A Comparison of 3D Global CTM Results with Tropospheric Columns of NO_2 and HCHO Measured by GOME

A contribution to subproject GLOREAM

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Summary

Columns of NO_2 and formaldehyde measured using data from the GOME instrument are compared to the results of the chemical transport model TOMCAT. Generally good agreement is found for NO_2 while the modelled distribution of HCHO compares reasonably well for formaldehyde although there are differences in the absolute values. The importance of the method used to separate tropopause columns from the total and the effect of the satellite overpass time on measurements are also highlighted.

Introduction

Tropospheric ozone is an important greenhouse gas whose global average radiative forcing makes it the third most important greenhouse gas (IPCC, 2001). It is produced by photochemical reactions in which NO_2 and hydrocarbons play a central role. NO_2 has high temporal and spatial variability making it difficult to use the results of sparse in-situ and remote sensing measurements to test our understanding of its transport and chemistry. Therefore satellite measurements of NO_2 and other chemical species involved in ozone photochemistry such as formaldehyde (HCHO) may provide valuable insights for our understanding of ozone photochemistry on a global scale.

Objectives

Here we compare the results from a chemical transport model to measurements from the GOME instrument and investigate the sensitivity of the results to the methods used to compare model predictions to satellite data. The influence of the method used to separate tropospheric and stratospheric contributions to the NO_2 column and the effect of the diurnal variation of NO_2 concentrations are also considered.

Activities

The Global Ozone Monitoring Experiment (GOME) is a UV/visible spectrometer on board the European satellite ERS-2. GOME is a 4 channel double monochromator covering the wavelength range of 230 - 800 nm with a spectral resolution of 0.2 - 0.4 nm. ERS-2 was launched into a polar sun-synchronous orbit in April 1995. It passes over the equator at 10:30 local time. With a ground pixel size of 40 x 320 km^2 (40 x 960 km^2 for the back scan) GOME reaches global coverage at the equator within 3 days. The main objective of GOME is the global measurement of ozone columns, but other trace gases such as NO_2, and HCHO can be retrieved from the spectra as well (Burrows, 1999). In this study tropospheric columns of NO_2 (Richter and Burrows, 2001) and HCHO (Wittrock et al., 2000) have been evaluated using

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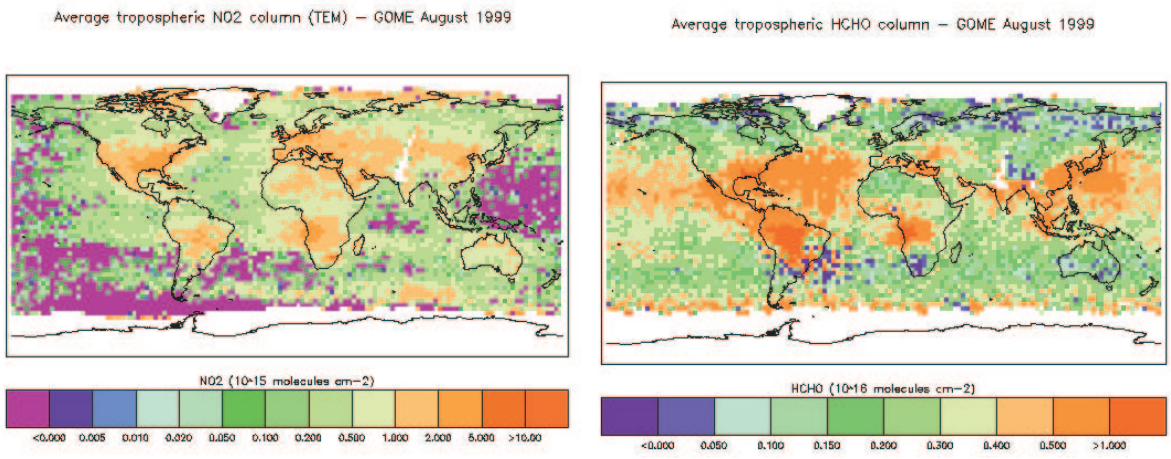
only cloud free pixels and gridded at the same resolution as the TOMCAT model. The tropospheric column of NO_2 was calculated by the tropospheric excess method (TEM). In this method a zonal mean of the vertical column of NO_2 over a clean area (in this case the Pacific region (180-190 degrees longitude band) is subtracted from all the data. This method assumes that over clean areas the tropospheric column is negligible and that there is little variation of the vertical column as a function of longitude.

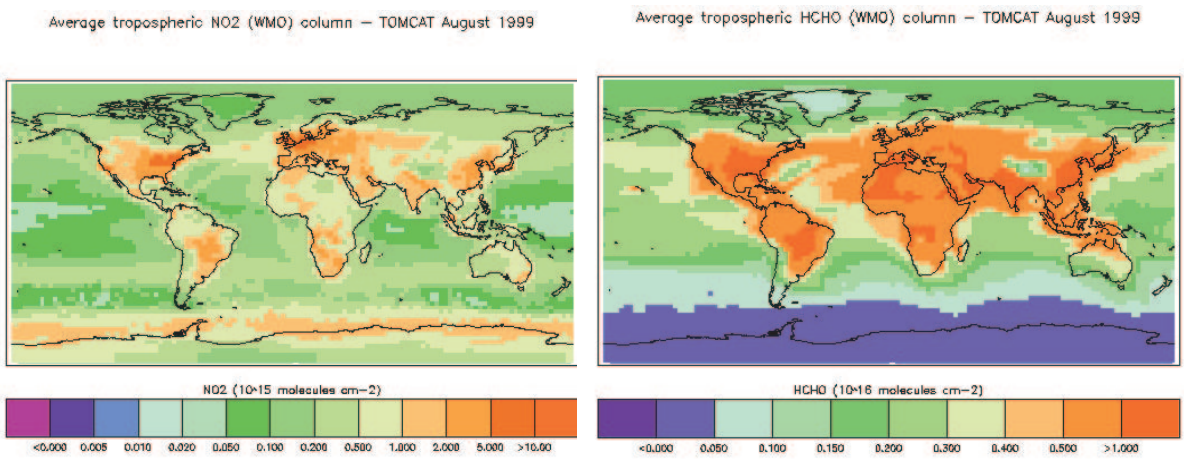
The global off-line grid point Chemistry Transport Model (CTM) TOMCAT (Law et al., 1998) was run for August 1999 using meteorological data from ECMWF. The model has a horizontal resolution of ~2.8 x 2.8 degrees (~310km x 310km) and 31 levels in the vertical from the ground to 10 hPa. The model contains 48 chemical species and uses a non-local vertical mixing scheme for the boundary layer (Holtslag and Boville, 1993) from which diffusion coefficients were also used to calculate dry deposition rates. The emission rates used are based on the IPCC Third Assessment Report. These were calculated from an extrapolation of the EDGARV2.0 to give emission rates for the year 2000. Concentrations of NO_2 and formaldehyde were output every timestep for those longitudes where the local time was 10:30 +/- 15 minutes to ensure a valid comparison with GOME results. Every six hours the global chemical fields were also output enabling a diurnal average to be calculated also. When calculating the tropospheric column two different methods were used. In the first (used only for NO_2) a similar procedure was used as for the GOME data (TEM). A zonal mean of the 180-190 degrees longitude band was subtracted from all the columns. In the second method the height of the tropopause was diagnosed using the WMO definition (a lapse rate of less than 2 ^{o}C/km) and the columns below this height were calculated.



Figures 1 and 2. Total tropospheric columns of NO_2 and HCHO as measured by GOME in August 1999.

Results

Areas which are white in these plots indicate areas where there are insufficient data to calculate a tropospheric column. It can be seen that for NO_2 the major areas in the Northern Hemisphere where high concentrations are observed are the East Coast of the USA, Western Europe and Eastern China which correspond to regions of high industrial emissions. In the Southern Hemisphere the high concentrations in South America and Southern Africa appear to correspond to biomass burning regions. Similar high concentration regions can be seen in the formaldehyde data however here it appears that the areas where there is biomass burning are more important than the industrialised areas.



Figures 3 and 4. TOMCAT model results for NO_2 and HCHO.

The tropospheric NO_2 columns shown here are determined in the same way as the GOME data shown above (TEM). The formaldehyde columns are found by summing the concentrations on all levels up to the WMO tropopause The comparison of the data for NO_2 is in general quite good. The highest concentrations of NO_2 are seen in the same places and are of the same order of magnitude in size. However the total columns over the Eastern USA and Western Europe do appear to be slightly higher in the model whereas concentrations in the more southerly latitudes are lower than those observed by GOME. Insufficient boundary layer mixing or too weak convection in the model may in part explain the large columns over industrialised areas in the Northern Hemisphere. Alternatively, the GOME data may be too low in some regions as a result of residual clouds that shield part of the troposphere from view (Richter and Burrows, 2001). TOMCAT appears to have lower concentrations over the oceans. Possible reasons for this include a lifetime for NO_2 which is too short, secondary sources of NO_x which are missing from the model or insufficient mixing of air from polluted to unpolluted regions.

The comparison for formaldehyde is not as close but large columns of HCHO are seen in the same areas in the two data sets with generally higher concentrations being seen in the TOMCAT results especially over polluted regions. This may indicate that the industrial emissions in the TOMCAT model are too high and need to be compared to emissions used by other models. The region of high concentrations over North America which is not seen in the GOME results also deserves further investigation.

In order to test the importance of the methods used to output and test the data two additional sets of data are presented here. Figure 5 is the NO_2 column calculated by summing the data to the tropopause. In Figure 6 the NO_2 column amounts are calculated from a diurnal average of the data as opposed to the data which were output at the appropriate local time.

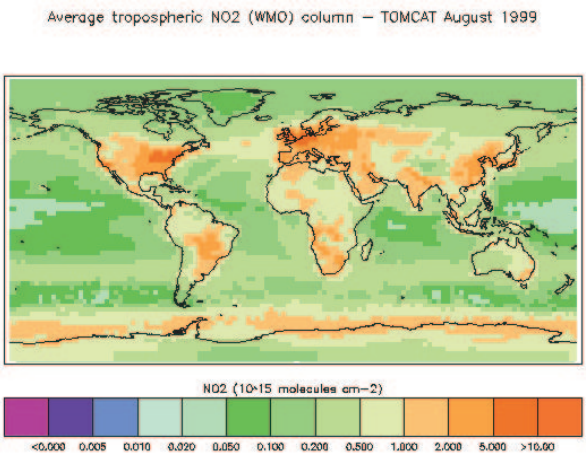


Figure 5. NO_2 column in TOMCAT up to the WMO tropopause.

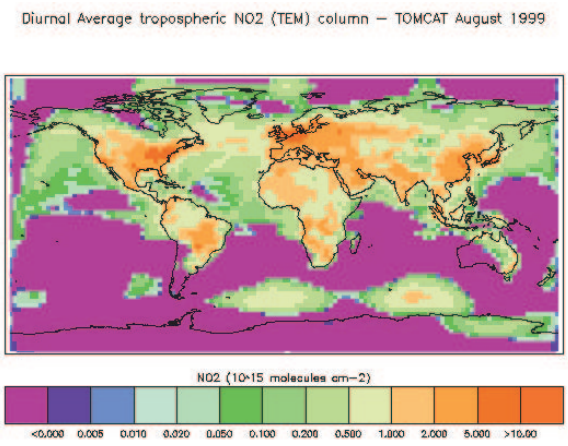


Figure 6. NO_2 column from a diurnal average.

The columns found in TOMCAT up to the WMO defined tropopause are not very different from those calculated by the TEM method over polluted regions. However over oceans the background concentrations are higher and more spatially smooth. The elevated concentrations at more southerly latitudes seen in the TEM results and the GOME results can also be seen but are more of a narrow band and are further south. This indicates the need to consider this feature in more detail by looking at height profiles in the model and examining how the use of the TEM method effects the interpretation of the results. These features may simply be a side effect of the method used to subtract stratospheric concentrations or could show descent of NO2 from the stratosphere. The diurnal average results are in broad agreement with the data output at the correct local time. However it can be seen that in certain areas such as the North Pacific region to the south of Alaska substantial increase in the total column (~0.5 x 10_{15} molecules cm^{-2}) are seen. This illustrates the importance of ensuring that the overpass time of GOME is accounted for in comparisons of models and data.

Conclusions

In general the satellite and model results compare well for NO_2 but TOMCAT gives larger concentrations of HCHO than GOME. The diurnal average concentrations of NO_2 are substantially different from those which were output at 10:30 in certain regions. The method used to calculate the tropospheric column in the model did not make a large difference to the results over polluted regions, however when columns up to the WMO tropopause were calculated in TOMCAT this gave higher and more spatially smooth concentrations over oceans.

Acknowledgements

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