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Keith E. Grant
Donald J. Wuebbles

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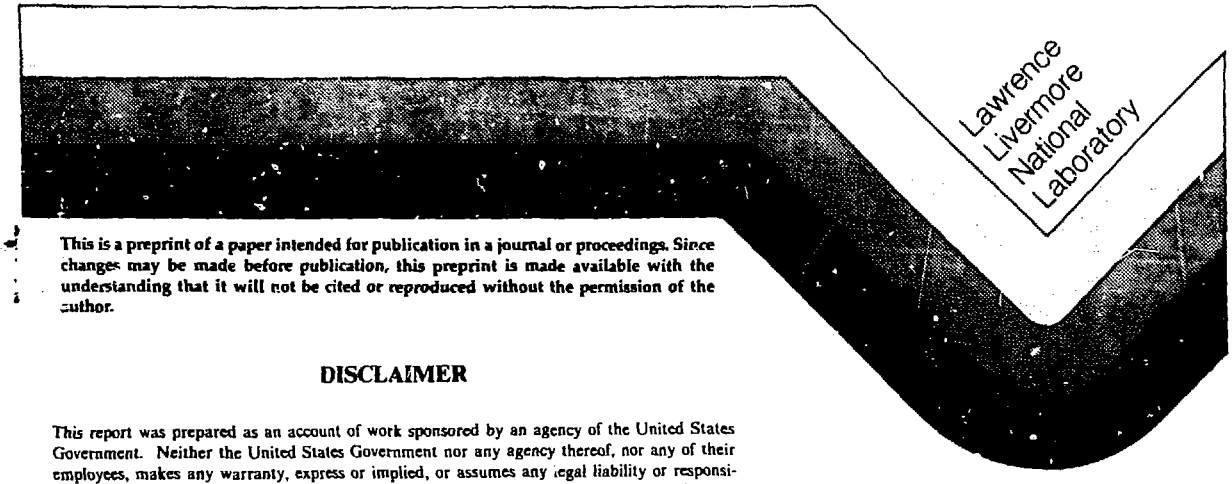

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A TWO DIMENSINAL MODELING STUDY OF THE SENSITIVITY OF OZONE TO RADIATIVE FLUX UNCERTAINTIES

Keith E. Grant and Donald J. Wuebbles
Lawrence Livermore Laboratory, University of California
Livermore, California, USA

ABSTRACT

Radiative processes strongly effect equilibrium trace gas concentrations both directly, through photolysis reactions, and indirectly through temperature and transport processes. We have used the LLNL 2-D chemical-radiative-transport model to investigate the net sensitivity of equilibrium ozone concentrations to several changes in radiative forcing. Doubling CO_2 from 300 ppmv to 600 ppmv resulted in a temperature decrease of 5 K to 8 K in the middle stratosphere along with an 8% to 16% increase in ozone in the same region. Replacing our usual shortwave scattering algorithms with a simplified Rayleigh algorithm led to a 1% to 2% increase in ozone in the lower stratosphere. Finally, modifying our normal CO_2 cooling rates by corrections derived from line-by-line calculations resulted in several regions of heating and cooling. We observed temperature changes on the order of 1 K to 1.5 K with corresponding changes of 0.5% to 1.5% in O_3. Our results for doubled CO_2 compare favorably with those by other authors. Results for our two perturbation scenarios stress the need for accurately modeling radiative processes while confirming the general validity of current models.

1. INTRODUCTION

The time and space dependent evolvement of trace gas concentrations within a 2-D chemistry-transport model is strongly affected by the effects atmospheric radiative processes have on temperatures, transport, and photolysis reactions. Changes to shortwave and infrared fluxes perturb model chemistry and transport calculations via a number of interconnecting mechanisms. Shortwave flux densities control photolysis rates, directly affecting trace gas concentrations. Changes to either short wave or infrared fluxes perturb net heating rates, changing the diabatic circulation and temperature profiles calculated by the model. Transport changes directly change the latitude-altitude distribution of space and time varying trace gas concentrations, in turn feeding back upon radiative absorption. Temperature changes perturb chemical reactions rates, also modifying trace gas concentrations and feeding back upon radiative fluxes.

We have investigated the net sensitivity of equilibrium ozone concentrations, estimated using the LLNL two-dimensional chemical-radiative-transport model, for several changes in radiative forcing. The radiative scenarios used were chosen to be representative of physically motivated modeling concerns from our continuing model development, rather than sensitivity studies based on statistical treatment of individual parameter uncertainties.

2. DESCRIPTION OF LLNL TWO-DIMENSIONAL MODEL

The LLNL zonally averaged two-dimensional chemical-radiative-transport model currently determines the atmospheric distributions of 31 chemically active atmospheric trace constituents in the troposphere and stratosphere. The model domain extends from pole to pole, and from the ground to 0.56 mb (approximately 0 to 54 km). The sine of latitude is used as the horizontal coordinate with uneven increments corresponding to about 10 in latitude. The vertical coordinate corresponds to the natural logarithm of pressure with resolution of approximately 3 km.

Approximately 95 chemical and photochemical reactions are included in the model. Reaction rates, solar flux data, absorption cross-sections, and quantum yields are based on the latest NASA panel recommendations of Demore et al. (1987), with the exception of the rate for $NO_2 + O$, which is based on prior recommendations. Photodissociation rates, including the effects of multiple scattering, are computed as a function of time at each zone, with optical depths consistent with calculated species distributions.

The diabatic circulation for the ambient atmosphere is determined using net heating rates calculated in an internally consistent way with the derived species distributions. The technique for deriving the diabatic circulation is similar to that used by Solomon et al. (1986): the vertical velocity is determined from the zonally averaged residual Eulerian thermodynamic equation, while the horizontal velocity is determined using the equation for mass continuity.

The net heating rates are determined using accurate solar and infrared radiative models. The solar model includes absorption and scattering effects for O_3, O_2, and NO_2 at ultraviolet and visible wavelengths, and for H_2O, CO_2, and O_2 in the near infrared. The solar model for visible and UV wavelengths uses the Sagan and Pollack (1967) two-stream model to calculate reflection and transmission operators for scattering of diffuse incident radiation by a single layer. Scattering from the solar beam is calculated for each layer using the delta-Eddington technique (Joseph et al., 1976). Merging of individual layers, including multiple-scattering, is accomplished via a flux formulation of the adding technique (Harshvardhan et al., 1987). The longwave emission and absorption by O_3, CO_2, and H_2O are included in the infrared submodel.

Temperatures for the ambient atmosphere vary continuously, over the annual cycle, based on the reference model of Barnett and Corney (1985). The derived diabatic circulation depends strongly on the temperature distribution: by using observed temperatures for the ambient atmosphere, a more accurate representation of the diabatic circulation can be derived. The model determined net radiative heating rates, and resulting diabatic circulation, compare well with those derived

from LIMS data (Kiehl and Solomon, 1986; Rosenfield et al., 1987; Solomon et al., 1986).

For the perturbed atmosphere, a perturbation form of the thermodynamic equation is solved for the changes in stratospheric temperatures resulting from changes in the distributions of ozone and other radiatively active constituents. Using this approach, the diabatic circulation is assumed to be unchanged in the perturbed atmosphere from that calculated for the ambient.

Turbulent eddy transport is parameterized through diffusion coefficients K_{yy} and K_{zz}. In the current version of the model, a value of K_{yy} of 2×10^9 cm^2 s^{-1} is assumed at all stratospheric altitudes and latitudes, and values of $1 \times 10^{3-5}$ in the troposphere, with a transition region at the tropopause. Values of K_{zz} are 1×10^3 cm^2 s^{-1} in the lower stratosphere, increasing slowly with altitude based on gravity wave modeling studies.

The continuity equation for each individual species is solved using a variable time step, variable order, implicit technique for solving stiff numerical systems with strict error control. Advection terms are treated accurately using the two-dimensional transport algorithm of Smolarkiewicz (1984). The diurnal averaged concentrations for each species at each zone are calculated at each time step. Accurate diurnal calculations are used to derive time-varying factors for each chemical and photochemical reaction included in the diurnal averaged version of the model.

3. RADIATIVE PERTURBATION SCENARIOS

As a baseline for comparison, we have used a year's seasonal cycle for a chemically ambient atmosphere. A CO_2 mixing ratio of 300 ppmv was chosen for this baseline. Although this is lower than the ambient CO_2 mixing ratios in current general use, it was compatible with available data for line-by-line CO_2 cooling rates (Fels and Schwarzkopf, private communication, 1987). These cooling rates were used in deriving one of our perturbation scenarios. For both the ambient and radiatively perturbed runs described below, the 2-D model was run for sufficient model years that the ozone concentrations had reached steady-state, apart from a seasonal cycle. All scenarios were run for clear-sky conditions, with seasonally varying, latitude dependent ground albedos.

As a first radiative perturbation scenario, the LLNL model was run to radiative-transport equilibrium for doubled CO_2 (600 ppmv), while allowing only stratospheric temperatures to respond to the radiative forcing. A second doubled CO_2 scenario was then run in which full chemical-radiative-transport equilibrium was allowed to occur. Since doubling CO_2 is a relatively standard radiative perturbation, these scenarios allowed comparison between the response of the LLNL model and the responses of other 2-D models.

A third scenario was created by modifying the present shortwave multiple-scattering submodel to simulate the simpler Rayleigh multiple-scattering formulation of Luther (1980). Luther's model was, until recently, used in LLNL models to calculate photolysis rates. Similar models are being used in other 2-D models. In current LLNL models, we have switched to the more rigorous scattering model, described above, which is able to treat clouds and aerosols as well as being more accurate at larger zenith angles. For this scenario, the radiation scattered out of a layer was approximated as the total single-scattering extinction minus the corresponding gaseous absorption. Diffuse radiation was assumed to have an effective zenith angle of approximately 53°, and scattered radiation was partitioned equally in the forwards and backwards directions. This scenario was run without temperature feedback, so the changes in equilibrium ozone were forced entirely by changes in photolysis rates.

For the final scenario in this study, we added corrections to the heating rates calculated by the longwave submodel internal to the LLNL 2-D model. These corrections were based on line by line calculations, which are expected to give a more accurate representation of cooling rates than those obtainable from any band model. An earlier version of the longwave submodel used by the LLNL 2-D model was described in Harshvardhan et al. (1987). The current version has improved treatments of absorption by CO_2 and doppler broadening of H_2O in the upper stratosphere (Harshvardhan, private communication, 1987). Using this improved longwave submodel we calculated CO_2 cooling rates for four vertical profiles with corresponding line-by-line calculations of infrared cooling rates for 300 ppmv CO_2. The two sets of cooling rate profiles were differenced to obtain cooling rate corrections for the McClatchey et al. (1972) midlatitude summer, midlatitude winter, tropical and subarctic winter profiles. After averaging the temperature and cooling rate correction profiles to the LLNL 2-D model vertical grid, singular value decomposition was used with these data to develop a linear model that related vertical profiles of cooling rate corrections to model generated temperature profiles. Cooling rate corrections generated by this linear model were then added to the cooling rates calculated by the longwave submodel in the LLNL 2-D model during model execution. This scenario was forced by temperature feedback to chemical-radiative-transport equilibrium.

4. RESULTS

Figure 1 shows the total column ozone as a function of month and latitude as calculated by the LLNL 2-D model for an ambient atmosphere with 300 ppmv CO_2. This distribution generally compares well with observations, although it shows too much total ozone in the tropics. The ozone distributions for all the other scenarios discussed, will be relative to the ozone for this atmosphere.

In Figure 2, we show the change in temperature for July, resulting from doubling CO_2 to 600 ppmv and allowing only the stratospheric temperatures to come into radiative-transport equilibrium (i.e. no chemistry feedback). No changes were made to tropospheric or surface temperatures. The temperature changes shown are forced only by changes in longwave cooling. The observed cooling increases with altitude with little latitudinal variation in the lower and middle stratosphere. In the upper stratosphere, there is slightly stronger (2 K) cooling at the winter pole and weaker cooling in the tropics. These results are in general agreement with those of Fels et al. (1980).

In Figure 3, we show (for July) the temperature change and percent ozone change, resulting from doubling CO_2 with full chemistry feedback. Allowing for somewhat different lower boundary conditions and a change in CO_2 of 300 ppmv versus 350 ppmv, these results compare closely with those of Brasseur and Hitchman (1988). Our temperature changes for doubled CO_2 with chemistry feedback also compare closely with those reported by Haigh (1984). Relative to Figure 2, Figure 3a shows less cooling (3 K) in the upper stratosphere, except slightly greater cooling (1 K) at the winter pole. These changes are explained by increased infrared cooling due to increased ozone, overshadowed by increased shortwave heating except at the winter pole. The changes in ozone in Figure 3 are, in general anticorrelated with the temperature changes. In the lower stratosphere the ozone increase shows a much greater latitudinal gradient, being smallest in the tropics and largest at the summer pole. The ozone increase peaks around 45 km at 18 to 22 percent.

Figure 4 shows the percent change in ozone for July caused by changing the 2-D model scattering algorithm to the simple Rayleigh treatment described above. In summary, an increase

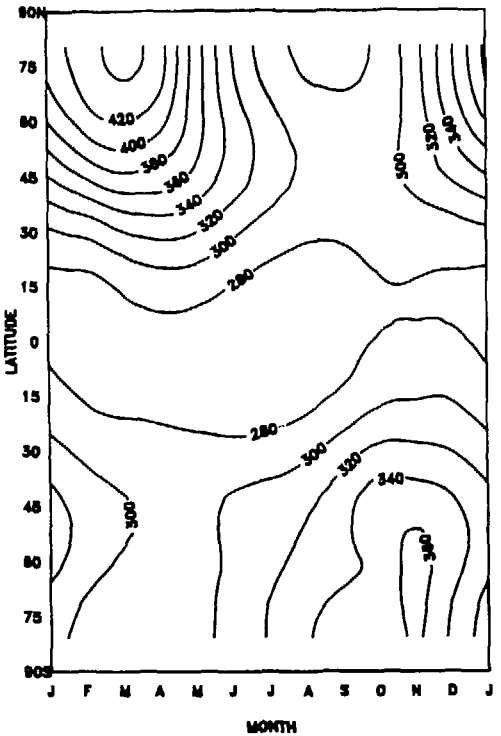

Fig. 1. Total ozone in Dobson units for an ambient atmosphere with 300 ppmv CO_2.

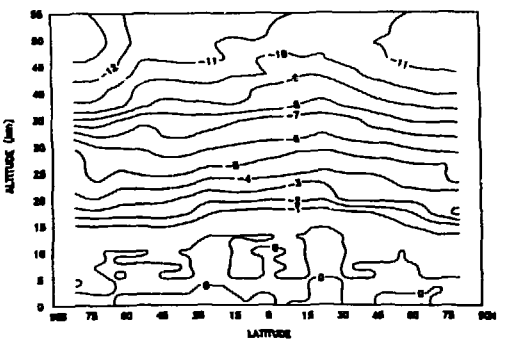

Fig. 2. Temperature change for July in response to doubling CO_2 without chemistry feedback.

in ozone is observed, peaking in the lower stratosphere at 1.5 to 2.5 percent and becoming largest in the summer hemisphere for small solar zenith angles. This increase is attributable to a increase in the photolysis of O_2. This case displays the importance of scattering algorithm choice, even under the simplest clear-sky conditions. It differs from the other scenarios presented here, in that it is driven solely by change in photolysis rates without accompanying temperature feedback.

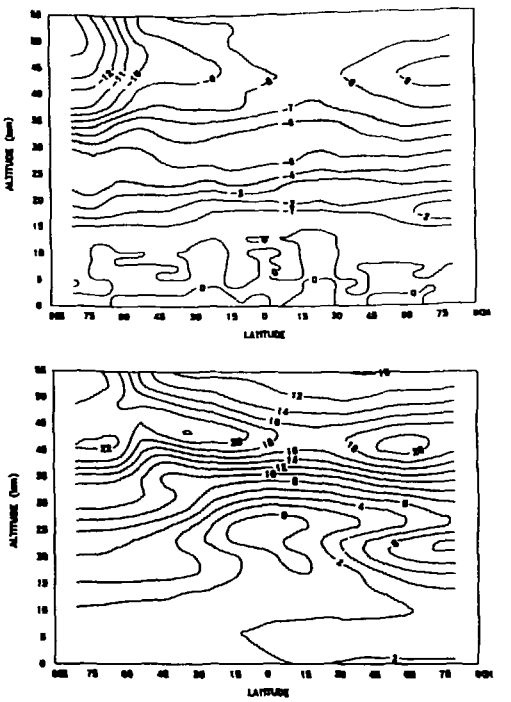

Fig. 3. (a) Temperature change for July in response to doubling CO_2 with chemistry feedback (b) Ozone percent change for July in response to doubling CO_2 with chemistry feedback.

radiation package. As described above, these correction terms were generated as a function of temperature profile from a linear model based on 300 ppmv CO_2 line-by-line calculations. The general features created by this correction include: slight warming (1 K) just above the polar tropopause, a region of slight cooling (1.0 to 1.5 K) between 20 km to 35 km, warming (0.5 K) between 35 km and 45 km, and cooling (1 to 3 K) above 45 km. A region of stronger cooling (greater than 3 K) occurs above 45 km at the winter pole. The ozone response in Figure 5 is anticorrelated with the temperature change, and of the order of one percent for each degree.

5. CONCLUSIONS

The studies described here should not be construed as being quantitatively predictive of errors which would be obtained in estimating ozone changes for chemical perturbation scenarios. For instance, the radiative perturbations of scenarios three (Figure 4) and four (Figure 5) would be expected to have a smaller effect on relative ozone than on absolute ozone concentrations. Nor are these studies in any sense comprehensive. Rather, they form part of our ongoing process of studying and evaluating the sensitivity of the LLNL 2-D model to changes and unavoidable tradeoffs in its radiative parameterizations (e.g. the most accurate and physically correct radiation models are far too slow for inclusion in chemical-radiative-transport models). However, The good agreement of our results for doubled CO_2 with those for other 2-D models suggests that the magnitude and form of the effects we saw for small radiative changes is generally valid. The response of

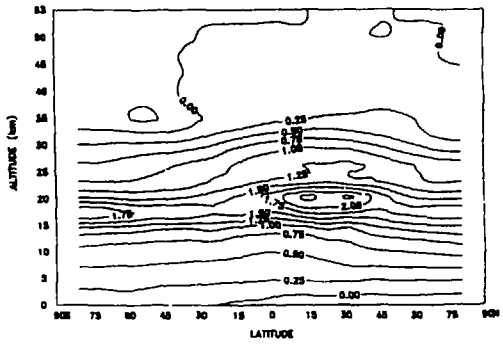

Fig. 4. Percent ozone change for July in response to replacing the shortwave scattering treatment with a simplified Rayleigh scattering algorithm.

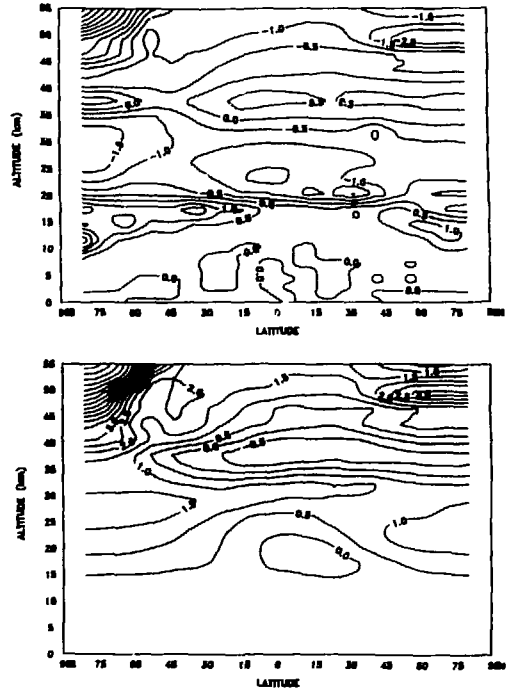

Fig. 5. (a) Temperature change for July in response to adding corrections to the normal CO_2 longwave cooling rates. These corrections were derived using line-by-line calculations for 300 ppmv CO_2. (b) Ozone percent change for July in response to adding above corrections.

ozone and temperature changes to the small radiative perturbations of scenarios three and four, relative to the responses for doubled CO_2 with chemical feedback in scenario two, provide an incomplete yet valid benchmark of the magnitude of error in ozone concentration predictions attributable to current radiative transfer approximations. Our results stress the importance of accurately modeling radiative processes when accurate distributions of trace gas concentrations are required. They also indicate, however, that model predictions for total ozone are not unduly susceptible to the reasonable approximations required for computationally efficient radiation models.

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