Top-down estimate of surface flux in the Los Angeles Basin using a mesoscale inverse modeling technique: assessing anthropogenic emissions of CO, NO_x and CO_2 and their impacts

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1. INTRODUCTION

The Los Angeles (LA) basin, a large urban area with emissions from mobile sources, industry and agriculture, is a challenging region for chemical-transport models. Previous studies have shown reductions in CO and NOx emission factors from mobile sources (*Bishop and Stedman*, 2008; Dallman and Harley, 2010) and from point sources (Frost et al., 2006) in the US. However, those large reductions over time result in substantial uncertainties in surface emission estimates of ozone precursors based on bottom–up inventories. Beside ozone precursors, urban areas are significant sources of greenhouse gases (Gurney et al., 2009). Duren and Miller (2012) stressed that accurate emission estimates based on top-down approaches for the largest cities in the world are needed to better assess the carbon emission trends.

In May and June 2010, NOAA organized and led the CalNex intensive field campaign that took place in the Los Angeles basin and Central Valley (Ryerson et al., 2013).

In Brioude et al. (2013), we published an analysis that used in-situ aircraft measurements from 6 NOAA P-3 flights in 2010 along with the Weather Research and Forecasting (WRF) mesoscale model in an application of an inversion technique to estimate and improve the CO and NOy surface fluxes from the U.S. Environmental Protection Agency's (EPA's) National Emission Inventory (NEI) for the reporting year 2005. We also used CO2 measurements to estimate urban CO2 emissions from the Los Angeles megacity and the South coast Air basin (SoCAB).

We applied the same inversion techniques as in Brioude et al. (2011; 2012) for Houston, Texas.

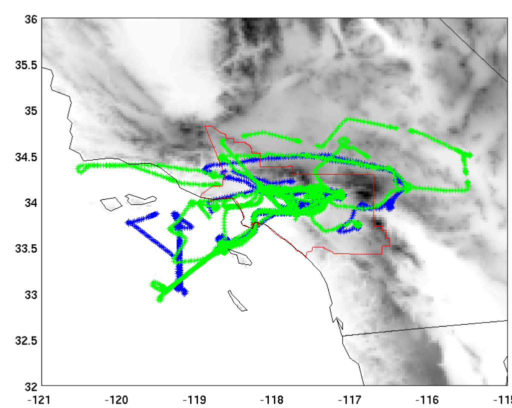

Fig. 1: Map of the domain showing the flight tracks from the 3 weekday flights (blue) and 3 weekend flights (green) of the NOAA P-3 aircraft during CalNex used in this study. From Brioude et al. (2013). The red trace lines out the South coast Air Basin.

We use 3 weekday and 3 weekend flights in our inversions to evaluate differences in surface emissions between weekday and weekend. The inversions using each FLEXPART-WRF transport model with 3 different parameterization settings

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were applied for each individual flight. The results were combined to estimate the mean and uncertainty of surface flux in the LA county and SoCAB.

To address the question of interannual emission trends, we employed aircraft measurements from a single NOAA P-3 flight during the ITCT 2002 campaign over the LA Basin to calculate the surface emissions in May 2002. We used three configurations of the WRF mesoscale model and FLEXPART to simulate the transport between the surface sources and the location of the aircraft measurements. The NEI 2005 inventory was used as a prior estimate for CO and NOx emissions in the inversion method. The CO2 posterior estimates were calculated from the flux ratio inversion method (Brioude et al., 2012), which allows the calculation of a posterior at mesoscale without a prior estimate.

This abstract shows a summary of the principal results of Brioude et al. (2013).

2. RESULTS

2.1 *CO*

Compared to the NEI 2005 inventory, the daytime CO emissions from LA County during weekdays are reduced by 43%, with an uncertainty of 6% (Figure 2). The SoCAB emissions are reduced by 37% ± 10%. The posterior CO emissions were higher by 15% compared to California Air Resources Board (CARB) 2008 inventory.

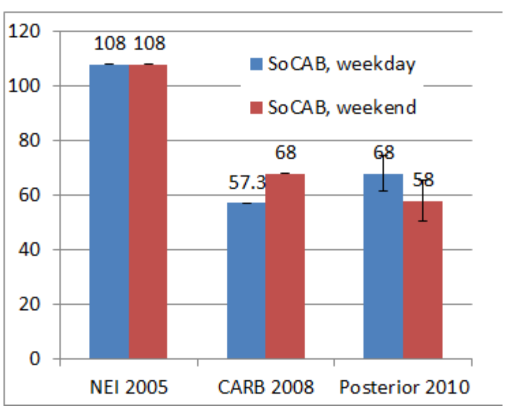

Fig. 2. Daytime CO surface emissions (kg/s) in the South coast air basin (SoCAB) in NEI 2005, CARB 2008 and the CO posterior. Weekday and weekend values are also shown.

2.2 N*Oy*

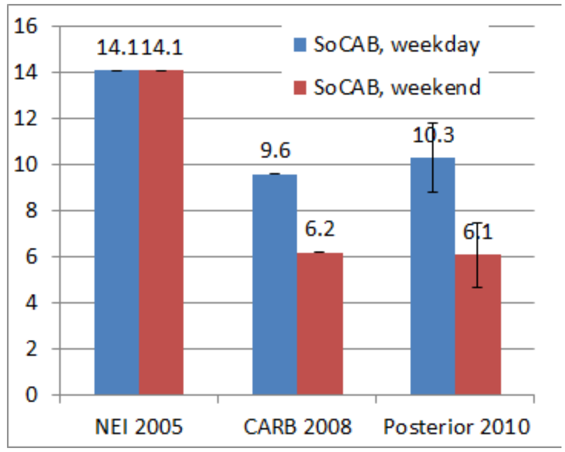

Fig. 3: Same as Fig2, but valid for NOy emissions.

Compared to the NEI inventory, the daytime weekday NOy posterior emissions are reduced by 32% ± 10% in Los Angeles County, while emissions in the SoCAB region are reduced by 27% ± 15%.

The difference between weekday and weekend is 43% in LA County and 40% in the SoCAB area. Those results agree with Pollack et al. (2012), who found a reduction of 34% to 46% based on in situ measurements. No weekend information is shown for NEI 2005 to highlight the fact that the same prior was used for both weekday and weekend flights.

Compared to CARB 2008, the posterior NOy was lower by 6% in LA County but higher by 7% in the SoCAB region, all within the uncertainty range of the inversion.

2.3 *CO_2*

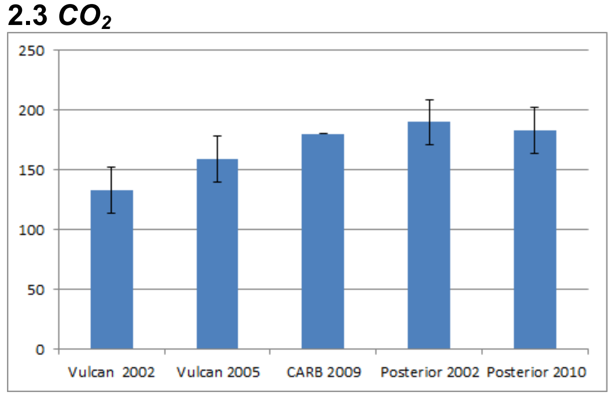

Fig. 4: Yearly average CO2 emission (Tg/year) in SoCAB in Vulcan 2002 and 2005, CARB 2009 and the posteriors in 2002 and 2010. A multiplicative coefficient of 0.78 is applied to the daytime CO2 posterior estimates to convert them into yearly average estimates based on Vulcan diurnal profile

CO2 posterior estimates are based on the flux ratio inversion method (Brioude et al., 2012a), which allows estimates of CO2 (or any species) at mesoscale without using a prior estimate. The linear relationships of CO2 with co-emitted species like CO and NOy, and their surface flux posterior estimates are used in the flux ratio inversion to calculate CO2 emission estimates for weekdays and weekends

The 2010 CO2 posterior estimate is higher than Vulcan by 31 to 44% in LA County and 15 to 38% in SoCAB. The 2010 posterior estimate in SoCAB is in agreement with CARB 2009.

2.4 *CO, NOy and CO_2 posteriors in 2002*

Trends between 2002 and 2010 were also evaluated by calculating surface fluxes in May 2002 using one weekday flight during the ITCT 2002 campaign over the LA Basin using NEI 2005 as a prior. Differences between the posteriors in 2002 and 2010 are driven by changes in observed concentrations and model uncertainties. From 2002 to 2010, the CO emission in the posteriors decreased by 42%±6% in LA County and 41%±10% in SoCAB. NOy emission in the posteriors decreased between 2002 and 2010 by 36%±10% in LA County and 37%±15% in SoCAB. CO2 emission in the posteriors increased between 2002 and 2010 by 10%±14% in LA County, but decreased by 4%±10% in SoCAB during the same time period. These variations are within the uncertainty range of our calculations.

The CO, NOy and CO2 2002-2010 emission trends found in the posteriors are in agreement with other observation-based studies (Warneke et al., 2012; McDonald et al., 2012).

2.5 Improvement in WRF-Chem chemistry

To evaluate the posterior estimates, we used them along with NEI and the gridded CARB inventories in WRF-Chem v3.4 Eulerian model simulations of the same CalNex P-3 flights considered in the inversion calculations. In Figure 5, results with the CO and NOy posteriors are shown using VOCs emission from NEI and CARB. The column "No Chemistry" on Figure 5 refers to the results without chemistry but with transport of boundary conditions in WRF-Chem only. For details on the chemistry options used, biogenic VOC fluxes and additional details, see Ahmadov et al. (2012).

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

CO discrepancy with NOAA P3 in-situ measurements are largely reduced using the CO posterior compared to NEI 2005. The CO error using the posterior is slightly larger than the one using CARB.

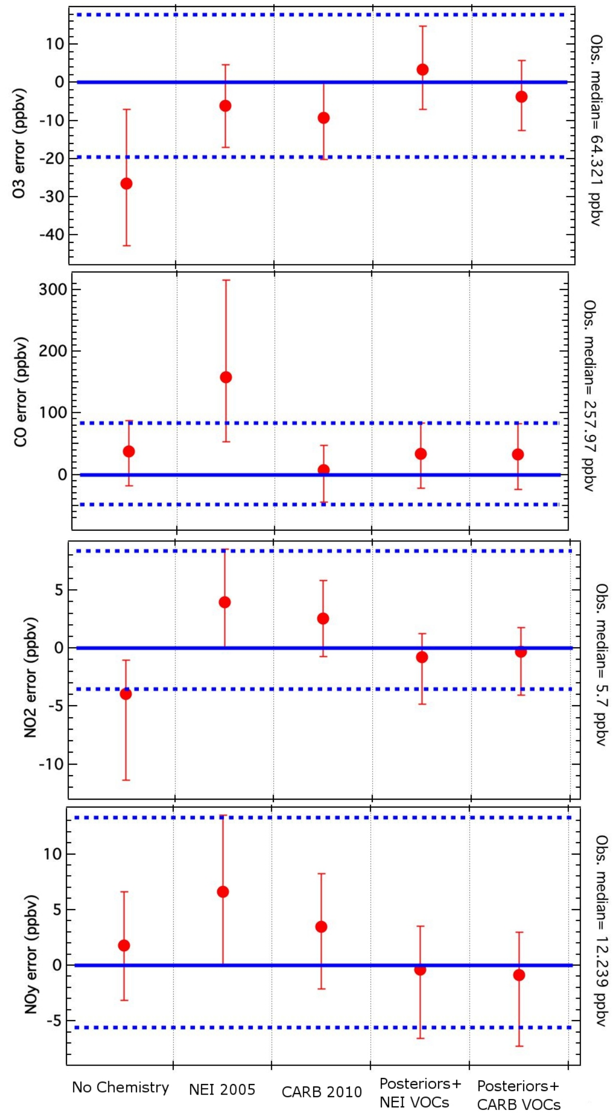

Fig. 5: Differences in Ozone, CO, NO2 and NOy mixing ratio between WRF-Chem and in-situ measurements from the NOAA P3 aircraft in 2010 using different emission inventories.

The median NO2 and NOy errors using the posterior are statistically insignificant. Ozone chemistry was also evaluated with the WRF-Chem simulations. The ozone error is -6ppb using NEI and -8 using CARB inventory. The ozone error ranges between -3 and +3ppb using the CO and NOy posteriors and NEI or CARB VOCs.

3. CONCLUSIONS

We have shown that the transport models and the inversion techniques were successful in improving bottom-up CO, NOy and CO2 inventories. Using the CO and NOy posteriors, ozone chemistry of the LA basin is improved in WRF-Chem. VOC emissions in Los Angeles can be estimated with a good accuracy using the CO posterior estimates from this study and observed CO-VOC emission ratios (Borbon et al., 2012). We have also shown that it is possible to evaluate the decadal change of CO2 and other anthropogenic species in a megacity. For further details and proper reference of this work, see Brioude et al. (2013).

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