What can we learn from HCHO column measurements from space? Hydrocarbon emissions, error analysis, and OMI-GOME comparison

D. B. Millet (dbm@io.harvard.edu)[1], D. J. Jacob[1], T. Kurosu[2], K. Chance[2], S. Turquety[1], R. C. Hudman[1], S. Wu[1], A. Fried[3], J. Walega[3], B. G. Heikes[4], D. R. Blake[5], H. B. Singh[6], B. E. Anderson[7], and A. D. Clarke[8]

[1]Harvard University [2]Harvard-Smithsonian Center for Astrophysics [3]NCAR [4]University of Rhode Island [5]U.C. Irvine [6]NASA Ames Research Center [7]NASA Langley Research Center [8]University of Hawaii at Manoa

Measurements of formaldehyde columns from space provide constraints on emissions of volatile organic compounds. Quantitative interpretation requires characterization of errors associated with HCHO column retrievals and the relationship of these columns to VOC emissions. Here we use aircraft measurements from the summer of 2004 to determine the local relationships between HCHO columns and VOC emissions, quantify the uncertainties in satellite measurements of HCHO columns due to the mass factor computation, and draw conclusions regarding the mapping of VOC emissions from space.

1. Distribution of HCHO Over North America

The objective of INTEX-A (July 1 – August 15, 2004) was to observe the chemical outflow from North America and infer constraints on chemical sources and export. Here we use HCHO concentrations measured aboard the NASA DC-8 aircraft and simulated using the GEOS-Chem chemical transport model over North America and the adjacent oceans during INTEX-A.

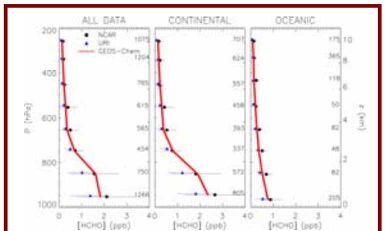

Figure 1. Mean simulated (lines) and observed (symbols) HCHO vertical distributions during INTEX-A.

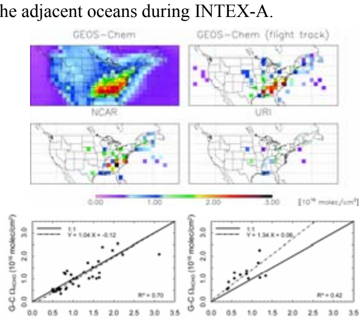

Figure 2. HCHO columns (Ω_{HCHO}) over North America during INTEX-A. Top left panel: GEOS-Chem simulated columns averaged over the INTEX-A period. Other panels: columns computed from simulated and observed [HCHO] during the DC-8 vertical profiles.

2. Relating HCHO Columns to VOC Emissions

We use the data from the INTEX-A aircraft profiles to determine how column HCHO data from space can be interpreted in terms of the underlying reactive VOC emissions.

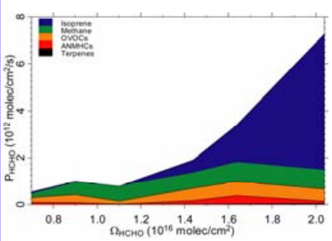

Figure 3. Relationship between measured Ω_{HCHO} and HCHO production rate from different precursors. Isoprene is the dominant source of Ω_{HCHO} variability.

Result: Variability in Ω_{HCHO} over North America in summer is mainly determined by isoprene emission. Satellite retrievals of Ω_{HCHO} can therefore be used reliably as a proxy for isoprene emissions over North America.

3. HCHO Yield from Isoprene

At steady state and in the absence of horizontal transport, the column integral HCHO is related to those of precursors *i* by Eqn. 1. The slope of a plot of Ω_{HCHO} vs. Ω_{ISOP}, normalized by k_{ISOP}/k_{HCHO}, thus estimates the molar yield of HCHO production from isoprene oxidation.

$$\Omega_{HCHO} = \frac{1}{k_{HCHO}} \sum_i k_i Y_i \Omega_i$$

Eqn. 1

k_{HCHO}, k_i: Column-average rate constants for chemical loss of HCHO and precursor *i*

Y_i: Molar HCHO yield from the oxidation of species *i*

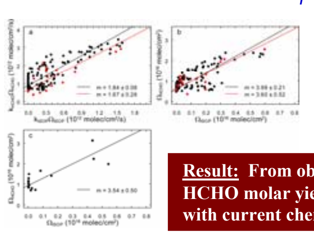

Figure 4. Simulated (a, b) and observed (c) relationships between HCHO and isoprene columns. Model results shown for the entire INTEX-A domain (black) and for the locations and times of the DC-8 vertical profiles (red).

Result: From observed HCHO and isoprene profiles we find an HCHO molar yield from isoprene oxidation of 1.6 ± 0.5, consistent with current chemical mechanisms.

4. Uncertainty in HCHO Column Data from Space

The dominant source of error in HCHO retrievals is the air mass factor (AMF), which defines the relationship between the HCHO slant and vertical columns.

Goal: Employ the extensive mapping of HCHO over North America from INTEX-A to quantify the uncertainties and bias in the AMF calculation.

Approach: Calculate AMFs separately based on measurements and model results for each DC-8 vertical profile. The comparison statistics then give a measure of the corresponding error in retrieved satellite HCHO vertical columns.

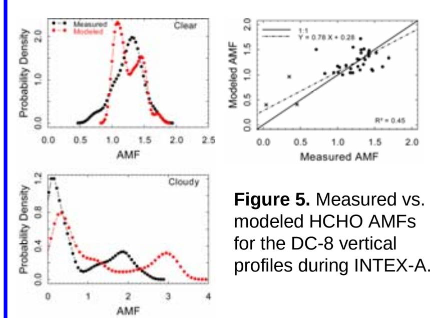

Figure 5. Measured vs. modeled HCHO AMFs for the DC-8 vertical profiles during INTEX-A.

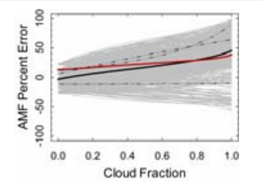

Figure 6. Error in the modeled AMF as a function of cloud fraction. Solid black line: mean bias; red line: SD in the bias.

5. A Comparison of HCHO Columns from OMI, GOME and GEOS-Chem

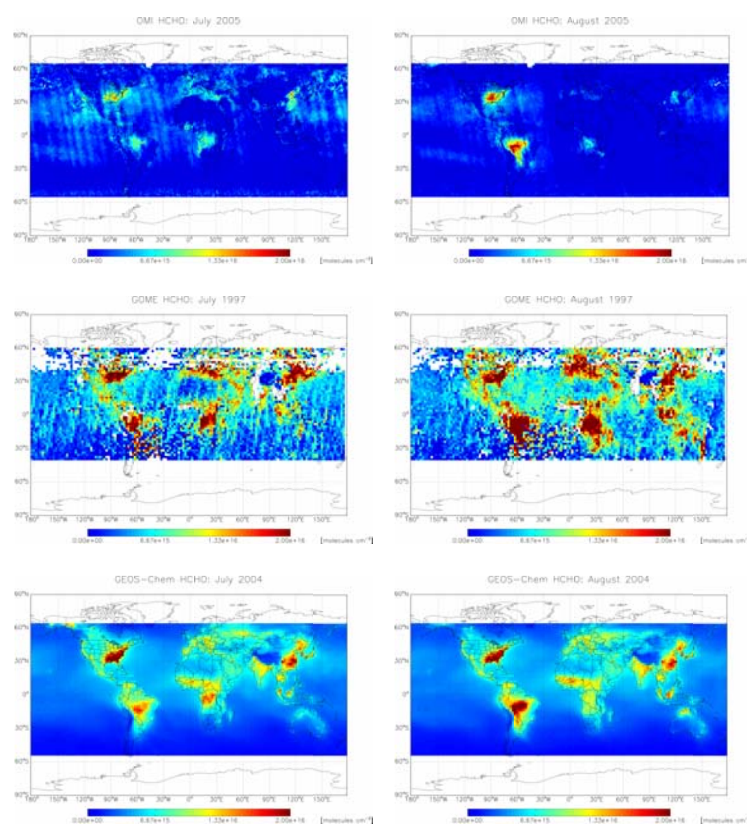

Figure 7. HCHO columns as measured by OMI and GOME, and simulated using GEOS-Chem, for July and August. Note different years for the three datasets.

How accurately we can infer isoprene emissions from HCHO column measurements made from space depends mainly on the retrieval errors and uncertainties in the HCHO yield. The HCHO yield calculated here has an estimated uncertainty of 30%, similar to the differences between yields calculated using different chemical models. With the retrieval errors calculated above, this results in a 1σ uncertainty in isoprene emissions derived from satellite measurements of HCHO columns of 40%. This compares favorably errors associated with extrapolating leaf and plant-level emission data. The overall approach therefore offers a useful and independent means of inferring surface emissions of isoprene.