

# BIOGENIC VS. ANTHROPOGENIC SOURCES OF CO OVER THE UNITED STATES

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## 1. Introduction

Parrish, [2006] using urban ambient measurements of the CO/NO<sub>x</sub> ratio along with fuel sales data find that the CO source from on-road transport, which makes up 60% of total CO emissions in the United States, is overestimated in by 50% in current inventories.

Here, we use aircraft observations from the ICARTT campaign over eastern North America in summer 2004, interpreted with a global 3-D model of tropospheric chemistry (GEOS-Chem) to better quantify the total U.S. anthropogenic CO source. Observations in the continental boundary layer imply a U.S. anthropogenic CO source of 37 Tg yr<sup>-1</sup>, 60% lower than specified in the total U.S. EPA inventory for 1999, and lower in summer than the biogenic CO source from the oxidation of VOCs.

## 2. GEOS-Chem Model and ICARTT

### OBSERVATIONS

ICARTT was a coordinated multi-aircraft atmospheric chemistry field program over eastern North America and the North Atlantic in July-August 2004 [Fehsenfeld et al., 2006; Singh et al., 2006]. We use CO observations from INTEx-NA and ITCT 2k4 aircraft and surface measurements from Chebogue Point on the southern tip of Nova Scotia (44°N, 66°W) [Millet et al., 2006b].

### GEOS-Chem MODEL

#### ANTHROPOGENIC EMISSIONS\*

EPA NEI 99 Anthropogenic Emissions over U.S.

#### BIOGENIC EMISSIONS\*\*

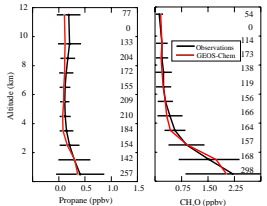
GEIA inventory [Guenther et al., 1995]. (Note: During ICARTT, E. U.S. Emissions of isoprene from GEIA - MEGAN ~ 10%)

**GEOS-CHEM**  
3D Model of Tropospheric (NO<sub>x</sub>-CO-hydrocarbon-O<sub>3</sub>) Chemistry  
30 σ-levels, 2°x2.5°

\*NO<sub>x</sub> emissions from stationary sources reduced by 50% as in Hudman et al., [2007].

\*\*Monoterpene yield 0.2 CO per atom C [Duncan et al., 2007]. Biogenic alkene (C<sub>2</sub>=3) emissions are assumed to be 10% of isoprene on a per molecule basis. Biogenic acetone emissions are as in Jacob et al., [2002].

### SIMULATION OF CONVECTION AND VOC EMISSIONS



Boundary layer ventilation is constrained by vertical profile of short-lived VOCs [Propane and Xiao et al., (2007ab) for C<sub>2</sub>H<sub>6</sub>, C<sub>2</sub>H<sub>4</sub>]

Biogenic VOCs are well-constrained by successful simulation of formaldehyde to within 30% [Millet et al., [2006a]

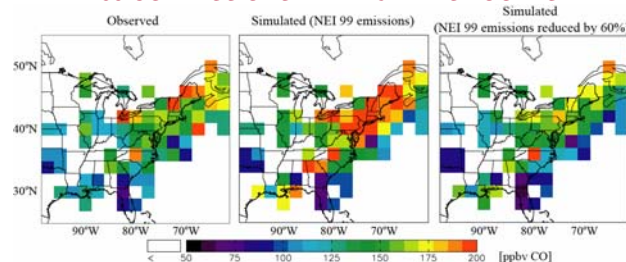
We conclude that the CO simulation bias reflects excessive anthropogenic emission in the model, and this is consistent with the spatial pattern of the bias.

## ACKNOWLEDGEMENTS

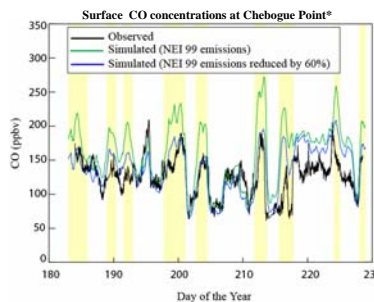
EPA NEI 99 emissions were provided by Alice Gilliland (U.S. EPA). This work was supported by the NASA Global Tropospheric Chemistry Program and the NOAA Office of Global Programs.

## 3. Observational constraints on U.S. Anthropogenic CO source

**BOTH AIRCRAFT AND SURFACE DATA SUGGEST NEI 99 CO EMISSIONS ARE 2.5 TIMES TOO HIGH!**



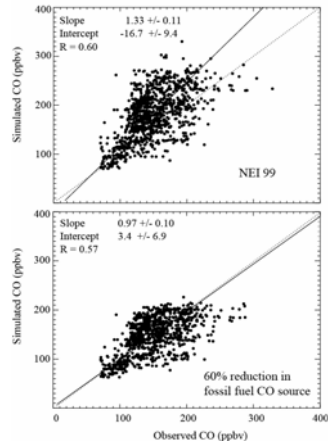
The model using NEI 99 emissions shows a consistent overestimate across eastern North America of 20-50 ppbv that matches the spatial distribution of the anthropogenic CO source.



Surface observations at Chebogue Point show a similar overestimate, as previously discussed by Millet et al. [2006b].

\* Yellow bands are periods of North American (NA) outflow diagnosed from the model (NA fossil fuel > 45 ppbv).

### Simulated vs. observed CO concentrations at 0-1.5 km altitude\*



The regression line indicates an overestimate of 33 ± 11%. We find that correcting the bias requires reducing anthropogenic CO emissions by 60% relative to the NEI 99 values

Applying this decrease also improves the simulated geographical distribution and the simulation at Chebogue Point (see above figures).

\*Regression lines are derived from the reduced-major-axis (RMA) method allowing for errors on both variables, and 95% confidence intervals are determined by bootstrap resampling (n=sample size).

## 4. Emissions and region of influence

CO sources in the contiguous United States for July 1 – August 15, 2004

Source Type	Magnitude (Tg CO) Constrained / NEI99
Fuel and industry	6.4 / 13.3*
Vegetation	9.1 ± 2.7**
Biomass Burning	0.16***
Total	15.8

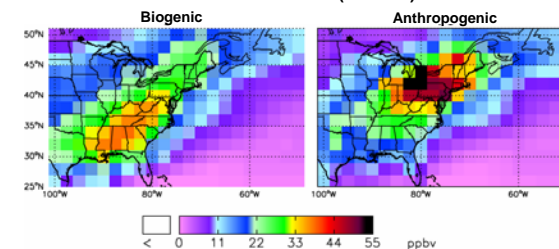
\* including a secondary source of 1.8 Tg from oxidation of anthropogenic VOCs, calculated for the purposes of this table by assuming VOC-specific CO yields from Duncan et al., [2007]. The anthropogenic emission of CO (4.6 Tg CO) represents a 60% reduction from the NEI 99 inventory.

\*\* including 6.7 Tg from isoprene oxidation and 2.4 Tg from oxidation of other biogenic VOCs (acetone, monoterpenes, >C3 alkenes). For isoprene we assume a 2.3 molar yield of HCHO from isoprene oxidation as constrained by the ICARTT observations [Millet et al., 2006a], and further assume 100% conversion of HCHO to CO. For the other biogenic VOCs we use the CO yields from Duncan et al., [2007].

\*\*\* North American fires during ICARTT were mainly outside the contiguous United States; Alaskan and Canadian fires produced 19 Tg CO during this period [Turquety et al., 2007].

Remarkably, the anthropogenic CO source in the United States is now lower than the biogenic source during the summer.

### CO Enhancements (0-2.5 km)\*



\* Calculated as difference between standard simulations and shutting off Biogenic VOCs (left) and NEI 99 Anthropogenic CO (reduced by 60%) and VOCs (right) for 130°-70°W, 25°-50°N. Canadian and U.S. boxes bordering Canada are from Bey et al., [2001] and are not reduced.

Global burden of CO due to anthropogenic emissions is larger, despite lower emissions due to non-linearity in OH → X5 OH increase when biogenics emissions shut off.

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