How Well Can We Model Tropospheric Ozone Production in Current Global Models?

O. Wild

Frontier Research System for Global Change, 3172-25 Showa-machi, Kanazawa-ku, Yokohama, Kanagawa 236-0001, Japan

Abstract. Current chemical transport models can reproduce the general features of the tropospheric ozone distribution reasonably well, but substantial uncertainties in the importance of chemical and physical terms in the ozone budget remain. We focus here on the sensitivity of ozone production to model resolution and to the treatment of a number of meteorological processes, and demonstrate that the resulting variations in the chemical time scales of regional and global O_3 production may have significant implications for the simulation of the impacts of surface precursor sources on air quality and on climate.

Introduction

Recent model comparisons and assessment reports have demonstrated that the current generation of global chemical transport models can reproduce the general features of the tropospheric ozone distribution reasonably well, and agree on the tropospheric burden, about 300 Tg, within about 10% [*Prather et al.*, 2001]. However, this agreement hides large differences in chemical production, deposition and influx from the stratosphere which reveal that significant uncertainties remain in our understanding of the tropospheric ozone budget. While these three tendencies are interdependent, coupled by meteorology and transport, we focus here on chemical production, which dominates the budget, and explore its sensitivity to a number of factors including meteorology, model resolution, and the treatment of chemical processes and aerosol. Particular emphasis is placed on how well the chemical time scales for production are modelled, as the calculated impacts on air quality, tropospheric oxidizing capacity and global climate are strongly influenced by how well the timing and location of production can be simulated.

Model Studies

Model studies described here use the Frontier Research System for Global Change version of the University of California, Irvine global chemical transport model (FRSGC/UCI CTM) [*Wild and Prather*, 2000]. The model is driven with meteorological data for 2001 generated with the European Centre for Medium-Range Weather Forecasts (ECMWF) Integrated Forecast System at TL159L40 resolution, and is used here at T21 ($5.6°\times5.6°$), T42 ($2.8°\times2.8°$) and T63 ($1.9°\times1.9°$) resolutions. We focus on springtime ozone formation in East Asia and evaluate simulations against the extensive measurements of ozone and its precursors made during the NASA Transport and Chemical Evolution over the Pacific (TRACE-P) measurement campaign in Spring 2001 [*Jacob et al.*, 2003]. Model simulations of O_3 during the campaign have been evaluated against ozonesonde, lidar and satellite measurements, and the influx of stratospheric air into the troposphere over this region is captured well [*Wild et al.*, 2003].

The zonal mean instantaneous net O_3 production along all aircraft flight tracks over the Western Pacific during the campaign is shown in Figure 1 along with photochemical steady-state box model calculations driven by observed precursor concentrations and photolysis rate constants using the Georgia Tech/NASA Langley box model [*Crawford et al.*, 1999]. O_3 destruction dominates below 6 km south of 27°N in clean, marine air masses; north of this region there is strong production in continental outflow from China, Korea and Japan. There is significant O_3 production in the upper troposphere over the whole region but this is notably smaller in the CTM than in the observation-driven box model. Along with underestimation of NO_x, and with an overestimation of O_3 over source regions found in comparisons with ozonesonde data, this suggests that the time scales for production are too short in the CTM.

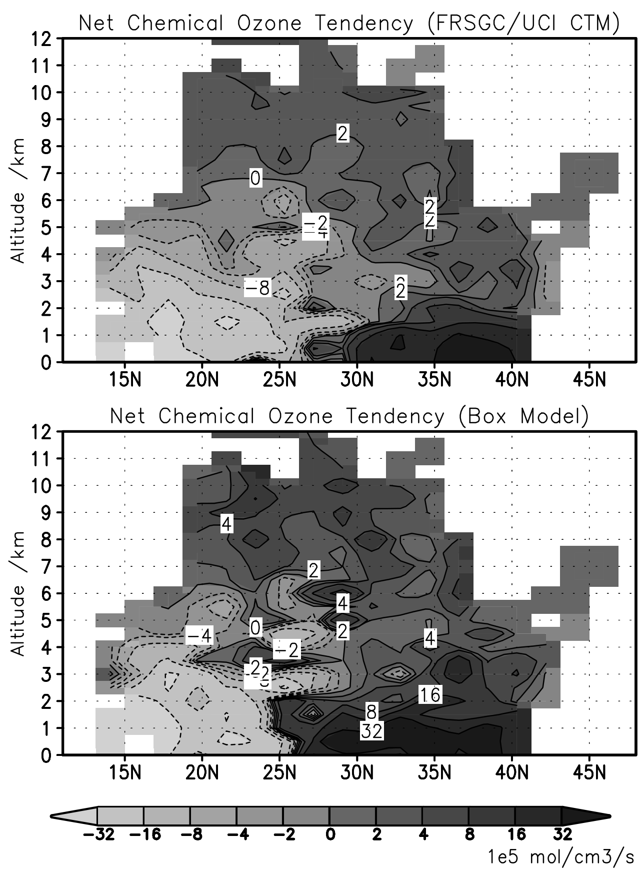

Figure 1. Net O_3 production along aircraft flight tracks during the TRACE-P campaign from FRSGC/UCI CTM (upper panel) and from steady-state box model calculations forced by measurements (lower panel) [*Wild et al.*, 2004].

Sensitivity of Ozone Production

Figure 2 shows the impact of model resolution on springtime O_3 production in the boundary layer over

East Asia. The greater mixing of precursors implicit at the coarser resolution leads to overestimation of O_3 production [e.g., *Sillman et al.*, 1990]; there is 10% greater production at T21 than T63 resolution, and the series converges only slowly suggesting that production is still overestimated at T63 resolution. The impact of boundary layer mixing is shown in the same figure, comparing bulk-mixed and K-profile treatments; in the K-profile scheme, where vertical mixing is less rapid, O_3 production is 12–15% less.

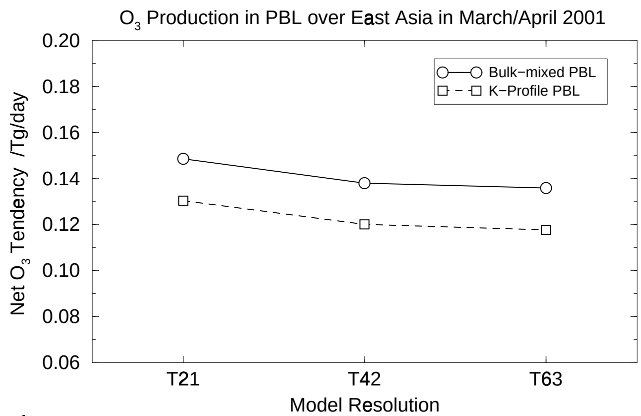

Figure 2. Mean net O_3 production below 750 hPa over East Asia in Spring 2001 at different model resolutions.

The time scales for O_3 production govern how much occurs close to source regions where impacts on air quality may be large, but where the lifetime is shorter, and how much reaches the free troposphere where it may have a larger impact on oxidizing capacity and climate. This is assessed by examining the O_3 production over six weeks following a 5-day pulse of precursor emissions over East Asia in March. Almost 50% of gross production occurs in the regional boundary layer for the standard run (T21, K-profile PBL). The time scales for production are examined by calculating the mean additional mass of O_3 over the region and over the globe, broadly representative of the impacts on air quality and climate, respectively. Figure 3 shows the sensitivity of these regional and global O_3 perturbations to different model treatments.

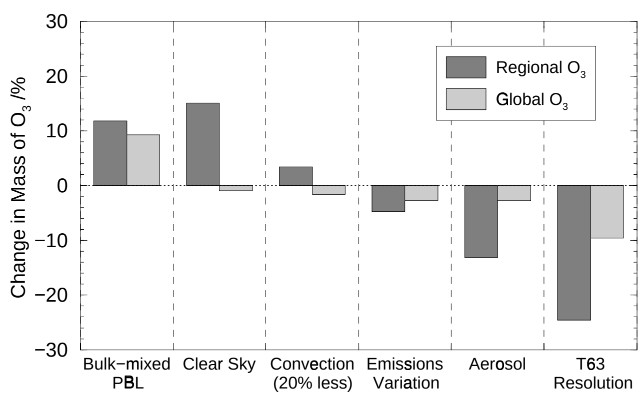

Figure 3. Changes in the regional and global O_3 perturbation due to East Asian emissions in early March compared with a standard T21 K-profile simulation.

Assuming a rapid, bulk mixing of the boundary layer leads to 10% greater impacts from O_3 on both air quality and climate. In contrast, using a higher resolution reduces the calculated impacts by 25% and 10%, respectively. Omitting cloud cover leads to shorter time scales for production and a bigger regional O_3 impact, but the impact on global O_3 is not greatly affected. However, inclusion of the photolytic impact of aerosols, a significant uncertainty in current models, leads to a substantial lengthening of chemical time scales and a reduction in total production. Providing a diurnal variation in anthropogenic emissions, still not well characterised for most parts of the globe, also leads to a modest reduction in production due to the different distribution of NO_x in the PBL. While these scenarios are largely illustrative, it is clear that large uncertainties in simulated O_3 production remain, and that production in current, coarse-resolution models may be biased high.

Conclusions

While current models can reproduce the global O_3 distribution and the variations in O_3 production reasonably well, considerable uncertainty in many processes influencing production remains. In particular, the chemical time scales may not be well simulated, and thus attribution of O_3 impacts from specific sources or regions may be significantly in error. While recent advances such as inclusion of aerosol are expected to lead to better simulation of O_3 production, further improvements are required if the environmental impacts are to be estimated with greater reliability. Comprehensive aircraft measurement campaigns, such as TRACE-P, will remain vital in assessing future progress.

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