

Initial review of “Scattering and Absorption  
Cross-sections of Atmospheric Gases in the  
Ultraviolet-Visible Wavelength Range (307 – 725  
nm)” by Quanfy He, Zhen Fang, Ofir Shoshamin,  
Steven S. Brown and Yinon Rudich.

The research presented in the manuscript is interesting, and certainly relevant to the journal Atmospheric Chemistry and Physics. It is somewhat unfortunate that nitrogen is used for the calibration of the mirror reflectance, as nitrogen itself is a highly interesting gas for these type of measurements. However, the manuscript needs to be improved significantly before it is ready for publication.

I have doubts about the direct measurements of the oxygen absorption bands presented rather prominently in the abstract (the delta and gamma overtone bands and the B-band) should be included at all. The spectral resolution of the instrument is simply not suitable to produce a meaningful result for these bands. I interpreted the “0.8 nm resolution” statement as a FWHM value, and produced a plot of high resolution cross sections of oxygen (HITRAN; modelled concentration is 21 % O<sub>2</sub> in N<sub>2</sub> at 1018 hPa and 294 K and includes pressure and temperature broadening) and overplotted a Gaussian spectral response function with FWHM of 0.8 nm, see figure 1. This combination cannot produce a meaningful result. The range of absorption cross sections spans several orders of magnitude within the spectral response of the instrument. In a CRDS setup this would lead to a multi-exponential decay, an underdetermined problem. I’m not sure the situation of interpreting the output of a BBCES instrument when a wide range of absorptions is present within a spectral pixel is any better. At best the result depends on the spectral resolution, making it far less useful to others. Also note that this is before taking any broadband collision induced effects into account, but that is probably the least of your worries in this case. Taking out these three figures leaves enough material for an article that is worth publishing, so I do not see this as a significant drawback or a hindrance to the publication of the remaining results. The other features (O<sub>2</sub>-O<sub>2</sub> absorption and Rayleigh scattering) have spectral features that are much wider than 0.8 nm, and as such can be investigated with this instrument. I’m not enough of an expert to judge the methane absorption features in this spectral range, but I would consider these suspect as well.

This means a substantial rewrite of section 3.3, to reduce the section on how the O<sub>2</sub>-O<sub>2</sub> absorption was isolated from the oxygen absorption and Rayleigh losses. Please note that at line 286 it is mentioned that the extinction cross section of the oxygen monomer is linearly correlated with the oxygen concen-

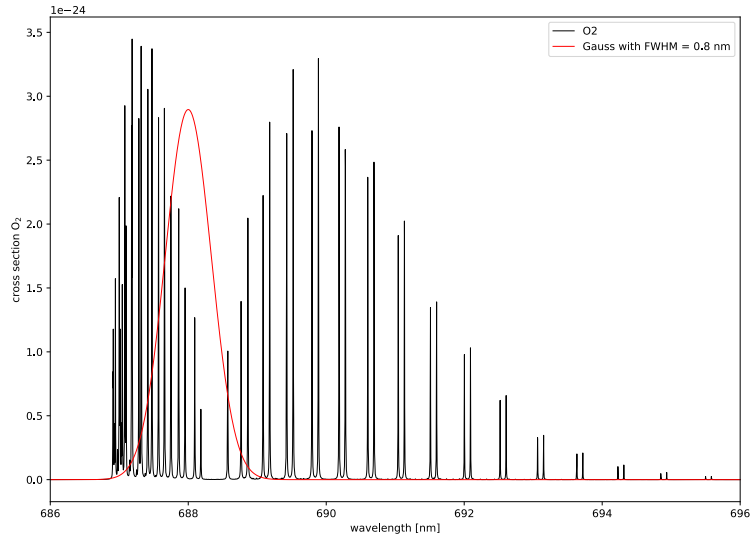


Figure 1: Oxygen B-band absorption cross sections from HITRAN. The modelled concentration is 21 % O<sub>2</sub> in N<sub>2</sub> at 1018 hPa and 294 K and includes pressure and temperature broadening. In red a Gaussian spectral response function with FWHM of 0.8 nm is shown.

tration. Be very careful with that statement, as the wings of the oxygen lines also show a  $p^2$  dependence due to pressure broadening.

At the end of section 3.3 there is a contradiction: on line 326 the 630 nm is listed as within 1.1 % of Thalman and Volkamer (2013), while on line 333 the same wavelength is listed as “this method cannot derive the cross-sections of CIA of O<sub>2</sub>-O<sub>2</sub> at 630 and 688nm”. Either alter the statement or explain better what is going on here.

I should note there that I don’t have access to He et al., 2018, so some of my remarks may have been covered there. However, some details on the experimental setup should be mentioned briefly here as an aid to the reader. I’ll indicate what I’d like to see added in the technical remarks below.

In the method section a lot of the equipment is mentioned. I’m missing some crucial information on the pressure sensor, the temperature probe and how the gas was mixed at specific mixing ratios, and the error introduced there. I do not see any discussion of the wavelength calibration. For the CRDS this is likely inherent in the used source (not mentioned here either) but for the BBCEs the calibration method for the spectrometer should at least be mentioned.

In section 2.2 I’m missing some details on the CRDS technique. There are a lot of methods to initiate a ring down of an optical cavity, and this details is not in the description. The reference (Bluvshstein et al., 2012) uses a 20 Hz Nd:YAG laser, whereas here the manuscript reads: “Over 1000 decay time measurements are monitored and averaged on a second basis”, suggesting that a much faster laser system was employed here.

In the conclusion (line 446 to 449) a reference is made to a wavelength range that is not part of this study at all. On line 441 nitrogen is explicitly mentioned as a gas of interest for atmospheric observations in particular for Lidar analysis, also including a wavelength that is outside the scope of the article. While I appreciate the outlook for future studies, please explicitly mark these statements as such, as they are not part of the conclusions of this study.

Finally, the choice of colours hinders accessibility for many colourblind people. There are combinations of colours for use in figures that will make the results more readable for colourblind people. Printing in black and white will quickly show where the use of colours should be improved.

## 1 Specific remarks

Line 43: please refer to the original year of publication in the reference (1899), not the year of the re-issue of the collected papers. Anyone familiar with the subject matter will be confused by Strutt 2009, but at the same time know immediately what Strutt 1899 is.

Line 50 (and several other locations throughout the manuscript): really weird line breaks.

Line 74: Nitrogen should have subscript “2” not “e”.

Line 99 in the Methods section: I found Bluvshstein et al., 2016 (doi:10.5194/amt-9-3477-2016) to be the end point of a chain of references for the method that starts with the two that are mentioned. I suggest to use this reference (in addition or instead of). Sending readers into a rabbit hole to chase the methods is not nice.

Line 226: In the results and discussion I read: “The reflectivity of the cavity mirrors, measured across the entire range using the difference in Rayleigh scattering of N<sub>2</sub> and He, was very stable throughout the experiments”. I expect to find this in the method section, how this was determined.

Line 231: the reflectivities and the losses have their relative order interchanged, please maintain order for readability.

Line 285: It is worthwhile to note what transitions of the O<sub>2</sub>-O<sub>2</sub> absorption features occur here, and especially that both molecules leave the interaction in an electronically excited state, at least for the shorter wavelengths.

## 2 Figures and captions

Line 466: Caption does not match labels in figure. Given the magnitude of the signal in the figure, the error is in the caption, not the figure.

Figure 3: missing units on the axes.

Figure 4: consider listing tabular material in a table rather than a figure legend.

Figure 5: the unit of panel (c) cannot be correct, there must be a density involved here.

Figure 6: the methane percentage is mentioned, but not the cell density for 100 % methane concentration.

Figure 6 & 7: suggest to add markers to figure 6 indicating where the wavelengths are that are shown in figure 7.

Figure S3: missing units on the axes

Figure S4: in black white the traces look identical.