 2009, 29 January 2010, and 29 March 2010). Each pair had more than 200 profiles for both  
25 SR and SS. In all cases, SR and SS measurements were taken about 5 days apart. Unlike the  
26 solar occultation measurements, SMILES observed the 10°S-10°N region almost  
27 continuously (see Section 2.4). Consequently, the changes in background ozone  
28 concentration; i.e.,  $O_{3\_BG}$  in Equation (4), were defined as the time series of ozone  
29 concentration over the region 10°S-10°N to which a five-day running mean was applied.  
30 Then,  $SSD_{seas}$  was assessed using Equation (3) and  $SSD_{cor}$  was obtained from Equation (6).  
31 The average of the four  $SSD_{cor}$ s will be used in Section 4.1, while the individual  $SSD_{cor}$ s are  
32 used in Section 4.2 in the context of a discussion of their seasonal dependence. Next, the

1 second kind of SSD is the SSD caused by diurnal variations. This quantity is the difference in  
2 ozone levels between 1800 LT (for SS) and 0600 LT (for SR), as derived from hourly diurnal  
3 ozone-concentration variations in the SMILES data averaged over the period of operation  
4 (October 2009–April 2010). The method for extracting hourly diurnal variations from  
5 SMILES data was described in S13. Note that the times of SR and SS are 1800 LT ( $\pm 0.3$  hr)  
6 and 0600 LT ( $\pm 0.3$  hr), respectively, for the region  $10^{\circ}\text{S}$ – $10^{\circ}\text{N}$ .

7 For the SD–WACCM output for satellite coincidences (HALOE, SAGE II, and ACE–FTS),  
8 the same method was applied as that used for each satellite dataset. In contrast, for the full-  
9 grid SD–WACCM output, diurnal variations as a function of LT were extracted for each  
10 latitude and altitude grid point, for each month, as follows. First, three hourly diurnal  
11 variations in Universal time (UT) are calculated at each longitude, latitude and altitude grid  
12 point. Then, along each latitude band, data at the same local time are averaged to obtain  
13 diurnal variations as a function of LT (e.g., 0000 LT data are the average of (0000 UT,  $0^{\circ}\text{E}$ ),  
14 (0300 UT,  $315^{\circ}\text{E}$ ), (0600 UT,  $270^{\circ}\text{E}$ ), (0900 UT,  $225^{\circ}\text{E}$ ), (1200 UT,  $180^{\circ}\text{E}$ ), (1500 UT,  
15  $135^{\circ}\text{E}$ ), (1800 UT,  $90^{\circ}\text{E}$ ) and (2100 UT,  $45^{\circ}\text{E}$ )). Using this data, as for SMILES, the SSD  
16 caused by diurnal variations, that is, difference between 1800 and 0600 LT, was calculated.  
17 For SD–WACCM, as was done by S13, the contributions from vertical transport and  
18 photochemistry are estimated, by integrating Equation (1). Diurnal variations of vertical wind  
19 ( $w'$ ) and photochemical generation or loss ( $S'$ ) are both derived from the full-grid SD-  
20 WACCM output.

21

## 22 **4 Results**

### 23 **4.1 Annual Mean**

24 Figure 5a shows the annual mean vertical profiles of the SSD in ozone mixing ratio in the  
25 region between  $10^{\circ}\text{S}$  and  $10^{\circ}\text{N}$ , using data from the three solar occultation measurements and  
26 from SD–WACCM output during each satellite coincidence. We see that HALOE (blue solid  
27 curve) and ACE–FTS (green solid curve) agree well with each other for the entire  
28 stratosphere; SD–WACCM results at times of satellite coincidence (blue and green dashed  
29 curves) are also quantitatively consistent in the stratosphere (below  $\sim 55$  km). In the lower  
30 stratosphere, at an altitude of around 25 km, the SR profiles exhibit 0.05–0.1 ppmv more  
31 ozone than the SS profiles. This difference gradually decreases with increasing altitude and

1 reaches zero at about 30 km. In the upper stratosphere, at 40–45 km, the SS profiles exhibit up  
2 to 0.2 ppmv more ozone than the SR profiles. Figure 5b shows the ratio of the SSD to the  
3 average ozone mixing ratio during the analysis period. The SSD is between  $-2\%$  and  $-3\%$  at  
4 around 20 km, and gradually increases to  $5\%$  at an altitude of 45 km. The SSD near the  
5 stratopause is quantitatively consistent with the HALOE result of *Brühl et al.* (1996),  
6 although they only analyzed two latitude bands at around  $18^\circ\text{N}$  and  $18^\circ\text{S}$ .

7 SAGE II (red solid curve) shows a similar result to HALOE and ACE–FTS, at least  
8 qualitatively, but the SSD in the upper stratosphere is approximately twice as large as those  
9 resulting from the other datasets. The SAGE II results are essentially consistent with the  
10 results of McLinden et al. (2009) and Kyrölä et al. (2013), although the latter may be partially  
11 contaminated by seasonal/interannual ozone variations (see Section 3). Considering that the  
12 SD–WACCM results at SAGE II measurement locations (red dashed curve) are consistent  
13 with those at both HALOE and ACE–FTS measurement locations, we suggest that the  
14 anomalous SAGE II values were not due to the differences in the measurement  
15 locations/times compared with HALOE or ACE–FTS. Possible causes will be discussed in  
16 Section 5.

17 There is a disagreement in SSD between satellite measurements and SD–WACCM above 55  
18 km (Figure 5). This may be due to the difference in diurnal variations between the model and  
19 observations, as shown out by S13 for the altitude region between 50 km and 60 km. S13 also  
20 found that such a difference was attributed to the difference in the lowest altitude of the  
21 dominance of mesospheric diurnal variations that shows a marked day–night contrast (i.e.,  
22 ozone concentration maximizes (minimizes) during the nighttime (daytime)). Another  
23 possible reason for the disagreement in SSD is that the strong ‘horizontal’ gradient of ozone  
24 concentration associated with the rapid ozone changes at SS/SR in the mesosphere was not  
25 considered in the retrieval of satellite measurements (Natarajan et al., 2005).

26 Figure 5 also shows the two SMILES results. One is the SSD deduced from the solar zenith  
27 angle (black solid curve), while the other is the difference between 1800 and 0600 LT caused  
28 by diurnal variations (black dot-dashed curve). We see that the HALOE and ACE–FTS results  
29 are strongly supported by the SMILES results, although the SMILES SSD is about  $2\%$   
30 smaller than that based on HALOE and ACE–FTS data at an altitude of approximately 45 km.  
31 This may be because SMILES observations have only four SR–SS pairs in total whereas the  
32 HALOE and ACE–FTS have 86 and 24 pairs, respectively. SD–WACCM results at SMILES



1 locations also exhibit a smaller SSD compared with SD–WACCM results at HALOE and  
2 ACE–FTS locations (not shown). It should be noted that there is an agreement of the two  
3 results derived from the SMILES data. Also, Figure 6 compares the SSD from SD-WACCM  
4 results at HALOE measurement locations (gray solid curve) and that caused by diurnal  
5 variations from full-grid SD-WACCM (black solid curve), and again shows a good agreement.  
6 These findings indicate that the SSD is predominantly caused by natural diurnal variations in  
7 ozone concentrations (i.e., not by instrumental/retrieval bias).

8 Based on the finding that the SSD is consistent between satellite measurements (at least  
9 HALOE and ACE-FTS) and SD-WACCM, SD-WACCM data are further examined. Figure 7  
10 shows diurnal variations in ozone mixing ratio at 42 km, 32 km and 26 km, as derived from  
11 full-grid SD-WACCM data (black solid curve). The contribution from vertical transport by  
12 atmospheric tides (red solid curve) and that from photochemistry (blue solid curve) are also  
13 shown (see Equation (1) and Section 3). As shown by S13, the sum of the two processes  
14 (black dashed curve) explain almost all characteristics of diurnal variations (black solid curve).  
15 At 26 km, ozone levels reach a maximum in the morning and a minimum in the afternoon in  
16 the lower stratosphere mainly due to tidal vertical transport, resulting in a negative SSD. At  
17 32 km, ozone levels show a significant diurnal variation during the daytime due to  
18 photochemistry but they are almost zero at SR and SS, resulting in almost zero SSD.  
19 Chapman mechanism and NO<sub>x</sub> chemistry are related to the photochemical variations  
20 (Pallister and Tuck, 1983; Schanz et al., 2014). At 42 km, ozone levels show a diurnal  
21 variation caused by both vertical transport and photochemistry. But, as is the case at 32 km,  
22 photochemical processes do not contribute to the SSD significantly. In contrast, the vertical  
23 transport generates higher ozone levels in the late afternoon than in the morning; this causes a  
24 positive SSD. The vertical transport exhibits only small variations at 32 km because of the  
25 small vertical gradient in the background ozone concentration, i.e.,

26 
$$\frac{\partial[\overline{O_3}]}{\partial z} \approx 0 \quad (7)$$

27 in Equation (1) (see figure 7d of S13).

28 Figure 8 shows the amplitude and phase of diurnal migrating tide in vertical wind averaged  
29 between 10°S and 10°N, as derived from SD-WACCM data. The amplitude exponentially  
30 increases with altitude. The phase basically shows the downward progression but it is almost  
31 constant around 40 km possibly due to the presence of trapped modes excited by ozone

1 heating (e.g., Sakazaki et al., 2013b). Consequently, the phase of vertical wind is similar in  
2 the whole stratosphere; thus, the sign of SSD (i.e., negative at 20-30 km and positive at 35-50  
3 km) is determined by that of vertical gradient in the background ozone concentration.

4 Figure 6 also shows the contributions from vertical transport and photochemistry to the  
5 annual-mean SSD profile. It is clearly demonstrated that the SSD in the tropical stratosphere  
6 is mainly caused by vertical transport while the contribution from photochemistry is  
7 negligible. Note that a further analysis of SD-WACCM indicated that the photochemical  
8 variations cause a SSD of  $\sim 0.1$  ppmv at high latitudes ( $>40^\circ$ ), where they are not zero at SR  
9 and SS (not shown; cf. Schanz et al., 2014). The relationship between the SSD and vertical  
10 tidal winds will be further demonstrated in the next section, where we will examine the  
11 seasonal variations of the SSD.

## 12 **4.2 Seasonal Variations**

13 This section examines seasonal variations in the SSD. It should be emphasized here that by  
14 calculating the seasonal variations in SSD using solar occultation measurements, we could  
15 obtain quasi-observational evidence of seasonal variations in stratospheric vertical tidal winds,  
16 which are difficult to observe directly. This section mainly use HALOE data, because the  
17 SSD based on the HALOE measurements covers a wider range of months. Figure 9 shows the  
18 month–altitude distribution of the SSD, from HALOE and SD–WACCM at HALOE locations.  
19 In the upper stratosphere at altitudes of 40–50 km, both datasets show that the SSD reaches a  
20 maximum during the Northern Hemisphere winter between December and February (0.2–0.3  
21 ppmv; see also Figure 4). In the lower stratosphere, at 20–30 km, the SSD reaches a minimum  
22 twice, in March–April and again in August–September.

23 These seasonal variations can be confirmed on the basis of other datasets. Figure 10 shows the  
24 seasonal variations in the SSD averaged between altitudes of 40 and 45 km and also between  
25 22 and 28 km, as derived from the three solar occultation measurements, as well as the  
26 SMILES and SD–WACCM data at satellite measurement locations. In the upper stratosphere  
27 (see Figure 10a), all data are consistent in the sense that the SSD reaches a maximum between  
28 December and February. Note that the SSD based on SAGE II data is larger by a constant  
29 amount (ca. 0.2 ppmv) than that based on the other datasets, for the entire year. In the lower  
30 stratosphere (see Figure 10b), all data (except for those from ACE–FTS) also agree  
31 reasonably well in that the SSD reaches a minimum between March and April, while another

1 minimum is discernable between August and November. The disagreement seen for the  
2 ACE–FTS data may be caused by the smaller number of SR–SS pairs, or because  $SSD_{\text{seas}}$  was  
3 estimated using HALOE data rather than ACE–FTS data (see Section 3).

4 Based on the finding that the seasonal dependence of SSD in the SD–WACCM data is  
5 consistent with those in the satellite data, the full-grid SD–WACCM data are further analyzed  
6 to interpret the seasonal variations of SSD from the perspective of those of diurnal variations  
7 in ozone concentration. Figure 11a shows the month–altitude distribution of the difference in  
8 ozone mixing ratio between 1800 LT (for SS) and 0600 LT (for SR); i.e., the SSD due to  
9 diurnal variations, as derived from the full-grid SD–WACCM data (see also the black dashed  
10 curve in Figure 10). The seasonal dependence shown in Figure 11a agrees well with that in  
11 Figure 9b, both in the upper stratosphere (the positive-SSD region) and in the lower  
12 stratosphere (the negative-SSD region). We conclude that the seasonal dependence of the  
13 SSD is mainly caused by that in the diurnal variations in ozone level. Figure 11b shows the  
14 seasonal variation of the SSD related to vertical tidal transport only (Equation 1). Good  
15 agreement between Figure 11a and b again indicates that the SSD can be largely explained by  
16 diurnal variations caused by vertical tidal transport. It is also suggested that the seasonal  
17 dependence of the SSD is caused by that of the tidal vertical winds.

18 Although tides are mainly composed of diurnal and semidiurnal components, the former  
19 cause the SSD, because the semidiurnal cycle does not cause any difference between two  
20 measurements taken 12 hours apart (i.e., normal SR and SS at 0600 and 1800 LT). Figure 12  
21 shows the tropical month–latitude distributions of the diurnal migrating tide in the vertical  
22 winds at 42 and 26 km, derived from the SD–WACCM results. At an altitude of 42 km, the  
23 amplitude is greatest during the Northern Hemisphere winter, while a small secondary  
24 maximum is discernable in the period August–September between 20°S and 10°S. At 26 km,  
25 we see similar double maxima, one in February–March and the other in August–September.  
26 The seasonal variations in vertical wind are consistent with those in temperature (e.g., Wu et  
27 al., 1998; Sakazaki et al., 2012). Sakazaki et al. (2013b) attributed the double maxima to the  
28 seasonal dependence of the background zonal winds; the meridional advection of the zonal  
29 wind momentum peaks twice a year, producing the tidal seasonal variation in the stratosphere.  
30 We conclude that the seasonal variation in the vertical tidal wind causes the variation of the  
31 vertical transport of ozone (see Figure 11b), and, consequently, this results in the seasonal  
32 variation of the SSD (see Figure 11a). The secondary maximum in August–September at an

1 altitude of 42 km, as seen in the vertical wind, does not appear in the SSD, probably because  
2 we only analyzed the SSD in the region 10°S–10°N.

3

## 4 **5 Discussion and Concluding Remarks**

5 We quantified the SSD (sunset-minus-sunrise difference) for the tropical stratosphere based  
6 on three different measurements of solar occultation (SAGE II, HALOE, and ACE–FTS)  
7 supplemented by data from SMILES measurements and the SD–WACCM model. To quantify  
8 the SSD, we addressed the importance of removing the effects of sampling issues on the SSD,  
9 particularly contamination by seasonal variations in ozone level. If contamination by seasonal  
10 variations in ozone concentration is not removed in advance (i.e., if monthly composites are  
11 calculated for  $SSD_{org}^i$ ), the SSD shows seasonal variations that are contaminated by signals  
12 associated with the SAO. These sampling issues should be taken into account when correcting  
13 the SSD to create homogenized datasets based on solar occultation measurements.

14 All data, except for those from SAGE II, indicated that the SSD is up to +0.2 ppmv in the  
15 upper stratosphere at altitudes of 40–45 km, and up to -0.1 ppmv in the lower stratosphere at  
16 around 25 km. SMILES and SD–WACCM results suggested that the SSD is caused by natural  
17 diurnal variations in ozone concentration, particularly variations related to vertical transport  
18 by tidal winds. SAGE II data showed qualitatively similar results, including the seasonal  
19 dependence, but the SSD in the upper stratosphere was twice as large as in the other datasets.

20 The SSD also shows seasonal variations: it reaches a maximum during the Northern  
21 Hemisphere winter at an altitude of 42 km, and minima in March–April and September–  
22 October near an altitude of 26 km. Analysis of SD–WACCM output showed that the seasonal  
23 variation in the SSD follows that of the tidal vertical winds. We believe that we have obtained  
24 the first quasi-observational evidence of the tidal vertical wind and its seasonal dependence in  
25 the tropical stratosphere. Therefore, we suggest that the ozone SSD is not a bias, but a useful  
26 proxy for the vertical tidal wind.

27 Nevertheless, there is an uncertainty about the magnitude of SSD; the SSD from SAGE II  
28 data is twice as large as that from other four datasets. With the analysis of SD–WACCM  
29 output, this difference is found not due to the difference in measurement locations or times  
30 among the datasets. The retrieval procedure in SAGE II has been greatly updated in SAGE II

1 version 7 data (Damadeo et al., 2013). We have so far no clear answer to the difference, and  
2 there may be several possibilities as discussed in the following.

3 A SSD could occur if the measurement “altitude” is mistakenly defined at SR and/or SS. But,  
4 the observed uncertainty in SSD of ~4% (the difference in SSD between SAGE II and the  
5 others at 42 km) corresponds to the uncertainty in altitude of ~400 m (at 42 km, the ozone  
6 mixing ratio changes 1% per 100 m in vertical); this seems unrealistic, because an uncertainty  
7 of defined altitude is considered 20 m for SAGE II, 100 m for HALOE, and 150 m for ACE-  
8 FTS.

9 [Another related issue may be the reproducibility of tides in SD-WACCM and reanalyses.](#)  
10 [Satellite measurements use, more or less, \(re\)analysis data for the altitude registration and/or](#)  
11 [the retrieval process. It is found that the amplitude of diurnal tide in SD-WACCM and](#)  
12 [reanalyses is up to ~50% smaller in the upper stratosphere compared to than in data from the](#)  
13 [Sounding of the Atmosphere using Broadband Emission Radiometry \(SABER\) measurements](#)  
14 [\(version 2.0 data\) \(not shown; see Sakazaki et al. 2012 for the comparison between SABER](#)  
15 [and reanalyses\). This could affect any of the satellite datasets, but a further study is needed for](#)  
16 [a more quantitative discussion.](#)

17

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6

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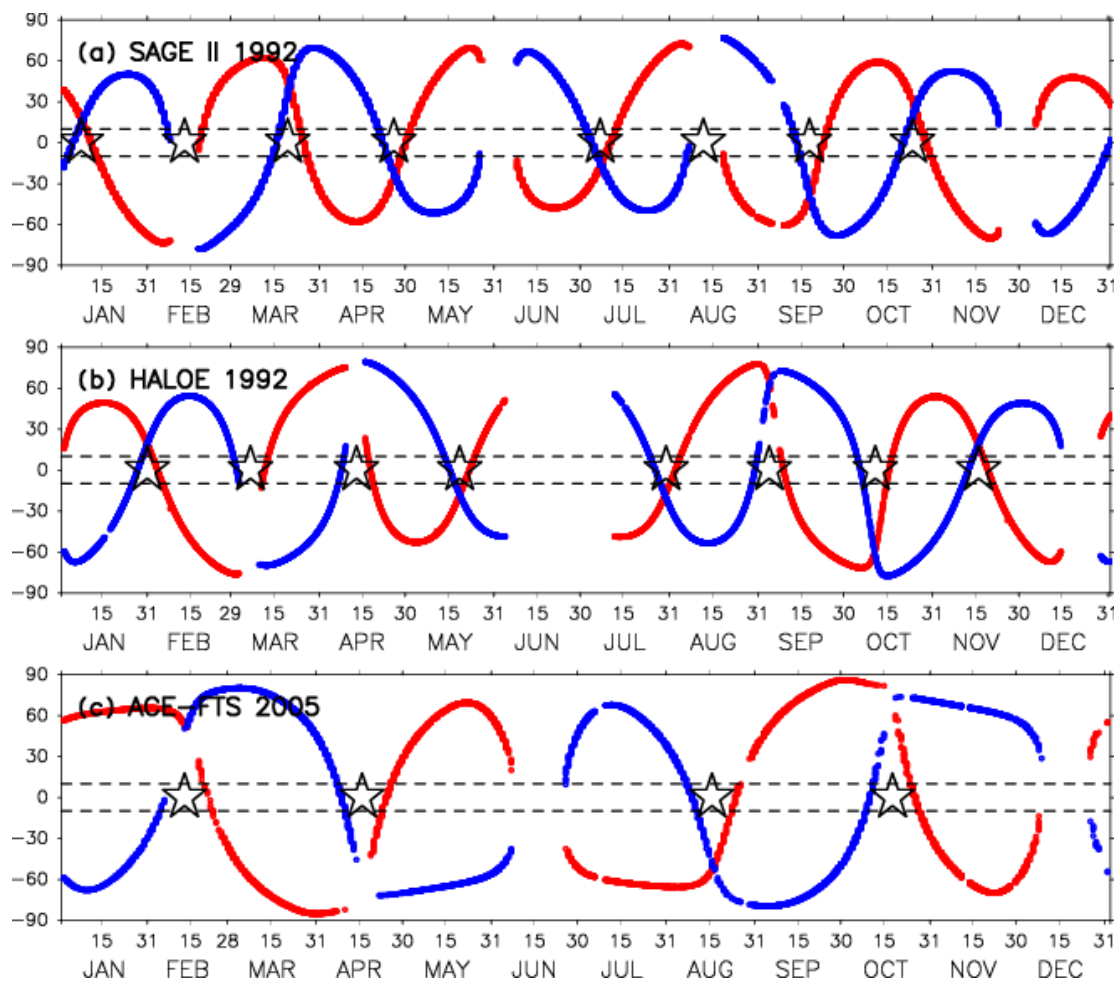
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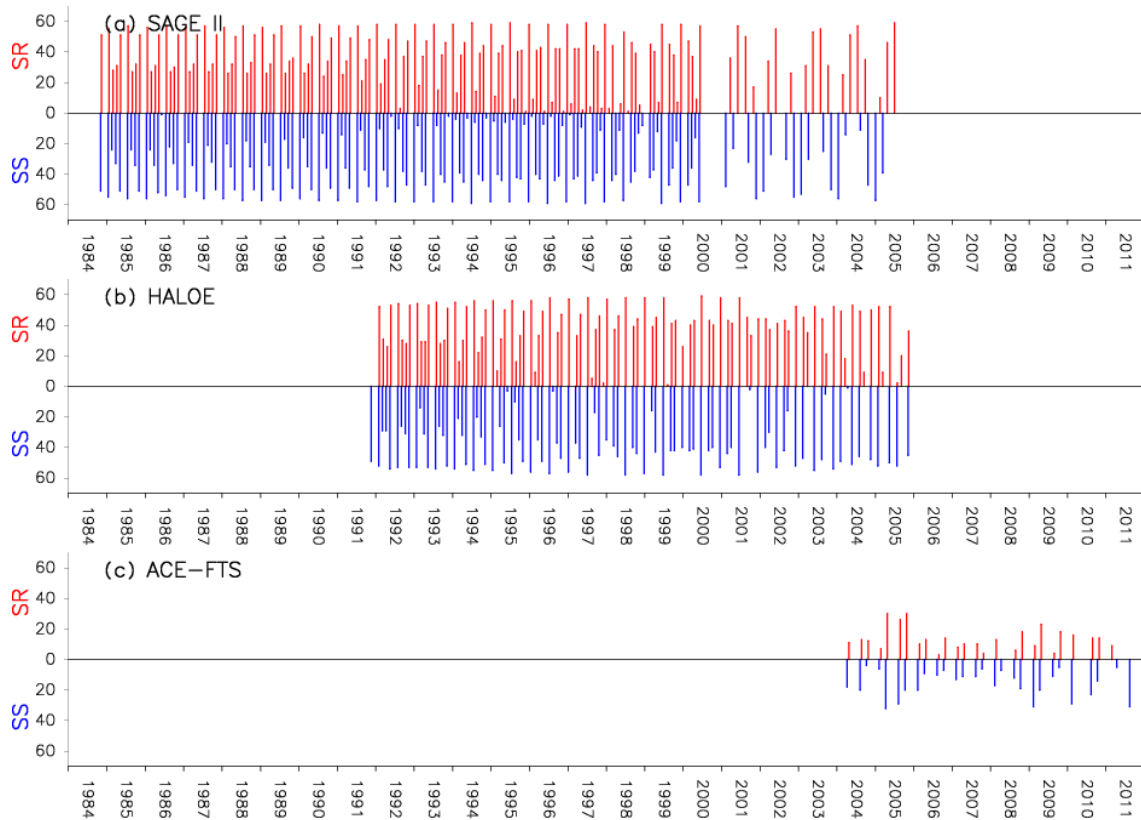
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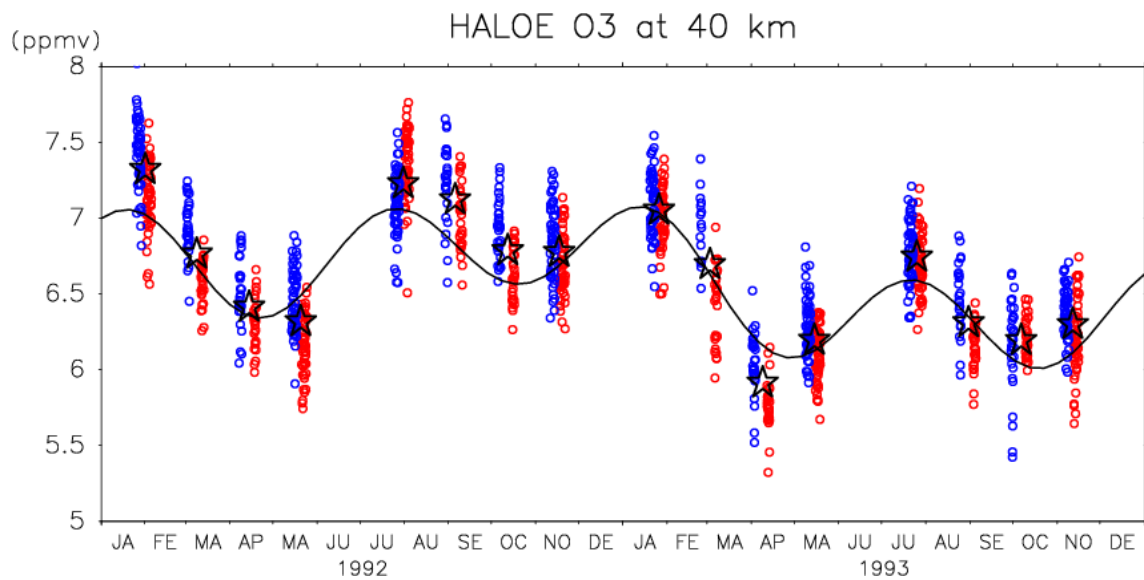
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Figure 1. Measurement tracks of (a) SAGE II in 1992, (b) HALOE in 1992, and (c) ACE-FTS in 2005. Red and blue closed circles show SR and SS measurements, respectively. Open stars indicate SR-SS measurement pairs. Dashed lines denote 10°S/N.



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Figure 2. Number of profiles obtained for observations at 10°S–10°N from (a) SAGE II, (b) HALOE, and (c) ACE-FTS. Red and blue bars show the number of SR and SS profiles, respectively.

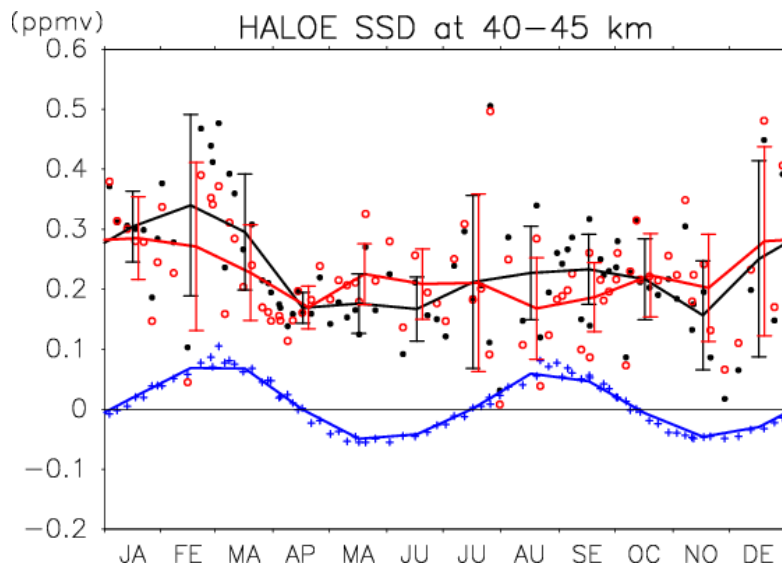


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2 Figure 3. Time series of the ozone mixing ratio observed by HALOE during the period 1992–  
 3 1993 at an altitude of 40 km (between 10°S and 10°N). Red and blue open circles show  
 4 individual measurements at SR and SS, respectively. Open stars denote the average values of  
 5 SR and SS, for each SR–SS pair. The solid curve shows the best-fit function for the average  
 6 values (see Equation 4).

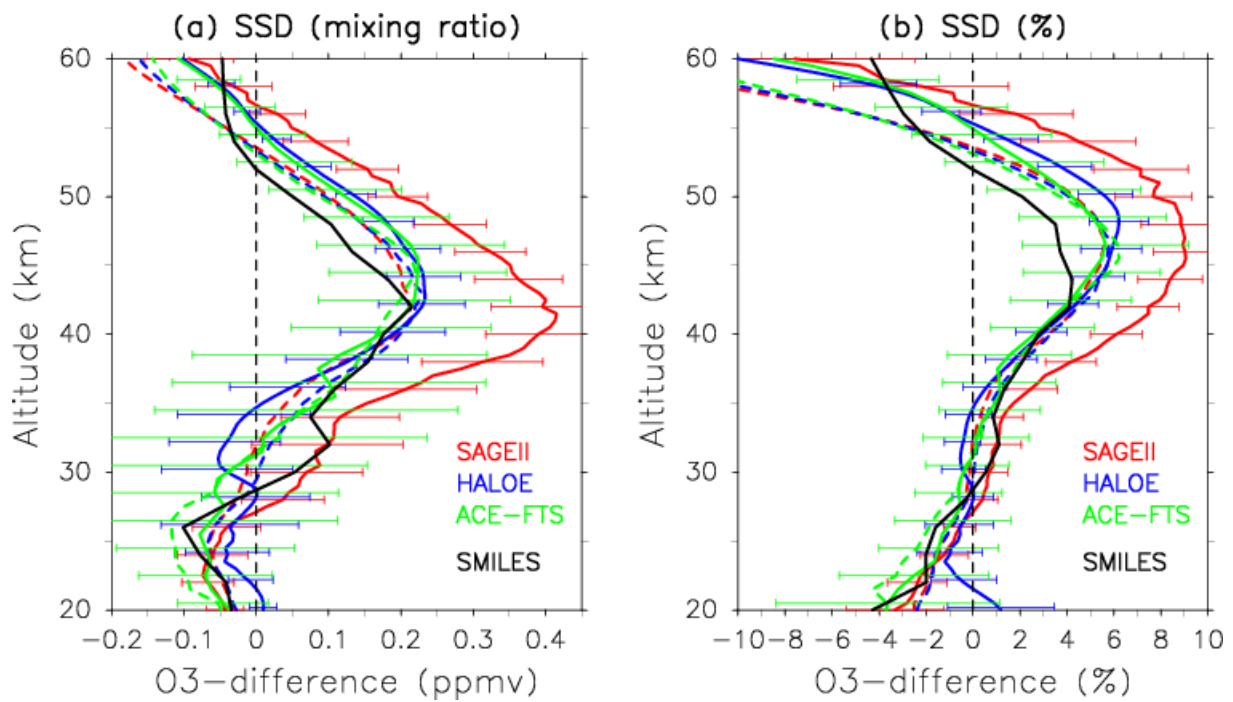
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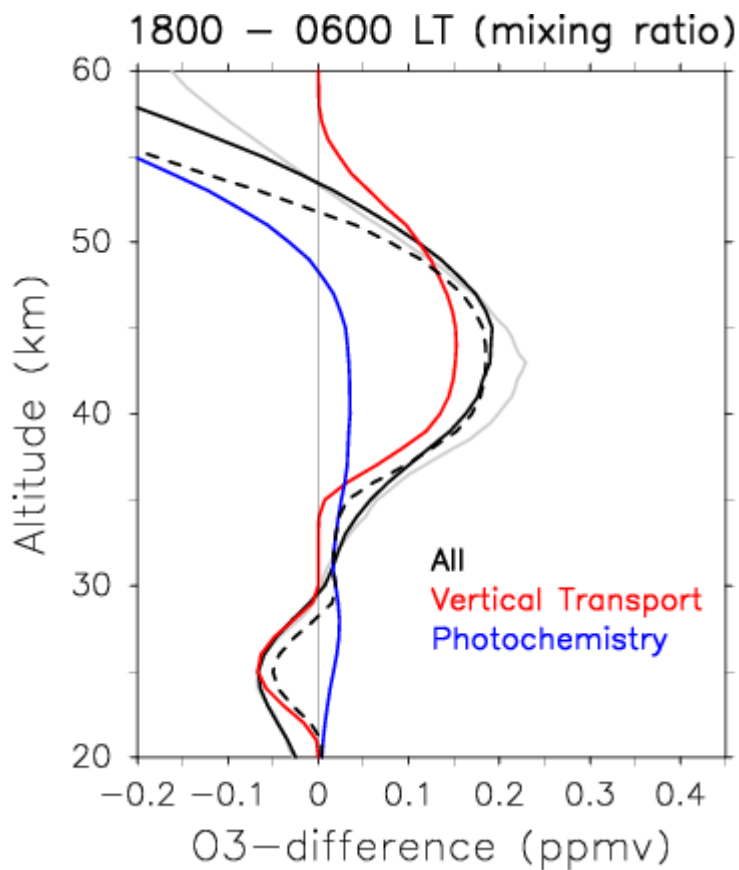
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 2 Figure 4.  $SSD^{i_{org}}$  (black closed circles),  $SSD^{i_{seas}}$  (blue crosses), and  $SSD^{i_{cor}}$  (red open circles)  
 3 averaged at an altitude of 40–45 km for individual SR–SS pairs sorted by time of year,  
 4 derived from HALOE data during the period 1990–2005. Black, blue, and red curves denote  
 5 monthly averaged  $SSD_{org}$ ,  $SSD_{seas}$ , and  $SSD_{cor}$ , respectively. Vertical bars show one standard  
 6 deviation for the monthly  $SSD_{org}$  and  $SSD_{cor}$ .

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 2 Figure 5.  $SSD_{cor}$  for  $10^{\circ}S-10^{\circ}N$  in (a) ozone mixing ratio (ppmv), and (b) SSD ratio to the  
 3 daily mean (%), derived from SAGE II (red solid curves), HALOE (blue solid curves), and  
 4 ACE-FTS (green solid curves). Red, blue, and green dashed curves denote SD-WACCM  
 5 results at SAGE II, HALOE, and ACE-FTS coincidence, respectively. Black solid curves  
 6 show the SMILES result (SR and SS are defined by those profiles with a solar zenith angle  
 7 between  $80^{\circ}$  and  $100^{\circ}$ ). Black dot-dashed curves show the difference between 1800 and 0600  
 8 LT, calculated using SMILES data and based on 1-hourly diurnal variations. Horizontal bars  
 9 for SAGE II, HALOE and ACE-FTS show 95% confidential levels in  $t$ -test. For the statistical  
 10 test, the error is defined as the standard deviation for the monthly SSD; this quantity has been  
 11 propagated to the error in annual-mean. Then, the  $t$ -test has been made with the degrees of  
 12 freedom being the total numbers of SS-SR pairs for each dataset.  
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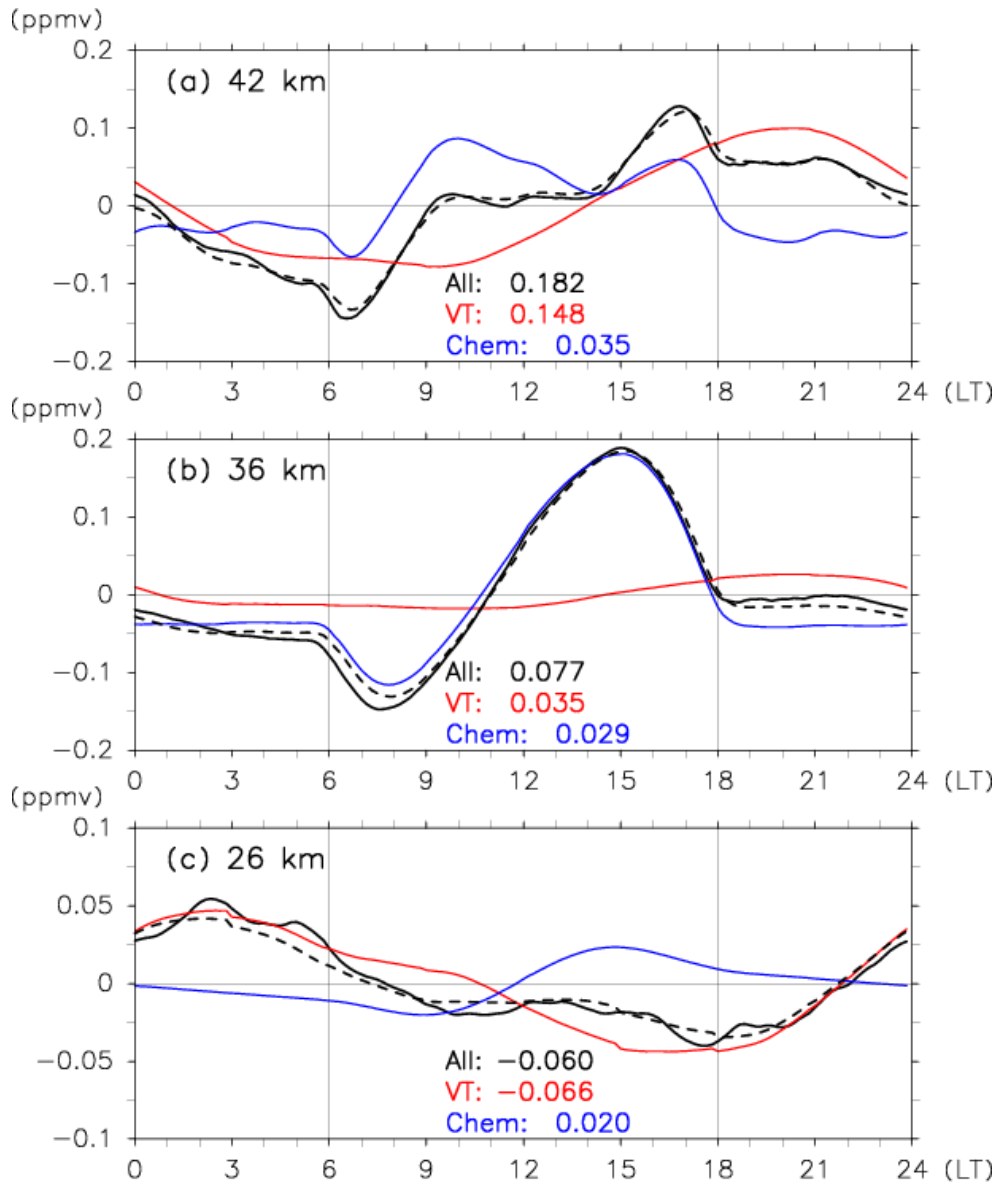


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2 Figure 6. As Figure 5a, but for the difference between 1800 and 0600 LT, as deduced from  
 3 diurnal variations in ozone concentration based on full-grid SD-WACCM between 2008 and  
 4 2010. Solid curve is for the diurnal variations, red solid curve is for the contribution from the  
 5 vertical transport, blue solid curve is for the contribution from photochemical processes, and  
 6 the dashed solid curve is for the sum of tidal vertical transport and photochemical processes  
 7 (see Equation (1) and Section 3 for details). Solid gray curve shows the SSD in SD-WACCM  
 8 data at HALOE measurement locations (the same as blue dashed curve in Figure 5) for a  
 9 reference.

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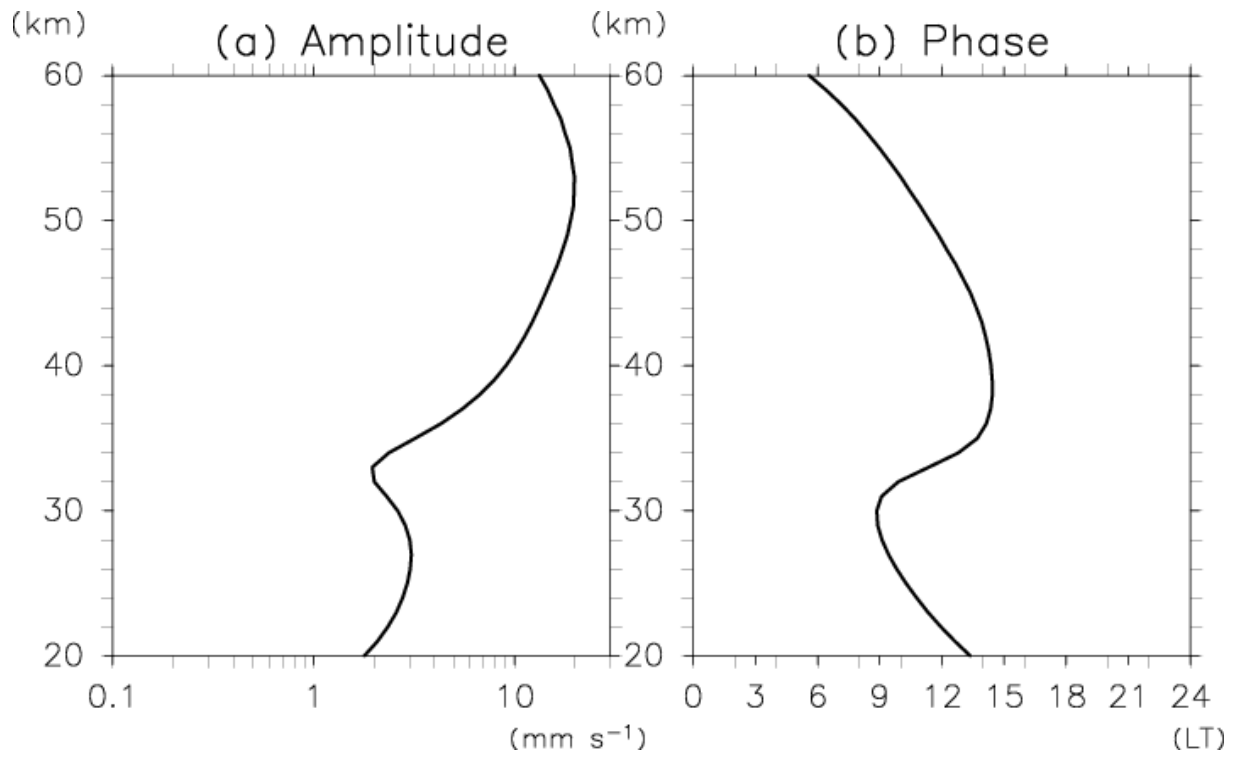


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2 Figure 7. Diurnal variations in ozone mixing ratio averaged over 10°S-10°N at altitudes of (a)  
3 42 km, (b) 36 km, and (c) 26 km. Black solid curves are SD-WACCM. Red solid curves show  
4 the contribution from tidal vertical transport, and blue solid curves show the contribution from  
5 photochemical processes (see Equation (1) and Section 3), while black dashed curves show  
6 the sum of the two processes. SR and SS are denoted by thin black solid lines at 0600 LT and  
7 1800 LT, respectively. The differences between 1800 and 0600 LT in ozone concentration  
8 (ppmv) from diurnal variations (“All”), tidal vertical transport (“VT”), and photochemical  
9 processes (“Chem”), are shown in each panel.

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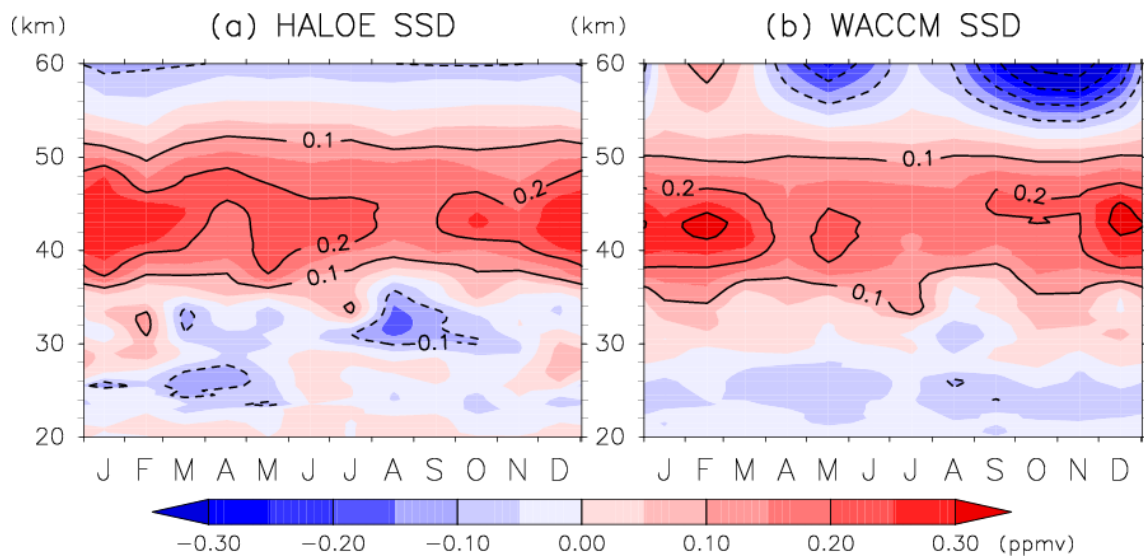


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3 Figure 8: Vertical profiles of (a) amplitude and (b) phase for diurnal migrating tide in vertical  
4 wind, averaged between 10°S and 10°N, as derived from SD-WACCM data during 2008-  
5 2010.

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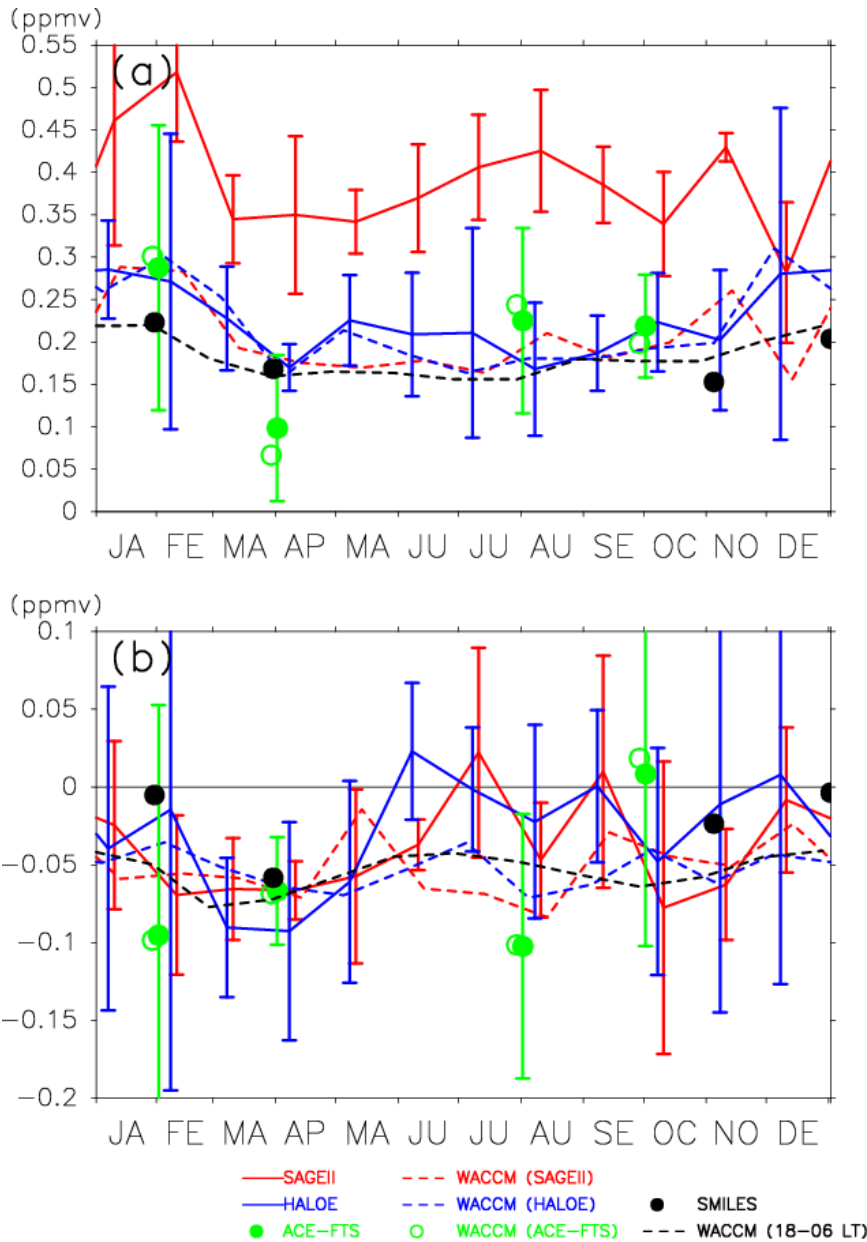


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2 Figure 9. Month–altitude distributions of the SSD for the latitude range 10°S–10°N, derived  
 3 from (a) HALOE and (b) SD–WACCM at HALOE measurement locations. Contour intervals  
 4 are 0.1 ppmv.

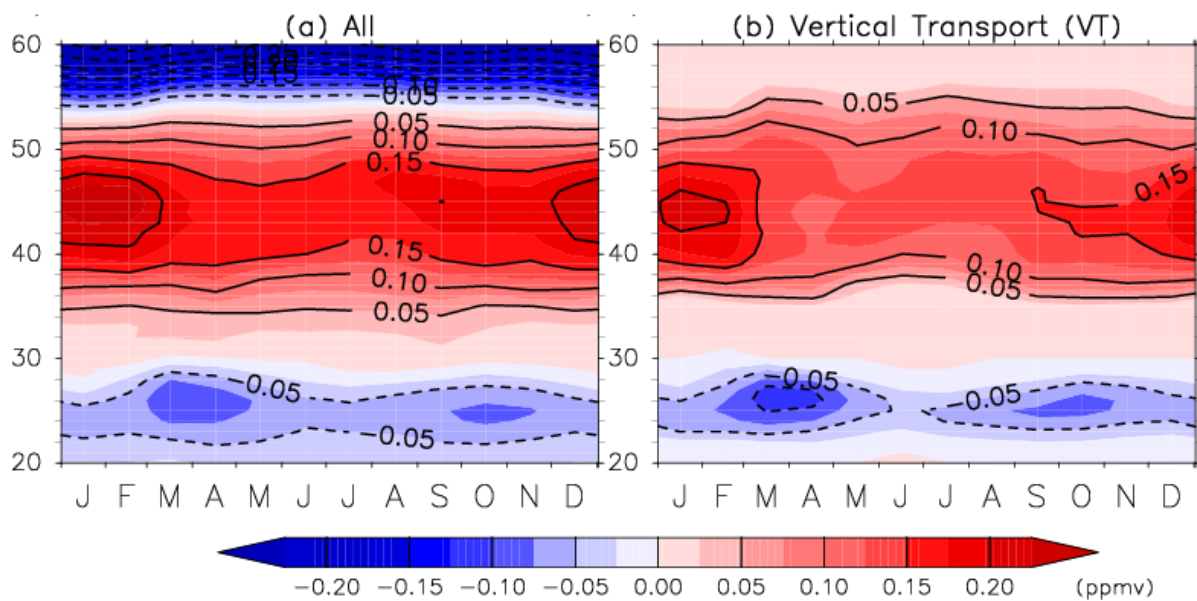
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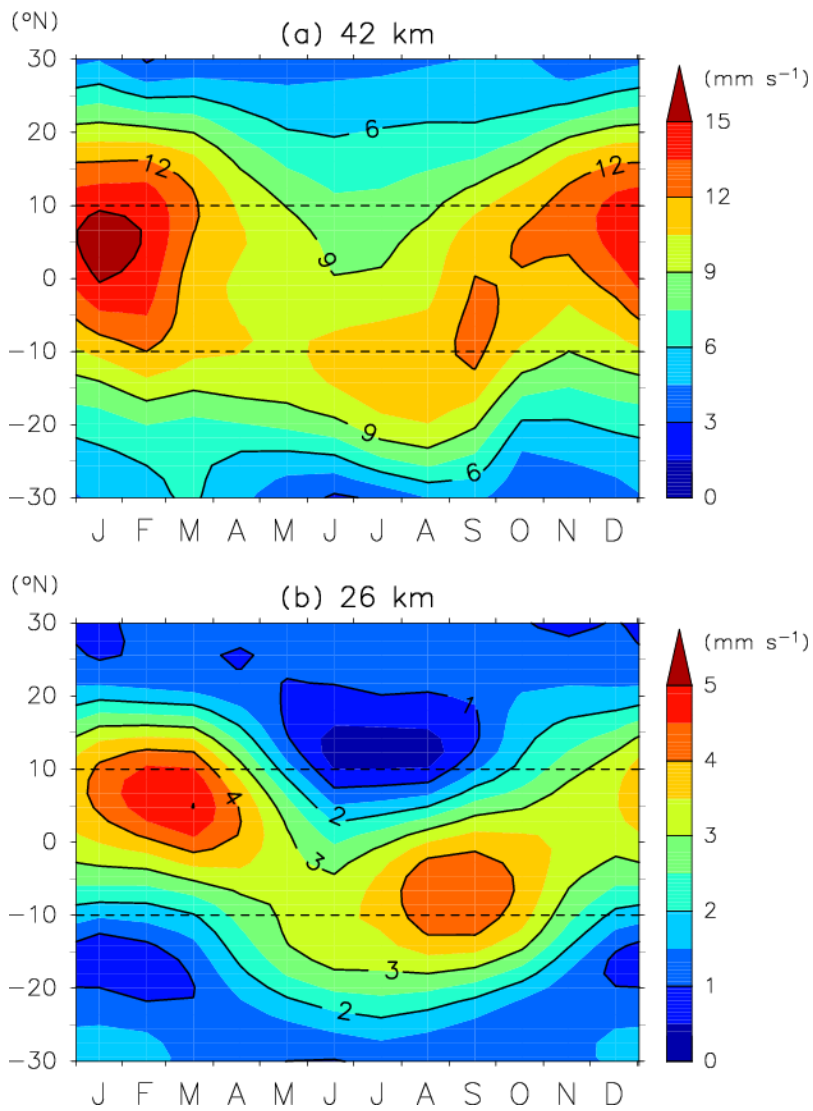


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 2 Figure 10. Seasonal variation of O<sub>3</sub> SSD<sub>cor</sub> at altitudes of (a) 40–45 km and (b) 22–28 km,  
 3 obtained from (red solid curve) SAGE II, (red dashed curve) SD–WACCM at SAGE II  
 4 locations, (blue solid curve) HALOE, (blue dashed curve) SD–WACCM at HALOE locations,  
 5 (green closed circles) ACE–FTS, (green open circles) SD–WACCM at ACE–FTS locations,  
 6 and (black closed circles) SMILES. The black dashed curve shows the difference in ozone  
 7 between 1800 and 0600 LT, as deduced from diurnal variations in ozone concentration based  
 8 on full-grid SD–WACCM between 2008 and 2010. Vertical bars for SAGE II, HALOE and  
 9 ACE-FTS show 95% confidential levels with *t*-test.

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 2 Figure 11. (a) As Figure 9, but for the difference in ozone mixing ratio between 1800 and  
 3 0600 LT, deduced from diurnal variations in ozone concentration based on full-grid SD-  
 4 WACCM during the period 2008–2010. (b) As (a), but for the contribution from tidal vertical  
 5 transport (see Equation 1).  
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 2 Figure 12. Month–latitude distributions of the amplitude of the diurnal migrating tide in  
 3 vertical winds at altitudes of (a) 42 km and (b) 26 km, derived from SD–WACCM data for the  
 4 period 2008–2010.

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