Geophysical Research Abstracts
Vol. 15, EGU2013-1398-1, 2013
EGU General Assembly 2013

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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We present top-down estimates of anthropogenic CO, NO_x and CO_2 surface fluxes at mesoscale using a Lagrangian model in combination with three different WRF model configurations, driven by data from aircraft flights during the CALNEX campaign in southern California in May–June 2010. The US EPA National Emission Inventory 2005 (NEI 2005) was the prior in the CO and NO_x inversion calculations. The flux ratio inversion method, based on linear relationships between chemical species, was used to calculate the CO_2 inventory without prior knowledge of CO_2 surface fluxes. The inversion was applied to each flight to estimate the variability of single-flight-based flux estimates. In Los Angeles (LA) County, the uncertainties on CO and NO_x fluxes were 10% and 15%, respectively. Compared with NEI 2005, the CO posterior emissions were lower by 43% $\pm$ 6% in LA County and by 37% $\pm$ 10% in the South Coast Air Basin (SoCAB). NO_x posterior emissions were lower by 32% $\pm$ 10% in LA County and by 27% $\pm$ 15% in the SoCAB. NO_x posterior emissions were 40% lower on weekends relative to weekdays. The CO_2 posterior estimates were 183 $\pm$ 18 Tg yr^{-1} in SoCAB. A flight during ITCT in 2002 was used to estimate emissions in the LA Basin in 2002.
From 2002 to 2010, the CO and NO_x posterior emissions decreased by 41% and 37%, respectively, in agreement with previous studies. Over the same time period, CO_2 emissions increased by 10% $\pm$ 14% in LA County but decreased by 4% $\pm$ 10% in the SoCAB, a statistically insignificant change. Overall, the posterior estimates were in good agreement with the California Air Resources Board (CARB) inventory, with differences of 15% or less. However, the posterior spatial distribution in the basin was significantly different from CARB for NO_x emissions. WRF-Chem mesoscale chemical-transport model simulations allowed an evaluation of differences in chemistry using different inventory assumptions, including NEI 2005, CARB 2010 and the posterior inventories derived in this study. The biases in WRF-Chem ozone were reduced and correlations were increased using the posterior from this study compared with simulations with the two bottom-up inventories, showing that improving the spatial distribution of ozone precursor surface emissions is also important in mesoscale chemistry forecasts.