

Atmosphere-based “Top-down” Emission Estimates of HFC-134a and HCFC-22 for the United States (U.S.) over Multiple Years

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HFC-134a and HCFC-22 are potent greenhouse gases; HCFC-22 is also an ozone-depleting gas. Uncertainty in their national emissions reported from inventory-based “bottom-up” estimates is not well determined due to incomplete understanding of the emissive processes and the size of their existing stocks and reserves. This study uses our atmospheric measurements from multiple surface and aircraft sites across the U.S. continent and in the Pacific Basin from 2008 - 2012, along with a newly-developed regional inverse model, to constrain U.S. national emissions of HFC-134a and HCFC-22 over this period. A suite of synthetic-data experiments were conducted to optimize the model design, assess model-data mismatch errors and a prior flux error covariance matrix given by the maximum likelihood estimation method, and test the sensitivity of derived national fluxes to a range of priors that have different emission magnitudes and distributions than the “synthetic-true” emission. Limitations in synthetic-data experiments led us to explore the influence of background and transport uncertainties on derived national emissions in real-data inversions. With multiple backgrounds, multiple priors, and multiple transport models driven by three different meteorological fields, derived national emissions of HFC-134a and HCFC-22 using actual observations agree within 20% on an annual basis. Results also consistently show seasonal variations with summer emissions higher than winter by 20 – 50 % (Figure 1). The HFC-134a emission estimated for the entire U.S. from our work is comparable to that reported by US Environmental Protection Agency (EPA) whereas HCFC-22 emission derived from our work is lower and shows a more rapid decline than that reported by U.S. EPA.

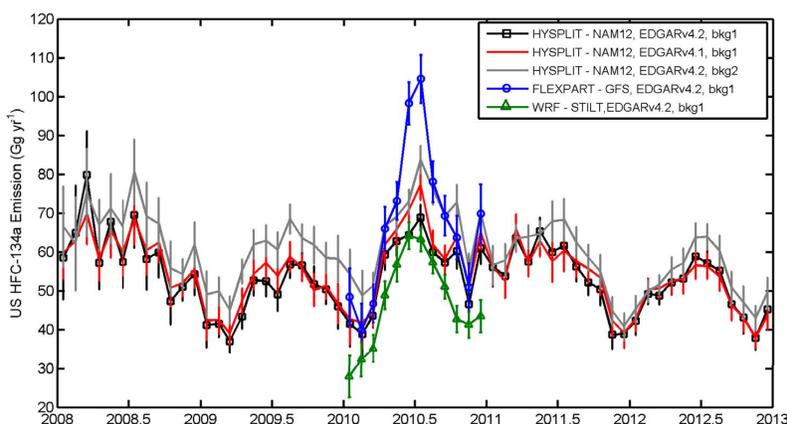


Figure 1. U.S. national emissions of HFC-134a estimated from this study using multiple transport models and meteorological data (HYSPLIT-NAM12, FLEXPART-GFS, and WRF-STILT), multiple priors (EDGARv4.2 and EDGARv4.1) and multiple backgrounds (bkg 1: the lowest 10th percentile at the surface sites and average mixing ratios between 3000 – 7000 masl at aircraft sites; bkg 2: average background mixing ratios from 500 particles that transported backward in time and intersected with an empirical “curtain” which represents North American HFC-134a background concentrations as a function of time, latitude and altitude).