MODEL SIMULATIONS OF FORMALDEHYDE COMPARED WITH GOME OBSERVATIONS

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Introduction

Formaldehyde (HCHO) is an **important indicators of hydrocarbon emissions and photochemical activity**

HCHO sources

- oxidation of Methane provides constant HCHO source; tropospheric NMHC emissions, biomass burning, fossil fuel combustion

HCHO sinks

- reaction with OH, photolysis

→ HCHO is a good test for model oxidation mechanism and emission scenarios

→ HCHO could be used as proxy for biogenic emissions (isoprene and monoterpene)

Experiment

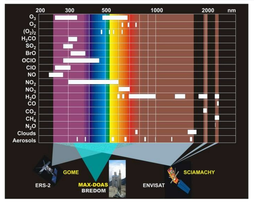

Fig 1: Spectral coverage of GOME and other UV/vis instruments.

GOME instrument

- launch in April 1995 aboard ERS-2
- on sun-synchronous orbit
- global coverage within 3 days
- spatial resolution 320x40 km^2
- global data from July 1995 to March 2003

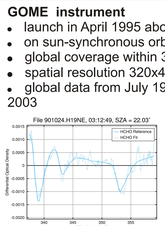

***Fig 2:** Example of the laboratory (thick line) and observed (dotted line) DOAS fit spectrum for formaldehyde (HCHO) using one GOME measurement.*

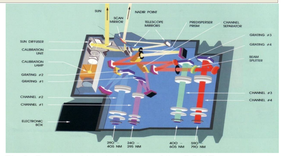

Fig 3: Functional block diagram of the GOME spectrometer optical system [ESA, 1995].

Data retrieval

- Differential Optical Absorption Spectroscopy (**DOAS**) yields slant columns = averaged absorption along all contributing light paths
- DOAS fitting window: 337.5 to 359 nm, daily solar irradiance as background (see Fig. 2)
- conversion to vertical columns using air mass factors (AMF) calculated by radiative transfer model **SCIATRAN** (Rozanov et al.)
- constant background between 200 and 220°E near to the equator assumed (normalisation) to account for instrumental drifts/inhomogenities - lookup table for AMF taking into account albedo, orography, aerosol and trace gas profile shape (in total 48.000 scenarios) using external information
- only GOME data with cloud fraction less than 20 percent have been used in this study

Selected References / Other RETRO Contributions to this Conference

- D. Hauglustaine et al., A MULTI-MODEL SIMULATION OF THE TROPOSPHERIC COMPOSITION OVER THE LAST 40 YEARS: oral A-09464
- S. Rast et al., MULTIMODEL ANALYSIS OF THE SENSITIVTY [...] TO EMISSIONS: oral A-04450
- S. Rast et al., ANALYSIS OF TRENDS [...] SIMULATED WITH MOZECH: poster A-04463
- S. Szopa et al., TRENDS AND VARIABILITY OF TROPOSPHERIC OZONE [...] OVER EUROPE [...]: poster A-03926
- N. Savage et al., STUDIES OF SEASONAL CYCLES AND SENSITIVITY [...]: poster A-04221
- T. van Noije et al., MODEI SIMULATIONS OF TROPOPHERIC NO2 COMPARED WITH GOME RETRIVALS [...]: poster A-08247
- F. Wittrock et al., GLOBAL OBSERVATIONS OF FORMALDEHYDE AND GLYOXAL WITH [...] UV/VIS INSTRUMENTS: poster A-08191
- A. Vik et al., THE RETRO DATABASE FOR OBSERVATIONS: poster A-07737

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- Wittrock, F. et al. (2006c), Global observations of Formaldehyde and comparison to model simulations, manuscript in preparation.

Model vs GOME

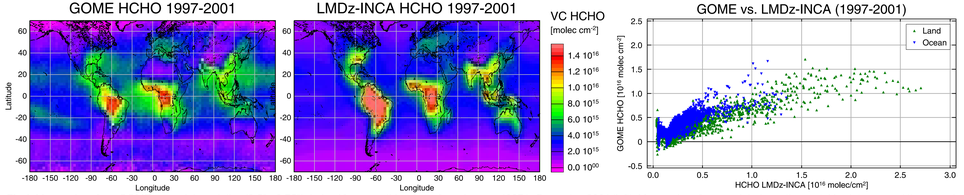
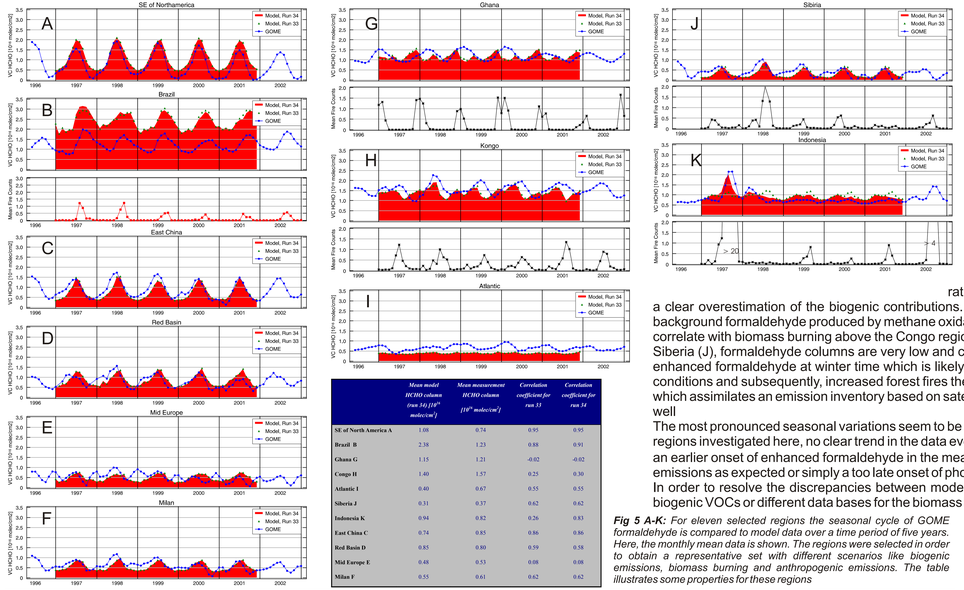


Fig 4: Mean values from GOME on model resolution (2.5° x 3.75°) derived from monthly means from January 1997 to December 2001 (left). Mean values from model – here, the run taking into account the annual variability of biomass burning was utilised (run 34) (middle). The scatter plot (right) shows the correlation between GOME observation and the Model results. The correlation coefficient is 0.81 overall, while it is for the continents 0.89 and the oceans 0.83, respectively. The slope is 0.64 for the continents and 1.05 for the ocean.



	Mean model HCHO column (run 34) [10^{16} molec/cm^2]	*Mean measurement HCHO column [10^{16} molec/cm^2]*	*Correlation coefficient for run 33*	*Correlation coefficient for run 34*
SE of North America A	1.08	0.74	0.95	0.95
Brazil B	2.38	1.23	0.88	0.91
Ghana G	1.15	1.21	-0.02	-0.02
Congo H	1.40	1.57	0.25	0.30
Atlantic I	0.40	0.67	0.55	0.55
Siberia J	0.31	0.37	0.62	0.62
Indonesia K	0.94	0.82	0.26	0.83
East China C	0.74	0.85	0.86	0.86
Red Basin D	0.85	0.80	0.59	0.58
Mid Europe E	0.48	0.53	0.08	0.08
Milan F	0.55	0.61	0.62	0.62

***Fig 5 A-K:** For eleven selected regions the seasonal cycle of GOME formaldehyde is compared to model data over a time period of five years. Here, the monthly mean data is shown. The regions were selected in order to obtain a representative set with different scenarios like biogenic emissions, biomass burning and anthropogenic emissions. The table illustrates some properties for these regions*

Model Description

Several global chemistry-transport models (TM5, LMDz, CTM2, MOZECH, p-TOMCAT) recently performed a multiannual run as part of the European project RETRO (REanalysis of the TROpospheric chemical composition over the past 40 years - http://retro.enes.org). All models apply the same anthropogenic, biogenic and biomass burning emissions of ozone precursor gases. The formaldehyde fields from the models are analysed daily at 10:30 local time, close to the overpass time of the satellite. Because the model runs become available very recently this study shows only results with an older run of the LMDz-INCA model, which is characterized in the following:

- LMDz (Laboratoire de Meteorologie Dynamique, zoom) is a grid point General Circulation Model (GCM)
- horizontal resolution of 3.75 degrees in longitude and 2.5 degrees in latitude, 19 vertical sigma-p levels extending from the surface to 3 hPa
- Interactive Chemistry and Aerosols (INCA) model integrated into LMDz
- INCA simulates tropospheric chemistry, emissions and deposition of primary tropospheric trace species including non-methane hydrocarbons
- INCA chemical scheme includes 85 chemical species and 303 chemical reactions, details in Hauglustaine et al., [2004] and Folberth et al., [2004].
- The simulation used in this study was performed using the nudged version for meteorology (i.e. meteorological fields are relaxed toward the ERA40 reanalysis). Regarding the emissions, anthropogenic emissions are from the EDGAR V2.0 inventory (Olivier et al. 1996), biomass burning emissions are those of Van der Werf et al. (2003 and 2004), biogenic emissions are from GEIA and aircraft emissions are provided by the NASA. The output is for the time of the GOME overpass.Two different runs were carried out: one with a static scenario for biomass burning (run 33), the other one considering the interannual variability of biomass burning (run 34).

Results

The oak forests in the so-called Ozarks of south eastern North America (A) are efficient emitters of isoprene . Depending on light intensity as well as growing season (May to August) formaldehyde production is expected to be largest in summer which is reflected by measurement and model both in magnitude and in timing. In Brazil (B) the amplitude and also the time of maximum formaldehyde of the seasonal variation is also reproduced nicely by the model.The maximum formaldehyde correlates with the burning season in this region in summer. However, over the time period of five years the average model column is almost a factor of two larger. The year-round background values are caused by biogenic emissions which seem to be significantly overestimated in the model. For the measurements, cloud contamination over the rain forest could be a reason for a systematic underestimation, but a study on the dependence of HCHO columns on cloud fraction does not give any indication for such an effect. This point will have to be revisited once satellite measurements with higher spatial resolution and thus less cloud problems become available, e.g. OMI. G and H illustrates the situation in Africa: Due to the ITCZ, the dry season and consequently the burning season are shifted in time in the two selected regions of Ghana and Congo. The measurements of formaldehyde display a seasonal cycle with a maximum at the time of biomass burning. Although the amplitude of model and measurement of this seasonal cycle as well as the average column agree well, the phase of the measurements is completely missed and rather exhibits a double maximum indicating problems in the parameterisation of the biomass burning events in the model for both regions and a clear overestimation of the biogenic contributions. In addition to the continental regions, also an area over the Atlantic close to Africa was selected (I): Here, the model only shows the background formaldehyde produced by methane oxidation whereas GOME sees on average a 60 percent higher column. It is notable, that the small seasonal variation above the ocean does not correlate with biomass burning above the Congo region. This might be a hint that rather biogenic emissions than plumes of biomass burning cause the elevated levels of HCHO above water. In Siberia (J), formaldehyde columns are very low and close to the detection limit. Although a correlation with the fire counts can be observed, in particular in 1998, the measurements also show enhanced formaldehyde at winter time which is likely caused by albedo artefacts similar to e.g. Mid Europe (E). Indonesia (K) is a region that is influenced by ENSO effects resulting in drier conditions and subsequently, increased forest fires there. In the years 1997 and 2002 El Nino events lasted for several months causing high formaldehyde columns. Ergo, only the model run 34 which assimilates an emission inventory based on satellite fire counts is able to reproduce the enhanced formaldehyde in 1997. For both regions, the overall magnitude of HCHO is captured quite well

The most pronounced seasonal variations seem to be connected to regions with a distinct growing season in summer as in south eastern North America (A) and eastern China (C). There is, for all regions investigated here, no clear trend in the data even in those areas (East China) where this was clearly identified for NO_2 (Richter et al., *Nature* 2005). Regions with biogenic emissions reveal an earlier onset of enhanced formaldehyde in the measurements compared to the model in the growing season. Reasons for this could be either an earlier start in the year of biogenic isoprene emissions as expected or simply a too late onset of photochemical production of formaldehyde in the model.

In order to resolve the discrepancies between model runs and GOME observations, case studies are necessary where single parameters like emission inventories of isoprenes and other biogenic VOCs or different data bases for the biomass burning events are investigated in more detail. This will be carried out in the near future as part of the RETRO project.

Conclusions

GOME measurements have been compared in detail with results from the LMDz-INCA model as part of the European project RETRO. Overall, excellent agreement was found over the continents, but significant underestimation in the model over the oceans. Here, the satellite measurements show clearly enhanced columns in some areas affected by continental outflow, indicating either a longer than modelled lifetime of HCHO or more probably in-situ production by decomposition of long-lived organic compounds. The seasonal variation of modelled HCHO columns shows excellent agreement with the measurements in some regions (North America), underestimation of the overall amplitude in South America and varying degrees of agreement over different parts of Africa and Asia, indicating model deficiencies in the parameterisation of the emission of biogenic precursors and their timing. In regions with strong biomass burning, the use of measurement-based fire distributions improves the agreement between model and observation as expected. A similar study using all models within RETRO is in preparation and will be published soon.

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