Scattering and Absorption Cross-sections of Atmospheric Gases in the Ultraviolet-Visible Wavelength Range (307 - 725 nm)

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Abstract

Accurate Rayleigh scattering and absorption cross-sections of atmospheric gases are essential for understanding the propagation of electromagnetic radiation in planetary atmospheres. Accurate extinction cross-sections are also essential for calibrating high finesse optical cavities and differential optical absorption spectroscopy and for accurate remote sensing. In this study, we measured the scattering and absorption cross-sections of carbon dioxide, nitrous oxide, sulfur hexafluoride, oxygen, and methane in the continuous wavelength range of 307–725 nm using Broadband Cavity Enhanced Spectroscopy (BBCES). The experimentally derived Rayleigh scattering cross-sections for CO₂, N₂O, SF₆, O₂, and CH₄ agree with refractive index-based calculations, with a difference of 1.5% and 1.1%, 1.5%, 2.9%, and 1.4% on average, respectively. The O₂-O₂ collision-induced absorption and absorption by methane are obtained with high precision at the 0.8 nm resolution of our BBCES instrument in the 307–725 nm wavelength range. New dispersion relations for N₂O, SF₆, and CH₄ were derived using data in the UV-vis wavelength range. This study provides improved refractive index dispersion relations, n-based Rayleigh scattering cross-sections, and absorption cross-sections for these gases.
1. Introduction

The dominant interactions of gas-phase molecules with light in Earth’s atmosphere can be divided into absorption, where the light energy is converted to internal energy and generally (at atmospheric pressures) transferred to the surrounding environment as either as heat or as photoemission, and light scattering where the gases redistribute the light energy in the atmosphere. The knowledge of light extinction (scattering + absorption) by gases is essential for predicting the radiative transfer in the atmospheres of the Earth and other planets. In addition, the light extinction by gases is widely used for determining the effective optical pathlength of high-finesse optical cavities that measure trace gases and aerosols (Washenfelder et al., 2013; Washenfelder et al., 2008; Wilmouth and Sayres, 2019; Jordan et al., 2019) and for Differential Optical Absorption Spectroscopy (DOAS) to infer information about the light extinction properties of aerosols and clouds in the open atmosphere (Baidar et al., 2013; Platt and Stutz, 2008). The interaction of light with a wavelength much larger than the size of a molecule/particle gives rise to the scattering of light, which is known as Rayleigh scattering (Strutt, 1899). Rayleigh scattering accounts for scattering, local field effects (Lorentz–Lorenz) (Strutt, 1920) as well as depolarization from the non-sphericity of molecule/particles (King correction factor) (King and Eve, 1923; Strutt, 1918). For gas with known refractive index ($n_\nu$) and King correction factor ($F_k(\nu)$), the wavelength-dependent Rayleigh scattering cross-section ($\sigma_\nu$, cm$^2$ molecule$^{-1}$) can be calculated as follows (Sneeep and Ubachs, 2005):

$$\sigma_\nu = \frac{24\pi^3\nu^4}{N^2}\left(\frac{n_\nu^2-1}{n_\nu^2+2}\right)^2 F_k(\nu)$$  \hspace{1cm} (1)

where $N$ is the number density of the gas (molecules cm$^{-3}$) and $\nu$ is the wavenumber of the light (cm$^{-1}$). Note that the cross-section contains the gas number density but is not in fact dependent on the number density since the refractive index also appears in the expression. This $n$-based method is an advantageous approach for calculating Rayleigh scattering cross-sections, but it is vital to note that the accuracy of the calculated cross-sections depends on the experimentally-determined refractive index and the King correction factors. In particular, cautions should be used when applying a dispersion formula derived from measurements in one wavelength region to calculate Rayleigh scattering cross-sections in a different wavelength range.
Direct experimental measurement of Rayleigh scattering cross-sections is essential given the potential uncertainties in \( n \)-based calculations. While measurements of the King correction factors and refractive index for gases are well known from the literature (Cuthbertson and Cuthbertson, 1932; Leonard, 1974; Strutt, 1920; Vukovic et al., 1996; Hohm, 1993), there are only a few direct measurements of Rayleigh scattering cross-sections (Ityaksov et al., 2008a, b; Jordan et al., 2019; Naus and Ubachs, 2000; Sneep and Ubachs, 2005; Thalman and Volkamer, 2013; Thalman et al., 2014; Wilmouth and Sayres, 2019; He et al., 2018; Fuchs et al., 2009), especially measurements with a continuous spectrum from ultraviolet to visible.

Rayleigh scattering cross-section measurements were previously performed at a single wavelength (e.g., 458 nm, 532 nm, 632.8 nm) using Nephelometry (Shardanand and Rao, 1977) and cavity-ring down spectroscopy (CRDS) (Ityaksov et al., 2008a, b; Naus and Ubachs, 2000; Sneep and Ubachs, 2005; He et al., 2018). More recently, advanced Broadband Cavity Enhanced Spectroscopy (BBCES) was used to determine the Rayleigh scattering cross-sections of gases such as Ar, CO\(_2\), O\(_2\), SF\(_6\), and CH\(_4\). The BBCES technique enables the measurement of Rayleigh scattering cross-sections over a broad wavelength range. Thalman et al. (2014) performed measurements over selected wavelength regions between 350 and 660 nm using six BBCES cavities for N\(_2\), Ar, and O\(_2\). The BBCES were calibrated with He and N\(_2\) using Rayleigh scattering cross-sections calculated using their refractive index and from cavity-ring down measurements, respectively. They found a good agreement with \( n \)-based values to within 0.2±0.4%. Recent studies using BBCES with 30 nm spectral range were also used for Rayleigh scattering cross-section measurement in the UV wavelength region and demonstrated excellent agreement with \( n \)-based values for Ar and CO\(_2\) (Wilmouth and Sayres, 2020, 2019). Recently, Rayleigh scattering cross-sections for CO\(_2\) were measured using BBCES at visible wavelengths between 400 and 650 nm, and agreement with \( n \)-based values was within 2.4% on average. To the best of our knowledge, there is no direct continuous wavelength measurements of extinction cross-sections of gases that covers the ultraviolet across the entire visible range (300–725 nm) as shown in Table 1.

In this study, we used a recently-developed BBCES instrument to measure the extinction cross-sections of CO\(_2\), N\(_2\)O, SF\(_6\), O\(_2\), and CH\(_4\) continuously across the wavelength region 307–725 nm. All of the measurements were done at a single pressure to eliminate effects due to alignment. This requires the use of two gases with different Rayleigh cross-sections for the calibration of the BBCES instrument since the reference state is not vacuum. In this study, He and N\(_2\) were used to...
calibrate the system. By using the $n$-based calculated Rayleigh scattering cross-sections of He and $\text{N}_2$ to calibrate the path length of the optical cavity, the other cross-sections can be determined relative to the difference between these two gases. We report high accuracy Rayleigh scattering cross-sections for all five gases and compared our results with previous $n$-based values. New dispersion relations for $\text{N}_2\text{O}$, $\text{SF}_6$, and $\text{CH}_4$ are derived by incorporating data obtained by this study, extinction cross-section data in the deep UV, and previously available scattering cross-section data in the visible wavelength range.

2. Methods

2.1 Extinction measurement using BBCES

The BBCES systems used in this study are analogous to our previous studies (He et al., 2018; Washenfelder et al., 2016; Bluvshtein et al., 2016). Briefly, our BBCES consists of two channels, one in the UV (BBCES$_{\text{UV}}$, 307-350 nm) and one in the UV-vis range (BBCES$_{\text{vis}}$, 338-725 nm). The two channels of the BBCES share a laser-driven Xenon arc lamp source (LDLS EQ−99CAL, Energetiq Technology, Inc., MA, USA) coupled with a high transmission UV-Vis optical fiber from which the light is collimated and focused (BBFIBERX-600-1M, Energetiq Technology, Inc., MA, USA). The light source was purged with high purity $\text{N}_2$ and cooled by an aluminum block (with 15°C circulating water inside) to maintain stable optical power output. The UV light from the fiber is reflected by a low-pass dichroic mirror and filtered into the BBCES$_{\text{UV}}$ channel, which has a cavity with two 2.5 cm diameter, 1 m radius of curvature mirrors, with manufacturer’s reported reflectivity of 0.9995 (per pass loss = 500 parts per million, ppm) at the nominal center wavelength of 330 nm (Advanced Thin Films, Boulder, USA). The transmitted UV-vis light from the beam splitter is reflected and filtered into the BBCES$_{\text{vis}}$ channel consisting of two 2.5 cm, 1 m radius of curvature mirrors (FiveNine Optics, USA) with manufacturer’s reported reflectivity above 0.9993 (loss < 700 ppm), see Figure S1. The light emerging through the rear mirror of the cavity was focused using a 0.1 cm F/2 fiber collimator (74−UV, Ocean Optics, Dunedin, FL, USA) into a high transmission UV-vis optical fiber which directs the light into a high-performance spectrometer (QEPro, Ocean Insight, USA). Before gas measurement, the wavelength of the spectrometer was calibrated using an HG-1 mercury argon calibration light source (Ocean Insight, USA) within the wavelength range of 302.15−727.29 nm. During these experiments, a 300 line
mm$^{-1}$ grating and a 200 µm entrance slit width were used. The CCD array is a back-illuminated detector with 1024×56 pixels (Hamamatsu S7031-1006, Japan) thermo-electrically cooled to $-10$ °C to reduce thermal noise. Individual spectra at a wavelength resolution of 0.8 nm were acquired with 3.0 s integration time, and a total of 150 spectra were recorded during each measurement.

During the extinction measurements, the entire 94.0 ± 0.1 cm pathlength between the mirrors was filled with He, N$_2$, CO$_2$, N$_2$O, SF$_6$, or CH$_4$. The gases were obtained from several vendors (Airgas, Linde) with the following purities: He, 99.995%; N$_2$, 99.999%; N$_2$O, 99.999%, CO$_2$, 99.999%; SF$_6$, 99.999%; CH$_4$, 99.999%.

The reflectivity of the mirrors ($R(\lambda)$) can be determined as a function of wavelength ($\lambda$) by taking into account the difference in the extinction due to known literary data of Rayleigh scattering coefficient ($\alpha_{Ray}^{gas}$) by two different gases such as N$_2$ ($\alpha_{Ray}^{N_2}(\lambda)$) and He ($\alpha_{Ray}^{He}(\lambda)$) (Washenfelder et al., 2008).

\[
\frac{1-R(\lambda)}{d} = \frac{I_{N_2}(\lambda)(\alpha_{Ray}^{N_2}(\lambda)) - I_{He}(\lambda)(\alpha_{Ray}^{He}(\lambda))}{I_{He}(\lambda) - I_{N_2}(\lambda)}
\]

(2)

where $d$ is the length of the cavity filled by the gas. In this study, the studied gas filled the entire length of the cavity (94.0 ± 0.1 cm) since no purge flows were used. $I_{gas}$ is the light intensity measured by filling the cavity with high purity N$_2$ ($I_{N_2}(\lambda)$) and He ($I_{He}(\lambda)$). Rayleigh scattering ($\alpha_{Ray}^{gas}$) is the combined product of Rayleigh scattering cross-section ($\sigma$) and the gas number density ($N$) during the measurements. Rayleigh scattering cross-sections of N$_2$ and He are calculated using the data in Table 1. Figure S1 shows typical examples of light intensity when the BBCES cavities are filled with pure N$_2$. Reflectivity measurements were repeated every three sample measurements to track the stability of the system.

Once the reflectivity is determined, it is possible to calculate the wavelength-dependent extinction cross-sections of other gases ($\sigma(\lambda)$) as follows:

\[
\sigma(\lambda) = \left[ \left( \frac{1-R(\lambda)}{d} \right) \left( \frac{I_{He}(\lambda) - I_{gas}(\lambda)}{I_{gas}(\lambda)} \right) + \left( \frac{I_{He}(\lambda)}{I_{gas}(\lambda)} \right) \left( \alpha_{Ray}^{He}(\lambda) \right) \right] / N
\]

(3)

Where $N$ is the number density of the gas during the measurements, and $I_{gas}(\lambda)$ is the light intensity when a target gas fills the cavity. During our experiments, the purge flow of the high reflection
mirrors was shut down to ensure that the cavity was filled with target gas completely. To measure
the extinction cross-sections of CO$_2$, N$_2$O, and SF$_6$, the cavity is filled with pure target gas. Mass
flow controller controlled O$_2$/CH$_4$ flow was mixed with He in a 2 m Teflon tube ($\Phi = \frac{1}{4}$ inch) to
generate a gas mixture with total flow rate of 500 mL min$^{-1}$. For O$_2$ experiments, measurements
were performed for O$_2$ + He mixtures by varying the O$_2$ percentage between 10$\%$ and 100$\%$ with
a 10$\%$ step. The CH$_4$, measurements were performed for CH$_4$ + He mixtures with CH$_4$ percentage
ranges between 10$\%$ to 100$\%$ with a 10$\%$ step. Additional measurements were also performed for
15$\%$, 25$\%$, 35$\%$, and 45$\%$ CH$_4$.

2.2 Extinction measurements using cavity-ring down systems (CRDS) at 404 nm and 662 nm.

To obtain independent measurements for the extinction cross-sections and to cross-validation of
the BBCES technique, we conducted CRD measurements at two fixed wavelengths of 404 nm and
662 nm. CRDS is a highly sensitive technique and uses a different measurement principle than
BBCES. The CRDS measured the decay rate of light due to extinction rather than an absolute
absorbance (as in the BBCES) and thus immune to shot-to-shot source light fluctuations. A
detailed description of the CRD method for light extinction measurement can be found in
Bluvstein et al. (2016) and He et al. (2018). Briefly, diode lasers (110 mW 404 nm diode laser,
iPulse, Toptica Photonics, Munich, Germany; 120 mW 662 nm diode laser, HL6545MG, Thorlabs
Inc., NJ, USA) are used as the light source of these CRDS. The 404 nm and 662 nm lasers are
modulated at 1383 Hz and 500 Hz with a 50$\%$ duty cycle. The diode lasers are optically isolated
by quarter waveplates (1/4 $\lambda$) and polarizing beam splitters to prevent damage to the laser head by
back reflections from the highly reflective CRDS mirror. The back-reflected light beam is directed
into a photodiode, which serves as an external trigger source. Light transmitted through the back
mirror of the cavity is collected by an optical fiber and detected by a photomultiplier tube (PMT),
which samples at a rate of 10 to 100 MHz. The time-dependent intensity data is acquired with a
100MHz card (PCI-5122, National Instruments, USA) and processed by data acquisition software
in Labview. An exponential curve is fitted to each intensity decay data set (Figure S2). Over 1000
decay time measurements are monitored and averaged on a second basis. The residual of the fit
for the averaged intensity decay is obtained and further normalized to the averaged intensity. The
derived relative residuals (Figure S2) show no apparent structure with other time constants,
validating the application of CRDS as a good measure of extinction. The resultant 1 Hz decay time
is averaged over one measurement duration of five minutes with standard error as the measurement uncertainty.

All of the CRDS measurements were performed under room temperature and pressure downstream from the BBCES instrument. The gas temperature (K-type thermocouple) and cavity pressure (Precision Pressure Transducer, Honeywell International Inc., MN, USA) were recorded for gas number density \( (N) \) calculation. During the CRDS measurements, the full cavity was filled with the investigated gases (He, CO\(_2\), N\(_2\)O, SF\(_6\), O\(_2\), CH\(_4\), or gas mixtures (O\(_2\) + He and CH\(_4\) + He)).

The extinction cross-section \( (\sigma(\lambda)) \) of the studied gas was measured relative to that of He and was calculated by equation (4):

\[
\sigma(\lambda) = \frac{L}{c} \left( \frac{1}{\tau_{gas}} - \frac{1}{\tau_{He}} \right) + \sigma_{He}
\]

Where \( L \) is the total length of the cavity \( (l) \), \( c \) is the speed of light, and \( \tau_{gas} \) and \( \tau_{He} \) are the ring-down time of the cavity when it is filled by target gas or by the reference gas, He.

### 2.3 Data processing

For comparison, the scattering cross-sections of the gases investigated in this study were also calculated with Equation (1) based on the refractive index and the King correction factors available in the literature that are listed in Table 1. The King correction factors are taken as unity for monoatomic molecules and spherical molecules (with regards to the depolarization) but deviates for non-spherical molecules. For the 307–725 nm wavelength range of this study, the \( n \)-based calculated Rayleigh scattering cross-sections from largest to smallest are SF\(_6\), N\(_2\)O, CO\(_2\), CH\(_4\), N\(_2\), O\(_2\), and He.

The extinction of O\(_2\) + He mixtures \( (\alpha_{O_2+He}) \) consists of the extinction by O\(_2\) \( (\alpha_{O_2}) \) and He \( (\alpha_{He}) \), and the O\(_2\)–O\(_2\) collision-induced absorption \( (\alpha_{O_2-O_2}) \). The extinction of O\(_2\) and He is a combined product of extinction cross-section \( (\sigma_{gas}) \) and gas number density \( (N_{gas}) \). Thus \( \alpha_{O_2+He} \) can be described with the following equation:

\[
\alpha_{O_2+He} = \alpha_{O_2} \times N_{O_2}^2 + \sigma_{gas} \times N_{O_2} + \sigma_{He} \times N_{He}
\]

Where \( N_{O_2} \) and \( N_{He} \) are the number density of the O\(_2\) and He in the cavities. Performing a 2nd order polynomial fit to the extinction obtained by the BBCES with respect to the gas number density
thus yields the extinction cross-section of O$_2$ and the O$_2$-O$_2$ collision-induced absorption (CIA) cross-section.

In addition to the results from 2$^{nd}$ order polynomial fitting, we also used data from pure O$_2$ measurement to calculate the extinction by O$_2$ and by CIA of O$_2$–O$_2$. The real refractive index of O$_2$ ($n_{O_2}$) derived from extinction data measured in the wavelength regions where there is no absorption was fitted using the generalized expression of $(n_{O_2} - 1) \times 10^8 = A + \frac{B}{C-\nu^2}$. Based on the refractive index, the scattering cross-sections of O$_2$ in the wavelength range of 307-725 nm were further calculated. By subtracting the scattering cross-section of O$_2$ from the measured total extinction, we derived the CIA of O$_2$–O$_2$. However, the O$_2$ absorption bands at 580, 630, and 690 nm overlaps with those of O$_2$–O$_2$ collisions. Additional corrections are thus needed to split the absorption by O$_2$ and O$_2$–O$_2$ collision, which is out of the scope of this study.

Methane has weak vibrational overtone absorption in the UV-vis wavelength range that is comparable to or greater than its Rayleigh scattering. Previous high-resolution spectroscopy studies have identified smooth and unstructured absorption bands across the UV-visible range (Giver, 1978; Smith et al., 1990). The spectral features are substantially broader than 0.8 nm, thus the absorption by CH$_4$ can be measured by our BBCES. The measured extinction coefficients of CH$_4$+He mixtures ($\alpha_{CH_4+He}$) are linearly correlated with the number concentration of the CH$_4$ ($N_{CH_4}$) as described by the following equation:

$$\alpha_{CH_4+He} = \sigma_{CH_4} \times N_{CH_4} + \sigma_{He} \times N_{He}$$

A linear fit was used for deriving the extinction cross-section of CH$_4$. The absorption between 300 and 400 nm is negligible as compared to the Rayleigh scattering. Thus extinction data in this UV wavelength range were used to calculate the real part of the refractive index of CH$_4$ which was further fitted utilizing the expression of $(n_{CH_4} - 1) \times 10^8 = A + \frac{B}{C-\nu^2}$. By applying this dispersion relation, the Rayleigh scattering cross-sections in the entire wavelength range of 307–725 nm were derived. Finally, the CH$_4$ absorption cross-sections were calculated by subtraction of the scattering cross-section from the extinction cross-section.

2.4 Error Propagation for BBCES
The uncertainty for BBCES measurements can be assessed by the propagation of the errors associated with the measurements. The pressure (±0.01%), temperature (±0.1%) and cavity length (94.0 ± 0.1 cm) are combined with the Rayleigh cross-section uncertainties for N$_2$ (±1%) as well as uncertainty in the measurements of the spectral signal by the spectrometer (<0.2%) to get an overall relative uncertainty for the mirror reflectivity curve of ±1.03%. This uncertainty is further propagated to the target gas by consideration of the uncertainties of pressure, temperature, and spectral intensity of the target gas measurements. The overall 1-σ uncertainty of the gas extinction cross-section is 1.1%. The precision of the mass flow controllers is 0.5 mL min$^{-1}$. When the total flow rate is 500 mL min$^{-1}$, the resulted uncertainty of the gas concentration (10–100%) varies from 0% to 1.0%. Thus, the overall 1-σ uncertainty of extinction coefficients measured for CH$_4$+He and O$_2$+He varies from 1.1% to 1.5%. The detailed wavelength-dependent uncertainties were calculated due to the wavelength-dependence of the spectral intensity. The results are shown and discussed in later sections.

3 Results and Discussion

3.1 Performance of the optical system

The reflectivity of the cavity mirrors, measured across the entire range using the difference in Rayleigh scattering of N$_2$ and He, was very stable throughout the experiments. The measured mirrors reflectivity curves are shown in Figure S1. The mean peak reflectivity of the BBCES$_{UV}$ mirrors was 0.999328±0.000006 (672±6 ppm) at 330 nm, with a corresponding effective optical pathlength of 1.40±0.01 km. The reflectivity curve of the BBCES$_{Vis}$ is much more structured, with reflectivity ranging between 0.999224±0.000010 and 0.9999550±0.0000006 (776±10 ppm > loss > 45±0.6 ppm) over a wide wavelength range of 338–725 nm. The reflectivity of the BBCES$_{Vis}$ is much higher than that of our previous system (He et al., 2018) and also covers a much broader wavelength range. Thus the effective pathlength of the BBCES$_{Vis}$ varies between 1.3 and 20.4 km, guaranteeing a high sensitivity of the extinction measurement. The mean uncertainty in the effective pathlength across the measured wavelengths as determined from the mirror reflectivity was ±1.03%, which is predominantly due to the uncertainty in the Rayleigh scattering cross section for N$_2$ derived from n-based calculation.

3.2 Rayleigh scattering cross-sections of CO$_2$, N$_2$O, SF$_6$. 

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Figure 1 shows the extinction cross-sections of CO$_2$, N$_2$O, and SF$_6$ measured by the BBCES. The extinction cross-sections of these gases monotonically decrease with increasing wavelength, and no absorption (i.e., no structured extinction larger than the smoothly varying Rayleigh curve) is observed in the wavelength range of 307–725 nm, indicating that the measured extinction is due solely to the Rayleigh scattering of these gases. The wavelength-dependent relative standard deviations of the measurements for each gas are shown in Figure 1d. The mean 1-σ uncertainty of the reported cross sections for all three gases across the 307–725 nm wavelength range is 1.5% for CO$_2$, 1.1% for N$_2$O, and 1.5% for SF$_6$. As mentioned above, the derived uncertainty originates predominantly from the uncertainty in the N$_2$ Rayleigh scattering cross-section. Uncertainty in the Rayleigh cross-sections of each gas varies with wavelength and generally tracks the light intensity spectra, which is a combined product of light source spectrum and the mirror reflectivity profile. The uncertainty is much higher when the transmitted light intensity is low (Figure S1).

The BBCES measured Rayleigh scattering cross-sections for these three gases agree well with those obtained by our CRDS operating at 404 nm and 662 nm, with deviations smaller than 1.6%. Table 2 listed the Rayleigh scattering cross-sections at several wavelengths obtained by the BBCES measurements (Exp) and by the theoretical calculations using the refractive index and F$_k$(v) values from Table 1 (n-based). The relative differences between these two sets of results are within 1.4%.

Figure 1a–c shows a comparison of the measured Rayleigh scattering cross-sections for CO$_2$, N$_2$O, and SF$_6$ with n-based calculations and with previous experimental results from the literature. There are a few measurements for the Rayleigh scattering cross-sections for CO$_2$ which cover a wide spectral range (Jordan et al., 2019; Shardanand and Rao, 1977; Sneep and Ubachs, 2005; Wilmouth and Sayres, 2019; He et al., 2018). There are fewer Rayleigh scattering measurements for N$_2$O and SF$_6$ in the studied wavelength range. The measured Rayleigh scattering cross-sections for CO$_2$, N$_2$O, and SF$_6$ are in excellent agreement with n-based calculation. The wavelength-dependent difference of our experimentally derived Rayleigh scattering cross-sections with n-based calculations are shown in Figure 1e. The mean ratios of our measurements to the n-based values for the entire wavelength range of 307–725 nm are 1.00±0.01, 0.99±0.01, and 1.01±0.01 for CO$_2$, N$_2$O, and SF$_6$, respectively. Notably, while our results for N$_2$O agree well with the n-based calculations, previous results obtained by CRDS at 532 nm (Sneep and Ubachs, 2005) and by absorption spectroscopy in the wavelength of 300–315 nm (Bates and Hays, 1967) do not agree...
well with the n-based calculations. The measurements between 300 and 315 nm were first published by Bates and Hays (1967), who obtained the results from a doctoral thesis. However, the results from our BBCES system are in good agreement with the n-based calculations and with experimental results from independent CRDS measurements, thus increasing the confidence in our measured values.

3.3 Scattering and absorption cross-sections of O₂.

The UV-vis spectra of gas-phase molecular oxygen are characterized by discrete structured absorption bands due to the electronic transition \((b^1 \Sigma_g^+(\nu' = 1/2/3) \leftarrow \Sigma_g^-(\nu'' = 0))\) of O₂ monomer, broader unstructured CIA of O₂–O₂, and structured dimer bands from the bound van der Waals O₂ dimer (Newnham and Ballard, 1998). Under atmospheric conditions, the O₂–O₂ CIA bands are frequently described as "O₄" bands, although absorption by O₂ dimer is thought to be significant only under very low-temperature conditions (Thalman and Volkamer, 2013; Long and Ewing, 1973). Within the wavelength range investigated in this work, the molecular oxygen B band at 688 nm \((b^1 \Sigma_g^+(\nu' = 1) \leftarrow X^3 \Sigma_g^-(\nu'' = 0))\), γ overtone band at 629 nm \((b^1 \Sigma_g^+(\nu' = 2) \leftarrow X^3 \Sigma_g^-(\nu'' = 0))\), and δ overtone band at 580 nm \((b^1 \Sigma_g^+(\nu' = 3) \leftarrow X^3 \Sigma_g^-(\nu'' = 0))\) overlap with O₂–O₂ CIA bands of \(^1\Sigma_g^+(\nu = 1)\), \(^1\Delta_g + ^1\Delta_g (\nu = 0)\), and \(^1\Delta_g + ^1\Delta_g (\nu = 1)\), respectively. These absorption bands can only be resolved by a high-resolution spectroscopic technique.

Absorption cross-sections of the B, γ, and δ bands were convoluted from the HITRAN database (Gordon et al., 2017) by considering the temperature, pressure, and instrument’s wavelength resolution. The wings of the oxygen lines also show a quadratic dependence on the pressure due to pressure broadening. However, due to the minimal O₂ absorption contribution below 680 nm and the low instrument wavelength resolution, the extinction cross-section of the O₂ monomer can be treated as linearly correlated with the O₂ concentration. Moreover, the O₂–O₂ CIA cross-section is correlated with the square of the O₂ concentration. Therefore, these cross-sections can be retrieved from measurements at different O₂ concentrations. Due to the discrete structured absorption bands and the instrument’s wavelength resolution, the range of absorption cross-sections spans several orders of magnitude within the spectral response of the instrument, limiting the relevance of the absorption cross-sections for other researchers. These results are not further discussed here. However, the data for broader unstructured CIA of O₂–O₂ are still useful for various applications.
Figure 2 shows the wavelength-dependent extinction coefficients of O$_2$+He mixtures. He was used in these experiments to minimize extinction contributions from Rayleigh scattering. Nine absorption peaks centered at 344 nm (CIA), 360 nm (CIA), 380 nm (CIA), 446 nm (CIA), 477 nm (CIA), 532 nm (CIA), 577 nm (δ overtone and CIA), 629 nm (γ overtone and CIA), and 688 nm (B band and CIA) were observed in the wavelength range of 307–725 nm. The absorption coefficients of the central wavelengths for the first eight peaks increase non-linearly with O$_2$ concentration while that of the 688 nm peak increases in a more linear manner, indicating that the O$_2$ B band absorption dominates the last absorption peak while the other peaks are mostly associated with CIA of O$_2$–O$_2$.

The extinction coefficients obtained by the BBCES correlated well with those measured by the CRD, with slopes of 0.990 ($R^2=0.9994$) and 0.993 ($R^2 = 0.9996$) at the wavelengths of 404 nm and 662 nm, respectively (Figure 3). This excellent agreement between the instruments further substantiates the BBCES measurements and suggests that the accuracy of the BBCES is better than estimated in the error propagation above, where the N$_2$ refractive index was the largest uncertainty. As explained in the data processing section, the measured extinction coefficients were fitted with a 2nd order polynomial (selected wavelengths at the peaks of the CIA absorption bands are shown in Figure 4). At 476.7, 577.2, and 629.2 nm, the absorption is from the CIA of O$_2$–O$_2$.

The fit generates positive values matching the absorption cross-section of O$_2$–O$_2$ CIA. At 687.7 nm where strong B-band absorption appears, the fit yields a small negative coefficient for O$_2$–O$_2$ CIA.

Figure 5a shows the extinction cross-section measured for 100% O$_2$. These results agree well with previously reported results by Jordan et al. (2019). For wavelengths where no absorption is detected, the measured extinction cross-sections agree well with $n$-based calculations. Figure 5b-c shows the determined extinction cross-sections for molecular O$_2$ and the absorption cross-sections of O$_2$–O$_2$ CIA. For wavelength ranges without O$_2$ bands, our extinction cross-sections agree well with $n$-based values with an average deviation of 2.8%. The absorption cross-sections for O$_2$–O$_2$ CIA derived in this study mostly agree well with literature data from Thalman and Volkamer (2013). The differences are within 1.1% at 477, 532, 577, and 630 nm but larger deviations were found at 344 (4.2%), 360 (-29%), 380 (-21%), and 446 (4.2%) nm. These absorption bands are the lowest intensity bands and therefore have the largest relative uncertainties in either measurement.
The Rayleigh scattering cross-sections of molecular O\(_2\) derived from the 100\% O\(_2\) measurement agree well with \(n\)-based calculations with an average difference of 1.2\%. CIA of O\(_2\)--O\(_2\) calculated from this single measurement matches the results from the fitting method. Due to strong absorption from O\(_2\) B band and \(\gamma\) overtone band, this method cannot derive the cross-sections of CIA of O\(_2\)--O\(_2\) at 630 and 688 nm.

3.4 The scattering and absorption cross-sections of CH\(_4\).

CH\(_4\) has weak absorption in the UV–vis wavelength range, and these bands dominate the photographic spectra of planets such as Uranus and Neptune. Figure 6 presents the wavelength-dependent extinction coefficients of CH\(_4\)+He mixtures. A total of eleven absorption bands were detected in the wavelength range of 307–725 nm. The extinction coefficients increase as a function of increasing CH\(_4\) concentration. Extinction coefficients obtained by the BBCES correlated well with those measured in parallel by the CRDS, with slopes of 1.002 (\(R^2=0.9999\)) and 0.99 (\(R^2=0.999\)) at the wavelengths of 404 nm and 662 nm (Figure S3). The excellent agreement between these three systems further supports the accuracy of BBCES extinction measurements over a wide working range. The measured extinction coefficients were linearly fit against the CH\(_4\) number concentration. Figure 7 shows the fitted curves at five selected wavelengths. The extinction coefficients have a linear correlation with CH\(_4\) concentration (\(R^2 > 0.9988\)) without exception. The calculated slopes represent the extinction cross-sections of CH\(_4\) and also indicate a wide dynamic range of our BBCES.

The extinction cross-sections for CH\(_4\) retrieved from concentration-dependent measurements are plotted in Figure 8a. BBCES results from this study agree well with results from previous studies using BBCES (Jordan et al., 2019; Wilmouth and Sayres, 2019) and CRDS (Sneep and Ubachs, 2005). Previous studies using a Nephelometer (Shardanand and Rao, 1977) and interferometer (Cuthbertson and Cuthbertson, 1920; Watson et al., 1936) obtained the scattering cross-sections of CH\(_4\). The BBCES measures the extinction cross-section. For wavelengths where extinction is dominated by Rayleigh scattering (< 475 nm), our BBCES results agree well with the results from Nephelometer and interferometer measurements. In this study, the refractive index of CH\(_4\) was calculated using the extinction data in the wavelength range of 307–400 nm. The calculated refractive index was fitted to the general expression:

\[
(n_{CH_4} - 1) \times 10^8 = 5476 + \frac{4.1579 \times 10^{14}}{1.1568 \times 10^{10} - \nu^2}
\]  
(7)
As shown in Figure 8b, our calculated scattering cross-sections are in good agreement with those derived from the newest refractive index developed by Wilmouth and Sayres (2020) (Table 2), with an average difference of 1.4%. The absorption cross-section, which is the difference between the total extinction and the Rayleigh scattering cross-section, is shown in Figure 8c. At most spectral ranges, our results are in good agreement with the results from previous studies (Giver, 1978; Smith et al., 1990). For example, the difference as compared to the results from Giver (1978) at 542, 576.4, 598, 619, 665.7, and 703.6 nm is 4.0% on average. At several wavelength regions (e.g., 520–536 nm, 580–605 nm), the results from Fink et al. (1977) differ from all of the other studies. In the wavelength range of 400–725 nm, absorption contributes up to 99.7% of the CH<sub>4</sub> extinction.

3.5 Dispersion relations for N<sub>2</sub>O, SF<sub>6</sub> and CH<sub>4</sub>.

SF<sub>6</sub>: Wilmouth and Sayres (2020) found that their measured Rayleigh scattering cross-sections for SF<sub>6</sub> in the ultraviolet range were much lower than those from the n-based expression of Sneep and Ubachs (2005). To better constrain the dispersion formula when extrapolated over a broad wavelength range, we employed an alternative fit of the form \( A + B/(C - \nu^2) \) to our data. The Rayleigh scattering derived refractive index in the wavelength range of 264–297 nm and 333–363 nm by Wilmouth and Sayres (2020, 2019), and direct refractive index measurement at 632.99 nm (Vukovic et al., 1996) were used (Figure 9a). The resulting dispersion relation for SF<sub>6</sub> in the wavelength range of 264–725 nm is

\[
(n_{SF_6} - 1) \times 10^8 = 22871 + \frac{8.0021 \times 10^{14}}{1.6196 \times 10^{10} - \nu^2}
\] (8)

N<sub>2</sub>O: Sneep and Ubachs (2005) derived the refractive index based on polarizability measurements using interferometer at five single wavelengths (457.9, 488, 514.5, 568.2, 647.1 nm) by Alms et al. (1975). In this study, we calculated the refractive index of N<sub>2</sub>O from the Rayleigh scattering cross-sections in the wavelength range of 307–725. Based on this refractive index data set, the dispersion relation (Eq (9)) for N<sub>2</sub>O was retrieved for a much broader wavelength range (Figure 9b) compared to that generated by Sneep and Ubachs (2005).

\[
(n_{N_2O} - 1) \times 10^8 = 23154 + \frac{1.534 \times 10^{14}}{6.5069 \times 10^9 - \nu^2}
\] (9)

CH<sub>4</sub>: The previous study by Wilmouth and Sayres (2019) has shown that their measured Rayleigh scattering cross-sections for CH<sub>4</sub> are in substantial disagreement (22%) with those calculated from
the refractive index recommended by Sneep and Ubachs (2005). Sneep and Ubachs (2005) formulated the refractive index of CH\textsubscript{4} based on interferometric measurements at wavelengths of 325, 543.5, 594.1, 612, and 633 nm by Hohm (Hohm, 1993). However, the Rayleigh scattering cross-sections calculated from their refractive index are much higher than all the measured values listed in Figure 9b. Using Rayleigh scattering cross-sections in the wavelength range of 264–297 nm, 333–363 nm (Wilmouth and Sayres, 2019, 2020), 307–400 nm from this study, and single wavelength measurements which are not impacted by absorption (Cuthbertson and Cuthbertson, 1920; Shardanand and Rao, 1977; Watson et al., 1936), we derived the dispersion formula for the refractive index of CH\textsubscript{4} in the combined UV/visible range (Figure 9c) as follows:

\[
(n_{CH_4} - 1) \times 10^8 = 7327.7 + \frac{4.1884 \times 10^{14}}{1.2208 \times 10^{10} - \nu^2} \tag{10}
\]

The calculated Rayleigh scattering cross-sections using the dispersion relations derived in this study were compared with those derived from previously recommended formulations listed in Table 1 (Figure 9). The difference increases significantly at the longer wavelength in the region of 320–725 nm (Figure S4). The average deviations are 0.8%, 0.9%, and 1.6% for SF\textsubscript{6}, N\textsubscript{2}O, and CH\textsubscript{4}, respectively. Notably, the difference for CH\textsubscript{4} is much more significant than for the other two gases. This study uses additional measurements and literature data in the wavelength ranges of 307-333 nm and 363–400 nm than those used by Wilmouth and Sayres (2020). Therefore this fit captures well our BBCES measurements (Figure 9d), and also the Wilmouth and Sayres (2020, 2019) data.

Conclusions and Implications

Rayleigh scattering cross-sections between 307 and 725 nm were determined for CO\textsubscript{2}, N\textsubscript{2}O, SF\textsubscript{6}, O\textsubscript{2}, and CH\textsubscript{4} by simultaneous BBCES and CRDS measurements. Extinction coefficients obtained by the BBCES show high consistency with those measured by parallel CRDS at 404 and 662 nm (Figure 3 and figure S3), demonstrating that the BBCES measurements provide results with both a wide wavelength range and high accuracy. Comparison of our measurements with n-based calculations for these gases in the entire wavelength range of this study yields excellent agreement with relative differences of 1.5% and 1.1%, 1.5%, 2.9%, and 1.4% on average, respectively. The O\textsubscript{2}-O\textsubscript{2} CIA cross-sections obtained from the BBCES measurements are compared with those published by Thalman and Volkamer (2013). The relative differences are within 1.1% at 477, 532, 577, 630 nm. Larger relative differences occur at the weak bands at 344...
The absorption cross-sections of CH$_4$ in the wavelength range of 400-725 nm agree well with those documented by Giver (1978).

Rayleigh scattering cross-sections of CO$_2$ determined using BBCES in this study, and in other studies, have shown that the refractive index recommended by Sneep and Ubachs (2005) is suitable for use in the wavelength range of 307–725 nm. By incorporating the refractive index data from previous studies, we developed new dispersion relations for the refractive index of N$_2$O (307-725 nm), SF$_6$ (264–725 nm), and CH$_4$ (264–671 nm). The new dispersion relation for CH$_4$ captures the measurements from BBCES more adequately.

Previous studies measured the Rayleigh scattering and absorption cross-sections of CO$_2$, N$_2$O, O$_2$, SF$_6$, and CH$_4$ at narrow spectral ranges or single wavelengths. In this study, we used BBCES that covers the broad wavelength range of 307–725 nm to measure total extinction (the sum of absorption and scattering). The measurements validate that refractive index-based methods for calculating Rayleigh extinction cross-sections are accurate and provide new fits over more continuous and extended wavelengths range than available in the literature to constrain such methods. The Rayleigh scattering cross-sections reported here are useful in several applications. These include calibration standards based on extinction for optically-based instruments, such as those designed for aerosol optical properties measurements or trace gas concentrations in the field (Jordan et al., 2019; Min et al., 2016; Bluvshtein et al., 2017), especially when high-refractive index gases are used for improved calibration. They will also improve the accuracy of Rayleigh scattering parameterizations for major greenhouse gases in Earth’s atmosphere, CO$_2$, CH$_4$, and N$_2$O. Accurate quantitative measurements of Rayleigh scattering coefficients and absorption cross-sections of atmospheric gases such as molecular N$_2$, O$_2$, CO$_2$ and the CIA of O$_2$–O$_2$ cross-sections in the UV-NIR range are of particular importance for the application of Rayleigh LIDAR systems, especially at the Nd:YAG laser harmonics 1064, 532 & 366 nm. These systems analyze the molecular backscattering contributions to the LIDAR’s attenuated backscatter signals to retrieve the atmospheric profile of aerosols and clouds in the planetary boundary layer (Tomasi et al., 2005; Herron, 2007). Recent NASA satellite missions have also aimed to measure global carbon dioxide concentrations with high precision (0.25%) (Drouin et al., 2017). These CO$_2$ global missions use the O$_2$–O$_2$ CIA underneath the structured O$_2$ A-band (760 nm) to evaluate the solar radiation double pathlength in the Earth atmosphere and to determine the atmospheric pressure. The measurements in this study validate the existing literature on the extinction of O$_2$ collision...
complexes and molecular oxygen bands, and can be used for calibration purposes in both remote sensing and *in-situ* spectroscopic applications in the atmosphere. In the future, gas extinction measurements at extended wavelengths (near-infrared) and for additional gases (e.g., \( \text{N}_2 \)) will improve the spectroscopic applications in atmospheric studies.

**Data availability.**

Data are available upon request from the corresponding author (yinon.rudich@weizmann.ac.il).

**Author contributions.**

Q.H., S.S., and Y.R. designed this study. Q.H., Z.F., and O.S. conducted the experiments. Q.H. prepared the draft and all of the co-authors reviewed it and provided comments.

**Competing interests.**

The authors declare that they have no conflict of interest.

**Acknowledgments**

This research was partially supported by the US-Israel Binational Science Foundation (BSF grant #2016093). Dr. Q. H. is supported by the Koshland Foundation and the Center for Planetary Sciences, Weizmann Institute of Science. Dr. Z.F. is supported by SAERI initiative of the Weizmann Institute.
Figure 1. Rayleigh scattering cross-sections of CO$_2$ (a), SF$_6$ (b), and N$_2$O (c). Panel (d) shows the relative standard deviations as a function of wavelength for each gas. The relative difference in the cross-sections obtained by the BBCES and calculations from the refractive index are displayed (e).
Figure 2. Wavelength-dependent extinction coefficients of O₂ + He mixtures as a function of O₂ concentration. The colored lines represent the extinction coefficients measured by BBCES, and markers represent results from CRDS.
Figure 3. Correlations between the extinction coefficients (unit, cm\(^{-1}\)) measured by the BBCES and CRDS.

\[
\begin{align*}
\text{BBCES (404 nm)} & : y = 0.990 x + 2.320e-9, \quad R^2 = 0.9994 \\
\text{BBCES (662 nm)} & : y = 0.993 x + 1.161e-10, \quad R^2 = 0.9996
\end{align*}
\]
Figure 4. 2nd order polynomial fit of extinction coefficients measured by the BBCES. The O$_2$ concentration-dependent extinction coefficients are contributed by the extinction coefficients of O$_2$ ($\sigma_{O_2}$), He ($\sigma_{He}$), and the O$_2$-O$_2$ CIA cross-sections ($\sigma_{O_2-O_2}$).
Figure 5. Wavelength-dependent extinction cross-sections of 100% O₂ (a), extinction cross-sections of O₂ (b), and O₂-O₂ CIA cross-section (c).
Figure 6. Wavelength-dependent extinction coefficients of CH$_4$ + He mixtures as a function of CH$_4$ mixing ratio. The colored lines represent extinction coefficients obtained from BBCES and markers represent results from CRDS. Measurements were performed with CH$_4$ percentage within 10% and 100% with a 10% step. Moreover, BBCES measurements were also performed for 15%, 25%, 35%, and 45% CH$_4$. The number concentration of 100% methane was 2.50143×10$^{19}$ molecules cm$^{-3}$. Data at selected wavelengths (vertical lines) are shown in Figure 7.
Figure 7. The relationship between BBCES measured extinction coefficients of CH$_4$+He mixtures and CH$_4$ concentration. The selected wavelengths were located in Figure 6 by vertical lines.
Figure 8. Extinction (a), scattering (b), and absorption (c) cross-sections of CH$_4$. 
9. Real refractive index \( (n) \) for SF\(_6\) (a), N\(_2\)O (b), and CH\(_4\) (c). Comparison of refractive index from this work with previous studies (Cuthbertson and Cuthbertson, 1920; Naus and Ubachs, 2000; Shardanand and Rao, 1977; Snee and Ubachs, 2005; Vukovic et al., 1996; Watson et al., 1936; Wilmouth and Sayres, 2019, 2020) over the wavelength range of 264–725 nm. The green line represents the dispersion relation given in Table 1. The black line represents the dispersion relation given in Eq. (8–10) derived from a fit to our data and references results. The \( n \) values for Shardanand and Rao (1977), Snee and Ubachs (2005), Naus and Ubachs (2000) were calculated from their reported Rayleigh scattering cross-sections. Refractive index data from Snee and Ubachs (2005) are not used in the fitting since these results are away from others. Panel (d) is a close-up view of the panel (c) in the wavelength range of 264–363 nm.
Table 1. Refractive index and King correction factors for calculating Rayleigh scattering cross-sections and available measurements in the wavelength range of 300–725 nm. Measurements for He and N$_2$ are not summarized in this table.

<table>
<thead>
<tr>
<th>Gas</th>
<th>Refractive index and King correction factors</th>
<th>Measurements</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>(n-1) $\times 10^8$</td>
<td>$F_0(\nu)$</td>
</tr>
<tr>
<td>He</td>
<td>2283 + $1.8102 \times 10^{13}$</td>
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<tr>
<td></td>
<td>+ $1.5342 \times 10^{10} - \nu^2$</td>
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<tr>
<td>N$_2$</td>
<td>5677.465 + $3.188174 \times 10^{12}$</td>
<td>1.034 + 3.17 $\times 10^{-12} \nu^2$</td>
</tr>
<tr>
<td></td>
<td>+ $1.44 \times 10^{10} - \nu^2$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$\times \frac{(1289089^3 - \nu^2)}{579925}$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>+ $\frac{(89223.8)^2 - \nu^2}{5.3354}$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>+ $\frac{(75037.5)^2 - \nu^2}{4.3244}$</td>
<td></td>
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<tr>
<td></td>
<td>+ $\frac{(67837.7)^2 - \nu^2}{1.218145 \times 10^{-5}}$</td>
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<tr>
<td></td>
<td>+ $\frac{(2418.136)^2 - \nu^2}{10^{-2}}$</td>
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<tr>
<td>CO$_2$</td>
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<td>39417-55340</td>
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<td>$\times \frac{1.133 \times 10^{10} - \nu^2}{10^{-2}}$</td>
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<td>N$_2$O</td>
<td>46890 + $4.12 \times 10^{-6} \nu^2$</td>
<td>$3.3462 + 70.8 \times 10^{-12} \nu^2$</td>
</tr>
<tr>
<td></td>
<td>$\times \frac{2.7692 - 47.2 \times 10^{-12} \nu^2}{10^{-2}}$</td>
<td></td>
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<tr>
<td>SF$_6$</td>
<td>18611.4 + $8.9566 \times 10^{14}$</td>
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<tr>
<td></td>
<td>+ $1.680 \times 10^{10} - \nu^2$</td>
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<td>O$_2$</td>
<td>20564.8 + $2.480899 \times 10^{13}$</td>
<td>$1.09 + 1.385 \times 10^{-11} \nu^2$</td>
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<tr>
<td></td>
<td>+ $4.09 \times 10^{-9} - \nu^2$</td>
<td>+ $1.448 \times 10^{-20} \nu^4$</td>
</tr>
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</table>

Unless noted, the refractive index is scaled to 288.15 K and 1013.25 hPa. $N = 2.546899 \times 10^{19}$ molecules cm$^{-3}$. Due to limited space, only the first name of each reference is shown in the table. $^a$ The refractive index was obtained at 273.15 K and 1013.25 hPa, $N = 2.68678 \times 10^{19}$ molecules cm$^{-3}$ is used in Eq. (1)
Table 2. The Rayleigh scattering cross-sections ($10^{-27} \text{ cm}^2$) calculated from the refractive index (n-based) and obtained from BBCES (Exp) of selected wavelengths.

<table>
<thead>
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<th>$\lambda$(nm)</th>
<th>CO$_2$</th>
<th>SF$_6$</th>
<th>N$_2$O</th>
<th>O$_2$</th>
<th>CH$_4$</th>
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<td>41.6</td>
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<td>4.08</td>
<td>10.55</td>
<td>10.4</td>
<td>5.551</td>
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</table>
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