The authors have addressed many of my comments since the first review, and the manuscript is definitely improved. However, I still have some significant concerns that need to be addressed before the paper can be published.

We thank the Reviewer for these comments and suggestions. We will address them in the replies below.

--Major Comments:

(1) My biggest concern is with the uncertainty. The highly structured reflectivity curve of the mirrors significantly limits the accuracy of the Rayleigh scattering cross sections in this study and therefore limits the conclusions that can be drawn from this work. The unfortunate reality is that the very thing that is newest about this paper (the very broad wavelength mirrors) is the thing that seems to cause the most problems. The limitations on accuracy due to the structured reflectivity is abundantly clear in Figure 9. The Pink data simply cannot be used to provide any constraint on distinguishing the green (previous work) and black (this study) fit lines, especially for SF6 and CH4. Perhaps the green and black fit lines are different enough for N2O to warrant producing a new dispersion relation. But for SF6 and CH4, the results of this study can only be used to say that they validate the existing n-based expressions.

Reply: We agree with the reviewer that the structure in the derived refractive indices that arises from the residual structure in the reflectivity curves is a limitation. However, we respectfully disagree that the data are therefore of no value to the literature, as implied by the statement that we could not distinguish our own fit from the prior data. That these two fits are indeed very close together, and both well within the error bounds estimated for the BBCES data, speaks to the accuracy of the BBCES data and the prior determinations. The agreement between the CRDS data and the BBCES data at 405 nm and 662 nm is 0.24% and 0.8%, both values well within the stated uncertainty of the BBCES data in the figure. CRDS is an absolute method that does not suffer from the apparent issue of the structured mirror reflectivity. Moreover, the large number of data points in the BBCES data easily compensates for the uncertainty in the structured mirror reflectivity when fitting a smooth function to the data. For the case of SF6, for example, there are 327 points in the BBCES data and 2 additional CRDS points. The total number of literature data points across this wavelength range is 8 prior to the recent data from Wilmouth, and 57 including these recent data. The sparseness of the literature does not allow one to assess if there may have been systematic errors in these measurements similar to the structured mirror reflectivity artifact in the BBCES data, so any potential artifacts of this nature in the literature data are simply unknown. The improvement in the refractive index fit on going from 57 literature data points to 57+329 = 386 is a factor of 2.5.

To illustrate the importance of the denser wavelength coverage, we present in the figure below the same refractive index fit using our raw BBCES data and a series of fits to more highly averaged data. As the averaging increases, the apparent structure in the data decreases, but the fit does not change. We have added these figures as a supplement to the paper in the discussion surrounding Figure 9. We also illustrate this in the revised manuscript at the end of Section 3.5.

"The structure in derived refractive indices that arises from the residual structure in the reflectivity curves is a limitation. However, the agreement between the CRDS data and the BBCES data at 405 nm and 662 nm is 0.24% and 0.8%, both values well within the stated uncertainty of the BBCES data. CRDS is an absolute method that does not suffer from the apparent artifact due to the structured mirror reflectivity. Moreover, the large number of data points in the BBCES data easily compensates for the uncertainty in the structured mirror reflectivity when fitting a smooth function to the data. Figure S5–7 show the refractive index fit using our raw BBCES data and a series of fits to more highly averaged data. As the averaging increases, the apparent structure in the data decreases, but the fit does not change." (Line 484-492)

1.SF_6

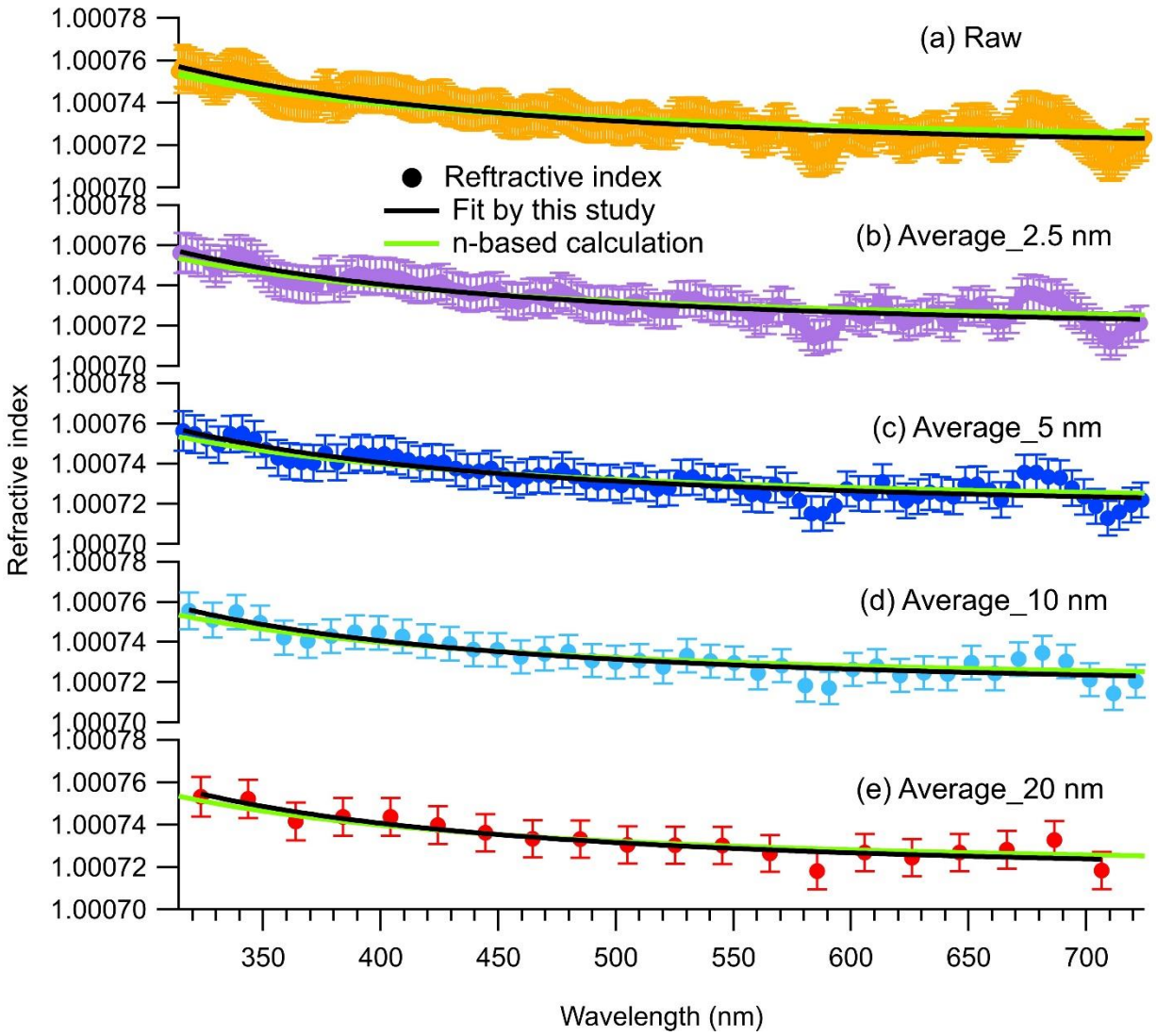

Table 1. The difference between the fit and n-based calculations

Types	Mean	Min	Max	STD
Raw	-5.28E-07	-2.26E-06	3.68E-06	1.57E-06
Ave_2.5	-5.11E-07	-2.17E-06	3.50E-06	1.50E-06
Ave_5	-5.37E-07	-2.26E-06	3.59E-06	1.58E-06
Ave_10	-5.30E-07	-2.26E-06	3.52E-06	1.60E-06
Ave-20	-3.07E-07	-1.92E-06	3.22E-06	1.51E-06

2.N_2O

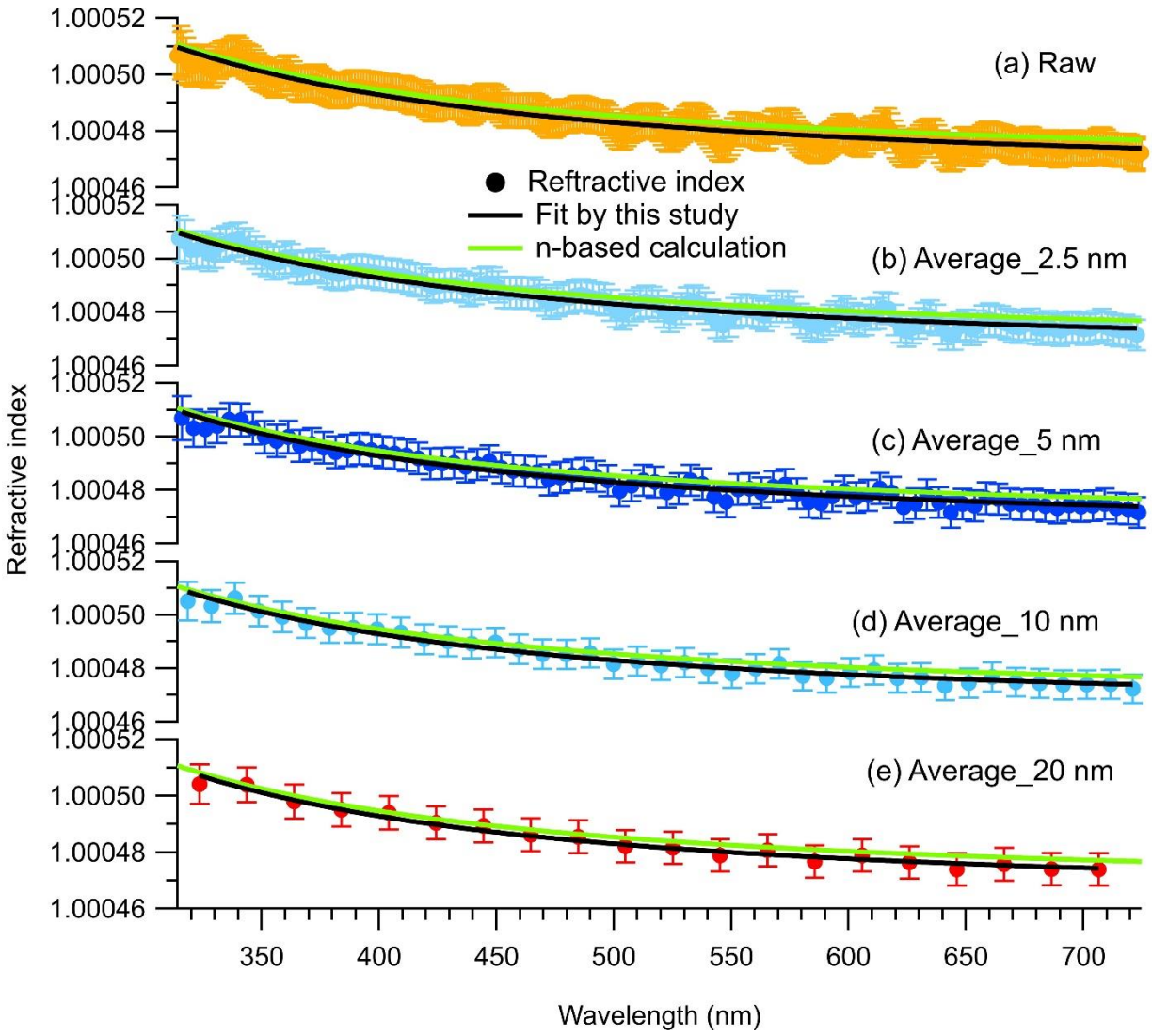

Table 2. The difference between the fit and n-based calculations

Types	Mean	Min	Max	STD
Raw	-2.30E-06	-2.87E-06	-9.79E-07	5.05E-07
Ave_2.5	-2.30E-06	-2.88E-06	-9.82E-07	5.09E-07
Ave_5	-2.32E-06	-2.91E-06	-9.76E-07	5.28E-07
Ave_10	-2.30E-06	-2.91E-06	-9.47E-07	5.50E-07
Ave-20	-2.26E-06	-2.81E-06	-1.09E-06	5.10E-07

3.CH_4

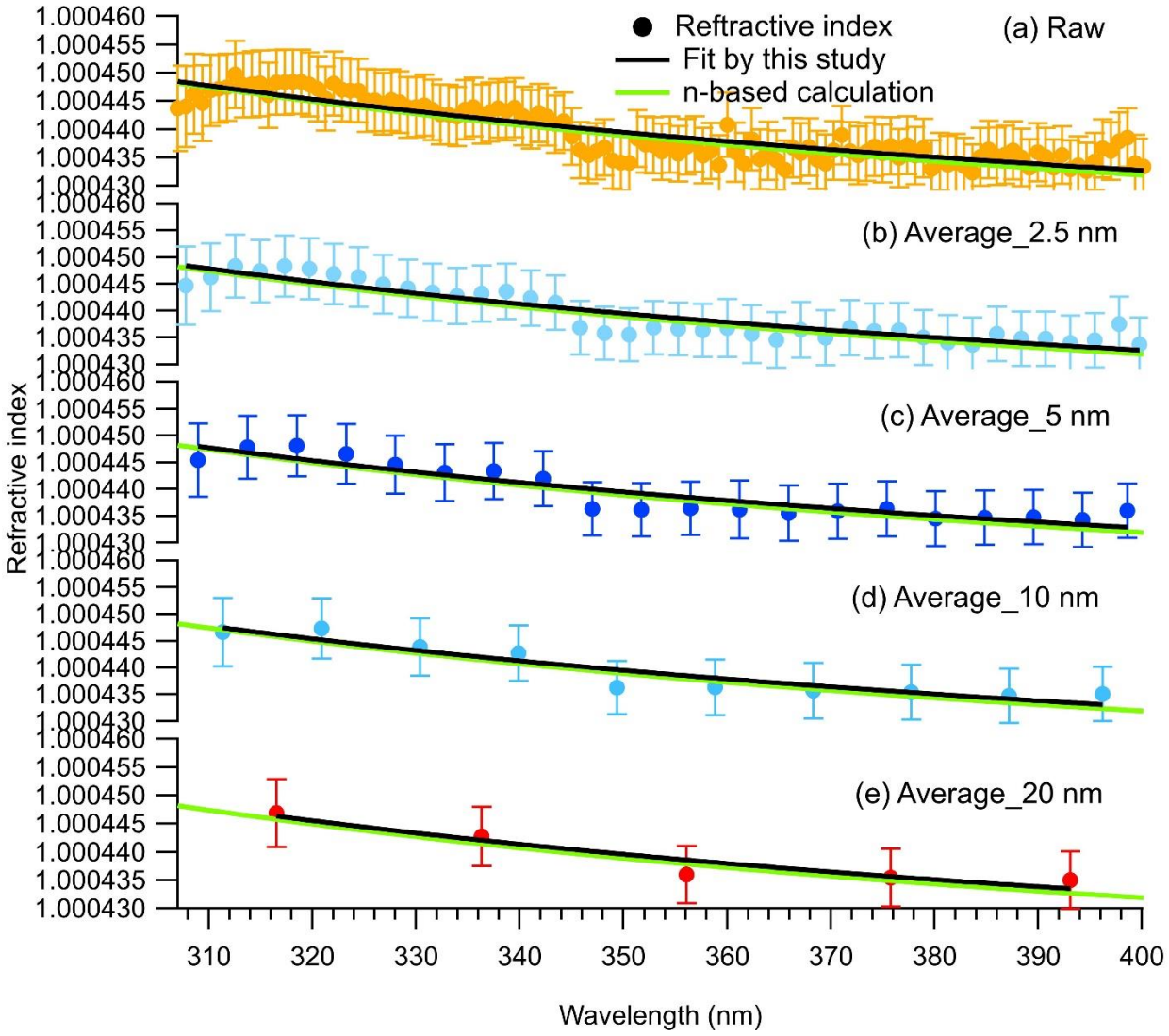

Table 3. The difference between the fit and n-based calculations

Types	Mean	Min	Max	STD
Raw	5.81E-07	2.87E-07	7.97E-07	1.47E-07
Ave_2.5	5.87E-07	4.32E-07	7.00E-07	1.40E-07
Ave_5	6.06E-07	3.22E-07	8.20E-07	1.54E-07
Ave_10	6.03E-07	3.94E-07	7.63E-07	1.24E-07
Ave-20	6.03E-07	5.90E-07	7.94E-07	8.14E-08

Moreover, as now detailed in lines 254-259 of the revised manuscript, there are additional uncertainties beyond those treated in the paper that the authors cannot quantify and did not even attempt to estimate. Because the uncertainty of the N2 reference cross section is unknown at wavelengths greater than 468 nm and because the mirror reflectivity is highly structured, the authors do not know their true uncertainties, and

the values in the paper represent a lower limit. Indeed, because N2 is used to determine the pathlength, all of the Rayleigh scattering cross sections reported in this study have unknown uncertainties >468 nm.

Reply: We respectfully disagree. Naus and Ubachs (2000) conducted CRD measurement of Rayleigh scattering cross section [$\sigma(v)$] for N_2 in the wavelength range of 560-650 nm. They also calculated the Rayleigh scattering cross section for N_2 based on the refractive index calculated from the dispersion relation developed by Peck and Khanna (1966) [$(n-1)\times 10^8 = 6498.2 + \frac{307.4335\times 10^{13}}{14.4\times 10^9 - v^2}, \lambda = \frac{1}{v} > 468\,nm$]. The Rayleigh scattering cross sections were fitted to a function of $\sigma(v) = \bar{\sigma} \times v^{4+\varepsilon}$, where ε is 0.0624 for N_2. The values for $\bar{\sigma}_{exp}$, obtained after several (weighted) fitting and data-analyzing procedures, allow for a comparison between the observed ($\bar{\sigma}_{exp}$) and the calculated values ($\bar{\sigma}_{cal}$) of the Rayleigh scattering cross section. They found that values for $\bar{\sigma}_{exp}$ is deduced with accuracies of ~1%. The cross sections calculated based on dispersion relation by Peck and Khanna (1966) and the dispersion relation used by this study agree with each other with an average difference of 0.08% (0% to 0.12%) in the wavelength range of 468-725 nm. Thus, the uncertainty of the n-based Rayleigh scattering cross sections used by this study is about 1%. Moreover, recent CRD measurements by Thalman et al. (2014) agree well with calculations based on the dispersion relation used by this study with relative differences within 0.5% (See the table below). All of this supports that the uncertainty of the Rayleigh scattering cross section of N_2 is about 1%. We add the following sentences in the manuscript as "Rayleigh scattering cross section of N_2 is validated up to 468 nm with an uncertainty of 1%. Rayleigh scattering cross sections measurement by CRDs agree well with those calculated from the refractive index with relative difference within 1% in the wavelength range of 468–650 nm (Naus and Ubachs, 2000; Thalman et al., 2014). Thus, an uncertainty of 1% was assigned for N_2 Rayleigh scattering cross section in the wavelength range of 307–725 nm." (Line 245-250)

We further point out that the BBCES measurements at 662 nm agree with the absolute CRDS measurements from this data set to within 0.9%. This agreement between the two measurements, one relative to the Rayleigh cross section of N_2 and the other absolute, demonstrates the accuracy of the BBCES data beyond 468 nm.

Rayleigh scattering cross sections ($cm^{-3} \times 10^{27}$) of N_2 from *n*-based calculations and CRDs measurements for selected wavelengths.

Wavelength	n-based	CRD measured	Relative difference (%)	Reference
405.8	16.146	16.1	-0.3	Thalman et al. 2014
532.2	5.297	5.32	0.4	Thalman et al. 2014
404	16.45	16.42	-0.2	This lab
662	2.181	2.18	0.0	This lab

Naus Hans, Ubachs Win. Experimental verification of Rayleigh scattering cross sections. Opt. Lett. 2000; 25(5): 347–9.
Peck Edson R., Khanna Baij Nath. Dispersion of Nitrogen. J. Opt. Soc. Am., 1966; 56:1059–63.

For these reasons, I would state even more strongly than in my previous review that there is no point in publishing new dispersion relationships in this paper that are indistinguishable within uncertainty from the dispersion relationships already in the literature. The authors take this approach for CO2, opting not to report a new dispersion relationship, and they should take a consistent approach with SF6 and CH4, given

that the uncertainties are too large to say anything other than the SF6 and CH4 measurements in this study agree with the dispersion relationships currently in the literature.

Reply: Once again, we respectfully disagree. We are in fact somewhat confused by this recommendation. Indeed, our data and analysis agrees with what is already in the literature. This is not a reason not to report it. Quite to the contrary, it is common practice to publish measurements of the same quantity made by different approaches to test the understanding of basic physical and chemical properties (see, for example, the kinetics literature (JPL and IUPAC panels)). Furthermore, our data set has greater spectral density and breadth of wavelength coverage than any currently available data set or in fact the sum of all of the literature reported to date. To simply not report a valid result arising from such a data set taken by a new and different technique would be negligent. This statement is valid regardless of the degree of agreement or lack thereof with the existing literature.

2) On Line 457, it is stated that the average deviations in the dispersion relationships between the literature and this study for SF6 and CH4 are 0.1% for both gases. On first read, this level of agreement would seem to be almost impossibly close given the large uncertainties in the present data set. But upon closer inspection, this remarkable agreement in reality is simply due to the fact that the authors are just repeating the method of Wilmouth and Sayres (2020) for determining the dispersion relations and adding in their data from this study. Therefore, the 0.1% agreement essentially just means that one gets the same dispersion relation using the Wilmouth and Sayres (2020) method for SF6 and CH4 regardless of whether or not the data from the present study is included in the fit. Again, there is no justification here for providing new dispersion relationships for SF6 and CH4.

Reply: We have added a sentence clarifying how this was done in the revised manuscript as **"**The fit performed by this study combines the data set used by Wilmouth and Sayres and the data acquired by this study. These small deviations support that our data acquired by the BBCES at a wide range agrees well with literature data.**"** (Line 472-474)

(3) The authors have a new approach from their previous manuscript of presenting the differences between their Rayleigh scattering cross sections and the n-based values as x +/- y%, as in lines 22-23 of the Abstract and in several places in the text. For example, for CO2, the difference is listed at 0.37 +/- 1.24%. Some explicit discussion when this approach is first presented (around line 306) regarding why the standard deviation is so much larger than the mean would be helpful. There are also too many significant figures being used in this approach. I recommend at most 2 significant figures rather than 3, e.g., with CO2, it should be listed at 0.4 +/- 1.2%.

Reply: Thank you for this suggestion. We agree that it is reasonable to keep one figure after decimal point. We changed this in the revised manuscript. (Line 22-23, 257, 260, 294, 295, 317, 324, 386, 426, 500, 501)

The relative difference (RD) was calculated as RD = (measured value-n-based calculation)/n-based calculation. As we can see from the figures, the measured cross section can be bigger or smaller than the n-based values. Thus, we have positive and negative values for the RD. This causes the mean of the RD to be smaller than the standard deviation. Statistically, there is no direct correlation between the mean and the standard deviation value.

(4) Based on lines 241-247, the authors appear to be using their precision as if it were their accuracy. The variation in the temperature and pressure over the 150 scans is the precision, not the accuracy. This new

text now makes it clear why the pressure uncertainty is listed at the unrealistic value of +/-0.01%, as I noted in the previous review. I would be extremely surprised if the pressure gauge used in this study were capable of 0.01% accuracy. Please update the uncertainties with reasonable estimates for accuracy, not using the precision.

Reply: We re-calculated the uncertainty of our data. That is, we calculated the overall uncertainty as the root sum squares of the standard deviation of the measurements, and the uncertainties in the effective pathlength, the temperature, the pressure, and the photon counting uncertainty in the spectra. We revised Section 2.4 as follows:

"The uncertainty for BBCES measurements can be assessed by the propagation of the errors associated with the measurements. Rayleigh scattering cross section of N_2 is validated up to 468 nm with an uncertainty of 1%. Rayleigh scattering cross sections measurement by CRDs agree well with those calculated from the refractive index with relative difference within 1% in the wavelength range of 468–650 nm (Naus and Ubachs, 2000; Thalman et al., 2014). Thus, an uncertainty of 1% was assigned for N_2 Rayleigh scattering cross section in the wavelength range of 307–725 nm. Each parameter (temperature, pressure, light intensity) was measured 150 times for each gas. The standard deviation of the measurements (<0.3%) is combined with the uncertainties in the pressure (±0.3%), temperature (±0.1%), 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 effective pathlength curve of ±1.1%. This uncertainty is further propagated to the target gas by consideration of the uncertainties of pressure (±0.3%), temperature (±0.1%), and spectral intensity (⩽0.2%) of the target gas measurements, and the standard deviation of the measurements (<1.2%). The overall 1-σ uncertainty of the gas extinction cross section is within 1.7%.**"** (Line 245-260)

We also updated the figures accordingly and relevant text in the manuscript.

(5) As stated in (2) above, the dispersion relationships for SF6 and CH4 in this study appear to have been calculated using the exact same method and data sets as Wilmouth and Sayres (2020) except that the data from the present study were included in the fit, and "all sets of data were weighted equally" according to the text. This latter point is stated as if it were important, but it is never actually explained in the text what weighting the data sets equally means.

Does weighting the data sets equally mean that the points from all of the data sets were combined, and then the fit obtained? If so, then the data set with the most points will have far more weight than the ones with fewer points, such that they will not be truly weighted equally. More specifically, with this approach, the Vukovic, Watson, and Cuthbertson studies will be mostly irrelevant in the fits because the BBCES studies have so many more data points.

Alternatively, weighting them equally could mean that each data set was normalized according to how many points were in the data set, such that each study got equal weight. The downside of this approach is that the older Watson and Cuthbertson studies are probably less accurate, but they would be given equal weight with the newer studies.

Either way, it should be clearly stated what was done. Or alternatively, as strongly recommended above, delete the dispersion relationship calculations altogether for the reasons outlined in points (1) and (2).
Reply: During the fitting, we weighted each data point equally instead of weighting each data set equally. More specifically, the points from all of the data sets were combined, and then the fit was obtained. We agree with the reviewer that Vukovic, Watson, and Cuthbertson studies will weigh less in the fitting due to

fewer data points from these studies. Since the BBCES measurements cover a much wider wavelength range, these results which contain more data points should have heavier weight during the fitting. We revised this in the manuscript as "All data points were weighted equally." (Line 445)

--Additional Comments/Corrections:

Line 72 and many other Lines in the manuscript: I mentioned this in my previous review, but the problem persists. The authors use "BBCES" as if it were the name of their experimental setup. If the sentence doesn't make sense with "Broadband Cavity Enhanced Spectroscopy" substituted in place of "BBCES", the sentence should be edited. In general, anywhere in the paper that the expression "the BBCES" or "our BBCES" is written, it is incorrect. For example, "the BBCES setup" or "our BBCES instrument" could be stated instead.

Reply: We thank the reviewer for this suggestion. We agree that the results are obtained from the "BBCES system" instead of only "BBCES". The same case for CRD. We changed accordingly throughout the revised manuscript.

Lines 282-284. These values are not the "1-sigma uncertainty" as stated in the text. They appear to be from a root sum of squares calculation that includes the N2 reference cross section with the precision of the temperature, precision of the pressure, and precision of the test gas spectra. The true 1-sigma uncertainty is much large than the numbers presented in this sentence.

Reply: In the revised manuscript, the 1-sigma uncertainty was calculated as the root sum square of the uncertainties for the effective pathlength (1.1%), the temperature (0.3%), the pressure (0.1%), the photon counting of the spectra (0.2%) and the standard deviation of the measurements (<1.2%). We updated the text and figures accordingly. (Line 257-260, 294-295, Figures 1,5,8,9)

Line 429 and Line 438: Saying "for 288.15K and 1013.25 hPa" is ambiguous as written in these sentences, as it sounds like the measurements were made at these conditions. These T and P values should appear a bit later with the dispersion relationships to be clear that n is being defined at 288.15K and 1013.25 hPa in order to be consistent with past studies.

Reply: To make this sentence accurate, we changed it to "In our study, the refractive index of SF_6 in the wavelength range of 307-725 nm was calculated from the measured Rayleigh scattering cross section. The calculated refractive index was scaled to 288.15 K and 1013.25 hPa to be consistent with past studies." (Line 440-442)

Line 430: Delete "better" and Line 431: Delete "alternative". This is the standard form of the fit; it is not better or alternative to what has been done before.

Reply: These two words were deleted in the revised manuscript. (Line 442, 444)

Line 447: To avoid ambiguity, replace "their refractive index" with "Sneep and Ubachs (2005)".

Reply: Replaced. (Line 460)

Line 475: Please repeat the list of gases in order before “respectively”. As it is, this sentence is referencing a gas list from two sentences prior.

Reply: Added. (Line 501-502)

Table 1: The He equation is still incorrect. There is a missing decimal in 1.8102.

Reply: Thanks for the careful reading. This has been revised.

Figure 5b: Missing right parenthesis in figure caption

Reply: We added the right parenthesis in panel (b)

Figure 9, panel (c): Is the green fit line plotted for CH4? It is not visible on the plot.

Reply: It is plotted. However, the green fit line overlaps with the black fit line.

--Minor Corrections:

Line 80: Change “is” to “are”

Reply: Changed. (Line 82)

Line 84: Add “applicable” after “CH4” and change “264” to “250”

Reply: Changed. (Line 86-87)

Lines 135-136: Delete the sentence beginning “In this study…” as it simply repeats what was just stated on Lines 126-127.

Reply: This sentence was deleted in the revised manuscript. (Line 139-140)

Lines 273-275: The sentence beginning “The mean uncertainty…” could be deleted as it repeats what was already stated on Lines 246-247.

Reply: This sentence was deleted in the revised manuscript. (Line 283-286)

Line 292: Change “listed” to “lists”

Reply: Changed. (Line 303)

Line 299: Add “, 2020” after “2019”

Reply: Added. (Line 310)

Line 301: Change “calculation” to “calculations”

Reply: Changed. (Line 312)

Line 309: Define variables in the equation

Reply: The sentence has been changed to “…described in the form of $\sigma = A \times \lambda^B$, where σ and λ are the cross section and the wavelength.”. (Line 320-321)

Line 310: Change “fitted” to “fit”

Reply: Changed. (Line 321)

Line 359: Change “slops” to slopes

Reply: Changed. (Line 371)

--Note
A few of the changes indicated in the authors’ response to the previous review were never actually made in the revised manuscript.

Reply: We thank the reviewer for point out this. We checked the manuscript and updated the manuscript.

1. In the abstract, we changed the last sentence to “This study provides dispersion relations for refractive indices, n-based Rayleigh scattering cross sections and absorption cross sections based on more continuous and more extended wavelength ranges that available in the current literature.” (Line 26-29)
2. Change 630 nm to 629 nm. (Line 388)
3. Change “Figure 9b” to “Figure 9c”. (Line 465)

--Final Suggestion

If someone were to summarize what’s new about this paper in a single statement, it would be that the measurements were made with such broad mirrors. I think some perspective text at the end of the paper discussing pros and cons of using mirrors like these would be appropriate. It seems clear that there is a tradeoff one must make here between speed (acquiring a broad wavelength range quickly) and accuracy (the structured reflectivity curve is limiting).

Reply: We agree with this suggestion. While the BBCES instrument provides data at a wide wavelength range, the structured mirror reflectivity also induces some uncertainties to the wavelength ranges where the reflectivity changes significantly. We add this in the revised manuscript as “We also note that while acquiring data at a broad wavelength range quickly using a single BBCES instrument, uncertainties were also observed in the wavelength ranges where the mirror reflectivity changed significantly. However, with appropriate averaging, this can be minimized without compromising the accuracy. There is a tradeoff between obtaining data at a wide wavelength range and ensuring high accuracy data. New mirrors with a smoother reflectivity curve will improve the performance of BBCES instrument.” (Line 540-546)