Multi-tracer Measurements and Inversions Quantifying California's GHG Emissions Budget

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  CH4 measurement network and emission inventory development
- UCSD-Scripps: R. Keeling
  GHG measurements at background sites
- LLNL: C. Zhao, T. Guilderson
  Inverse model and $^{14}CO_2$ analysis
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  AGES CO measurements
- AER Inc.: J. Eluszkiewicz, T. Nehrkorn
  WRF-STILT development
- JPL: C. Frankenberg
  SCIAMACHY CH_4 retrievals
- Caltech-UHouston: S. Newman, B. Leffer
  Pasadena Lidar PBL depths

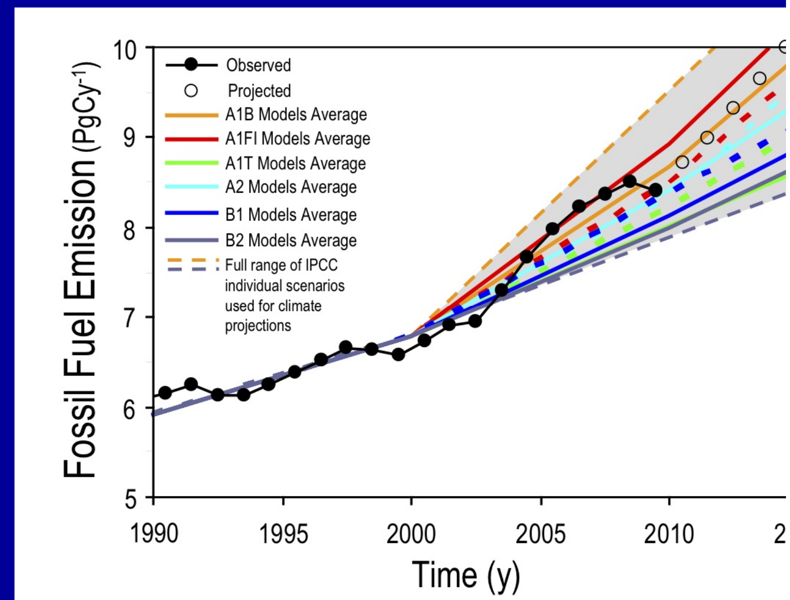
Outline

- Need for GHG emissions control validation
- Initial work for CEC and US-DOE in Central California
- Progress on CARB Inverse Modeling Project
- Discussion of New Research

Emissions Reduction Validation is Crucial for Future Climate Control

- **Global GHG emissions near IPCC A1F1 "high growth" scenario**
- **UNFCC reporting of GHG emissions currently optional**
- **Progress at Copenhagen on emissions reductions agreements limited, in part, by lack of verification capability. President Obama: "[verification] must, however, ensure that an accord is credible, and that we are living up to our mutual obligations."**
- **National Academy study: "Verifying Greenhouse Gas Emissions: Methods to Support International Climate Agreements"**

Net Global Carbon Dioxide Emissions (pgC y



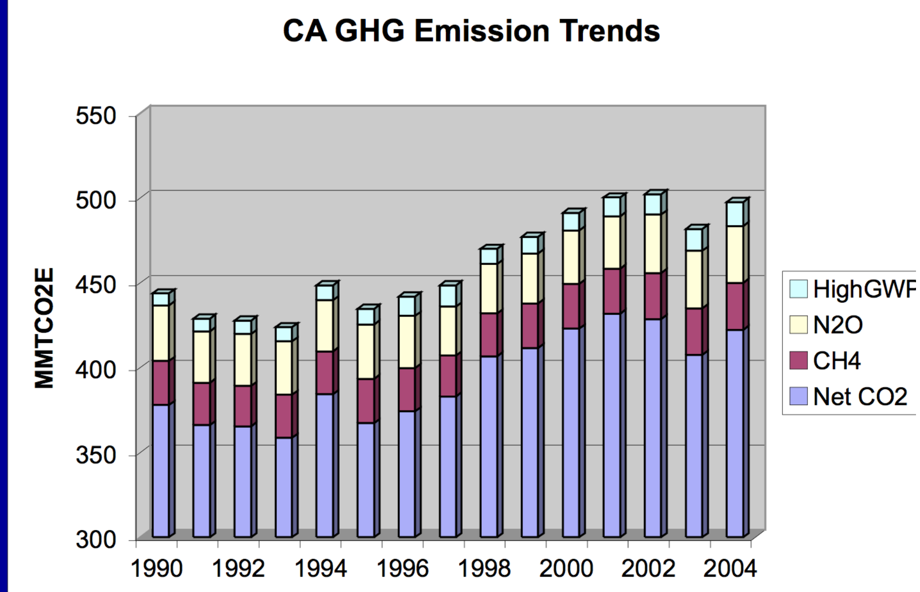
Global Carbon Project
Adapted from Raupach et al. 2007, PN

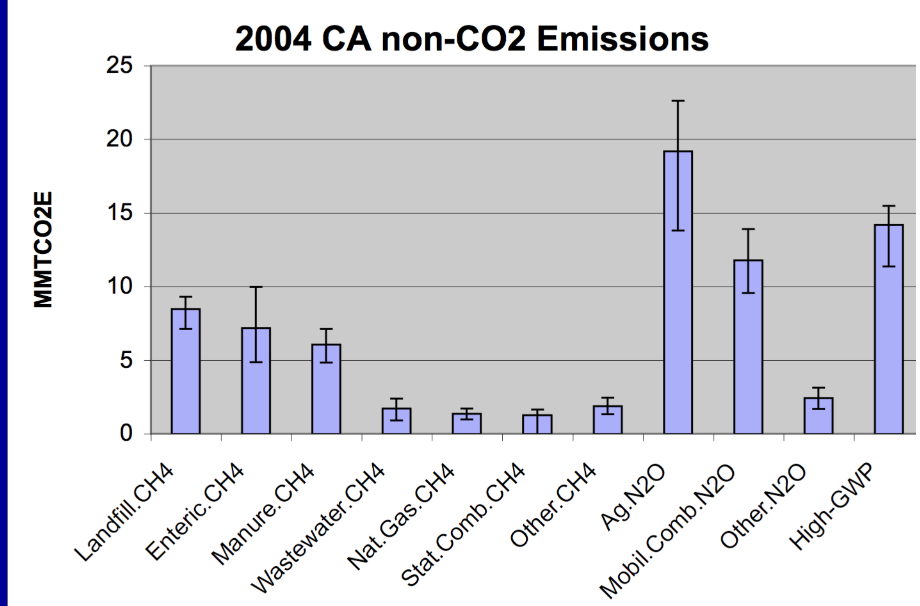
California emissions reductions policy (AB-32) drives the need for validation capability today

California GHG Emissions

- California is the first state in US to legislate GHG controls
  - AB-32: Return to 1990 levels by 2020
- Sum of non-CO_2 GHG emissions comparable to fossil fuel CO_2 but
  - Industrial and biological sources are not readily metered
  - Uncertainties in inventories are large (even using US average fractional error estimates)
- Atmospheric inverse method provides independent check

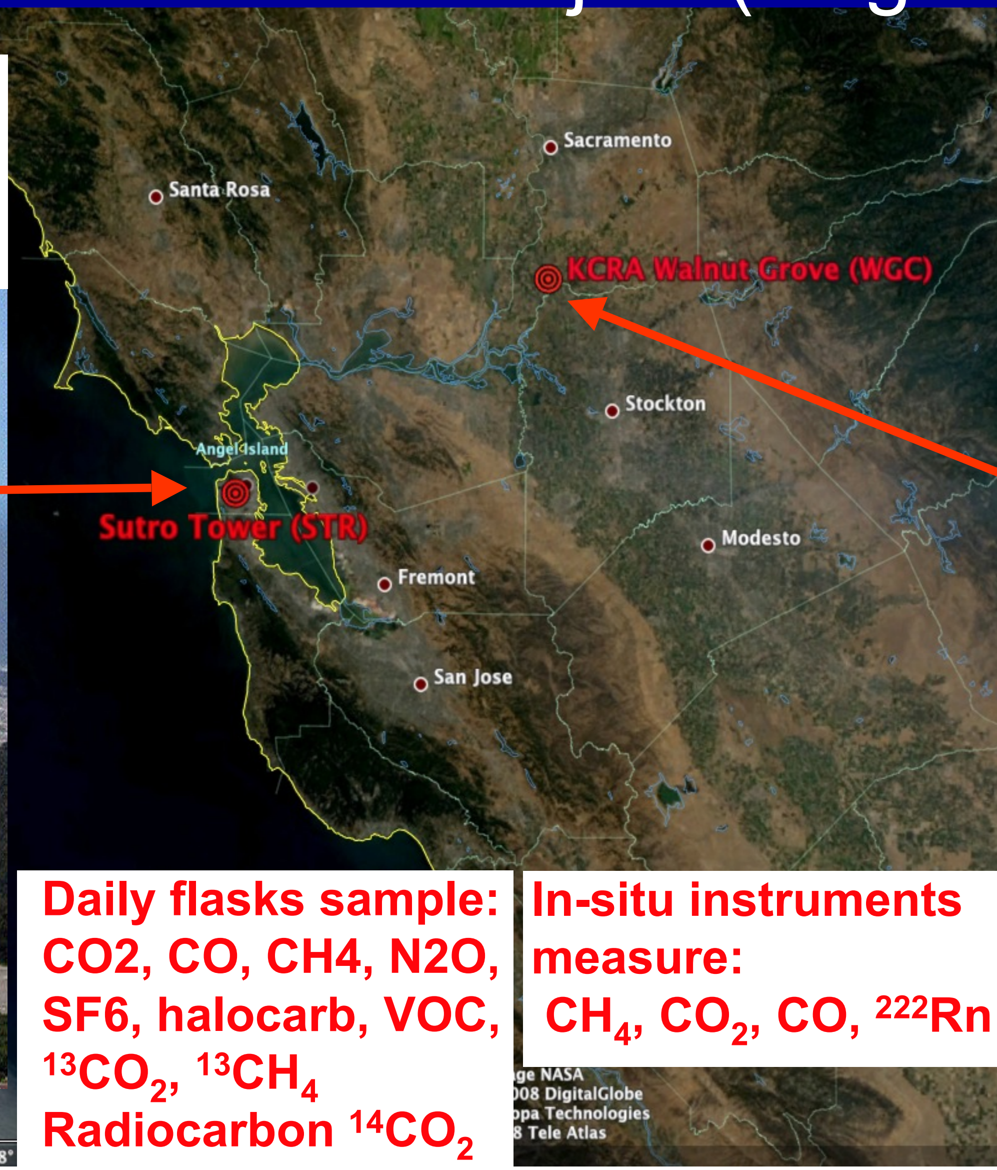
CEC, 2006 ; USEPA, 2007





Initial Work for CEC Began in 2003 California Greenhouse Gas Emissions Measurement Project (calgem.lbl.gov)

Sutro Tower (232 m agl) Oceanic + urban



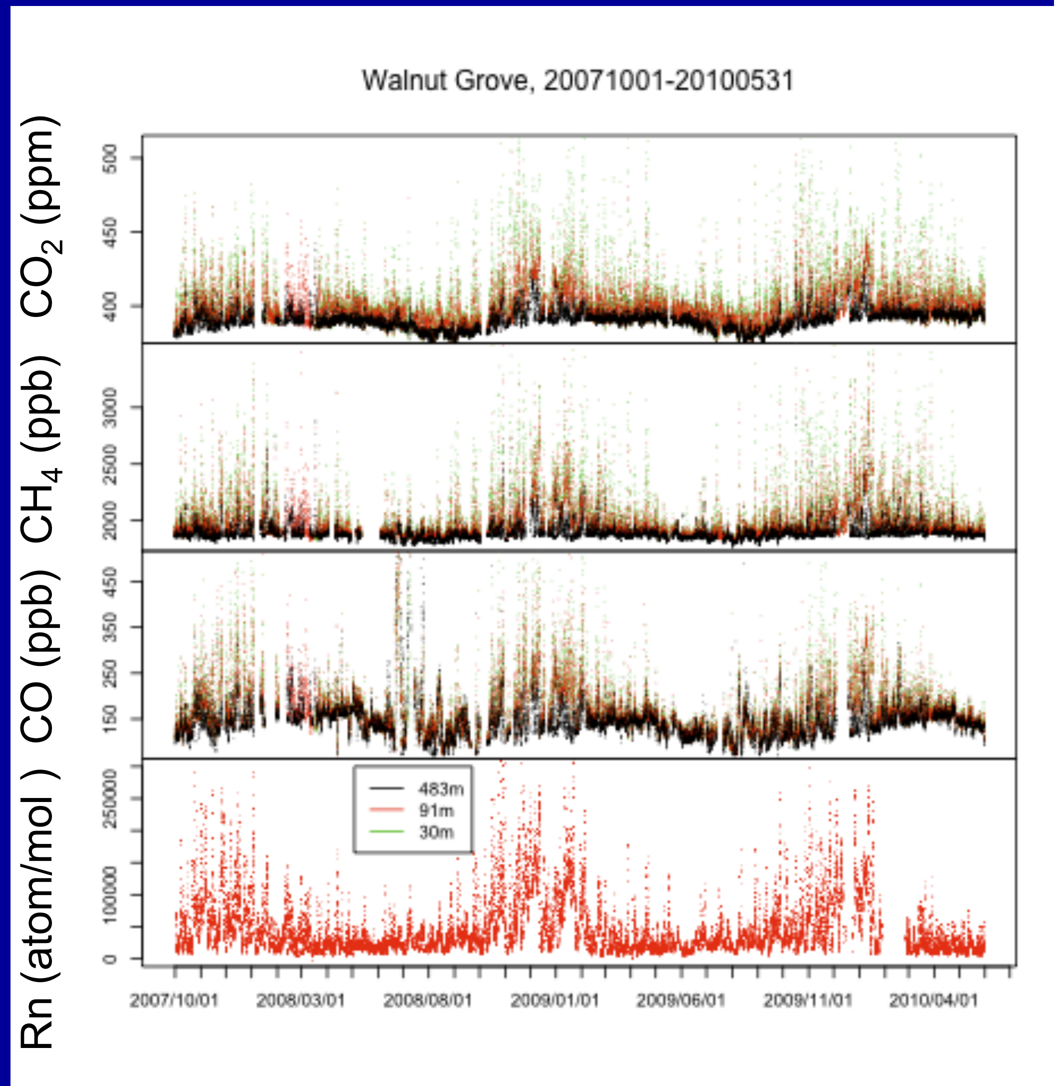
Walnut Grove (483 m agl) Central Valley + Bay Area

Daily flasks sample: CO2, CO, CH4, N2O, SF6, halocarb, VOC, $^{13}CO_2$, $^{13}CH_4$ Radiocarbon $^{14}CO_2$

In-situ instruments measure: CH_4, CO_2, CO, ^{222}Rn

In-situ Measurements at Walnut Grove

- Elevated mixing ratios above background (black) indicate strong regional-local emissions
- Synoptic variations offer opportunity to extract emissions information
- Multi-year time series will probes inter-annual variations in emissions

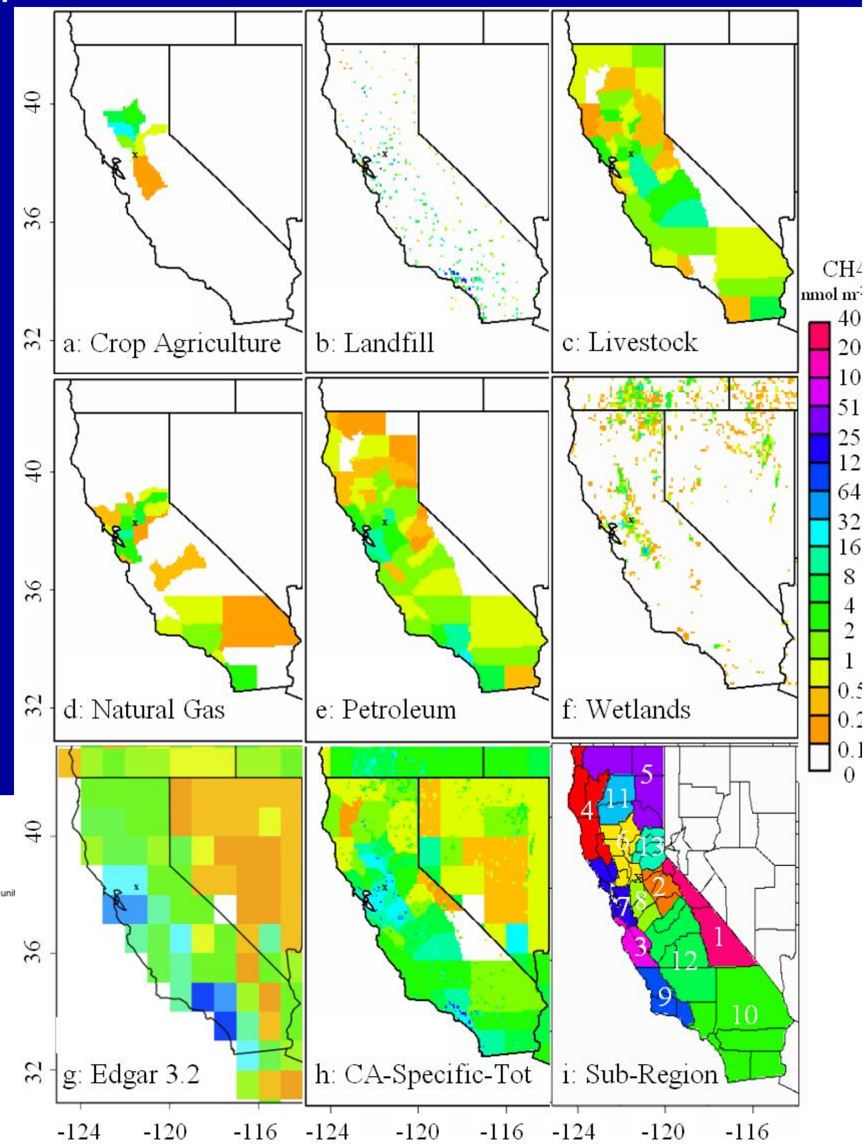


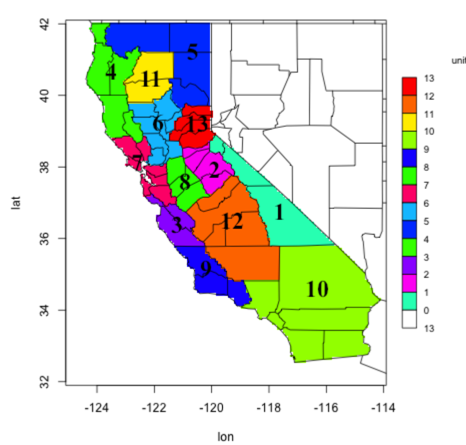
Inverse Model Estimates of Central California CH_4 and N_2O Emissions

- Emissions Inventories
  – Incorporating data-driven updates
- Meteorological Model
  – Advancing accuracy and resolution
- Estimates of CH_4 and N_2O Emissions
  – Single tower CH_4 over seasons
  – Initial work with N_2O

Early *a priori* CH_4 Emission Maps

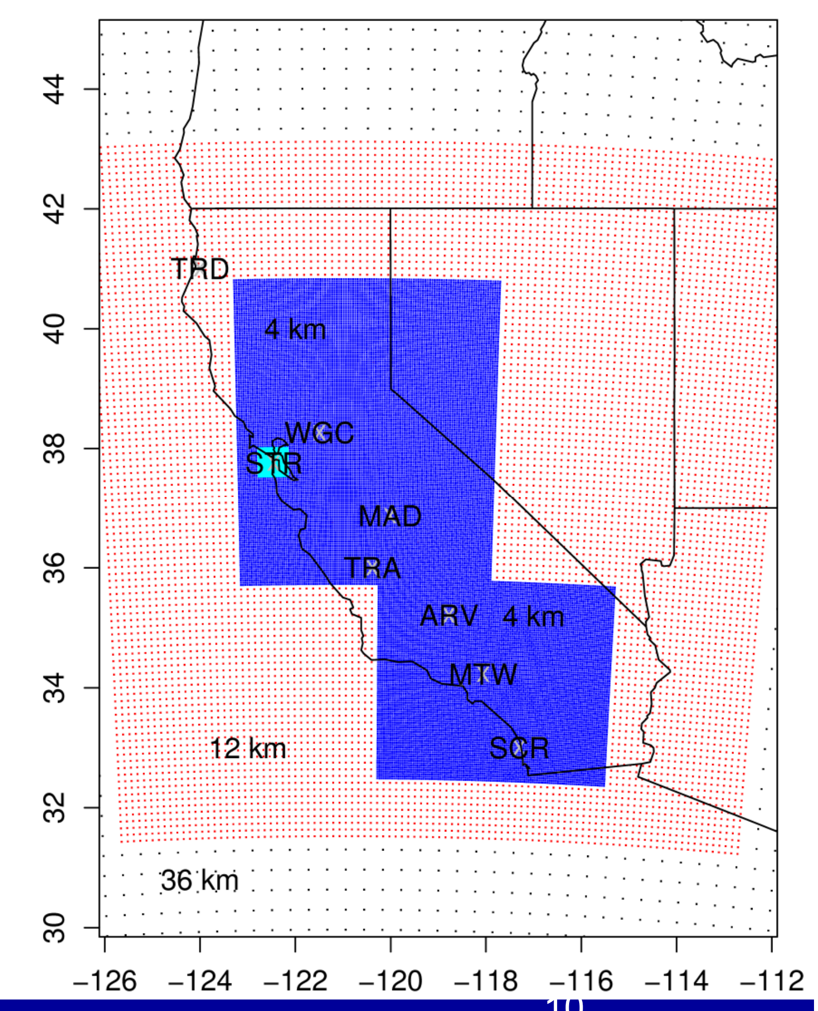
- Landfill point sources (CARB)
- Crop Agriculture (Salas et al., 2006)
- 10 km wetland fluxes (Potter et al.)
- County level livestock (USDA)
- County level natural gas dist./use
- County level petroleum refining and use (CARB VOC data)
- EDGAR3.2 (1x1degree)
  - Landfills and petroleum extraction and refining ~ 2 x CA estimates
- Regions for spatial analysis (Fischer et al., 2009)





Meteorological Model for CA Domain

- Weather Research Forecast Model (WRF)
  - Domains (extension of Zhao et al., 2009):
    - 36 km (W. US), 12 km (CA)
    - 4 km (Central Valley)
    - 1.3 km (Sutro, WGC)
  - NARR boundary forcing and internal nudging
  - Daily runs, hourly output



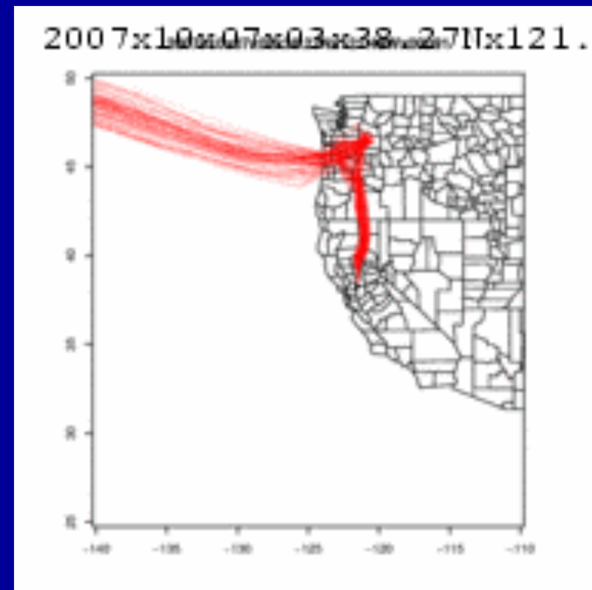
WRF-STILT Footprints for WGC Tower

- Footprint from ensemble of particle trajectories, p

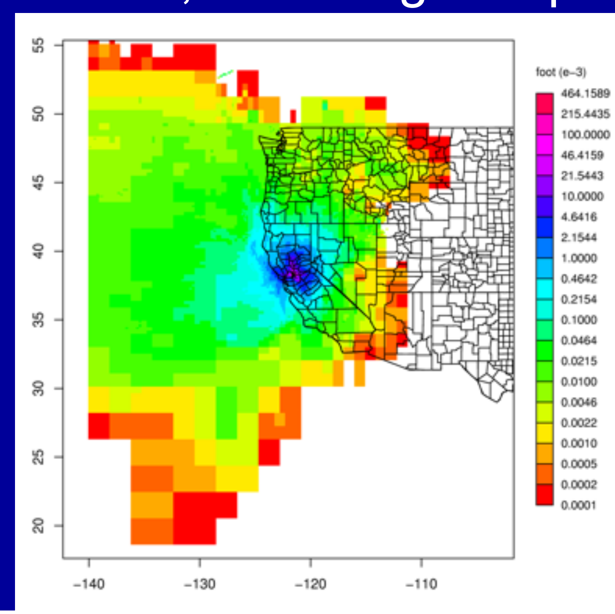
$$f(\underline{X}_r, t_r \mid x_i, y_j, t_m) \propto \sum_{p=1..N} \left(\frac{\Delta t}{Zi} \right) \Big| i, j, m, p$$

- Largest surface influences (purple) for Bay Area and Central Valley
- Predict local CH_4 signals

$$C_l(\underline{X}_r, t_r) = \sum_{i,j,m} f(\underline{X}_r, t_r \mid x_i, y_j, t_m) \cdot F(x_i, y_j)$$



Oct-Dec, 2007 avg. footprint

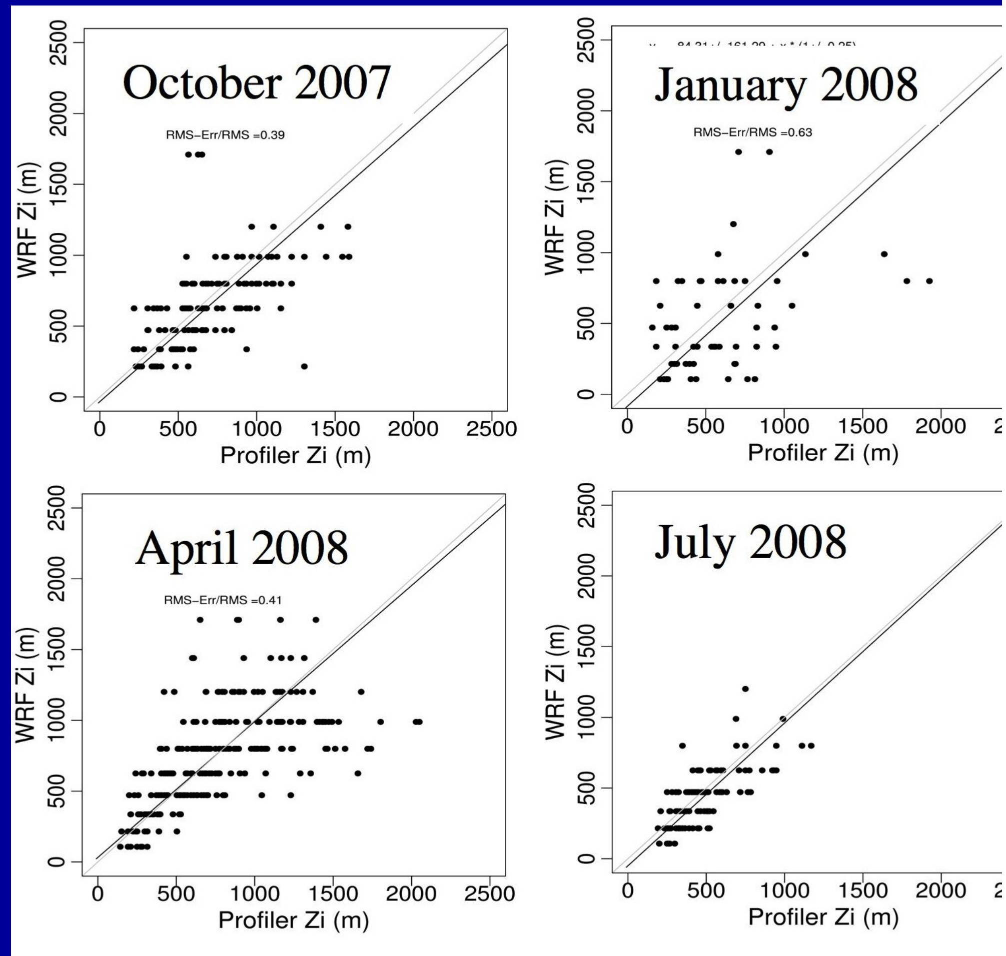


Uncertainty Estimation

- Quantify error sources
- Propagate errors through modeling system to provide quantitative uncertainties
  - Boundary layer ~ 25 %
  - Wind Velocity ~ 10%
  - GHG background ~ 15 %
  - Inventory resolution ~ 8 %
  - Other ~ 8%
- Quadrature sum ~ 32%

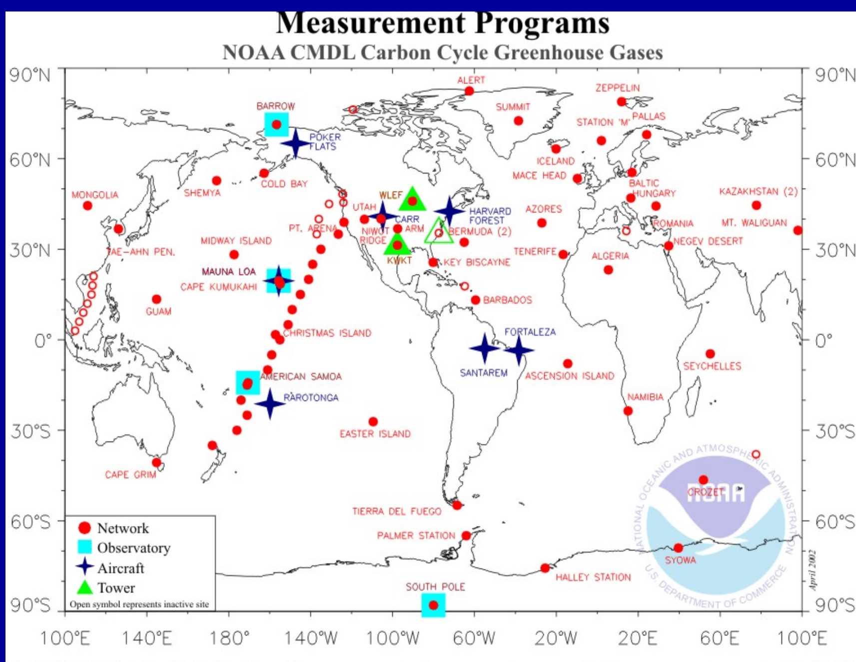
of signal for individual time points

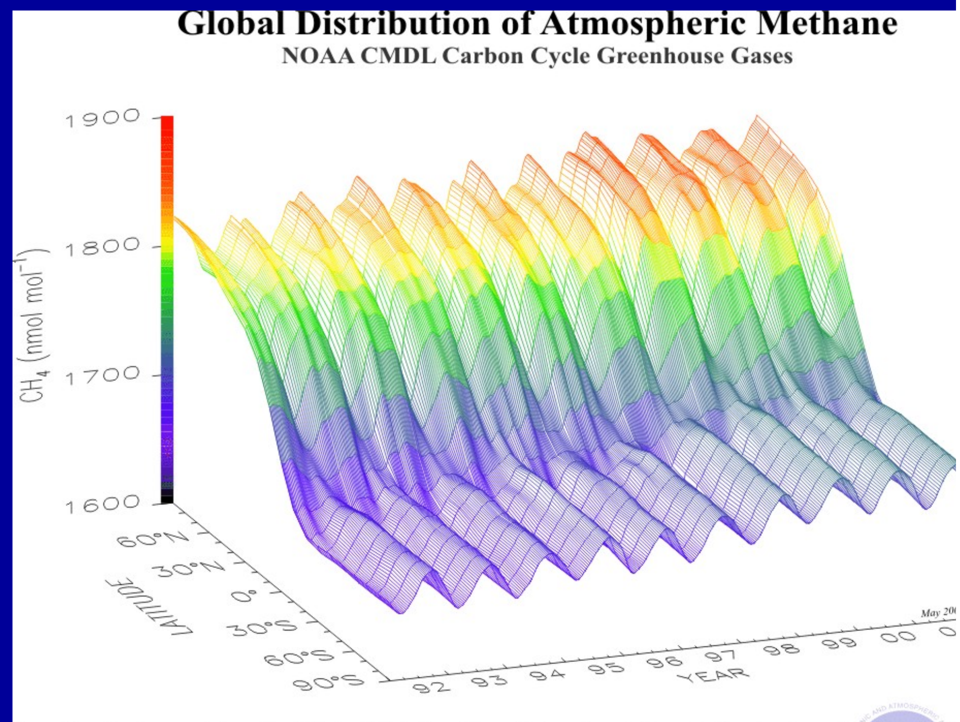
WRF-STILT versus Profiler PBL Depth



Global CH_4 Background

- Global monitoring provides California in-flow boundary condition
- CH_4 exhibits latitudinal gradient due to northern hemisphere sources



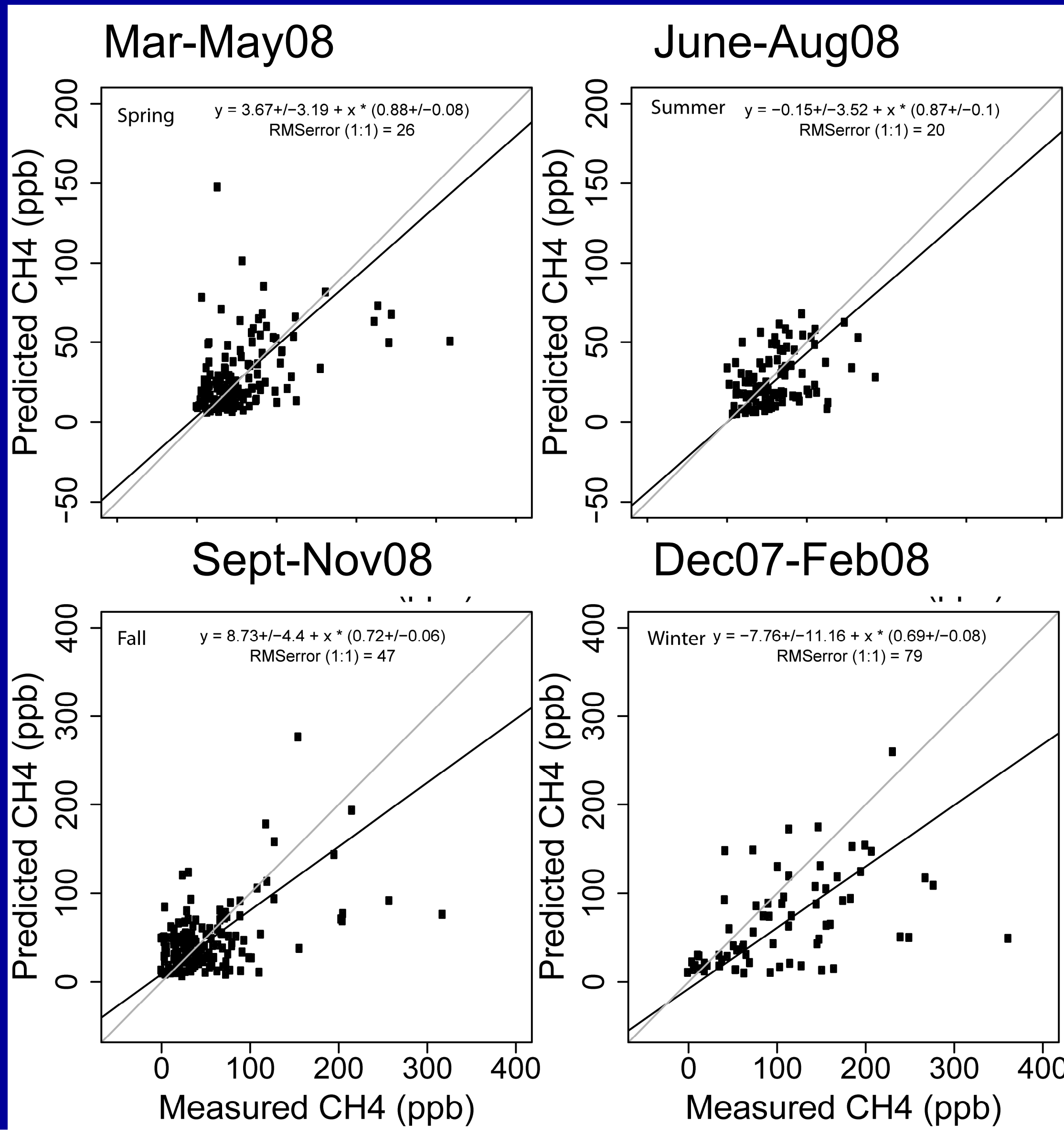


(NOAA-ESRL Global Monitoring)

Comparison of Measured and Predicted CH_4 at Walnut Grove by Season

- Scatter approximately consistent with estimated uncertainties

- CH_4 emissions appear under-estimated in CA inventory for most periods



Bayesian Linear Inverse Model

- Local signal $C_{local} = C_m - C_{bg} \sim \Sigma \lambda_i F_i f + \varepsilon$
  - Write cost function for $C_{local} = y$
  - Balance predicted-measured mismatch with error S_ε, and posterior-prior mismatch with error S_{prior}

$$\underline{J} = (\underline{\underline{y - K\lambda}})^T \underline{\underline{S}}_\varepsilon^{-1} (\underline{\underline{y - K\lambda}}) + (\lambda - \underline{\lambda}_{prior})^T S_{prior}{}^{-1} (\lambda - \underline{\lambda}_{prior})$$

where K λ =

$$\sum_{i,j,m} f(\underline{X}_r, t_r | x_i, y_j, t_m) \cdot F(x_i, y_j, t_m)$$

  - If errors random ($\varepsilon \sim N(0,\sigma)$), solution algebraic

$$\underline{\hat{\lambda}} = (\underline{\underline{K}}^T \underline{\underline{S}}_\varepsilon^{-1} \underline{\underline{K}})^{-1} (\underline{\underline{K}}^T \underline{\underline{S}}_\varepsilon^{-1} \underline{y} + \underline{\underline{S}}_{prior}^{-1} \underline{\lambda}_{prior})$$

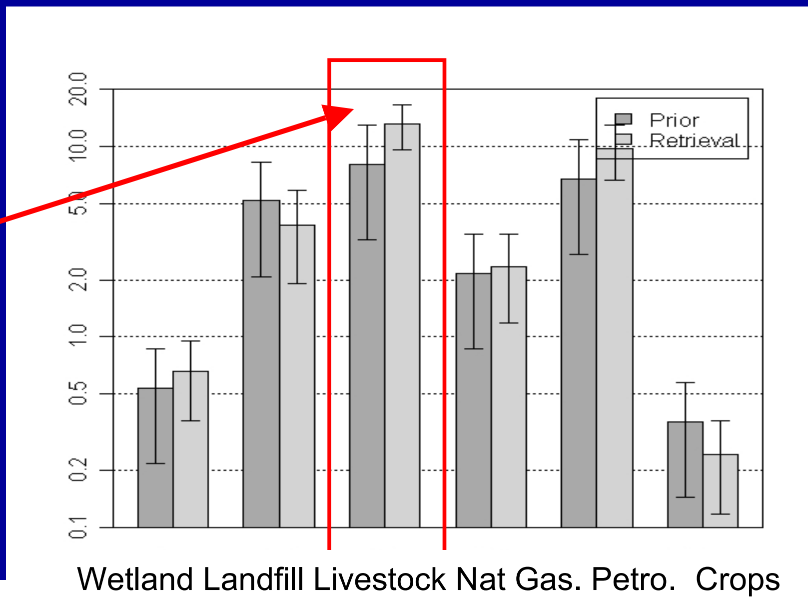
$$\underline{\underline{\hat{S}_\lambda}} = (\underline{\underline{K}}^T \underline{\underline{S}}_\varepsilon^{-1} \underline{\underline{K}} + \underline{\underline{S}}_{prior}^{-1})^{-1}$$

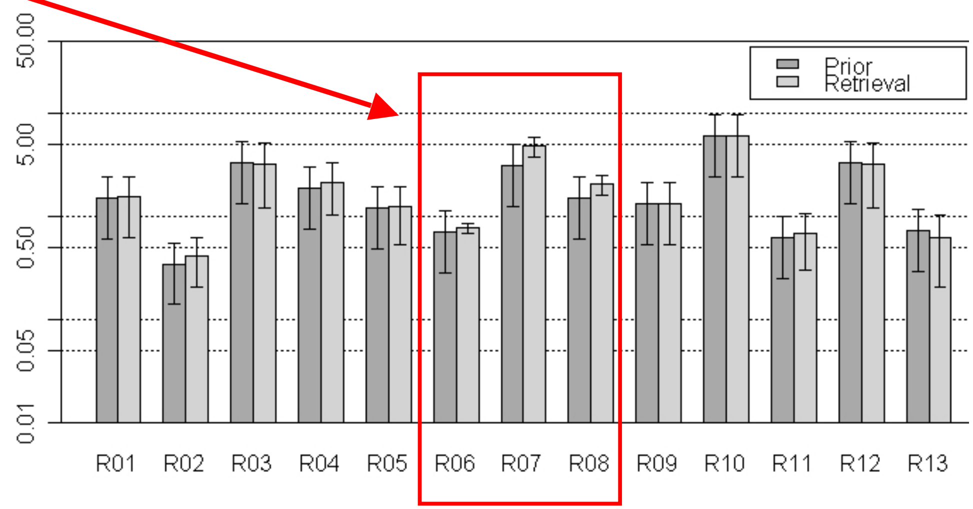
Oct-Dec07: Estimated CH_4 Emissions
(MMT CO_{2equiv} yr^{-1})

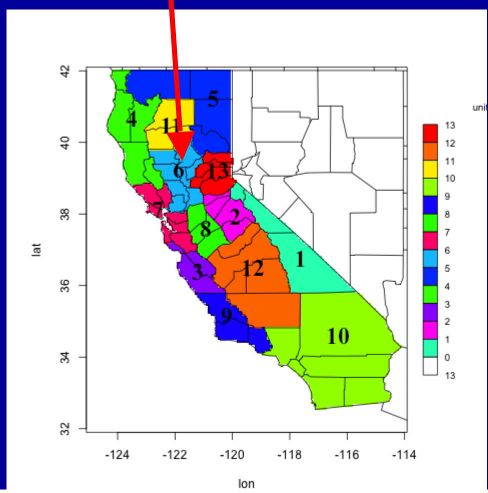
- Baysian estimate of scaling factor for each emission source or region (Zhao et al., 2009)

- Source analysis: only livestock significantly different from prior (x 1.6 ± 0.15)

- Region analysis:
  - errors reduced only for regions 6, 7, 8 near tower

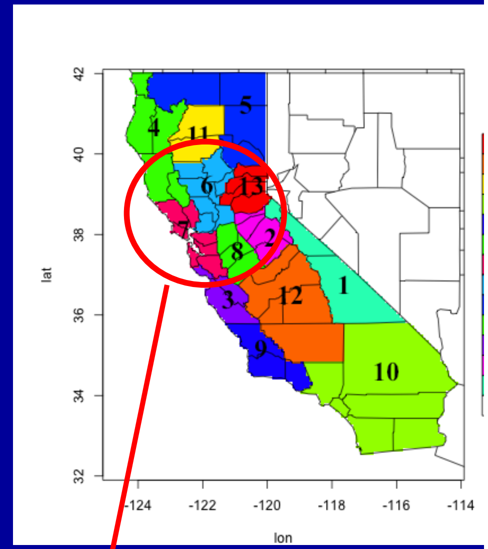


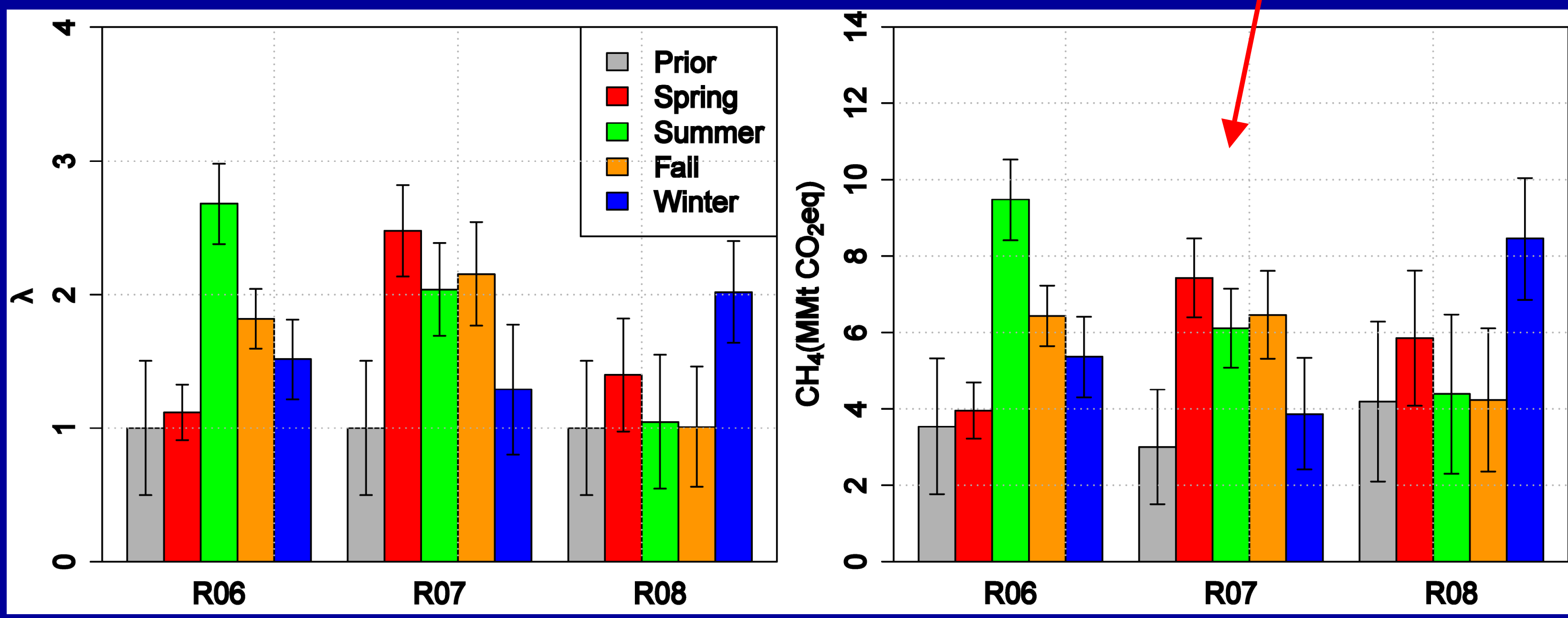




Capturing Seasonality of CH_4 Emissions

- Emissions from regions 6,7,8 show statistically significantly higher emissions in warm months
  - Seasonality of rice and wetlands in regions 6,7
  - Exception is region 8, dominated by livestock

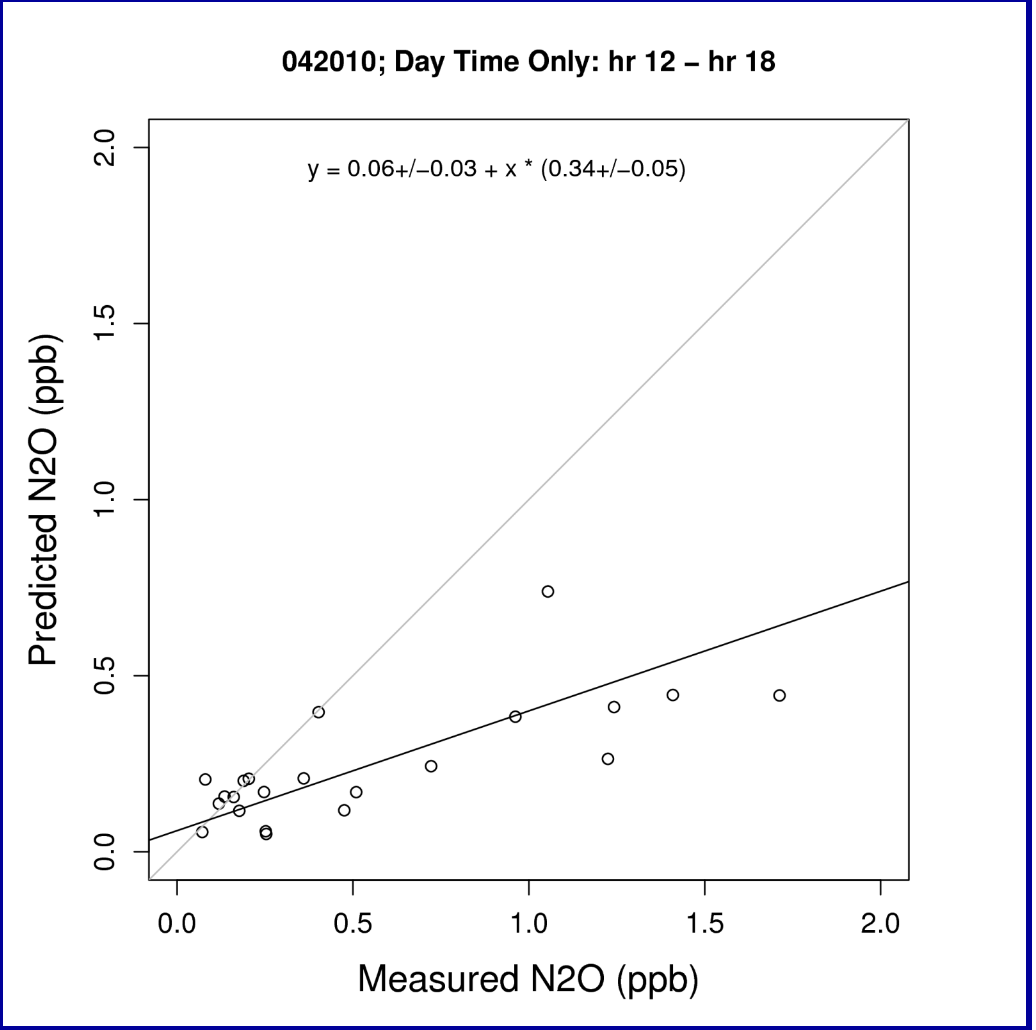




Measured and Predicted N_2O

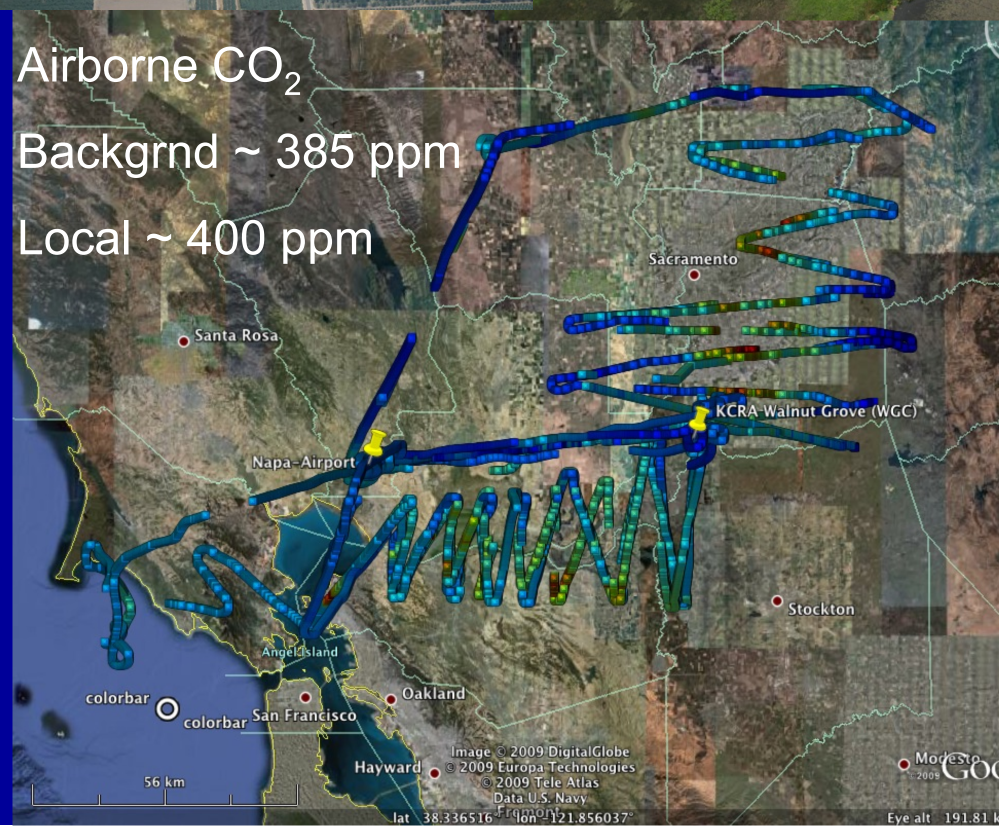
Initial continuous N_2O measurements April, 2010

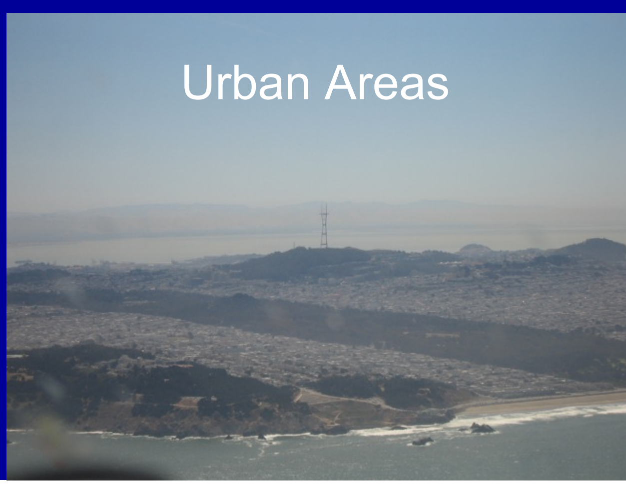
- Comparison with existing prior model (Edgar 3.2)
- Slope suggests actual emissions ~ 3 x higher than inventory
- Need to extend analysis 2007- present



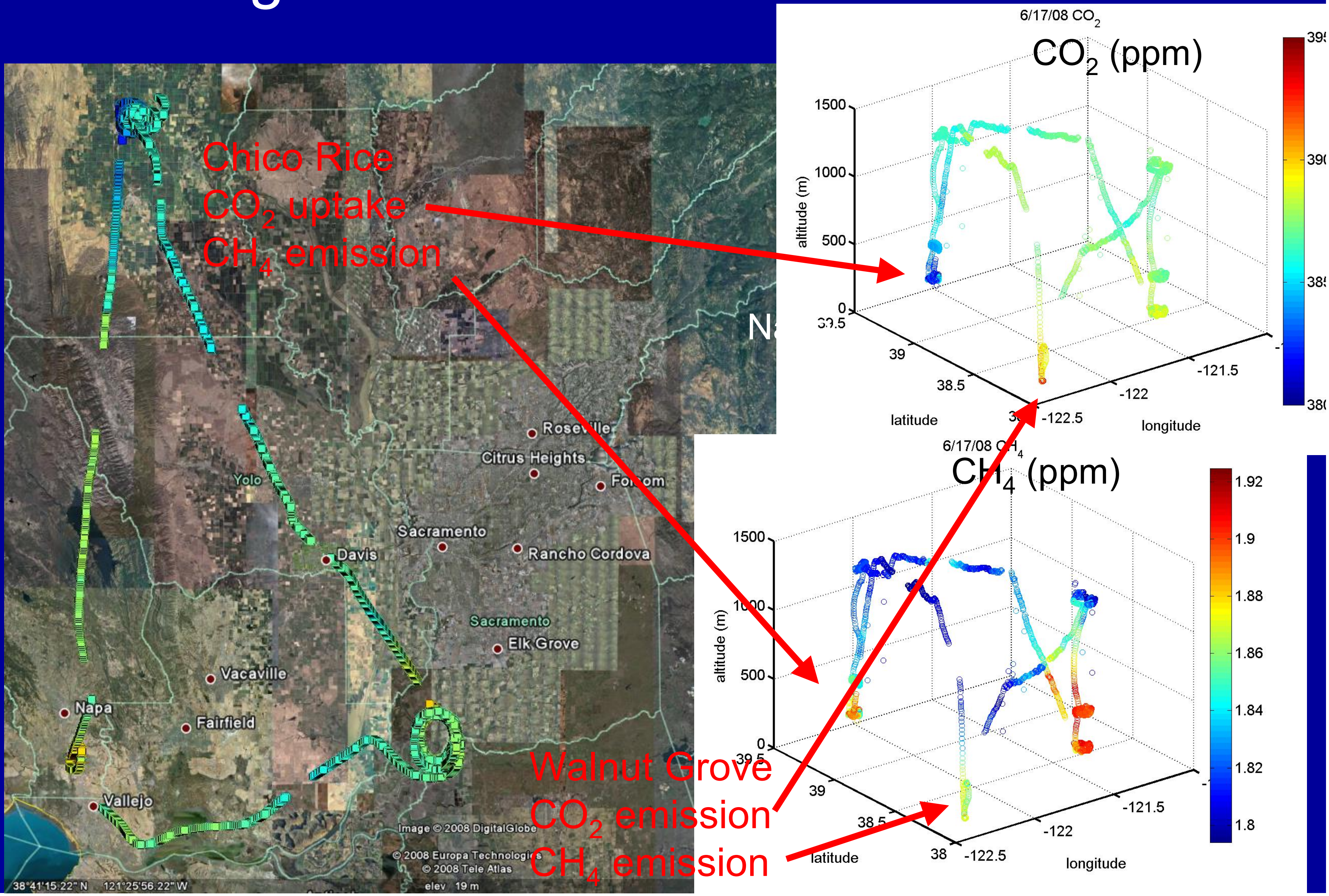
Identifying GHG Sources with Airborne Greenhouse Gas Survey (AGES)

- June, 2008 and March, 2009 campaigns from Napa CA
- Observations of Sac. river delta, Sacramento urban area, and Central Valley agriculture
- Follow-on work being planned with UCB (PI Goldstein)
- Multi-species measurements
  - Insitu CO_2, CH_4,CO
  - Flask CO_2, CH_4, CO, N2O, $^{14}CO_2$





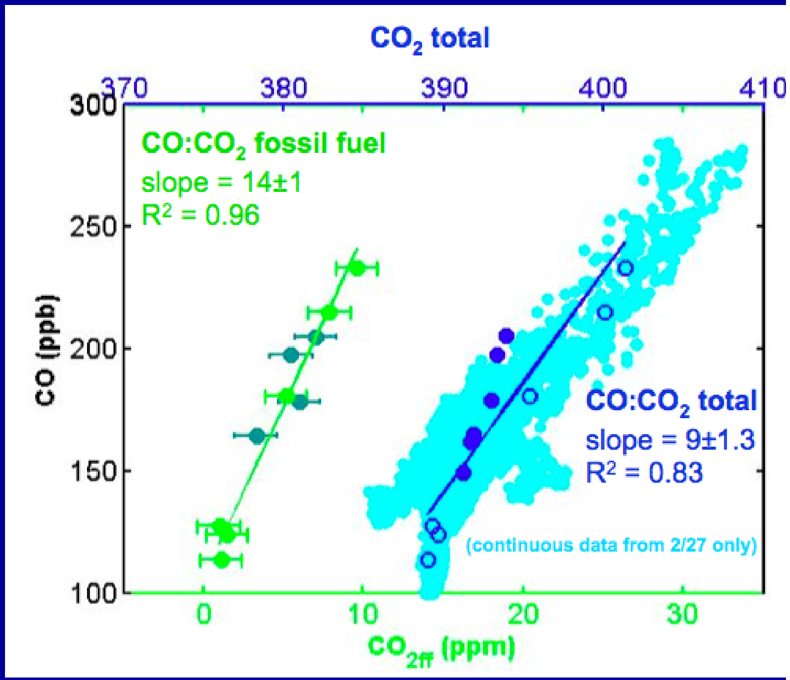
Contrasting CO_2 and CH_4 Signals over Rice and WGC



Airborne Estimate of Sacramento Fossil Fuel CO_2

- AGES flights also measured Sacramento fossil fuel CO_2 using $^{14}CO_2$, HGWPs
- Continuous fossil fuel CO_2 ($ffCO_2$) from tight correlation of flask $CO:ffCO_2$
- Slab-model estimate of Sacramento CO_{2ff} emissions consistent with CARB and Vulcan $ffCO_2$ inventories

$$Q = \int_{acrossplume} \int_{z=0}^{Z_{PBL}} u(y,z) n(z) CO_{2ff}(y,z) dy dz$$



2009 Sacramento CO_{2ff} Emissions (MtC yr

AGES	Vulcan	CARB
3 (2-6)	3.0	2.6

(Turnbull et al., 2011)

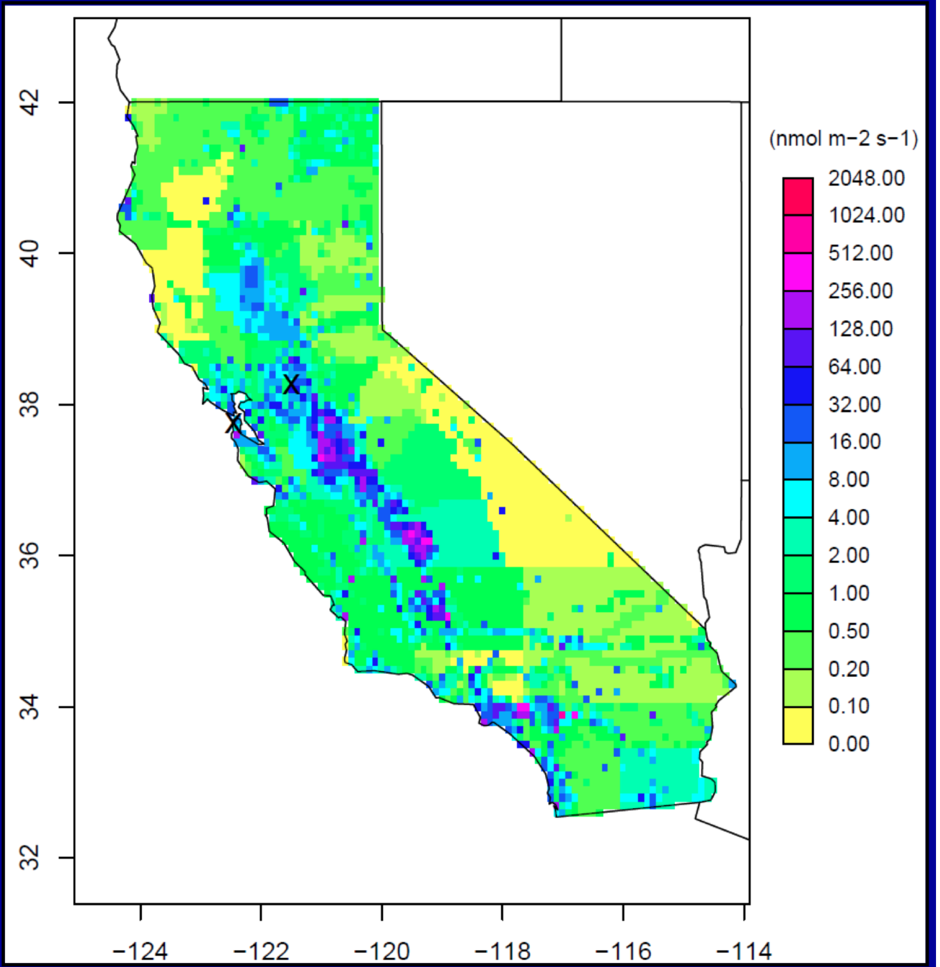
New CH_4 Inverse Modeling with CARB

- Enhanced CH_4 emission maps
- Improved meteorological model
- Initial inverse analysis of multi-site Central Valley CH_4 data

Enhanced *a priori* CH_4 Emission Maps

- Facility specific livestock counts and updated emission factors (DWR, CEC, CARB)
- GIS driven natural gas pipelines, storage, and use (CEC, CARB)
- Facility specific petroleum refining and distribution (CARB)
- Facility specific waste water treament (CARB)
- Mobil source CH_4 model (following Vulcan fossil fuel CO_2, Gurney et al., 2009)

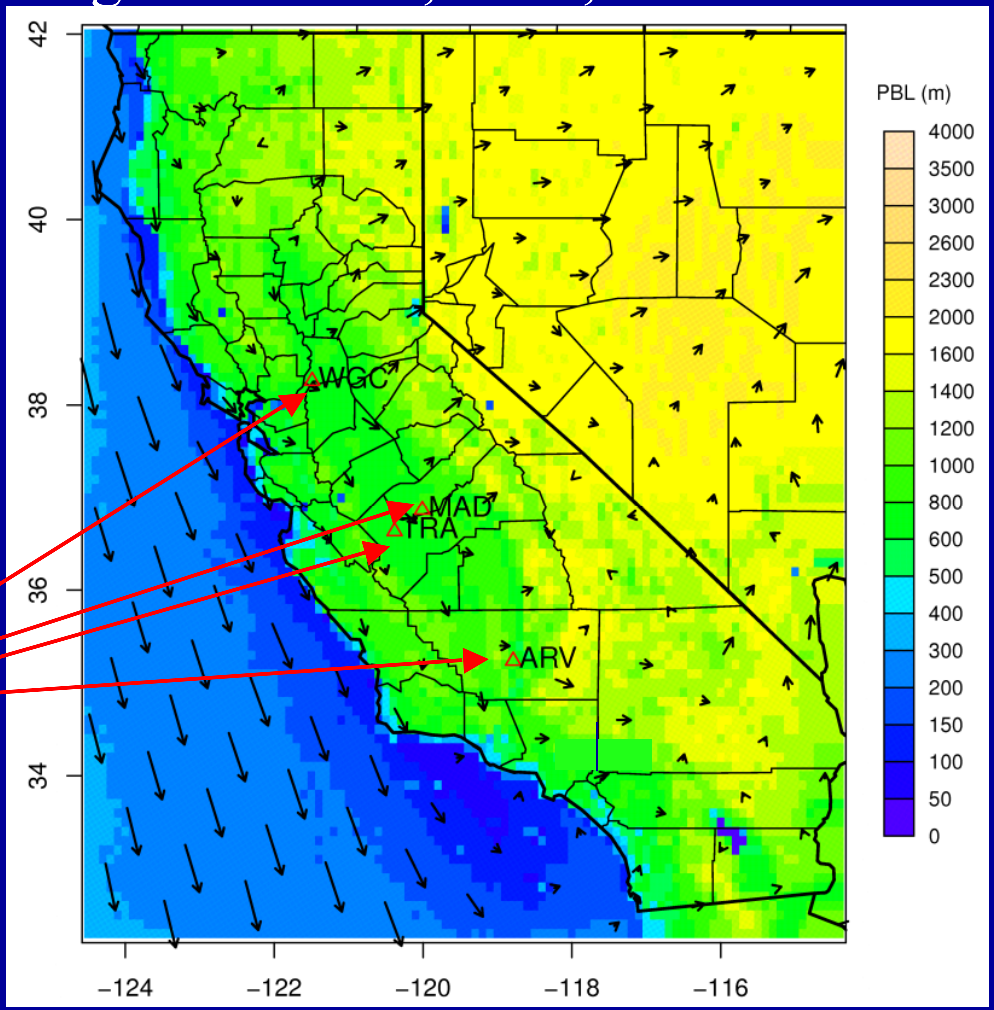
Map of California specific CH_4 emissions at 0.1 degree resolution



Improved Meteorological Model

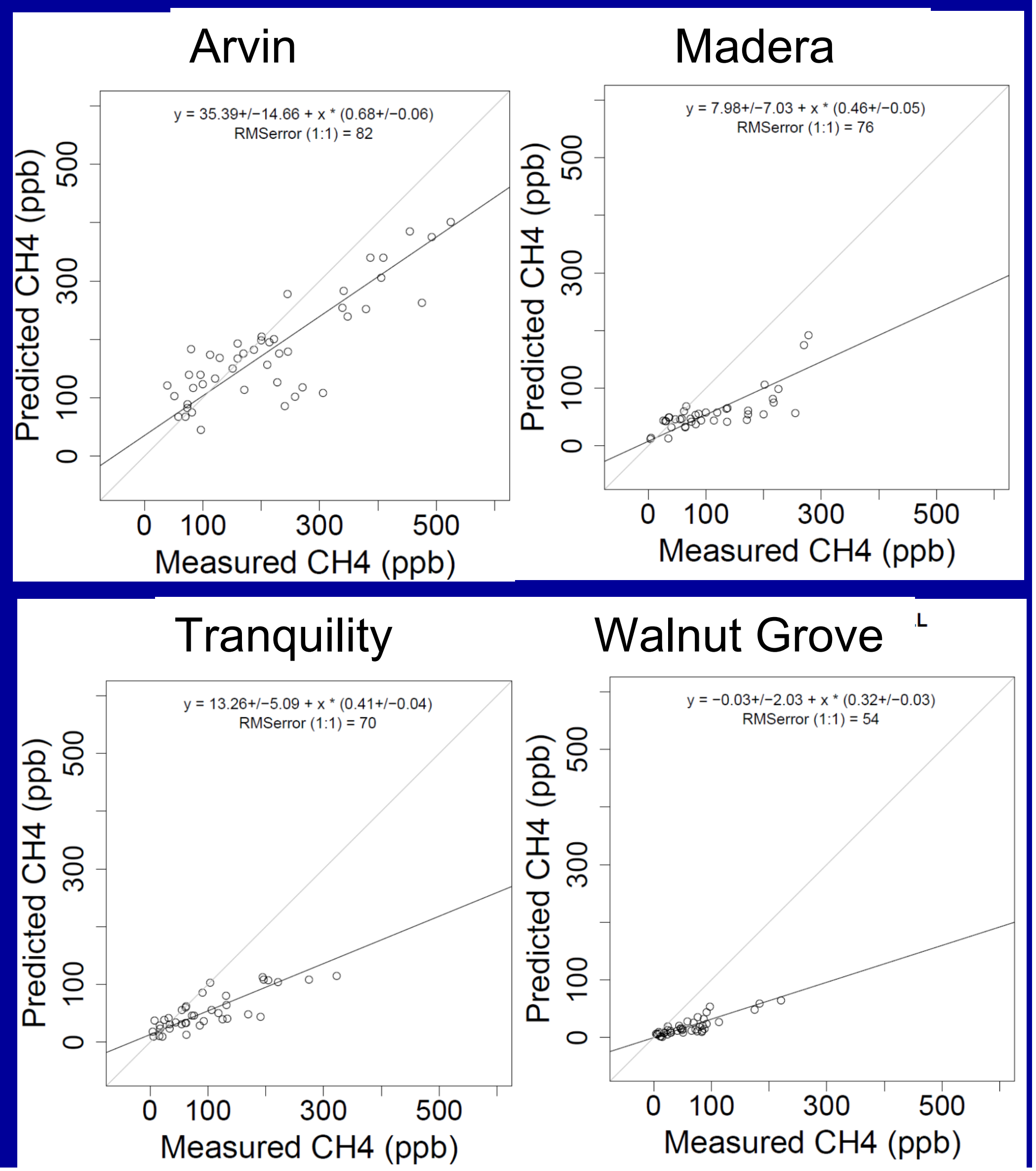
- New land surface model improves energy budget by incorporating irrigation practices
- PBL estimates are improved in native model
- Apply to CARB Central Valley CH_4 network

Predicted monthly mean winds and PBL heights for June, 2010, 1000 local.



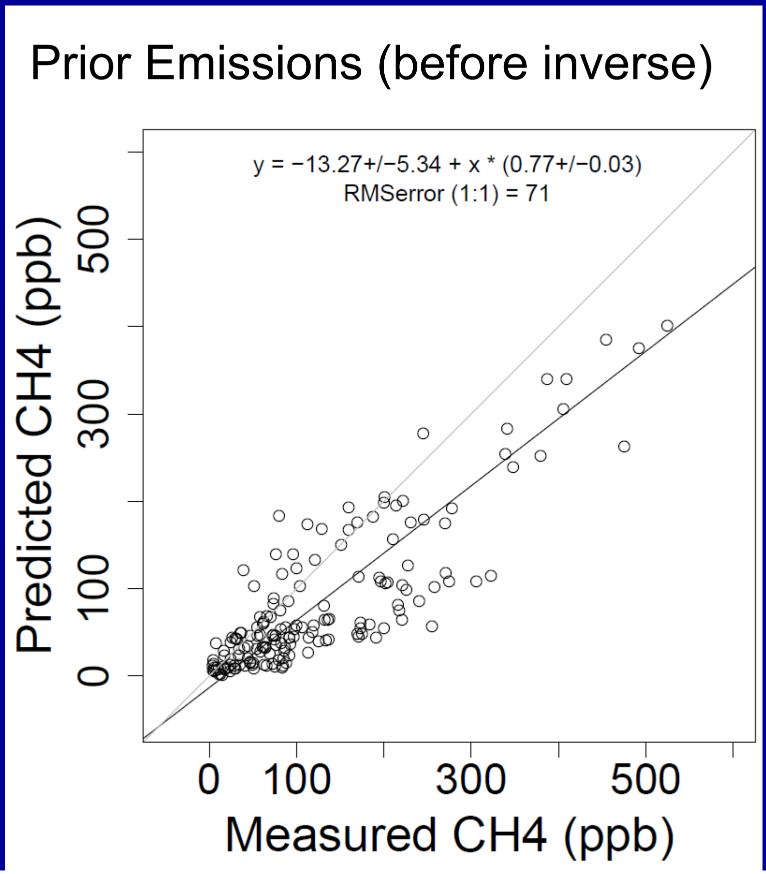
Measured and Predicted Midday CH_4 Signals for CARB CH_4 Network September, 2010

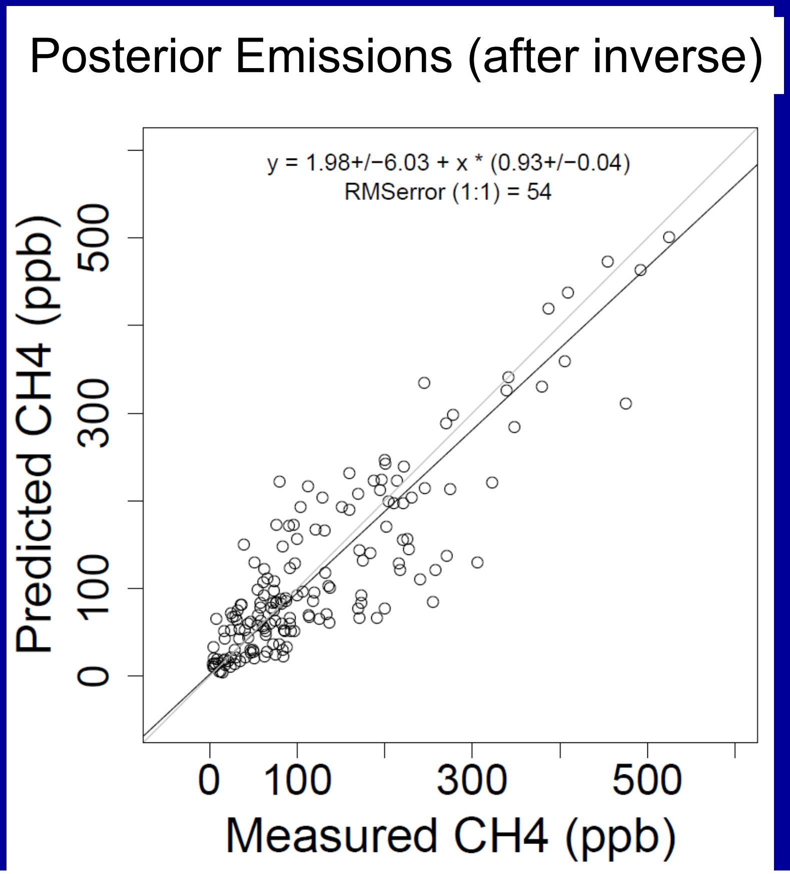
- N-S Gradient in San Joaquin Valley
- Significantly higher CH_4 signals at Arvin
- Madera and Trinity sites comparable
- Lower signals at Walnut Grove



Prior and Optimized CH_4 Signals for Sept., 2110

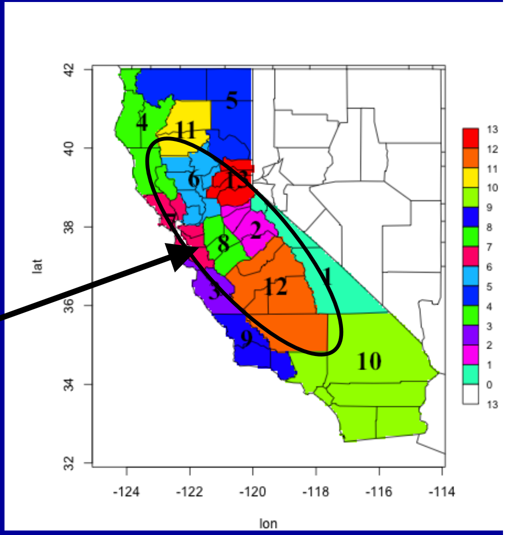
- Composite data from all sites
- Scale prior emissions in 13 regions to obtain best posterior match

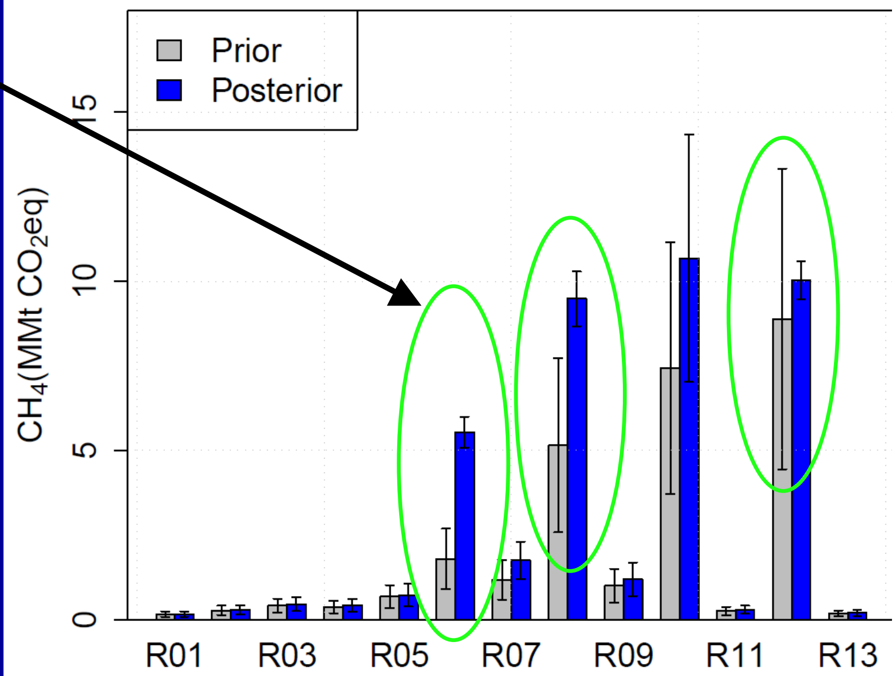




Initial Results from CARB CH_4 Network

- CARB sites significantly reduce uncertainties for key regions
  - Capture most of Central and South San Joaquin valley
- Future towers planned:
  - Capture North Valley (rice agriculture)
  - Need for sampling in SoCal Air Basin





Conclusions

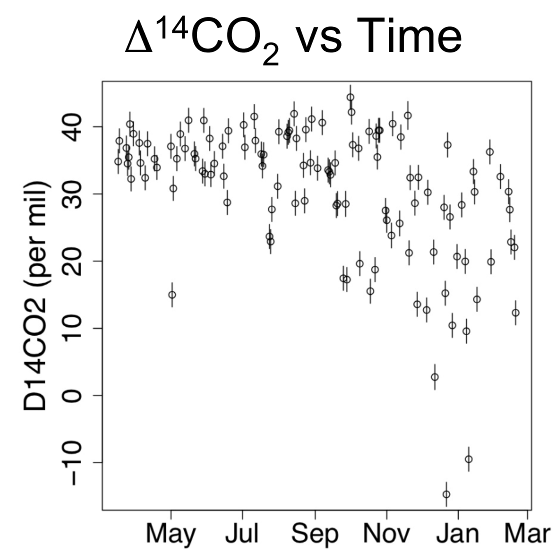
- Atmospheric measurements and inverse modeling provide a powerful independent constraint for emissions inventory validation
- Careful attention to uncertainties essential for quantitative emission inventory assessment
- Network of towers are effective in capturing regional CH_4 emissions from much of California's Central Valley
  - CH_4 emissions are modestly underestimated
  - N_2O emissions significantly underestimated

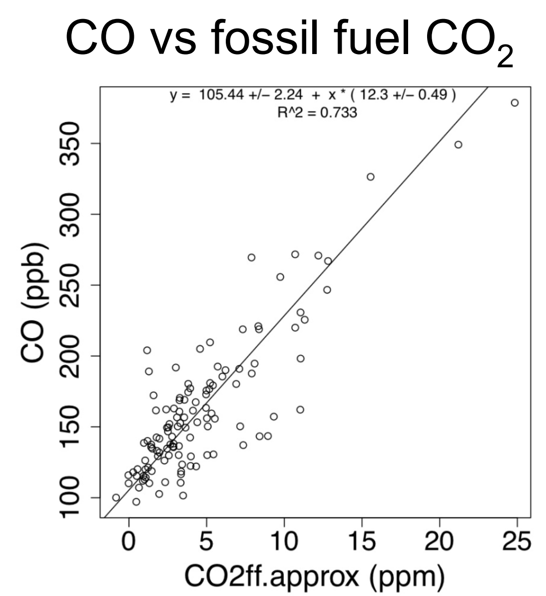
Suggested Research to Address CARB needs for emission inventory validation:

- Extend measurement records and capabilities at existing sites
  - Multi-species approach provides source attribution
- Improve inverse modeling framework
  - Hierarchical Bayesian models to identify unexpected emissions
  - Assimilation of network meteorology to reduce transport uncertainty
- New key sites and measurements for CARB network
  - Close gaps in spatial coverage to cover important regions

Extending capabilities at key sites
Radiocarbon $^{14}CO_2$ at Walnut Grove

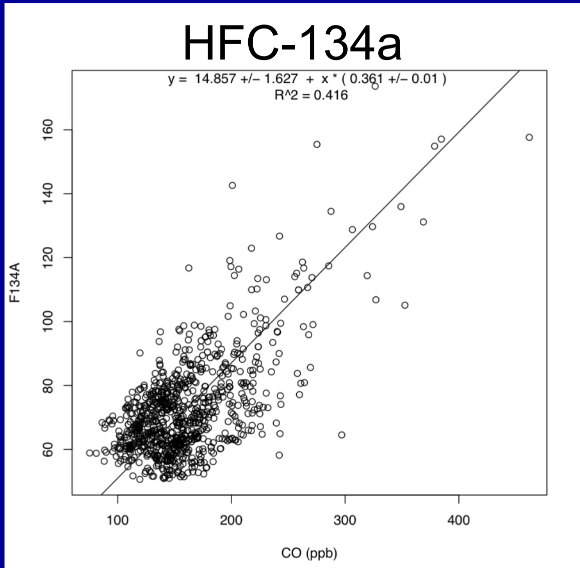
- Daily flask sampling
- 2-3 weekly sub-samples for $^{14}CO_2$ 2009 - present
- First year analysis shows large depletion in winter, likely due to decreased mixing
- Excellent correlation of CO to fossil fuel CO2
  - Use CO as continuous proxy for fossil CO_2
  - Need additional tracers of biomass combustion

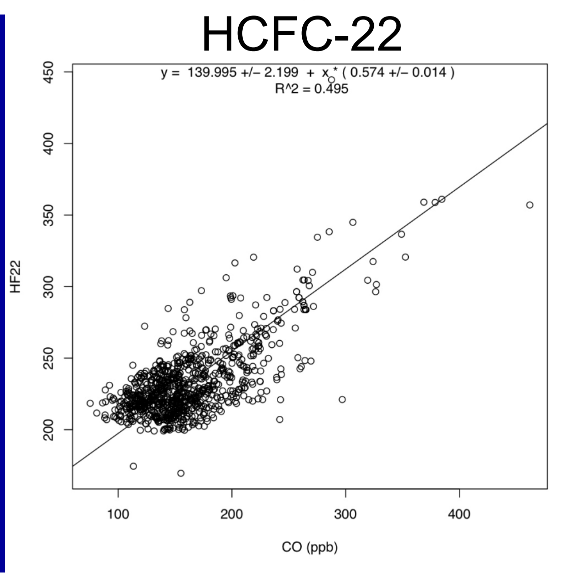




Extending Key Measurements: High Global Warming Potential Measurements at Walnut Grove

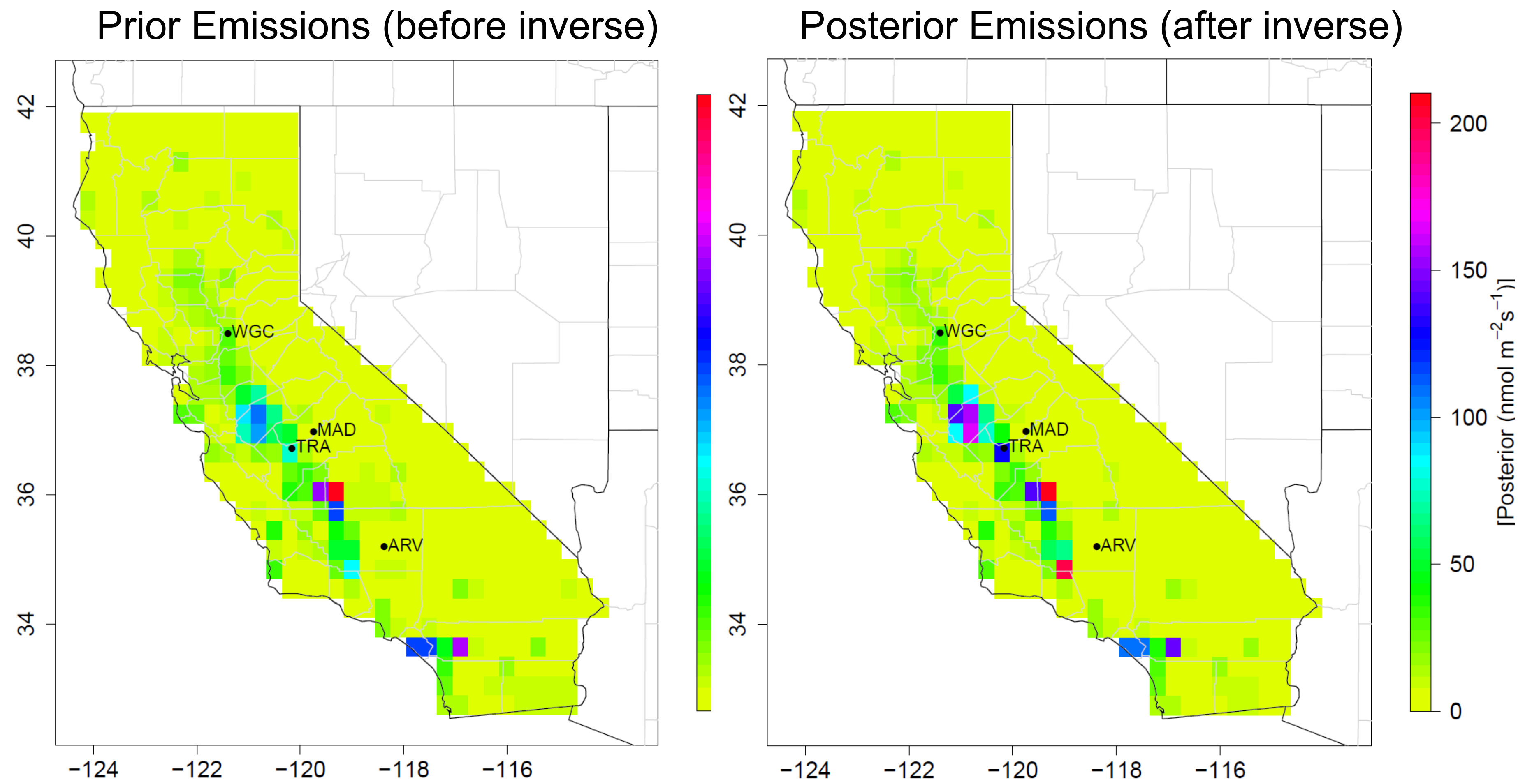
- All major warming gases accurately measured
- Inter-annual trends due to global background
- Synoptic variations reveal regional sources
- High correlation of some HWGP to CO or fossil fuel CO_2 may allow emission factor analysis of emissions (e.g., Hsu et al., 2009)
- Other gases may yield inverse model estimates of HWGP emissions





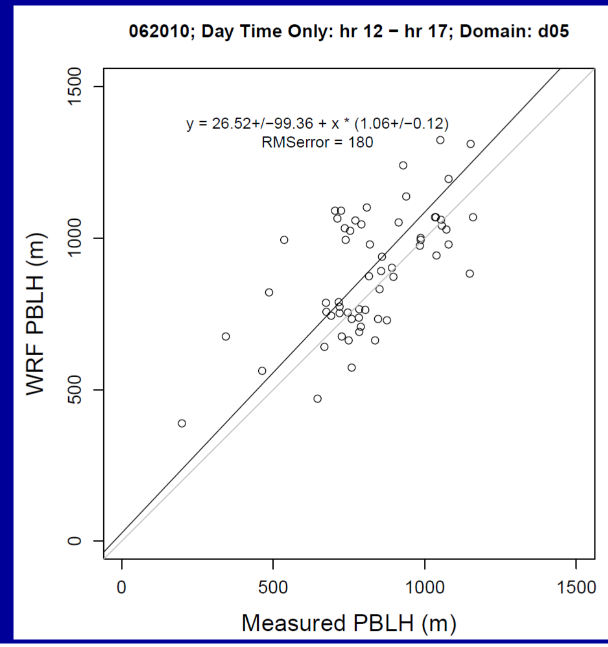
Improving Inverse Model Framework: Hybrid Model Maps Prior and Pixel-Based CH_4

- Identifies spatial features of CH4 emissions beyond prior
- Requires larger data sets (e.g., aircraft, satellite)

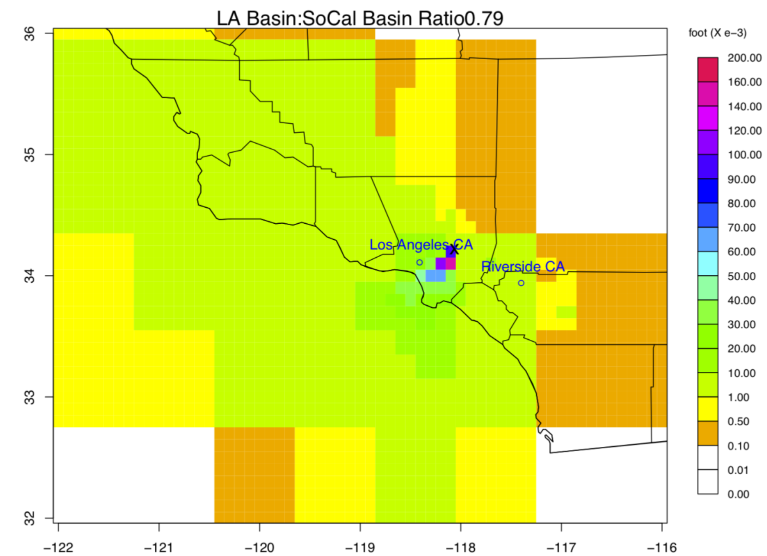


Expand CARB Network to South Coast

- Caltech/U-Houston Lidar measurements confirm accurate daytime PBL in WRF model



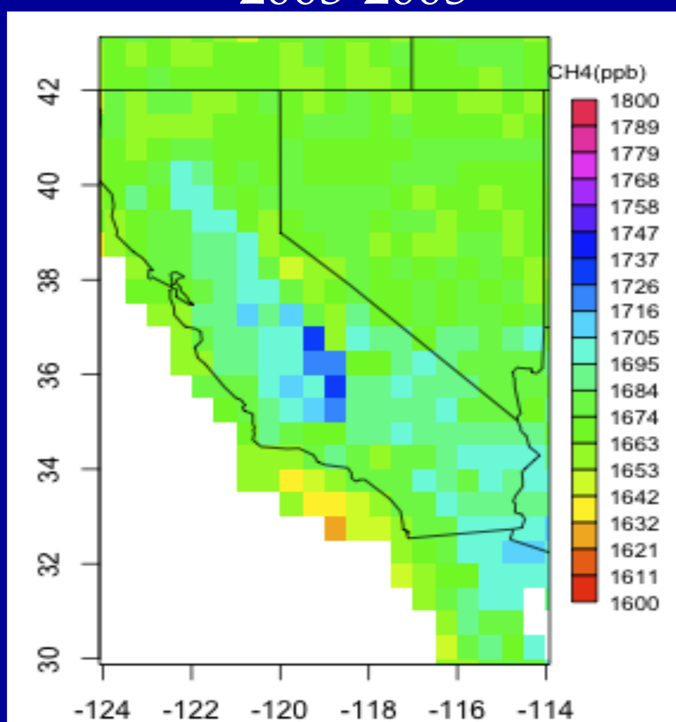
- Footprint model shows 80% of SoCal influence at Mt Wilson limited to Los Angeles County
- Proposed site near UC Riverside (San Bernadino) will provide coverage of larger area containing CH_4 source



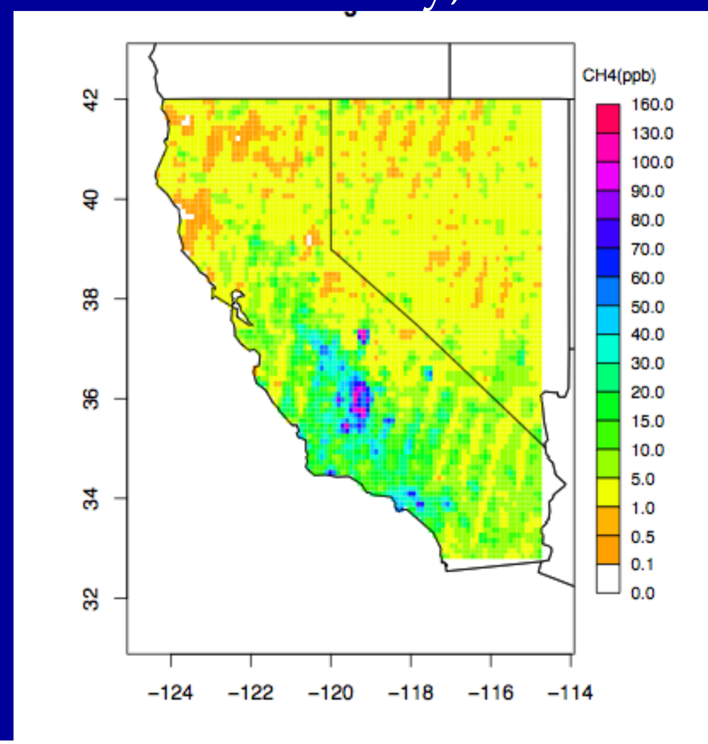
Including Key Measurements: Initial Comparison of SCIAMACHY Column Mean CH_4 Retrievals and Prior Model

- Satellite tracks cover CA require long integrations
- Clear spatial correlation of measurement and prediction

SCIAMACHY CH_4
2003-2005



Predicted Column CH_4
March-May, 2005




Thank You

Fossil Fuel CO_2 Observed with Radiocarbon

Measure $\Delta^{14}CO_2$ & $^{12}CO_2$ in local and background air

$$\Delta^{14}C = [(^{14}C/^{12}C)_{air}/(^{14}C/^{12}C)_{std} - 1] * 1000 ‰$$

Δ_{ff} = -1000 ‰ (fossil is radiocarbon free), Δ_{bck} & Δ_{eco} ~ 60 ‰

Apply mixing model to determine fossil fuel CO_2, C_{ff}

1) $C_{obs} = C_{bck} + C_{ff} + C_{eco}$ --- total carbon, and

2) $\Delta_{obs} C_{obs} = \Delta_{bck} C_{bck} + \Delta_{ff} C_{ff} + \Delta_{eco} C_{eco}$ --- radiocarbon

⇒Accurate radiocarbon measurements allow accurate estimate of fossil fuel CO_2

$\Delta14C \sim 2.8$ ‰ => C_{ff} = 1 ppm