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.

J. Brioude

Contributing authors:
Modeling and inventories: W.M. Angevine, R. Ahmadov, S-W Kim, S. Evan, S.A. McKeen, E.-Y Hsie, G.J. Frost, M. Trainer

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
- 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

Met fields

- WRF+ECMWF (4x4km), MYJ Every 30 min
- WRF+ECMWF (4x4km), MYNN Every 1h
- WRF+ECMWF+UCM (4x4km), YSU Every 1h

Transport Model

FLEXPART-WRF (24h transport)

4D least square method

Simulated observations

Observation (NOAA P3 measurements)

Observation

Control parameters

Surface inventory (NEI 2005)

Posterior

$\varepsilon_{\text{obs}} =$ simulated observations uncertainty

$\varepsilon_{\text{b}} =$ surface inventory uncertainty

Inverse modeling = reducing and balancing $\varepsilon_{\text{b}}$ and $\varepsilon_{\text{obs}}$
Log normal distribution assumed for CO and NOx observations and parameters

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

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

\[
W = \left[ \frac{H(i,j)x(j)}{\sum_{j} H(i,j)x(j)} \right]_{i,j}
\]
No bias found in the meteorology

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
- 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

![NOx flux in Posterior, weekday](image1.png)

![NOx flux in Posterior, weekend](image2.png)

![NOx flux in NEI 2005](image3.png)

![Differences(%) Posterior-NEI](image4.png)

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 CO2 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 CO2.
CO2 flux estimates

183±18 Tg/year of CO2 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.

<table>
<thead>
<tr>
<th>Daytime emission (kg s⁻¹) in 2010</th>
<th>CO</th>
<th>NOy</th>
<th>CO2</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Weekday</td>
<td>Weekend</td>
<td>Weekday</td>
</tr>
<tr>
<td>LA County</td>
<td>NEI 2005</td>
<td>69</td>
<td>69</td>
</tr>
<tr>
<td></td>
<td>Posterior</td>
<td>39 ± 2.2</td>
<td>32 ± 6.4</td>
</tr>
<tr>
<td></td>
<td>CARB 2008</td>
<td>34.1</td>
<td>39.7</td>
</tr>
<tr>
<td>SoCAB</td>
<td>NEI 2005</td>
<td>108</td>
<td>108</td>
</tr>
<tr>
<td></td>
<td>Posterior</td>
<td>68 ± 6.6</td>
<td>58 ± 7.6</td>
</tr>
<tr>
<td></td>
<td>CARB 2008</td>
<td>57.3</td>
<td>68.0</td>
</tr>
</tbody>
</table>

Table 5. Total daytime emissions of CO, NOy, and CO2 in Los Angeles County and the SoCAB during weekdays for the posteriors in 2002 from the inversion technique applied in this study.

<table>
<thead>
<tr>
<th>Daytime emission (kg s⁻¹) in 2002 (weekday)</th>
<th>CO</th>
<th>NOy</th>
<th>CO2</th>
</tr>
</thead>
<tbody>
<tr>
<td>LA County</td>
<td>67.8 ± 2.2</td>
<td>9.6 ± 0.6</td>
<td>4160 ± 150</td>
</tr>
<tr>
<td>SoCAB</td>
<td>116 ± 5.4</td>
<td>16.3 ± 0.6</td>
<td>7730 ± 420</td>
</tr>
</tbody>
</table>
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

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

Anthropogenic emissions: EPA NEI-2005
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

Compared to Brioude et al. (2013), photolysis rate of O$_1^D$ has improved.

No bias level

NO$_2$

Obs. median = 12.239 ppbv

LA region, 200 - 700 meter, 10 am - 6 pm LT, All 6 flights


r=0.64  r=0.70  r=0.73  r=0.73  r=0.67

r=0.65  r=0.70  r=0.73  r=0.74  r=0.69

NO$_y$

LA region, 200 - 700 meter, 10 am - 6 pm LT, All 6 flights
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)
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