Measurement of fossil fuel derived carbon dioxide and other anthropogenic trace gases above Sacramento, California in Spring 2009

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Outline

How can we use aircraft observations of $^{14}CO_2$-based measurements of CO_2ff?

- Emission ratios to other species
  - evaluate inventories
- Combine with continuous CO measurements to obtain continuous CO_2ff
  - Partition observed CO_2 into fossil and bio portions
- Infer flux of CO_2ff from urban region
  - Compare with inventories

Calculation of recently added fossil fuel CO_2 mixing ratio from observations of CO_2 and $\Delta^{14}CO_2$

$$C_{obs} = C_{bg} + C_{ff} + C_r$$

$$C_{obs}\Delta_{obs} = C_{bg}\Delta_{bg} + C_{ff}\Delta_{ff} + C_r\Delta_r$$

$$C_{ff} = \frac{C_{obs}(\Delta_{obs} - \Delta_{bg})}{\Delta_{ff} - \Delta_{bg}} - \frac{C_r(\Delta_r - \Delta_{bg})}{\Delta_{ff} - \Delta_{bg}}$$

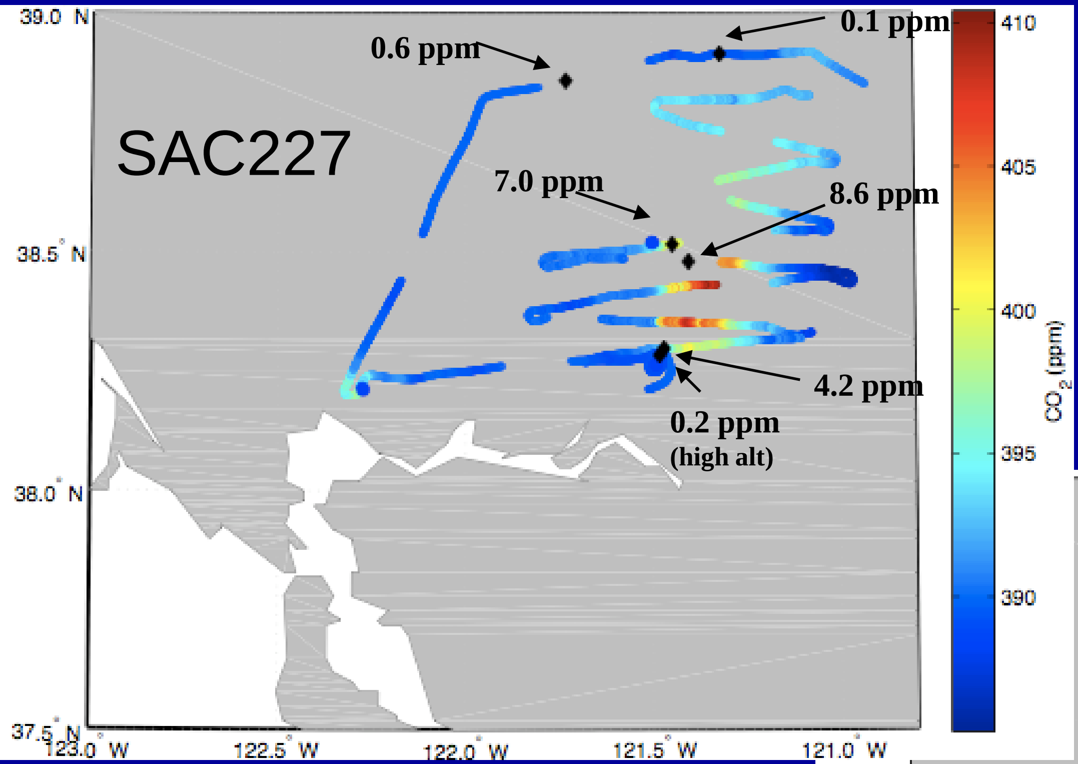
$$\Delta^{14}CO_2 = \left\{ \left(\frac{({}^{14}C/C)_{sa}}{({}^{14}C/C)_{std}} \right)_{BN[x]} - 1 \right\} \times 1000‰$$

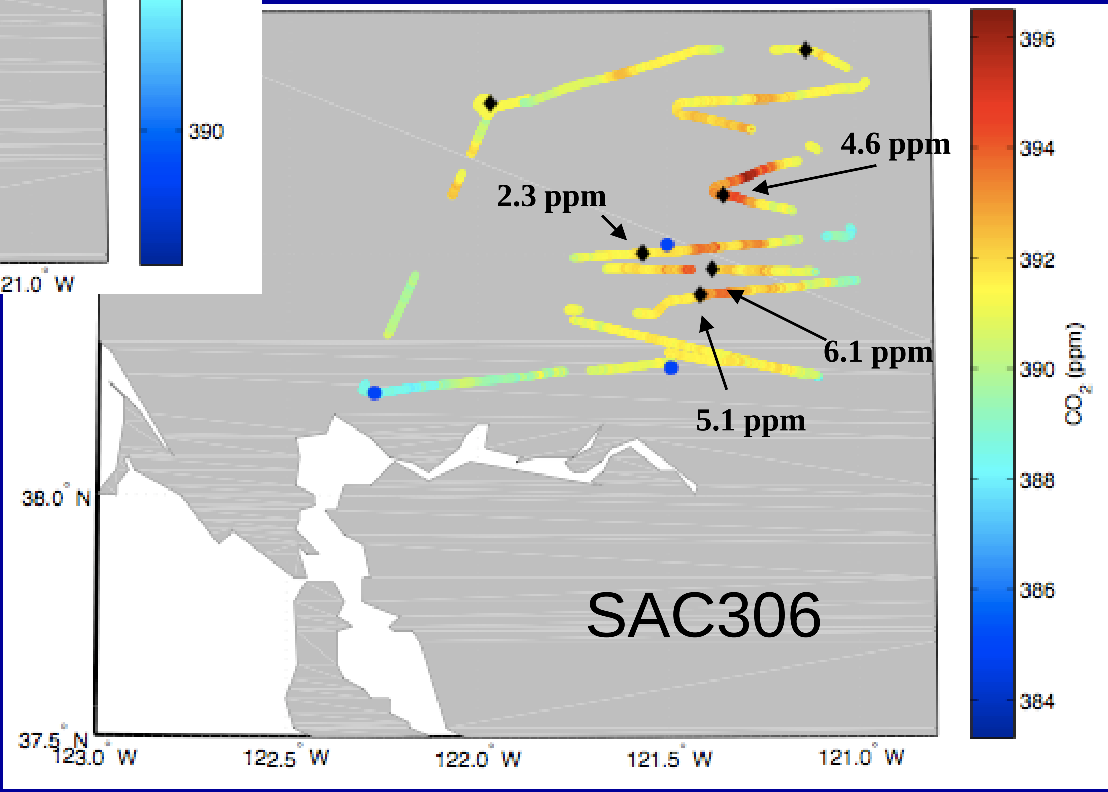
std=1.176×10^{-12} $^{14}C/C$

Use Δ_{bg}=47.01±2.5 ‰ (highest value in campaign)

Bias term = -0.2 ±0.1 ppm (winter estimate)

Two flights over Sacramento, Feb/Mar 2009



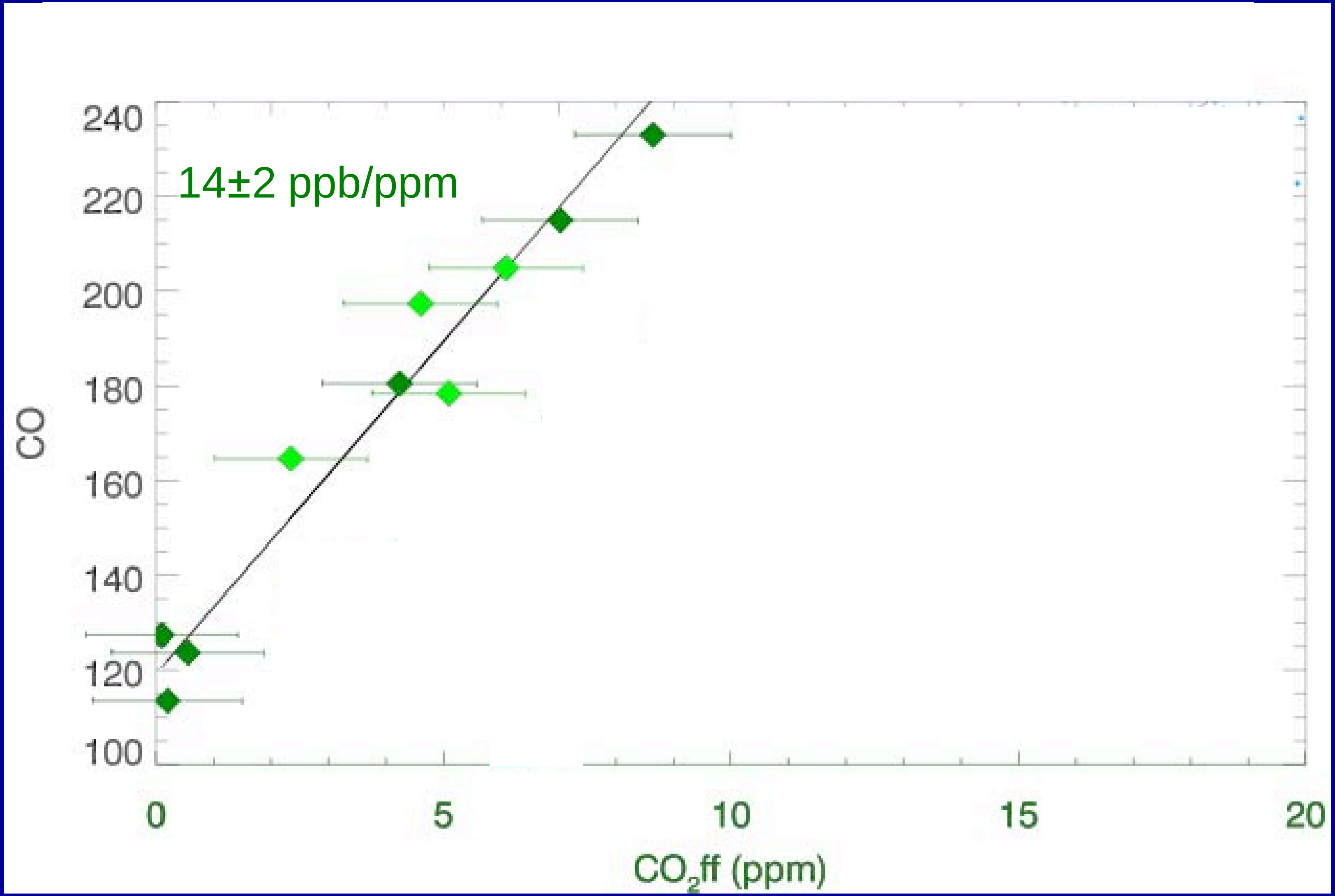


Continuous: CO_2, CH_4, CO

PFP flasks: $\Delta^{14}CO_2$, CO_2, CO, halocarbons, hydrocarbons...

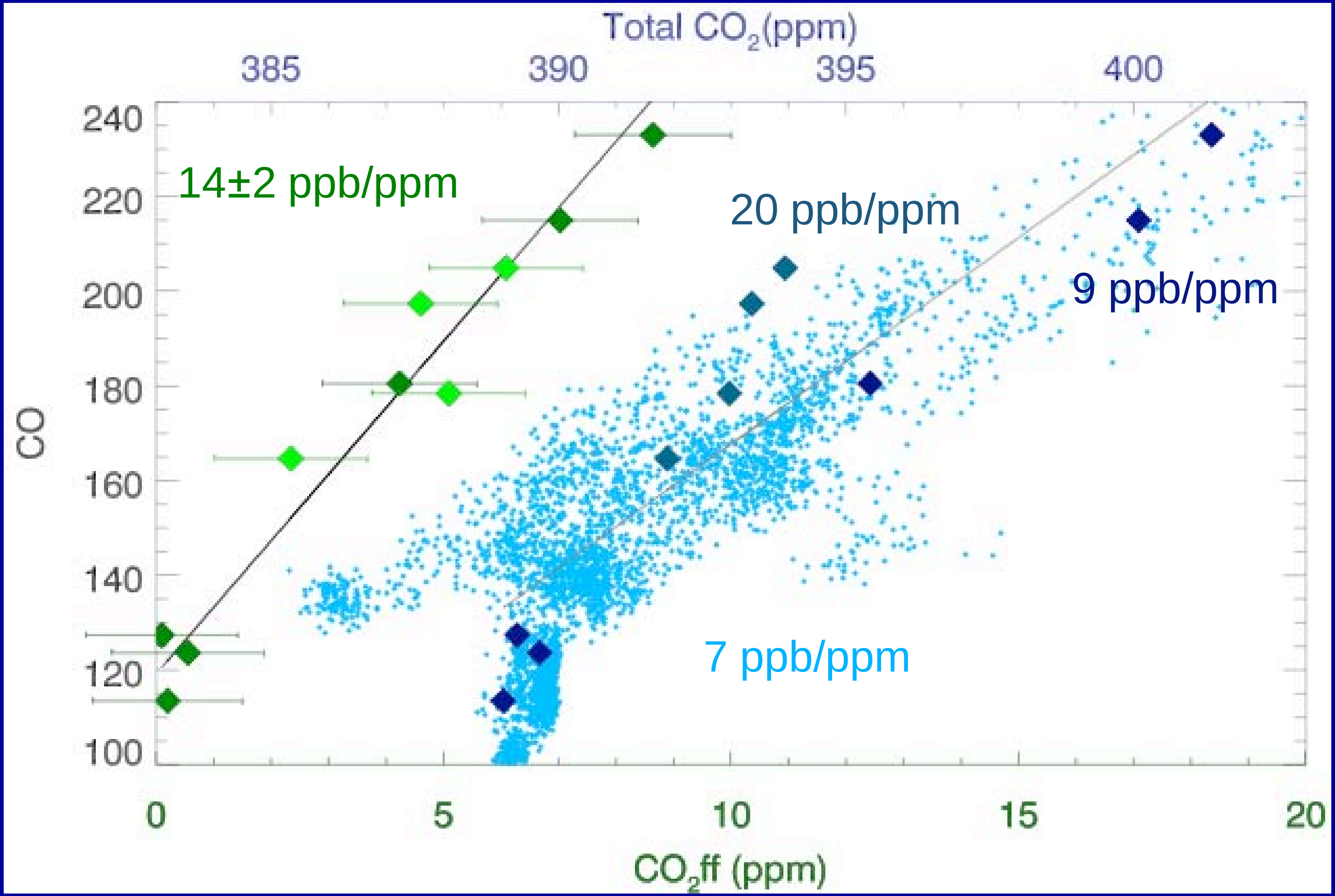
Calculate CO_2ff from $\Delta^{14}CO_2$ and CO_2 in flasks

Emission ratios: CO:CO_2ff



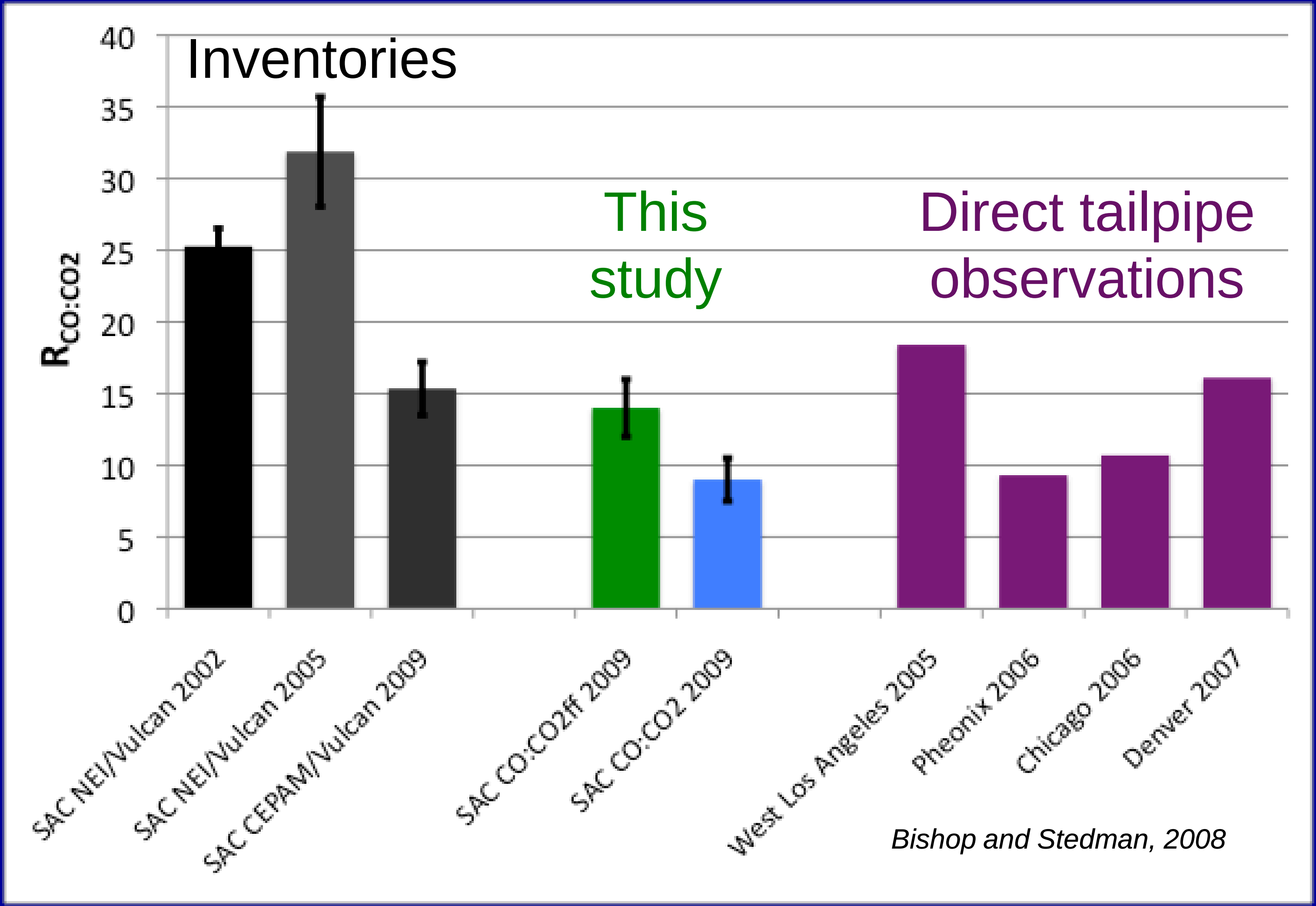
Very strong correlation between CO and CO_2ff

Emission ratios: $CO:CO_2ff$



Using CO_2 enhancement instead of CO_2ff gives a different emission ratio that varies between the two flights

Emission ratios: evaluation of inventories

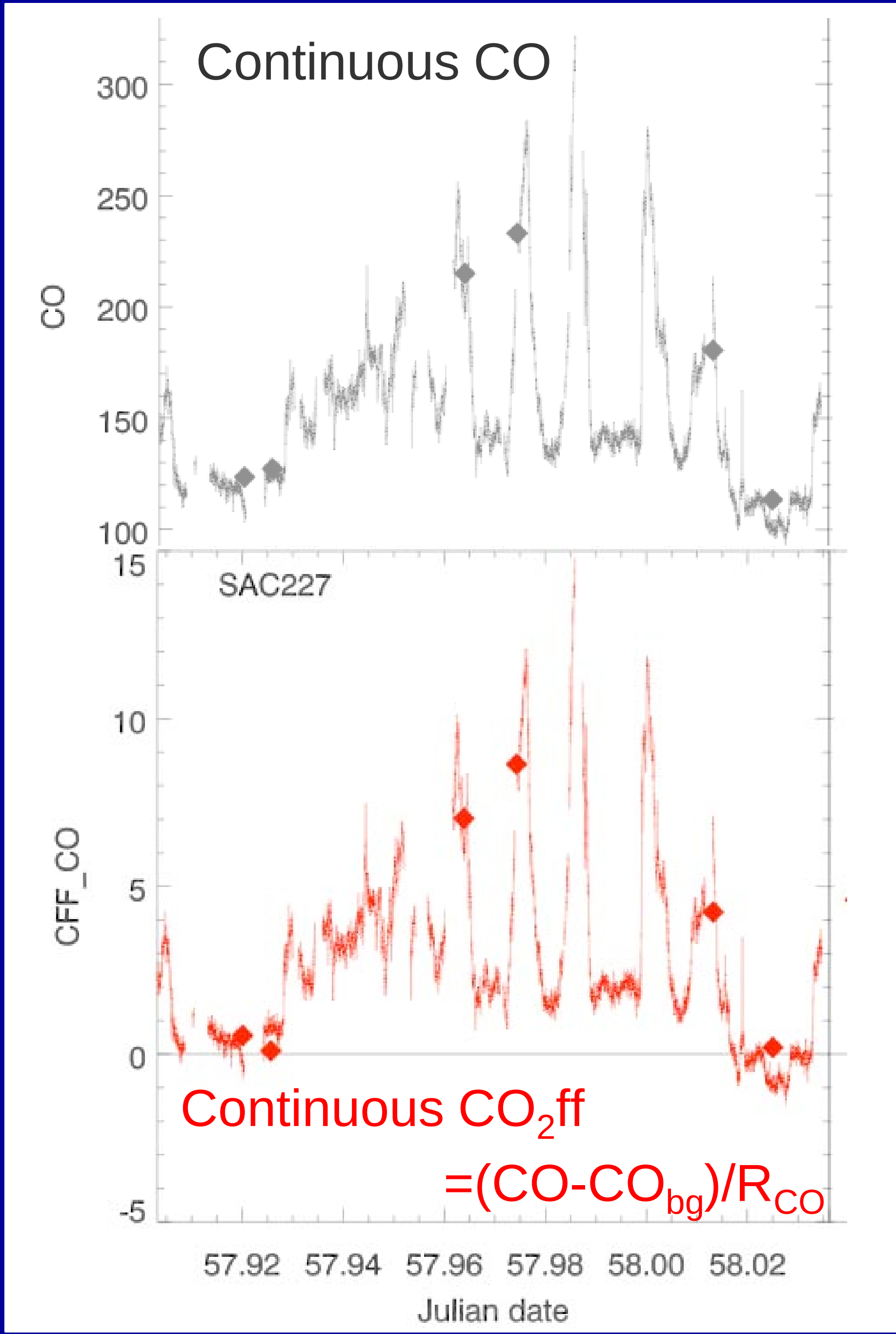


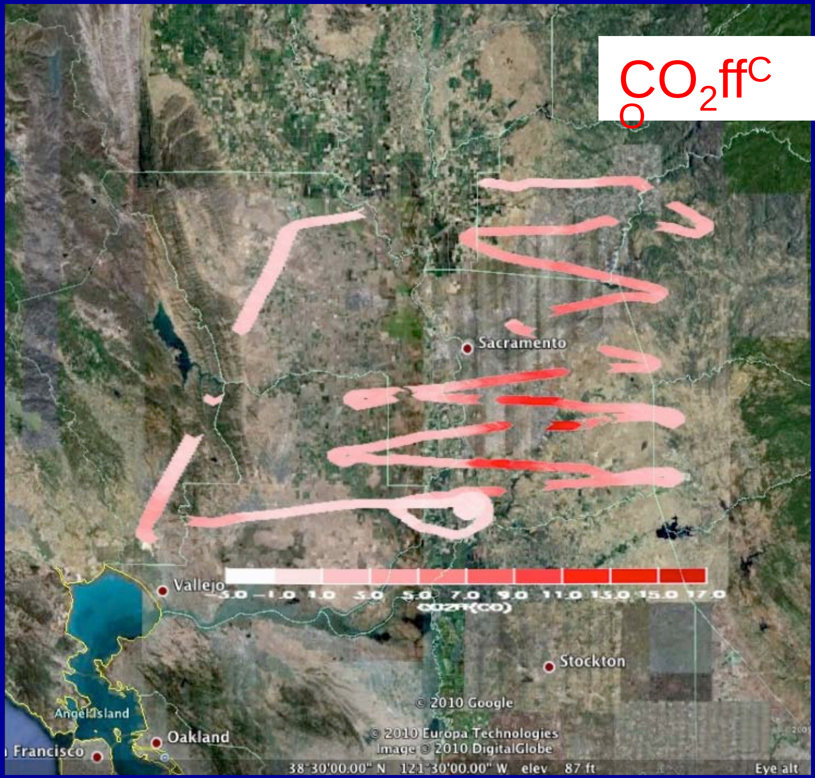
Our observed ratio is within the range of other observations

Excellent agreement with CEPAM/Vulcan CO/CO_2 inventories

NEI inventories appear to overestimate CO emissions

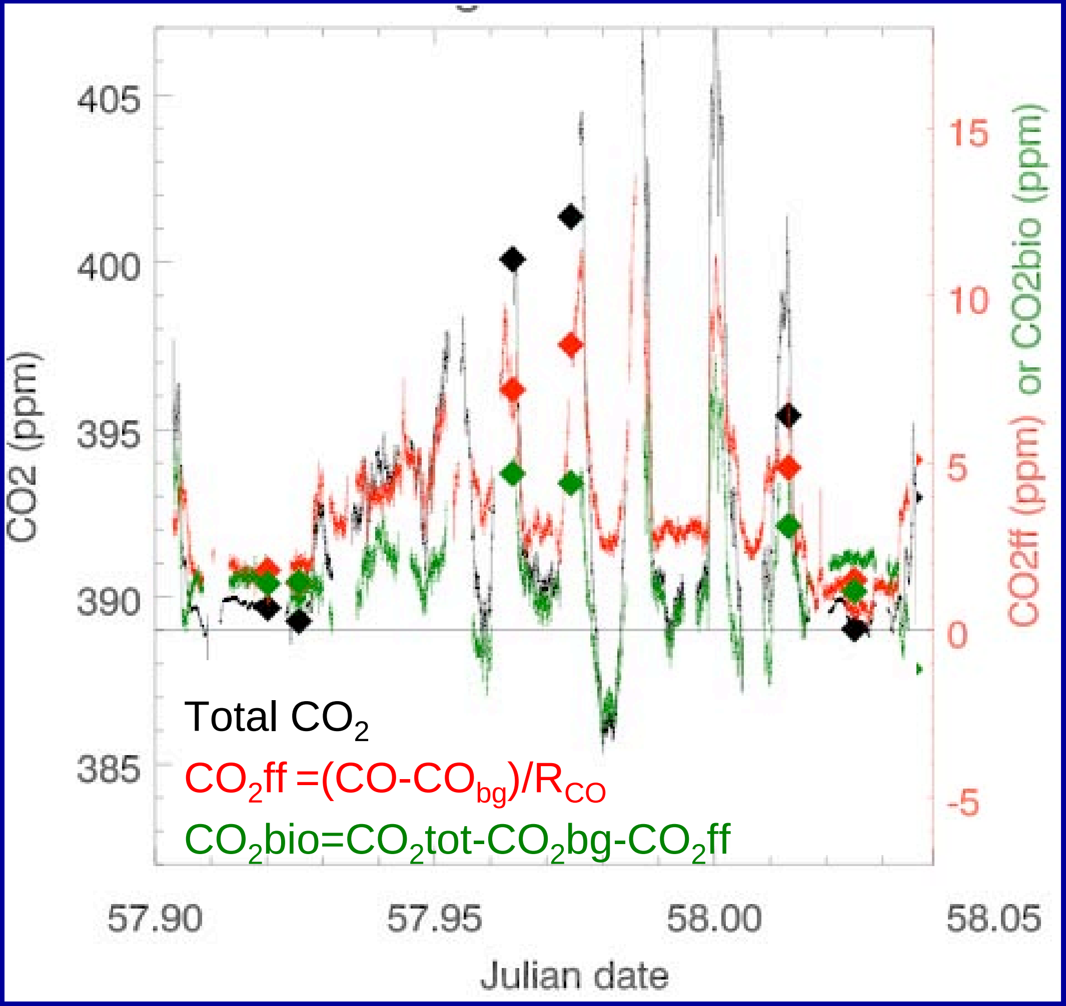
Continuous CO_2ff from continuous CO observations

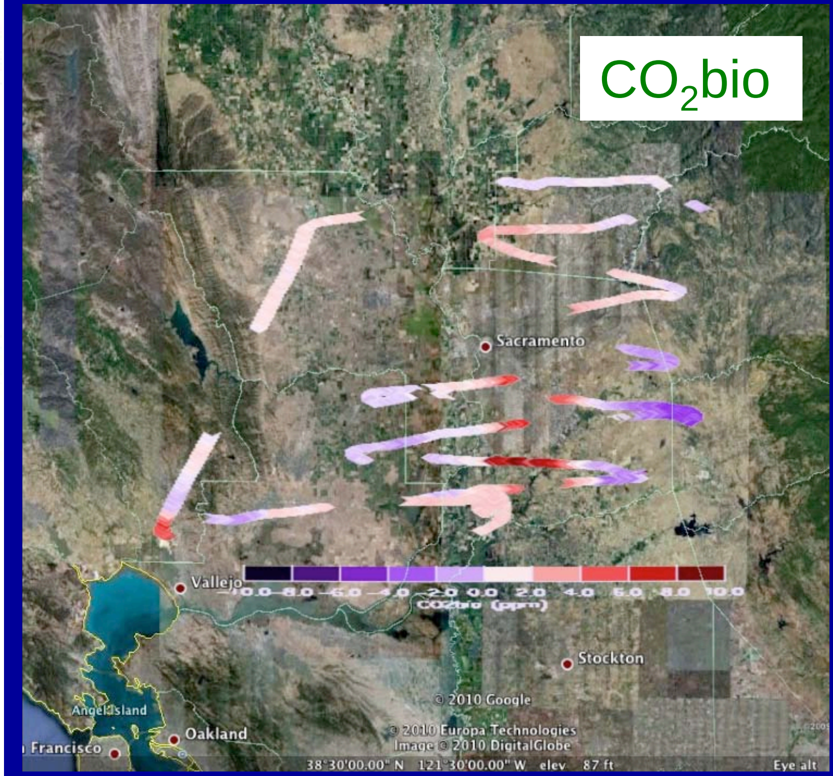




R_{CO} = 14±2 ppb/ppm from slope of CO:CO_2ff in these flights

Continuous CO_2ff: partitioning of CO_2

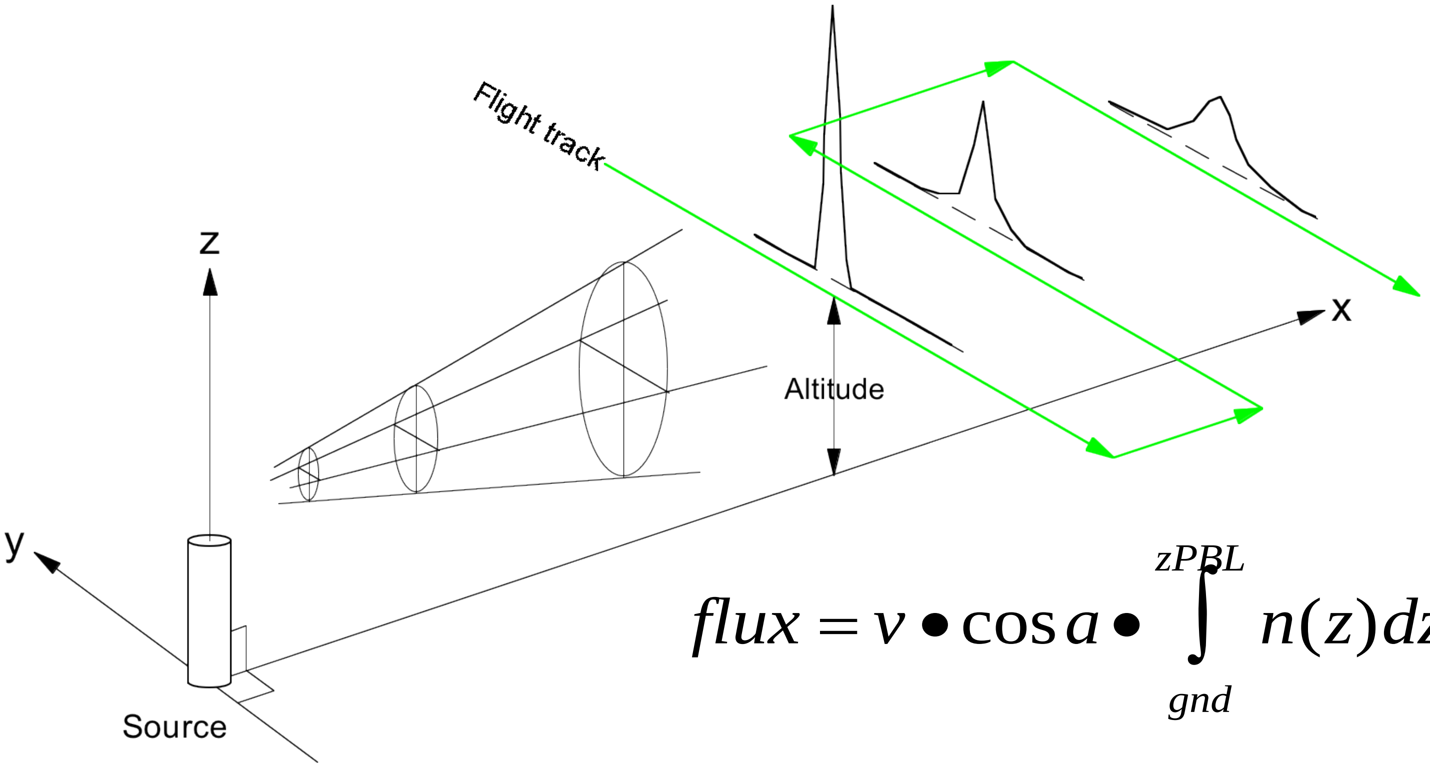




CO_2ff explains most, but not all, CO_2 variability over Sacramento

Variable CO_2bio contribution

Quantifying emissions using a mass balance approach



$$flux = v \bullet \cos a \bullet \int_{gnd}^{zPBL} n(z)dz \bullet \int_{-y}^{y} M(y)dy$$

variables:
v: wind at t = 0
a: angle between wind normal and track
z: height above sea level
n: atmospheric number density
M: mixing ratio enhancement

assumptions:
wind history is constant

pbl height known, flux well-mixed in vertical

vertical uniformity within mixed layer

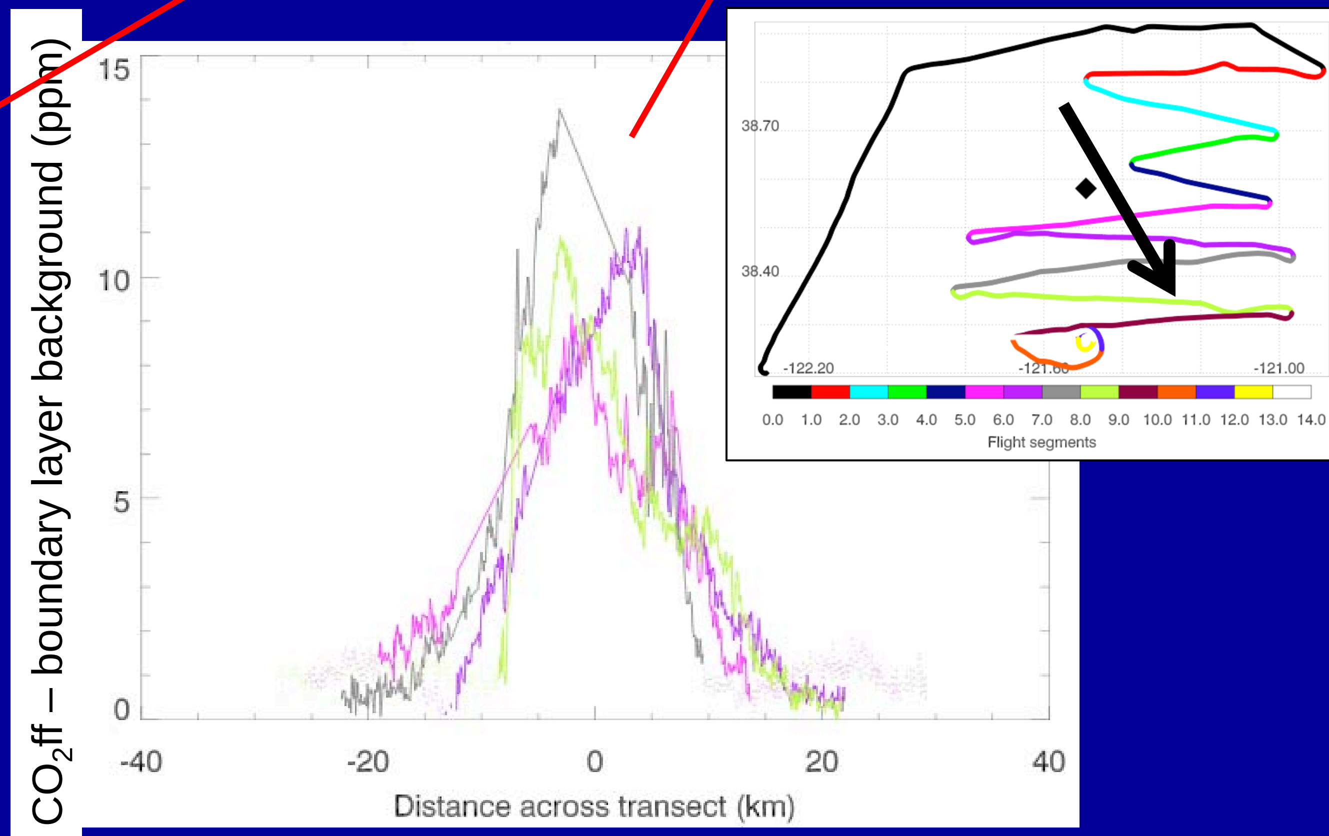
from Ryerson et al., 2001

Sacramento 2009: Quantifying CO_2ff emissions using a mass balance approach

$$flux = v \bullet \cos a \bullet \int_{gnd}^{zPBL} n(z)dz \bullet \int_{-y}^{y} M(y)dy$$

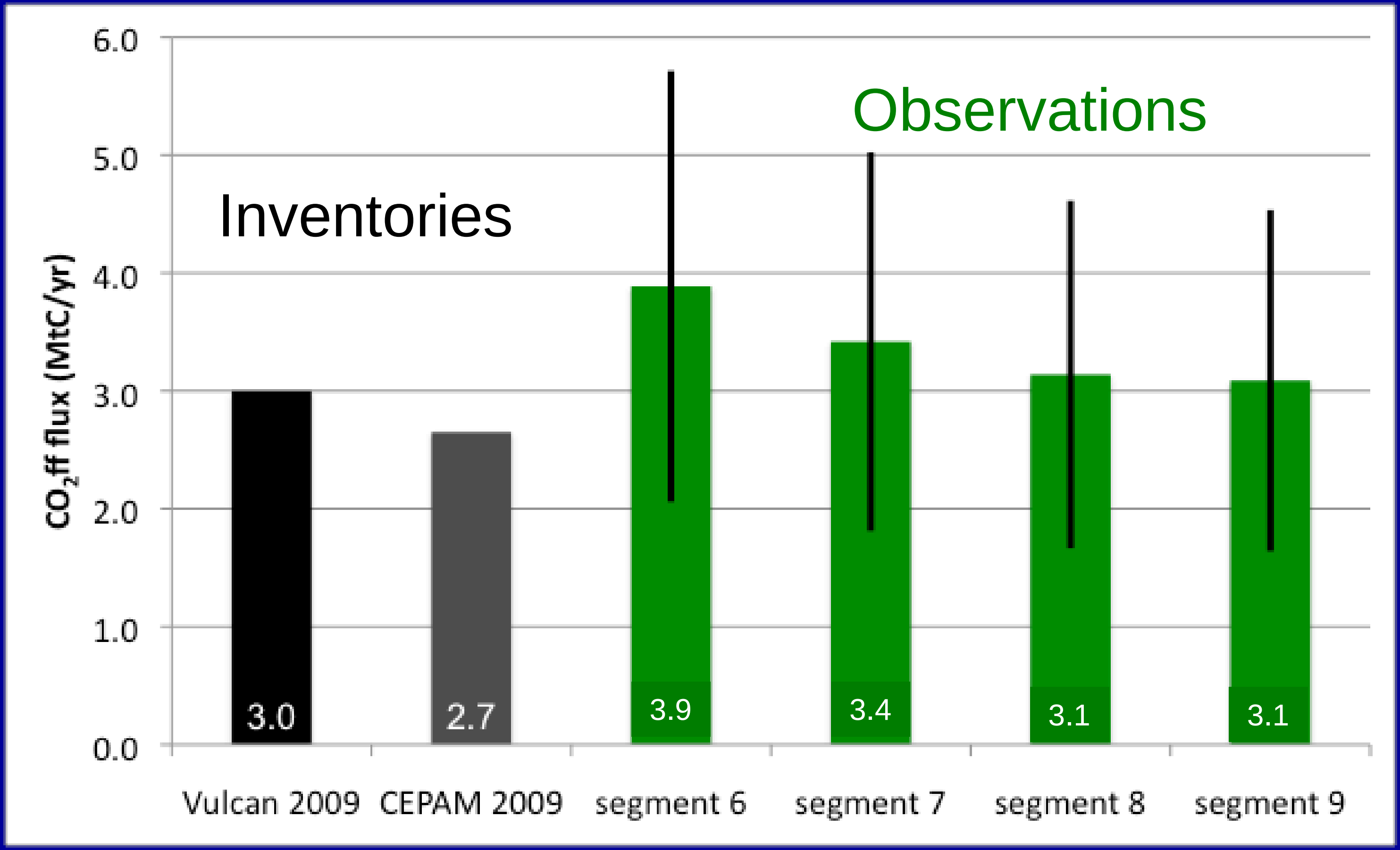
Wind speed
1.0 – 1.7 m/s
(NARR reanalysis)

PBL height estimates:
1500 m (NARR reanalysis)
750 m (flight vertical profile



Calculate flux independently for each downwind transect

Sacramento 2009: Calculated CO_2ff flux



Large uncertainties mostly due to uncertainty in wind speed and pbl height

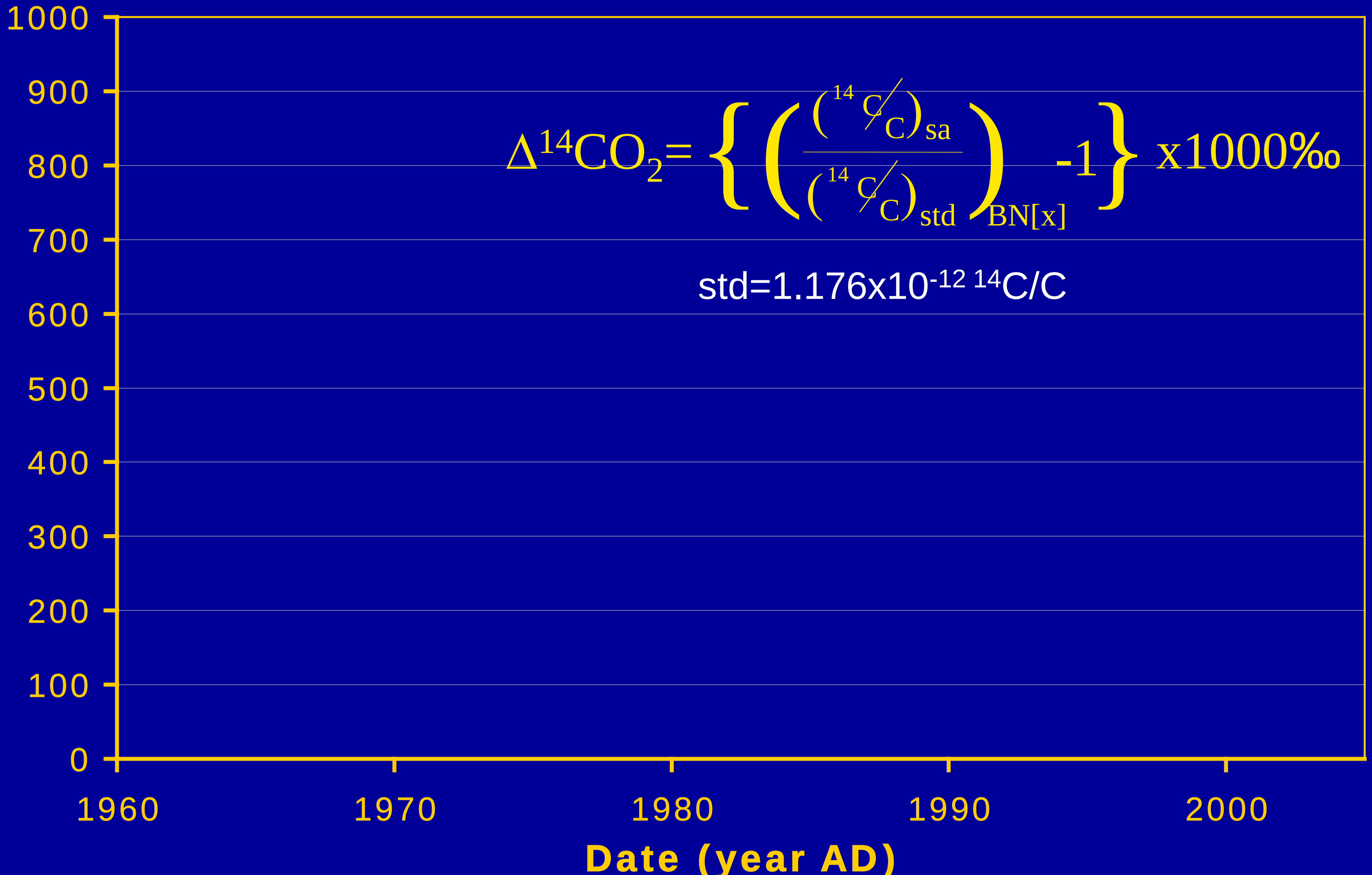
Good agreement with inventories

Conclusions

- A few $^{14}CO_2$ measurements can go a long way
- Can evaluate bottom up inventories using emission ratios
  - Good agreement with recent inventories – (Vulcan for CO_2ff, CEPAM for CO)
  - NEI CO inventories too high
  - without ^{14}C-based CO_2ff, would get “wrong” emission ratios
- Combine flask $^{14}CO_2$ measurements with continuous CO to get continuous CO_2ff and CO_2bio
  - Large variability in CO_2bio contribution
- Estimate urban area CO_2ff flux using mass balance
  - Large uncertainties in this example due to wind and pbl height uncertainties
  - Could be improved, with met measurements, more flights, and more sophisticated models (eg WRF-CHEM)

Time evolution of $\Delta^{14}CO_2$

Northern Hemisphere records from Germany and Colorado



$$\Delta^{14}CO_2 = \left\{ \left(\frac{\left({}^{14}C/C\right)_{sa}}{\left({}^{14}C/C\right)_{std}} \right)_{BN[x]} - 1 \right\} \times 1000‰$$

$std = 1.176 \times 10^{-12}\ {}^{14}C/C$

Levin and Kromer, 2004 and Turnbull et al., 2007

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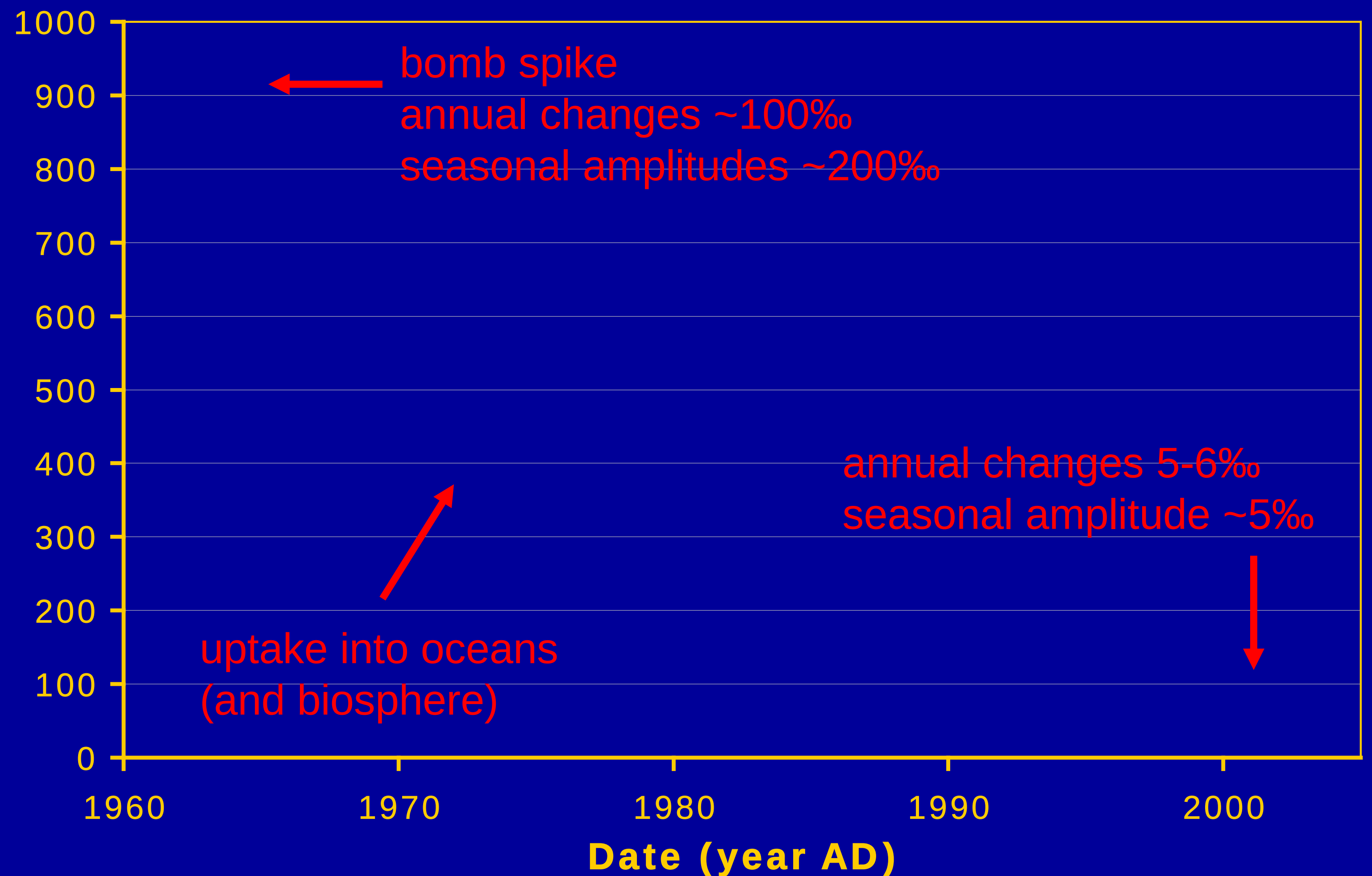

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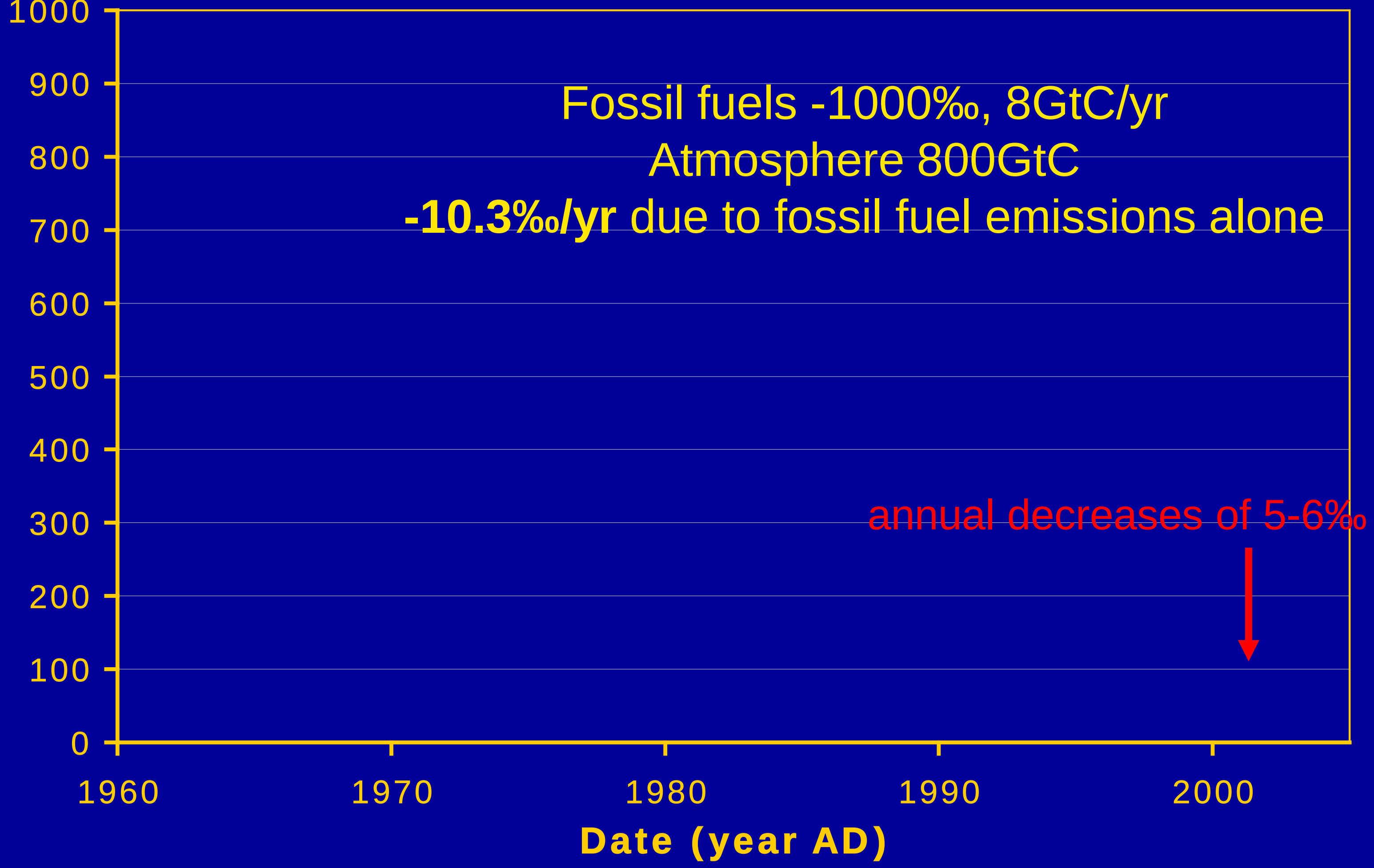

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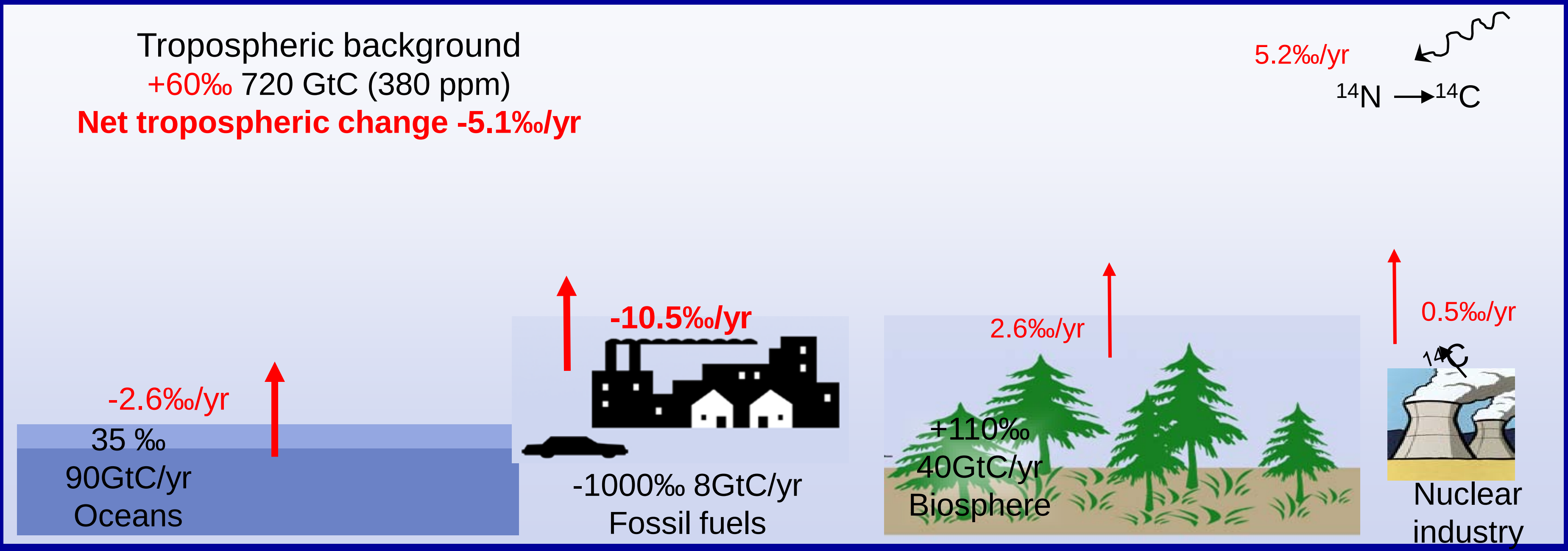
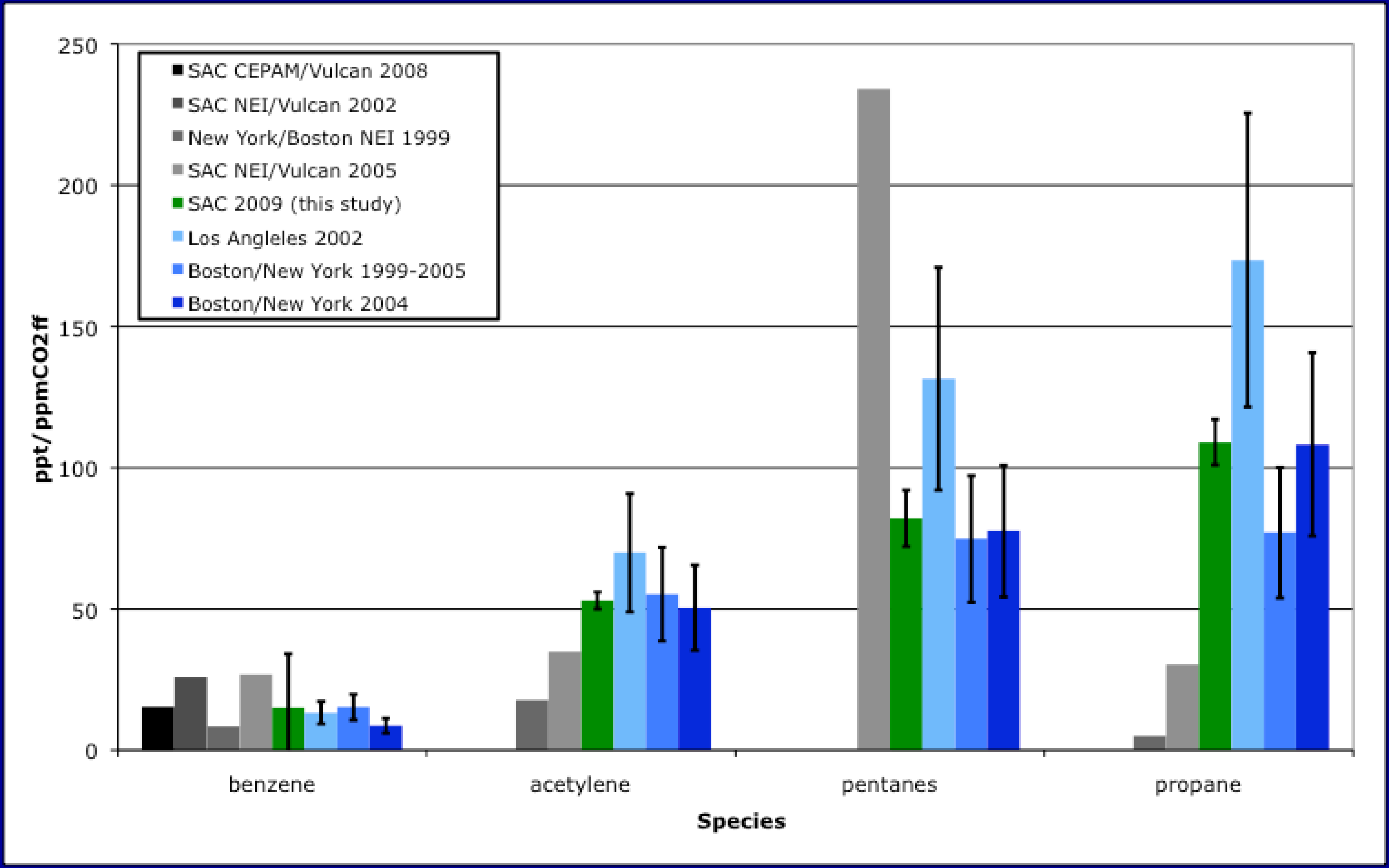
^{14}C in the global carbon cycle



$$\Delta\Delta^{14}CO_{2(source)} \approx (\Delta_{source} - \Delta_{atm}) \frac{C_{source}}{C_{atm}}$$

Emission ratios: evaluation of hydrocarbon inventories



Our observations generally agree with other observational studies

Sometimes large differences from inventories

Correlate tracer methods carbon monoxide

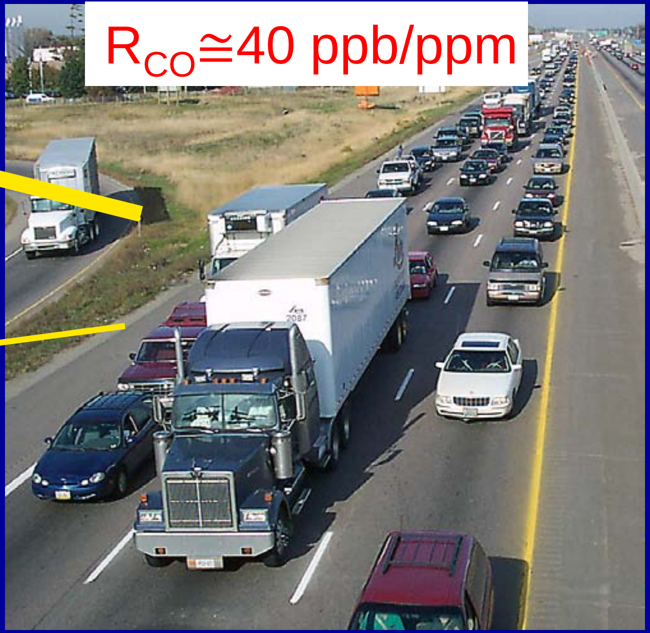
$$CO_{obs} = CO_{bg} + CO_{ff} + CO_{other}$$

$$R_{CO} = \frac{CO}{C_{ff}}$$

$$C_{ff} = \frac{(CO_{obs} - CO_{bg})}{R_{CO}}$$

$R_{CO} \cong 0.1$ ppb/ppm

$R_{CO} \cong 40$ ppb/ppm



CO_2

CO

CO co-emitted with fossil fuel CO_2