# Intercomparison of Simultaneously Obtained Infrared (4.8 μm) and Visible (515−715 nm) Ozone Spectra Using ACE-FTS and MAESTRO

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Laboratory ozone absorption spectra were measured simultaneously in the visible (515−715 nm) and infrared (2070−2140 cm$^{-1}$) spectral regions using SCISAT-1's MAESTRO (Measurement of Aerosol Extinction in the Stratosphere and Troposphere Retrieved by Occultation) and ACE-FTS (Atmospheric Chemistry Experiment−Fourier Transform Spectrometer) spectrometers. An intercomparison of these measurements was used to assess the relative accuracy of HITRAN absolute line strengths, for which there was a 4% change between the 2000 and 2004 versions. Results reported here show that Chappuis band cross section strengths are more consistent with the HITRAN 2004 4.8 μm band line strengths than with the 2000 compilation.

## 1. Introduction

Ozone is a molecule of prime importance to atmospheric science because of its role as an absorber of solar UV radiation and as a tropospheric pollutant. It has absorption features in the UV, visible and infrared regions of the electromagnetic spectrum that make it accessible to a variety of ground-based, airborne and satellite-based remote sounding instruments. High-accuracy visible cross sections and infrared line strengths, obtained through laboratory measurements, are essential for performing accurate atmospheric measurements. It is difficult to measure the exact amount of ozone in a viewing path when performing laboratory measurements, so discrepancies can exist in the absolute strengths obtained by various research groups. In the infrared, a recent comparison of four measurement data sets from the 10 μm band[1] showed that the retrieved absolute strengths of three of them disagreed significantly with the HITRAN 2000[2] line strengths. This led the compilers of the HITRAN 2004 database to decrease line strengths by 4% for all infrared bands. In the Hartley−Huggins and Chappuis bands, a recent comparison of cross section data sets[3] shows an agreement on the order of 2−3%.

An opportunity arose during the prelaunch testing of the two spectrometer instruments on board Canada's SCISAT-1 satellite to compare the absorption by ozone in the visible and infrared. This type of measurement is challenging and rarely performed, primarily because of the difficulty in creating a stable, high-powered illumination source spanning the wide spectral range required for simultaneous visible and infrared absorption band measurements. Ozone amounts obtained with both instruments using different sets of visible cross sections and infrared line data were compared.

## 2. Experimental Apparatus

SCISAT-1, a small Canadian satellite, was successfully launched on August 12, 2003. It carries two instruments, ACE-FTS and MAESTRO, which are designed to measure vertical profiles of various gases in the stratosphere by observations of solar occultations.[4] A sequence of absorption spectra of the earth's atmosphere is recorded during sunrise and sunset. The two instruments operate in the infrared (750−4400 cm$^{-1}$) and UV−visible (280−1000 nm) regions of the spectrum, respectively. The ACE-FTS is a 0.02 cm$^{-1}$ unapodized resolution (25 cm maximum optical path difference) Michelson interferometer with InSb and HgCdTe (MCT) detectors, designed and built by ABB−Bomem Inc. MAESTRO comprises two holographic grating spectrometers with 1024-pixel detector arrays. One operates in the near UV (280−500 nm) with a resolution of 1.5 nm and the other in the visible (500−1000 nm) with a resolution of 2 nm. It was designed and built by the Meteorological Service of Canada and EMS Technologies, Inc.

The prelaunch characterization of ACE-FTS and MAESTRO took place at the University of Toronto's Instrument Characterization Facility (ICF) during February and March of 2003. To simulate orbital pressure and temperature conditions, the instruments were mounted on a replica of the SCISAT-1 baseplate and placed in the vacuum tank at the ICF. ACE-FTS and MAESTRO look at light coming from a common aperture and have coincident fields of view, two key features that made simultaneous laboratory spectral measurements of absorbing gases possible. A solar simulator source was constructed by combining infrared light from a high-temperature (3000 K) blackbody source with visible light from a calibrated 1000 W quartz-halogen bulb. It was initially planned that a xenon arc lamp would be used to provide illumination in the ultraviolet and visible regions, but it was found to be too temporally unstable for accurate measurements. Consequently, ozone absorption measurements by MAESTRO were confined to the Chappuis band because of lack of sufficient illumination at near-UV wavelengths from the quartz-halogen lamp.

Figure 1 illustrates the positioning of the components of the experimental apparatus. A germanium beam splitter was used

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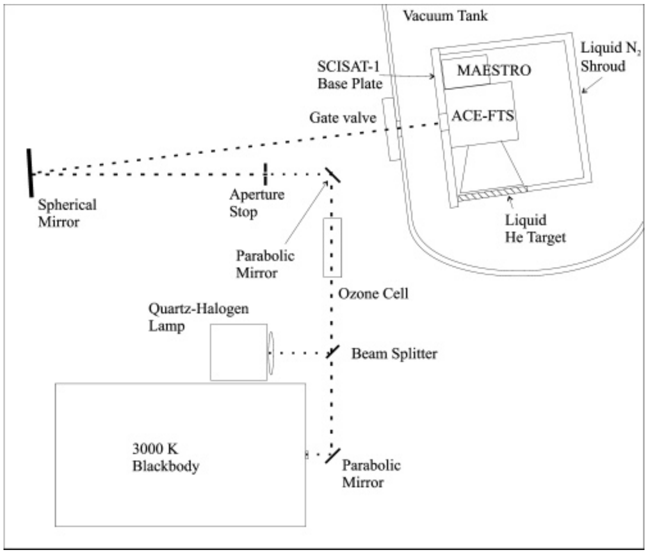

**Figure 1.** Experimental apparatus used for the ozone intercomparison measurements (not to scale).

to combine the beams from the two sources. The thermal runaway effect, a property of semiconductors characterized by the increase of the absorption coefficient with temperature, was avoided by the use of a cooling fan. This kept the beam splitter at a temperature of approximately 320 K, below the critical threshold.

The optical path (Figure 1) includes a segment where a gas cell can be inserted into the combined beam. A 20 cm long cell, previously developed for the characterization of the Odin Spectrometer and IR Imager System (OSIRIS) spectrograph,[5] was used for our measurements. Ozone was generated from molecular oxygen filling the cell at atmospheric pressure by applying a Tesla coil to the outside wall of the cell. The resulting electrical discharge between the Tesla coil and a coil inside the cell dissociates some of the oxygen molecules, leading to the production of ozone. The Tesla coil charger was applied for 30 min prior to making measurements.

The beam emerging from the gas cell was focused on an aperture stop, at the focus of a spherical mirror that produced a collimated beam with the same angular size as the Sun (9.6 mrad). The mirrors used to direct and focus the beams were made of uncoated gold for the blackbody beam and UV-enhanced aluminum for the combined beam. The gas cell and vacuum chamber windows were made of barium fluoride ($BaF_2$) to provide good spectral transmission from the UV to the infrared.

## 3. Gas Absorption Measurements

A series of absorption measurements of ozone was obtained, although only one set of measurements produced enough ozone for the accurate retrieval of amounts using the Chappuis band. Full cell measurements were bracketed by empty cell measurements, made with the ozone flushed out by pure oxygen. Comparisons of the two empty cell spectra were used to quantify the stability of the source. It was found that useful stability was achieved by allowing a 30 min warm-up period for the blackbody and quartz halogen sources before taking any measurements.

The photodissociation of ozone by light from the quartz–halogen source was found to be significant. The decrease in the amount of ozone with time was quantified for each of the individual spectral fits. Both instruments recorded approximately 300 spectra during a 20 min measurement period and measured a similar change in ozone amount.

The temperature of the ozone in the cell was measured using a thermocouple gauge placed on the outer surface of the cell. The use of this technique to determine the temperature inside the cell was validated by separate nitrogen dioxide ($NO_2$) absorption experiments performed using the same apparatus. Partial pressures of up to 900 Pa of nitrogen oxide (NO), generated by the photodissociation of $NO_2$, were measured. The temperatures retrieved from the relative strengths of NO absorption lines in the R-branch region of the fundamental band agreed with the thermocouple measurements to within 1 K.

Additional parameters required for spectral analysis were also obtained during the prelaunch test period: instrument line shape determination, field-of-view mapping, wavelength calibration and detector dark current generation rate.

## 4. Spectral Fitting Method

For both MAESTRO and ACE-FTS, the effective wavelength-dependent irradiance, $S(\lambda)$, incident on the detectors can be described as the convolution of the incident irradiance, $I(\lambda)$, with the normalized instrument line shape, $L(\lambda)$. Because $I(\lambda)$ is the irradiance of the solar simulator light going through the empty cell, then the full-cell irradiance is $I(\lambda)\,\tau(\lambda)$, where $\tau(\lambda)$ is the transmission by the ozone in the cell. The ratio of effective full-cell irradiance over effective empty-cell irradiance at a given wavelength is therefore

$$\frac{S_{\mathrm{FULL}}(\lambda)}{S_{\mathrm{EMPTY}}(\lambda)} = \left(\int_{\lambda_1}^{\lambda_2} I(\lambda')\,\tau(\lambda')\,L(\lambda-\lambda')\,\mathrm{d}\lambda'\right)\Big/\left(\int_{\lambda_1}^{\lambda_2} I(\lambda')\,L(\lambda-\lambda')\,\mathrm{d}\lambda'\right)$$

The integration limits $\lambda_1$ and $\lambda_2$ span the spectral range measured by the instrument, extended by the wavelength span of the instrument line shape $L(\lambda)$. This ratio was modeled for each measured frequency using modeled $\tau(\lambda)$, measured $L(\lambda)$, and a Planck blackbody function for $I(\lambda)$. The validity of using a blackbody function for the quartz-halogen bulb's irradiance was determined by comparison with externally calibrated spectra of the same bulb.

In the case of ACE-FTS, the modeled transmissions were obtained on a wavenumber grid ranging from 2070 to 2140 $\mathrm{cm}^{-1}$ with a resolution of 0.001 $\mathrm{cm}^{-1}$ by using the GENSPECT[6] line-by-line spectral toolbox. The GENSPECT algorithm used input line data from either HITRAN 2000 or 2004 and measured cell pressures and temperatures. For MAESTRO, the transmissions were calculated on a wavelength range 515–715 nm by using Beer's law with published cross sections obtained under the nearest possible pressure and temperature conditions.[7,8] The convolution with instrument line shape was done at a wavelength resolution corresponding to the smallest wavelength interval in the cross sectional data, approximately 0.1 nm for the Burrows et al.[8] data set and 0.01 for the Voigt et al.[7] data set.

The calculated full-over-empty cell irradiance ratios were then multiplied with measured, co-added empty-cell spectra to generate modeled full-cell spectra. Before these spectra were compared with the measured full-cell spectra, the latter were corrected for solar simulator fluctuations by intercomparison with the co-added empty-cell spectra in regions with no ozone absorption. Correction factors were thereby determined and applied to the measured full-cell spectra.

For MAESTRO, spectra from the visible spectrometer (500–1000 nm) were used, and for ACE-FTS spectra measured with

**TABLE 1: Error Sources for Individual MAESTRO and ACE-FTS Fitted Amounts**

| uncertainty source | MAESTRO | ACE-FTS |
|---|---|---|
| solar simulator fluctuations | 0.78% | 0.42% |
| wavelength dispersion | 0.06% | 0.02% |
| gas cell temperature | | 0.1% |
| instrument line shape | 0.01% | |
| **total uncertainty** | **0.78%** | **0.43%** |

the InSb detector were used. $\chi^2$ minimization of the difference between the measured and modeled full-cell spectra was accomplished iteratively by the adjustment of the normalized absorber amount and wavelength scaling parameters. The pixel-to-wavelength dispersion was modeled by a seventh-order Chebyshev polynomial for MAESTRO and a frequency point spacing parameter for ACE-FTS. InSb detector channeling, having a frequency of approximately 0.6 cm$^{-1}$ and a peak-to-peak magnitude of approximately 6% of the signal amplitude, was noticed in the ACE-FTS spectra. The channeling was successfully modeled as a cosine modulation proportional to the signal intensity. The best-fit channeling phase, frequency and magnitude were determined for each spectrum, which allowed for its removal prior to fitting.

## 5. Uncertainty Analysis

The effects of various sources of uncertainty were considered to determine the accuracy and precision of the ozone measurements. As shown in Table 1, the majority of the estimated uncertainty for both ACE-FTS and MAESTRO comes from random fluctuations in solar simulator irradiance. Although a correction factor applied to the measured full-cell spectra can successfully compensate for most of the fluctuations, small residual fluctuations remain. The analysis of a set of empty-cell spectra showed that these residual fluctuations appear to be random in frequency, amplitude and phase, and are therefore impossible to remove from the full-cell spectra. To estimate the uncertainty in the fitted ozone amount caused by these random fluctuations and from detector noise, the standard deviation of the difference between the individual measurements and the best-fit slope of ozone amount versus time was calculated. The values in Table 1 represent a 2$\sigma$ uncertainty.

The larger uncertainty for MAESTRO reflects a higher sensitivity to noise and intensity fluctuations due to the relatively low levels of absorption in the Chappuis band. Other error sources considered for MAESTRO include the uncertainty in the fitted wavelength dispersion coefficients and the instrument line shape. The instrument line shape uncertainty arises because the instrument exhibits a slight line shape change with wavelength and the line shape is determined from measurements using lasers at fixed wavelengths. However, the standard deviation of ozone amounts obtained by using different line shapes for convolution was found to be very small ($<$0.01%).

For ACE-FTS, the only significant error source besides intensity fluctuations is the uncertainty in the estimate of the temperature inside the cell, which was derived from thermocouple measurements. The estimated 1 K temperature uncertainty was found to correspond to a 0.1% uncertainty in the retrieved ozone amount. There is no significant effect of cell temperature uncertainty on the MAESTRO ozone measurements because the temperature dependence of the ozone cross section spectrum in the Chappuis band is very small.[7,8]

## 6. Results

Typical full and empty cell spectra obtained by both ACE-FTS and MAESTRO are shown in Figures 2 and 3. In these figures, the top plot represents a typical empty-cell spectrum and the one immediately beneath a full-cell spectrum.

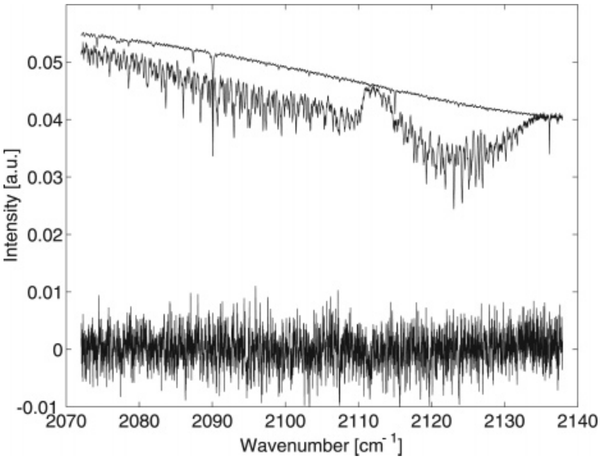

**Figure 2.** Sample full and empty cell spectra measured using ACE-FTS, with InSb detector channeling removed. The fit residual, multiplied by a factor of 10, is shown on the bottom.

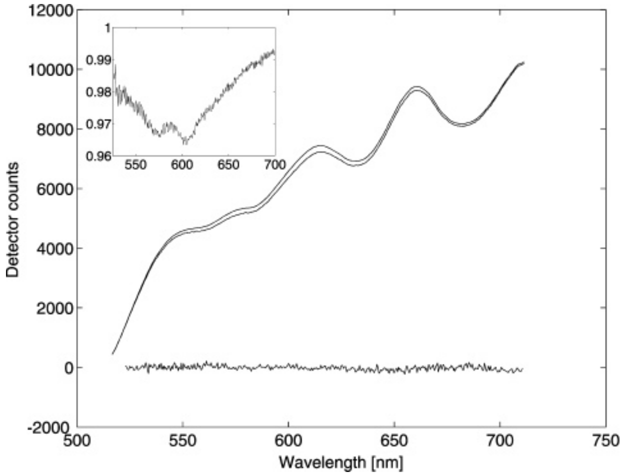

**Figure 3.** Sample full and empty cell spectra measured using MAESTRO. The low-frequency oscillations seen in both spectra are due to etaloning on the diode array detector. The inset on the upper left shows the transmission, obtained by dividing the full cell spectra by the empty cell spectra, as function of wavelength (nm). The full cell fit residual, multiplied by a factor of 10, is shown on the bottom.

Figure 4 shows the fit amounts, averaged in groups of 10, as a function of time for ACE-FTS and MAESTRO using different line data. The ozone amounts retrieved by MAESTRO using Voigt or Burrows cross sections were virtually identical; therefore the Burrows amounts were omitted from the figure for clarity. A linear ozone photodissociation trend was assumed; therefore the individual fitted amounts were simply averaged to obtain the values shown in Table 2.

It should be noted that ozone measurements were also obtained in the 10 $\mu$m band region using the ACE-FTS MCT detector. However, the obtained ozone amounts were too inaccurate for intercomparison with 4.8 $\mu$m band and Chappuis band results. Significant structures, probably due to differences in channeling between the full end empty cell measurements, were seen in the fit residuals. Channeling is complex in this spectral region and cannot be easily modeled and removed using a cosine function as was the case for the 4.8 $\mu$m band data. In addition, supplementary measurements have shown that irregular absorption features due to ice deposition on the detector optical system occur in this spectral region. Sufficient ice buildup to influence the fit results could have occurred in the time between

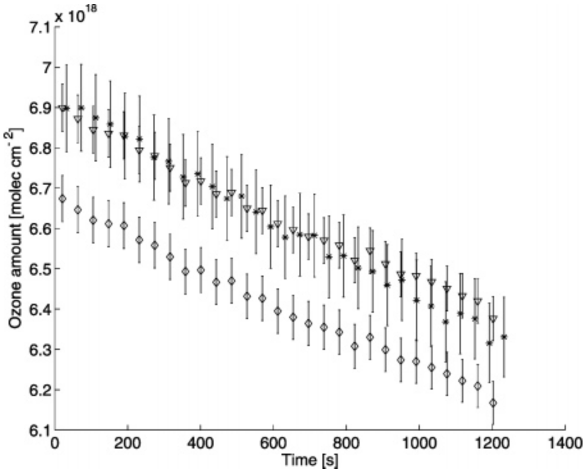

**Figure 4.** Fitted ozone amounts as function of time using MAESTRO and ACE-FTS. Triangles represent ACE-FTS fitted with HITRAN 2004, diamonds are ACE-FTS with HITRAN 2000, and asterisks are MAESTRO fits using Voigt et al.[7] cross sections.

**TABLE 2: Mean Ozone Amounts Obtained with ACE-FTS and MAESTRO Using Different Cross Section Data**

| | fit amt [molecules/cm$^2$ × $10^{18}$] | ratio (amt/Voigt) |
|---|---|---|
| MAESTRO with Voigt et al. cross sections | 6.61 ± 0.05 | 1.000 |
| MAESTRO with Burrows et al. cross sections | 6.61 ± 0.05 | 0.999 |
| ACE-FTS with HITRAN 2000 | 6.41 ± 0.03 | 0.970 |
| ACE-FTS with HITRAN 2004 | 6.63 ± 0.03 | 1.003 |

the full and empty cell measurements. It may be possible to retrieve 10 μm region ozone amounts with an appropriate channeling and ice deposition model, but that is beyond the scope of this paper.

## 7. Discussion

In a report commissioned by the European Space Agency,[9] Orphal has considered the various measurements of the visible and UV absorption cross sections of ozone. For the Chappuis bands he recommends the cross sections of Bogumil et al.[10] measured with the preflight model of the SCIAMACHY instrument. At room temperature in the Chappuis band region used in our work, the Bogumil cross sections differ by a negligible amount (<1%) from the work of Voigt et al.[7] and Burrows et al.[8] The Chappuis band ozone cross sections of Bogumil et al.,[10] Voigt et al.,[7] and Burrows et al.[8] all originate from the University of Bremen group of Burrows and produce essentially identical results for our work. For simplicity we have adopted the work of Voigt et al.[7] as the reference in our comparisons.

As shown in Table 2, the ozone amounts fitted using MAESTRO agree within uncertainty with the ACE-FTS amounts obtained using HITRAN 2004. This finding contrasts with the IR/UV intercomparison by Picquet-Varrault et al.[11] In that study, the HITRAN 2000 and 2004 cross sections were found to be about 1.2% and 5.3% lower, respectively, than the values derived from their experiment, assuming the UV cross sections are correct. However, that study used Huggins region UV (302.15−313.17 nm) and 10 μm band IR (950−1085 cm$^{-1}$) absorption data.

Considering our results and those of Picquet-Varrault et al.,[11] there are four possible explanations: (1) Both the Huggins cross sections used by Picquet-Varrault et al.[11] and the Voigt et al.[7] Chappuis band cross sections are correct; therefore the HITRAN 2004 infrared absorption strengths are correct in the 4.8 μm band but need to be increased in the 10 μm band. (2) The Huggins cross sections are too high and both HITRAN 2004 IR bands are correct. (3) The Chappuis cross sections are too low and both HITRAN 2000 IR bands are correct. (4) The Huggins and Chappuis band data and HITRAN 2004 all have problems and need to be changed (e.g., by 2% each) to obtain agreement. Clearly, further studies are required, as our work offers no guidance as to which of these possibilities is correct.

Our work has implications for a number of satellite missions, including ACE,[4] that measure ozone. For example, Walker et al.[12] have found that the ACE-FTS atmospheric ozone profiles obtained using the HITRAN 2004 line parameters are about 5−10% smaller than those obtained with a number of UV/visible satellite instruments such as SAGE III near the ozone peak at 20 km. At 20 km altitude, limb-viewing instruments such as SAGE III generally use the Chappuis band for their retrievals. As the Chappuis band cross sections agree with HITRAN 2004 line strengths (at least for the 4.8 μm ozone band), it suggests that these discrepancies are not due to problems with the spectroscopic parameters but have some other source.

## 8. Conclusions

This intercomparison study of simultaneous ozone absorption measurements in the Chappuis band (515−715 nm) and the 4.8 μm IR band suggests that the HITRAN 2004 line intensity values are correct, assuming the Chappuis band cross sectional data of Voigt et al.[7] to be correct. These findings are in contrast with the results obtained by Picquet-Varrault et al.,[11] which suggest that the HITRAN 2000 line intensity values are correct, on the basis of an intercomparison of the Huggins bands and the 10 μm bands. There is no intrinsic reason at this stage to believe that the "generally accepted" cross sections for the Huggins bands or the Chappuis band of ozone are more accurate than the "generally accepted" infrared line intensities (e.g., HITRAN 2004). More work is clearly required on the absolute line intensities of ozone to resolve these problems.

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