

Seasonal and interannual variability of North American isoprene emissions as determined by formaldehyde column measurements from space

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[1] Formaldehyde (HCHO) columns measured from space by solar UV backscatter allow mapping of reactive hydrocarbon emissions. The principal contributor to these emissions during the growing season is the biogenic hydrocarbon isoprene, which is of great importance for driving regional and global tropospheric chemistry. We present seven years (1995–2001) of HCHO column data for North America from the Global Ozone Monitoring Experiment (GOME), and show that the general seasonal and interannual variability of these data is consistent with knowledge of isoprene emission. There are some significant regional discrepancies with the seasonal patterns predicted from current isoprene emission models, and we suggest that these may reflect flaws in the models. The interannual variability of HCHO columns observed by GOME appears to follow the interannual variability of surface temperature, as expected from current isoprene emission models. *INDEX TERMS:* 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry. *Citation:* Abbot, D. S., P. I. Palmer, R. V. Martin, K. V. Chance, D. J. Jacob, and A. Guenther, Seasonal and interannual variability of North American isoprene emissions as determined by formaldehyde column measurements from space, *Geophys. Res. Lett.*, 30(17), 1886, doi:10.1029/2003GL017336, 2003.

[2] Isoprene ($\text{CH}_2=\text{CH}-\text{C}(\text{CH}_3)=\text{CH}_2$), a biogenic hydrocarbon whose main source is terrestrial vegetation [Guenther *et al.*, 1995], plays an important role in the production of tropospheric ozone. Isoprene emission depends largely on vegetation type, vegetation density (leaf area index), leaf age, light, and temperature [Guenther *et al.*, 2000; Geron *et al.*, 2000]. Current inventories of isoprene emission use functional dependences of these parameters that are extrapolated from limited isoprene flux measurements. Large uncertainties are associated with this approach [Pierce *et al.*, 1998]. Total global emission estimates of isoprene used in atmospheric chemistry models vary by at

least a factor of 2 [Guenther *et al.*, 1999] while the variation in regional emission estimates can be much larger.

[3] Satellite observations of formaldehyde (HCHO) columns from solar UV backscatter offer top-down constraints to reduce uncertainty in isoprene emission inventories [Palmer *et al.*, 2003]. The HCHO column enhancement over that contributed by the methane background is due to oxidation of reactive organic gases. Over North America during the growing season, isoprene is the main regional contributor to this HCHO enhancement [Lee *et al.*, 1998; Sumner *et al.*, 2001]. The lifetime of isoprene against atmospheric oxidation is typically less than one hour, and the lifetime of HCHO is a few hours, so that isoprene emission is collocated with and linearly related to the resulting HCHO enhancement over a smearing length scale of order 10–100 km [Palmer *et al.*, 2003]. Biogenic terpenes are thought to contribute little to HCHO columns because their HCHO yield per unit carbon is low [Orlando *et al.*, 2000]. We showed previously that HCHO column observations from the Global Ozone Monitoring Experiment (GOME) satellite instrument [Burrows *et al.*, 1999] over North America during July 1996 are consistent with *in situ* HCHO data and with the expected distribution of isoprene emission [Palmer *et al.*, 2003]. Biomass burning is also a major source of enhanced HCHO observed by GOME [Thomas *et al.*, 1998] but this source is not important over North America except for occasional wildfires. In this paper we further demonstrate the utility of HCHO column observations from GOME for mapping isoprene emission by examining the seasonal and interannual variability of these data over North America and comparing it to current understanding of isoprene emission patterns.

[4] The GOME instrument measures backscattered solar radiation (237–794 nm) in the nadir with a $320 \times 40 \text{ km}^2$ field of view. Global coverage is achieved in 3 days. “Slant” column HCHO along the line of sight is calculated from GOME spectra in the 336–356 nm wavelength region with a mean fitting uncertainty of $4 \times 10^{15} \text{ molec cm}^{-2}$ [Chance *et al.*, 2000]. Air mass factors (AMFs), used to convert slant to vertical columns, are calculated with the LIDORT radiative transfer model [Spurr, 2002] using local relative vertical profiles of HCHO concentration (shape factors) from the GEOS-CHEM chemical transport model, and accounting for aerosol and cloud effects as well as Rayleigh scattering [Palmer *et al.*, 2001; Martin *et al.*, 2002; Martin *et al.*, 2003a]. Using GOME cloud information [Kurosui *et al.*, 1999] we exclude observation scenes with cloud or snow cover fraction >40%; these represent about 40% of the total data over North America during the summer.

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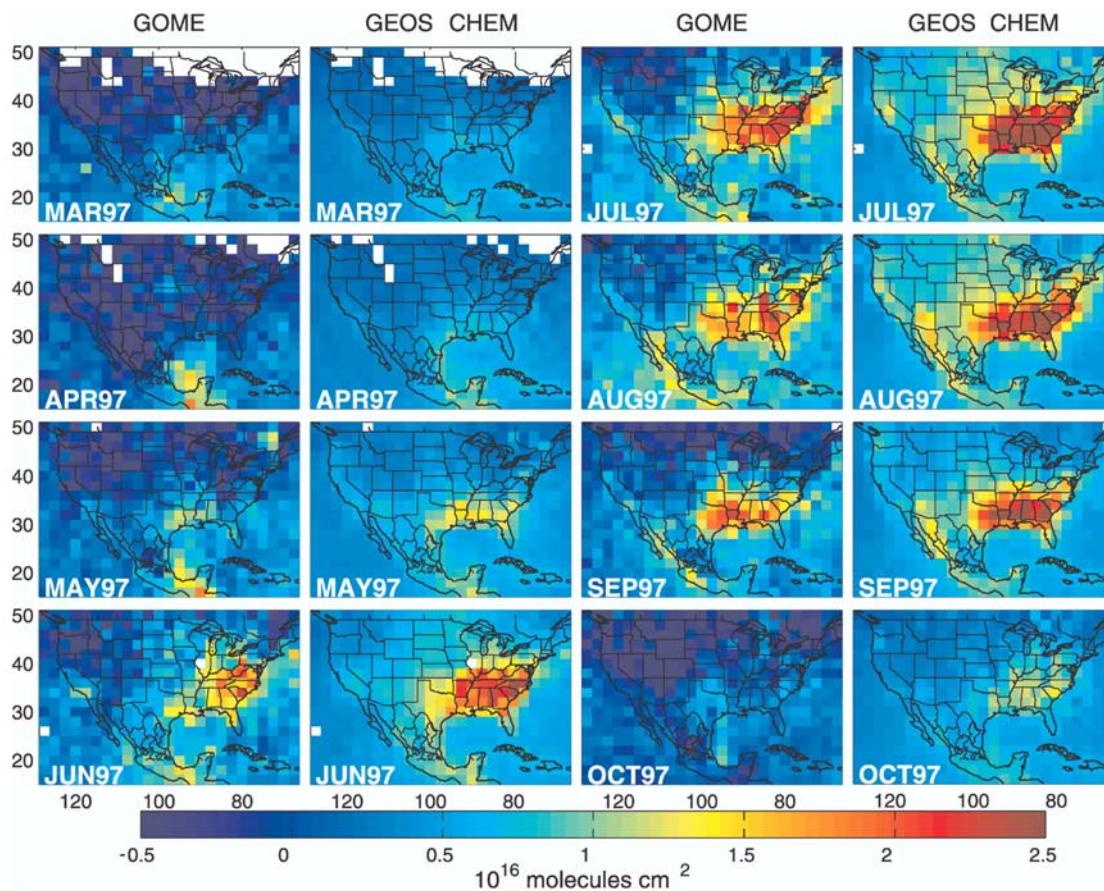


Figure 1. Monthly-mean HCHO columns (10^{16} molec cm^{-2}) over North America for March–October 1997. Data from the GOME satellite instrument (left) are compared to the GEOS-CHEM chemical transport model (right) sampled along the GOME orbit tracks at the GOME observation time. The GOME data have been mapped onto the GEOS-CHEM $2 \times 2.5^\circ$ model grid. The color scale is capped at 2.5×10^{16} molec cm^{-2} ; values as high as 3×10^{16} molec cm^{-2} are found over the southeastern United States in GEOS-CHEM in July and August. Negative HCHO columns in the GOME data are typically within the 4×10^{15} molec cm^{-2} fitting uncertainty [Chance *et al.*, 2000]. Values for November–February are generally below the GOME fitting uncertainty, both in the model and the observations, and are not included here. We exclude observation scenes with cloud or snow cover fraction $>40\%$ [Kurosu *et al.*, 1999]. White areas denote regions with persistent cloud or snow cover fraction greater than this cut-off value.

[5] The GOME diffuser plate, used to attenuate solar radiation, introduces a global uniform bias in the retrieved slant column that varies with solar day and has no apparent seasonal or interannual pattern [Richter and Wagner, 2001; Martin *et al.*, 2002]. There is also a zonally symmetric and seasonally varying pattern in the slant columns that we do not understand which results in slant columns over remote oceans that exceed the expected background due to methane oxidation by values approaching 1.0×10^{16} molec cm^{-2} at high latitudes in summer. We quantify these biases as the daily difference between GEOS-CHEM and GOME background HCHO columns over the central North Pacific as a function of latitude [Martin *et al.*, 2002; Palmer *et al.*, 2003], and subtract them from the results presented here.

[6] Figure 1 shows the monthly-mean GOME HCHO columns over North America during March–October 1997. Daily cloud data from GOME and HCHO shape factors for each observation scene were used in the AMF calculation to convert slant to vertical columns. We find a strong seasonal variation in HCHO columns correlated with the growing season and with high summertime temperatures. The

GOME HCHO columns are highest in the southeastern United States, consistent with the known distribution of isoprene emissions [Guenther *et al.*, 2000]. Also shown in Figure 1 are monthly mean HCHO columns from the GEOS-CHEM chemical transport model [Bey *et al.*, 2001], which uses isoprene emissions from the Global Emission Inventory Activity (GEIA) [Guenther *et al.*, 1995]. We use here GEOS-CHEM version 4.26 with additional model developments described in Martin *et al.* [2003b]. The model is broadly consistent with the observations in terms of both spatial and seasonal variations. It is biased high by 20% on average. Improvements in isoprene emission modeling have been made since the GEIA inventory, notably the accounting for the effect of leaf age and antecedent temperatures [Guenther *et al.*, 1999; Geron *et al.*, 2000]. These improvements will be incorporated into the model in future work.

[7] Outside of the growing season, observed and modeled HCHO columns are generally below the detection limit of 4×10^{15} molec cm^{-2} . The low winter values in the model reflect the relatively low emissions and long lifetimes of

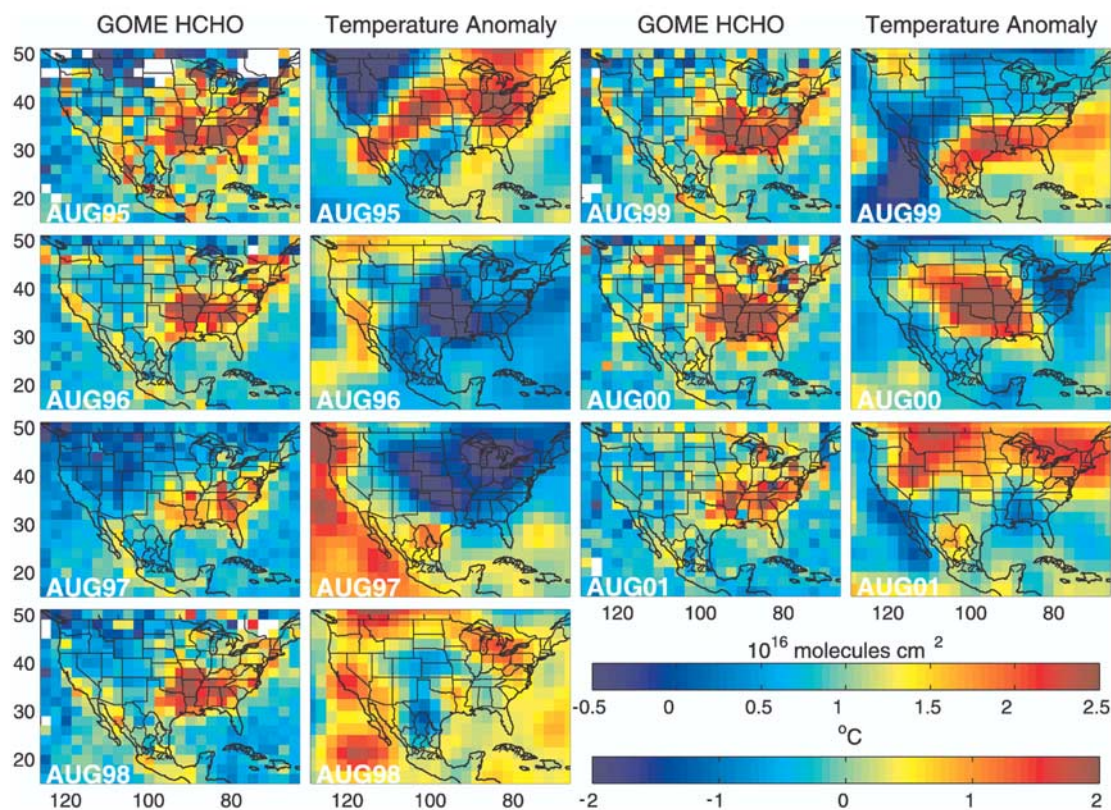


Figure 2. Interannual variability of August monthly mean HCHO columns (10^{16} molec cm^{-2}) from GOME over the 1995–2001 period (left), and corresponding anomalies in monthly mean surface air temperature ($^{\circ}\text{C}$) relative to the seven-year record (right). Surface air temperatures are from the National Center for Environmental Prediction [Kalnay *et al.*, 1996]. Air mass factors computed for August 1997 are applied to other years, so that the interannual variability in the GOME data is driven solely by the slant columns fitted from the observed radiances. We exclude observation scenes with cloud fraction $>40\%$ [Kurosu *et al.*, 1999]. The color scale of the GOME HCHO columns is capped at 2.5×10^{16} molec cm^{-2} ; values as high as 4×10^{16} molec cm^{-2} occur in 1995 and as high as 3.2×10^{16} molec cm^{-2} occur in other years. Negative HCHO columns are typically within than the 4×10^{15} molec cm^{-2} fitting uncertainty [Chance *et al.*, 2000].

anthropogenic hydrocarbons. Large values over southern Mexico during April–May 1997 in the GOME data are due to agricultural fires; these fires are accounted for in the GEOS-CHEM simulation on the basis of satellite observations of firecounts [Duncan *et al.*, 2003], but with apparently insufficient hydrocarbon emissions. Outside of this fire period the GOME HCHO columns over southern Mexico are low, consistent with the GEIA isoprene emission inventory.

[8] Figure 2 shows the interannual variability of observed monthly mean HCHO columns for August 1995–2001, together with the corresponding surface air temperature anomalies. Monthly mean AMFs from August 1997 were used to convert slant to vertical columns in other years. Comparison of monthly mean local AMFs over North America for September 1996 and 1997 suggests that interannual variation is generally within 10% so that the interannual variability in the GOME data is driven mainly by the slant columns fitted from the observed radiances. Cloud data specific to each GOME observation scene [Kurosu *et al.*, 1999] was used to exclude scenes with cloud or snow fraction $>40\%$. The general spatial pattern of HCHO columns from GOME in Figure 2 is reproduced from year to year. There is a 30% interannual variability over the eastern United States that appears to follow roughly variations in

surface air temperature. The HCHO columns during August 1997 are about 15% lower than average over the southeastern United States, and the corresponding negative temperature anomaly is about 1 K, consistent with the $\exp[(0.12 \text{ K}^{-1})T]$ temperature dependence of isoprene emission in the temperature range of interest [Guenther *et al.*, 1995]. The hottest August over the southeastern United States in the GOME record was in 2000, and HCHO columns for that month exceed 3.0×10^{16} molec cm^{-2} . The HCHO columns in August 2000 also show high values over the northwestern United States due to large wildfires.

[9] Not all of the interannual variability of GOME HCHO columns in Figure 2 can be explained by temperature (e.g., August 1996). Additional factors that might contribute to interannual variability of isoprene emissions include fires, water availability, and tree disease. Preliminary examination of satellite observations of firecount and drought indices did not reveal an obvious role for these factors but further investigation is needed.

[10] We have shown in this paper that satellite observations of the HCHO column from GOME are broadly consistent with our understanding of the seasonal and interannual variability of isoprene emission. This demonstrates the considerable potential for constraining isoprene emission inventories using satellite observations and im-

proving our understanding of the factors controlling these emissions. Extending our analysis to the rest of the world is clearly desirable but will require careful separation of biomass burning and biogenic contributions to HCHO. Uncertainty in the HCHO yields from isoprene oxidation under low-NO_x conditions [Palmer *et al.*, 2003], as frequently encountered in the tropics, is a key limitation that will need to be addressed through laboratory studies. Better understanding is also needed of the HCHO yields from oxidation of terpenes, sesquiterpenes, and other highly reactive organic compounds emitted by vegetation.

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