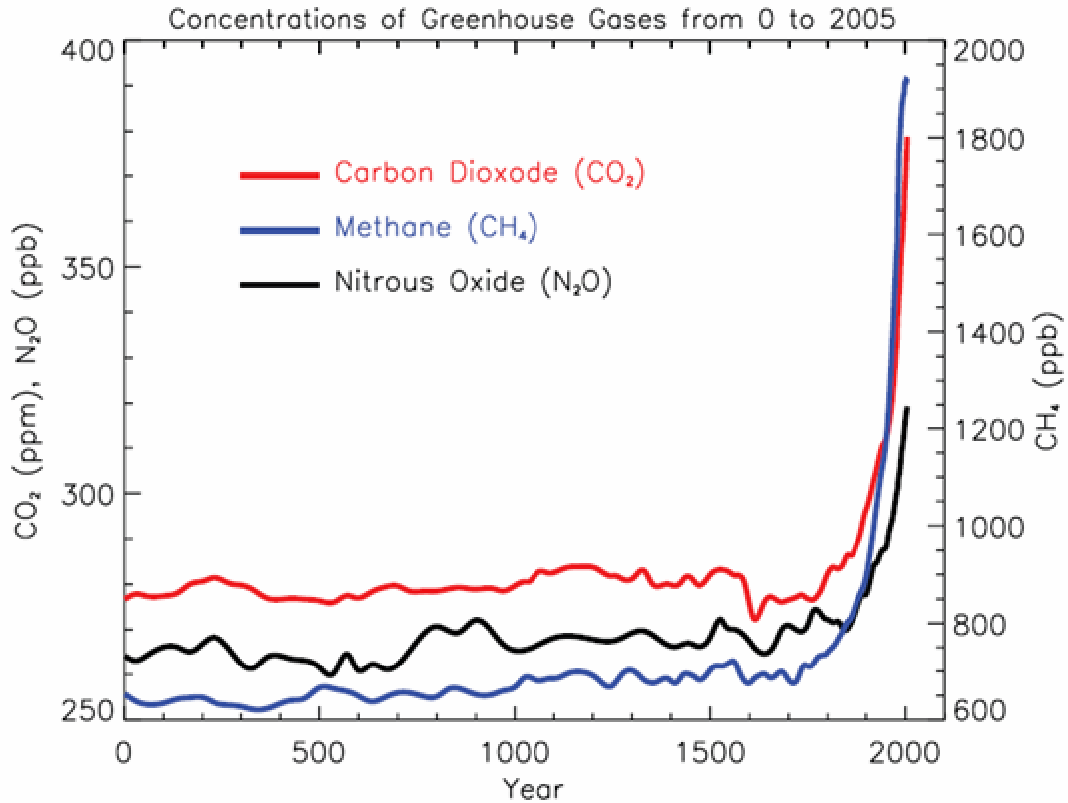
Emissions of CH_4 and CO in the Los Angeles Area

Debra Wunch, Paul Wennberg, Geoff Toon, Gretchen Keppel Aleks, Yael Yavin

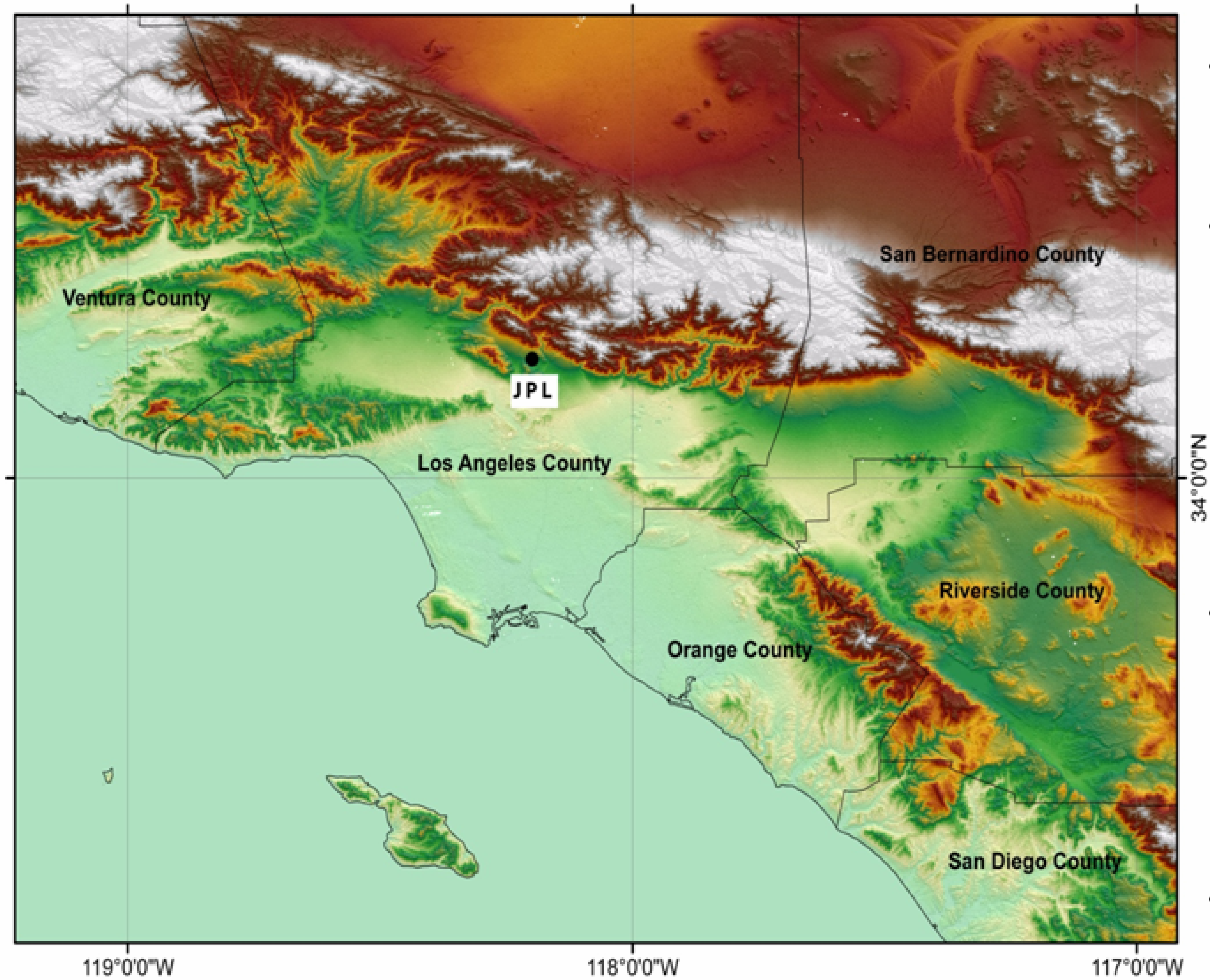
Motivation: Are Cities a Major Source of CH_4?

- CH_4 has the second-largest radiative forcing (0.48 W/m^2), after CO_2 (1.66 W/m^2)
- Total CH_4 in atmosphere is known (1775 ppb), as are the total emissions per year (582 Tg CH_4)
- Exactly where this CH_4 comes from is **not** known
- According to the IPCC:
  - 70% of the global source of methane is biogenic
    - methanogens in wetlands, rice paddies, ruminants, landfills, oceans and forests.
  - Non-biogenic sources include fossil fuel mining and burning, biomass burning, waste treatment and geological sources
- We had unique opportunity to look at a nearly year-long time series of CH_4, CO_2 and CO in a highly polluted urban area



FAQ 2.1, Figure 1. *Atmospheric concentrations of important long-lived greenhouse gases over the last 2,000 years. Increases since about 1750 are attributed to human activities in the industrial era. Concentration units are parts per million (ppm) or parts per billion (ppb), indicating the number of molecules of the greenhouse gas per million or billion air molecules, respectively, in an atmospheric sample. (Data combined and simplified from Chapters 6 and 2 of this report.)*

Instrument Location



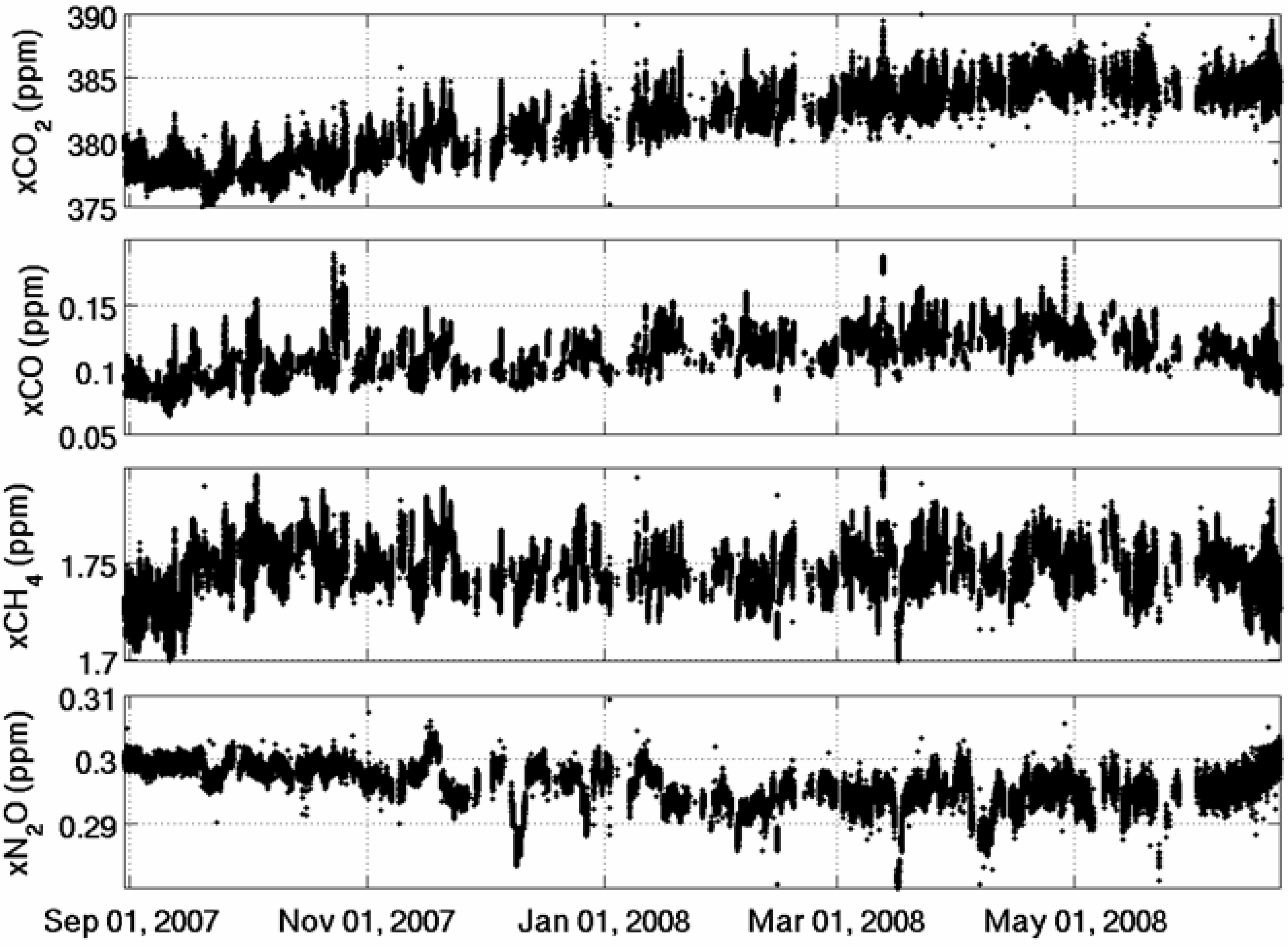
- TCCON container at JPL from Aug. 07 – Jun. 08
- In the South Coast air basin (SCB)
  - Highly polluted
  - 15 million inhabitants (~40% of California)
- Bounded on three sides by mountains, and the Pacific Ocean on the fourth
- Well-contained air mass

Time Series Measurements

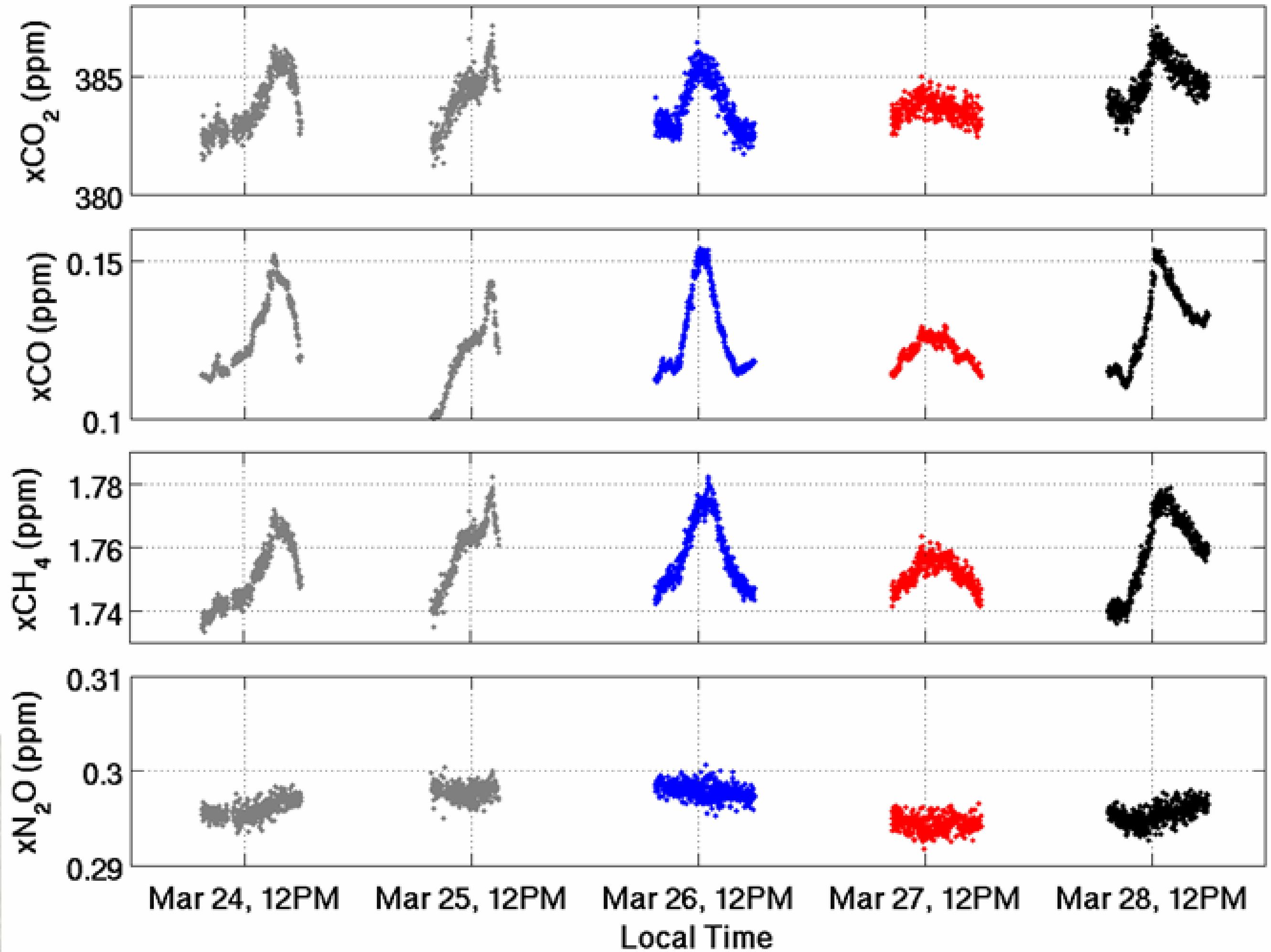
- Dry-air mole fractions (DMF):

$$xG = 0.2095\frac{column_G}{column_{O_2}}$$

- DMF insensitive to boundary layer height changes, surface pressure changes
- Slow, likely seasonal, changes apparent
- Faster, diurnal changes also visible



Diurnal Changes



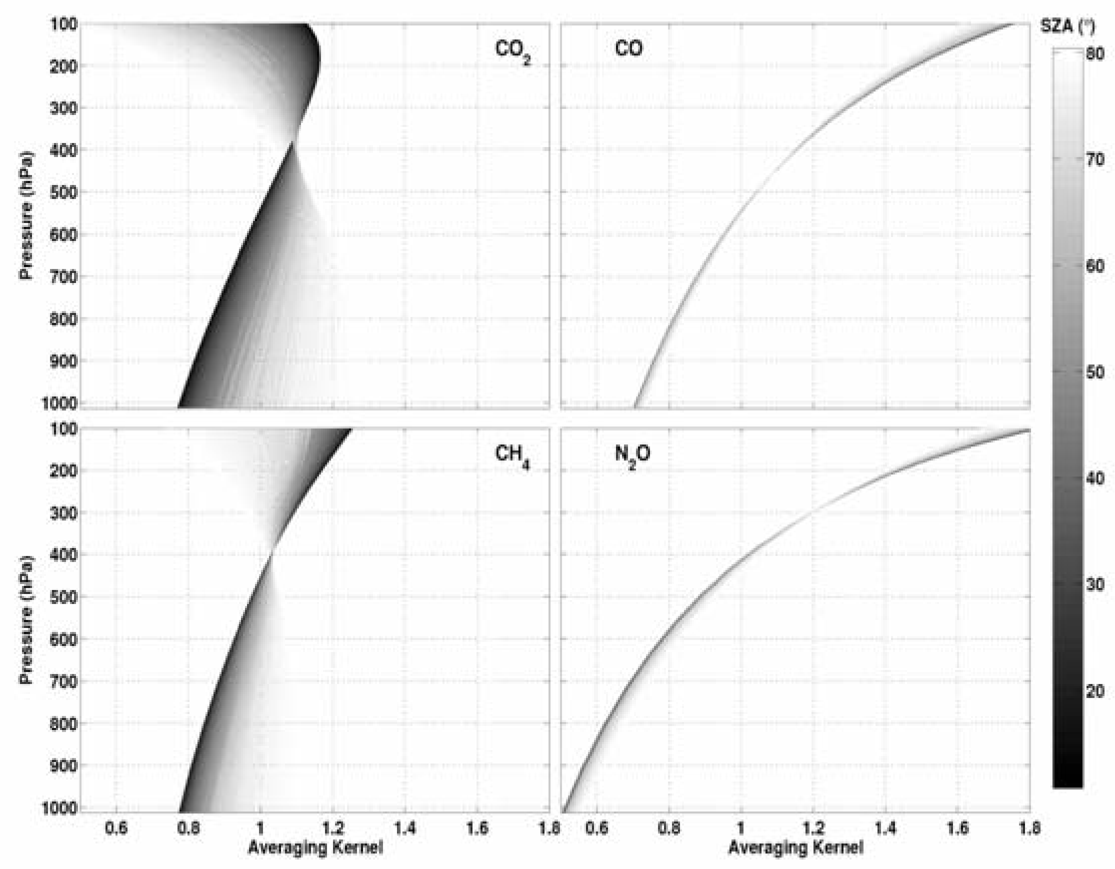
Diurnal signal seen in basin air (blue, black and grey curves)

- Build-up in morning until PBL reaches tops of surrounding mountains, then decrease seen

Smaller diurnal signal seen from air originating from (clean-air) Mojave Desert (red)

Analysis Method

- Compute daily anomalies by subtracting morning value at a certain SZA from its afternoon counterpart
  - Independent of any air mass dependency
  - Removes any seasonal cycle
- Assume all emissions originate from the surface layer
  - Divide anomalies by the averaging kernel value at the surface
- Correlate xCO and xCH_4 with xCO_2
  - Compute correlation slope of gas G wrt xCO_2 (α_G mol/mol)
  - Convert mol/mol into g/g using molecular mass ratio (M_G/M_{CO2})

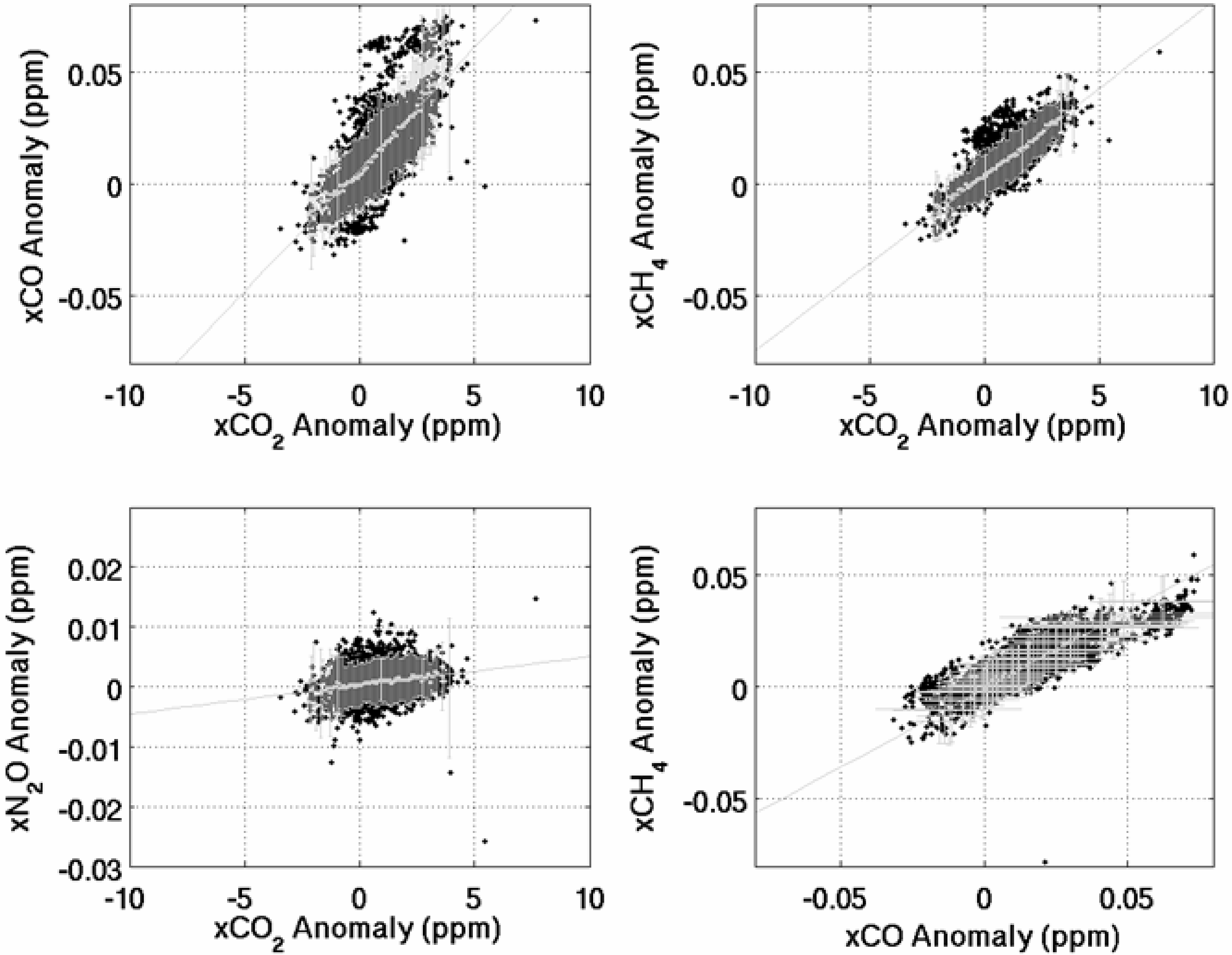


- Emissions of xCO and xCH_4 can be computed if emissions of xCO_2 are known

$$E_G^{SCB} = \alpha_G \frac{M_G}{M_{CO_2}} E_{CO_2}^{SCB}$$

Unknown

Correlation Results

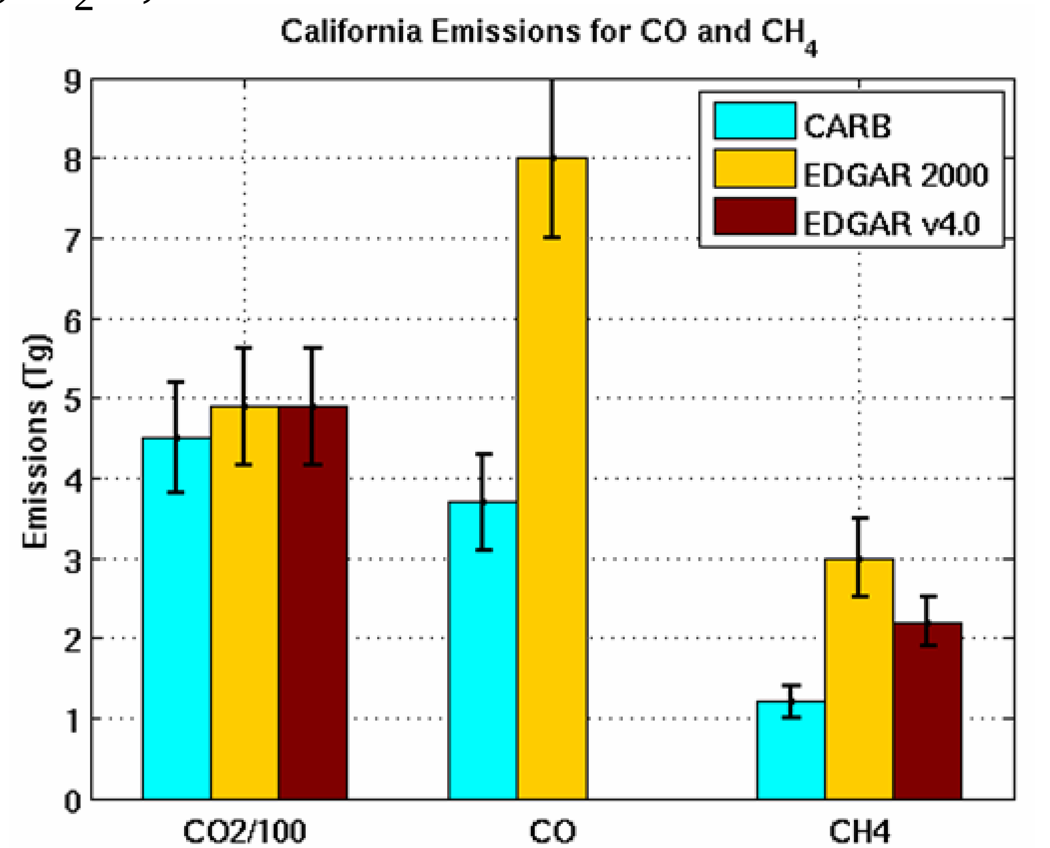


	xCO_2 (per mil) α_G	Mass Ratio (M_G/M_{CO_2})
xCO	11 ± 2	28/44
xCH_4	7.8 ± 0.8	16/44
xN_2O	0.5 ± 0.3	44/44

- Diurnal changes in the data show strong and significant correlation slopes (α_G in mol/mol) between the gas anomalies

Finding $E_{CO_2}^{SCB}$: Inventories

- California Air Resources Board (CARB)
  - State-wide emissions of CO_2, CH_4, N_2O, CO
    - SCB emissions of CO
  - Bottom-up accounting
    - Split into sector (agriculture and forestry, industrial, etc.)
- Emission Database for Global Atmospheric Research (EDGAR)
  - 2000: 1x1-degree global data
  - V4.0 (2005): 0.1x0.1-degree
  - No breakdown of data by source



- CARB and EDGAR CO_2 for SCB agree within error, giving

$$E_{CO_2}^{SCB} = 196 \pm 29 \text{ Tg CO}_2$$

SCB Emissions

- From the EDGAR/CARB CO_2 emissions and the correlation coefficients, emissions for CO, CH_4, N_2O are computed

$$E_G^{SBC} = \alpha_G \frac{M_G}{M_{CO_2}} E_{CO_2}^{SBC}$$

$$E_{CO_2}^{SCB} = 196 \pm 29 \ \text{Tg}\, CO_2$$

	xCO_2 (per mil) α_G	Mass Ratio (M_G/M_{CO_2})
xCO	11 ± 2	28/44
xCH_4	7.8 ± 0.8	16/44
xN_2O	0.5 ± 0.3	44/44

	State Wide Emissions		SCB Emissions		
	CARB	**EDGAR**	**CARB**	**EDGAR**	**FTS**
CO_2 (Tg CO_2)	450	489	194	197	196±29
CO (Tg CO)	4.124	9.799	1.240	4.346	1.4±0.3
CH_4 (Tg CH_4)	1.236	2.959	—	1.270	0.6±0.1
N_2O (Tg N_2O)	0.049	0.080	—	0.023	0.10±0.06

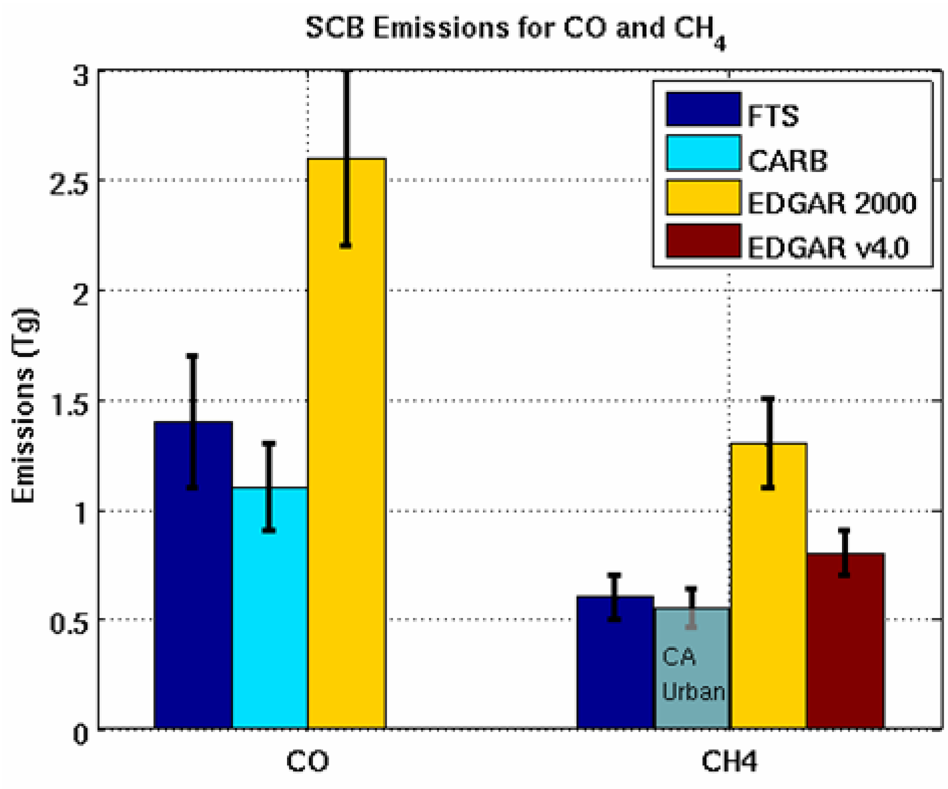
SCB Emissions

CO

- EDGAR 2000 overestimates SCB CO
  - Steady decrease in CO emissions since 2000, but EDGAR 2000 still reads high
- CARB and FTS estimates of CO agree well
  - CARB: 1.1 ± 0.2 Tg CO; FTS: 1.4 ± 0.3 Tg CO

CH_4

- EDGAR 2000 overestimates SCB CH_4
- EDGAR v4.0 agrees well
- CARB underestimates CH_4



	State Wide Emissions		SCB Emissions		
	CARB	EDGAR	CARB	EDGAR	FTS
CO_2 (Tg CO_2)	450	489	194	197	196±29
CO (Tg CO)	4.124	9.799	1.240	4.346	1.4±0.3
CH_4 (Tg CH_4)	1.236	2.959	—	1.270	0.6±0.1
N_2O (Tg N_2O)	0.049	0.080	—	0.023	0.10±0.06

Comparison with CARB SCB Emissions of CH_4

- CARB underestimates the SCB's contribution to CH_4
  - 54% of CARB California-wide CH_4 is from Agriculture & Forestry
  - 5% of California's Agriculture and Forestry occurs in SCB
  - Results in >90% of the state's non-Agriculture and Forestry CH_4 emissions from SCB
    - Very unlikely, since SCB only 50% of California's urban population
- CARB seems to be missing (or improperly-assigning) a source of CH_4 in the SCB

	State Wide Emissions		SCB Emissions		
	CARB	EDGAR	CARB	EDGAR	FTS
CO_2 (Tg CO_2)	450	489	194	197	196±29
CO (Tg CO)	4.124	9.799	1.240	4.346	1.4±0.3
CH_4 (Tg CH_4)	1.236	2.959	—	1.270	0.6±0.1
N_2O (Tg N_2O)	0.049	0.080	—	0.023	0.10±0.06

Conclusions (or Questions Raised)

- Is CARB missing (or improperly-assigning) a source of CH_4 in the SCB?
  - FTS measurements unable to determine source: require in situ measurements
- Do other urban areas produce similar correlation coefficients to the SCB?
  - Is there an un-catalogued CH_4 source from urban areas?
- If the correlation coefficient is similar to other urban regions, this could mean, as an upper limit, that 21%-34% of the global anthropogenic emissions could be due to the urban source.

References	Indicative ^{13}C, ‰[b]	Hein et al., 1997[c]	Houweling et al., 2000[c]	Olivier et al., 2005	Wuebbles and Hayhoe, 2002	Scheehle et al., 2002	J. Wang et al., 2004[c]	Mikaloff Fletcher et al., 2004a[c]	Chen and Prinn, 2006[c]	TAR	AR4
Base year		1983–1989		2000		1990	1994	1999	1996–2001	1998	2000–2004
Natural sources			**222**		**145**		**200**	**260**	**168**		
Wetlands	–58	231	163		100		176	231	145		
Termites	–70		20		20		20	29	23		
Ocean	–60		15		4						
Hydrates	–60				5		4				
Geological sources	–40		4		14						
Wild animals	–60		15								
Wildfires	–25		5		2						
Anthropogenic sources		**361**		**320**	**358**	**264**	**307**	**350**	**428**		
Energy						74	77				
Coal mining	–37	32		34	46			30	48[d]		
Gas, oil, industry	–44	68		64	60			52	36[e]		
Landfills & waste	–55	43		66	61	69	49	35			
Ruminants	–60	92		80	81	76	83	91	189[f]		
Rice agriculture	–63	83		39	60	31	57	54	112		
Biomass burning	–25	43			50	14	41	88	43[e]		
C3 vegetation	–25			27							
C4 vegetation	–12			9							
Total sources		**592**			**503**		**507**	**610**	**596**	**598**	**582**
Imbalance		+33								+22	+1
Sinks											
Soils	–18	26			30		34	30		30	30[g]
Tropospheric OH	–3.9	488			445		428	507		506	511[g]
Stratospheric loss		45			40		30	40		40	40[g]
Total sink		**559**			**515**		**492**	**577**		**576**	**581[g]**

Notes:

[a] Table shows the best estimate values.

[b] Indicative ^{13}C values for sources are taken mainly from Mikaloff Fletcher et al. (2004a). Entries for sinks are the fractionation, $(k_{13}/k_{12}-1)$ where k_n is the removal rate of $^nCH_4$; the fractionation for OH is taken from Saueressig et al. (2001) and that for the soil sink from Snover and Quay (2000) as the most recent determinations.

[c] Estimates from global inverse modelling (top-down method).

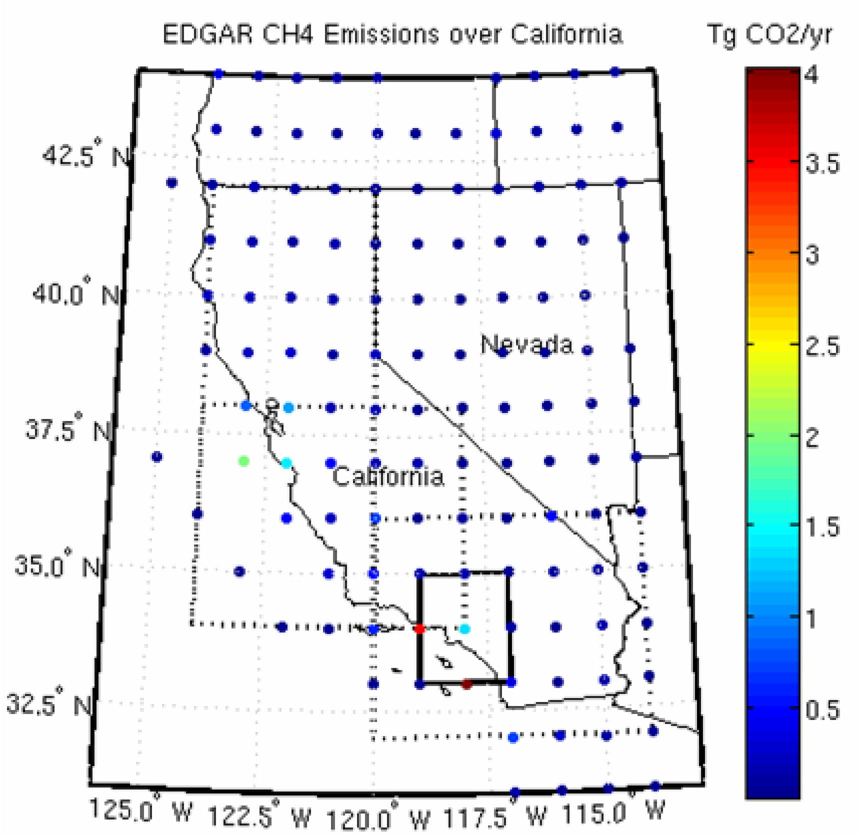
[d] Includes natural gas emissions.

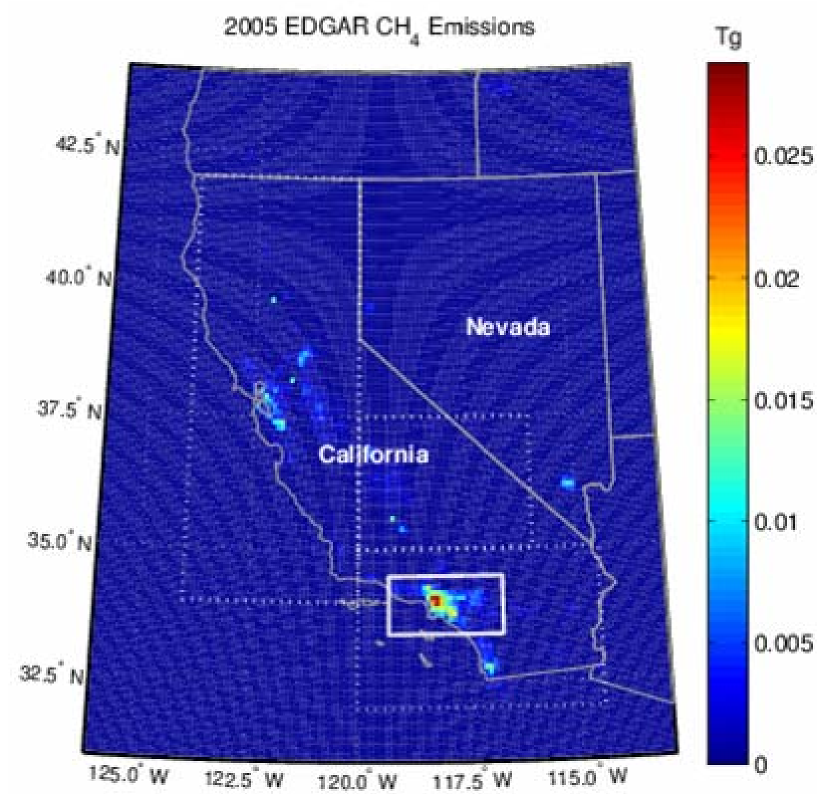
[e] Biofuel emissions are included under Industry.

[f] Includes emissions from landfills and wastes.

[g] Numbers are increased by 1% from the TAR according to recalibration described in Chapter 2.

Comparisons with EDGAR SCB CH_4



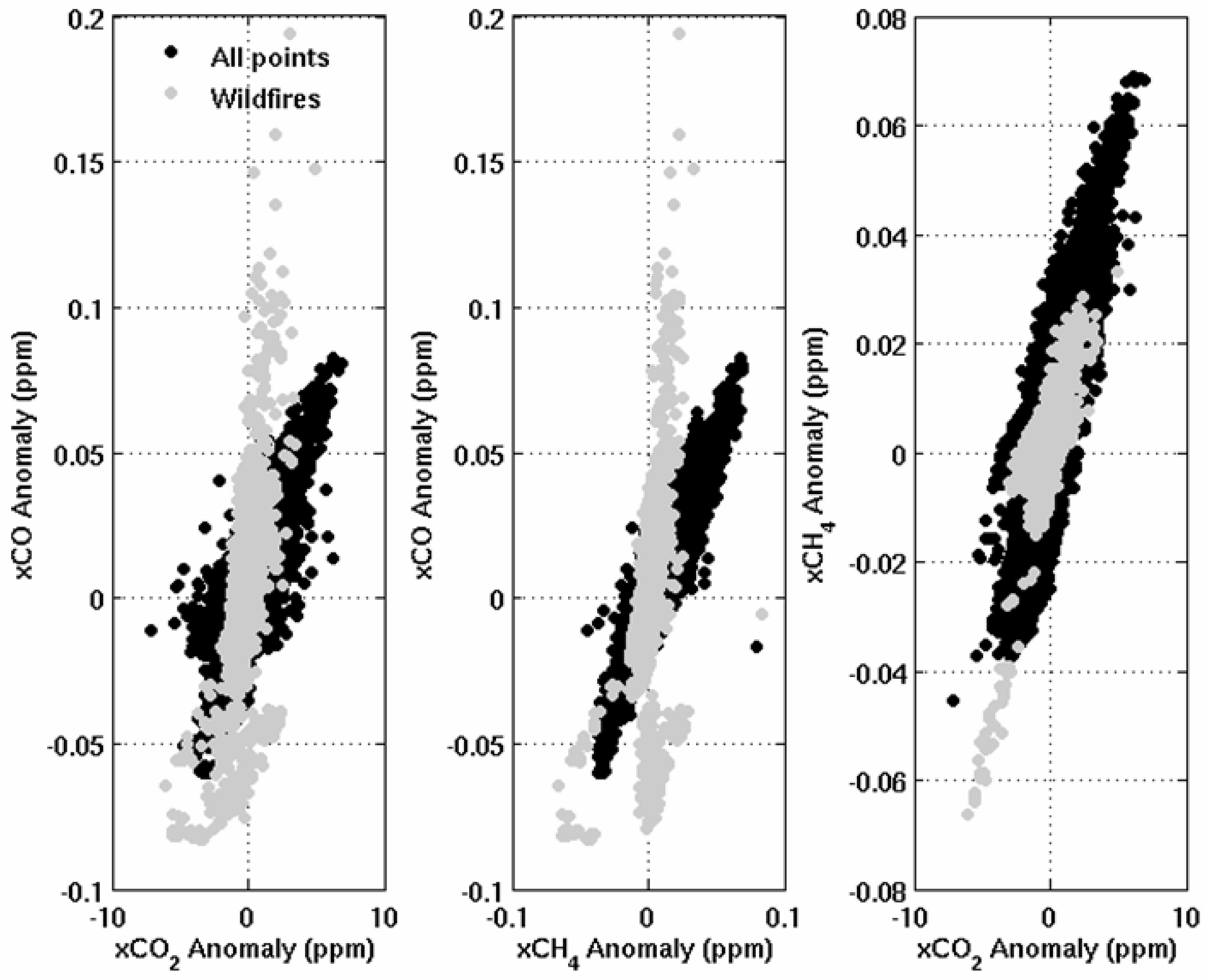


- EDGAR 2000 overestimates CH_4 in the SCB
- EDGAR v4.0 (released May 19, 2009)
  - 0.1x0.1 degree grid; 2005; only GHGs (no CO, yet)
- EDGAR v4.0 SCB:
  - 0.78 Tg CH4; 223.872 Tg CO2; 0.016 Tg N2O
- EDGAR v4.0 CA:
  - 2.24 Tg CH4; 488.743 Tg CO2; 0.049 Tg N2O
- EDGAR v4.0 is in *much* better agreement with the JPL FTS results

Data Filtering

Wildfires:

Days affected by wildfires were removed from the time series because they produce a very distinctly different xCO-xCO_2 correlation slope (grey)



Data Filtering

- Water contamination:
  - Ensure that the days are dry (do not conflate correlations in H_2O with correlations between the molecules of interest).
  - Maximum xH_2O in the spectrum is limited to 2600 ppm.
- Origin of Air
  - Ensure that the air originates from the SCB (i.e. is not the clean air from the north).
  - Limit data to days in which the change in xCO_2 is >2 ppm.
- Tropopause Height
  - DMF measurements are relatively insensitive to boundary layer height, however, xCH_4 and xN_2O are somewhat sensitive to tropopause height.
  - Accept days only when there are small anomalies in HF (<0.015 ppb).