The effect of updating reaction rate data on the tropospheric NO₂ column simulated by TM4 as compared to GOME retrievals for the year 2000

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Summary

Here we present a systematic comparison of tropospheric NO₂ columns for this study we have used the TM4 version applied in the recent Royal Society study on 'Ground-level ozone in the 21st Century: Evolution of the global atmospheric chemistry and its consequences for ozone levels and other air pollutants' (van Noije et al., 2006). As for the TM4 version, the model uses constant anthropogenic emissions from the Global Fire Emissions Database (GFED) and monthly emissions from biomass burning based on the 1997–2002 average carbon emissions from the Global Fire Emissions Database (GFEDv1). For comparison with GOME, we followed the procedure described by van Noije et al. (2006). Model output was analyzed at 10:30 local time, close to the overpass time of the ERS-2 satellite, and collocated with the measurements to account for sampling biases due to incoherent spatiotemporal coverage of the instrument. The 3-D concentration fields were converted to tropospheric columns by application of averaging kernels.

Update of reaction rate co-efficients

The TM4 model uses modified CB4 chemical mechanism of Houseau et al. (1999) for the description of the chemical evolution of the atmosphere. This contains a total of 16 photolysis rates, 67 gas-phase reaction rates and 4 reservoir species, for which the latest recommendations (Atkinson et al., 2004, 2006; Sander et al., 2006). This results in a substantial increase in the reservoir species representing organic nitrates and a substantial modification in NO₂, respectively. Biome-scale decompositions reveal that for regions influenced by a biomass burning season, this improved agreement between NO₂ columns from TM4 and GOME retrievals is partly due to the increase in the resident concentration of PAN and ORGNIT, which are important reservoir species for NO₂, and ultimately have an influence on the long-range transport of NO₂ away from the source regions. For the year 2000, the annual mean tropospheric NO₂ column density from the TM4 model with updated reaction rate co-efficients is shown in Figure 3.

Comparison with GOME

The seasonal cycle was analyzed in detail for different continental regions, six of which are shown in Figure 4 below. The regional difference introduced range between a 1.6% decrease in the tropospheric NO₂ column, although there is no difference for the region influenced by industrial sources (thus having low [NMHC]). For those regions dominated by strong NOx emissions there are reductions of ~10%. For regions dominated by strong NOx emissions there are reductions of ~10%

Regional analysis

We have analyzed the effect of updating reaction rate data has on the tropospheric NO₂ column calculated by the TM4 Chemical Transport Model. Figure 5 shows the seasonal cycle of tropospheric NO₂ column density for six different regions we have shown that for locations where the biomass burning season imposes a strong signature onto the NO₂ column, there is a corresponding increase in the concentrations and atmospheric lifetime of ORGNIT, an important reservoir species for NO₂, is thought to be the reason for the improvement in this region.

Future Work

The update to the TM4 model are still ongoing and the following work is planned in the near future which will have an impact on the simulation of tropospheric NO₂ columns:

• The introduction of an online routine for calculating tropospheric photolysis rates: this will allow the attenuation of light due to different aerosols which will affect JNO₂.
• The update of the photolysis frequencies of HCHO which will further perturb HOx chemistry.

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Figure 1. Annual mean tropospheric NO₂ column density for the year 2000 calculated by TM4 using the updated reaction rates. Percentage differences are shown against the distribution calculated using the original reaction rates.

Figure 2. Annual mean tropospheric NO₂ column density for the year 2000 calculated by TM4 using the updated reaction rates, together with the difference compared to the old schemes. Data have been smoothed to a horizontal resolution of 4° x 5°.

Figure 3. Annual mean tropospheric NO₂ column density averaged over the three GOME retrievals together with the difference between the TM4 model results using the updated reaction rates and the retrieval average. Data have been smoothed to a horizontal resolution of 4° x 5°.

Figure 4. Seasonal cycle in the tropospheric NO₂ column density for six different regions as calculated using three different retrieval algorithms. Comparisons are made against the results of simulations using the TM4 model with both the original and updated reaction rate data.

Figure 5. Seasonal cycle in the tropospheric NO₂ column density for six different regions as calculated using three different retrieval algorithms. Comparisons are made against the results of simulations using the TM4 model with both the original and updated reaction rate data.