

# Direct-Sun column ozone retrieval by the ultraviolet multifilter rotating shadow-band radiometer and comparison with those from Brewer and Dobson spectrophotometers

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A methodology for direct-Sun ozone retrieval using the ultraviolet multifilter rotating shadow-band radiometer (UV-MFRSR) is presented. Total vertical column ozone was retrieved in three stations: Mauna Loa, Hawaii, in the U.S., and Regina, Saskatchewan, and Toronto, Ontario, in Canada, from direct solar irradiances of the UV-MFRSR at 325-, 305-, 332-, and 311-nm channels (2-nm FWHM). The total uncertainty of ozone retrievals in this study is  $\pm 2.0\%$ . For Mauna Loa the mean ratios of the UV-MFRSR column ozone retrievals to the collocated Dobson and Brewer were 0.998 and 0.986 between May and September of 1999. The mean ratio of UV-MFRSR retrievals to the collocated Brewer retrievals was 1.012 in Toronto between April and August of 1999, and the mean ratio of retrievals of the UV-MFRSR to the collocated Brewer was 0.988 in Regina between June and September of 1999. Total vertical column ozone values for solar zenith angles of  $>70^\circ$  were not considered, because of the signal-to-noise ratio and the angular response of the instruments, and were not used in the evaluation. The advantages of total vertical column ozone retrieval using UV-MFRSR include relatively low cost, computer-controlled operation, automated calibration stability checks, and minimal maintenance. It allows for the real-time measurement of total vertical column ozone. The UV-MFRSR is being used at 28 sites across the United States and 2 sites in Canada that form the U.S. Department of Agriculture UV-B Radiation Monitoring and Research Program. This constitutes a unique network of total vertical column ozone measurement.

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## 1. Introduction

Both observations and predictions suggest the tendency of increased solar ultraviolet (UV) radiation, resulting from stratospheric ozone decreases.<sup>1-4</sup> Thus it becomes increasingly important to monitor the variation of ozone over large areas of the Earth's surface. Ground-based ozone retrievals provide finer spatial and temporal resolution compared with satellite data<sup>5</sup> and also *in situ* calibration. The spec-

tral measurements of the direct component of solar UV radiation are possible in the observational programs of various UV monitoring networks. Dobson and Brewer spectrophotometers have been used to estimate column ozone for more than 60 (Ref. 6) and 25 yr,<sup>1</sup> respectively. It is common to use four or more wavelengths to account for the effects of aerosols and sulfur dioxide. The U.S. Department of Agriculture (USDA) UV-B Radiation Monitoring and Research Program<sup>7</sup> began installing the ultraviolet multifilter rotating shadow-band radiometer (UV-MFRSR, Yankee Environmental Systems, Turners Falls, Massachusetts) in 1995 and now has 28 sites across the United States as well as 2 sites in Canada.

This paper describes a method for retrieving total vertical column ozone (called column ozone hereafter) by use of UV-MFRSR direct-Sun irradiances. Section 2 provides an experimental description of the three spectral instruments used in this study and outlines an algorithm for determining column ozone from the UV-MFRSR measurements. Section 3 pre-

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sents a comparison of UV-MFRSR ozone retrievals from Mauna Loa, Hawaii, with those from a collocated Dobson and a collocated Brewer and UV-MFRSR ozone retrievals at Regina, Saskatchewan, and Toronto, Ontario, in Canada with those from collocated Brewer spectrophotometers. Section 4 covers sources of the uncertainty of column ozone retrievals, and, finally, Section 5 provides conclusions.

## 2. Experimental Description

### A. Introduction

Total column ozone values were retrieved for 4 months (May–September 1999) with the UV-MFRSR and the Dobson and the Brewer spectrophotometers at Mauna Loa, Hawaii (19.539 °N, 155.578 °W) and for four months (April–August 1999) and three months (June–September 1999) with data from the UV-MFRSR and the Brewer spectrophotometers at Toronto, Ontario (43.78 °N, 79.47 °W), and Regina, Saskatchewan (50.197 °N, 104.7 °W). Daily average values of the total column ozone were used in this study. Data for solar zenith angles (SZAs) of >70° were not used in the evaluation because of the low signal-to-noise ratio and the angular response of the UV-MFRSR instruments.

### B. Instrumentation

The UV-MFRSR<sup>7</sup> is a seven-channel UV version of the visible multifilter rotating shadow-band radiometer described by Harrison *et al.*<sup>8</sup> This instrument uses 2-nm nominal FWHM bandwidth ion-assisted-deposition filters at 300-, 305-, 311-, 317-, 325-, 332-, and 368-nm nominal center wavelengths to measure total horizontal and diffuse horizontal irradiances. There is filter-to-filter variation in this nominal wavelength center ( $\pm 0.5$  nm), and each is characterized to an accuracy of  $\pm 0.02$  nm as described in Subsection 2.C. The direct beam is obtained by subtraction of the diffuse horizontal from the total horizontal irradiance and includes the corrections described by Harrison *et al.*<sup>8</sup> The measurement is completed in less than 5 s at all wavelengths. All three components are recorded every 20 s and averaged to 3-min intervals. Data from each day under all sky conditions were used during study periods. Only the direct-Sun irradiances were used to retrieve the total column ozone in this study. Since we average the direct beam measurement every 3 min, there is always enough data with sufficient direct beam signal some time during every day to allow for retrieval even during cloudy days.

Both Brewer and Dobson spectrophotometers used in this study made direct-Sun total ozone measurements at the selected stations. The Brewer spectrophotometer determined the total column ozone by measuring the direct beam through a sloping side quartz window, using a prism that points directly to the Sun.<sup>9–11</sup> The Brewer measures routinely at 306.3, 310.1, 313.5, 316.8, and 320.0 nm with a FWHM resolution of 0.56 nm. The total column ozone is calculated on the basis of relative intensities

at these wavelengths with the Bass and Paur<sup>12</sup> ozone absorption coefficients at a constant temperature of 228.3 K. The retrieval accuracy of a well-maintained Brewer is approximately  $\pm 1\%$ , and the precision is better than  $\pm 1\%$  (Ref. 13).

For the Dobson ozone spectrophotometer the total column ozone was determined by measurement of the relative intensities at pairs of wavelengths in the UV region. The pairs of wavelengths typically refer to an A-wavelength pair, 305.5 and 325.4 nm, and a D-wavelength pair, 317.6 and 339.8 nm.<sup>14</sup> The wavelength pairs are chosen from one wavelength that is relatively unaffected by ozone absorption and another that is strongly absorbed. The reproducibility for direct-Sun ozone retrievals in Dobson is approximately  $\pm 1\%$ , and the absolute accuracy is estimated as  $\pm 3\%$ .<sup>5</sup> As with the Brewer spectrophotometer we calculate the total column ozone, using the Bass and Paur<sup>12</sup> absorption coefficients, but both the Dobson and our method use the ozone-weighted mean temperature of 227 K (Ref. 15) in the calculation.

### C. Direct-Sun Ozone Retrieval Using the UV-MFRSR

Attenuation of direct-solar irradiance through the atmosphere can be described by the Beer–Lambert law.<sup>16</sup> This law was applied for constructing the method of UV-MFRSR ozone retrieval from direct normal irradiances. The theory assumes that the measured intensity of direct-solar irradiance  $I_\lambda$  at the Earth's surface, for some narrow interval about a wavelength  $\lambda$ , can be approximated as

$$I_\lambda = I_{0\lambda} \exp[-\alpha_\lambda \mu \Omega - \beta_\lambda m(P/P_0) - \delta_\lambda \sec Z], \quad (1)$$

with the following parameters:

- $I_\lambda$  direct normal irradiance at the ground at wavelength  $\lambda$ , in  $\text{W}/\text{m}^2/\text{nm}$ ;
- $I_{0\lambda}$  extraterrestrial irradiance, in  $\text{W}/\text{m}^2/\text{nm}$ ;
- $\alpha_\lambda$  ozone absorption coefficient (base  $e$ ) at wavelength  $\lambda$ , in  $\text{cm}^{-1}$ ;
- $\mu$  ratio of the actual and the vertical paths of solar radiation through the ozone layer;
- $\Omega$  total column ozone expressed in Dobson units (1 DU =  $10^{-3}$ -cm pure ozone at standard temperature and pressure);
- $\beta_\lambda$  Rayleigh scattering coefficient (molecular optical depth) at wavelength  $\lambda$ ;
- $m$  air mass corresponding to solar zenith at the time of the direct normal irradiance measurement;
- $P$  observed station pressure, in millibars;
- $P_0$  mean sea level pressure, in millibars;
- $\delta_\lambda$  scattering coefficient (optical depth) of aerosol particles at wavelength  $\lambda$ ;
- $Z$  SZA, in degrees.

For the uncalibrated voltages, which are proportional to  $I$ , measured by the detector the equation is

$$V_\lambda = V_{0\lambda} \exp[-\alpha_\lambda \mu \Omega - \beta_\lambda m(P/P_0) - \delta_\lambda \sec Z], \quad (2)$$

where  $V_\lambda$  is the measured voltage at the ground at wavelength  $\lambda$ , in millivolts, and  $V_{0\lambda}$  is the extraterrestrial voltage intercept at zero air mass, in millivolts.

The direct-Sun total column ozone retrievals are made with the UV-MFRSR by measurement of the relative intensities of selected pairs of UV wavelengths, as with the Dobson spectrophotometer.<sup>6</sup> The first pair used in this study consists of 305- and 325-nm wavelengths. Light from both wavelengths is attenuated owing to scattering by air molecules and dust particles in passing through the atmosphere to the instrument. Construction of mathematical equations generating the total column ozone from direct observation of the Sun were developed by Dobson<sup>14</sup> and summarized by Komhyr.<sup>6</sup> The total column ozone expressed in Dobson units made from a single pair wavelength ( $\lambda$  and  $\lambda'$ ) is

$$\Omega = \frac{N - (\beta - \beta')m(P/P_0) - (\delta - \delta')\sec Z}{(\alpha - \alpha')\mu} \times 10^3, \quad (3)$$

where

$$N = \ln \frac{V_{0\lambda}}{V_{0\lambda'}} - \ln \frac{V_\lambda}{V_{\lambda'}}.$$

In this study the double-pair wavelengths, 305 and 325 nm and 311 and 332 nm, approximately correspond to the *A* and the *C* pairs for the Dobson. Using the double-pair wavelengths allows for better correction for aerosol scattering. Since the wavelength difference of both pairs is nearly equal, and aerosol extinction varies little over the 305–332-nm wavelength range, the assumption is made that  $(\delta - \delta')_1 - (\delta - \delta')_2 = 0$ , which is equivalent to assuming that  $\delta$  has close to a linear dependence on wavelength. The bias in the ozone retrievals in this study with this assumption is at most  $\pm 0.44\%$ . The absorption by ozone, therefore, is the major factor on the relative intensities of the double-wavelength pairs. The total column ozone  $\Omega$  was computed as

$$\Omega = \frac{N_1 - N_2 - [(\beta - \beta')_1 - (\beta - \beta')_2]m(P/P_0)}{[(\alpha - \alpha')_1 - (\alpha - \alpha')_2]\mu} \times 10^3, \quad (4)$$

where

$$N_1 = \ln \frac{V_{0(305 \text{ nm})}}{V_{0(325 \text{ nm})}'} - \ln \frac{V_{(305 \text{ nm})}}{V_{(325 \text{ nm})}'},$$

$$N_2 = \ln \frac{V_{0(311 \text{ nm})}}{V_{0(332 \text{ nm})}'} - \ln \frac{V_{(311 \text{ nm})}}{V_{(332 \text{ nm})}'},$$

$$\mu = \frac{R + h}{[(R + h)^2 - (R + r)^2 \sin^2 Z]^{1/2}},$$

where  $R$  is the mean earth radius (6371.229 km);  $r$  is the height of the station above mean sea level, in kilometers;  $h$  is the height of the ozone layer above mean sea level at station location (20 km); and  $Z$  is the SZA.

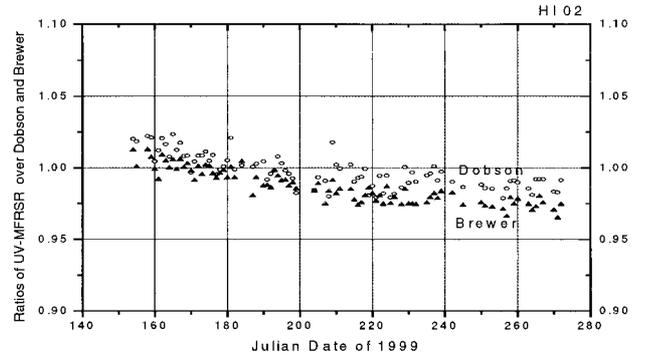


Fig. 1. Ratios of the UV-MFRSR column ozone to Dobson and Brewer between May and September, 1999, at Mauna Loa, Hawaii. The mean ratio of the UV-MFRSR divided by the Dobson was 0.998 with the standard deviation 0.012 and by the Brewer was 0.986 with the standard deviation 0.011.

Total column ozone was retrieved for five 3-min periods with the highest irradiances at 311 nm and SZA less than  $70^\circ$  for days during the study period with at least five such irradiances greater than  $0.0025 \text{ W/m}^2/\text{nm}$ . Total column ozone was determined by the average of these five retrievals. The spectral response function (SRF) of each of the four channels was measured by the Central Ultraviolet Calibration Facility (CUCF)<sup>17</sup> to an accuracy of  $\pm 0.02 \text{ nm}$ . The ozone absorption coefficients of Bass and Paur<sup>12</sup> at a constant temperature of 227.0 K were used.

### 3. Comparison Results of Direct-Sun Column Ozone Retrievals

Total column ozone was retrieved from UV-MFRSR measurements for each day from Julian dates 150–270, 1999, in Mauna Loa, Hawaii, by unit 386; from Julian dates 120–240, 1999, in Toronto, Ontario, by unit 387; and from Julian dates 160–250, 1999, in Regina, Saskatchewan, by unit 287. The UV-MFRSR's SRFs were measured at the CUCF in Boulder, Colorado, on Julian date 134, 1999, for unit 386; Julian date 42, 1999, for unit 387; and Julian date 28, 1999, for unit 287. The direct-Sun ozone retrievals were compared with those from collocated Brewer and Dobson spectrophotometers in the three locations.

The results of these comparisons are shown in Figs. 1, 2, and 3, where the ratio of UV-MFRSR to Dobson and to Brewer ozone measurements in Mauna Loa and the ratio of UV-MFRSR to Brewer in Toronto and Regina, respectively, are plotted as a function of time. Table 1 provides the summary of ozone retrieval comparison results. For Mauna Loa the mean ratio of the UV-MFRSR column ozone retrievals to the Dobson and the Brewer was 0.998 with a standard deviation of 0.012 (3.2 DU) and 0.986 with a standard deviation of 0.011 (2.9 DU). The mean ratio UV-MFRSR retrievals to the Brewer retrievals was 1.012 with a standard deviation of 0.028 (9 DU) in Toronto, and the mean ratio of retrievals of the UV-MFRSR to

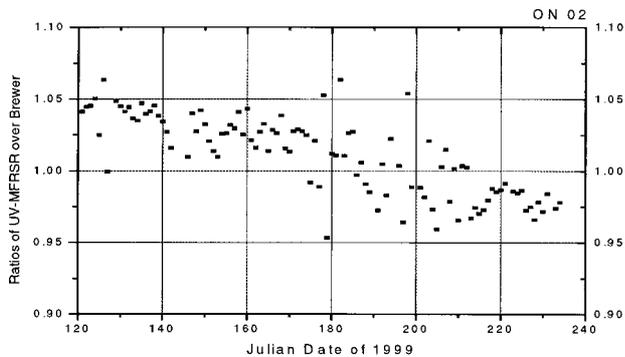


Fig. 2. Ratios of the UV-MFRSR column ozone to Brewer between April and August, 1999, at Toronto, Ontario. The mean ratio of the UV-MFRSR divided by the Brewer was 1.012, and the standard deviation was 0.028.

the Brewer was 0.988 with a standard deviation of 0.025 (7.5 DU) in Regina. The mean of the ratios demonstrates the systematic difference among three types of measurement, and the standard deviation about the mean indicates the random differences. The uncertainties of the UV-MFRSR direct-Sun ozone retrievals are explored below.

#### 4. Uncertainty Analysis of the UV-MFRSR Direct-Sun Ozone Retrievals

Potential instrumental error occurs if the signal-to-noise ratio of any of the four channels becomes too small. The detection limit of the 305-nm channel of

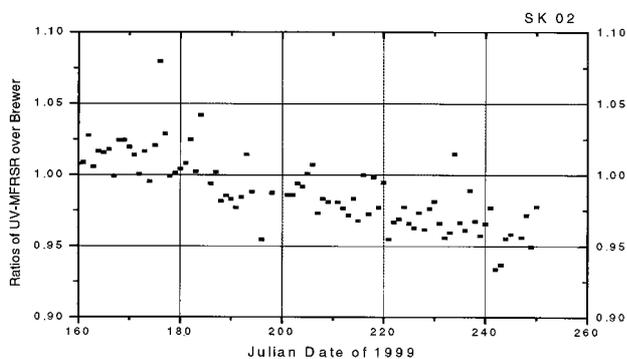


Fig. 3. Ratios of the UV-MFRSR column ozone to Brewer between June and September, 1999, at Regina, Saskatchewan. The mean ratio of the UV-MFRSR divided by the Brewer was 0.988, and the standard deviation was 0.025.

Table 1. Summary of Direct-Sun Ozone Retrieval Comparison Results

Ratio	UV-MFRSR/ Dobson	UV-MFRSR/ Brewer	Station
Mean May–Sept. 1999	0.998	0.986	Mauna Loa
Standard deviation	0.012	0.011	—
Mean Apr.–Aug. 1999	—	1.012	Toronto
Standard deviation	—	0.028	—
Mean June–Sept. 1999	—	0.988	Regina
Standard deviation	—	0.025	—

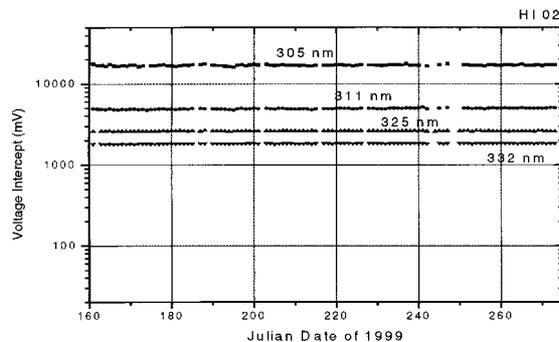


Fig. 4.  $V_0$  time series for unit 386 at Mauna Loa, Hawaii.

the UV-MFRSR is SZA  $80^\circ$ , owing to the poor signal-to-noise ratio and the angular response of the detector.<sup>5</sup> The column ozone retrievals made in this study restrict cases in which SZA  $< 70^\circ$ . The UV-MFRSR's direct beam is corrected for its measured angular response.<sup>7</sup> Although the direct beam can be corrected for SZA  $> 70^\circ$ , we consider the correction to be less reliable because of the large corrections necessary. Thus poor angular response is the dominant reason to use only SZA  $< 70^\circ$ .

The extraterrestrial voltage intercepts used in this study were generated by the Langley method<sup>17</sup> to extrapolate voltages of each wavelength to their value above the Earth's atmosphere. The range of air masses 1.2–2.2 was used for 305 and 311 nm, and 1.5–3.0 was used for 325 and 332 nm. The  $V_0$  used for a particular day was predicted from a single least-squares regression of all the  $V_0$  points at a particular wavelength obtained during the instrument deployment period at the selected location. This method accounts for any possible changes in each channel's responsivity. Figure 4 shows a time series of  $V_0$  values with an instrument (unit 386) that made measurements in Mauna Loa, Hawaii, during the study period. In the figure are four separate wavelengths, and each point is a separate Langley event at that wavelength. The extrapolations of  $V_0$  are within a standard deviation of the mean 0.17% or smaller at these four wavelengths. There are errors associated with voltage measurement, and we estimated the worst case as an overall uncertainty of  $\pm 1.57\%$  for all channels of UV-MFRSR used in this study.

In the calculation for UV-MFRSR column ozone retrievals, ozone absorption coefficients were computed at a temperature of 227.0 K. This temperature is representative of the average column temperature weighted by the ozone concentration. The real temperature changes with the height, latitude, and season. The Dobson and the Brewer use ozone-weighted temperatures of 227.0 and 228.3 K, respectively. The difference in ozone absorption coefficients over all channels in the model is  $\sim 2.4\%$  larger at 228.3 K than at 227.0 K. If the ozone absorption coefficients at 228.3 K were used in the model, less ozone would be retrieved from the UV-

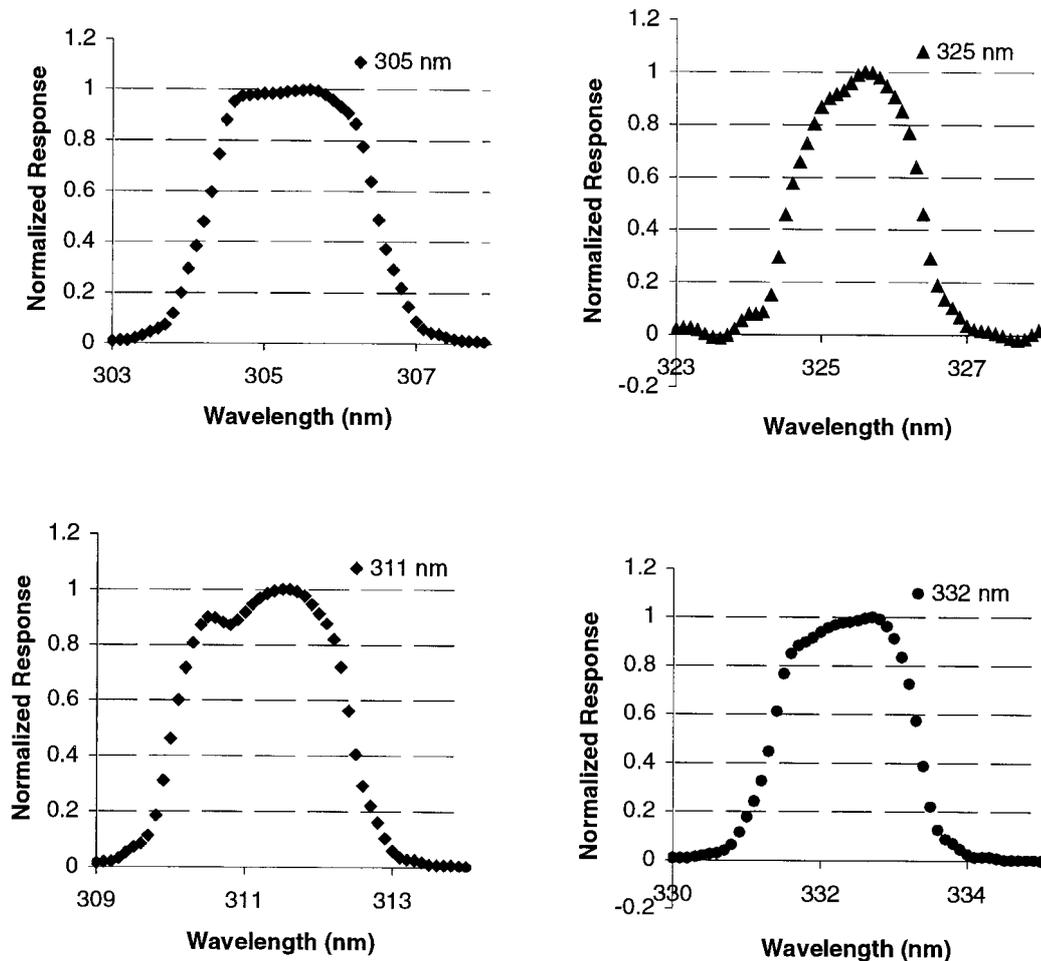


Fig. 5. Spectral response functions for 305-, 325-, 311-, and 332-nm channels for unit 287 used in Regina, Saskatchewan, during the study period.

MFRSR, which will change the bias compared with both the Dobson and the Brewer.

The ozone absorption coefficients and Rayleigh scattering coefficients in the calculation are functions of wavelength. The ozone absorption coefficients decrease with increasing wavelength. Changes or errors in measurement of SRFs will produce errors in the column ozone estimates. The SRFs of each channel of the units used in this study were characterized at the CUCF, Boulder, Colorado. Figure 5 gives the measured SRF of 305-, 325-, 311-, and 332-nm channels of unit 287 that was used in Regina, Saskatchewan. Bigelow and Slusser<sup>18</sup> evaluated the stability of UV-MFRSRs by comparing the reproducibility of each unit's SRF with its previous SRF each time an instrument is returned for calibration and indicated that the ion-assisted-deposition filters employed in the UV-MFRSR are stable. The combined shift and uncertainty of remeasured SRFs have been shown to be  $<0.1$  nm. The CUCF performed 34 repeat filter SRFs, which allow for statistical assessment of the SRF's stability. Since the CUCF's wavelength measurement accuracy and repeatability is better than  $\pm 0.02$  nm and does not change over time,<sup>17</sup> all paired differences reflect real shifts in a

channel's SRF. The median shift in SRF ranged from 0.016 to 0.037 nm with a standard deviation of between 0.061 and 0.103 for the four channels (Table 2). A wavelength shift (or error in characterization) of  $\pm 0.1$  nm was assigned for all channels to determine the possible errors in ozone absorption coefficients and Rayleigh scattering coefficients in the model. An uncertainty of  $\pm 0.1$  nm in SRF resulted in an uncertainty of  $\pm 1.13\%$  in ozone retrievals because of ozone absorption coefficients and  $\pm 0.23\%$  because of Rayleigh scattering coefficients.

The effects of variations in atmospheric pressure should be accounted for in the direct-Sun ozone retrievals model. Variation of surface pressure by

Table 2. Actual Shift of Wavelength Centroid as Measured by the CUCF

No. of Heads	Wavelength (nm)	Median	Mean	Standard Deviation
34	305	0.017	0.019	0.064
34	311	0.021	0.028	0.103
34	325	0.026	0.027	0.072
34	332	0.037	0.035	0.061

**Table 3. Factors Leading to Uncertainties of the UV-MFRSR Direct-Sun Ozone Retrievals**

Factor	Uncertainty in Ozone Retrievals (%)
Uncertainty due to $V_0$	$\pm 1.57$
Repeatability of voltage measurement	$\pm 0.5$
Uncertainty due to wavelength shift or measurement error for Rayleigh scattering coefficients	$\pm 0.23$
Uncertainty due to wavelength shift or measurement error for ozone absorption coefficients	$\pm 1.13$
Uncertainty due to atmospheric pressure	$\pm 0.02$
Uncertainty due to time stamp	$\pm 0.09$
Uncertainty due to aerosol distinction assumption	$\pm 0.44$
Total uncertainty	$\pm 2.0$

$\pm 20$  mbars, typical of the extremes in pressure because of synoptic patterns in the atmosphere, resulted in uncertainty in ozone retrievals of  $\pm 0.02\%$ . The air mass calculations used in the model depend on the SZA. The uncertainty on the setting of the time stamp is  $\pm 15$  s, which is approximately the duration of measurement. This causes uncertainty in air mass determination, which introduces a bias of  $\pm 0.09\%$  in column ozone retrievals. The timing uncertainty is a small contribution to the overall uncertainty budget.

It is assumed that these uncertainties described above are randomly distributed and independent and that they may be combined by use of the root-mean-squares method.<sup>19</sup> The total uncertainty in the absolute accuracy of the direct-Sun ozone retrievals is  $\pm 2.0\%$ . Factors leading to uncertainties of the UV-MFRSR direct-Sun ozone retrievals in this study are given in Table 3.

### 5. Conclusions

Total column ozone measured by the UV-MFRSR was found to agree with those from collocated Brewer and Dobson spectrophotometers in three locations within the combined uncertainties of all instruments. This agreement suggests that the total column ozone can be measured by use of the direct irradiance of solar UV radiation from the UV-MFRSR with an accuracy approaching the other two instruments. The uncertainty in UV-MFRSR direct-Sun ozone retrievals is  $\pm 2.0\%$ . The comparisons show the mean total column ozone measured for this research to be 0.2% (0.53 DU) smaller than concurrently measured Dobson data and  $\sim 1.4\%$  smaller (3.6 DU) than measured Brewer at Mauna Loa station, and  $\sim 1.2\%$  higher (3.8 DU) than measured Brewer data at Toronto and 1.2% smaller (3.7 DU) than measured Brewer data at Regina stations. Although more data is needed for conclusive results, these results demonstrate that the UV-MFRSR could be a viable alternative to the

widely used Dobson and Brewer spectrophotometers for total column ozone determination. In addition to total column ozone, the UV-MFRSR measures total and diffuse irradiances as well as the aerosol optical depth.

This paper is dedicated to David Bigelow, who died 10 June 2000. His passion for the environment and quality measurements will remain an inspiration to all who knew him. This study was funded by the USDA (agreements 98-34263-6888 and 99-34263-8566) and NASA (contract NCC5-288). The research results reported in this paper would not have been possible without the dedicated help of Bill Durham and George Janson in maintaining all the monitoring equipment in the network and Bill Davis and Becky Olson for maintaining a steady stream of daily measurement data from the network. Special thanks are due to Vitali Fioletov and Gloria Koenig for their kind assistance in supplying total ozone data taken with the Dobson and the Brewer instruments. The assistance of Roberta Brouwer, who helped make this paper more readable, is appreciated. The authors acknowledge David Wardle and the two reviewers for their excellent suggestions.

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