Top-down estimate of surface flux in the Los Angeles Basin using a mesoscale inverse modeling technique: assessing anthropogenic emissions of CO,NOx and CO2 and their impacts.

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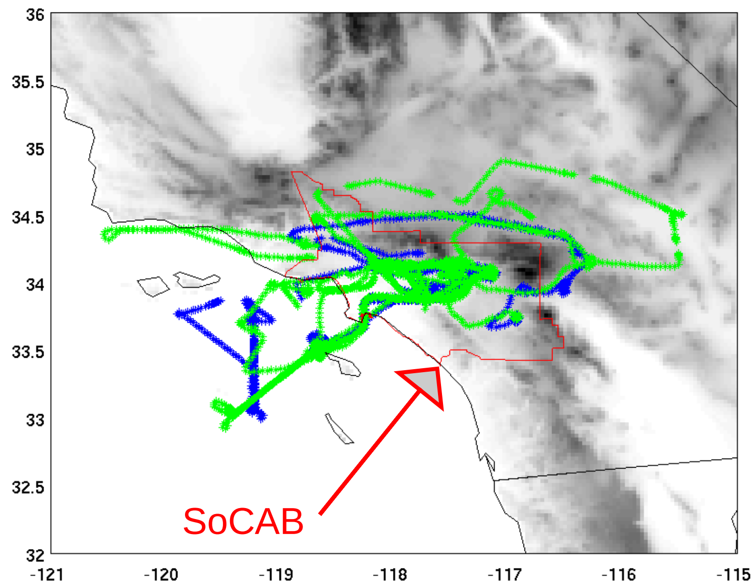
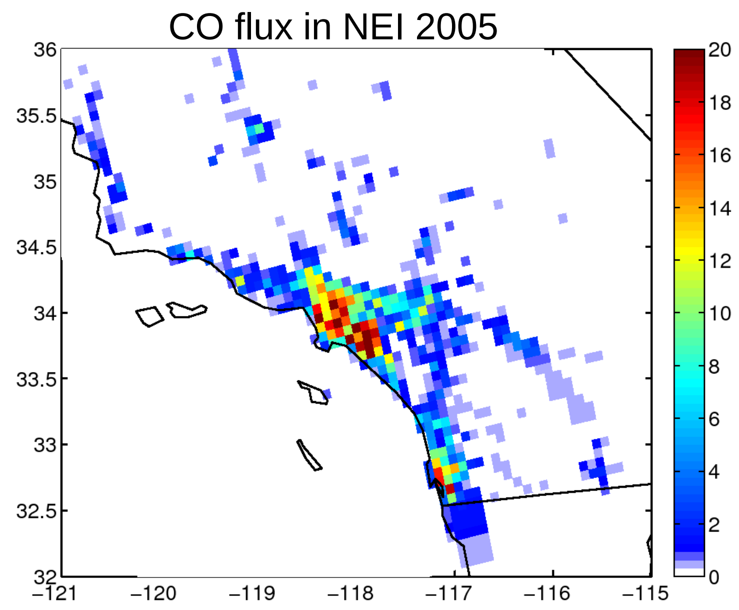
For further details, see J. Brioude et al. (2013, ACP)

Outline

1. Observations and inversion method
2. Differences in CO,NOy and CO2 between the posterior and existing inventories (NEI, CARB)
3. Trend between 2002 and 2010
4. Application: Ozone Chemistry in the LA basin with WRF-Chem

CALNEX 2010 and ITCT 2002





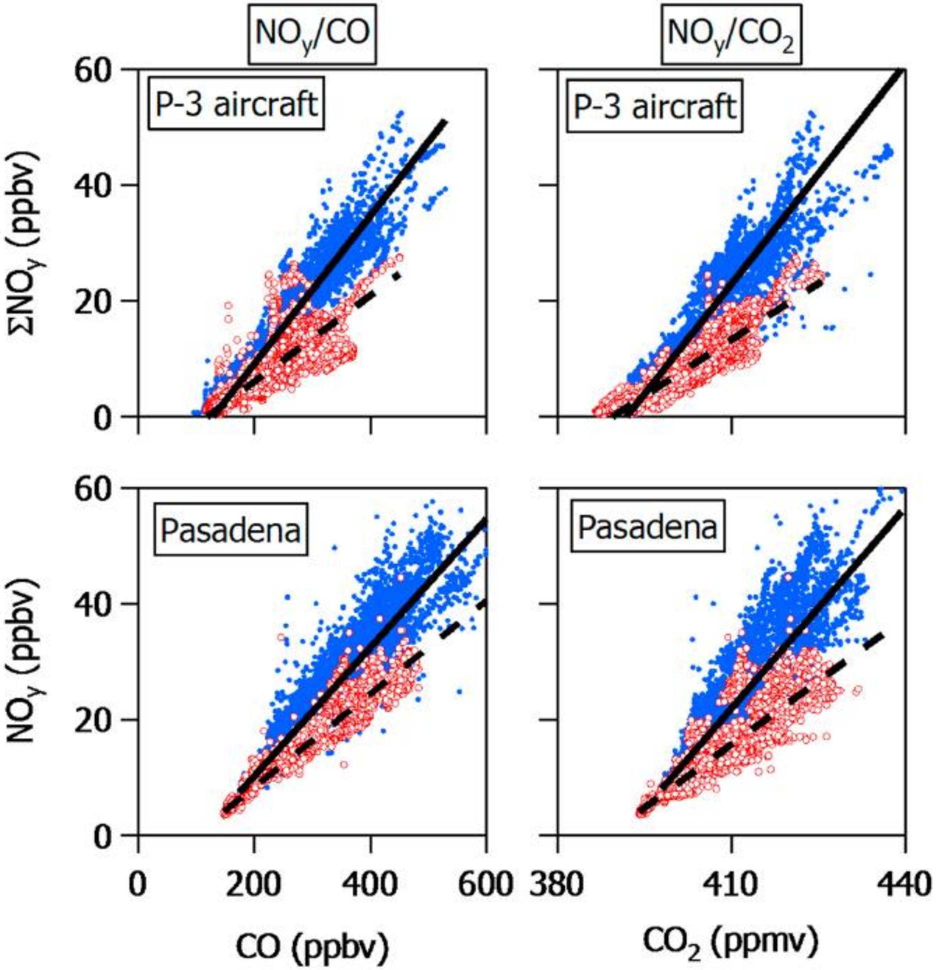
- 6 flights (3 weekday flights in blue, 3 weekday flights in green) during CALNEX 2010 are used to evaluate LA basin anthropogenic emissions
- 1 flight during ITCT 2002 is used to evaluate the emissions in 2002

- CO,NOy and CO2 tracers in FLEXPART are assumed passive.

=> Variability of single-flight-based inversion is about 10 to 15%, depending on the chemical species used

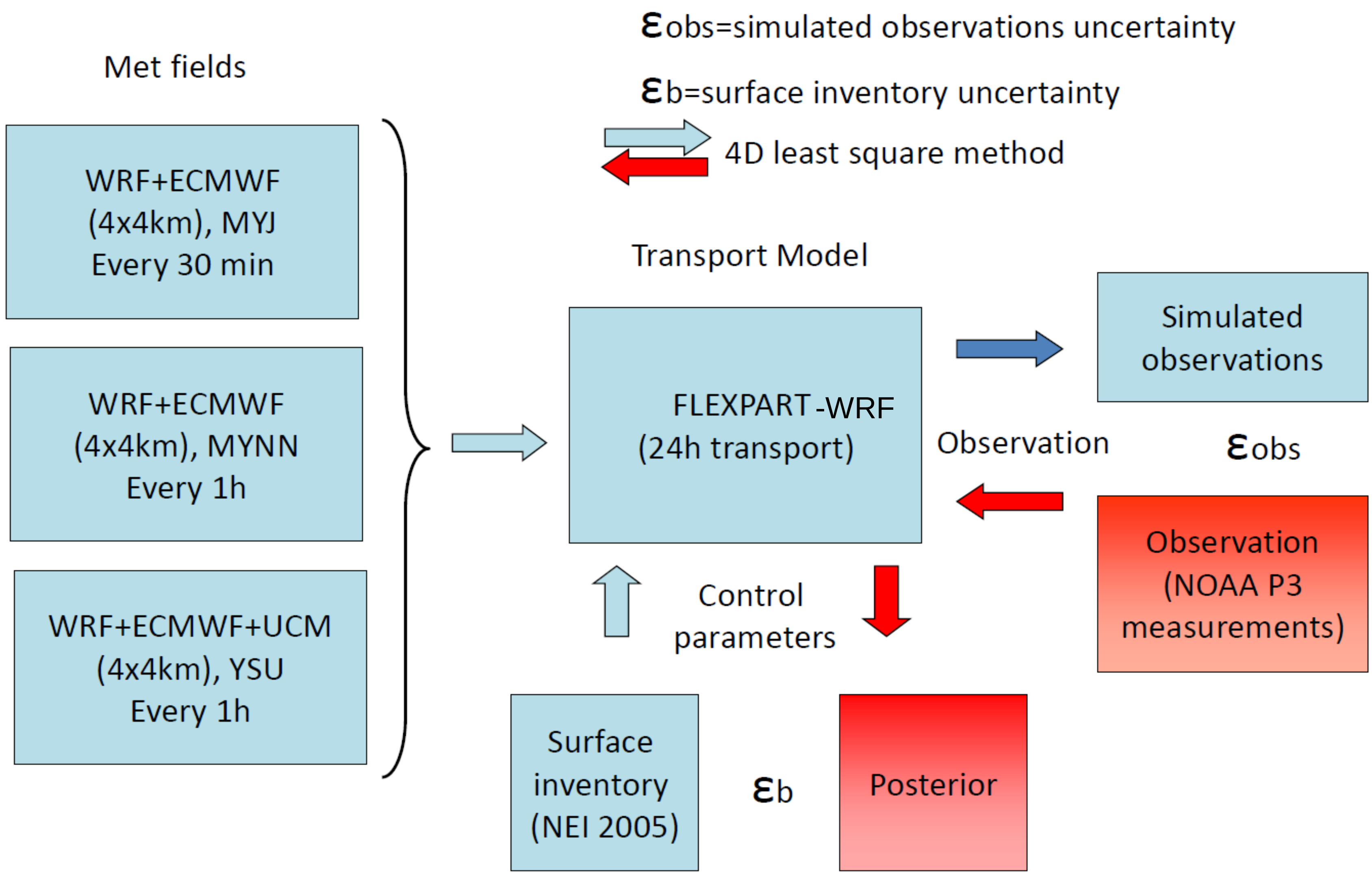
Weekday/weekend effects (at least in the US)

- Large reductions in NOx emissions on weekends relative to weekdays result in higher weekend ozone production efficiencies (Pollack et al., 2012). Due to differences in truck traffic.



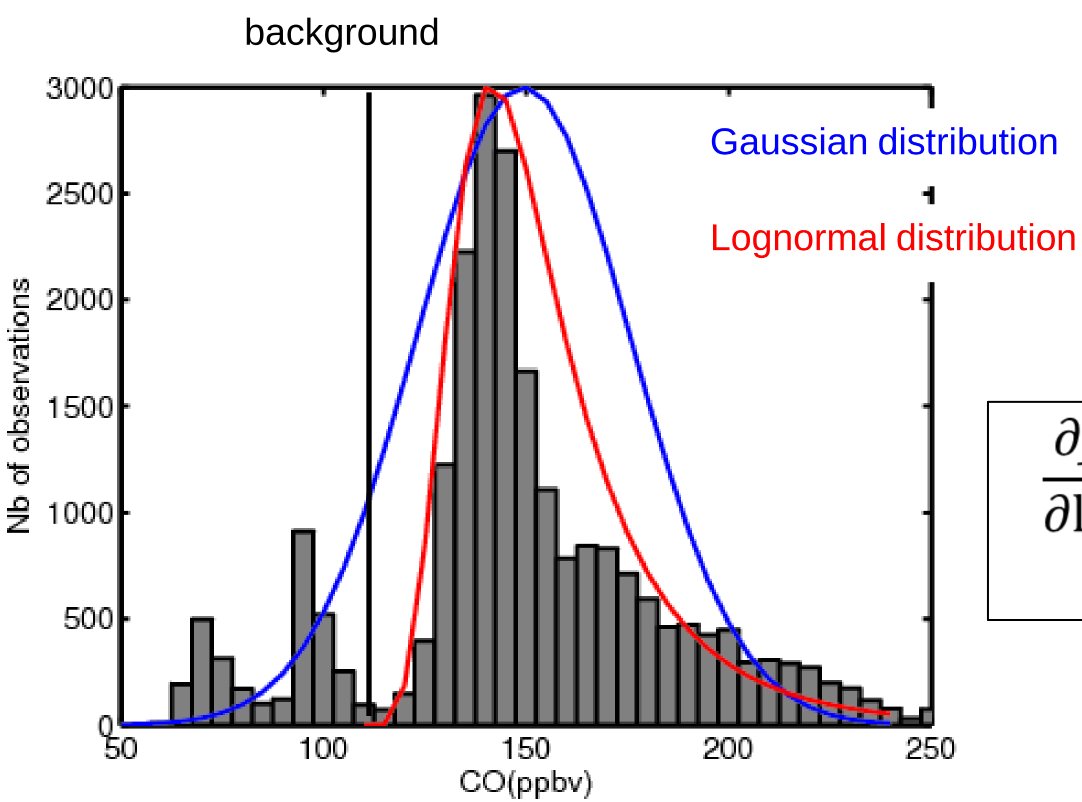
Red=weekday, Blue=weekend

Lagrangian inversion method



Inverse modeling = reducing and balancing εb and εobs

Log normal distribution assumed for CO and NOx observations and parameters

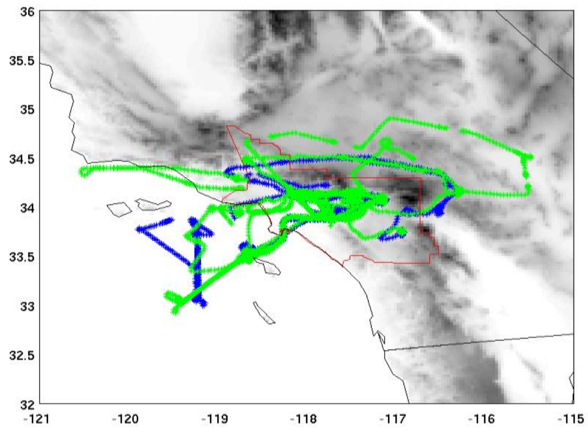


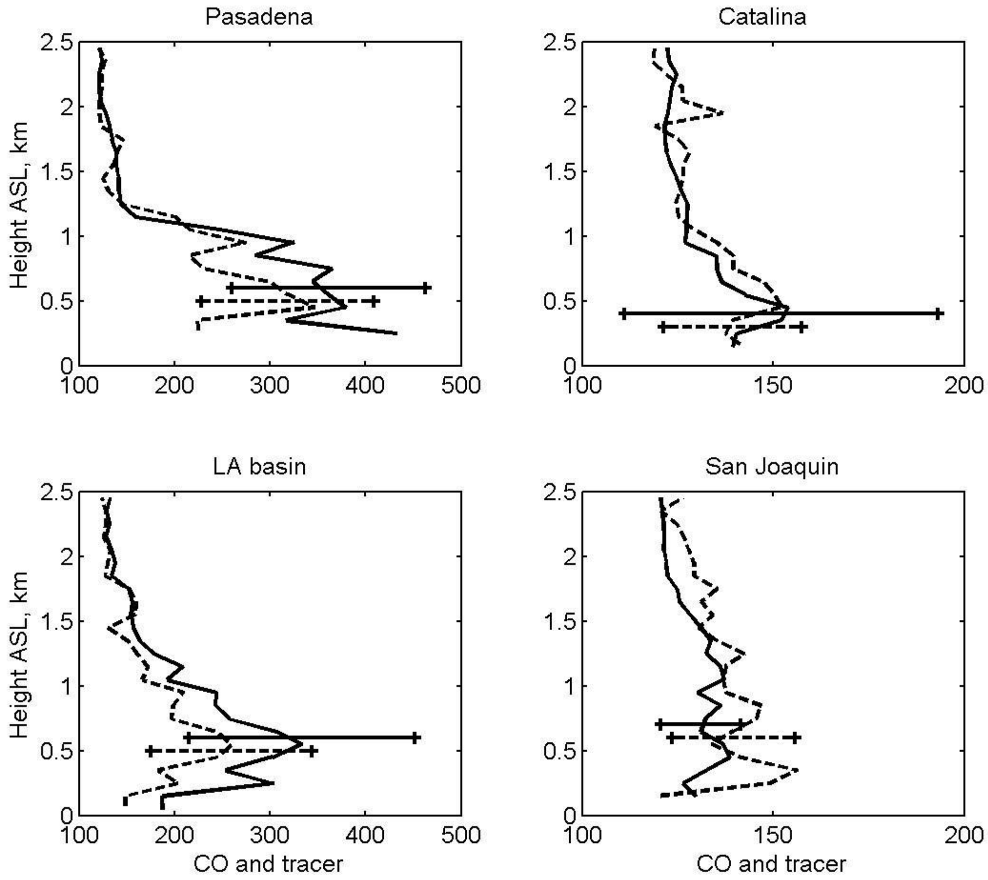
$$\frac{\partial J(x)}{\partial \ln(x)} = -WR^{-1}(\ln y_o - \ln(Hx)) + B^{-1}(\ln x - \ln x_b)$$

$$W = \left[\frac{H(i,j)x(j)}{\sum_j H(i,j)x(j)} \right]_{i,j}$$

Gaussian distributions let observations be negative below background.
Lognormal distributions are closer to reality

No bias found in the meteorology





Observed CO

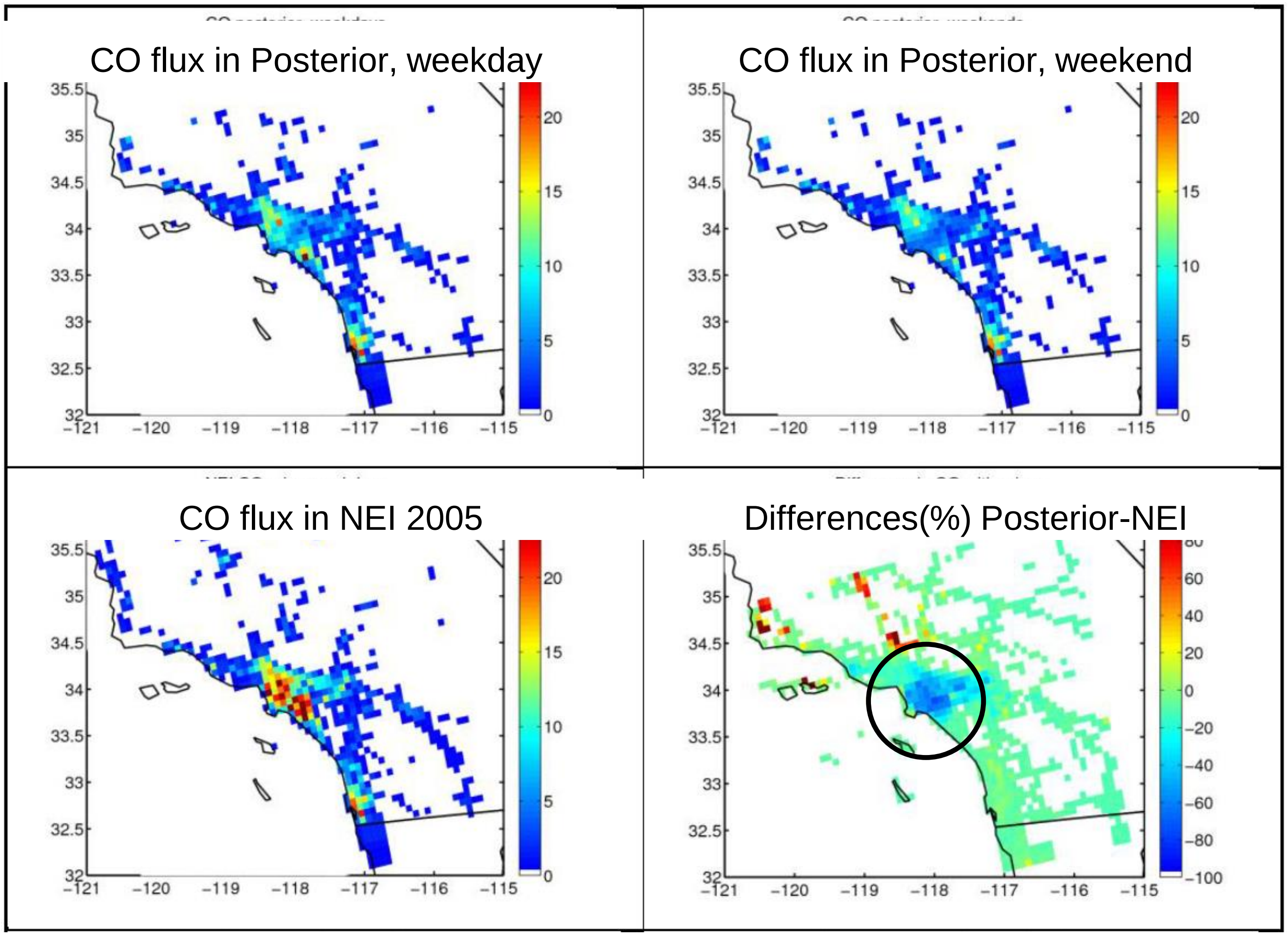
Simulated CO by FLEXPART

No particular bias in the PBL height
Wind speed uncertain by +- 1m/s

From Angevine et al., (2012)

Optimization of CO surface fluxes at mesoscale

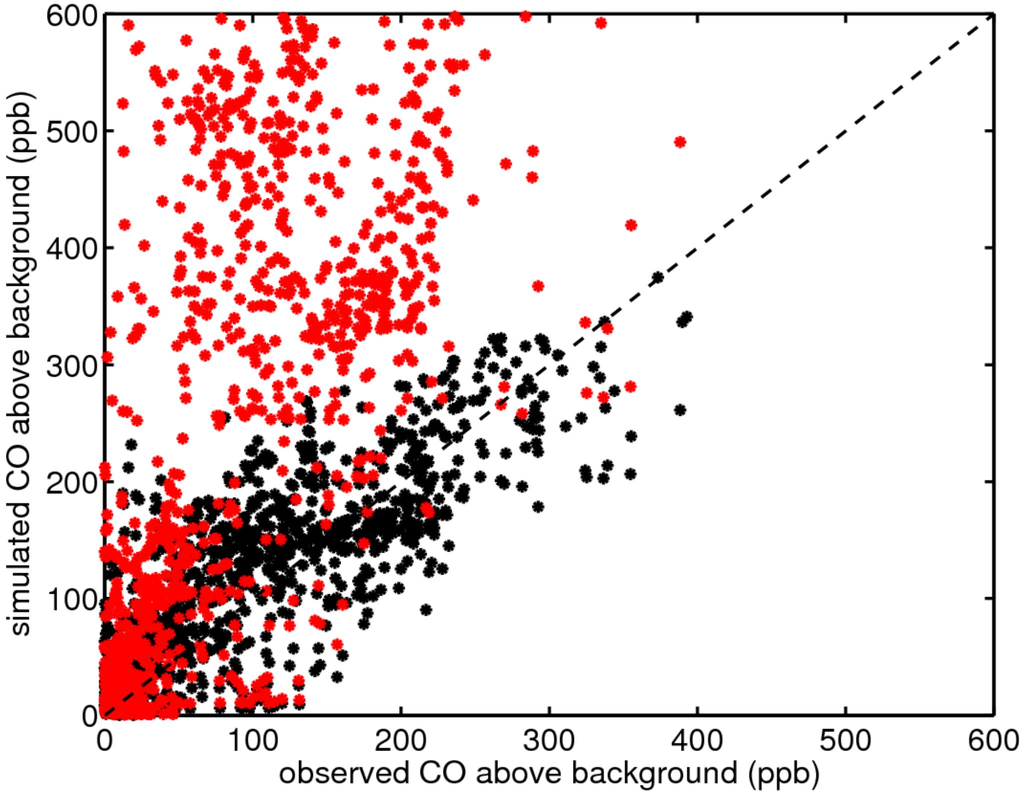
- NEI 2005 used as a prior for weekday and weekend flights



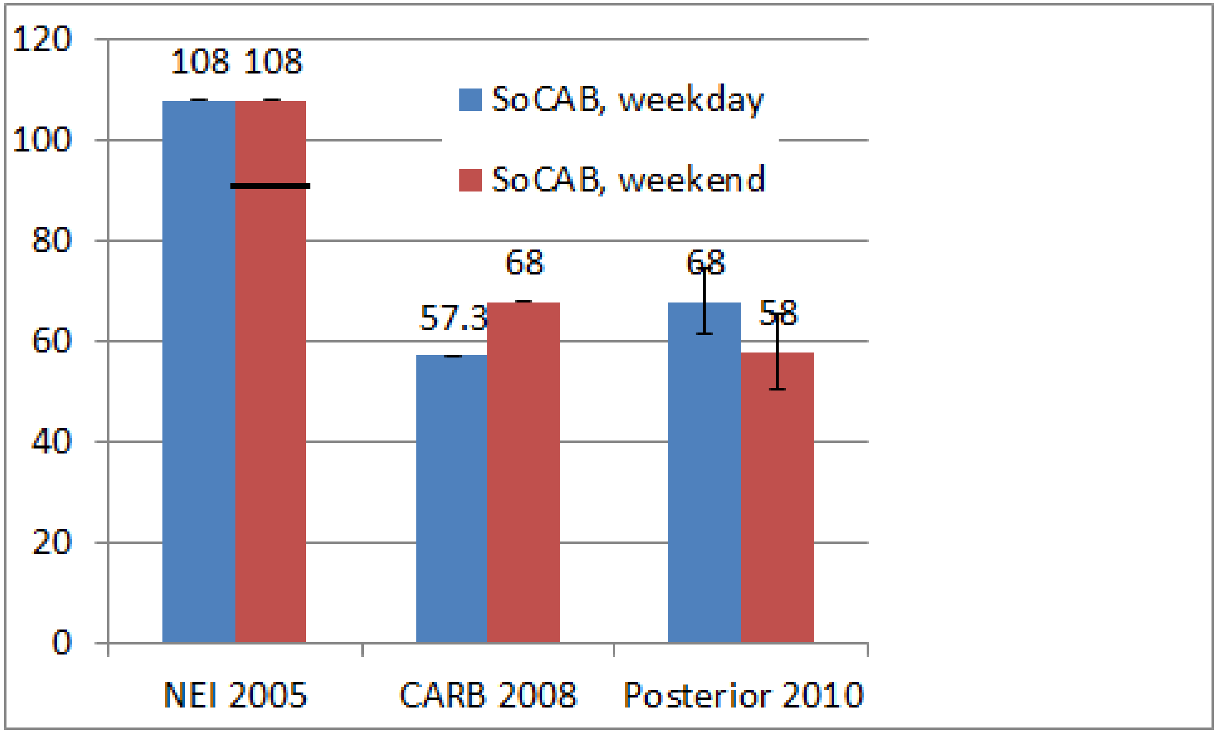
reduction of ~40% in LA county.

Simulated vs Observed CO above background

- Using NEI
- Using Posterior



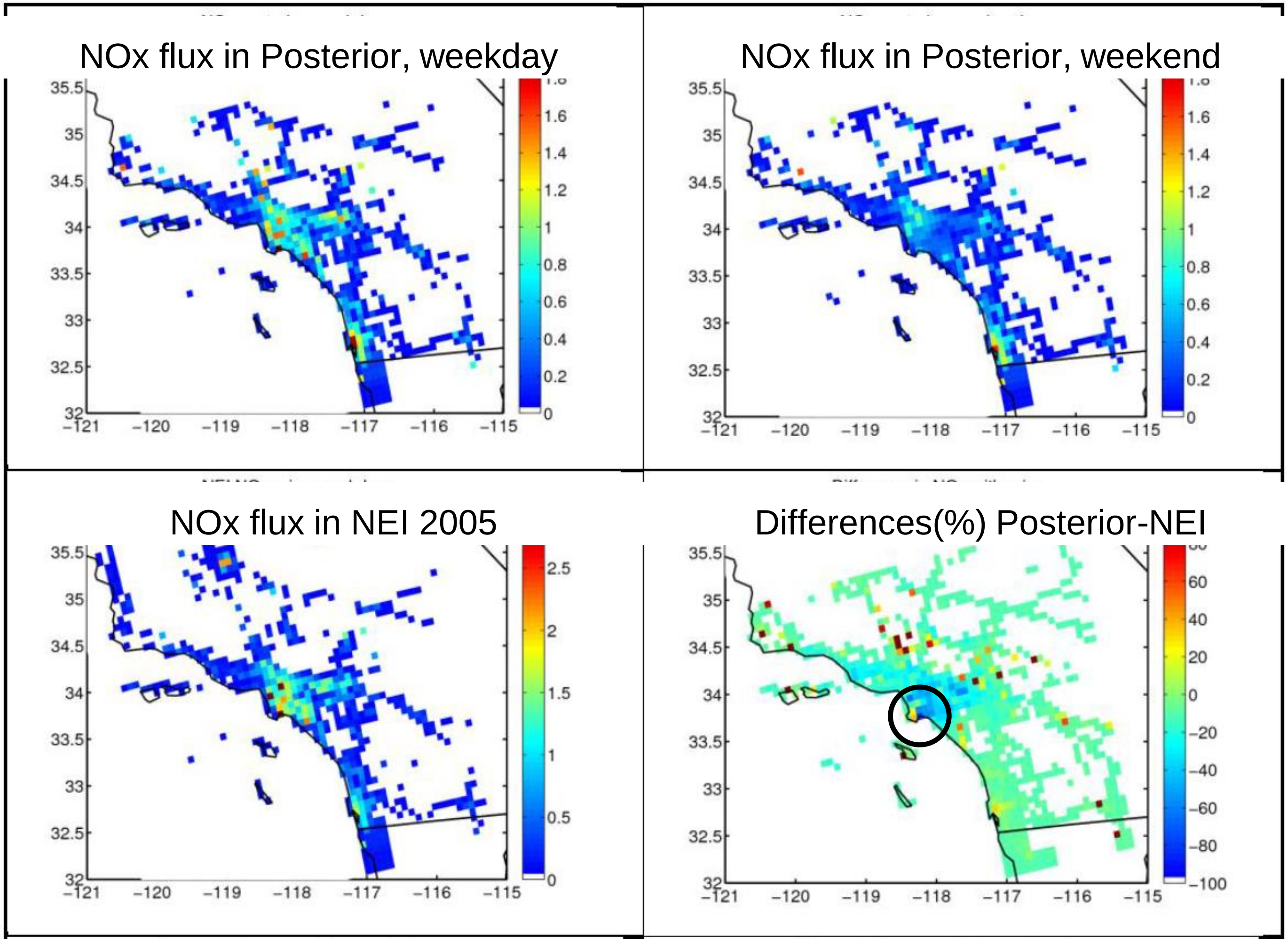
CO flux estimates



- Posterior close to CARB 2008, but lower by 37% compared to NEI 2005
- Weekend effect of -15% in the posterior, consistent with Pollack et al (2012) and NEI(-19%). Opposite sign in CARB.

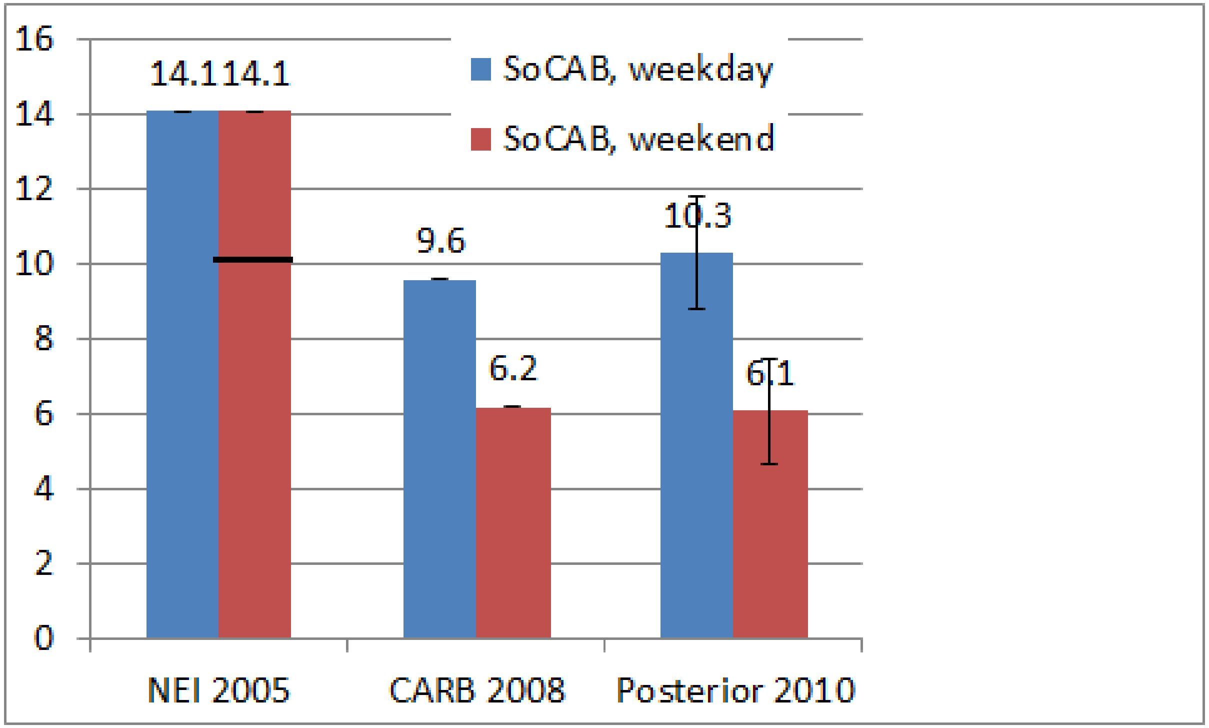
Optimization of NOx surface fluxes at mesoscale

- NEI 2005 used as a prior for weekday and weekend flights



reduction of ~30% in LA county.
factor of 5 reduction the in the Port of LA

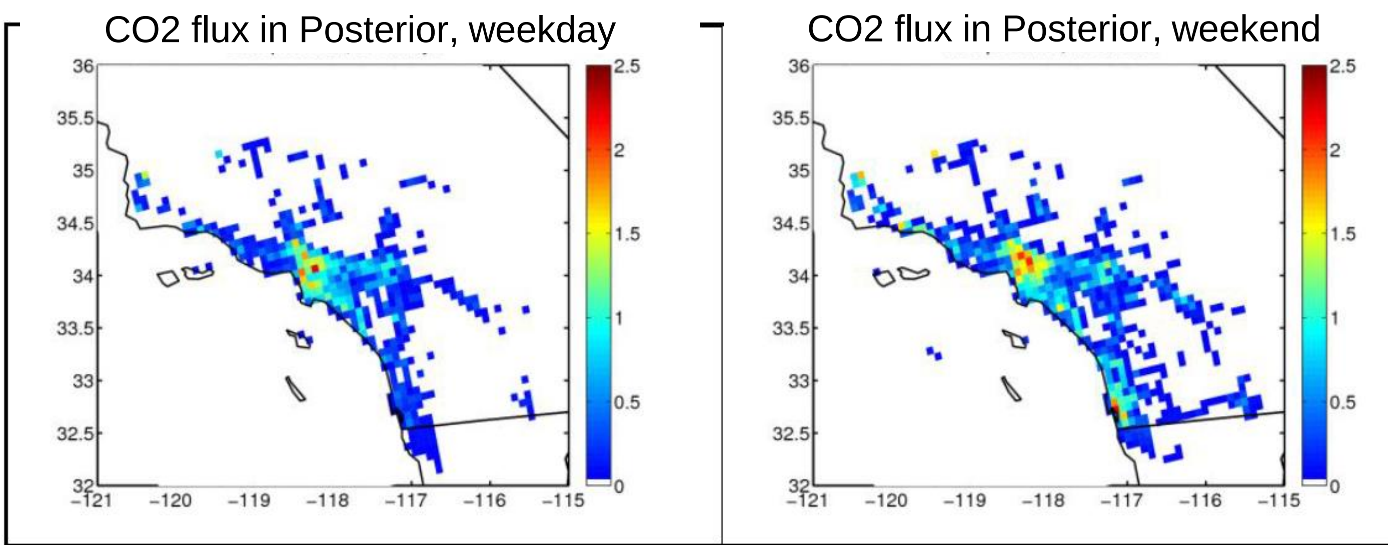
NOx flux estimates



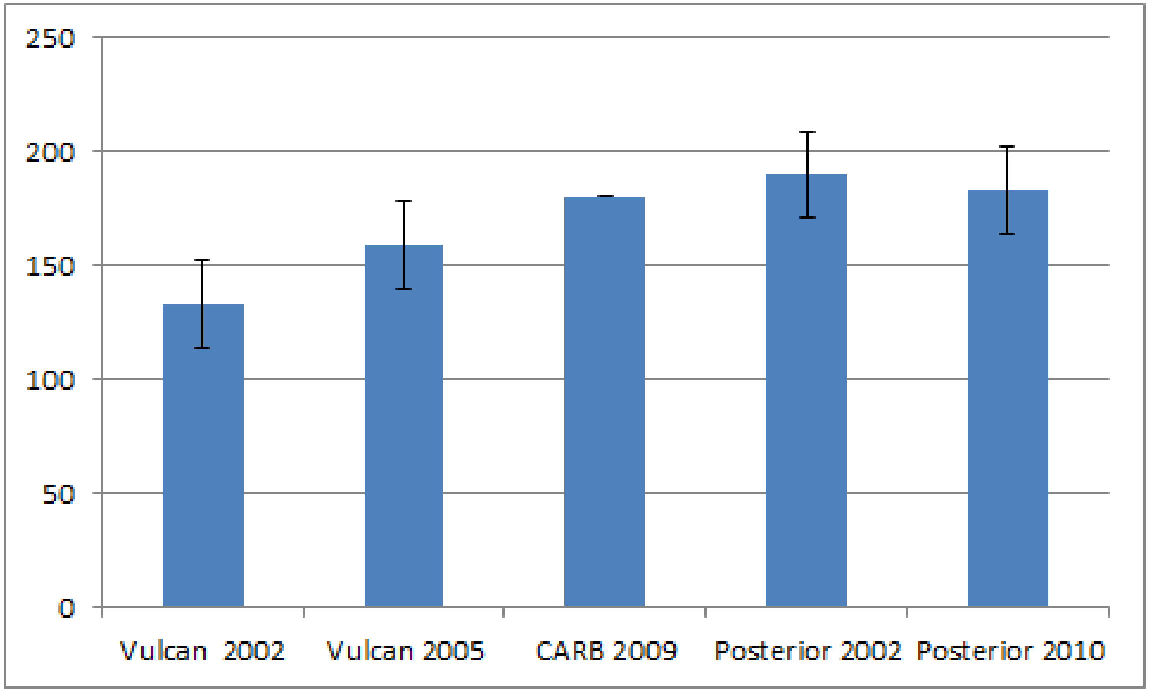
- Posterior close to CARB 2008. Lower by 27 to 40% compared to weekday NEI 2005.
- Strong weekend effect of -40% in the posterior. In agreement with weekend effect in NEI (-29%) and CARB (35%).
- Difference with CARB 2008 statistically insignificant

Optimization of anthropogenic CO_2 fluxes at mesoscale

- No prior estimates used. We used the flux ratio inversion method (Brioude et al., 2012, JGR) based on CO,NOx best estimates and linear correlations with CO_2

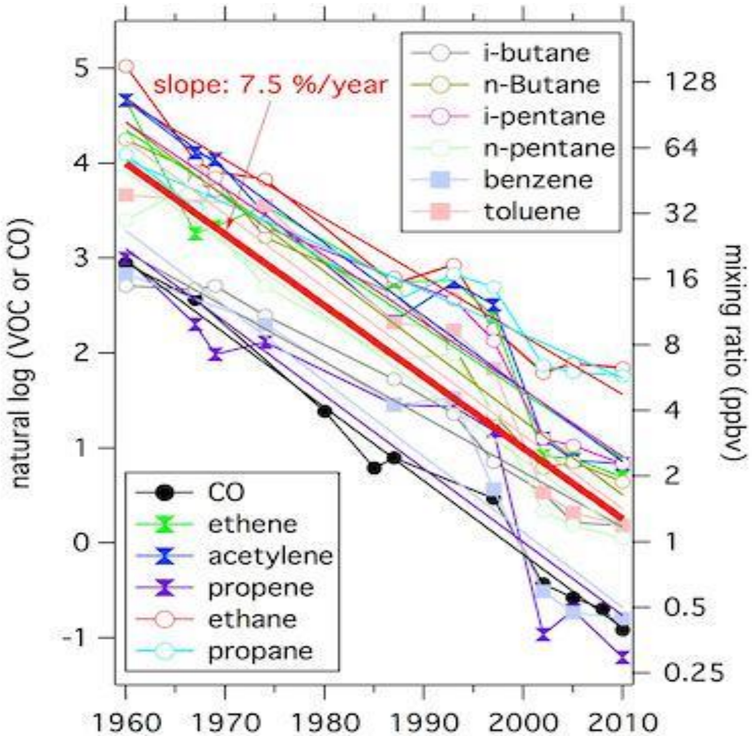


CO2 flux estimates

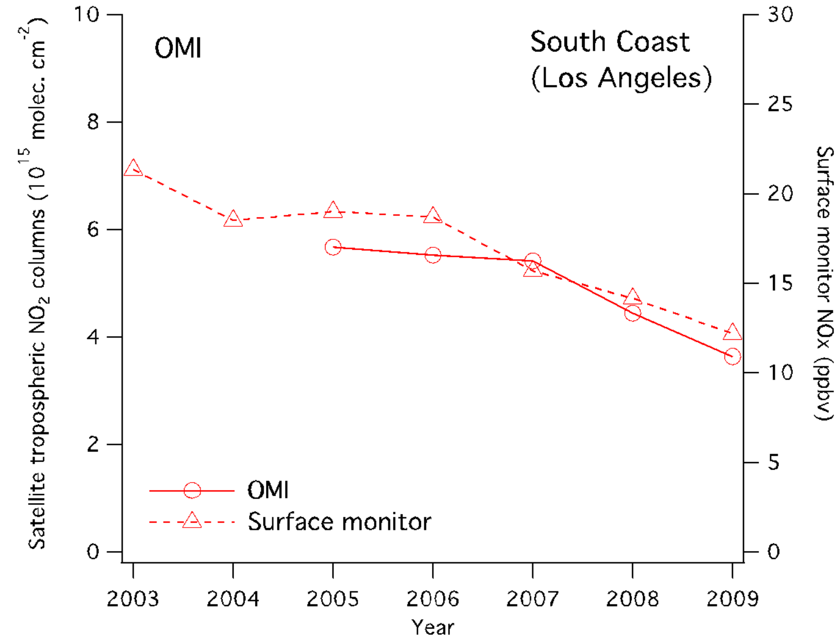


183±18 Tg/year of CO_2 based on the posterior. Good agreement with CARB. Higher than Vulcan by 15 to 38%.

Strong reduction in NOx and VOC emission



Warneke et al., 2012 JGR



Bishop and Stedman, 2008; Dallman and Harley, 2010 for NOx (Courtesy Si-Wan Kim.)

Reduction of 7.8% per year

Reduction of 37% within the past 8 years (McDonald et al., 2012)

Los Angeles from 2002 to 2010

Reductions of 41% in CO emission and 37% in NOx emission found in the posterior between 2002 and 2010 No trend found in CO2 surface fluxes. Consistent with published studies. The CO2 trends (+10 %± 14 % in LA, -4 %± 10 % in SoCAB) are statistically insignificant.

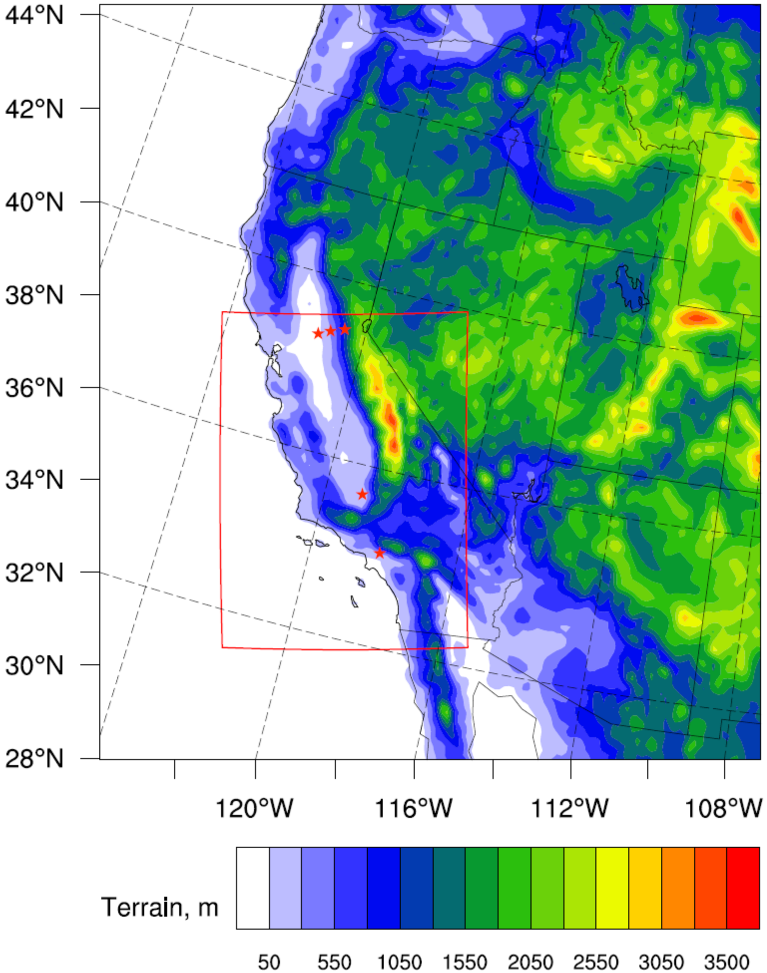
Daytime emission ($kg\,s^{-1}$) in 2010		CO		NO_y		CO_2	
		Weekday	Weekend	Weekday	Weekend	Weekday	Weekend
LA County	NEI 2005	69	69	9.0	9.0	N/A	N/A
	Posterior	39 ± 2.2	32 ± 6.4	6.1 ± 0.6	3.5 ± 0.9	4590 ± 290	4930 ± 670
	CARB 2008	34.1	39.7	6.5	4.2	N/A	N/A
SoCAB	NEI 2005	108	108	14.1	14.1	N/A	N/A
	Posterior	68 ± 6.6	58 ± 7.6	10.3 ± 1.5	6.1 ± 1.4	7440 ± 390	8200 ± 700
	CARB 2008	57.3	68.0	9.6	6.2	N/A	N/A

Table 5. Total daytime emissions of CO, NO_y and CO_2 in Los Angeles County and the SoCAB during weekdays for the posteriors in 2002 from the inversion technique applied in this study.

Daytime emission ($kg\,s^{-1}$) in 2002 (weekday)	CO	NO_y	CO_2
LA County	67.8 ± 2.2	9.6 ± 0.6	4160 ± 150
SoCAB	116 ± 5.4	16.3 ± 0.6	7730 ± 420

Inventory evaluation in 4x4km WRF-Chem runs

WRF-Chem model Domains
D1: Western US (12x12km2 resolution)
D2: California (4x4km2 resolution)



WRF-Chem model version
3.4 (released in February, 2012)

Domains: Western US
Number of vertical levels: 41

<u>Simulation period: May/1-June 30, 2010</u>
Meteorological I.C. and B.C.: ECMWF
Idealized Chemical I.C. and B.C. for 12km resolution domain (D1): clean maritime condition

<u>Anthropogenic emissions: EPA NEI-2005</u>
Biogenic emissions: BEIS3.14
Chemical mechanisms: RACM-SOA

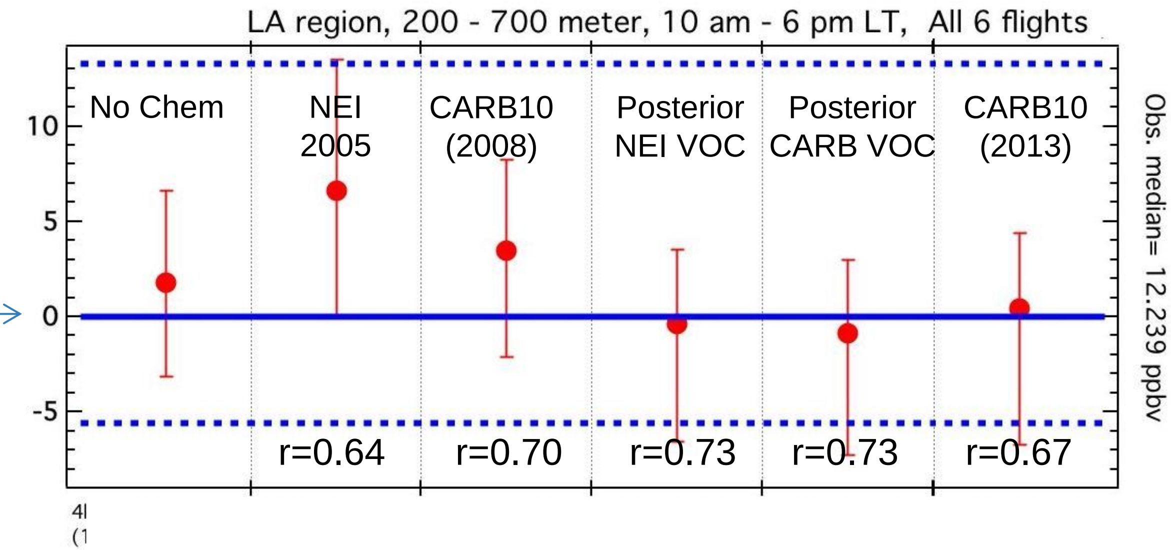
Cumulus parameterization for D1 only
MYNN Planetary Boundary Layer model
Noah Land surface model

Credit: Ravan Ahmadov, Stuart McKeen

Inventory evaluation in 4x4km WRF-Chem runs

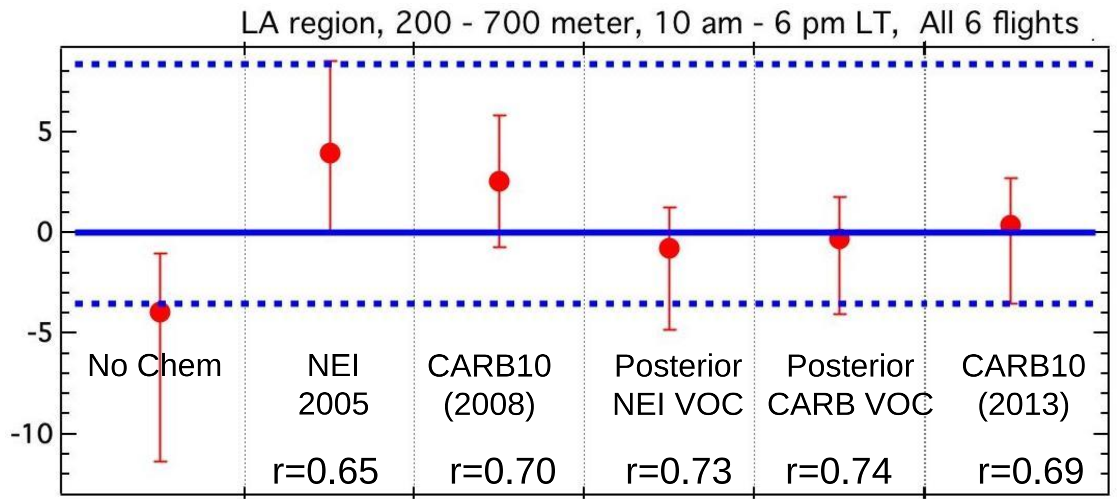
NO2

No bias level

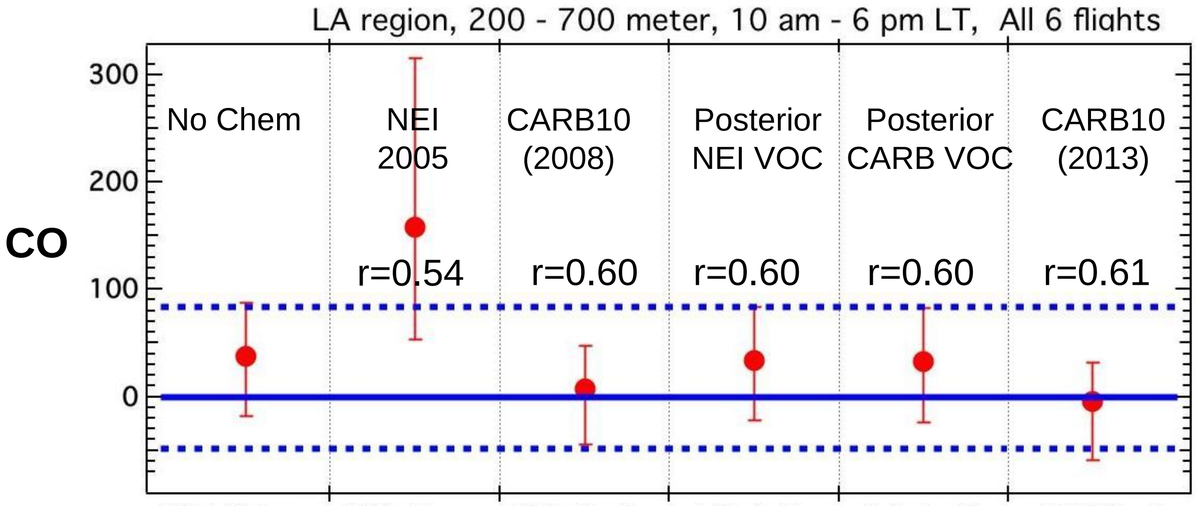


Compared to Brioude et al. (2013), photolysis rate of O^1D has improved.

NOy



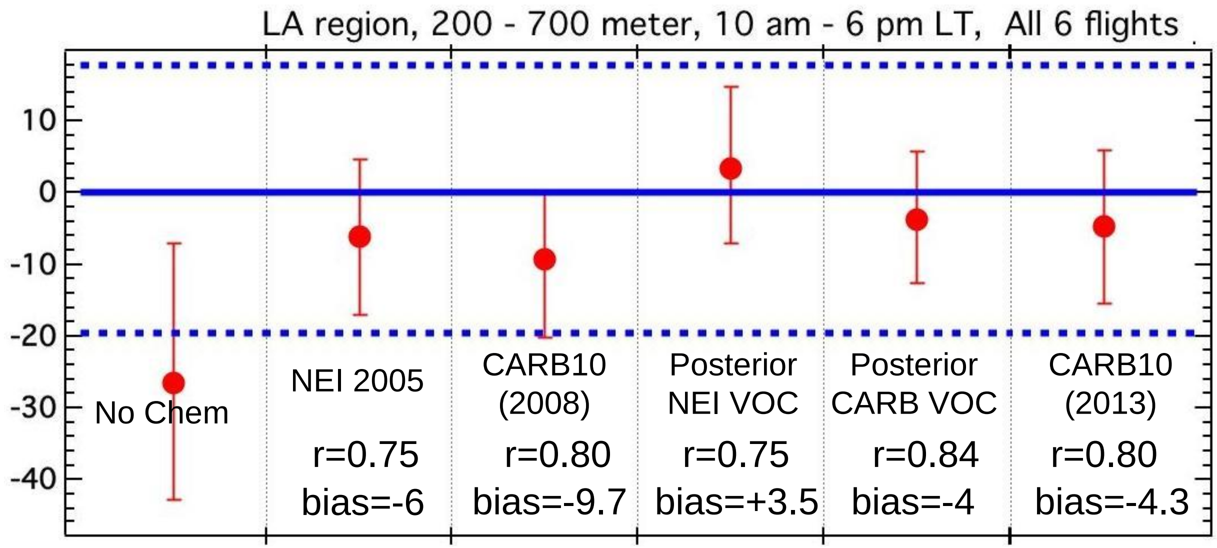
Inventory evaluation in 4x4km WRF-Chem runs



CO discrepancy with NOAA P3 in-situ measurements are largely reduced using the CO posterior compared to NEI 2005. The best results are found using CARB 10 (version 2008 or 2013)

Inventory evaluation in 4x4km WRF-Chem runs

Ozone



Ozone chemistry was also evaluated with the WRF-Chem simulations. The ozone error is -6ppb using NEI and -10 using CARB inventory. The ozone error ranges between -4 and +3.5ppb using the CO and NOy posteriors and NEI or CARB VOCs.

- In WRF-Chem, biases reduced and correlations improved using CO and NOy posteriors

Conclusions

- The inversion seems to do a decent job in estimating surface fluxes of CO, NOy and CO_2 at mesoscale
- Trend in the posteriors between 2002 and 2010 matches the trends in the observations
- NEI 2005 inventory agrees within 40% for CO and NOy posterior emissions in 2010
- Good Agreement with CARB 2008 and 2010
- Single-flight-based inversions have an uncertainty of ~15% and can be used to evaluate existing bottom-up inventories
- For further information, see Brioude et al., 2013, ACP