^{14}C-based emission estimates for halocarbons and other gases across the U.S.

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An extension of the analysis in:

J.B. Miller, S.J. Lehman, S.A. Montzka, C. Sweeney, B.R. Miller, A. Karion, C. Wolak, E.J. Dlugokencky, J. Southon, J.C. Turnbull, P.P Tans, *JGR-A, 117*, D08302, doi:10.1029/2011JD017048, 2012.

Goal:

Derive atmosphere-based estimates of national emission magnitudes for chemicals influencing climate, ozone, & air quality.

Approach:

use multiple techniques…

For Today:

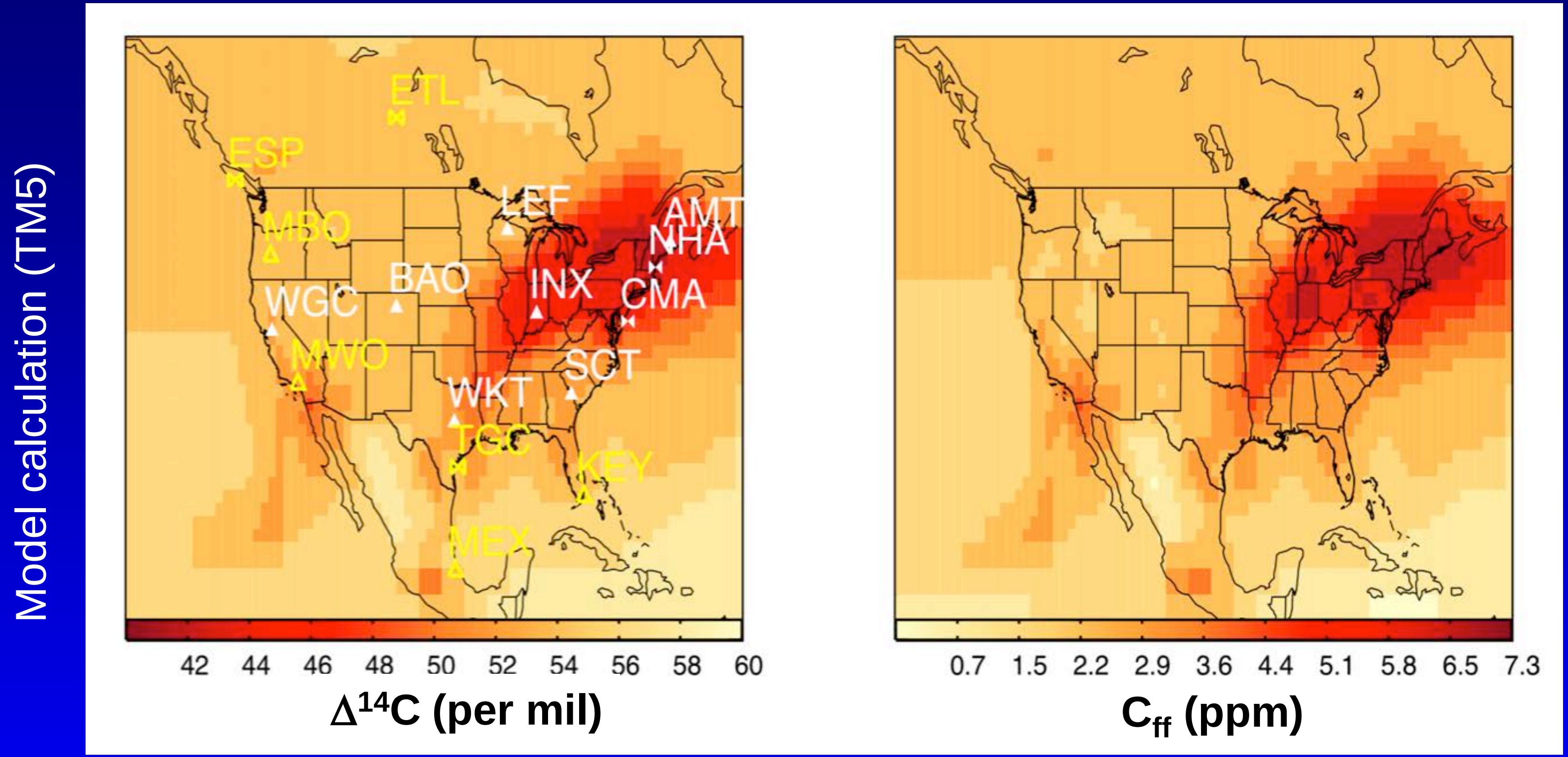
$$\boxed{\textbf{Emissions}(X_1) = [\Delta X_1 / \Delta X_2] \times \textbf{Emissions}(X_2)}$$

where:

* ΔX_2 = fossil-fuel CO_2 (C_{ff}) derived from measurements of $^{14}CO_2$

* Emissions(X_2) = fossil fuel emissions from the VULCAN inventory (Gurney *et al*., 2009)

$\Delta^{14}CO_2$ is a useful proxy for C_{ff}

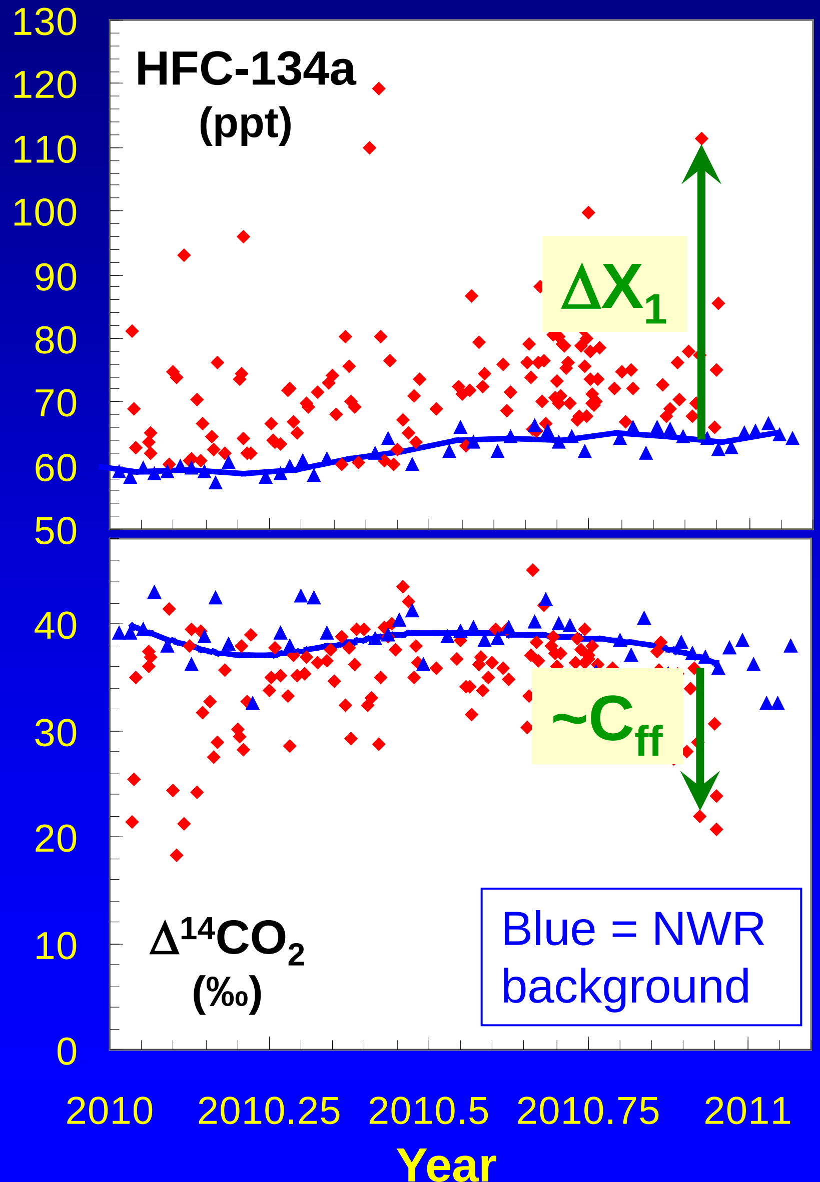


* In a model, distribution of C_{ff} dominates the $\Delta^{14}CO_2$ signal over NH land area
  → nuclear power and respiration influences are small
  → figures here are scaled according to mass balance relation of -2.7‰/ppm

* In practice, measurement precision allows determination of C_{ff} within ±1 ppm

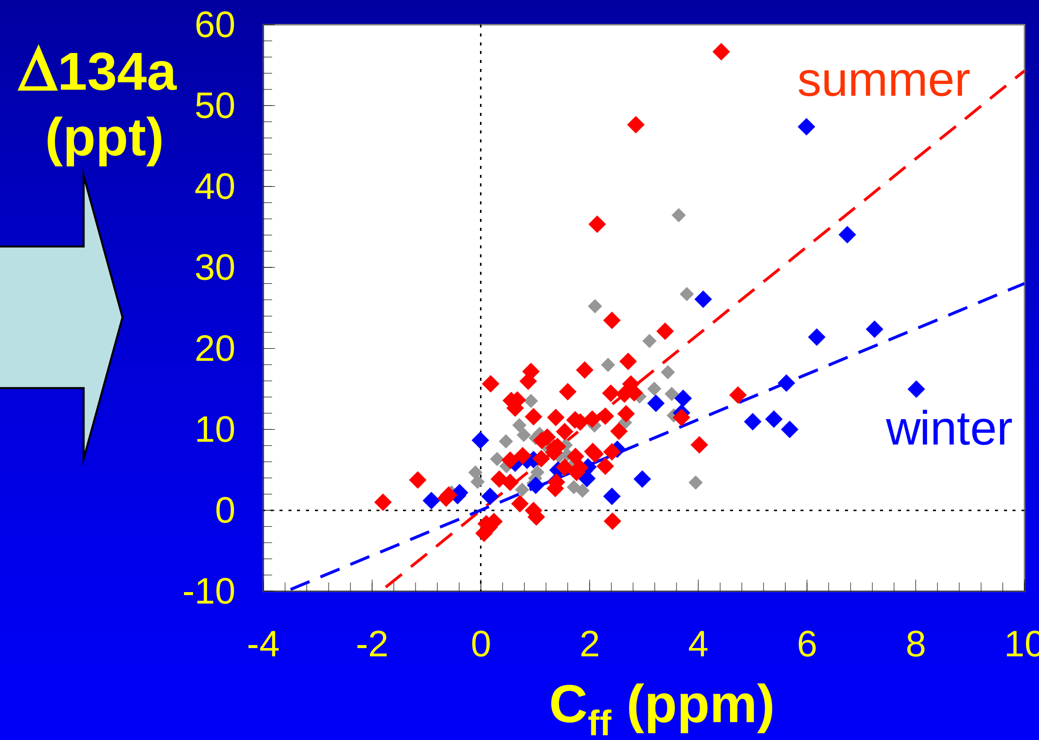
Deriving ΔX_1 and C_{ff} from air sample measurements

Observations at WKT (Texas) relative to background site

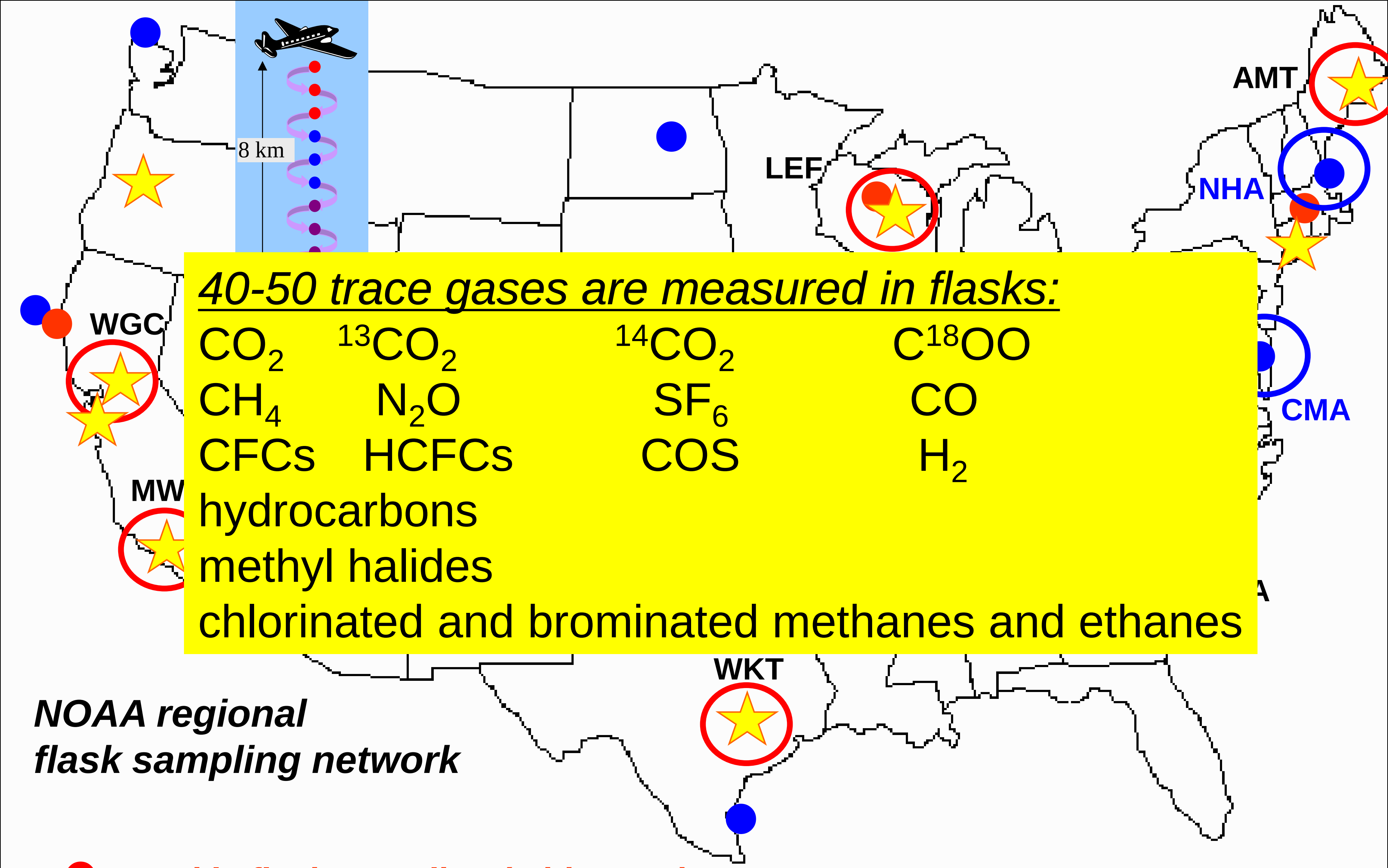


Apparent Emission Ratio
(as median of point-by-point enhancements)

HFC-134a *vs.* C_{ff} at WKT (2010)



$C_{ff} \approx (\Delta^{14}C_{obs} - \Delta^{14}C_{bkgd}) / -2.7$ ‰ / ppm +…



__40-50 trace gases are measured in flasks:__

CO_2 $^{13}CO_2$ $^{14}CO_2$ $C^{18}OO$

CH_4 N_2O SF_6 CO

CFCs HCFCs COS H_2

hydrocarbons

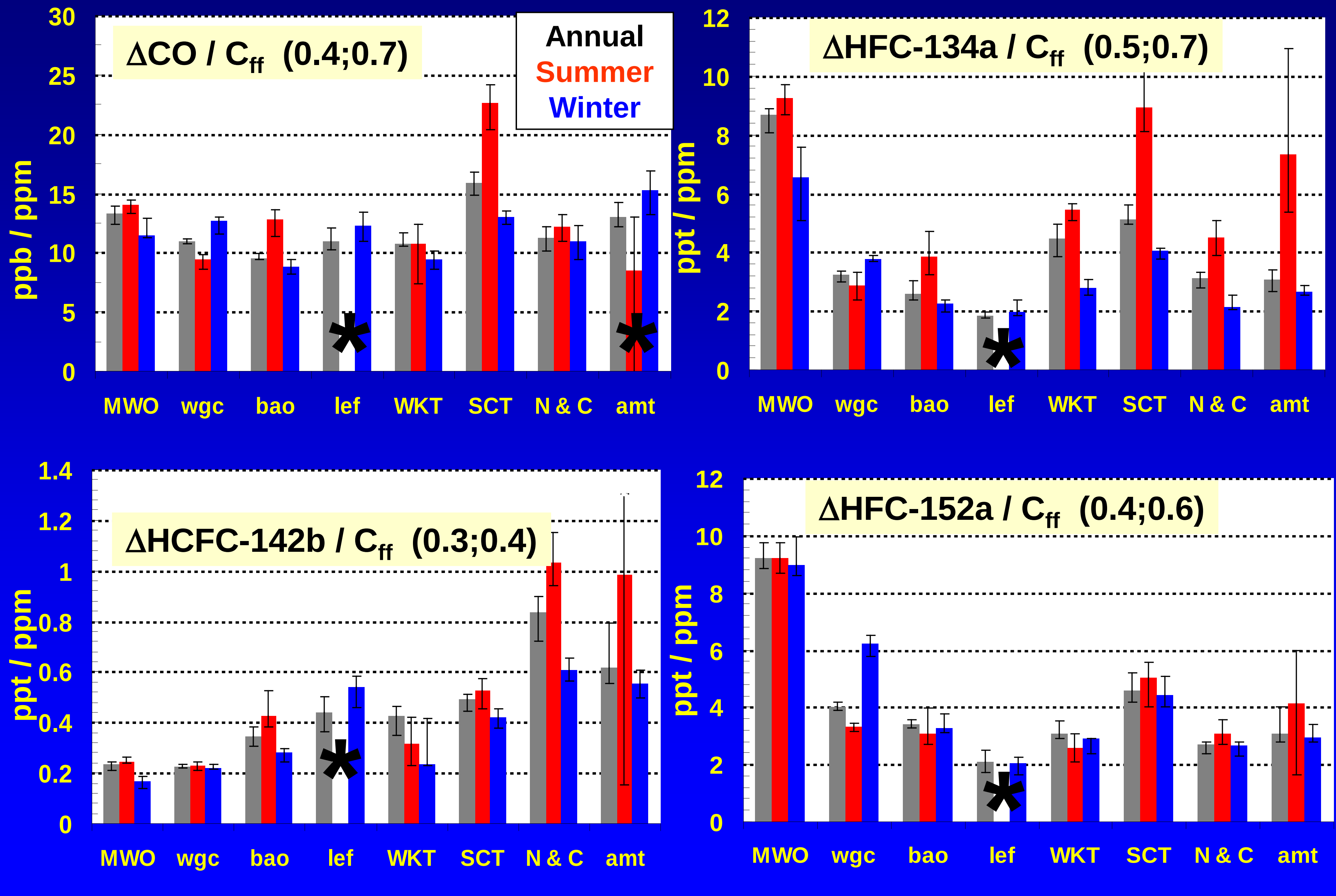
methyl halides

chlorinated and brominated methanes and ethanes

NOAA regional flask sampling network

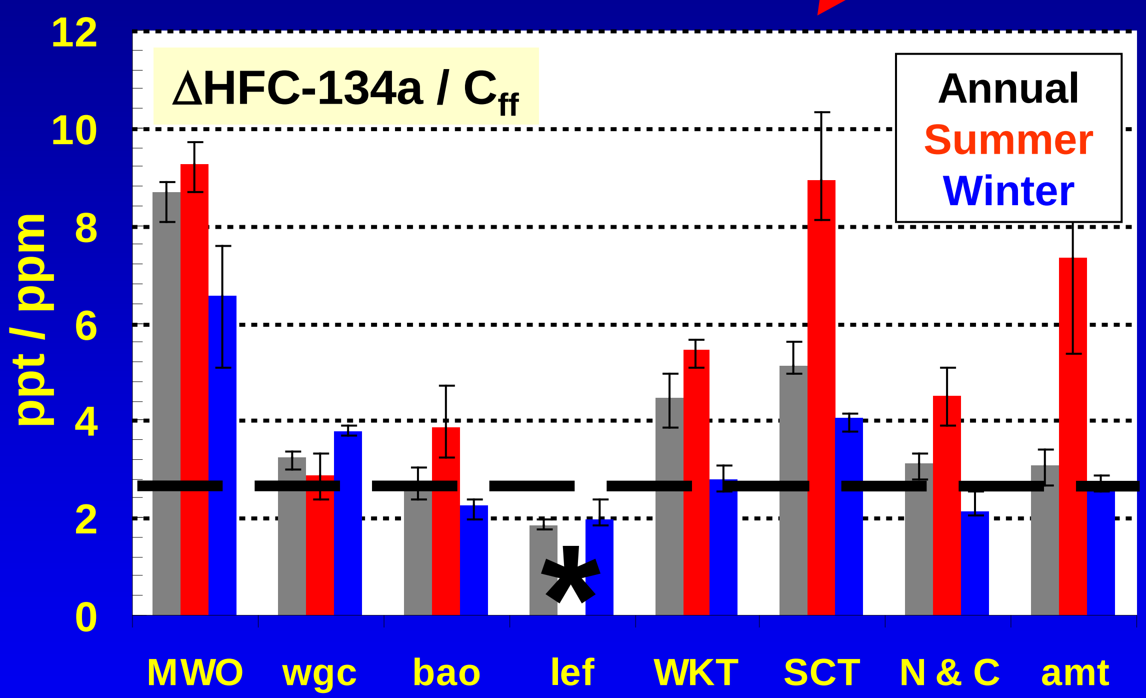
Weekly flask sampling (mid 1990s)

Apparent Emission Ratios ($\Delta X_1 / C_{ff}$) :



Deriving absolute emission rates:

$$\text{Emissions}(X_1) = [\Delta X_1 / C_{ff}] \times \text{Emissions}(C_{ff})$$



$\times$

1.6 PgC yr^{-1} ??

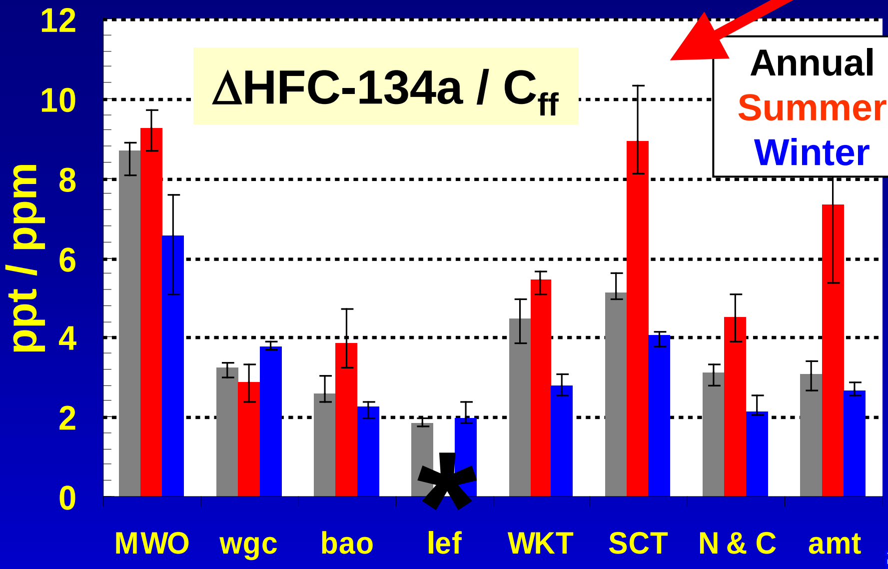
Annual total US C_{ff}

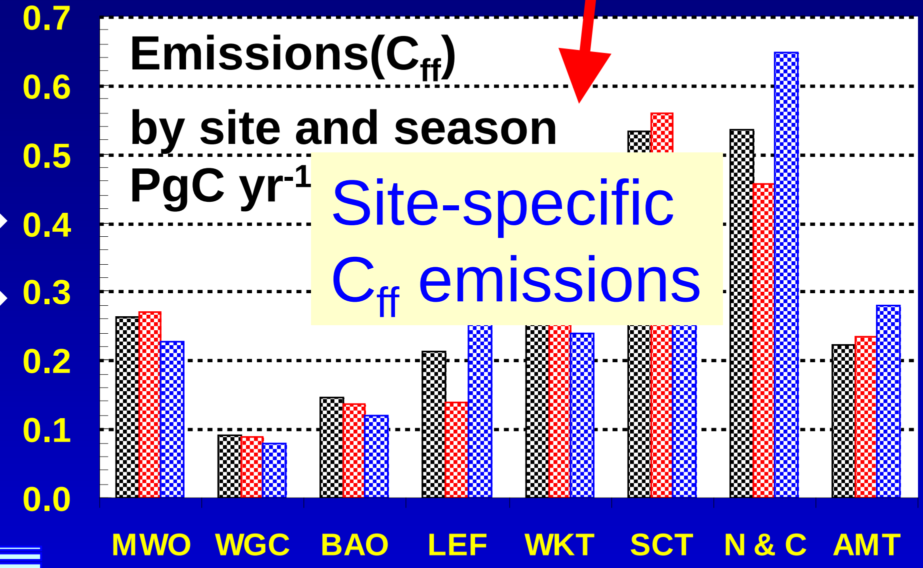
Emissions (HFC-134a) ≈ 43 Gg yr^{-1}

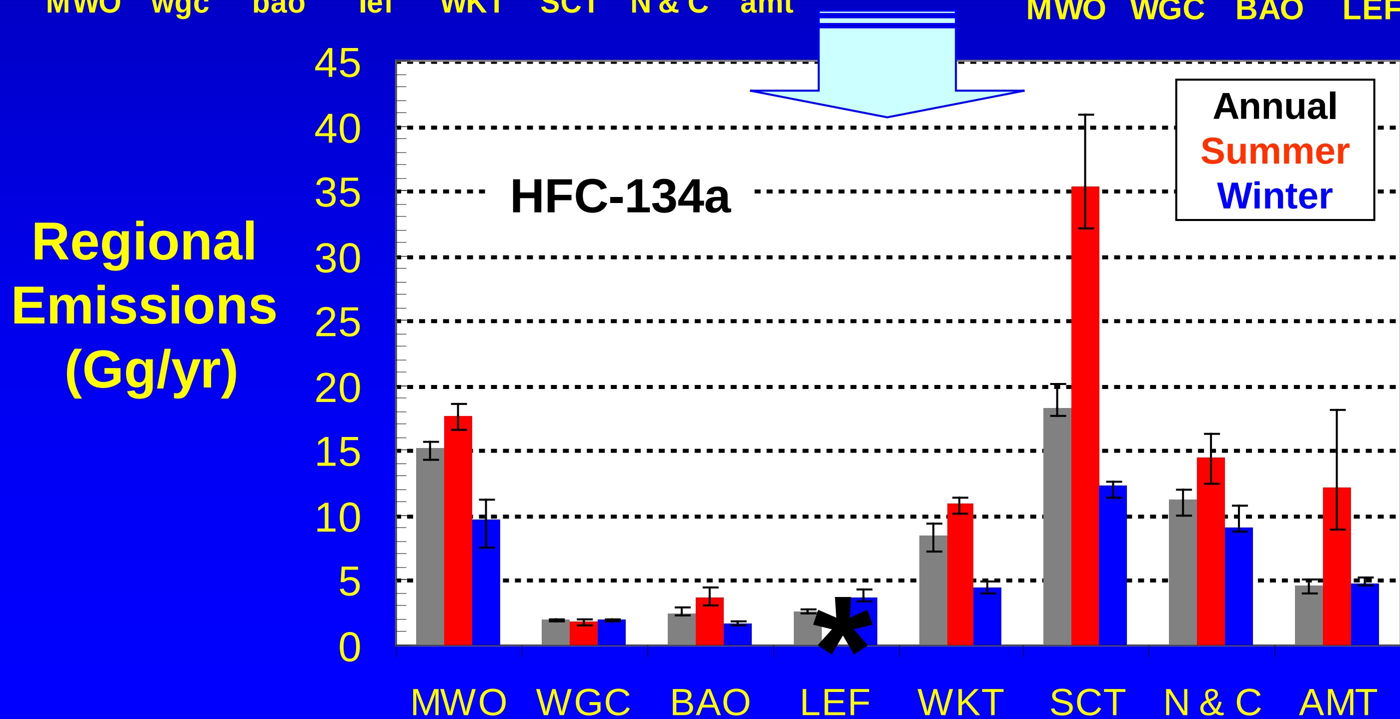
from NHA & CMA alone ≈ 46 Gg yr^{-1}

(Miller *et al*., 2012)

$$\text{Emissions}(X_1) = [\Delta X_1 / C_{ff}] \times \text{Emissions}(C_{ff})$$







HFC-134a

Annual Emiss:

(annual basis)

≈58 Gg yr^{-1}

(seasonal basis)

≈65 Gg yr^{-1}

Annual national emissions:

Chemical	Miller *et al.** 2006-2009	**this work**** 2010	EPA * 2005-2009	EDGAR * 2005-2009	
CO	41 (16-73)	48	77	62	Tg yr^{-1}
HFC-134a	46 (10-86)	65	55	70	Gg yr^{-1}

* As reported in Miller *et al*. (2012) from CMA & NHA only

** Scaled to total US C_{ff} emission of 1.6 PgC yr^{-1}

For California:

		this work	CARB estimate for 2010
CO	Tg yr^{-1}	3.4	3.5
HFC-134a	Gg yr^{-1}	6.9	6.0

…from *ave $\Delta X_1/C_{ff}$ at WGC and MWO in 2010* and *Vulcan C_{ff} for CA in 2002 scaled by EIA to 2010.*

Refinements planned for the future:

1) Improving our estimates of:
   * background concentrations
   * emission ratios
   * surface sensitivity (footprints)

2) Adding observations at new sites to improve spatial coverage.

3) Performing inverse modeling analyses of _all_ measurements.
   → with verification potentially provided by ^{14}C.

4) Provide estimates of inter-annual emission changes.

Conclusions

From atmospheric measurements of a suite of chemicals affecting climate, ozone, and air quality at nine US sites during 2010:

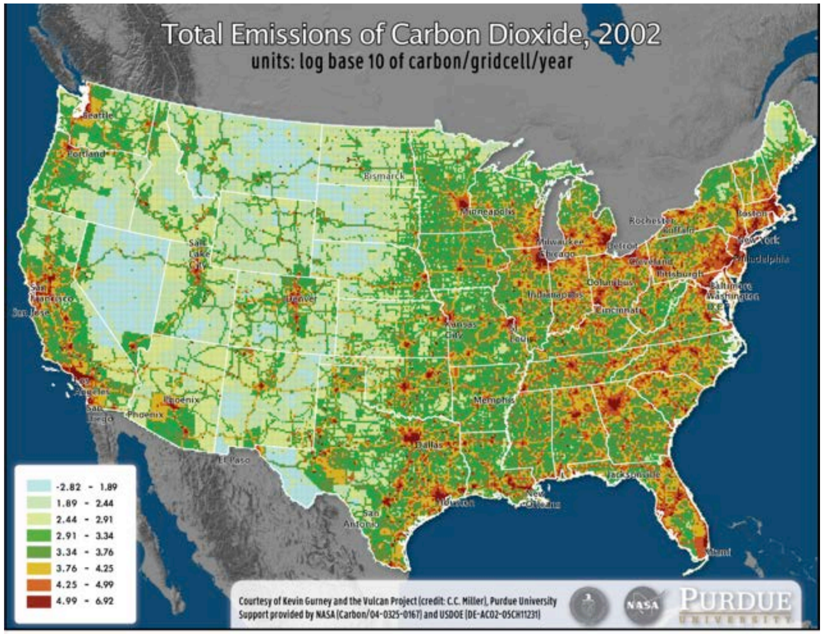
* ***State-wide and national scale emissions were derived*** (based on measured atmospheric co-variations with fossil-fuel CO_2 and the VULCAN C_{ff} inventory)

* ***Substantial variations noted across seasons and space*** (necessary to characterize for deriving accurate and representative, top-down national emission magnitudes)

* Future work will focus on maintaining the observational network; refining our approach, defining robust uncertainties, and comparing results among multiple techniques.

Deriving site-specific C_{ff} emissions

Fossil-fuel emission inventory



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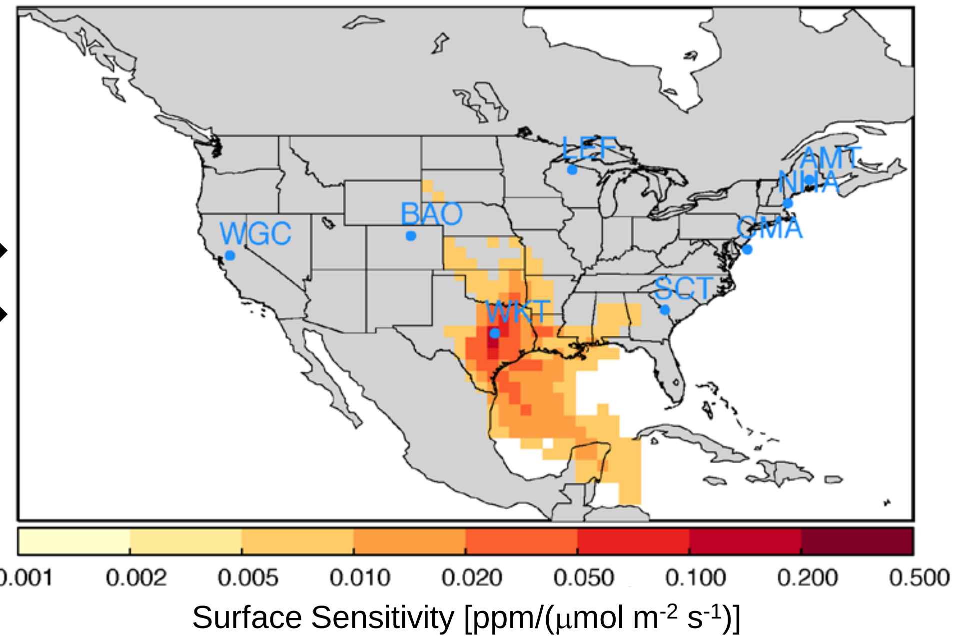
Site sensitivity to surface emissions

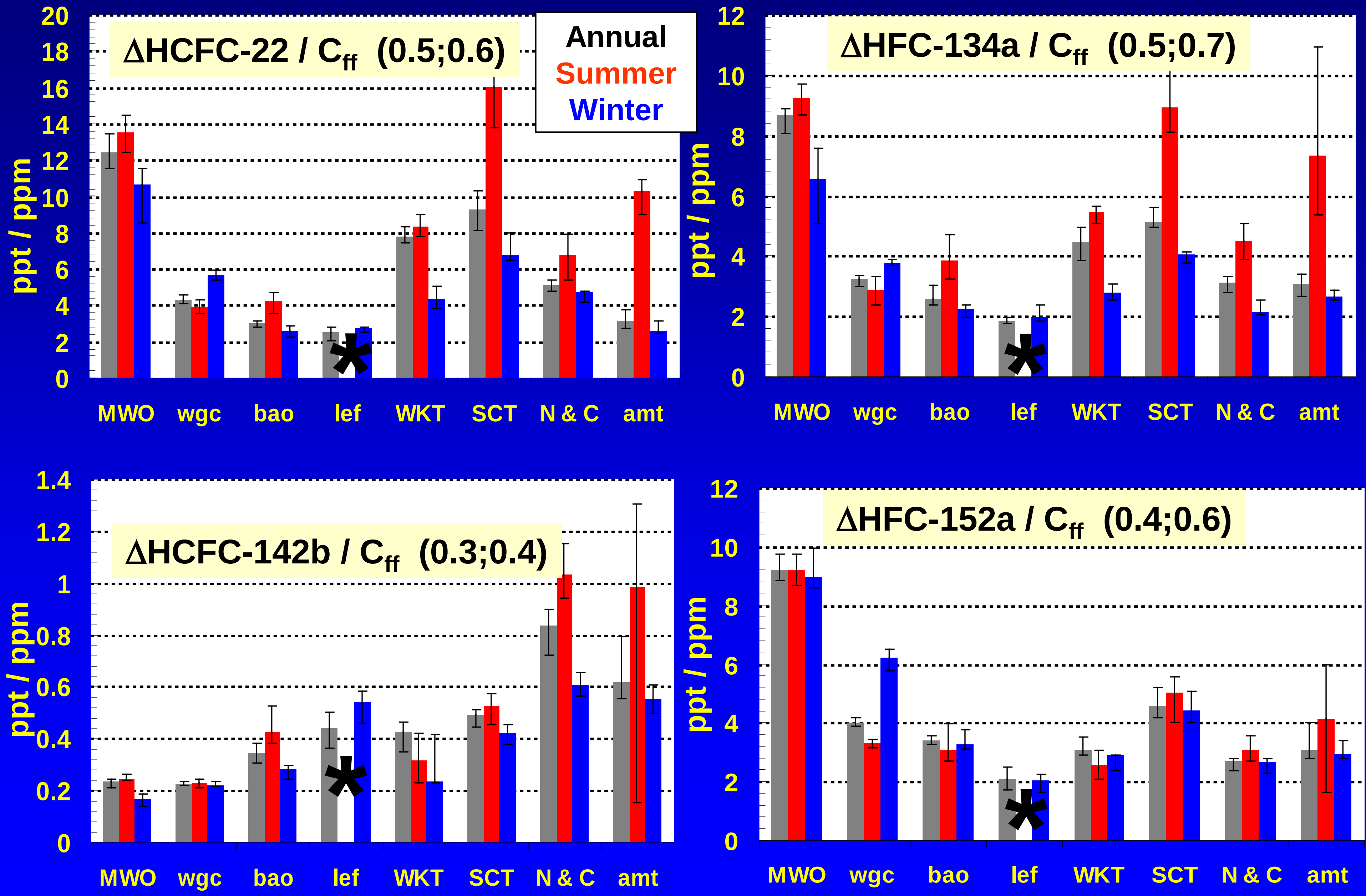


Gurney, K.R., *et al*. (2009), *Environ. Sci. Technol, 43*, doi:10.1021/es900806c; 2008 emissions used.

Footprint calculated with STILT Lagrangian trajectory model driven by WRF winds at 10 km resolution

Site-specific C_{ff} emissions can be derived by convolving the Vulcan fossil-fuel emission inventory with site- and season-specific surface sensitivity footprints.

Apparent Emission Ratios (ΔX_1 / C_{ff}) :



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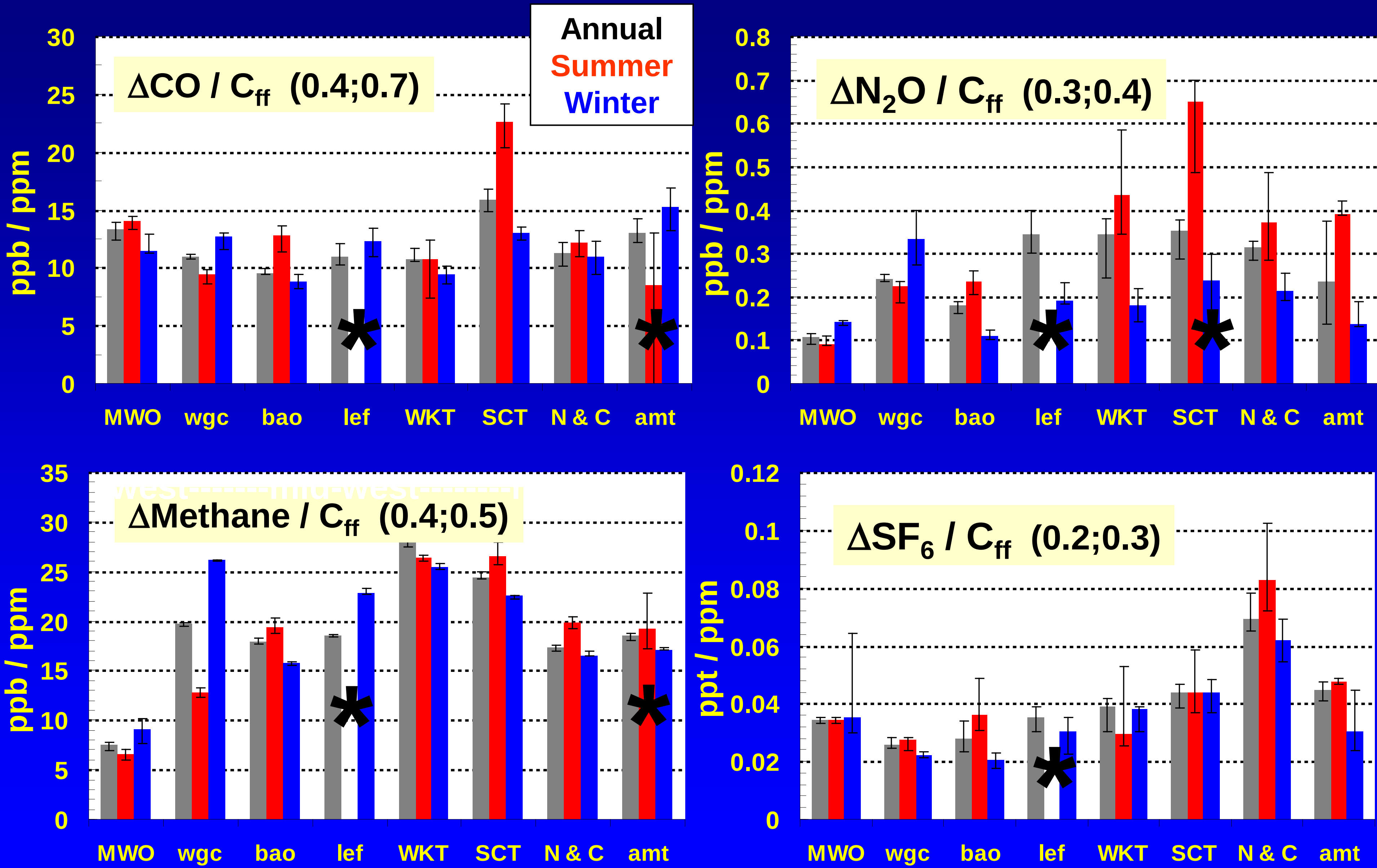

<u>Annual national emissions</u>:

Chemical	Miller *et al.**	this work**	EPA *	EDGAR *	
Years	2006-2009	2010	2005-2009	2005-2009	
CO	41 (16-73)	48	77	62	Tg yr^{-1}
SF_6	1.4 (0.7-3.0)	0.9	0.7	1.8	Gg yr^{-1}
HFC-134a	46 (10-86)	65	55	70	Gg yr^{-1}
CH_4	39 (18-69)	41	32	26	Tg yr^{-1}
N_2O	1.7 (0.7-3.6)	1.8	1.0	1.0	Tg yr^{-1}
Sites>>	CMA & NHA	nine	All US	All US	

* As reported in Miller *et al*. (2012)

** PRELIMINARY for 2010; Scaled to total US C_{ff} emission of 1.6 PgC yr^{-1}