



Tangent height determination from the N₂-continuum for the Atmospheric Chemistry Experiment Fourier transform spectrometer

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ABSTRACT

With inadequate information from sensors onboard the SCISAT satellite to generate accurate pointing information, tangent heights for the solar occultation measurements collected by the Atmospheric Chemistry Experiment Fourier transform spectrometer (ACE-FTS) are determined from features in the measured spectra. For version 4.0 processing of the ACE-FTS data, tangent heights below 18 km are generated from the N₂ collision induced absorption continuum near 2500 cm⁻¹. Because this tangent height determination requires no assumptions on CO₂ concentrations in this altitude region (unlike previous processing versions), ACE-FTS version 4.0 includes retrieved CO₂ volume mixing ratio profiles below 18 km as a standard product.

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1. Introduction

Developed under the auspices of the Canadian Space Agency, the Atmospheric Chemistry Experiment (ACE) is a satellite-based mission for remote sensing of the Earth's atmosphere [1,2]. Housed on board the small science satellite SCISAT, it was launched August 12, 2003 into a circular, highly-inclined orbit (650 km altitude, 74° inclination to the equator). The measurement technique employed is solar occultation, the geometry of which is depicted in Fig. 1. Using the Sun as a light source, the instruments collect a series of atmospheric absorption measurements as the Sun rises or sets from the orbiting satellite's perspective, providing up to 30 measurement opportunities per day.

The primary instrument on SCISAT is the Atmospheric Chemistry Experiment Fourier transform spectrometer (ACE-FTS), featuring high resolution (± 25 cm maximum optical path difference, 0.02 cm⁻¹ resolution) and broad spectral coverage in the mid infrared (750–4400 cm⁻¹), with a signal-to-noise ratio ranging from just under 100:1 up to ~400:1 [3].

The analysis of an ACE-FTS occultation begins with the determination of instrument pointing information for all of the measurements [4], typically expressed in terms of tangent height, which is defined as the distance of closest approach to the Earth's surface by a measured solar ray (see Fig. 1). In the absence of accurate pointing information from the satellite itself, tangent heights must be inferred from the ACE-FTS measurements. One common

practice is to assume a CO₂ volume mixing ratio (VMR) profile and then derive the pointing geometry required in forward model calculations to reproduce a set of CO₂ spectral features in the measurements [4,5], which is the approach employed in all previous ACE-FTS processing versions. This, however, prohibits the retrieval of an independent CO₂ VMR profile at low altitude. One can only at best reproduce the original, assumed profile.

Note that at high altitude (above ~60 km), retrieval of CO₂ VMR [6,7] is possible because accurate relative pointing information can be derived from knowledge of the satellite orbit. At low altitude, atmospheric refraction, clouds, and aerosols impact instrument pointing, and tangent heights cannot simply be calculated from the satellite-Earth-Sun geometry, necessitating the use of ACE-FTS spectra to derive pointing information.

An alternative approach for tangent height determination makes use of N₂ features in the infrared spectrum rather than CO₂. The VMR profile for N₂ is well-known, and, unlike CO₂, does not vary significantly with time, season, or location in the altitude region of interest (between 5 and ~20 km), thereby requiring fewer assumptions in the analysis inputs. A number of studies have investigated using the N₂ collision induced absorption (CIA) continuum in the analysis of ACE-FTS spectra, generating corrections to retrieved ACE-FTS version 2.2 or version 3.0 tangent heights (depending on the study) and then retrieving CO₂ VMR profiles [8–10]. These studies all avoided occultations that contained cloud signatures in the spectra. For the upcoming processing version of ACE-FTS data (version 4.0), tangent heights below 18 km will be derived using the N₂ continuum, including cloud-containing occul-

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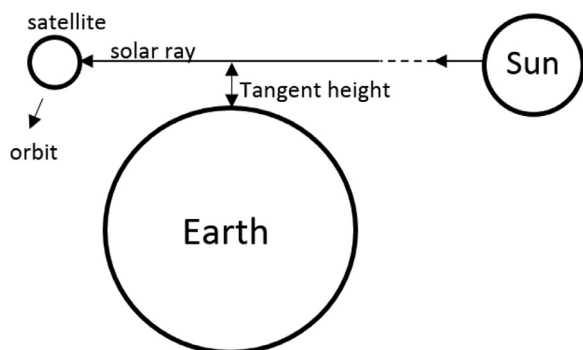


Fig. 1. Diagram (not to scale) depicting the geometry of solar occultation measurements.

tations, thereby enabling the retrieval of low-altitude (between 5 and 18 km) CO_2 from ACE-FTS measurements.

As a potent greenhouse gas exhibiting steadily increasing concentrations in large part due to human activity, atmospheric CO_2 carries a high priority for routine monitoring. Information on CO_2 is currently provided by a number of sources, including (but not limited to) the Total Carbon Column Observing Network (TCCON) [11], a ground-based network of instruments measuring a number of carbon-containing atmospheric constituents, as well as the CarbonTracker modeling system [12] administered by the National Oceanic and Atmospheric Administration (NOAA), which assimilates a network of measurements on the Earth's surface, on towers, from aircraft, and from ships on the ocean. Several satellite missions have measured the column-average dry-air mole fraction of atmospheric CO_2 , notably the Orbiting Carbon Observatory 2 (OCO-2) mission [13], the Greenhouse gases observing satellite (GOSAT) mission [14], and the Scanning Imaging Absorption Spectrometer for Atmospheric Cartography (SCIAMACHY) mission [15]. Measurements of tropospheric CO_2 have also been reported by the Atmospheric Infrared Sounder (AIRS) mission [16] and the Tropospheric Emission Spectrometer (TES) mission [17].

The bulk of CO_2 data are either local (*in situ*) measurements or column amounts. The ACE-FTS results will provide CO_2 altitude profiles extending no lower than 5 km, from which it would be challenging to derive information on CO_2 sources and sinks. However, the profile information is complementary to the bulk of existing CO_2 data, a fact that could be exploited to improve knowledge of CO_2 distributions and improve flux inversions [18].

2. Calculating the N_2 continuum

2.1. Collision induced absorption cross sections

N_2 - N_2 CIA normalized absorption coefficients ($B_{\text{N}_2-\text{N}_2}$) are available in the HITRAN compilation [19]. They consist of a set of five out of the six measurements originally described in Lafferty et al. [20] covering the temperature range 228–272 K, and another set of five measurements from Baranov et al. [21] covering a higher temperature range: 300–362 K. The data are plotted in Fig. 2, with units expressed in $\text{cm}^{-1} \text{ amagat}^{-2}$, converted from the units in HITRAN ($\text{cm}^5 \text{ molecule}^{-2}$) using the factor of 1.385×10^{-39} provided in Richard et al. [19]. In the plot label, the amagat unit is abbreviated as Am.

Previous studies of the N_2 continuum in ACE-FTS spectra [8–10] did not employ the set of higher temperature data. This additional information should allow for improved characterization of temperature dependences.

Performing a quality check on the data set, the curve for temperature 233.7 K exhibits unexpected behavior at higher wavenumbers, with the measured signal leveling off above $\sim 2650 \text{ cm}^{-1}$, making for an increasing high bias relative to the expectations from the curves for other temperatures, as can be seen in Fig. 3. This poses a difficulty for accurate calculation of N_2 CIA in this wavenumber region. The 233.7 K curve also exhibits significantly higher noise levels than the other curves. This temperature will therefore be excluded from the analysis that follows.

Fig. 4a plots the data from HITRAN in the vicinity of a wavenumber where the curves exhibit a minimum in temperature dependence. The high temperature data set features a compact relationship among the curves for different temperatures, with a

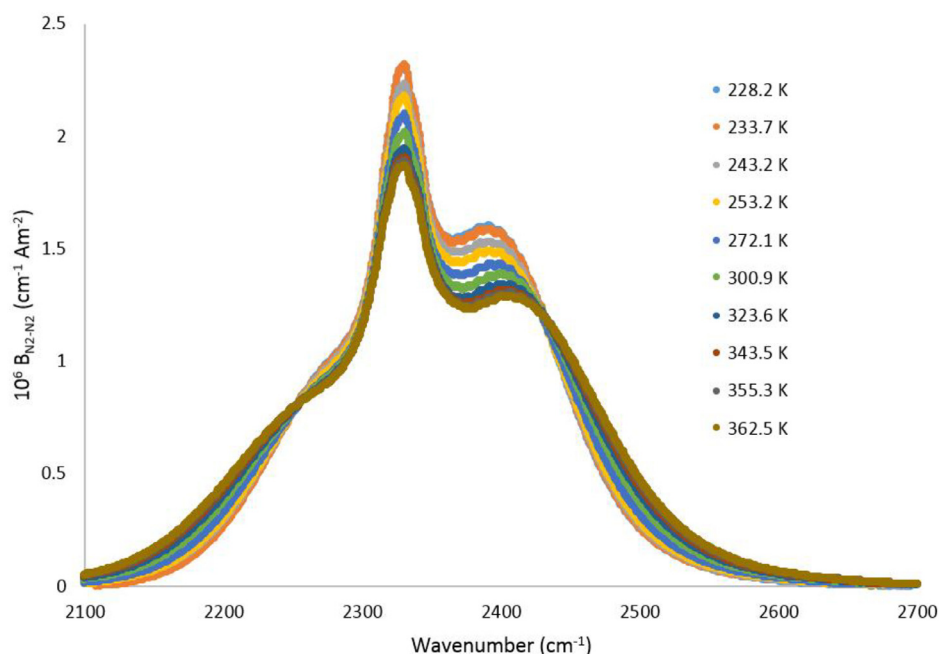


Fig. 2. The set of infrared N_2 - N_2 collision induced absorption normalized absorption coefficients from HITRAN.

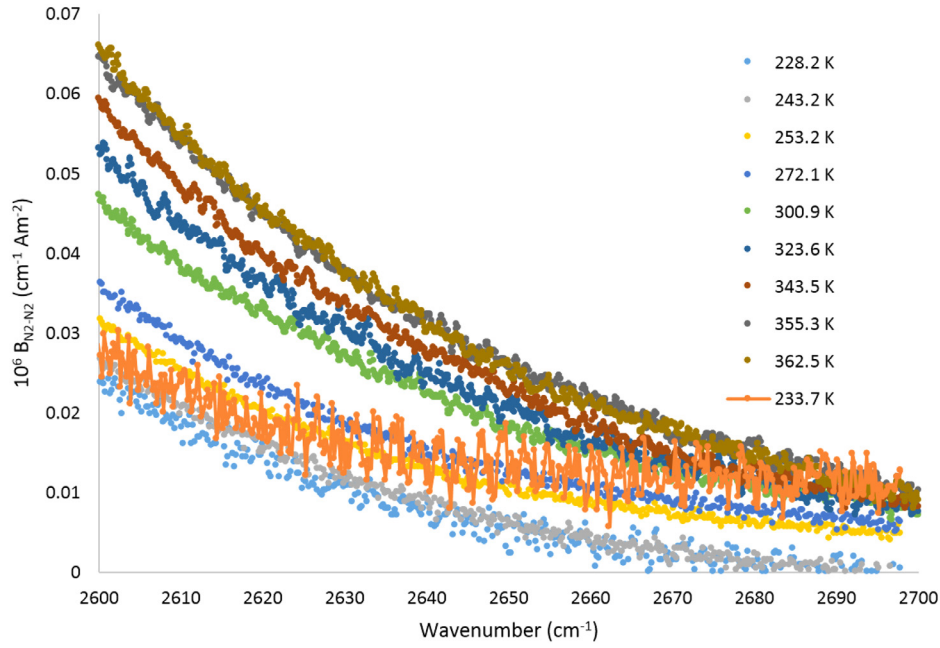


Fig. 3. The high wavenumber wing of the N_2 - N_2 collision induced absorption normalized absorption coefficients. The curve for temperature 233.7 K displays anomalous behavior.

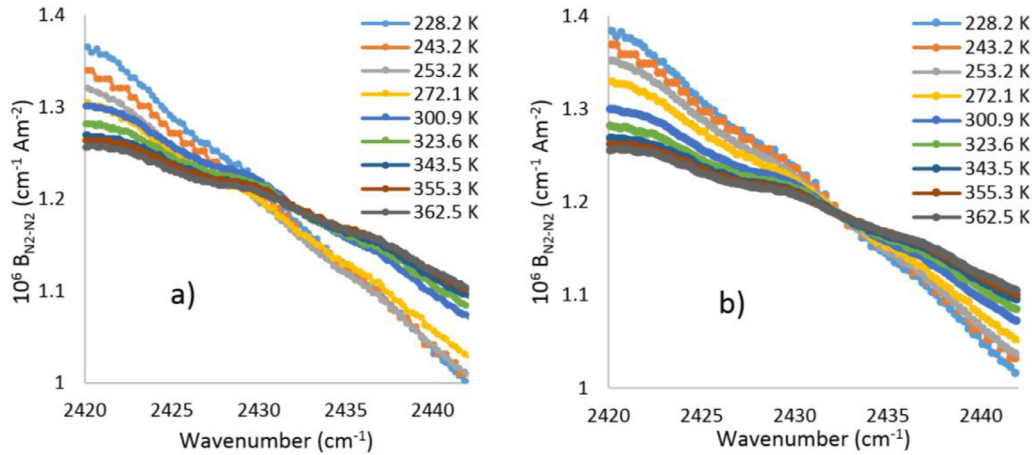


Fig. 4. N_2 - N_2 CIA normalized absorption coefficient curves in the vicinity of a point with minimal temperature dependence: (a) using the original data from HITRAN, and (b) after applying a scaling to achieve improved internal consistency.

(roughly) common value near 2432.5 cm^{-1} , which represents a crossover point such that absorption coefficient decreases with increasing temperature for wavenumbers below that point and increases with increasing temperature at higher wavenumbers. The low temperature data set also has a crossover point in the same vicinity, but it is less well-defined and has a clear low bias relative to the high temperature data set.

To improve the internal consistency among the different curves, all were scaled to obtain the same absorption coefficient at 2432.5 cm^{-1} , where the target value at that wavenumber is taken as the average from the five curves in the high temperature data set. Applying a scaling presumes that inconsistencies more likely arise from calibration issues, as opposed to something like detector nonlinearity, which could yield offsets in the measurements.

The scaling factors determined in this process are provided in Table 1. The adjusted curves in Fig. 4b exhibit much more regular behavior as a function of temperature in the vicinity of the

Table 1

Scaling factors applied to the cross section curves from the HITRAN compilation.

Temperature (K)	Scaling factor
228.2	1.0147
243.2	1.0217
253.2	1.0251
272.1	1.0206
300.9	0.9996
323.6	1.0003
343.5	1.0002
355.3	0.9991
362.5	1.0007

crossover point. Justification for applying this scaling is perhaps best illustrated by looking at the integrated intensities (calculated

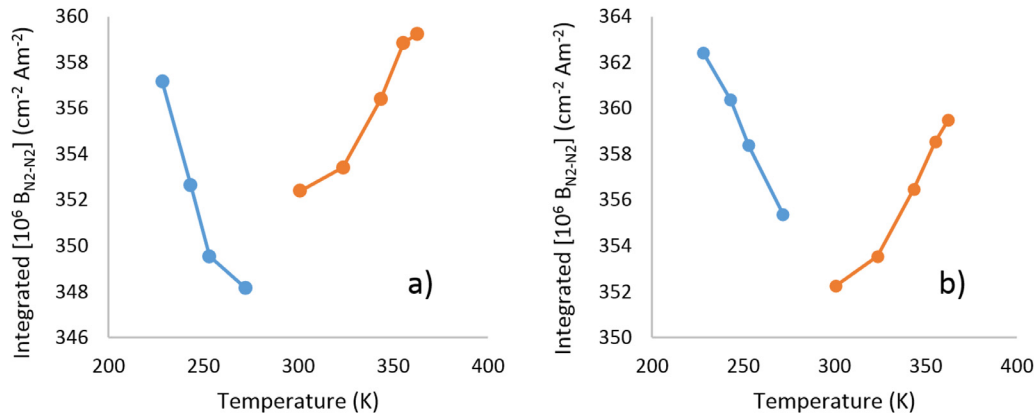


Fig. 5. Integrals of the normalized absorption coefficient curves: (a) using the original curves from HITRAN, and (b) using the curves scaled to improve internal consistency.

over the range 2130–2650 cm^{-1}) of the different curves as a function of temperature, shown in Fig. 5. With the original data (in Fig. 5a), there was a distinct step in integrated intensity between the two data sets. After the curves were scaled, integrated intensity appears to be more consistent with a single, unified function for the two data sets (see Fig. 5b).

2.2. Empirical function for temperature dependence: 2130–2610 cm^{-1}

The analysis of ACE-FTS data involves calculating the N_2 CIA continuum for a variety of pressure and temperature conditions. For ease of calculation, the N_2 normalized absorption coefficients are expressed as an empirical function of temperature on a fixed wavenumber grid, following the procedure described in Lafferty et al. [20]. To calculate N_2 CIA, absorption coefficient values for the given temperature are populated on a fixed wavenumber grid using the empirical function, and then the values at the required wavenumber points are determined through interpolation (e.g., using cubic splines). The empirical function often serves to extrapolate in temperature, in that the lowest temperature in the data set is 228.2 K, whereas temperatures near the tropopause can fall below 200 K. Any troubles with the empirical function will likely compound when extrapolating.

Lafferty et al. [20] suggested the following empirical function to account for the temperature dependence of the N_2 CIA absorption coefficients:

$$B_{N_2-N_2}(\sigma, T) = B_{N_2-N_2}^0(\sigma) \exp\left[\beta_{N_2-N_2}^0(\sigma)\left(\frac{1}{T_0} - \frac{1}{T}\right)\right], \quad (1)$$

where σ is wavenumber, T is temperature, and the reference temperature, T_0 , is taken as 296 K. They employed a fixed wavenumber grid that extended from 2125 to 2600 cm^{-1} with a grid spacing of 5 cm^{-1} . However, this grid spacing fails to fully capture the weak (1 to 2% amplitude) structure discussed in Lafferty et al. [20]. We therefore employ the following variable-width wavenumber grid: from 2130 to 2300 cm^{-1} with 5 cm^{-1} spacing, from 2300 to 2550 cm^{-1} with 2 cm^{-1} spacing, and from 2550 to 2740 cm^{-1} with 5 cm^{-1} spacing. The regions from 2130 to 2610 cm^{-1} and 2610 to 2740 cm^{-1} will be treated in separate fashions, as will be described later.

The absorption data were fit to the expression in Eq. (1), a least-squares analysis determining two parameters (the amplitude $B_{N_2-N_2}^0(\sigma)$ and the temperature coefficient $\beta_{N_2-N_2}^0(\sigma)$) at each grid point over the range 2130–2610 cm^{-1} , a total of 344 parameters (all determined simultaneously) for the variable-width grid configuration described above. The fitting residuals for the wavenumber region of greatest interest for the analysis (2400 to

2610 cm^{-1}) are provided in Fig. 6. They are expressed as Observed/Calculated (rather than Observed – Calculated) to give a better indication of the percentage errors. It is clear that the empirical function in Eq. (1) does not adequately represent the data with the higher temperature curves added to the mix. The inability to accurately reproduce the fitting data is particularly concerning when the empirical function needs to extrapolate in temperature.

It appears an additional parameter in the empirical function is required to fully capture the temperature variation. The proposed change is the addition of a quadratic term in the exponential:

$$B_{N_2-N_2}(\sigma, T) = B_{N_2-N_2}^0(\sigma) \exp\left[\beta_{N_2-N_2}^0(\sigma)\left(\frac{1}{T_0} - \frac{1}{T}\right) + \delta_{N_2-N_2}^0(\sigma)\left(\frac{1}{T_0} - \frac{1}{T}\right)^2\right]. \quad (2)$$

The residuals achieved with the form of the empirical function in Eq. (2) are shown in Fig. 7, and are within 1% for the region of greatest interest, compared to discrepancies up to roughly 8% for the residuals in Fig. 6. Note that large scatter in the residuals at higher wavenumbers results from the fact that the signal is low in this region, amplifying the residuals when they are expressed as Observed / Calculated.

The fitted parameters (a total of 516, with 3 parameters for each grid point) on the variable-width wavenumber grid for the wavenumber range 2130–2610 cm^{-1} are plotted in Fig. 8. These data are provided in Table A1 in Appendix A.

Residuals similar to those in Fig. 7 can be achieved by adding a constant to the empirical function in Eq. (1) instead of using the quadratic term in Eq. (2). However, the fitted constant term and amplitude $B_{N_2-N_2}^0(\sigma)$ exhibit high anti-correlation, which could signal problems for large extrapolations in temperature below the minimum temperature in the data set (228.2 K).

2.3. Empirical function for temperature dependence: 2610–2740 cm^{-1}

As mentioned previously, a different treatment of the data is employed at higher wavenumbers. Above about 2650 cm^{-1} , where the curves for the lowest two temperatures drop close to zero, the empirical function in Eq. (2) no longer works well, and fitting the data to that form yields results of dubious quality. The low temperature curves are near their upper wavenumber limit ($\sim 2697 \text{ cm}^{-1}$) and are therefore perhaps pushing the limitations of the measurement system. The high temperature data extend up to 3000 cm^{-1} and are presumably more reliable in this wavenumber region. However, it would be dangerous to ignore completely the

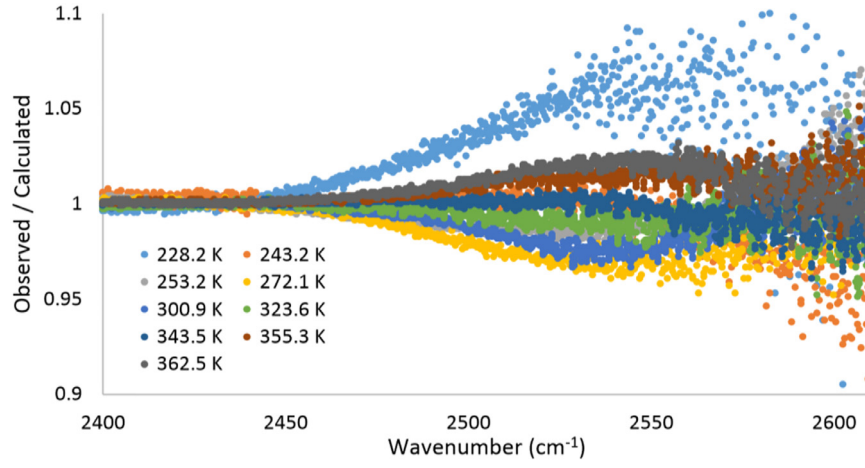


Fig. 6. Residuals (expressed as Observed / Calculated) for the empirical function from Eq. (1).

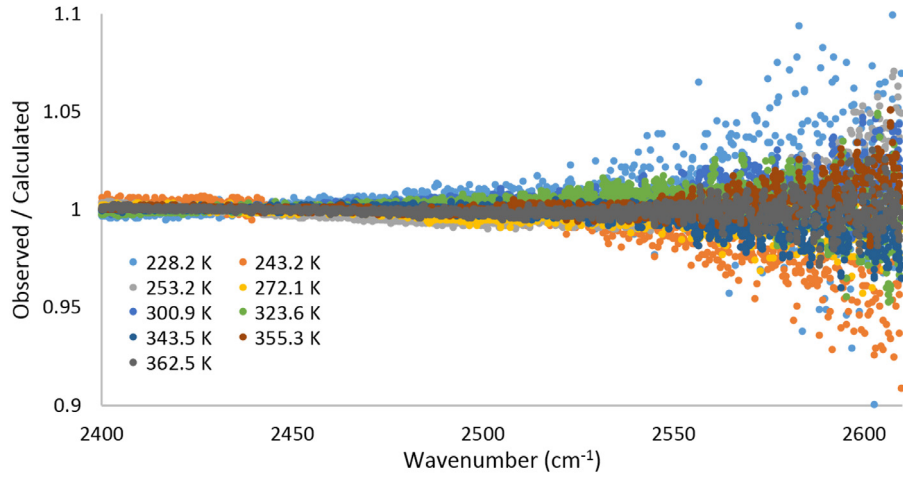


Fig. 7. Residuals (expressed as Observed / Calculated) for the empirical function from Eq. (2).

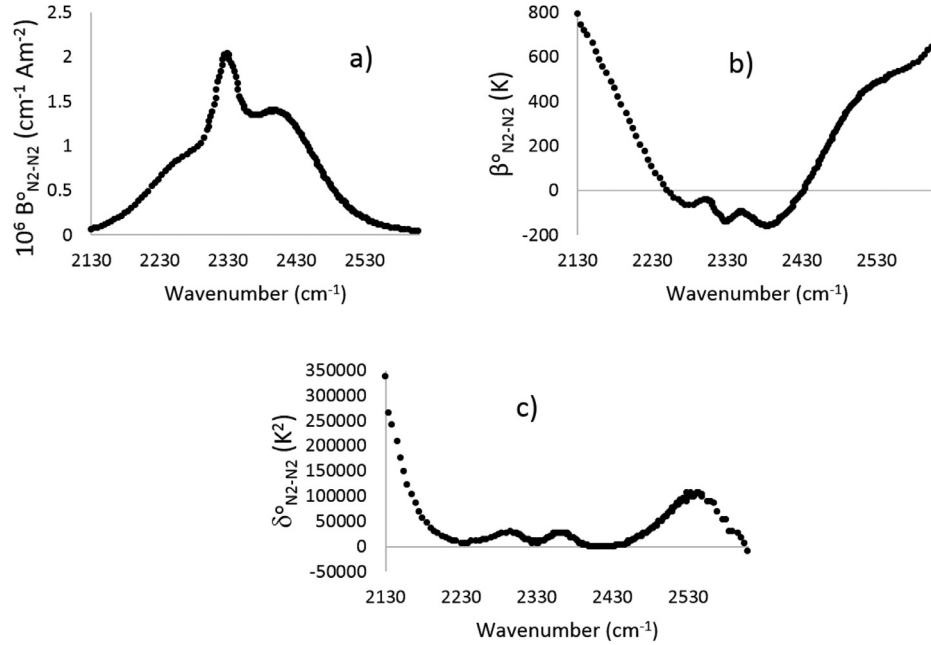


Fig. 8. Fitted parameters for the N_2-N_2 CIA normalized absorption coefficients using the empirical function in Eq. (2): (a) amplitude $B_{N_2-N_2}^0$ (σ), (b) linear term temperature coefficient $\beta_{N_2-N_2}^0$ (σ), and (c) quadratic term temperature coefficient $\delta_{N_2-N_2}^0$ (σ).

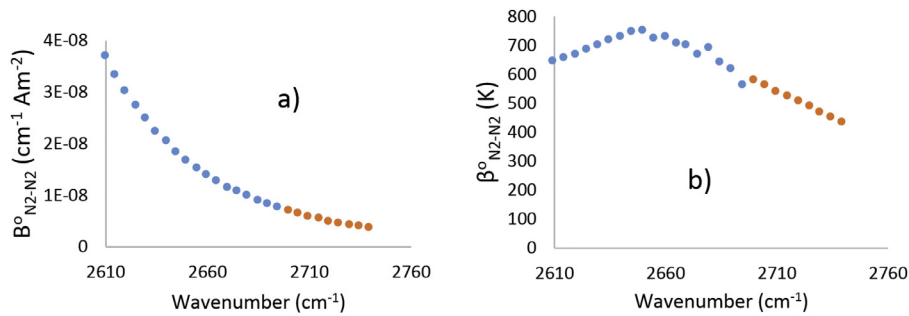


Fig. 9. Fitted parameters for the N_2-N_2 CIA normalized absorption coefficients using the empirical function in Eq. (1): (a) amplitude $B^0_{N_2-N_2}(\sigma)$, and (b) linear term temperature coefficient $\beta^0_{N_2-N_2}(\sigma)$. Blue points are the results from fitting the curves for the seven highest temperatures and orange points are extrapolations from the results in the range 2650–2695 cm^{-1} .

low temperature data, particularly because the temperatures associated with ACE-FTS measurements fall in that regime, not in the range of the high temperature data set.

The break point of 2610 cm^{-1} was chosen for the low wavenumber treatment in Section 2.2 because the variation with wavenumber for the fitted empirical parameters from Eq. (2) starts to change dramatically above that point, a symptom of the growing loss of internal consistency among the different curves with increasing wavenumber.

The curves for the two lowest temperatures (228.2 and 243.7 K), which drop close to zero above 2650 cm^{-1} , are clear outliers when attempting to fit this region. These two curves are therefore discarded for the high wavenumber analysis, and the remaining seven curves are fitted to the empirical function in Eq. (1) on a wavenumber grid with 5 cm^{-1} spacing, which is sufficiently accurate to reproduce the fitting data in this region. The function from Eq. (2), with the quadratic term in the temperature coefficient, does not provide significant improvement in fitting residuals, and the extra parameter could also amplify extrapolation errors, which is a concern because the lowest temperature employed in determining the empirical function for this wavenumber region was 253.2 K. The fitted parameters as a function of wavenumber are shown in Fig. 9.

The wavenumber range for the two curves from the low temperature set (253.2 and 272.1 K) employed in this analysis only extend up to 2697 cm^{-1} , but the analysis of ACE-FTS spectra requires calculations of the N_2 continuum up to ~ 2740 cm^{-1} . Fitting only the five highest temperature curves above 2697 cm^{-1} yields excessively noisy results in the absence of smoothing constraints. Therefore, results above 2697 cm^{-1} in Fig. 9 (shown in orange) are derived from linear extrapolation of $\ln(B^0_{N_2-N_2}(\sigma))$ and $\beta^0_{N_2-N_2}(\sigma)$ using the results between 2650 and 2695 cm^{-1} for the two parameters. The empirical parameters for the high wavenumber region are presented in Table A2 in Appendix A.

Unfortunately, this approach leads to a minor discontinuity in calculated N_2-N_2 CIA at 2610 cm^{-1} for temperatures less than ~ 250 K (recall the lowest temperature employed in the high wavenumber region was 253.2 K). An extra step is therefore required to ensure continuity between the two regions at lower temperatures. The calculated curve for wavenumbers above 2610 cm^{-1} are scaled as follows:

$$B^{scaled}(\sigma, T) = \frac{B^{low}(2610 \text{ cm}^{-1}, T)}{B^{high}(2610 \text{ cm}^{-1}, T)} B^{high}(\sigma, T), \quad (3)$$

where B^{low} is the calculated N_2-N_2 CIA normalized absorption coefficient at the given temperature for the low wavenumber region (2130–2610 cm^{-1}), using the empirical function in Eq. (2) as described in Section 2.2, and B^{high} is the calculated value at the given

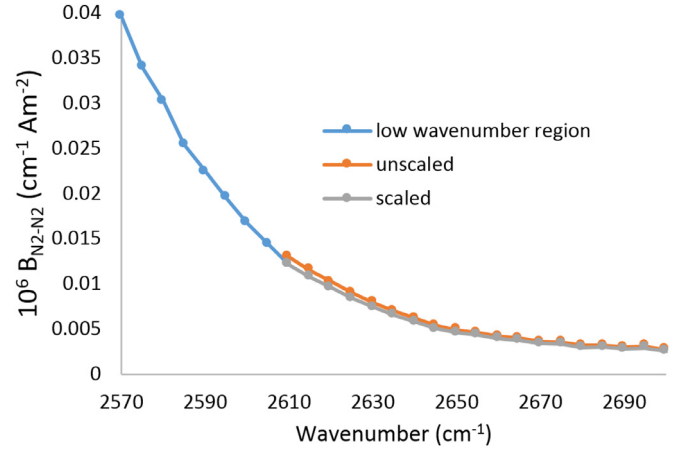


Fig. 10. The N_2-N_2 CIA normalized absorption coefficient for temperature 200 K, calculated using the expression in Eq. (3). The high wavenumber portion of the curve (above 2610 cm^{-1}) is scaled to avoid discontinuity.

temperature in the high wavenumber region (2610–2750 cm^{-1}), making use of the empirical function in Eq. (1) as described in this section. Fig. 10 displays the effect of this scaling on the calculated N_2-N_2 CIA normalized absorption coefficient at temperature 200 K.

2.4. Calculation for N_2 -Air

We now have the capacity to calculate the N_2-N_2 CIA normalized absorption coefficient for any temperature relevant to the low Earth atmosphere and wavenumbers between 2130 and 2750 cm^{-1} , with the caveat that temperatures below 228.2 K represent an extrapolation. However, the Earth does not consist of pure nitrogen. Dry air atmospheric composition is 78.09% N_2 , 20.95% O_2 , 0.93% argon, 0.04% carbon dioxide and trace amounts of other gases. The contribution to the calculation from other atmospheric constituents must be taken into account.

Ideally, independent measurements would be used to calculate the contributions from other collision partners (e.g., O_2 and argon), but such data are not currently available. The CIA normalized absorption coefficient for N_2 in air is

$$B_{N_2-Air}(\sigma, T) = 0.7809 B_{N_2-N_2}(\sigma, T) + 0.2095 B_{N_2-O_2}(\sigma, T) + 0.0093 B_{N_2-Ar}(\sigma, T) + \dots \quad (4)$$

Lafferty et al. [20] assume a relative collision efficiency between N_2 and O_2 as collision partner as follows:

$$B_{N_2-O_2}(\sigma, T) = B_{N_2-N_2}(\sigma, T) * \left[1.294 - 0.4545 \frac{T}{T_0} \right], \quad (5)$$

where T_0 is taken as 296 K. The above expression assumes no difference in wavenumber dependence for the two collision partners, although Lafferty et al. [20] characterize the relative efficiency as being “practically” independent of wavenumber, which implies the simple form in Eq. (5) may introduce errors in the shape of the curve.

Lafferty et al. [20] ignored argon but assumed an atmospheric composition with 79% N_2 , thereby implicitly assuming argon was an equivalent collision partner to N_2 , with no differences in shape as a function of wavenumber or temperature dependence. For the current study, a similarly rough correction is applied via a simple scaling factor with no temperature or wavenumber dependence, accounting for the N_2 -Ar contribution by applying a 1 percent increase in the calculated contributions from N_2 - N_2 plus N_2 - O_2 .

$$B_{N_2-Air}(\sigma, T) \approx 1.01 * [0.7809B_{N_2-N_2}(\sigma, T) + 0.2095B_{N_2-O_2}(\sigma, T)]. \quad (6)$$

It has been shown that H_2O collisions with N_2 can contribute significantly near the surface, where water concentrations are largest [22]. The effects of N_2 - H_2O collisions will not be included in the current study.

Thus, the absorption coefficient for N_2 CIA in the atmosphere is calculated as follows:

$$\alpha(\sigma, P, T) = 1.01 * \left(\frac{P}{P_0} \frac{273}{T}\right)^2 \left(0.8215 - 0.074356 \frac{T}{T_0}\right) B_{N_2-N_2}(\sigma, T), \quad (7)$$

where P_0 is 1 atm, the pressure at standard temperature and pressure (STP), 273 K is temperature at STP, and T_0 is again taken as 296 K. $B_{N_2-N_2}(\sigma, T)$ is calculated as described in Section 2.2 for wavenumbers between 2130 and 2610 cm^{-1} and as described in Section 2.3 for wavenumbers between 2610 and 2740 cm^{-1} . Recall that the scaling factor of 1.01 in Eq. (7) is applied to account for the N_2 -Ar contribution. It was kept separate from the other numerical factors in order to investigate its effect in the analysis of ACE-FTS spectra. It was found that a slightly larger scaling factor improved agreement between tangent heights determined from the N_2 continuum compared to tangent heights generated from CO_2 , but that may have been compensating for other errors (e.g., errors in the CO_2 spectroscopic parameters) rather than improving the calculated N_2 continuum. For the analysis of ACE-FTS, a scaling factor of 1.015 (instead of 1.01) was employed to obtain slight improvement in the internal consistency with CO_2 .

As a measure of the sensitivity of the analysis to this scaling factor, a change of roughly one percent (e.g., from a value of 1.015 to 1.025) imparts a difference of 30 to 35 m in derived tangent height.

Note that pressure in Eq. (7) is implicitly dry-air pressure, since we have assumed dry-air values for the volume mixing ratios of the various constituents.

From the absorption coefficient in Eq. (7), the spectrum is calculated as follows:

$$I(\sigma) = I_0(\sigma) e^{-\int \alpha(\sigma, P(z), T(z)) dz}, \quad (8)$$

where $I_0(\sigma)$ is the calculated gas phase spectrum. The integration in Eq. (8) is performed along the path traveled by the light ray. If the light ray travels through a region of constant pressure and temperature, the argument of the exponential is simply $-\alpha(\sigma, P, T) * L$, where L is the distance traveled through the medium, the standard Beer's law expression.

2.5. Altitude grid for forward model calculations

Calculating a spectrum to analyze limb geometry measurements like those from the ACE-FTS involves forward model calculations:

tracing the path of a solar ray as it travels through the Earth's atmosphere and summing the total absorption experienced during transit. Integrating the calculated signal is discretized by dividing the atmosphere into layers, where pressure and temperature within a layer are assumed constant, and summing the contributions from all layers through which the solar ray passed. The integral in Eq. (8) is thus approximated as:

$$\int \alpha(\sigma, P(z), T(z)) dz \approx \sum_i \alpha(\sigma, P_i, T_i) L_i, \quad (9)$$

where P_i and T_i are the pressure and temperature, respectively, within layer i , and L_i is the distance traveled through the layer.

Forward model calculations for analyzing gas phase lines in ACE-FTS spectra are performed on a 1 km altitude grid. Previous studies of N_2 CIA in ACE-FTS measurements [8–10] retained that altitude grid for calculating the CIA spectrum. Note in Eq. (7) that the absorption coefficient for N_2 CIA varies as the square of the pressure. Atmospheric pressure varies exponentially with altitude, and this strong variation with pressure causes a significant loss in accuracy for N_2 CIA calculated on a 1 km grid. A finer altitude grid of 100 m is therefore suggested for the calculation.

Fig. 11 shows the sort of discrepancies one can expect from using a 1 km grid in the wavenumber range of greatest interest for ACE-FTS spectra. For a measurement toward the top of a layer on the 1 km grid, such as the measurement at tangent height 13.6 km in occultation sr10063 in Fig. 11a, calculating the transmittance using the 1 km altitude grid overestimates the N_2 continuum, as seen in the ratio of the curves calculated on the two different altitude grids in Fig. 11b. Conversely, the calculated transmittance for a measurement with tangent height near the bottom of the layer will be underestimated when using a 1 km altitude grid, as seen for the measurement at tangent height 13.1 km in occultation sr10063 in Fig. 11c, where the ratio of the two calculated curves is presented in Fig. 11d.

Deficiencies in the calculation on the 1 km grid arise because the pressure at the center of the layer is not representative of the average value experienced by the solar ray as it traverses the layer. This is true even for calculation of gas phase lines, which is why version 4 of the ACE-FTS processing software employs a 100 m altitude sub-grid within the 1 km grid tangent layer (the lowest layer traversed by the solar ray) when fitting gas phase lines. The effect is amplified in the N_2 continuum calculation because of the quadratic pressure dependence. We therefore employ a 100 m altitude grid for N_2 continuum calculations over the full altitude range (from the tangent height up to 100 km), not just within the tangent layer on the 1 km grid as we do for forward model calculations for gas phase lines. The improved calculation fidelity from switching to the 100 m altitude grid significantly reduces scatter in tangent heights determined from the N_2 continuum.

3. Analysis of ACE-FTS spectra

3.1. Residual spectra

ACE-FTS version 3.6 results can be used to remove all known contributions to the spectrum in the vicinity of the N_2 continuum, making it easier to evaluate needs or challenges associated with analyzing this spectral region. “Residual spectra” are generated by dividing an ACE-FTS measurement by a gas phase spectrum calculated from version 3.6 results, and then averaging all data points in 2 cm^{-1} wide bins for which the calculated transmittance is greater than 0.8 (to emphasize the baseline and avoid division by zero for saturated lines). The residual spectrum thus contains all contributions to the spectrum not included in the calculation, such as aerosol extinction, far wing absorption from the nearby strong CO_2 ν_3 band, and absorption from the N_2 continuum.

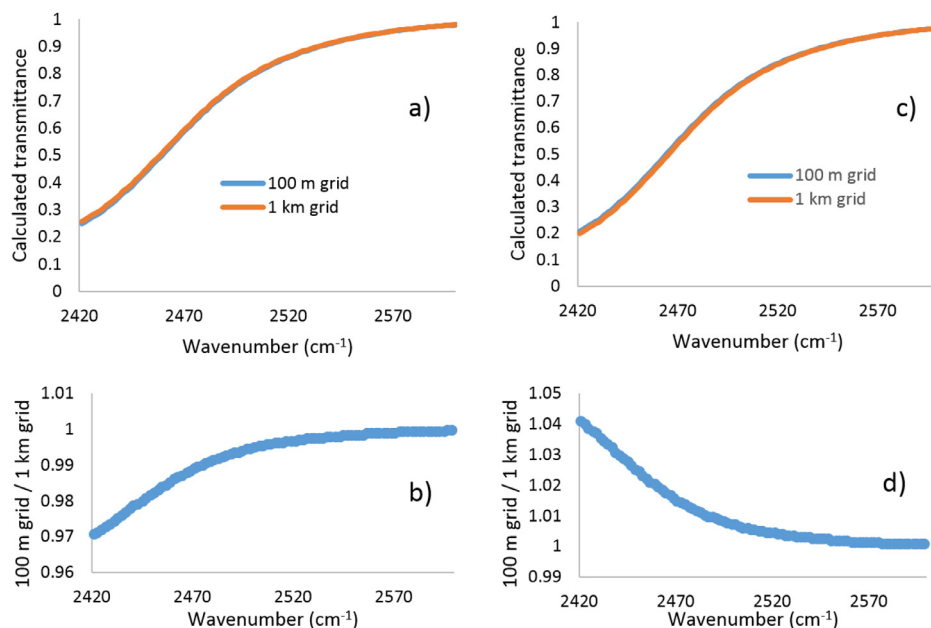


Fig. 11. (a) The calculated N₂ continuum transmittance for tangent height 13.6 km in occultation sr10063 using a 100 m altitude grid (in blue) and a 1 km altitude grid (in orange). (b) The ratio of the calculations on the two altitude grids for the measurement at 13.6 km. (c) The calculated N₂ continuum transmittance for tangent height 13.1 km in occultation sr10063 using a 100 m altitude grid (in blue) and a 1 km altitude grid (in orange). (d) The ratio of the calculations on the two altitude grids for the measurement at 13.1 km.

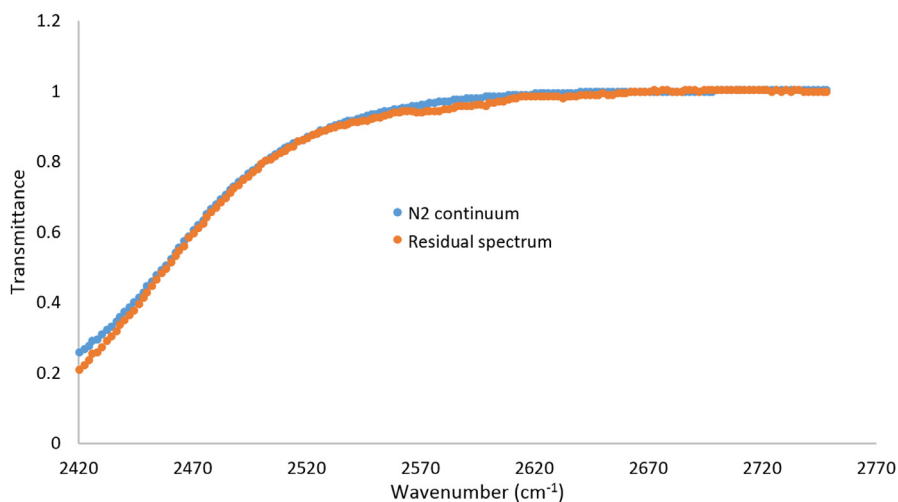


Fig. 12. Residual spectrum (in orange) for the measurement with tangent height 13.6 km from occultation sr10063, along with the N₂ continuum transmittance (in blue) calculated from ACE-FTS v3.6 geometry and pressure and temperature profiles.

Fig. 12 shows the residual spectrum for the measurement at tangent height 13.6 km from occultation sr10063, along with the calculated N₂ continuum transmittance for the measurement, based on the tangent height and the pressure and temperature profiles from ACE-FTS version 3.6 processing. A scaling was applied to the calculated N₂ continuum to match the baseline near 2740 cm⁻¹. The agreement between the two curves is fairly good, although a slight high bias in the N₂ continuum curve suggests that the tangent height generated from this spectral feature will be slightly lower than that determined from CO₂ in version 3.6 processing. There are, however, two problems clearly evident in Fig. 12 that impact the retrieval strategy.

The first problem is the divergence of the two curves at the low wavenumber end of the plot in Fig. 12. In this region, there is a contribution to the spectrum (changing rapidly with wavenumber) associated with the nearby CO₂ ν_3 band, a well-known effect [23]. Unfortunately, there is no means to readily calculate this contribution to the spectrum sufficiently accurately for our purposes, and so we simply avoid this region of the spectrum, limiting the analysis region to be above 2497 cm⁻¹. Because the N₂ continuum signal is increasing in the spectral region below 2497 cm⁻¹, there is potential to significantly improve the analysis if an accurate calculation for the CO₂ far wing contribution to this spectral region becomes available.

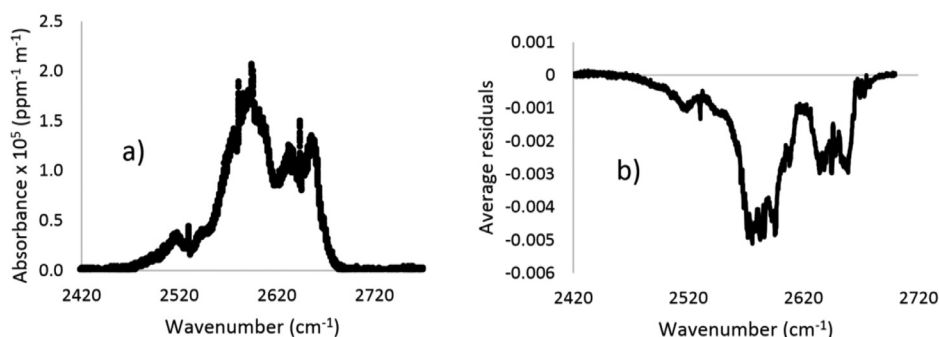


Fig. 13. (a) HNO₃ absorbance from Pacific Northwest National Laboratory. (b) Average residuals from ACE-FTS spectra from roughly 50 occultations for wavenumbers greater than 2565 cm⁻¹, and a scaled version of the PNNL curve for wavenumbers less than 2565 cm⁻¹.

The second problem visible in Fig. 12 is a collection of broad, weak spectral features near the center of the region, with peak absorption of the strongest feature around 2580 cm⁻¹, indicating something missing in the calculated spectrum. There is a set of weak HNO₃ bands in this spectral region that are missing from the HITRAN line list. Fig. 13a shows the HNO₃ absorbance in the region provided by the Pacific Northwest National Laboratory (PNNL) [24], while Fig. 13b shows the average residuals from a set of ACE-FTS spectra for wavenumbers greater than 2565 cm⁻¹. Residuals below that point were excessively noisy and were replaced by a scaled version of the PNNL curve. There are some differences in the shapes of the spectra in Fig. 13a and b that arise from the differences in resolution (0.1 cm⁻¹ for PNNL versus 0.02 cm⁻¹ for the ACE-FTS) and the fact that the relative intensities among the various bands depend on temperature, but the missing contribution to the calculated spectrum evident in the ACE-FTS residuals clearly comes primarily from HNO₃.

HNO₃ poses a greater problem than CO₂ far wing absorption because one cannot exclude the spectral region containing HNO₃ from the analysis while still providing adequate coverage of the N₂ continuum spectral feature. No spectroscopic constants are available for HNO₃ in this region, but it is possible to roughly account for the contribution from HNO₃ using the curve in Fig. 13b. HNO₃ volume mixing ratio (VMR) peaks in the stratosphere, and so the shape of the spectrum should remain similar for measurements at low altitudes, i.e., below the VMR peak. The HNO₃ transmittance spectrum in a particular measurement can therefore be approximated by

$$HNO_3 \text{ transmittance} \approx 1 - t_{HNO_3} * I_{HNO_3}(\sigma), \quad (10)$$

where σ is wavenumber, $I_{HNO_3}(\sigma)$ is the spectrum in Fig. 13b, and t_{HNO_3} is an empirical parameter whose value is fitted from the spectrum. When calculating the spectrum, the expression on the right hand side in Eq. (10) is multiplied by the right hand side of Eq. (8).

This is a very rough approximation that ignores the changing relative intensity among the different HNO₃ bands with temperature, and changes in spectral shape as a function of altitude, but it is better than simply ignoring the contribution of HNO₃ to the spectrum, which is currently the only other option. Spectroscopic constants for calculating HNO₃ in this spectral region would greatly simplify and improve the analysis.

3.2. Tangent height determination

While it was convenient to evaluate the N₂ continuum analysis in residual spectra, tangent height determination occurs near the beginning of the processing chain for an occultation, before all

of the information required to generate residual spectra is available. We must therefore develop an analysis approach for the “raw” transmittance spectra.

The analysis of ACE-FTS spectra involves the use of microwindows, a collection of narrow spectral regions containing information on the target molecule with minimal contributions from other molecules. In this case, microwindow selection emphasized information on the apparent baseline, to best track the broad N₂ continuum spectral feature. Regions with strong H₂O absorption were avoided because water lines tend to have large residuals in ACE-FTS spectra [25]. The set of microwindows employed in the determination of tangent heights from ACE-FTS spectra are provided in Table 2. The microwindows at the highest wavenumbers contain little contribution from the N₂ continuum, which provides a means for calibrating the baseline (i.e., either accounting for aerosol contributions to the spectrum or for small offsets in transmittance calibration). Note that a number of contiguous microwindows could in theory be combined, but they were chosen as shown in order to accommodate the memory configuration of the pressure/temperature retrieval software, which expects relatively narrow microwindows.

The interferers in the microwindow set are N₂O, CH₄, CO₂, O₃, and HDO. VMR profiles need to be fitted for these molecules in order to minimize their impact on the results. There are also N₂ quadrupole lines in the microwindows, but the VMR profile for this molecule is a fixed quantity (a constant 0.7809 as a function of altitude) and is not adjusted in the analysis.

Pressure and temperature profiles at low altitude were fixed to outputs from the analysis run of the weather service model at the Canadian Meteorological Center [26,27], which provides data up to roughly 30 km.

The measurements are analyzed in an “onion-peeling” type approach [28], starting at a measurement well above the altitude range of interest (around 25 km), fitting for the tangent height, a scaling factor for the baseline, the empirical scaling factor for HNO₃ (t_{HNO_3} from Eq. (10)), and scaling factors times the input guess VMR profiles for all of the interferers. Processing then proceeds sequentially downward in altitude, determining the tangent height for each measurement, the baseline and HNO₃ scaling factors, and the VMR profiles of the interferers for the altitude range between the measurement and the measurement directly above it, with the VMR profiles above that fixed to the results of previous steps, as is the standard approach with the onion-peeling method.

Fig. 14 shows the fitting results for a particular measurement (at tangent height 13.6 km in occultation sr10063). A number of lines clearly do not fit perfectly, in part due to the shape of the VMR profile above the highest analyzed measurement being not quite right for this occultation. Points below 0.85 times the max-

Table 2
Microwindow set for analysis of the N₂ continuum.

Center (cm ⁻¹)	Width (cm ⁻¹)	Lower Limit (km)	Center (cm ⁻¹)	Width (cm ⁻¹)	Lower Limit (km)
2497.80	0.80	5	2575.45	0.28	5
2498.85	1.30	5	2580.38	0.24	5
2500.10	1.20	5	2592.45	0.70	5
2501.05	0.70	5	2595.10	0.40	5
2501.95	1.10	5	2603.20	0.60	5
2502.90	0.80	5	2611.40	0.60	5
2503.80	0.60	5	2612.60	0.80	12
2505.15	0.30	5	2616.00	1.20	5
2505.90	1.20	5	2620.80	0.60	5
2506.75	0.50	5	2626.65	0.90	5
2507.65	1.30	5	2636.63	0.34	5
2508.85	1.10	5	2644.75	0.30	12
2510.05	1.30	5	2650.60	1.00	5
2511.00	0.60	5	2653.50	1.00	5
2513.40	0.60	5	2662.75	0.70	12
2516.90	0.80	5	2668.21	0.30	5
2518.90	1.00	5	2670.63	0.30	5
2521.67	0.50	5	2677.05	0.50	12
2524.00	0.60	5	2684.10	0.40	5
2528.24	0.36	5	2692.20	0.40	12
2531.62	0.44	5	2700.30	0.80	5
2538.85	0.50	12	2704.04	0.40	12
2539.85	0.50	5	2713.57	0.34	5
2545.87	0.26	5	2719.34	0.40	12
2552.40	0.40	5	2725.03	0.30	12
2560.42	0.20	5	2731.95	0.50	5
2561.37	0.22	5	2739.90	0.40	5
2569.41	0.30	12	2746.45	0.70	5

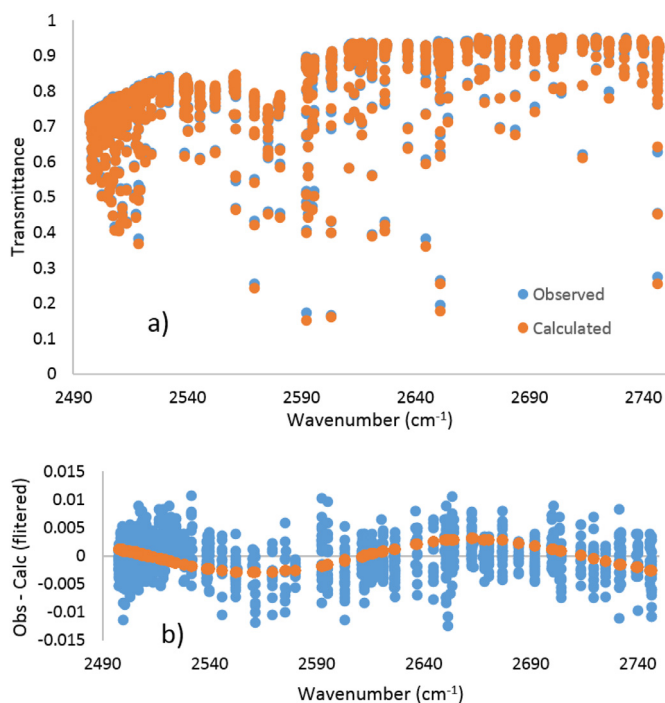


Fig. 14. (a) The observed and calculated spectra in the selected microwindows for tangent height 13.6 km from occultation sr10063. (b) In blue are the residuals (observed – calculated) for the fitting, filtering out all points less than 0.85 times the maximum value in the microwindow to focus on the baseline. In orange is a sine curve (generated by hand) that roughly matches the shape of the residuals.

imum transmittance in the microwindow are excluded from the residual plot in Fig. 14b in order to better see how well the fitted N₂ continuum tracks the baseline, which is the target of the analysis. The residuals show possible evidence of channeling effects in

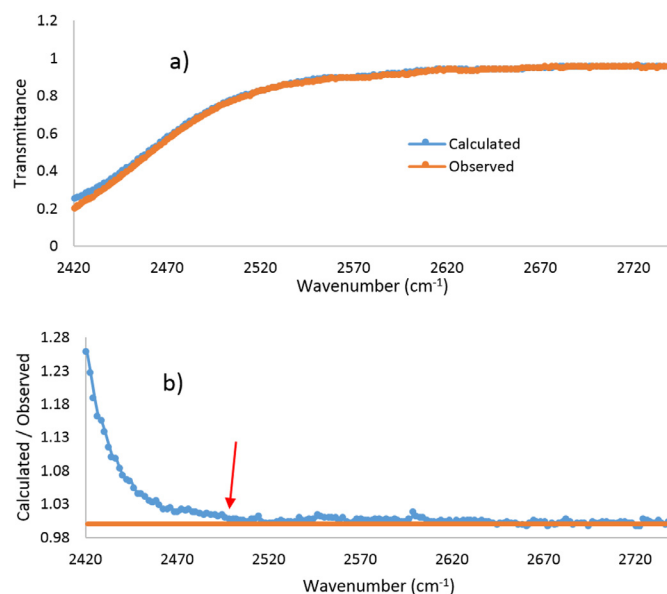


Fig. 15. (a) The residual spectrum (in orange) plus the calculated N₂ continuum and fitted HNO₃ (in blue) from the ACE-FTS version 4 analysis for the measurement with tangent height 13.6 km in occultation sr10063. (b) The ratio of the two curves (in blue), with a line at 1.0 (in orange) shown for reference. The arrow indicates the lower wavenumber limit of the microwindows employed in the analysis.

the spectrum, exhibiting the classical sinusoidal pattern associated with such effects, as indicated by the orange curve in Fig. 14a. This could be accounted for by fitting for a sinusoidal term multiplying the baseline.

However, one can use the version 4 fitting results to generate residual spectra and evaluate the behavior outside the range of the microwindows employed in the analysis. As can be seen in Fig. 15, there may be small contributions from CO₂ far wing absorption

in the microwindows. The arrow in Fig. 15b indicates the lower wavenumber limit of the microwindows, and recall that the divergence in the two curves in Fig. 15a is due primarily to CO₂ far wing absorption missing in the calculation. A visual inspection of the blue curve in Fig. 15b, the ratio of the residual spectrum and the fitted contribution from N₂ continuum plus HNO₃, suggests that low levels of CO₂ far wing absorption may extend into the range of microwindows. It is possible the apparent sinusoidal feature in the residuals is a coincidence, a consequence of the fitted N₂ continuum and HNO₃ compensating for the missing information in the calculation (i.e., CO₂ far wing absorption), in which case it would be inappropriate to fit for a sinusoidal channeling term. This issue will be investigated for future processing versions.

As mentioned previously, a baseline scaling is applied in the analysis, in part to account for aerosol extinction in the region, implicitly assuming that aerosol effects are flat and featureless in the vicinity of the N₂ continuum. Investigation of various types of aerosols that sometimes appear in ACE-FTS measurements (e.g., polar stratospheric clouds, cirrus clouds, sulphate aerosols resulting from volcanic eruptions, etc.) yielded no indication of significant spectral structure expected in this wavenumber region. However, over the ~ 250 cm⁻¹ range of the N₂ continuum microwindows, these aerosols will generally exhibit a slow variation of extinction with wavenumber. The simplest means to account for this would be to determine a linear term for the apparent baseline (a “baseline slope”).

Unfortunately, including a baseline slope term in the analysis introduced a large offset in retrieved tangent heights (roughly 200 m near 18 km tangent altitude), even in the absence of significant aerosol extinction, suggesting that the term was perhaps compensating for problems in the N₂ continuum calculation, thereby imparting a systematic error in retrieved tangent heights. This tangent height offset generated a step in retrieved VMR profiles for all molecules near the boundary between the altitude region where tangent heights are generated from the N₂ continuum (below 18 km) and the altitude region where tangent height separations are determined from CO₂ (above 18 km).

Note that the boundary of 18 km was chosen based on where the N₂ continuum signal dropped low enough to start causing minor problems in some occultations. This boundary may be pushed higher in future processing versions, particularly if the means to accurately calculate the contribution from CO₂ far wing absorption becomes available.

While the observed step in retrieved VMRs near 18 km could be at least partly a consequence of problems in the spectroscopic constants for CO₂ lines employed in the analysis above 18 km, the fact that the baseline slope term imparts such a large change (200 m in tangent height in the absence of obvious aerosol contributions to the spectra) is troubling. We therefore exclude this term from the analysis, which means we are left with a simple baseline scaling term to account for aerosol extinction in the vicinity of the N₂ continuum.

At lower altitudes (below about 7 km), fitting residuals increased significantly as a consequence of removing the baseline scaling term. Also, occultations containing very large aerosol extinction (e.g., from polar stratospheric clouds or volcanic sulphate aerosols) may suffer errors in tangent height determination, the magnitude of which will depend on the properties and quantity of aerosol particles. Future processing versions will try to incorporate a baseline slope term to improve results for such occultations, but that may involve finding improvements for the N₂ CIA calculation, for example perhaps having a more accurate calculation for the N₂-O₂ contribution if it has a different shape than N₂-N₂ CIA.

Fig. 16 shows how well the fitted N₂ continuum plus HNO₃ reproduces the residual spectrum calculated from the ACE-FTS version 4 fitting results for the measurement at 6.9 km from oc-

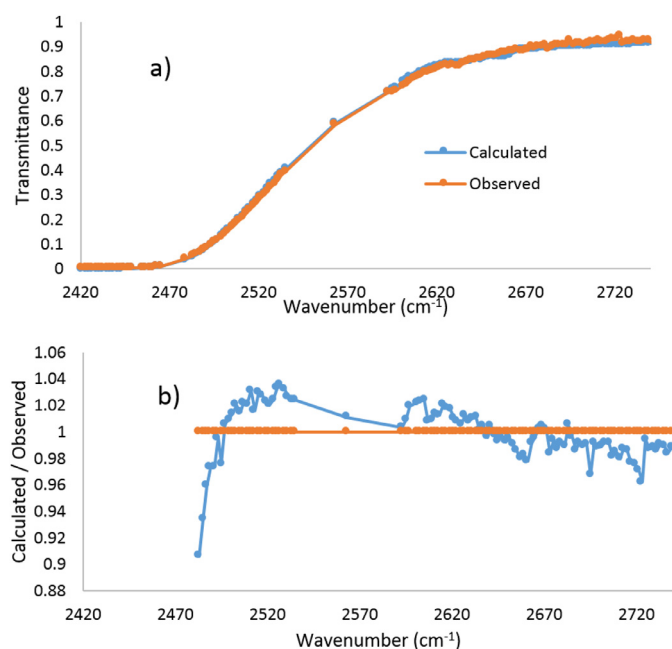


Fig. 16. (a) The residual spectrum (in orange) plus the calculated N₂ continuum and fitted HNO₃ (in blue) from the ACE-FTS version 4 analysis for the measurement with tangent height 6.9 km in occultation sr10063. Gaps in the plot arise from bins where there were no data points above 0.8 in the calculated transmittance. (b) The ratio of the two curves (in blue), with a line at 1.0 (in orange) shown for reference.

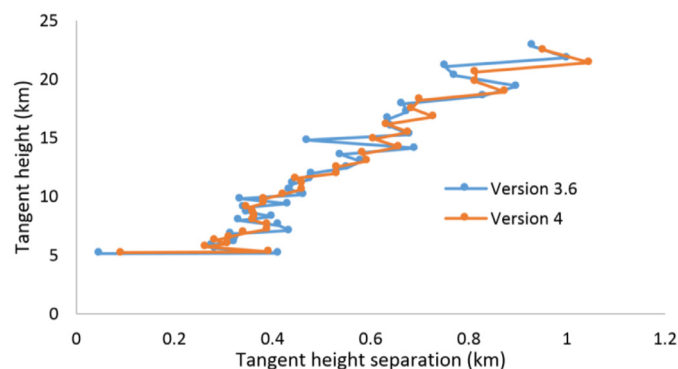


Fig. 17. The separations between subsequent tangent heights for occultation sr10063.

cultation sr10063. A baseline slope term would significantly improve the residuals. Note, however, that the ratio plot in Fig. 16b for wavenumbers above ~ 2650 cm⁻¹ is relatively flat (ignoring the structure that comes primarily from elevated residuals for H₂O lines in the region), further evidence that the baseline slope term was not necessarily accounting for wavenumber variation of aerosols in the spectrum, but was instead compensating for errors in the calculation (e.g., the previously mentioned possibility of problems in the shape of the N₂-O₂ contribution). The baseline term may also have been compensating for the missing CO₂ far wing absorption, which could be stronger at these lower altitudes.

The missing N₂-H₂O CIA contribution in the calculated signal does not explain the systematic residuals in Fig. 16. A rough estimate (based on information provided in Hartmann et al. [29]) of the contribution of N₂-H₂O CIA to the measurement indicates a peak absorption in the microwindow set of roughly one-third of a percent, much smaller than the observed residuals. For

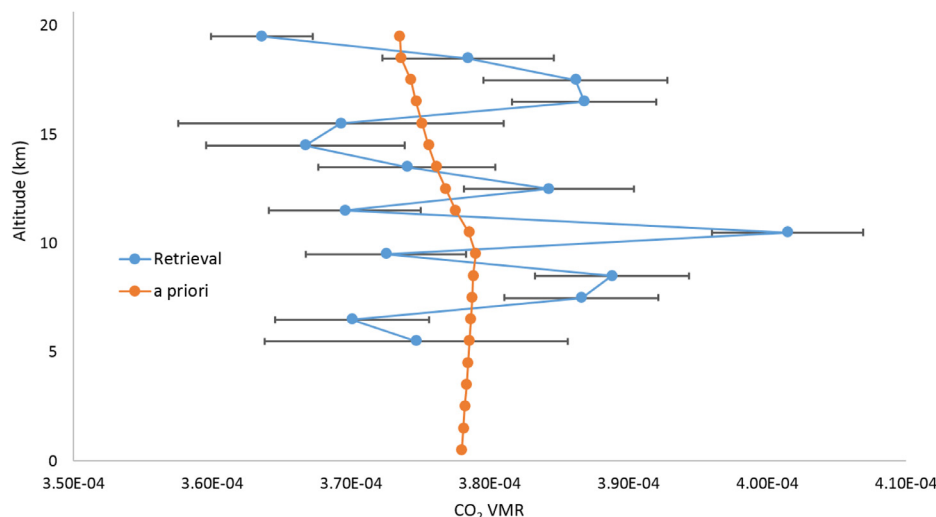


Fig. 18. ACE-FTS version 4 retrieval of low altitude CO_2 for occultation sr10063 (in blue), along with the expectations from an *a priori* profile (in orange) generated for the date and location of the occultation. Error bars are 1-sigma statistical least squares fitting errors.

the lowest measurement (tangent height ~ 5.2 km) in this occultation (sr10063), the peak absorption was estimated to be less than one percent. This missing contribution to the calculated spectrum would equate to a low bias in the derived tangent height of roughly 25 m for the measurement near 5.2 km. Note that this occultation has moderate levels of H_2O (relative to other measured ACE-FTS occultations). For tropical occultations with significantly higher H_2O VMR, it is estimated that N_2 - H_2O absorption could range as high as 5 to 10% in extreme cases for tangent heights near 5 km. This would lead to errors in derived tangent height of up to 250 m, much larger than our target accuracy of 50 to 100 m. This effect declines with increasing tangent height, thanks to the rapid change of H_2O VMR with altitude in the troposphere. Future processing versions will look to incorporate N_2 - H_2O CIA into the analysis.

The ACE-FTS suntracker mirror controls instrument pointing by finding the center of radiance of the sun [3]. In the absence of thick clouds or aerosol layers that can cause jumps in the pointing, tangent height separations between successive measurements (i.e., the altitude difference between the tangent height for a given measurement and the tangent height for the measurement just above it) should vary relatively smoothly with altitude. Plotting tangent height separations as a function of tangent height, as is presented in Fig. 17, provides a means to evaluate how smoothly the tangent heights are varying. Note that the separation between tangent heights decreases with decreasing altitude in this plot due to refraction effects, which get more pronounced with increasing atmospheric density. It is clear from Fig. 17 that the variability in tangent heights determined from the N_2 continuum is reduced compared to the tangent heights from the previous ACE-FTS processing version, which were determined from CO_2 .

4. Low-altitude CO_2 retrievals

As mentioned previously, employing the N_2 continuum to generate tangent heights at low altitude from ACE-FTS spectra (rather than CO_2) opens up the possibility of retrieving CO_2 at low altitude (below 18 km). Recall that the pressure and temperature information below 18 km is taken from the analysis run of the Canadian weather service model. Many of the available CO_2 lines (i.e., unsaturated and not too highly blended with spectral features from other molecules) for this altitude region have large lower state en-

Table 3

Microwindow set for low altitude CO_2 retrievals from the ACE-FTS.

Center (cm^{-1})	Width (cm^{-1})	Lower Limit (km)	Upper limit (km)
1986.09 ^a	0.30	10	20
2620.81 ^b	0.46	12	20
3160.15	0.50	5	20
3161.62	0.40	5	20
3204.70	0.40	5	20
3206.36	0.40	5	20
3301.20 ^a	0.40	5	10
3301.50	0.40	5	20
3315.08 ^c	0.40	5	10
3330.02	0.28	12	20
3342.93 ^c	0.30	10	20
3344.80	0.60	5	20
3350.50 ^d	0.40	5	20
3377.04	0.24	10	20

^a Outside microwindow to improve results for interferer H_2^{17}O .

^b Outside microwindow to improve results for interferer ^{18}OCO .

^c Outside microwindow to improve results for interferer H_2O .

^d Outside microwindow to improve results for interferer H_2^{18}O .

ergies, making them very sensitive to temperature. Small errors in temperature for data from the weather service model can lead to significant errors in retrieved temperature for CO_2 lines with strong temperature sensitivities, and so such lines were avoided as much as possible in the selected microwindow set.

The microwindows employed for low altitude CO_2 retrievals from the ACE-FTS are presented in Table 3. The signal-to-noise ratio for the microwindows in Table 3 that contain CO_2 is generally low (around 100:1) compared to the CO_2 microwindows employed the pressure/temperature analysis and high altitude CO_2 retrievals (300:1 to 400:1), and so some mild averaging of retrieved low altitude CO_2 VMR profiles might be beneficial for scientific analysis. The interferers in this microwindow set, retrieved simultaneously using a global analysis approach [4], are H_2O , H_2^{17}O , H_2^{18}O , $^{13}\text{CO}_2$, ^{18}OCO , O_3 , N_2O , and HCN . Note that version 4 results are reported up to 18 km (the upper altitude limit for which tangent heights are determined from the N_2 continuum), but the retrievals extend up to 20 km, providing a small overlap that enables detection of discontinuities between the two altitude regions (above and below 18 km).

Fig. 18 shows the retrieved low altitude CO₂ VMR profile for occultation sr10063. A model that accounts for variations of CO₂ with time, location, season and altitude [30] is used to calculate an *a priori* VMR profile for the molecule that is employed in the retrieval of pressure and temperature above 18 km. The errors bars in Fig. 18 are the 1-sigma statistical errors from the least squares fitting process. The retrieved profile agrees well with the expectations from the model, typically within 2-sigma error bars (excluding the point at 10.5 km). The error bars in Fig. 18 are somewhat large (compared to typical ACE-FTS results for main-isotopologue targets) as a consequence of the relatively low signal to noise ratio for the microwindows employed in the retrieval and the small number of microwindows. Improved precision can be achieved, however, through averaging results from multiple occultations. An assessment of low altitude CO₂ from ACE-FTS version 4.0 retrievals will be reported in a future publication.

5. Conclusions

A process has been developed for determining ACE-FTS measurement tangent heights from the N₂ collision induced absorption continuum near 2500 cm⁻¹. It makes use of high temperature N₂-N₂ CIA normalized absorption coefficients that were not employed in previous studies of the N₂ continuum in ACE-FTS spectra. This provided improved temperature dependence in the calculation of the N₂ continuum. With the high temperature absorption coefficients included in the analysis, small corrections were applied to the curves for the low temperature set, and a refinement was made to the empirical function employed to account for temperature dependence in the calculation.

The current analysis accounted for HNO₃ absorption in this spectral region, which has not been done in previous studies of the N₂ continuum. It also employed a finer altitude grid in forward model calculations, which represents a significant calculation accuracy improvement over previous studies.

This approach has been implemented in the ACE-FTS version 4.0 processing software. With no information from CO₂ employed in the determination of tangent heights below 18 km from ACE-FTS measurements, independent retrievals of CO₂ VMR profiles in that altitude region become possible and will be included as a standard product in the upcoming ACE-FTS version 4.0 data set.

Acknowledgments

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Appendix

The parameters for the empirical function (in Eq. (2)) used to describe the temperature dependence of the N₂-N₂ collision induced absorption normalized absorption coefficients for the wavenumber range 2130–2610 cm⁻¹ are presented in Table A1. Excess significant digits are supplied to guard against rounding errors.

The parameters for the empirical function (in Eq. (1)) used to describe the temperature dependence of the N₂-N₂ collision induced absorption normalized absorption coefficients for the wavenumber range 2610–2750 cm⁻¹ are presented in Table A2. Excess significant digits are supplied to guard against rounding errors. The curve derived from these parameters should be scaled according to Eq. (3) in order to avoid a discontinuity with the curve below 2610 cm⁻¹ calculated from the information in Table A1.

Table A1

Parameters for the empirical function (in Eq. (2)) describing the temperature dependence of the N₂-N₂ collision induced absorption normalized absorption coefficients for the wavenumber range 2130–2610 cm⁻¹.

Wavenumber (cm ⁻¹)	$B_{N_2-N_2}^0$ (cm ⁻¹ Am ⁻¹)	$\beta_{N_2-N_2}^0$ (K)	$\delta_{N_2-N_2}^0$ (K ²)
2130	5.879E-08	793.37	336,862
2135	6.900E-08	742.81	263,744
2140	7.951E-08	722.43	241,379
2145	9.149E-08	695.58	206,169
2150	1.059E-07	658.19	174,091
2155	1.218E-07	623.20	148,504
2160	1.400E-07	587.70	121,132
2165	1.604E-07	555.11	103,421
2170	1.825E-07	521.78	85,140
2175	2.079E-07	488.22	69,935
2180	2.350E-07	454.24	56,747
2185	2.646E-07	419.01	45,935
2190	2.976E-07	383.15	36,793
2195	3.314E-07	347.78	30,426
2200	3.693E-07	311.57	24,773
2205	4.085E-07	276.13	19,432
2210	4.499E-07	241.18	16,464
2215	4.927E-07	206.07	11,999
2220	5.358E-07	173.12	10,790
2225	5.808E-07	138.84	9587
2230	6.231E-07	108.05	7650
2235	6.664E-07	78.58	7756
2240	7.092E-07	50.44	6967
2245	7.466E-07	26.73	8001
2250	7.850E-07	4.22	9261
2255	8.187E-07	-15.90	9942
2260	8.485E-07	-32.56	11,948
2265	8.769E-07	-46.77	13,541
2270	9.037E-07	-57.59	15,104
2275	9.295E-07	-62.73	18,426
2280	9.558E-07	-62.53	22,596
2285	9.902E-07	-62.44	24,653
2290	1.032E-06	-56.49	27,115
2295	1.088E-06	-49.26	28,330
2300	1.174E-06	-45.51	27,299
2302	1.214E-06	-44.45	26,906
2304	1.266E-06	-46.58	25,548
2306	1.320E-06	-48.51	23,994
2308	1.382E-06	-52.10	21,830
2310	1.453E-06	-59.19	20,169
2312	1.538E-06	-67.90	17,509
2314	1.630E-06	-81.10	15,118
2316	1.711E-06	-93.12	13,418
2318	1.774E-06	-103.46	12,438
2320	1.831E-06	-111.23	12,457
2322	1.897E-06	-120.20	10,421
2324	1.969E-06	-130.41	7819
2326	2.008E-06	-138.89	8048
2328	2.019E-06	-141.47	8856
2330	2.030E-06	-142.55	7391
2332	2.024E-06	-142.48	7239
2334	1.969E-06	-136.21	9013
2336	1.919E-06	-129.79	9380
2338	1.882E-06	-125.08	10,330
2340	1.839E-06	-120.79	10,646
2342	1.775E-06	-114.19	11,811
2344	1.694E-06	-106.72	14,523
2346	1.619E-06	-100.62	16,825
2348	1.555E-06	-96.13	19,297
2350	1.505E-06	-96.44	20,754
2352	1.467E-06	-99.21	22,913
2354	1.436E-06	-103.63	24,639
2356	1.411E-06	-107.49	25,584
2358	1.391E-06	-112.35	27,217
2360	1.377E-06	-117.31	27,201
2362	1.363E-06	-122.36	27,320
2364	1.351E-06	-126.77	26,811
2366	1.344E-06	-132.09	26,081
2368	1.344E-06	-136.57	24,691
2370	1.341E-06	-141.14	24,573
2372	1.337E-06	-144.88	22,304
2374	1.339E-06	-148.67	21,491
2376	1.346E-06	-153.48	17,737

(continued on next page)

Table A1 (continued)

Wavenumber (cm ⁻¹)	$B_{N_2-N_2}^0$ (cm ⁻¹ Am ⁻¹)	$\beta_{N_2-N_2}^0$ (K)	$\delta_{N_2-N_2}^0$ (K ²)
2378	1.347E-06	-156.02	16,590
2380	1.348E-06	-157.19	14,575
2382	1.357E-06	-159.36	12,204
2384	1.371E-06	-160.23	9690
2386	1.374E-06	-159.54	7675
2388	1.375E-06	-157.24	7380
2390	1.384E-06	-155.71	5775
2392	1.392E-06	-152.42	3968
2394	1.389E-06	-148.71	3273
2396	1.388E-06	-144.27	2736
2398	1.396E-06	-140.24	1695
2400	1.401E-06	-135.37	138
2402	1.392E-06	-128.90	266
2404	1.385E-06	-122.68	360
2406	1.387E-06	-116.81	-390
2408	1.381E-06	-109.99	-564
2410	1.365E-06	-102.02	497
2412	1.355E-06	-94.80	-104
2414	1.354E-06	-87.70	-40
2416	1.341E-06	-79.76	3
2418	1.320E-06	-70.93	-703
2420	1.306E-06	-61.74	-791
2422	1.296E-06	-53.40	-319
2424	1.273E-06	-43.13	760
2426	1.250E-06	-33.98	990
2428	1.234E-06	-24.54	455
2430	1.220E-06	-14.67	-100
2432	1.192E-06	-2.79	664
2434	1.167E-06	7.92	917
2436	1.148E-06	18.57	1544
2438	1.123E-06	30.51	2051
2440	1.093E-06	41.74	2562
2442	1.067E-06	53.00	2687
2444	1.046E-06	64.75	3711
2446	1.018E-06	76.89	4257
2448	9.868E-07	88.94	6104
2450	9.612E-07	100.82	7361
2452	9.357E-07	113.45	8078
2454	9.031E-07	126.29	9346
2456	8.749E-07	138.46	9791
2458	8.507E-07	150.67	11,650
2460	8.234E-07	163.60	13,517
2462	7.920E-07	176.94	15,290
2464	7.656E-07	189.66	18,750
2466	7.421E-07	201.69	17,954
2468	7.125E-07	215.10	20,249
2470	6.841E-07	228.22	23,523
2472	6.618E-07	241.02	24,741
2474	6.386E-07	253.79	25,257
2476	6.112E-07	264.66	26,992
2478	5.863E-07	278.04	30,462
2480	5.663E-07	290.01	31,426
2482	5.423E-07	300.47	34,731
2484	5.174E-07	312.17	36,574
2486	4.975E-07	323.79	37,824
2488	4.789E-07	334.64	40,355
2490	4.571E-07	344.27	43,271
2492	4.362E-07	354.28	47,836
2494	4.188E-07	365.32	49,259
2496	4.017E-07	373.61	48,076
2498	3.822E-07	381.74	55,171
2500	3.654E-07	391.60	59,015
2502	3.505E-07	401.06	60,253
2504	3.347E-07	407.54	65,079
2506	3.195E-07	414.56	68,051
2508	3.057E-07	423.62	70,308
2510	2.922E-07	431.50	75,662
2512	2.788E-07	435.98	77,033
2514	2.668E-07	441.49	77,546
2516	2.548E-07	451.51	85,071
2518	2.436E-07	455.65	82,929
2520	2.327E-07	458.25	92,032
2522	2.230E-07	464.32	90,466
2524	2.126E-07	469.48	96,934
2526	2.031E-07	475.40	94,467

(continued on next page)

Table A1 (continued)

Wavenumber (cm ⁻¹)	$B_{N_2-N_2}^0$ (cm ⁻¹ Am ⁻¹)	$\beta_{N_2-N_2}^0$ (K)	$\delta_{N_2-N_2}^0$ (K ²)
2528	1.950E-07	476.46	89,461
2530	1.860E-07	480.85	104,828
2532	1.779E-07	488.05	98,366
2534	1.702E-07	488.30	104,761
2536	1.632E-07	493.39	99,655
2538	1.564E-07	494.70	99,906
2540	1.493E-07	498.18	100,583
2542	1.431E-07	502.56	103,999
2544	1.371E-07	507.05	105,545
2546	1.315E-07	510.54	101,832
2548	1.258E-07	515.38	97,952
2550	1.207E-07	516.92	100,666
2555	1.088E-07	524.93	90,368
2560	9.828E-08	531.54	87,551
2565	8.871E-08	539.75	85,337
2570	8.037E-08	547.42	68,879
2575	7.289E-08	556.82	53,362
2580	6.582E-08	566.00	53,642
2585	5.982E-08	577.18	31,005
2590	5.413E-08	591.81	30,638
2595	4.903E-08	609.70	25,766
2600	4.446E-08	626.74	16,212
2605	4.018E-08	641.60	6819
2610	3.652E-08	666.64	-8678

Table A2

Parameters for the empirical function (in Eq. (1)) describing the temperature dependence of the N₂-N₂ collision induced absorption normalized absorption coefficients for the wavenumber range 2610–2750 cm⁻¹.

Wavenumber (cm ⁻¹)	$B_{N_2-N_2}^0$ (cm ⁻¹ Am ⁻¹)	$\beta_{N_2-N_2}^0$ (K)
2610	3.679E-08	643.96
2615	3.328E-08	654.51
2620	3.019E-08	664.77
2625	2.740E-08	683.79
2630	2.489E-08	700.86
2635	2.237E-08	716.61
2640	2.036E-08	728.35
2645	1.823E-08	745.81
2650	1.660E-08	750.31
2655	1.496E-08	723.10
2660	1.373E-08	728.83
2665	1.257E-08	704.91
2670	1.131E-08	701.09
2675	1.056E-08	668.43
2680	9.718E-09	687.32
2685	8.980E-09	636.51
2690	8.117E-09	614.43
2695	7.733E-09	560.80
2700	6.923E-09	577.13
2705	6.356E-09	558.86
2710	5.834E-09	540.60
2715	5.356E-09	522.34
2720	4.916E-09	504.08
2725	4.513E-09	485.81
2730	4.143E-09	467.55
2735	3.803E-09	449.29
2740	3.491E-09	431.03
2745	3.205E-09	412.76
2750	2.942E-09	394.50

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