INVERTING GOME FORMALDEHYDE COLUMN FOR BIOGENIC EMISSIONS OVER EUROPE

Gabriele Curci[1], Paul I. Palmer[2], Tzung-May Fu[2], Guido Visconti[1]

[1] Dipartimento di Fisica - CETEMPS, Università degli Studi dell'Aquila, via Vetoio 67010 Coppito – L'Aquila, Italy. [2]Atmospheric Chemistry Modelling Group, Division of Engineering and Applied Sciences, Harvard University, Pierce Hall, 29 Oxford Street, Cambridge, MA 02138.

e-mail: gabriele.curci@aquila.infn.it -- Tel: +39 0862 433087 -- Fax: +39 0862 433089 -- home-page: http://bobdylan.aquila.infn.it/Gabriele

U. L'Aquila

CETEMPS
cetemps.aquila.infn.it

Abstract

We analyse formaldehyde (HCHO) column data retrieved from GOME over the European continent. Formaldehyde is an important intermediate oxidation product of VOCs of anthropogenic and biogenic origin. We compare GOME HCHO column with model simulations focusing on the European continent. Our results suggest that the formaldehyde column over Europe is generally overestimated by the state-of-the-art chemistry and transport model GEOS-CHEM with respect to GOME column (Figure 1), possibly indicating biases in model VOC emissions.

We apply the method described by *Palmer et al.* [1] to constrain isoprene (a major biogenic VOC) emissions with HCHO column over Europe (Figure 3). We first apply the inversion only where the biogenic emissions are predominant over anthropogenic emissions and a clear relationship between HCHO column and isoprene emissions can be established. This is the case over Eastern Europe and other limited regions of Western Europe. Our updated "top-down" isoprene inventory in general revises low the emissions from the GEIA "bottom-up" inventory and is consistent with the new MEGAN biogenic emission inventory [2].

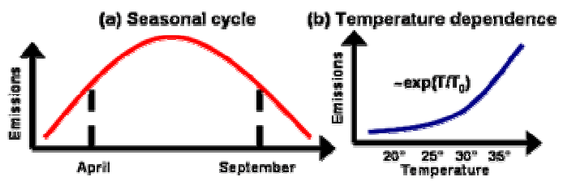
1. European Biogenic Emissions

Hydrocarbons other than methane are collectively called Volatile Organic Compounds (VOCs) in atmospheric chemistry studies. Natural sources due to biological activity contribute globally with 70% to VOC emissions, as opposed to 30% of anthropogenic plus biomass burning contribution. Europe is the only continent where anthropogenic VOC emissions overweigh biogenic. However, biogenic VOCs (BVOCs) can still play an important role with respect to ozone production locally (e.g. Mediterranean area) [3]. Current biogenic emission inventories are highly uncertain in Europe (up to a factor of 5 [3]) due to lack of data and human induced changes to landuse. We apply a top-down approach to estimate isoprene (major BVOC) emissions from satellite observations trying to reduce this uncertainty.

Biogenic emissions are typically modeled using base gridded emission fluxes E_0 corrected for environmental conditions [2]:

$$E = E_0 \, \gamma_{LAI} \, \gamma_T \, \gamma_{AGE} \, \gamma_{PAR} \qquad (1)$$

where γ_{LAI} (Leaf Area Index) correction accounts for vegetation density, γ_T is the correction exponentially dependent on temperature, γ_{AGE} accounts for the foliar age dependent emission efficiency, and γ_{PAR} (Photosynthetically Active Radiation) accounts for different illumination at different canopy levels. Equation (1) is fitted to observed data to match the main features of biogenic emissions, as schematically shown below: (a) seasonal cycle with null emissions in winter and a peak in summer and (b) exponential dependence on temperature



Major biogenic emission sources in Europe are forested areas in Mediterranean region and North-Eastern Europe and wetlands in Scandinavia and Russia.

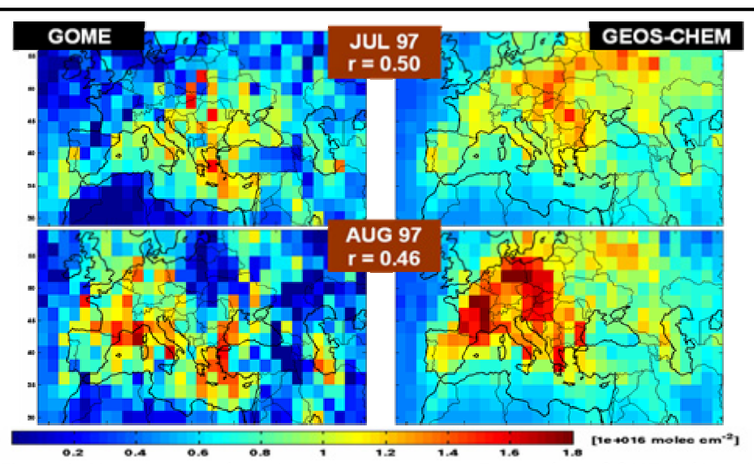


Figure 1: Monthly mean formaldehyde column over Europe from GOME (left) and GEOS-CHEM model (right) for July (top) and August (bottom) 1997. The correlation coefficients between observation and simulation are shown inset.

2. HCHO column from GOME

The GOME instrument onboard ERS-2 satellite measured HCHO columns in 1995-2001. Here we use the retrieved column product by *Chance et al.* [4] for summer 1997. Slant columns are converted to vertical columns by the Air Mass Factor (AMF) formulation by *Palmer et al.* [5] that accounts for shape of the HCHO vertical profile and scattering processes from aerosol and clouds, in addition to Rayleigh scattering. We use GOME HCHO column here as a proxy for VOC emissions (see below).

In Figure 1 we compare the monthly mean HCHO column over Europe observed by GOME and simulated by the GEOS-CHEM model. Cloud scenes with cloud fraction >40% are filtered out. The model generally overestimates the HCHO column with respect to GOME except in Central Europe and the Mediterranean area. It is especially striking the difference over Russia, where the model simulates a much higher column.

In Figure 2 we show the seasonal cycle of HCHO column over Europe as seen by GOME. A clear cycle with a peak in summer is visible over much of the European territory, indicating biogenic emissions as a main driver of the HCHO column seasonal variability. An exception is the industrialized area in Northern Europe, where anthropogenic VOCs play an important role in controlling the HCHO column.

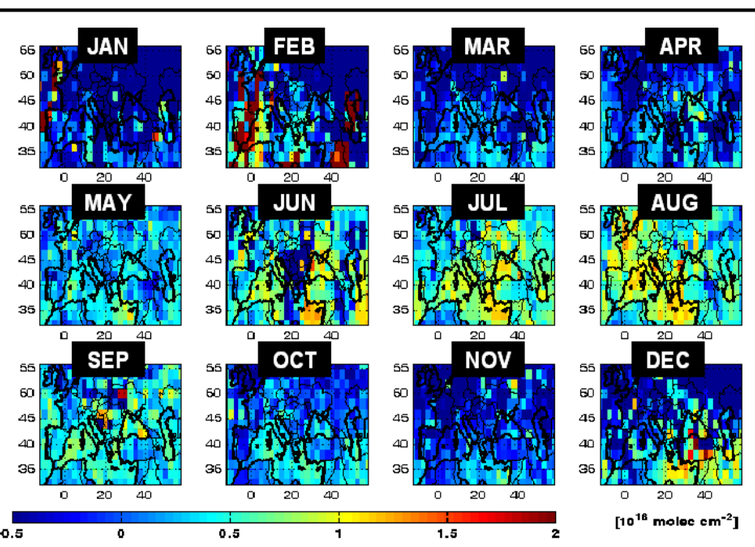


Figure 2: Monthly mean formaldehyde columns over Europe from GOME for the years 1996-97.

3. Estimating VOC emissions from HCHO observations

In the absence of transport HCHO column Ω at steady-state is linearly related to the sum of the underlying VOC emissions E_i scaled by their HCHO yields Y_i:

$$\Omega = \sum Y_i E_i \; / \; k_{HCHO} \qquad (2)$$

where k_{HCHO} is the loss rate constant for HCHO. Horizontal transport smears this local relationship spatially to a degree that depends on the wind speed and the time lag between VOC emission and HCHO production. Isoprene is highly reactive and the time lag is only a few hours, and the smearing length is <100 km, of the same order of the GOME pixel (40x320 km^2). We thus neglect the effect of transport in inverting Ω for isoprene emissions.

Under this assumption *Palmer et al.* [1] used local linear relationship between HCHO columns and isoprene emissions derived from the GEOS-CHEM CTM model [5] to infer local isoprene emissions from GOME HCHO over North America:

$$\Omega = S \, E_{isop} + B \qquad (3)$$

where S denotes the linear slope, E_{ISOP} is the local isoprene emission and B denotes a background HCHO determined by the oxidation of the other, generally, longer-lived VOCs.

We extend the method of *Palmer et al.* [1] here to Europe. From previous studies the VOC chemistry over the North American territory is known to be controlled by biogenic emissions, in particular in the South Eastern United States. This is not generally the case in Europe. We choose to apply the inversion for isoprene emission first in a region where the biogenic influence is demonstrated to be predominant.

We check for regions under biogenic control looking at the $HCHO/NO_2$ column ratio using observations of GOME. As shown by *Martin et al.* [6] this ratio could be used as a proxy for VOC/NO_x ratio into the lower layers of the troposphere. A high VOC/NO_x ratio implies that we are in a rural region [3]. Thus we look for a rural region that we also know to be forested. We found one such region in the Eastern part of Europe (see plot (a) of Figure 3) and we first apply the inversion of HCHO column for isoprene emission there.

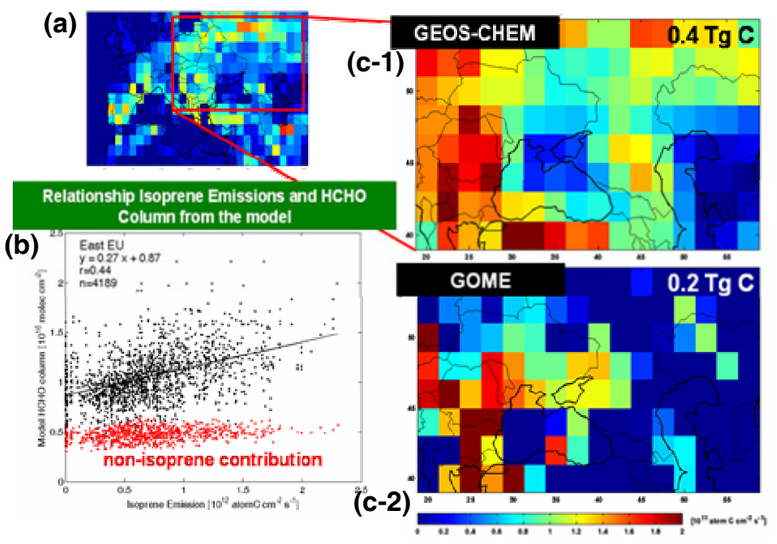


Figure 3: (a) Rural and forested region where we apply the inversion; (b) local linear relationship between isoprene emissions and HCHO columns from the model in region (a) (see equation (3)); (c-1) GEIA isoprene emissions implemented into the GEOS-CHEM model in region (a); (c-2) updated isoprene emissions deduced from GOME HCHO column for July 1997.

In plot (b) of Figure 3 we show the relationship deduced from model simulation between isoprene emissions and HCHO columns. The GEOS-CHEM model used here implements the GEIA inventory for biogenic emissions. The black points on the scatter plot are model values sampled at GOME pixel locations and are averages between 10-12am local time (GOME overpass hours). Cloud scenes with cloud fraction >40% are excluded. Slope, background values of the corresponding equation (3) and correlation coefficient are shown inset. The red dots on the plot show results from a run with null isoprene emissions. From the comparison with the standard run it is clear how the HCHO column in the model is largely controlled by isoprene emissions in this region.

In the two plots on the right we compare the isoprene emission distribution in GEIA inventory and as deduced from GOME HCHO observations. GOME results suggest a reduction of isoprene emissions in the upper and right part of the selected domain. The overall budget of isoprene emissions for the selected month (July 97) over the whole area is halved with respect to GEIA. This is also consistent with the recently released biogenic emission inventory MEGAN [2] that revises low the isoprene emission in this same region (not shown in this poster).

Future work will extend the method also to other European countries after a careful estimation of the anthropogenic contribution to HCHO column. The resulting updated isoprene emission inventory will be implemented into the model and will be evaluated against independent measurements of HCHO.

References

[1] Palmer et al., JGR 2003.

[2] Guenther et al., submitted to JGR 2005.

[3] Simpson et al., JGR 1999.

[4] Chance et al., GRL 2000.

[5] Palmer et al. JGR 2001.

[6] Martin et al., GRL 2004.

For further information on the GEOS-CHEM model please check on the Harvard group's web site:

http://www-as.harvard.edu/chemistry/trop/