Response to Reviewers

We thank the Editor and the two reviewers for their careful evaluation of our manuscript and for the constructive comments provided. The reviewers' insights have been extremely helpful in improving the clarity, rigor, and overall quality of our work. In response, we have revised the manuscript thoroughly to address all comments point by point. All changes made are highlighted in red in the revised version for ease of reference.

In particular, we have corrected the notation of the hydroxyl radical throughout the manuscript to conform to international conventions. We also clarified the experimental temperature conditions for the absorption cross-section measurements, explicitly stating these conditions in both the text and figure captions. Furthermore, the discussion regarding the discrepancy between experimentally derived absorption cross-sections and HITRAN line-by-line results has been substantially revised. Following the reviewers' recommendations, we performed a band-resolved analysis of the COF_2 absorption spectrum. This analysis demonstrates that the discrepancy is band-specific rather than indicative of a general limitation of line-by-line datasets. Finally, we have updated the manuscript to include the latest reference for the HITRAN database (HITRAN2024).

We also identified and corrected an error in the reference value of $AGWP_{CO2}(100)$ cited in the manuscript. The value previously stated as 8.064×10^{-14} W m^{-2} yr kg^{-1} (Smith et al., 2021) has been corrected to 8.95×10^{-14} W m^{-2} yr kg^{-1} (Smith et al., 2021) (main text, Lines 170–171). In addition, we improved the Data availability section to make the dataset easier for readers to access. The Zenodo statement has been updated and the DOI has been revised to reflect the newly issued record: https://doi.org/10.5281/zenodo.18015708 (main text, Line 411).

We believe that these revisions have significantly strengthened the manuscript and addressed all concerns raised by the reviewers.

Reviewer 1, Comment #1
Comment: An overall comment is related to OH radical that needs amended throughout the manuscript text, following an international adopted notation, i.e. •OH.
Response: We thank the reviewer for this important comment. Throughout the manuscript, the notation "OH radical" has been consistently revised to the internationally adopted radical notation "hydroxyl radicals (•OH)" at its first occurrence and "•OH" thereafter. All corresponding changes have been implemented in the main text and are highlighted in red for clarity.
Manuscript Changes: Lines 197, 202, 204, 207, 211–212, 342, and 345.

Reviewer 1, Comment #2

Comment:

Figure 2, is this temperature adjusted?

Throughout the manuscript authors make note of absorption cross-sections, yet it is not clear if their experimental data was recorded at room temperature or if they have taken a temperature profile investigations. The Earth's atmosphere temperature profile from sea-level up to the limit of stratopause varies from room temperature down to 213 K. A real assessment of F2CO molecule as to its radiative forcing is relevant as a function of temperature unless there is no significant change in such range. This needs to be properly addressed.

Response:

We agree with the reviewer that temperature effects are an important consideration in the interpretation of absorption cross-sections and radiative forcing. Although absorption cross-sections can exhibit temperature dependence, it is well established that integrated absorption cross-sections show little dependence on temperature. The apparent temperature dependence of absorption cross-sections mainly originates from changes in rotational state populations, which lead to broader spectral bands with reduced peak intensities at higher temperatures.

In the present study, the absorption cross-section (ACS) of COF_2 was measured under controlled laboratory conditions at a single, constant temperature of 296.77 K, and no temperature-dependent ACS measurements were performed. The experimental conditions are described in detail in the Methodology section (Section 2.1, FTIR Systems for Absorption Cross-Section and Atmospheric Lifetime Measurements).

From the reader's perspective, we acknowledge that the measurement temperature condition was not sufficiently explicit in the original presentation of Figure 2. To address this, we have revised both the caption of Figure 2 and the main text to explicitly state that the ACS spectrum was obtained under a single-temperature laboratory condition (296.77 K). This clarification ensures transparent interpretation of the ACS data and clearly defines the experimental scope and limitations of the present study. All changes are highlighted in red in the revised manuscript.

Manuscript Changes:

Line 222; **Figure 2** caption (lines 3–4).

Reviewer 2, Comment #1

Comment:

The authors have uncovered a significant issue with the HITRAN line-by-line data (it underestimates absorption by around 20%). I believe that they should spend a bit more time examining the cause of this discrepancy, as their explanation for the discrepancy is unconvincing. I do not think there is evidence of "an inherent limitation of line-by-line datasets" as the authors claim at lines 237-238. I think the problem may be specific to COF2. If I am wrong, then the authors need to present evidence of that "inherent limitation" beyond COF2, which would require much work, or reference to papers that support their statement.

I suggest that they break down the integrated absorption cross-section into the 4 distinct bands of COF2 included on the HITRAN database and compare it with their new measurements. Looking at the summary plots

on the HITRAN website, I suspect that most of the discrepancy originates from the band centred on 960 cm-1. In that band (unlike the other bands), lines weaker 2E-22 line strength units are ignored, and so the spectrum is truncated in both line strength and wavenumber space.

If the authors could demonstrate that this is the principal cause of the discrepancy, it could inspire future work to improve the line-by-line analysis of that band. If it is a more general problem with all sub-bands, then that would also be useful to demonstrate.

Response:

We would like to thank the reviewer for their insightful and constructive comments regarding the band-specific analysis of COF_2. We agree that our original attribution of the discrepancy to an "inherent limitation of line-by-line datasets" was overly broad. Following the reviewer's suggestion, we have performed a detailed breakdown of the integrated absorption cross-sections into the four distinct infrared bands represented in the HITRAN database. This analysis reveals that the discrepancy is indeed band-specific rather than a systemic failure of the line-by-line methodology.

The results of this decomposition show that for Band 1 (695–865 cm^{-1}), our measurements and HITRAN are in excellent agreement with a 0% relative difference, while the difference between PNNL and HITRAN is only 9%. This high level of agreement in Band 1 demonstrates that the line-by-line approach is highly effective when the underlying parameters are accurate and complete. In Band 2 (925–995 cm^{-1}), although we observe a difference of 18% relative to our work, the discrepancy compared to PNNL is more moderate at 10%, suggesting that while some issues exist, they are not the sole drivers of the overall underestimation.

The most significant and consistent discrepancies were identified in Band 3 (1175–1305 cm^{-1}) and Band 4 (1845–2005 cm^{-1}). In these regions, HITRAN underestimates the integrated absorption by approximately 17–19% compared to both our experimental results and the PNNL database. Given that the absolute absorption magnitude of these two bands is an order of magnitude larger than that of Bands 1 and 2, their underestimation dominates the total error. These findings strongly support the reviewer's hypothesis that the problem is specific to the COF_2 parameters in HITRAN, likely due to the omission of weak transitions or limited spectral coverage in those specific bands.

In light of this evidence, we have revised the manuscript to clarify that the discrepancy arises from the representation of specific COF_2 absorption bands in the HITRAN database rather than a general limitation of line-by-line models. We have replaced the problematic statement at lines 237–238 with a more nuanced discussion centered on the incompleteness of the line parameters in the 1175–1305 cm^{-1} and 1845–2005 cm^{-1} regions. We believe this revision accurately reflects the data and addresses the reviewer's concerns.

Manuscript Changes:

Lines 235–251 and Supporting Information (**Fig. S2** and **Table S1**).

Reviewer 2, Comment #2

Comment:

The present version of the manuscript does not provide a reference for HITRAN and this MUST be corrected. A slight complication is that a new version of HITRAN (HITRAN 2024) has only just (yesterday!) been released but I believe the COF2 data is unchanged from HITRAN 2020. The HITRAN website recommends this reference.

I. E. Gordon, L. S. Rothman, R. J. Hargreaves, F. M. Gomez, T. Bertin, C. Hill, et al., "The HITRAN2024 molecular spectroscopic database", J. Quant. Spectrosc. Radiat. Transfer, in press (2026). [doi:10.1016/j.jqsrt.2026.109807]

Response:

We agree with the reviewer's comment. The manuscript has been revised to include the recommended reference for the HITRAN database. In accordance with the HITRAN website recommendation, we have cited Gordon et al., "The HITRAN2024 molecular spectroscopic database," J. Quant. Spectrosc. Radiat. Transfer (in press). Although the COF_2 spectroscopic parameters used in this study are unchanged from HITRAN 2020, the reference has been updated to reflect the most recent HITRAN release. The corresponding citation has been added to the revised manuscript and is highlighted in red.

Manuscript Changes:

Lines 84, 236, and 460-461.