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Low-frequency contributions in the radiative efficiencies of HFC-236fa, HFC-245fa and HFC-43-10mee over the 225–298 K temperature range☆

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ABSTRACT

Hydrofluorocarbons (HFCs) are used as substitutes for ozone-depleting substances regulated under the Montreal Protocol. While having zero ozone depletion potential, HFCs strongly absorb infrared (IR) radiation, making them potent greenhouse gases. Vibrational modes associated with C–F stretching absorb strongly within the atmospheric window (750–1250 cm^{-1}), contributing substantially to radiative forcing. The low-frequency region (< 500 cm^{-1}), which accounts for approximately 16% of the Earth's thermal emission, has however remained largely unexplored mainly due to instrumental challenges. Here, we present the first experimental measurements of IR absorption cross-sections in the 150–500 cm^{-1} range for HFC-236fa, HFC-245fa, and HFC-43-10mee — three industrially relevant compounds with high global warming potentials (GWPs). The spectra were recorded at the Rutherford Appleton Laboratory using a high-resolution Fourier-transform infrared (FTIR) spectrometer in the temperature range between 225 and 298 K at resolution of 0.25 cm^{-1}. In addition, IR cross section spectra were simulated through quantum chemical (QC) calculations including a non-empirical treatment of anharmonic effects.

From the experimental results, we derived effective radiative efficiencies (EREs) in the low-frequency region of 0.001, 0.005, and 0.003 W m^{-2} ppb^{-1} for HFC-236fa, HFC-245fa, and HFC-43-10mee, respectively, and revised global warming potentials over 20-, 100-, and 500-year time horizons. Comparison with values reported in the WMO Ozone Assessment Report 2022 reveals minor differences for HFC-245fa and HFC-43-10mee, whereas their value for HFC-236fa shows a significant overestimation, corresponding to a discrepancy of approximately 360 units in the 100-year GWP. Theoretical predictions reproduced experimental band strengths with an overall average deviation of 4%, confirming the reliability of the computational approach even in the low-frequency region. This indicates that the QC technique is likely to provide reliable estimates for RE and GWP for similar compounds where measurements are not available.

These findings highlight that small variations in the treatment of low-frequency absorptions can propagate into substantial contributions in climate metrics, particularly for long-lived compounds. Overall, this study provides a consistent experimental–theoretical framework for quantifying the radiative forcing of HFCs and similar compounds and reduces current uncertainties in the estimation of their climate-relevant parameters.