

## Reviewer #1

We thank the reviewer for their helpful and constructive comments. Our responses and revisions to the manuscript are outlined in detail below.

**Author General Comment:** Several of the reviewer comments refer to the estimated uncertainties presented in our manuscript. Estimating uncertainties in the atmospheric metrics, although important, has generally been ignored in the literature to date. However, we believe that uncertainties should be a part of the discussion. A major hurdle in such a discussion, however, is that it is difficult to quantitatively estimate uncertainties in estimated quantities. In our manuscript, we have taken the approach of estimating uncertainties based on the performance obtained using the HCFC training dataset, i.e., what we call average behavior. We acknowledge that not all molecules follow “average” behavior, but the intention is that our analysis will provide a general perspective on the issue of metric uncertainty. Note that we emphasize that direct fundamental laboratory studies are preferred. However, in the absence of laboratory data theoretical methods can provide reasonably reliable results.

There was a misinterpretation by this reviewer of the range of GWPs reported in Table 2 as the uncertainty range of our analysis. The reported range is indeed large, which reflects the fact that the isomers of a HCFC family have different properties, while the uncertainties for the individual isomers is estimated to be much less, ~30-50%. This misinterpretation represents a thread for several of the other reviewer comments. We have revised the text in an attempt to avoid this misinterpretation by others.

A general comment from this and other reviewers was a lack of a historical perspective and citations for the development of applying theoretical methods to the quantitative calculation of infrared spectra, which was a major component of our study. To address this issue, we have added text to Section 2.3 that is relevant to the development of the theoretical methods and have cited some of the previous applications from our laboratory as well as from others as examples.

**Reviewer Comment:** The paper is a study by Burkholder et al. to synthetically estimate atmospheric rate constants (using certain models and approximations to estimate the atmospheric loss processes), as well as to use (*ab initio*) synthetic calculated infrared absorption spectra so as to ultimately derive estimates for the lifetimes and global warming potentials for a series of the C1-C3 hydrochlorofluorocarbons contained in the Kigali amendment to the Montreal protocol.

**Author Comment:** A minor point, the theoretical methods in our work used density functional theory (DFT) not *ab initio* methods.

**Reviewer Comment:** In that amendment, when accounting for all of the C1 to C3 conformers of the HCFCs, there are 274 species of interest, but of these only 15 have both the actual measured relevant kinetic rate constants and requisite quantitative infrared absorption spectra to derive accurate values for the RE and ultimately the associated GWP. It will take many years and more likely many decades until most or all of the experimental data are acquired for these 274 species, so any efforts to derive some interim values using estimates, models and theoretical calculations are clearly justified. Such is the motivation for the present work. The paper thus represents a seminal contribution for estimating the GWP values for these 259 species; it also represents a very great deal of work. To write such a paper requires significant expertise in

kinetics, atmospheric chemistry and modelling, IR spectroscopy, quantum chemical calculations, etc. The paper should clearly be published. We thus recommend publication with only minor modification.

**Author Comment:** We truly appreciate the reviewer comments. We would like to clarify, however, that the 274 species is the total number of “isomers” for the C1-C3 HCFCs. Each isomer may have a number of “conformers” (i.e., molecular configurations with slightly higher energy), that were included in our metric determinations. We considered conformers within 2 kcal/mole of the lowest energy geometry, which enables us to account for at least ~98% of the Boltzmann population distribution.

**Reviewer Comment:** This reviewer does have concerns leading us to criticise certain aspects of the study. Without pointing to a specific paragraph, the overall hue or colour of the narrative gives the reader an impression of the resultant data or GWP values indicate greater precision and accuracy than is perhaps warranted: The phrase “... such estimates are accurate enough for our study” is used quite often. The paper is in essence a (warranted) theoretical study that of necessity involves many approximations or extrapolations to derive certain data which are used with other data to calculate the lifetime and GWP values. Many of these data have non-trivial uncertainties and represent possible sources of significant error. However, in several places phrases to the effect “this study provides reliable policy relevant GWP metrics” imply significant accuracy. For example, inspection of column 5 in their Table 2 manifests their own estimates as the 100-year GWP values with ranges that at first blush appear to vary from 5x to 35x between the lower and upper bounds! While the data are clearly needed, the language should perhaps be modified to recognize the limitations of such calculated data.

**Author Response:** The reviewer has misinterpreted the range of values for the isomer GWPs presented in column 5 of Table 2 as the uncertainties associated with a given HCFC chemical formula. Second, the reviewer makes a reasonable point that some of the uncertainty statements in our paper are not quantitative. We have made an effort in this manuscript to convey to the reader that the metrics reported in this work are estimates that have uncertainty associated with them (the consideration of uncertainties in this type work is an issue often overlooked, or ignored, in previous studies and assessments). That said, it is not straightforward to assign accurate statistical uncertainties for molecules with “unknown” properties. However, we should be more careful in using phrases such as “suitable for this study”, which was meant to imply that these values represent a minor uncertainty in comparison with other sources of uncertainty. In our approach, we have recommended “average” uncertainties based on our comparisons with literature data, i.e., our training dataset. Note that metrics derived from accurate laboratory measurements still will lead to metric uncertainty, which can be substantial, due to the methods used to determine atmospheric lifetimes and radiative properties. For example, the well-mixed radiative efficiency estimation method given in Hodnebrog et al. (2013) is estimated to have a 25% uncertainty associated with the method. These other sources of uncertainty were not included in our analysis.

**Author Action:** We have removed the subjective “reliable” in several places as follows:  
*In the Abstract:* The results from this study provide ~~reliable~~ policy relevant GWP metrics for the HCFCs included in the Montreal Protocol in the absence of experimentally derived metrics.

*In the Introduction:* The objective of the present work is to provide a ~~reliable and~~ comprehensive evaluation of the atmospheric lifetimes, ozone depletion potentials (ODPs), GWPs, and global temperature change potentials (GTPs) for the HCFCs listed in Annex C of the amended Protocol.

*Section 2.3:* Similar approaches have been used in earlier studies for other classes of molecules with ~~reliable~~ good results, see Hodnebrog et al. (2013) and references cited within.

*Section 3.1:* This method of RE determination is, therefore, expected to provide ~~reliable~~ good estimates of REs in the absence of experimentally based determinations.

*Summary:* Although this work has provided ~~reliable a comprehensive set of estimated~~ of key metrics for the C<sub>1</sub>-C<sub>3</sub> HCFCs that presently do not have experimental data, careful direct fundamental laboratory studies of an intended HCFC would better define the critical atmospheric loss processes (reaction and UV photolysis) used to evaluate atmospheric lifetimes.

We have revised the use of “accurate” as follows:

*Section 2.1:* Therefore, this method of accounting for stratospheric loss is ~~acceptably accurate for our purpose~~ leads to only a minor uncertainty in the calculated global lifetime.

*Section 2.1:* As shown later, the O(<sup>1</sup>D) reaction is a minor loss process, <1%, for nearly all the HCFCs included in this study and, therefore, the estimation method used is not critical as this loss process is a minor contributor to the global lifetime ~~are sufficiently accurate for our study~~.

**Reviewer Comment:** Related to this phraseology aspect is the need for their intro/summary text to point out that more and better experimental data of several different kinds are needed: 1) Experimental measurements of atmospheric lifetimes are clearly needed; the laboratory of Dr. Burkholder and others are known for careful measurement of such rate constants and these are clearly warranted – while the SARs are good approximations, actual experimental data are required. 2) Even for relatively simple molecules such as the smaller HFCs, the *ab initio* Gaussian calculations still can vary significantly from the measured infrared cross sections (see below), and these IR data need to be measured experimentally as well, including to longer wavelengths as the authors point out. Current experimental IR absorption data can have accuracies of 3% or better. 3) Whilst measurement of such kinetic and IR absorption data for all of the 259 named molecules will likely not occur soon, even increasing the number of species in the training set (Table 1) from 15 to 25 or 35 or ? would obviously increase the accuracy and relevancy of the study. Such considerations need to be suggested in either the introduction or discussion sections, or both. A more appropriate slant to be taken throughout the manuscript would thus be words to the effect: “In the absence of such experimental data, the following study provides estimates of...”

**Author Response:** We agree with the reviewer sentiment whole heartedly. For compounds of interest direct careful laboratory studies are surely preferred. We have expressed this point of view in both the introduction and conclusion sections of our paper. It is also implicit in our approach that an improved (expanded) training dataset would be beneficial to the analysis presented in our work.

**Author Action:** None

**Reviewer Comment:** One other general comment is that in several spots the paper would do better to simply flesh out some of the details as to how the results and approximations are realised. This is especially true for the experimental and calculation sections. However, their error analysis is likely the best approach for a study of this magnitude.

**Author Response:** The results and approximations used in our work are based on the behavior of the training dataset, which for HCFCs is rather limited. We present our estimated uncertainties based on the “average” behavior (as discussed above) of the training dataset, although outliers are acknowledged. Note that the Betowski et al. study used a much larger training dataset that included a variety of classes of molecules to derive general properties that were applied to HCFCs. We argue that this approach resulted in a bias in their HCFC results (note that their theoretical results and those presented in this work are in good agreement).

**Author Action:** None

**Reviewer Comment:** Finally, the section of the manuscript that deserves a bit more attention is section 2.3 where the intensities of the calculated IR spectra are compared to the measured IR spectra over the spectral region 2000-500  $\text{cm}^{-1}$ . One initial question is how do the computed LWIR intensities compare with known values? Moreover, while not an expert in the Gaussian calculations, this reviewer fears that the 20% estimate as to the uncertainty in the IR absorbance spectra may perhaps be a bit too generous. The vibrational frequencies can now routinely be calculated with an accuracy of ca. 1 to 2% using such a basis set, and this is sufficient except in those cases where the predicted line frequency (depending on accuracy) is either adjacent to or obscured by an atmospheric absorption bands due to H<sub>2</sub>O, CO<sub>2</sub> or O<sub>3</sub>. However, the IR absorption intensities are far more difficult to predict with such accuracy - the eigenvalues (frequencies) to the nuclear motion equation are far less susceptible to approximations in the wavefunction than are the vibrational intensities (the eigenvectors). Besides the 20% estimate obtained from the training set, a more extensive discussion of the accuracy of the Gaussian results is likely justified.

**Author Response:** A thorough discussion of the uncertainties associated with theoretically calculated infrared absorption spectra is relevant to this work, and that of others, but is well beyond the scope of our manuscript. Because of the volume of calculations required in this work, we have used DFT methods that yield reasonable results without too much computing cost. We have based our uncertainty estimates on how well the DFT methods work for the training dataset. Future studies could build on the work presented in this manuscript. We have added text to the beginning of Section 2.3 to provide better perspective for the calculation methods (text given below). The next comment is related to this one.

**Reviewer Comment:** The authors have used DFT methods (B3LYP/6-31G(2df,p)) to estimate total IR cross sections, and have compared their results with a training set of molecules, to show that they are confident that their estimated cross sections are within about 20% of measured cross sections, for those that have experimental values. Their claimed accuracy would be made stronger if they had also compared their B3LYP method with a few standard small molecules that have been used to assess absolute accuracy of computed IR cross sections, as in Jemenez-Hoyos et al., PCCP, 2008, 6621-6629, and Halls and Schlegel, J Chem Phys, 1998, vol 109, 10587-10593, where the standard deviation of B3LYP computed IR cross section (by individual band) is shown to be about 11 km/mol (conversion to cross section is certainly known by the authors) for a set of molecules whose band strengths extend from less than 10 km/mol to about 90 km/mol. In addition

to these, more recent work has shown that for a set of alkanes, the absolute errors for IR band strengths were shown to be less than about 15% for B3LYP/6-31+G(d,p) computation compared to experiment, Williams et al., JQSRT, 2013, vol 129, 298-307.

It would be beneficial for the authors to review / cite some papers that have discussed the accuracy of the IR vibrational intensities, especially those that are conformer dependent. [A quick review of the literature pulled up the references listed below.] A similar discussion as to “quick and dirty estimate” may also be justified for two other areas: i) the neglect of the  $^{37}\text{Cl}$  isotopomers and ii) the SAR relationships used, as discussed on p. 7., lines 110 to 114.

**Author Response:** We had not reviewed the history of theoretical calculations and the application to molecules of atmospheric interest in our original submission. The other reviewers have also requested that this be included in our revised manuscript along with citation of some application papers.

**Author Action:** We have added “new” text with ample literature citations at the beginning of Section 2.3 “Theoretical Calculations”. The text includes the papers suggested by the reviewer as well as additional relevant material and some examples of work from our lab and others. We have also added the following text to address not including calculations with Cl-atom isotopes (note that including all permutations of Cl-atom isotopes would have expanded the depth of the present work tremendously, while not altering the general conclusions from this work): “In principle, substitution of  $^{35}\text{Cl}$  by  $^{37}\text{Cl}$  in a heavy molecule would lower the frequency of the C-Cl stretch by  $\sim 3\%$ ”.

**Text added to the start of section 2.3:**

Information about molecular vibrational frequencies, central to the interpretation of infrared spectra, thermodynamics, and many other aspects of chemistry, became amenable to computational determination in the early 1980s. It was recognized that computed harmonic frequencies derived via the second derivative of energy as a function of atomic position were systematically higher than observed fundamentals and scale factors were introduced (Hout et al., 1982; Pople et al., 1981). For Hartree-Fock frequencies these were typically  $\sim 0.9$  and accounted both for the influence of anharmonicity and deficiencies in the underlying quantum calculations. Frequencies based on methods incorporating electron correlation such as CCSD, CCSD(T) or certain functionals within density functional theory (DFT) often perform well for harmonic frequencies and are scaled by  $\sim 0.95$  to match fundamental vibrational modes. Such scaling has been updated as more methods appear (Alecú et al., 2010; Scott and Radom, 1996). Rather less information is available concerning the evaluation of absorption intensities for fundamental modes. Within the same harmonic approximation, implemented in popular quantum codes, the intensity is proportional to the square of the derivative of the dipole moment with respect to position. Halls and Schlegel evaluated QCISD results against experiment and their plot indicates deviations of up to around  $\pm 20\%$  and then used QCISD as a benchmark to evaluate a range of functionals (Halls and Schlegel, 1998). For B3LYP, they found differences from QCISD of around 10%. More recently, tests of the B3LYP functional found good performance for frequency and intensity (Jiménez-Hoyos et al., 2008; Katsyuba et al., 2013). Some prior work where similar methods have been applied to the infrared absorption for molecules of atmospheric interest include studies of fluoromethanes (Blowers and Hollingshead, 2009), unsaturated hydrofluorocarbons (Papadimitriou and Burkholder, 2016; Papadimitriou et al., 2008b), perfluorocarbons (Bravo et al., 2010), chloromethanes (Wallington et al., 2016),  $\text{SO}_2\text{F}_2$  (Papadimitriou et al., 2008a),

permethylsiloxanes (Bernard et al., 2017), and large survey studies such as by Kazakov et al. (Kazakov et al., 2012) and Betowski et al. (Betowski et al., 2015) to name a few.

Halls and Schlegel noted that real spectra may exhibit the influences of resonances, intensity sharing, and large-amplitude anharmonic modes. These can be partially accounted for in an analysis based on higher derivatives of the energy and the dipole moment, performed for instance within the framework of second-order vibrational perturbation theory (Barone, 2005). Advantages include treatment of resonances among vibrational levels and incorporation of overtones and combination bands. Examples of applications to molecules containing C-H and C-F bonds indicate excellent accord with experiment for band position and intensity, (Carnimeo et al., 2013) but for CH<sub>2</sub>ClF the intensity in the region involving C-Cl stretching nevertheless exhibits intensity errors of ~10% (Charmet et al., 2013).

**Additional text added within the section:**

We are not aware of prior studies of infrared spectra of HCFC conformers, but there have been prior theoretical studies of the conformers of other classes of molecule, such as for validation of observed infrared spectra used to deduce relative energies of carbonyl conformations (Lindenmaier et al., 2017) and comparison with measured infrared intensities for linear alkanes (Williams et al., 2013). The different errors and their trends for the intensities of C-H stretching and HCH bending modes indicate that a simple scaling approach, so successful for frequencies, will not work for intensities.

**and**

In fact the intensities of C-Cl stretches are a long-known problem for calculation (Halls and Schlegel, 1998).

**Minor Suggestions / Errata**

**Reviewer Comment:** Title: Perhaps insert “Estimates” into the title to read: “Global Warming Potential Estimates for the C1-C3 Hydrochlorofluorocarbons (HCFCs)...”

**Author Response:** Agee

**Author Action:** Title changed as follows: Global Warming Potential Estimates for the C<sub>1</sub>-C<sub>3</sub> Hydrochlorofluorocarbons (HCFCs) Included in the Kigali Amendment to the Montreal Protocol

**Reviewer Comment:** Pg. 3 line 61: Perhaps change first sentence to read “Due to the current dearth of such experimental data, the objective of the present work is to provide a best-effort and comprehensive evaluation...”

**Author Response:** We have used the phrase the “in the absence of experimental data...” in our manuscript, which we believe is a concise and accurate description.

**Author Action:** None

**Reviewer Comment:** p. 4. Line 63: Define the acronym GTP. 1, sent.21: In previous studies, emission factors have units of g kg<sup>-1</sup>, here the emission factors have units of g km<sup>-1</sup>. Please explicitly define the emission factor that you are estimating somewhere in the manuscript.

**Author Response:** GTP is defined on its first use on page 2. We are not addressing or discussing emission factors in our manuscript. So, we don't understand the origin of this comment.

**Author Action:** None

**Reviewer Comment:** p. 7 line 120: Insert “red” between “solid” and “symbols”

**Author Response:** This refers to the caption for Figure 1. Red is not necessary as there are only solid and open symbols on the graph.

**Author Action:** None

**Reviewer Comment:** p. 8 line 134–148. Almost every sentence in these two paragraphs mentions another source of error. Can the composite uncertainty be estimated at the end of this section 2.1?

**Author Response:** There are estimated uncertainties in the lifetimes and calculated infrared absorption spectra that propagate into the metrics as discussed in this text. Therefore, each parameter is considered separately and then combined to obtain an estimated uncertainty in the metric at the end of the discussion. Seeing that the uncertainties are estimated values, we feel that reporting uncertainty values in a table would place too much emphasis on the accuracy of these estimated values.

**Author Action:** None

**Reviewer Comment:** p. 8 line 153: It would be well to not only define FRF, but explain its meaning

**Author Response:** We have not included background material on the derivation or meanings of lifetime, ODP, RE, GWP, GTP, and FRF in our manuscript. Instead, we have provided the pertinent references that provide the detail necessary to fully understand these metrics.

**Author Action:** None

**Reviewer Comment:** p. 10 line 206. Is the comparison of experimental and predicted IR spectrum a “typical” result or is this one of the better matches?

**Author Response:** This comment refers to Figure 3. This result is typical. A comparison of the individual experimentally reported spectra and our calculated spectra for all molecules with experimental data is provided graphically in the SI.

**Author Action:** We have added the follow text following the introduction of Figure 3: “A comparison of experimental and theoretical spectra for all molecules with experimental data is provided in the SI (see Section 5).”.

**Reviewer Comment:** p. 11 Figure 3: Where do the experimental data come from in this Figure? One of the references?

**Author Response:** This comment applies to both Figure 3 and 4. The source of the experimental spectra are given in Table 1.

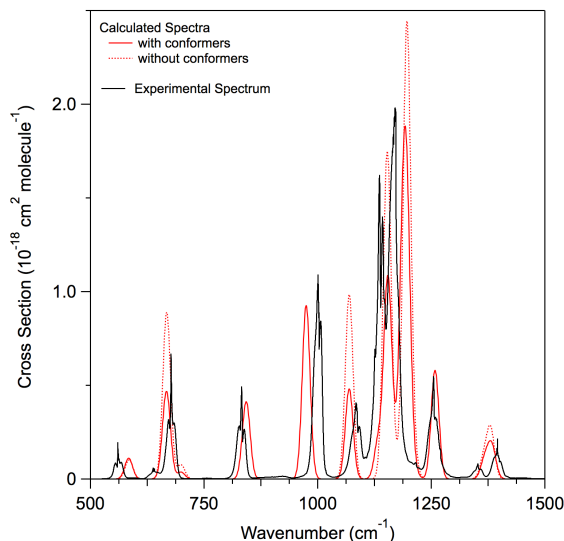
**Author Action:** We have reiterated the source of the experimental spectra in the captions for Figures 3 and 4 as follows: “... (see Table 1 for the source of the experimental spectrum)”.

**Reviewer Comment:** p. 11 Figure 3: The figure could facilitate a better comparison of predicted v. measured intensities if the y-axis were equally stretched in the two frames such that they were at the same factor, e.g. that 2.0 E-18 units were the same length in the top and bottom panels.

While quite weak, it appears there are two small bands in the experimental data near ca. 610 and ca. 1385  $\text{cm}^{-1}$  that are missed in the Gaussian calculations.

**Author Response:** The Gaussian calculation presented here considers only the fundamental vibration frequencies. Therefore, it is possible that weak combination or overtone bands are not included in the calculated spectra. The overall quality of the literature infrared absorption reference spectra was not explored as part of our study. The other reviewers have request that the panels for this figure be combined.

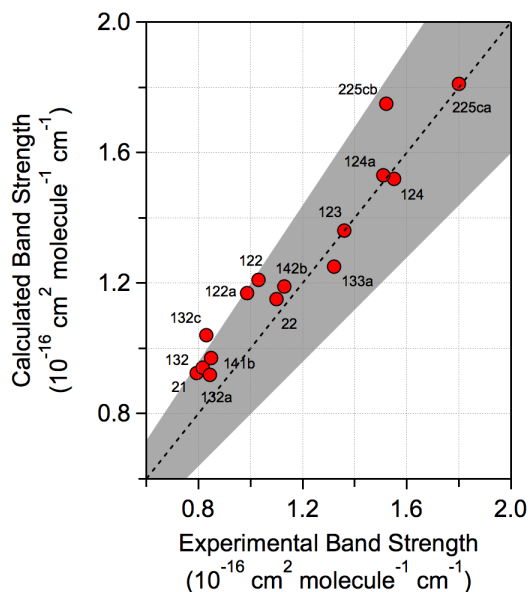
**Author Action:** Figure revised as follows:



**Reviewer Comment:** p. 12 Figure 4: It would be helpful to put the units ( $\text{cm}^2 \text{ molecule}^{-1} \text{ cm}^{-1}$ ) directly on the plot of this figure, not just in the legend.

**Author Response:** Okay

**Author Action:** Units removed from figure caption and figure revised as follows:





**Reviewer Comment:** p. 14 line 294: This sentence seems perhaps unjustified: While there are far fewer data for bands below  $500\text{ cm}^{-1}$ , it is curious that “the contribution of vibrational bands in this region to the RE is... usually minor, i.e.  $<1\%$ ”. While it is true there are often fewer fundamental below  $500\text{ cm}^{-1}$ , the ability to act as a greenhouse gas is also a function of the blackbody radiation of the earth and the blackbody curve near 295K maximizes near  $1000\text{ cm}^{-1}$ , with very appreciable intensity from 200 to  $1000\text{ cm}^{-1}$ . This sentence needs clarification or a reference to more extensive work. It may also suggest the need for more experimental measurements of IR intensities in the far-infrared.

**Author Response:** Our results would indicate that an experimental focus on far-infrared absorption bands is not presently warranted for HCFCs. This is because the band intensities in this region are usually much weaker than the C-F stretching region and our calculations show that the contribution to the RE would be small, less than 1% in nearly all cases. Stated another way: we are guiding experimental effort away from an unproductive effort, because it is not easy in the laboratory to go below  $500\text{ cm}^{-1}$  in the spectrum measurements.

**Author Action:** None

**Reviewer Comment:** p. 17 Figure 6: Good Figure-lots of information, which is well presented.

**Author Response:** Thanks

**Reviewer Comment:** p. 18 line 356: Establishing the reliability of the metrics based on the average behaviour can be quite an approximation. Perhaps include language that acknowledges this approximation.

**Author Response:** In general, we agree with this comment, but as discussed in response to other comments we do not feel that a more quantitative analysis is possible at this time for molecules lacking experimental data.

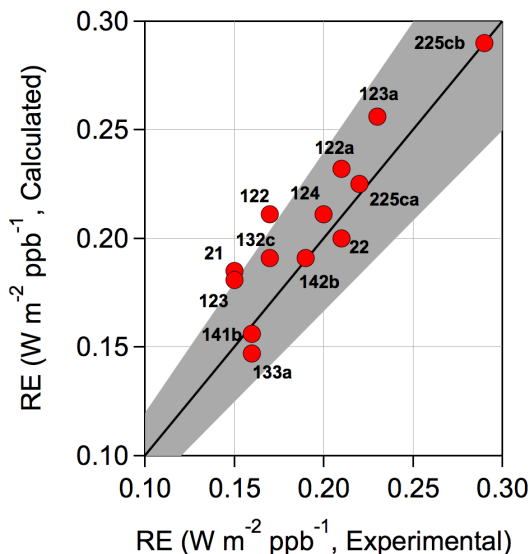
**Author Action:** None

**Reviewer Comment:** p. 18 line 370: This statement seems a bit skeptical and perhaps further explanations are needed to convince the readers that indeed the computed REs are within 10% of experimental values. Furthermore, a discussion of the estimated band strength uncertainty may strengthen the validity of the statement.

**Author Response:** The estimated RE uncertainty given in our manuscript is based on the average behavior found for the training dataset, which shows roughly a  $\sim 10\%$  uncertainty. Sources of uncertainty in the RE calculation arise from the calculated frequencies and band strengths. Combining these with the irradiance profile leads to some cancelation of error, although that is difficult to quantify other than in comparison of the calculated REs with the experimentally determined values. In our error discussion, we have used a 20% RE uncertainty.

**Author Action:** We have added a figure to the SI that shows the agreement between the lifetime corrected experimental and calculated REs for the training dataset (see below). The following text was added to Section 2.4: A comparison of the experimentally derived REs and the calculated values for the training dataset is given in Figure S3. Note that comparing ‘lifetime corrected’ REs increases the spread in the correlation between the experimental and calculated values because of differences between experimental and

calculated lifetimes and the correction factor. The figure shows the 20% correlation range, which is used in the metric uncertainty analysis. A general discussion of the uncertainties associated with band intensity calculation is now included in the introductory paragraphs to Section 2.3.



**Figure S3:** Comparison of experimental and calculated “lifetime corrected” radiative efficiencies (REs) for the training dataset HCFCs. The solid line is the 1:1 correlation. The gray shaded region represents  $\pm 20\%$  around the 1:1 line. Note that the spread in REs without applying the “lifetime correction” is  $\sim \pm 10\%$ .

**Reviewer Comment:** p. 18 line 375-377: Similar to the comment above, this sentence needs to be justified and it may be beneficial to have a discussion regarding the accuracy of the Gaussian results somewhere in the manuscript.

**Author Response:** We are basing our estimated uncertainties on the level of agreement with the training dataset values, which is pretty good. A comprehensive discussion of the uncertainties associated with the Gaussian calculations, more specifically, the calculations using DFT methods, is beyond the scope of this work. We have addressed this point in the revised Section 2.3 and in an earlier response regarding estimated uncertainties.

**Author Action:** None

**Reviewer Comment:** p. 18 line 378: This is not true for the spectral region 700-1300  $\text{cm}^{-1}$ .

**Author Response:** This comment regards the discussion that a change in the vibrational band centers primarily impacts the RE through the overlap with the irradiance profile. What is given in the manuscript is correct. This and the next comment are related.

**Author Action:** None

**Reviewer Comment:** p. 19 line 382: It might be valuable to emphasize or to provide additional details regarding this phenomenon.

**Author Response:** This comment refers to the possibility that the metric uncertainty might be greater if an absorbing molecule’s infrared spectrum was near the  $\text{CO}_2$  or  $\text{H}_2\text{O}$

absorption features. This issue has been mentioned in the text, but is difficult to quantify in general terms. This is a situation where laboratory studies would be needed.

**Author Action:** None

**Reviewer Comment:** p. 19 line 402-403: This sentence may benefit from some clarification. When compared to results from Betowski et al., it appears that the present results are 29% greater, which would suggest that the accuracy of the present results is at least  $\geq 29\%$ . There are many uncertainties presented in this section. It might be helpful to the readers to have a table displaying all of the uncertainty values in the SI.

**Author Response:** The reviewer has misinterpreted the results that are already included in the text. We actually agree very well with the raw HCFC results from the Betowski et al. study. The 29% difference is due to the band strength scaling factor developed in the Betowski et al. study that was based on an analysis of many classes of compounds. We argue that their band strength analysis yielded biased results for the HCFC class and that the 29% scaling factor should not be applied. This difference is, therefore, not representative of the uncertainty in the analysis.

**Author Action:** None

**Reviewer Comment:** p. 19 line 409-411: Might be beneficial to include how the present results compare to other experiments. Including more studies may strengthen this section of the manuscript.

**Author Response:** We do not agree with this comment. Basically, what the reviewer is suggesting is what was done in the Betowski et al. work. We already compare with all HCFC experiments, so other experiments would mean other classes of compound. We argue that this approach that was used in the Betowski et al. work led to a bias in the HCFC results. So, the approach used in the present work that limits the training dataset to HCFCs is most appropriate.

**Author Action:** None

**Reviewer Comment:** p. 20 line 425: The term “policy-relevant” is used in this sentence, yet the present results are off by a factor of 12. Again, adding language that acknowledges the limitations of such calculated data and the need for additional experimental data is likely justified.

**Author Response:** The present results are not off by a factor of 12. We believe that this comment is based on the misinterpretation of the range of GWPs given in Table 2 as described in a previous comments and responses.

**Author Action:** None

**Reviewer Comment:** p. 22 line 457: The need for additional laboratory studies is absolutely needed, and we suggest this point be emphasized throughout the paper.

**Author Response:** We agree that experimental studies are preferred over theoretically calculated or empirically derived values. We have emphasized this point in the most visible Introduction and Conclusion sections.

**Author Action:** None