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# Comparison of ozone profiles between Superconducting Submillimeter-Wave Limb-Emission Sounder and worldwide ozonesonde measurements

Koji Imai,[1] Masatomo Fujiwara,[2] Yoichi Inai,[3] Naohiro Manago,[4] Makoto Suzuki,[1] Takuki Sano,[1] Chihiro Mitsuda,[5] Yoko Naito,[6] Fumio Hasebe,[2] Takashi Koide,[1] and Masato Shiotani [1,3]

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[1] We compared ozone profiles measured by the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES) with those taken at worldwide ozonesonde stations. To assess the quality of the SMILES version 2.3 ozone data for 16–30 km, 601 ozonesonde profiles were compared with the coincident SMILES ozone profiles. The agreement between SMILES and ozonesonde measurements was generally good within 5%–7% for 18–30 km at middle and high latitudes but degraded below 18 km. At low latitudes, however, the SMILES ozone data showed larger values (~6%–15% for 20–26 km) than those at middle and high latitudes. To explain this bias, we explored some possible issues in the ozonesonde measurement system. One possibility is due to a pressure bias in radiosonde measurements with a pressure sensor, but it would be within a few percent. We also examined an issue of the ozonesonde's response time. The response time was estimated from ozonesonde measurements with ascending and descending profiles showing clear difference, by using the time lag correction method to minimize the difference between them. Our estimation shows 28 s on average which is a similar value derived by prelaunch preparation. By applying this correction to the original profiles, we found a negative bias of the ascending ozonesonde measurement more than 7% at 20 km in the equatorial latitude where the vertical gradient of ozone is steep. The corrected ozonesonde profiles showed better agreement with the SMILES data. We suggest that the response time of ozonesondes could create a negative bias, particularly in the lower stratosphere at equatorial latitudes.

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## 1. Introduction

[2] To demonstrate the high sensitivity of the 4 K cooled submillimeter limb sounder in the environment of outer space and to monitor the global distributions of middle-atmosphere trace gases, the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES) was developed and deployed on the Japanese Experiment Module (JEM) on the International Space Station (ISS) through the cooperation of the Japan Aerospace Exploration Agency and the National Institute of Information and Communications Technology. SMILES conducted high-sensitivity limb soundings for the middle atmosphere from 12 October 2009 to 21 April 2010 (see the overview by *Kikuchi et al.* [2010]). The SMILES Level 2 (L2) data-processing system [*Mitsuda et al.*, 2011; *Takahashi et al.*, 2010, 2011] retrieves vertical profiles of minor atmospheric constituents from the calibrated radiance observations (Level 1 data), and SMILES version 2.3 (hereafter v2.3) L2 products were released for public use in November 2012. In the extensive comparisons with existing satellite data sources, the SMILES ozone generally shows good agreements within 10% (30%) in the stratosphere (mesosphere) [*Imai et al.*, 2013]. In this study we separately compare the SMILES v2.3 ozone data with ozonesonde measurements, since we add some discussion about potential contributions of the pressure bias in radiosonde measurements and the time lag issue in ozonesonde measurements.

[3] Ozonesondes are balloon-borne in situ electrochemical instruments that continuously measure ozone concentrations

In the originally published version of this article, Figure 8 contained incorrect data. The corrected Figure 8 was inserted 11 FEB 2014. A related sentence on page 12,762 was also amended.

[1]Institute of Space and Astronautical Science, Japan Aerospace Exploration Agency, Sagamihara, Japan.
[2]Faculty of Environmental Earth Science, Hokkaido University, Sapporo, Japan.
[3]Research Institute for Sustainable Humanosphere, Kyoto University, Uji, Japan.
[4]Center for Environmental Remote Sensing, Chiba University, Chiba, Japan.
[5]Fujitsu FIP Corporation, Tokyo, Japan.
[6]Faculty of Science, Kyoto University, Kyoto, Japan.

Corresponding author: K. Imai, Institute of Space and Astronautical Science, Japan Aerospace Exploration Agency, Sagamihara, Japan. (imai.koji@jaxa.jp)


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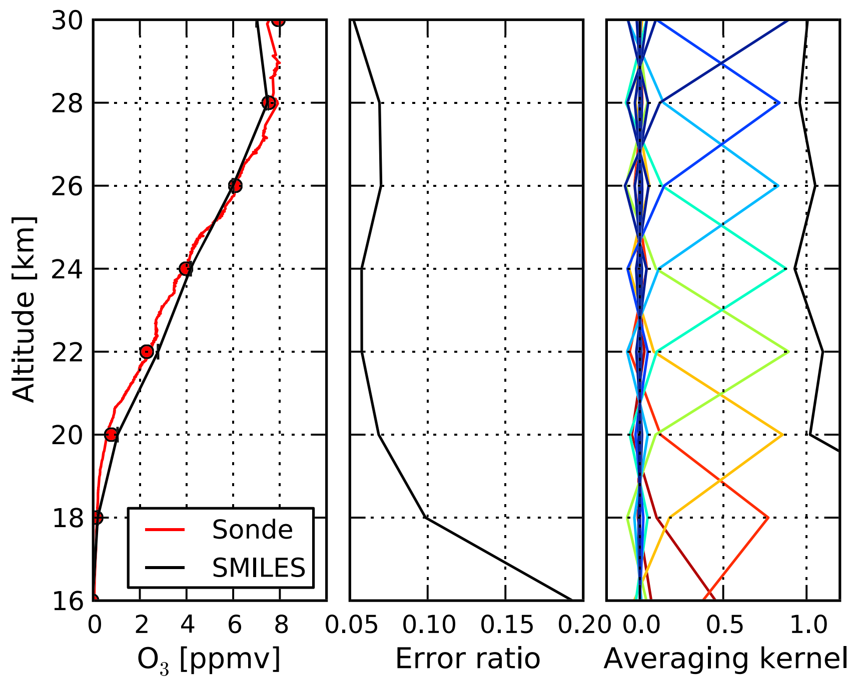


**Figure 1.** Example of (left) coincident SMILES and ozonesonde measurements, (middle) the error ratio, and (right) the SMILES ozone averaging kernels. (left) The solid red line is an ozonesonde profile taken at Hanoi, Vietnam (21.0°N and 105.8°E) launched at 7:53 UTC on 20 January 2010, and the red circles are values convolved with the SMILES ozone averaging kernels, while the solid black line is a SMILES ozone profile at 21.59°N and 97.87°E at 3:39 UTC. (middle) The solid black line is an error ratio calculated from the SMILES L2 retrieval system. (right) Colored solid lines show the corresponding vertical averaging kernels as a function of the retrieval level. The solid black line shows the integrated area under each of the colored curves.

from their release point on the ground up to the burst point of the meteorological rubber balloon, typically at 30–35 km. The ozonesondes currently used worldwide are well-established instruments whose measurement uncertainty has been evaluated in various field campaigns and laboratory experiments [e.g., *Smit et al*., 2007; *Thompson et al*., 2007, and references therein]. The ozonesonde profiles have been used for evaluating model calculations [e.g., *Tripathi et al*., 2005] and validating satellite data products [*Hoogen et al*., 1999; *Randall et al*., 2003]. Recently, such validation studies using the ozonesonde data have been carried out for the Microwave Limb Sounder (MLS) on the Earth Observing System Aura satellite [*Jiang et al*., 2007] and for the Fourier transform spectrometer and the Measurement of Aerosol Extinction in the Stratosphere and Troposphere Retrieved by Occultation onboard the Atmospheric Chemistry Experiment [*Hegglin et al*., 2008; *Dupuy et al*., 2009]. We also use the worldwide ozonesonde data to evaluate the measurement accuracy of the SMILES ozone data in the middle and lower stratosphere.

[4] For validation, we used the ozonesonde data taken from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC) [*Fioletov et al*., 2008], the Southern Hemisphere Additional Ozonesondes (SHADOZ) archives [*Thompson et al*., 2003a], and the Soundings of Ozone and Water in the Equatorial Region (SOWER) project [*Hasebe et al*., 2007]. An intensive SOWER campaign was conducted in January 2010, during which ~15 ozonesondes along with stratospheric water vapor sondes were launched in the western tropical Pacific. The ozonesonde sites of the SHADOZ project were mostly distributed in the tropical latitudes; the SHADOZ archives are available at http://croc.gsfc.nasa.gov/shadoz. Worldwide ozonesonde data of other stations are available at the WOUDC website, http://www.woudc.org/. Note that for early validation of the SMILES ozone measurements, ozonesonde profiles taken during the SMILES observation period were readily available by courtesy of the following sites: Ascension Island, Hanoi, La Réunion, Lindenberg, Naha, Natal, Sapporo, Tsukuba, and Wallops Island.

[5] The study is organized as follows: a description of the data including the handling procedure and the analysis method is presented in section 2, comparisons between the SMILES ozone data and the ozonesonde measurements are included in section 3, discussion about some issues in the ozonesonde system is presented in section 4, and a summary of this study is presented in section 5.

## 2. Data Description and Analysis Method

### 2.1. SMILES Ozone Measurements

[6] SMILES measured the Earth's limb from 12 October 2009 to 21 April 2010. Since the antenna beam is deflected 45° left from the direction of orbital motion, SMILES nominally covered latitudes from 38°S to 65°N on each orbit within a 93 min period. The antenna was scanned in elevation at a period of 53 s, and the total number of scans per day was about 1600. The nominal retrieved altitude range is from 8 to 100 km; the vertical grid step of the ozone product is 2 km in the altitude range 8–58 km and 3 km in the altitude range 58–100 km for the v2.3 products. An example of the SMILES single profile is shown in Figure 1 (left).

**Table 1.** Ozonesonde Site Information and the Number of Coincident Profiles for Comparisons Used in This Study

| No. | Location | Latitude | Longitude | Data Source | # |
|---|---|---|---|---|---|
| 1 | Lerwick | 60.1 | −1.2 | WOUDC | 10 |
| 2 | Legionowo | 52.4 | 21.0 | WOUDC | 20 |
| 3 | Valentia Observatory | 51.9 | −10.3 | WOUDC | 26 |
| 4 | UCCLE | 50.8 | 4.4 | WOUDC | 17 |
| 5 | Praha | 50.0 | 14.4 | WOUDC | 37 |
| 6 | Hohenpeissenberg | 47.8 | 11.0 | WOUDC | 77 |
| 7 | Payerne | 46.5 | 6.6 | WOUDC | 76 |
| 8 | Sapporo | 43.1 | 141.3 | WOUDC | 20 |
| 9 | Barajas | 40.5 | −3.6 | WOUDC | 30 |
| 10 | Ankara | 40.0 | 32.9 | WOUDC | 8 |
| 11 | Wallops Is. | 37.9 | −75.5 | WOUDC | 25 |
| 12 | Tsukuba | 36.1 | 140.1 | WOUDC | 29 |
| 13 | Isfahan | 32.5 | 51.7 | WOUDC | 5 |
| 14 | Naha | 26.2 | 127.7 | WOUDC | 27 |
| 15 | Hong Kong Observatory | 22.3 | 114.2 | WOUDC | 9 |
| 16 | Ha Noi, Vietnam | 21.0 | 105.8 | SHADOZ, (SOWER) | 2 (5) |
| 17 | Hilo, Hawaii, USA | 19.4 | −155.0 | SHADOZ | 31 |
| 18 | Alajuela, Costa Rica | 10.0 | −84.2 | SHADOZ | 2 |
| 19 | Paramaribo, Surinam | 5.8 | −55.2 | SHADOZ | 4 |
| 20 | Tarawa, Kiribati | 1.4 | 172.9 | SOWER | 6 |
| 21 | Biak | −1.2 | 136.1 | SOWER | 4 |
| 22 | Nairobi, Kenya | −1.3 | 36.8 | SHADOZ | 11 |
| 23 | Natal, Brazil | −5.5 | −35.3 | SHADOZ | 15 |
| 24 | Java Observatory, Indonesia | −7.5 | 112.6 | SHADOZ | 6 |
| 25 | Ascension Is. | −8.0 | −14.4 | SHADOZ | 31 |
| 26 | Pago Pago, American Samoa | −14.2 | −170.6 | SHADOZ | 17 |
| 27 | Suva, Fiji | −18.1 | 178.4 | SHADOZ | 6 |
| 28 | La Réunion Is. | −21.1 | 55.5 | SHADOZ | 16 |
| 29 | Broadmeadows | −37.7 | 145.0 | WOUDC | 24 |
| 30 | Macquarie Is. | −54.5 | 158.9 | WOUDC | 1 |
| 31 | Ushuaia | −54.9 | −68.3 | WOUDC | 2 |
| 32 | Marambio | −64.2 | −56.6 | WOUDC | 2 |
| | | | | Total | 601 |

[7] SMILES has three specified detection bands within the submillimeter-wave region: 624.32–625.52 GHz (Band A), 625.12–626.32 GHz (Band B), and 649.12–650.32 GHz (Band C). Since the brightest ozone emission line in the SMILES measurement bands is the line at 625.371242 GHz, Bands A and B were mainly used for ozone retrieval. Also, the spectrometer in Band A could be switched to different combinations of the three bands such as Bands A + B and Bands A + C; this situation is described in detail in *Kikuchi et al.* [2010]. Therefore, we noticed some differences in the SMILES measurement characteristics between Bands A and B and between the two spectrometers. These differences have been extensively investigated for the SMILES ozone product with the conclusion that the difference between the two bands is within 1% for the altitude range 28–64 km and increases at altitude below 28 km, reaching about 9% at 20 km. The difference between the two spectrometers is within 3% for the altitude range of 20–67 km [see *Imai et al.*, 2013]. As we do not know which band or which spectrometer is better, for our comparison with the ozonesondes, we used all the available SMILES ozone profiles. The larger number of coincident measurements also increases the stability of the statistics.

[8] The SMILES ozone product provides ozone concentration as volume mixing ratio with the "retrieval precision" [*Takahashi et al.*, 2010, 2011] at each SMILES altitude level and with related data screening flags for each profile. We refer to the error ratio ($S/S_a$) as that of the retrieval precision ($S$) divided by an a priori error ($S_a$); an example of which is shown in Figure 1 (middle). The retrieval precision stored in the data file is set as a negative value when the error ratio is larger than 50% of the a priori error. As described in *Takahashi et al.* [2010, 2011], the accuracy of the retrieved ozone profiles is worse than 10% below 14 km because of the uncertainty in the water vapor continuum emission and scattering by clouds. Thus, we only considered the altitude range from 16 km for the validation.

[9] The screening flags include information on the convergence status, the validity of the observation altitude range, and the field of view interference with the sun, the moon, and the ISS solar paddle, which are stored in the "status field" as a bit sequence. In this study, to assure the validity of the data, we only use the nonflagged profiles with positive retrieval precision. Subject to this condition, a total of 281,123 ozone profiles are available during the SMILES observation period from 12 October 2009 to 21 April 2010. This is about 94% of all SMILES profiles including the flagged ones.

[10] In the previous validation study of the SMILES v2.1 product based on coincidence statistics with satellite observations [*Imai et al.*, 2013], the comparisons of stratospheric ozone with correlative data show agreements that are generally within 10%. However, there are the following features of the SMILES ozone: (i) data quality is poor below 18 km, especially at lower latitudes and (ii) there is a positive bias of smaller than 8% below ~24 km in the equatorial latitudes. The positive bias is reduced in the v2.3 by improving the retrieval algorithm (see JEM/SMILES L2 Product Guide for further details).

## 2.2. Ozonesonde Measurements

[11] Ozonesonde measurements provide high vertical resolution ozone profiles; an example of an ozonesonde profile is shown in Figure 1 (left). The uncertainty and response time of the ozonesonde measurements are well established from extensive laboratory experiments [*Demuer and Malcorps*, 1984; *Bodeker et al.*, 1998; *Borchi et al.*, 2005; *Kerr et al.*, 1994; *Smit et al.*, 2007; *Thompson et al.*, 2007]. In general, the ozonesonde measurement uncertainty is estimated to be 5%–10% in the stratosphere, increasing at higher altitudes, particularly above 30 km. Thus, we use the altitude below 30 km. The response time, which is usually regarded as the *e*-folding time, is thought to be 20–30 s. In addition, previous examination of ozonesonde measurements suggests that station-to-station biases may exist among stations because of the differences in data-processing technique, electrochemical (KI) solution, and varying hardware [*Johnson et al.*, 2002; *Smit et al.*, 2007; *Thompson et al.*, 2003a, 2003b].

[12] We use ozonesonde profiles worldwide obtained during the SMILES observation period from 12 October 2009 to 21 April 2010. A total of 601 ozonesonde profiles are selected where the profile data is available up to 26 km; see Table 1 for ozonesonde site information including the location and the number of soundings used for the comparison. Figure 2 shows the locations of the ozonesonde sites whose data are used in this study. The ozonesonde observation sites are located mostly in the Northern Hemisphere; the SHADOZ network provides good coverage in the equatorial latitude, and the SOWER data supplements that of the SHADOZ. In section 2.3, the definition of the coincidence criteria will be described in detail.

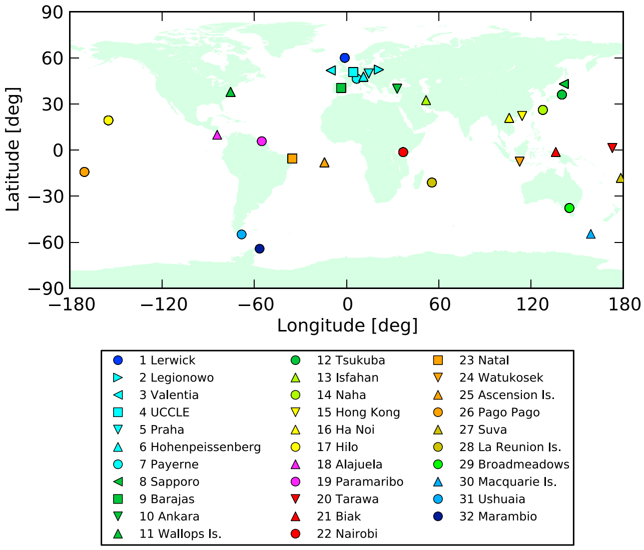


**Figure 2.** Global distribution of ozonesonde observation sites. The symbols for the sites are color coded in each 10° latitude bin. The number in the legend corresponds to the site number in Table 1.

### 2.3. Validation Methodology

[13] To find the coinciding SMILES and ozonesonde events, we used the location and time data of the ozonesonde at the launch and those of the SMILES observations at 30 km of the tangent point altitude. We defined the time and location criteria for the coincidence to be within ± 12 h, ± 2° latitude, and ± 10° longitude. If multiple coincidences for one ozonesonde profile were found, we selected the nearest measurement in space. In the measurement mode of Bands A + B, we used both profiles for the comparison. The criteria for the latitude and longitude are similar to those used in the validation study of Aura/MLS [*Jiang et al.*, 2007]. Though the criterion for the time used by *Jiang et al.* [2007] is on the same (geomagnetic time) day, we did not apply this one in our analysis. According to *Sakazaki et al.* [2013], even in the stratosphere, ozone amounts show diurnal variation; the peak-to-peak difference over the course of a day is ~5% in volume mixing ratio in the lower stratosphere at equatorial latitudes. However, since the SMILES measurements are evenly distributed on local time at each sonde launch site for this comparison study (not shown here), the influence of the sampling bias for the averaged profiles is negligible.

[14] In Table 1, the number of ozonesonde profiles used in this study is listed for each observation station, and Figure 3 shows the temporal and latitudinal distribution of the ozonesonde measurements used for the comparison. As indicated in Table 1 and Figure 3, there are a large number of coinciding events in the northern middle and high latitude. This is because there are many ozonesonde observation sites in Europe (Figure 2) and because the SMILES measurement density is higher at these latitudes [*Kikuchi et al.*, 2010].

[15] For comparisons, the altitude coordinate of the ozonesonde measurement was converted from geopotential height to geometric height to match the SMILES data; thus, the altitude shown in this study is expressed in geometric height. The flagged data points were removed from each altitude level. Then, we smoothed and degraded the high-resolution ozonesonde profiles by convolving the SMILES averaging kernels for the ozone; an example of the averaging kernels is shown in Figure 1 (right).

## 3. Results

[16] Figure 4 shows comparisons between the SMILES ozone and the ozonesonde profiles for four selected sites, Hohenpeissenberg (47.8°N, 11.0°E) at the northern high latitudes, Tsukuba (36.1°N, 140.1°E) at the northern midlatitudes, Natal (5.5°S, 35.3°W) in the equatorial latitudes, and Broadmeadows (37.7°S, 145.0°E) at the southern midlatitudes, where the number of profiles are relatively large. The average difference ($D$) (Figure 4 (middle)) and average relative difference (RD) (Figure 4 (right)) were derived by using reliable data from coincident profiles in the following equations:

$$D = \overline{Q_i - R_i}, \tag{1}$$

$$\mathrm{RD} = D/\left\{\left(\overline{Q_i} + \overline{R_i}\right)/2\right\}, \tag{2}$$

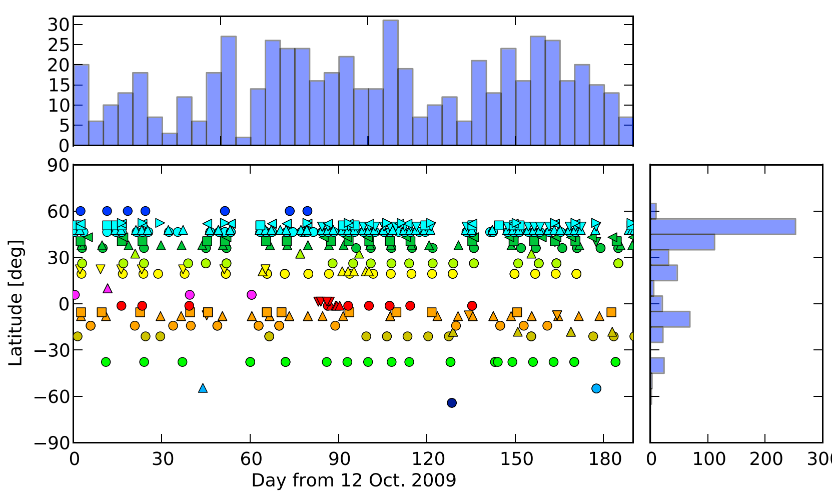


**Figure 3.** (bottom left) Temporal and latitudinal distribution of the coinciding ozonesonde measurements and the number of observations (top) for each 5 day period and (bottom right) for every 10° latitude bins. The symbols and the color in Figure 3 (bottom left) are the same as those in Figure 2.

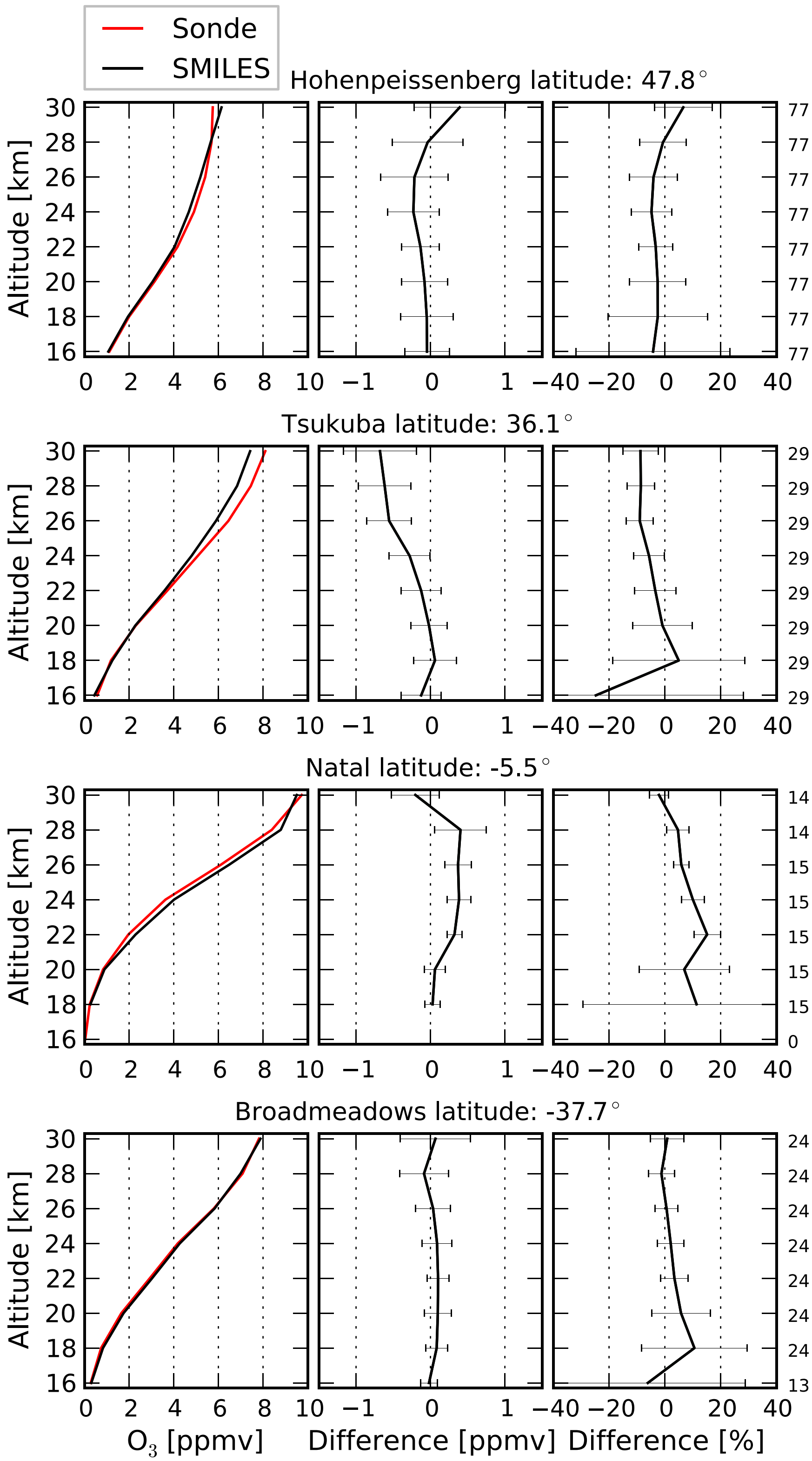


**Figure 4.** Comparisons of (left) average values, (middle) average differences, and (right) average relative differences for selected four ozonesonde sites at four representative latitudes. Relative differences are calculated by equation (2); ozonesonde are subtracted from SMILES. The error bars of average differences and average relative differences are standard deviations. The profile numbers for the statistics are shown on each SMILES altitude level on the right axes.

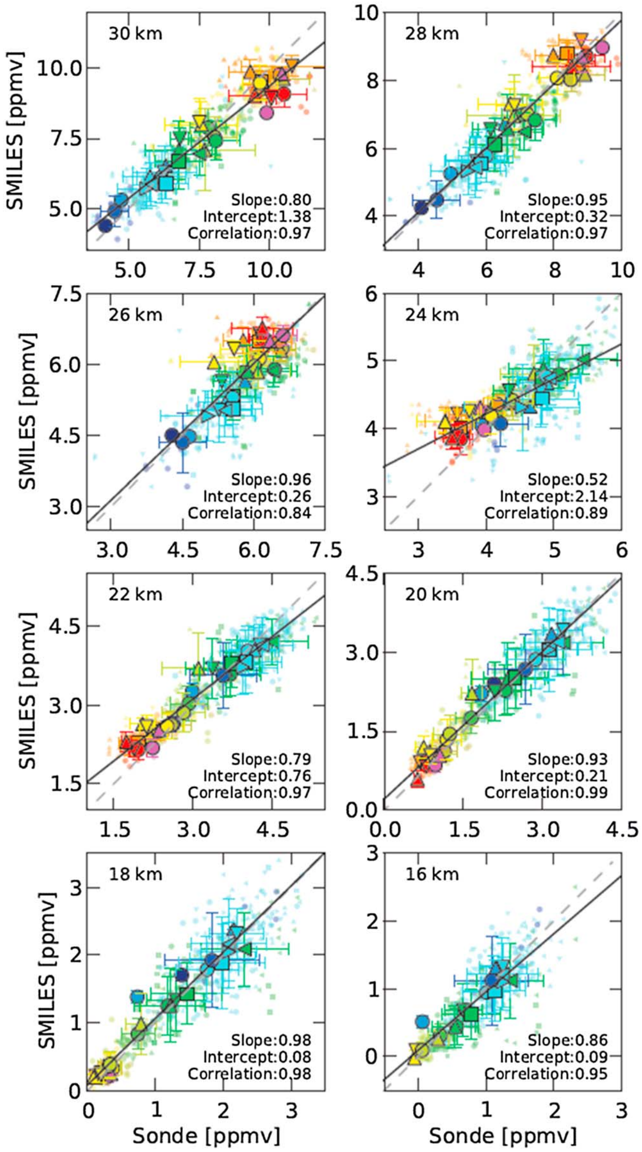


**Figure 5.** Scatterplots of ozonesonde measurements versus SMILES v2.1 ozone measurements for all the coinciding data from 16 to 30 km. The symbols are the same as those in Figure 2, but the colors are semitransparent with no borders. The average values and their standard deviations for each of the stations are also expressed in large opaque symbols with black border, and the error bars represent the standard deviation. The black solid lines are the best fit linear equations to the average values. The slope, intercept, and correlation derived from the best fit linear line are indicated in each figure.

where $Q_i$ and $R_i$ are the $i$th coincident pair of the SMILES and ozonesonde measurement values in volume mixing ratio, respectively, and the over bar denotes the average. The sample number of the ozone profiles, shown in the right of each figure, usually decreases for altitudes below 16 km and above 26 km. The decrease at lower altitudes, in particular at low latitudes, is due to some SMILES ozone profiles with low data quality which were excluded from the analysis. Conversely, the decrease at higher altitudes is due to lack of the ozonesonde measurements.

[17] At Hohenpeissenberg, we see a good agreement between SMILES and ozonesonde measurements within 5% in the altitude range from 16 to 28 km. At Tsukuba, the agreement is reasonable with an order similar to that of Hohenpeissenberg, but a negative bias of the SMILES measurements can be seen above 20 km. This negative bias decreases when the statistics are averaged for all the midlatitude stations (Figure 6). At Natal, the agreement is also reasonable, but we see a relatively large positive bias of the SMILES measurements throughout the height range of 18–26 km. This positive bias is still seen for the comparison using data from all the low-latitude stations (Figure 6) and will be discussed in relation to a potential contribution of the time lag of ozonesonde measurements in section 4. At Broadmeadows, we see a general agreement on an order similar to those of the other three stations.

[18] To extend our analysis for all the coincident profile pairs, in Figure 5, we show scatterplots of ozonesonde versus SMILES v2.3 ozone measurements from 16 to 30 km. In each of the plots, the latitudinal variations of the ozone mixing ratios are indicated by the shades of red in the low latitudes and blue in the high latitudes. As the color pattern switches, the latitudinal gradient is reversed around 24 and 26 km. The relation between the SMILES and ozonesonde measurements is generally good from the viewpoint of the latitudinal variation at fixed heights. The slope is close to one, and the intercept is close to zero at 18 and 20 km, though the two parameters deviate at 16 and 22 km. In particular, the deviation of rather higher values of the SMILES ozone mixing ratio at low latitudes is enhanced at 24 km and

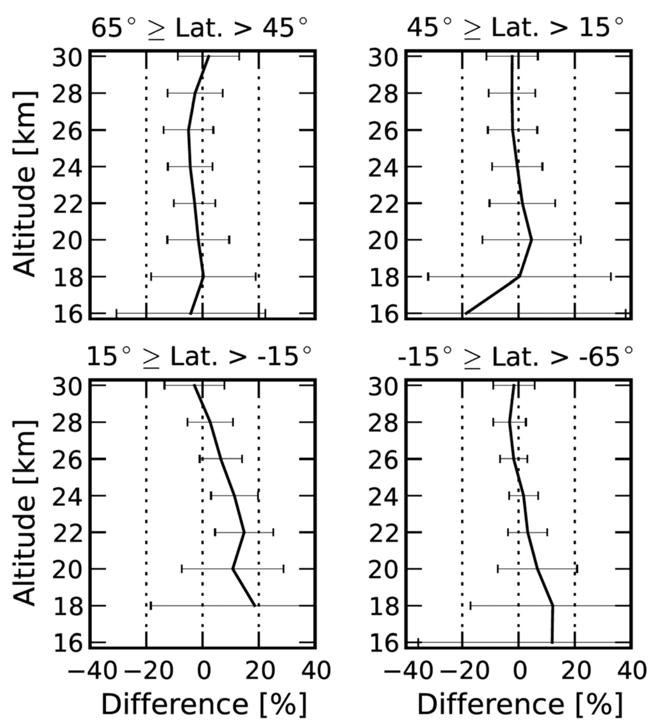


**Figure 6.** The relative differences averaged for the four latitude bands, 65°N–45°N, 45°N–15°N, 15°N–15°S, and 15°S–65°S using all ozonesonde sites. Relative differences are calculated by equation (2); ozonesonde are subtracted from SMILES. The error bars of the average relative differences represent standard deviations.

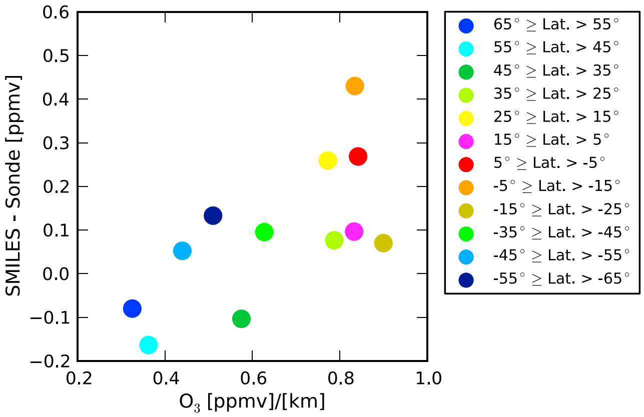


**Figure 7.** Relation between the vertical gradient of ozone at 25 km and the difference between the SMILES and ozonesonde measurements based on the average values for each 10° latitude bin, such as 65°N–55°N, 55°N–45°N, and so on. Red shades represent low latitudes, and blue shades represent high latitudes.

continues at 26 km while the latitudinal gradient is reversed. At 28 and 30 km, the agreement between the SMILES and ozonesonde measurements is resumed. Note the negative values of the SMILES ozone mixing ratio at 16 km and low latitudes even though they are not flagged.

[19] Regarding the differences in the equatorial latitudes, we note that the following sites with the relatively large number of coincidence events show more than 5% difference at 20–26 km (the average ozone values at 20, 22, 24, and 26 km are provided in parentheses; see Table 1 for the number of coincidence events): Nairobi (0.84, 2.0, 3.6, and 6.2 ppmv, SMILES 0.94, 2.1, 3.8, and 6.5 ppmv), Natal (0.84, 2.0, 3.6, and 6.1 ppmv, SMILES 0.90, 2.3, 4.0, and 6.5 ppmv), and Ascension Is. (0.75, 1.8, 3.4, and 5.8 ppmv, SMILES 0.94, 2.3, 4.1, and 6.5 ppmv).

[20] To summarize the results of the comparison between the SMILES and ozonesonde measurements, Figure 6 shows relative differences averaged for the four latitude bands, 65°N–45°N, 45°N–15°N, 15°N–15°S, and 15°S–65°S using all ozonesonde sites; these are basically similar to those in Figure 4 (right). At high northern latitudes, 65°N–45°N, the agreement is within 5% for 16–30 km. At the northern midlatitudes and southern latitudes, the difference is within ~3% for 22–30 km, and below 20 km, it gradually becomes worse. The most notable difference can be seen at the lower latitudes, 15°N–15°S, as the difference increases (~6–19%) with decreasing height below 26 km. Also, note that the difference changes its sign to negative (positive) below (above) the maximum ozone height. We discuss this issue in the next section.

## 4. Discussion—Some Possible Issues in the Ozonesonde System

[21] In the altitude range of 18–26 km at low latitudes, the SMILES ozone values showed a positive bias with respect to the ozonesonde measurements as shown in Figures 5 and 6. This tendency, a larger positive bias in the tropics, can also be seen in the previous validation study of the MLS ozone version 2.2 (hereafter v2.2) [*Jiang et al.*, 2007]. They reported that the comparisons with ozonesonde measurements at low latitudes between 20°S and 20°N show a positive bias with about 10% and 20% at altitudes of 24 km and 18 km, respectively. We confirmed this MLS characteristic by plotting figures similar to Figure 5 but for data from the ozonesondes and MLS v2.2 at 20, 22, 24, and 26 km (Figure A). The figure shows similar large values at the equatorial latitudes for the MLS ozone data.

[22] Conversely, the intersatellite validation study of MLS ozone did not find such a bias in these altitude ranges [*Froidevaux et al.*, 2008]. We confirmed this point by comparing the SMILES v2.3 with MLS v2.2 and found that the SMILES ozone mixing ratios at 20–24 km in the equatorial lower stratosphere show slightly larger values with less than ~7% than the MLS; moreover, they show some variation with latitude, but its size is less than half of the positive bias. For example, the relative differences between the SMILES and MLS measurements at the latitude band of 5°S–5°N are 7.4% at 22 km, 5.3% at 24 km, and 2.8% at 26 km; thus, these are not as large as shown in Figure 6. The detailed comparisons of the SMILES ozone profiles with other satellite measurements are presented by *Imai et al*. [2013].

[23] Before discussing this issue in detail, we show another expression of this positive bias in association with the latitudinal variation. Figure 7 displays the relation between the vertical gradient of ozone mixing ratios and the difference between the SMILES and ozonesonde on the basis of the average values for each 10° latitude bin, such as 65°N–55°N, 55°N–45°N, and so on. The result at 23 km is shown by differentiating values at 22 and 24 km. The red-toned colors present low latitudes and the blue-toned ones present high latitudes in a similar way as in Figure 2. It is clear that the difference becomes larger in lower latitudes. This suggests that the positive bias seen in the equatorial latitude may be related to the rapid change of ozone concentrations in the lower stratosphere.

[24] To explain this bias, we explored some possible issues in the ozonesonde measurement system. One is due to uncertainty in the height determination from a radiosonde with which an ozone sensor is equipped to measure pressure, temperature, and humidity. Recently, using a GPS receiver in the radiosonde system, height information is first derived very precisely and it is converted into pressure information. However, a fair number of ozonesondes are equipped with a radiosonde using a conventional pressure sensor, because of its established interface between an ozone sensor and a radiosonde. During the SOWER campaign in January 2010, they launched ozonesondes with a Vaisala RS80 radiosonde and water vapor sondes with a GPS module simultaneously.

[25] As already pointed out by *Inai et al*. [2009], pressure measurements by RS80 may introduce a positive bias in estimating the geopotential/geometric height about 300 m at 30 km; this is due to a negative bias of about 5% in pressure measurements in the height range from 20–30 km. We confirmed this bias from nine profiles to be 267 m on average in geometric height for those flights during the SOWER campaign. The height offset yields underestimation of ozone mixing ratios where ozone amounts are increasing with increasing height, and the pressure bias has an effect on the calculation of ozone mixing ratio from ozone partial pressure. Figure 8 shows percentage differences, averaged over nine ozonesonde observations during the SOWER campaign, between the two ozone mixing ratio profiles; one is with

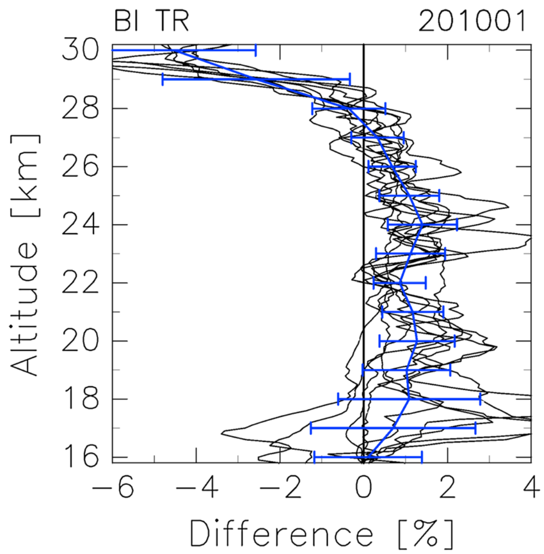


**Figure 8.** Percentage differences in ozone mixing ratio estimated from nine ozonesonde profiles during the SOWER campaign. They are calculated from differences between the two ozone mixing ratio profiles; one is with height information from GPS, and the other is with that based on radiosonde pressure and temperature measurements. One standard deviation is presented by a horizontal bar.

height information from GPS, and the other is with that based on radiosonde pressure and temperature measurements as calculated by *Inai et al*. [2009]. We see about 1% differences or underestimations of ozone mixing ratios for the height range from 20 to 24 km and negative biases less than 5% at 30 km. This issue does not affect the ozone amounts in the lower stratosphere so much, but it is critical above the peak of the ozone partial pressure at around 30 km, though the result may change depending on a radiosonde model.

[26] Another possible issue is related to the time lag in ozonesonde measurements. To investigate this bias at low latitudes, we took a close look at ascending and descending ozonesonde profiles. Figure 9 shows an example of the ozonesonde measurement conducted during the SOWER campaign at Tarawa, the republic of Kiribati (1.4°N and 172.9°E), launched at 10:41 UTC on 11 January 2010. The full height range is plotted in Figure 9 (left), and the limited height range for 20–25 km is plotted in Figure 9 (right). During this balloon flight, the ascent and descent data were recorded as shown by the thin blue line for the original ascending profile and the thin red line for the original descending profile in Figure 9. Since the ozone concentration is determined through an electric current measurement, which is proportional to the ozone number density, we plotted these lines in units of ozone partial pressure. Note that a typical ascent rate is 4–6 m/s while a descent rate (after balloon burst but with use of a parachute) can be approximately 4 times faster around the height range of 20–25 km.

[27] Before performing a detailed analysis, we look at ascending and descending temperature profiles recorded during the flight and found that there exist some differences between the two. They should be almost zero since the response time of the temperature sensor is negligible; this is probably due to hysteresis of the pressure sensor, resulting in miscalculation of altitude information during the descent. Therefore, we first adjust the altitude coordinate so as to minimize the difference between the ascending and descending temperature profiles for the altitude range of 20 to 25 km. The height difference for this case is 0.18 km, and an average of the difference is $0.22 \pm 0.08$ km. The thick green line is also for the descending profile of ozone, but the altitude is shifted as mentioned

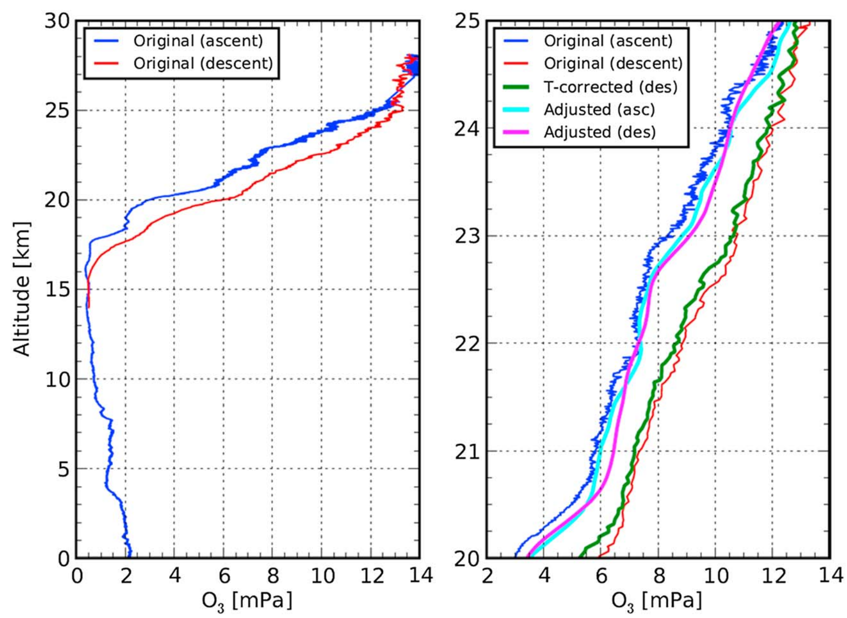


**Figure 9.** Example of the ozonesonde measurement conducted during the SOWER campaign at Tarawa, the Republic of Kiribati (1.4°N and 172.9°E), launched at 2:36 UTC on 6 January 2010. (left) Full altitude range and (right) 20–25 km altitude range. Original ascending (thin blue) and descending (thin red), T-corrected (thick green), adjusted ascending (thick magenta), and descending (thick cyan lines) profiles, respectively.

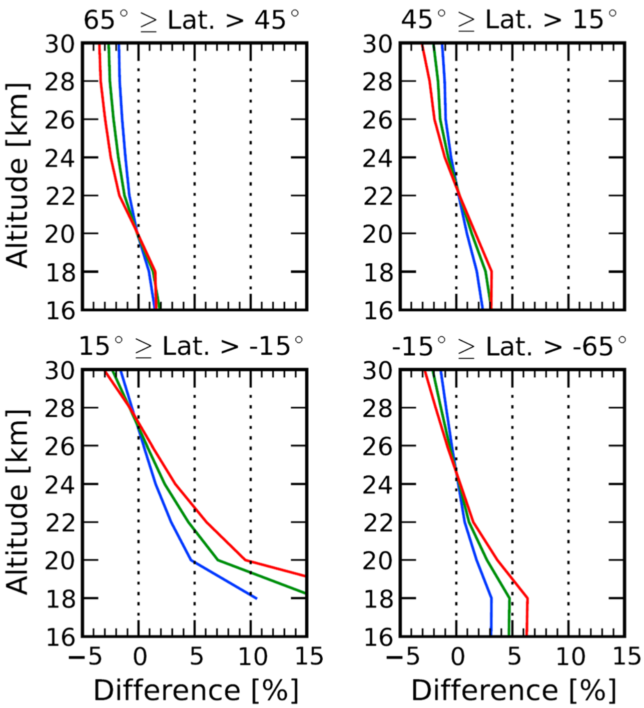


**Figure 10.** Correction amount to original mixing ratios averaged over the all ozonesonde profiles for the four latitude bands, 65°N–45°N, 45°N–15°N, 15°N–15°S, and 15°S–65°S with response times of 20 s (blue), 25 s (green), and 30 s (red).

above, and it is referred to as a T-corrected profile hereafter. Since the two lines of the ascending and the T-corrected descending profiles still do not overlap but differ considerably, it is suspected that the ozonesonde measurements could be delayed because of the sensor response time.

[28] To estimate the effect of the time delay, we applied a time lag correction which was proposed by *Miloshevich et al*. [2004] for humidity measurements of radiosondes. This is based on an assumption that the sensor responds approximately exponentially to a change in measurement value as described by the common "growth law equation."

$$\frac{dX_m}{dt} = k(X_a - X_m), \tag{3}$$

where $X_m$ is the measured ozone concentration, $X_a$ is the ambient ozone concentration, and $k$ is a constant. This equation can be solved for a step change in $X_a$ at time $t_s$ to give the measured ozone concentration as a function of time:

$$X_m(t) = X_a - (X_a - X_m(t_0))e^{-\Delta t/\tau}, \tag{4}$$

where $\tau = 1/k$ is the response time which is regarded as the $e$-folding time. We adjusted the response time that would minimize the difference between the ascending and T-corrected descending profiles for the altitude range of 20 to 25 km after smoothing the two profiles, assuming that the response time is constant during this altitude range.

[29] In Figure 9, the adjusted two profiles are plotted for the case when the difference between the two is minimized. We determined the response time as 32 s for this case. The SOWER campaign produced profiles with both ascending and descending profiles, which could be used for estimating the response time as indicated above. The response times we calculated for the six profiles in January 2010 ranged from 23 to 32 s with an average of about $28 \pm 4$ s, while an average of the response time of ground measurements is $25 \pm 3$ s. During the SOWER campaigns, the response time before the launch was measured in a conventional way as follows: First, air containing 5 μA level ozone is drawn into the sensor cell for 10 min; after the 10 min, the air with ozone is quickly replaced with ozone-free air; while ozone-free air is drawn, the time for 4.0 μA down to 1.5 μA is measured as the response time ($e = 2.72 \approx 4.0/1.5$). Therefore, the response time here roughly corresponds to the $e$-folding time for a stepwise change. This result suggests that the time lag correction method could be suitable to the ozonesonde profiles.

[30] To quantitatively estimate the effect of the response time, we applied the correction to all the ozonesonde profiles. Figure 10 shows the differences of corrected to original mixing ratios averaged over all the ozonesonde profiles for the four latitude bands similar to those in Figure 6. Since we could not assume that the above estimation for the response time could be applied to all the latitude bands and stations, we calculated profiles for three cases with response times of 20, 30, and 40 s. As we applied the correction to the ozone partial pressure measurement, it did not change at the altitude with the maximum ozone partial pressure. The tendency was positive and negative below and above the maximum height, respectively. The differences between the originals and the corrected profiles with response times of 30 s were mostly within ~5%. At the lower latitudes, however, they were over 7% with a response time of 30 s and almost 10% with a response time of 40 s at 20 km. The correction also influences the estimation of column ozone about 2.3 Dobson unit (DU) for 20–25 km in the equatorial latitudes with a response time of 30 s; this reaches 3.0 DU if we extend the height range for 18–25 km.

[31] With these corrections, the agreement between the SMILES and ozonesonde measurements is improved, particularly at low latitudes, although it is not perfect and there may still be some problems in the SMILES data. Moreover, the slight larger values of the ozonesonde measurements at higher altitudes may also be explained by the response time above the maximum of the ozone partial pressure. Thus, it could be understood that the altitude where the bias changes its sign in Figure 10 almost corresponds to that seen in Figure 6 at each latitude band. In addition, we have seen slight negative biases in the lower stratosphere resulting from pressure measurement errors of radiosondes, though they may depend on a specific model of the radiosondes. All these results suggest that the conventional ozonesonde system may produce a negative bias particularly at low latitudes in the lower stratosphere.

## 5. Summary

[32] This study presented results from the comparison of SMILES v2.3 ozone profiles and ozonesonde measurements in the lower stratosphere. To assess the SMILES v2.3 ozone data quality in the altitude range from 16 to 30 km, a total of 601 ozonesonde profiles from 32 ozonesonde stations were compared with coinciding SMILES ozone profiles. The ozonesonde data used in this study were from the WOUDC, the SHADOZ network, and the SOWER project campaign.

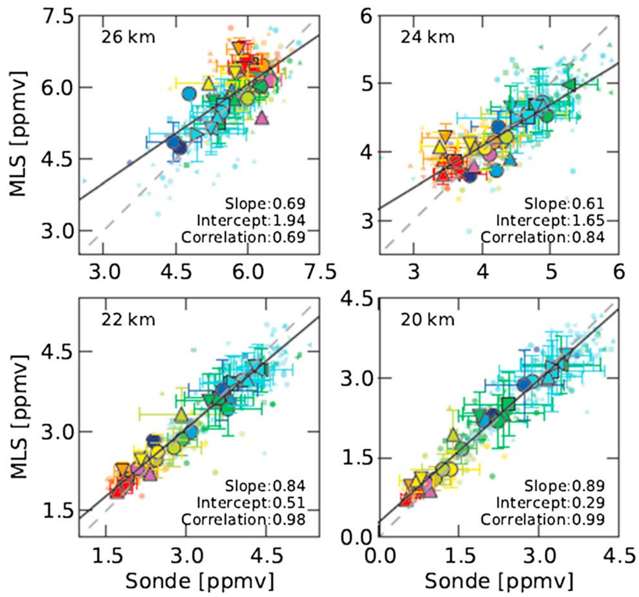


**Figure A1.** Same as Figure 5 but for ozonesonde and MLS v2.2 at 24 and 26 km. The symbols and the color in the panels are the same as those in Figure 2.

[33] The agreement between SMILES and ozonesonde measurements is good within 5%–7% for 18–30 km at middle and high latitudes but worsens below 18 km. At low latitudes, however, the SMILES ozone data show larger values (~6–15% for 20–26 km) than those at middle and high latitudes. This feature can also be seen in the ozone validation paper of the Aura Microwave Limb Sounder [*Jiang et al.*, 2007].

[34] To explain this bias at low latitudes, we explored some possible issues in the ozonesonde measurement system. As already pointed out by *Inai et al.* [2009], pressure measurements by conventional radiosondes may introduce an error in estimating the geopotential/geometric height. We found in the radiosonde system, Vaisala RS80, with a conventional pressure sensor that it overestimates height information about 300 m at 30 km because of the pressure bias about 5%. This height offset may yield about 2% underestimation of ozone mixing ratios for the height range 20–24 km and overestimation up to 6% at 30 km. This issue does not affect the ozone amounts in the lower stratosphere so much, but it is critical above the peak of the ozone partial pressure, though the effect depends on a radiosonde model.

[35] We also examined an issue of the ozonesonde's response time. To investigate this bias at low latitudes, we took a close look at ascending and descending ozonesonde profiles taken during the SOWER campaign. Since the differences between the ascending and descending ozonesonde profiles were evident, even after the height adjustment estimated from the ascending and descending temperature profiles, we applied a time lag correction which was proposed by *Miloshevich et al.* [2004] for humidity measurements of radiosondes. We estimated the response time that would minimize the difference between the ascending and corrected descending ozone profiles. The response time of our estimation was 28 s on average which is close to the ground measurements of 25 s and also within the value for electrochemical concentration cell sonde (20–30 s) [e.g., *Smit et al.*, 2007]. These results suggest the use of the time lag correction method would be appropriate.

[36] By applying this correction to the original profiles, we found a negative bias of the ozonesonde measurement more than 7% at 20 km in the equatorial latitude where the vertical gradient of ozone is steep. Also additionally, we may put 2% negative bias due to the bias from the radiosonde pressure sensor. The agreement becomes much better between the SMILES and the corrected ozonesonde profiles, but it is not perfect, and there may be other problems with the SMILES data. From these results, we suggest that the response time would provide a negative bias particularly at low latitudes in the lower stratosphere, though it is an order of the ozonesonde measurement uncertainty.

## Appendix A

[37] Figure A1 shows scatter plots of ozonesonde versus MLS v2.2 ozone measurements at 20, 22, 24 and 26 km. We used the same ozonesonde profiles coinciding with SMILES but reapplied the same criteria to find the coinciding MLS and ozonesonde events. The figure shows the similar tendency of a larger positive bias in the tropics as already seen in Figure 5.

[38] **Acknowledgments.** We would like to thank the project teams of following agencies for their support and for providing us the ozone profiles immediately after the soundings were taken: Naha, Sapporo, Tsukuba (the Japan Meteorological Agency), Ascension Island, Natal, Wallops Island (the NASA Goddard Space Flight Center Wallops Flight Facility), La Réunion (University of La Réunion), Lindenberg (the Deutscher Wetterdienst), and Hanoi (the Japan Agency for Marine-Earth Science and Technology, and National Hydro and Meteorological Service and Aero-Meteorological Observatory, Vietnam). We would also like to express our gratitude to the WOUDC, SHADOZ, and SOWER teams for providing the ozonesonde data used in this study. This research made use of data obtained from Data Archives and Transmission System (DARTS), provided by Center for Science-satellite Operation and Data Archive (C-SODA) at ISAS/JAXA. This study was partially supported by the Japanese Ministry of Education, Culture, Sports, Science, and Technology (MEXT) through Grants-in-Aid for Scientific Research (22310010) and the ISS Science Project Office of ISAS/JAXA.

## References

Bodeker, G. E., I. S. Boyd, and W. A. Matthews (1998), Trends and variability in vertical ozone and temperature profiles measured by ozonesondes at Lauder, New Zealand: 1986–1996, *J. Geophys. Res.*, *103*(D22), 28,661–28,681, doi:10.1029/98JD02581.

Borchi, F., J.-P. Pommereau, A. Garnier, and M. Pinharanda (2005), Evaluation of SHADOZ sondes, HALOE and SAGE II ozone profiles at the tropics from SAOZ UV-vis remote measurements onboard long duration balloons, *Atmos. Chem. Phys.*, *5*, 1381–1397, doi:10.5194/acp-5-1381-2005.

Demuer, D., and H. Malcorps (1984), The frequency-response of an electrochemical ozone sonde and its application to the deconvolution of ozone profiles, *J. Geophys. Res.*, *89*(ND1), 1361–1372.

Dupuy, E., et al. (2009), Validation of ozone measurements from the Atmospheric Chemistry Experiment (ACE), *Atmos. Chem. Phys.*, *9*, 287–343, doi:10.5194/acp-9-287-2009.

Fioletov, V. E., et al. (2008), Performance of the ground-based total ozone network assessed using satellite data, *J. Geophys. Res.*, *113*, D14313, doi:10.1029/2008JD009809.

Froidevaux, L., et al. (2008), Validation of Aura Microwave Limb Sounder stratospheric ozone measurements, *J. Geophys. Res.*, *113*, D15S20, doi:10.1029/2007JD008771.

Hasebe, F., M. Fujiwara, N. Nishi, M. Shiotani, H. Vömel, S. Oltmans, H. Takashima, S. Saraspriya, N. Komala, and Y. Inai (2007), In situ observations of dehydrated air parcels advected horizontally in the Tropical Tropopause Layer of the western Pacific, *Atmos. Chem. Phys.*, *7*, 803–813.

Hegglin, M. I., C. D. Boone, G. L. Boone, T. G. Manney, K. A. Shepherd, P. F. Walker, W. H. Bernath, P. H. Daffer, and C. Schiller (2008), Validation of ACE-FTS satellite data in the upper troposphere/lower stratosphere (UTLS) using non-coincident measurements, *Atmos. Chem. Phys.*, *8*(6), 1483–1499, doi:10.5194/acp-8-1483-2008.

Hoogen, R., V. V. Rozanov, K. Bramstedt, K.-U. Eichmann, M. Weber, and J. P. Burrows (1999), $O_3$ profiles from GOME satellite data - I: Comparison with ozonesonde measurements, *Phys. Chem. Earth Part C*, *24*(5), 447–452, doi:10.1016/S1464-1917(99)00071-9.

Imai, K., et al. (2013), Validation of ozone data from the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES), *J. Geophys. Res. Atmos.*, *118*, 5750–5769, doi:10.1002/jgrd.50434.

Inai, Y., F. Hasebe, K. Shimizu, and M. Fujiwara (2009), Correction of radiosonde pressure and temperature measurements using simultaneous GPS height data, *SOLA*, *5*, 109–112, doi:10.2151/sola.2009 028.

JEM/SMILES L2 Products Guide, [Available at http://smiles.tksc.jaxa.jp/l2data/pdf/L2dataGuide_130703.pdf.]

Jiang, Y. B., et al. (2007), Validation of Aura Microwave Limb Sounder Ozone by ozonesonde and lidar measurements, *J. Geophys. Res.*, *112*, D24S34, doi:10.1029/2007JD008776.

Johnson, B. J., S. J. Oltmans, H. Vömel, H. G. J. Smit, T. Deshler, and C. Kröger (2002), Electrochemical concentration cell (ECC) ozonesonde pump efficiency measurements and tests on the sensitivity to ozone of buffered and unbuffered ECC sensor cathode solutions, *J. Geophys. Res.*, *107*(D19), 4393, doi:10.1029/2001JD000557.

Kerr, J. B., et al. (1994), The 1991 WMO international ozonesonde intercomparison at Vanscoy, Canada, *Atmos. Ocean*, *32*, 685–716, doi:10.1080/07055900.1994.9649518.

Kikuchi K., et al. (2010), Overview and early results of the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES), *J. Geophys. Res.*, *115*, D23306, doi:10.1029/2010JD014379.

Miloshevich, L. M., A. Paukkunen, H. Vömel, and S. J. Oltmans (2004), Development and validation of a time-lag correction for Vaisala radiosonde humidity measurements, *J. Atmos. Oceanic Tech.*, *21*, 1305–1327.

Mitsuda, C., et al. (2011), Current status of level 2 product of Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES), in *Sensors, Systems, and Next-Generation Satellites XV*, vol. 8176, edited by R. Meynart, S. P. Neeck, and H. Shimoda, pp. 6 , Proc. SPIE, Bellingham, Wash.

Randall, C. E., et al. (2003), Validation of POAM III ozone: Comparisons with ozonesonde and satellite data, *J. Geophys. Res.*, *108*(D12), 4367, doi:10.1029/2002JD002944.

Sakazaki, T., et al. (2013), Diurnal ozone variations in the stratosphere revealed in observations from the Superconducting Submillimeter-Wave Limb-Emission Sounder (SMILES) onboard the International Space Station (ISS), *J. Geophys. Res. Atmospheres*, *118*, 2991–3006, doi:10.1002/jgrd.50220.

Smit, H. G. J., et al. (2007), Assessment of the performance of ECC-ozonesondes under quasi-flight conditions in the environmental simulation chamber: Insights from the Juelich Ozone Sonde Intercomparison Experiment (JOSIE), *J. Geophys. Res.*, *112*, D19306, doi:10.1029/2006JD007308.

Takahashi, C., S. Ochiai, and M. Suzuki (2010), Operational retrieval algorithms for JEM/SMILES level 2 data processing system, *J. Quant. Spectrosc. Radiat. Transfer*, *111*, 160–173.

Takahashi, C., et al. (2011), Capability for ozone high-precision retrieval on JEM/SMILES observation, *Adv. Space Res.*, *48*, 1076–1085.

Thompson, A. M., et al. (2003a), Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998–2000 tropical ozone climatology. 1. Comparison with Total Ozone Mapping Spectrometer (TOMS) and ground-based measurements, *J. Geophys. Res.*, *108*(D2), 8238, doi:10.1029/2001JD000967.

Thompson, A. M., et al. (2003b), Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998–2000 tropical ozone climatology. 2. Tropospheric variability and the zonal wave-one, *J. Geophys. Res.*, *108*(D2), 8241, doi:10.1029/2002JD002241.

Thompson, A. M., J. C. Witte, H. G. J. Smit, S. J. Oltmans, B. J. Johnson, V. W. J. H. Kirchhoff, and F. J. Schmidlin (2007), Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998–2004 tropical ozone climatology. 3. Instrumentation, station-to-station variability, and evaluation with simulated flight profiles, *J. Geophys. Res.*, *112*, D03304, doi:10.1029/2005JD007042.

Tripathi, O., et al. (2005), High resolution simulation of recent Arctic and Antarctic stratospheric chemical ozone loss compared to observations, *J. Atmos. Chem.*, *55*, 205–226, doi:10.1007/s10874-006-9028-8.

**Erratum**

In the originally published version of this article, several instances of text were incorrectly typeset. The following have since been corrected in the html, and are documented here:

In paragraph 34, the sentence "This height offset may yield about 2%" should read "This height offset may yield about 1%".
In paragraph 34, the sentence "overestimation up to 6%" should read "overestimation up to 5%".
In Figure 5 caption, "SMILES v2.1" should read "SMILES v2.3".
Figure 8 should appear as follow: