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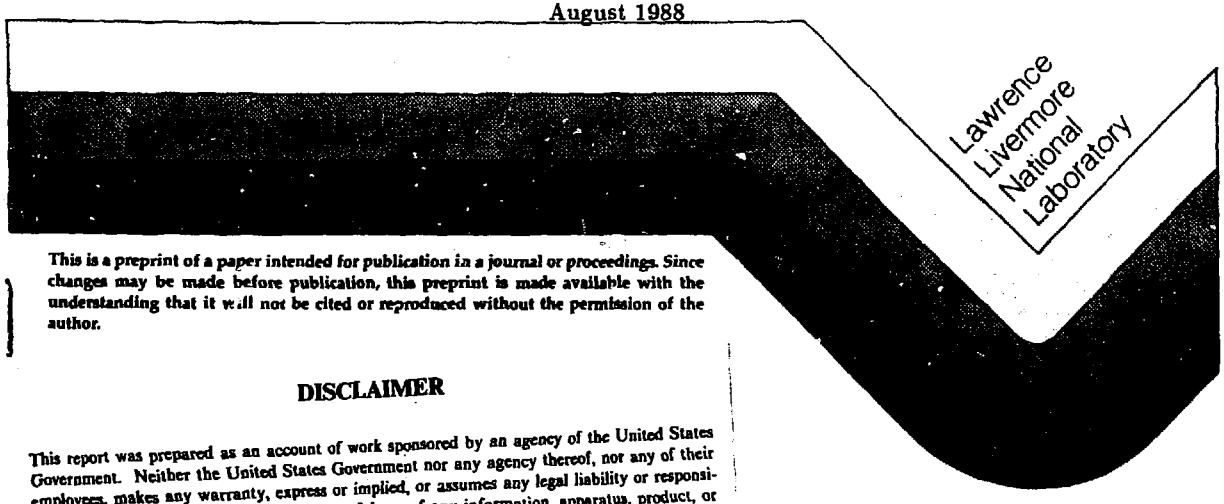

A TWO-DIMENSIONAL MODEL STUDY OF PAST TRENDS IN GLOBAL OZONE

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ABSTRACT

Emissions and atmospheric concentrations of several trace gases important to atmospheric chemistry are known to have increased substantially over recent decades. Solar flux variations and the atmospheric nuclear test series are also likely to have affected stratospheric ozone. In this study, the LLNL two-dimensional chemical-radiative-transport model of the troposphere and stratosphere has been applied to an analysis of the effects that these natural and anthropogenic influences may have had on global ozone concentrations over the last three decades. In general, model determined species distributions and the derived ozone trends agree well with published analyses of land-based and satellite-based observations. Also, the total ozone and ozone distribution trends derived from CFC and other trace gas effects have a different response with latitiude than the derived trends from solar flux variations, thus providing a "signature" for anthropogenic effects on ozone.

1. INTRODUCTION

At the Quadrennial Ozone Symposium in 1984, we presented a theoretical analysis, using the LLNL one-dimensional model, on the past trends in ozone and temperature expected from emissions of chlorofluorocarbons and other trace gases (Wuebbles, 1984). In this paper, we examine these trends using the LLNL two-dimensional model, which provides much more information on latitudinal and seasonal effects. Emissions and atmospheric concentrations of several trace gases important to global atmospheric chemistry, including CO_2, CH_4, N_2O and several chlorocarbons, are known to have increased substantially over recent decades. In addition, variations in ultraviolet radiation during the 11-year solar cycle are likely to have influenced upper stratospheric photochemistry. We also consider the effect of nitrogen oxides (NO_x) produced from the atmospheric nuclear test series of the late 1950s and early 1960s.

The LLNL two-dimensional chemical-radiative-transport model of the troposphere and stratosphere has been applied to analyzing the effects that these natural and human-related influences may have had on global ozone concentrations over the last three decades. Similar calculations were included in a recent international study of global trends (Watson, 1988); however, results presented here are based on an updated version of the model.

2. THE 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). 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 (Demore et al., 1987, except 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 of O_3, O_2, and NO_2 at ultraviolet and visible wavelenghts, and for H_2O, CO_2, and O_2 in the near infrared. The longwave emission and absorption by O_3, CO_2, and H_2O are included in the infrared submodel.

Temperatures for the atmosphere vary continuously over the annual cycle based on the reference data set of Barnett and Corney (1985). The derived diabatic circulation depends strongly on the temperature distribution; by using observed temperatures for the ambient atmosphere, an 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 all of the model calculations presented here, the diabatic circulation is assumed to be unchanged from that derived for a reference atmosphere corresponding to the mid-1980s. At the conference, we plan to discuss internally consistent perturbations to stratospheric temperatures; unfortunately results from this version of the model were not available by the time we had to write this paper.

3. HISTORICAL EMISSIONS AND INFLUENCES

Modeling past trends require estimates of the historical changes in trace gas emission and concentrations, variations in solar ultraviolet radiation, and NO_x produced from nuclear tests. Possible effects from other influences such as the El Chichon eruption, the Antarctic ozone hole, or possible changes in climate or atmospheric dynamics, were not included. As much as possible, available measurements and emissions evaluations were used in the development of the historical scenarios.

The historical emissions for the chlorocarbons $CFCl_3$, CF_2Cl_2, CCl_4 and CH_3CCl_3 were based on the expressions

developed by Wuebbles et al. (1984) from available Chemical Manufacturing Association data and other databases. The time history of CH_4 concentrations were based on measurements made since 1978 indicating a 1% per year increase in methane concentrations (Blake and Rowland, 1988; WMO, 1985) and data for earlier periods (e.g., spectral data and ice core data). Historical changes in N_2O surface mixing ratios were based on the data and analysis of Weiss (1981). The expression for increase in CO_2 concentrations developed by Wuebbles et al. (1984) was used; this was based on observations at Mauna Loa since 1958 and assumes a pre-industrial concentration of 270 ppmv.

Many uncertainties remain regarding the variations in ultraviolet radiation during the 11-year solar cycle. The variations assumed here are derived from the analyses of Heath and Schlesinger (1984, 1986). Their analyses, based on SBUV data, assume that the spectrum of solar rotation induced variations are preserved over the solar cycle and that the irradiance variations are linearly related to the ratio of the core to wing emissions of the MgII line. Their analyses produced variations in the ultraviolet flux from solar minimum to solar maximum of 9% at 205 nm, 4% at 250 nm, and 1% at 270 nm. These variations are much smaller than used in prior published modeling studies. For the model studies presented here, the solar cycle flux changes are assumed to have a sinusoidal variation with time. Magnitudes of respective solar cycles are based on F10.7 solar radio flux data (Heath and Schlesinger, 1984), with the magnitude of solar cycle 21 (1974–1985) taken to be 1.0 (i.e., same solar flux variations from solar min to solar max as given above).

Past modeling studies (e.g., Chang et al., 1979; Wuebbles, 1983) have indicated that nitrogen oxides produced from atmospheric nuclear tests could have had significant effects on stratospheric ozone during the early 1960s. In the calculations presented here, all reported tests are included, with the timing and yield of the various test devices based on Bauer (1979). The altitude for the bottom and top of the stabilized cloud are taken from the empirically-based analyses of Peterson (1970). Likewise, the distribution of NO_x formed within the cloud is based on Peterson (1970). There still remains uncertainty regarding the amount of NO produced per megaton of explosive energy; we assumed 0.67×10^{32} NO molecules/Mt based on the discussion in Chang et al. (1979).

4. RESULTS AND DISCUSSION

Figure 1 shows the model-determined annual and globally averaged changes in the total ozone for the period 1950–1986 relative to the total ozone amount calculated for 1960. Shown are three cases corresponding to the effects of trace gas emissions only, trace gases plus solar cycle, and finally, with nuclear test effects also included. These calculations indicate that trace gas emissions could have decreased total ozone by about one percent since 1960 (note that effects due to the Antarctica ozone hole, which are not included here, would tend to cause larger decreases). Prior calculations with the LLNL 1-D model suggest that the effects of temperature feedback, including stratospheric cooling due to the past growth in the CO_2 concentration, would tend to reduce the magnitude of the trend over this period. Inclusion of solar flux variations result in significant year-to-year changes in the total ozone amount. Although dependent on the particular cycle, the model estimates changes in total ozone of about 1.8% from solar minimum to solar maximum. These calculated global variations are consistent with available statistical analyses attempting to determine the solar cycle induced changes in total ozone from Dobson and satellite data (Reinsel et al., 1987, 1988; Oehlert, 1986). Likewise, the global trend in total ozone determined from trace gases is also consistent with published analyses of Dobson data for the 1970s and 1980s. (Angell and Korshover, 1983; Reinsel et al., 1987).

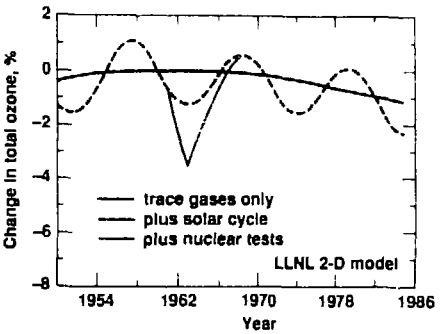

Fig. 1. Calculated global and annual-averaged changes in total ozone from 1950 to 1986 relative to total ozone for 1960 for three cases: (1) trace gases only, (2) trace gases plus solar cycle variations; and (3) with atmospheric nuclear test effects also included.

Figure 2 shows the model calculated distribution of total ozone for the year 1985. A comparison of this diagram with Dobson and satellite data indicates good general agreement. The model simulates the latitudinal contrast well, with maxima generally occurring approximately during early spring at high latitudes. The estimated high latitude minima are also in good agreement with observations. However, there is too much ozone calculated for the tropics.

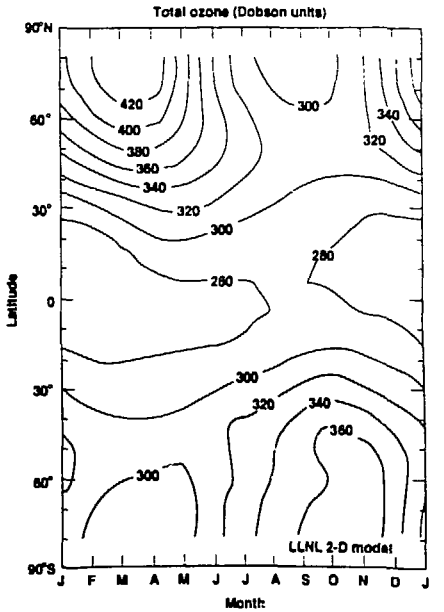

Fig. 2. Distribution of total ozone (in Dobson units) as a function of latitude and month calculated for model year 1985.

In agreement with past 1-D model studies, a significant decrease in total ozone (see Fig. 1) was calculated due to the atmospheric nuclear tests in the early 1960s. The maximum

effect of the nuclear tests on globally and annual averaged ozone occurred in 1963, with a maximum change of -2.3%. This is consistent with available analyses of Dobson data for this period (Reinsel, 1981; Chang et al., 1979). A solar minimum also occurs during this same time period. The recovery of ozone concentrations from the nuclear tests occurred over the next five years. Including the effects of solar cycle variations, the calculated total ozone increase from 1963 to 1970 is about 3.8%.

Figure 3 shows the model calculated changes in total ozone due to the nuclear tests for model year 1962 relative to 1960. Large ozone decreases are calculated in late 1962 at high northern latitudes, with a maximum decrease of 12.5% occurring poleward of 70°N in November 1962. This is consistent and showed a high correlation with a recent analysis of data from Northern Hemisphere Dobson stations (R. Bojkov, private communication, 1988). The largest changes occur at this time because of a significant number of high latitude (~75°N) tests by the USSR (eleven tests >5 Mt) and several tests at lower latitudes (2-17°N) by the US (two tests >5 Mt).

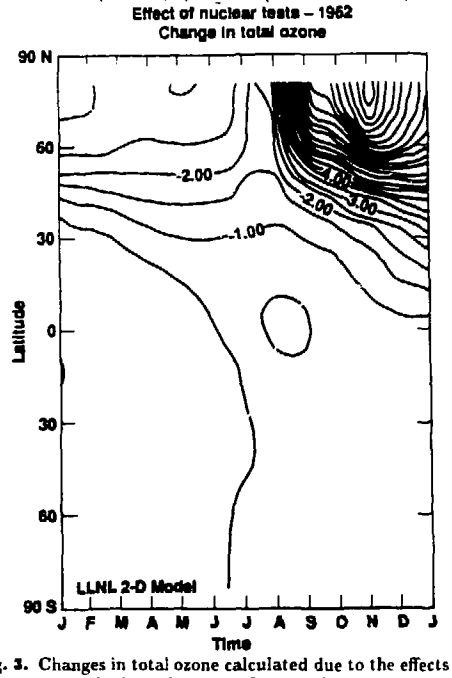

Fig. 3. Changes in total ozone calculated due to the effects of atmospheric nuclear tests for model year 1962.

Atmospheric nuclear testing was banned after 1962, although, as indicated earlier, the maximum global effect on total ozone occurred in 1963. Throughout the year, ozone at high northern latitudes continues to recover from the initial odd-nitrogen loading. Odd-nitrogen is also being transported to lower latitude and into the Southern Hemisphere, resulting in a 2% decrease in S.H. total ozone by late 1963. Later years continue to show gradual recovery of ozone concentrations at all latitudes.

The period from 1979 to 1986 is of particular interest because of the availability of satellite data. The trace gas emissions scenario gives a 0.6% decrease in globally-averaged total ozone for this time period. A much larger decrease in total ozone, -2.6%, is determined when solar cycle effects are included, reflecting the transition from solar maximum to solar minimum conditions. Analyses of SBUV data (Bowman, 1988; Reinsel et al., 1988) for the 1979-1985 period give much larger trends, but these analyses do not adequately account for apparent instrumental degradation over the period. The Reinsel et al. (1988) analysis does indicate a drift of data from SBUV relative to the Dobson data; when this drift is accounted for they determine a change of -0.35 ± 0.28% per year or -2.5% over this time period. This is in excellent agreement with the model determined trend, implying that most of the ozone decrease observed over this time period is related to solar cycle effects. In turn, an increase in total ozone from solar cycle effects would be expected over the next five years, until solar maximum is reached in 1990.

Figure 4 shows the change in total ozone and in local ozone (for January) calculated over the 1979-1986 period for the trace gas only scenario. A similar set of diagrams is shown in Fig. 5 for the trace gas plus solar cycle case. In each case, the largest changes in total ozone are found at the high latitudes in early spring, and the smallest changes occur in the tropics. However, these latitudinal and seasonal distributions of the changes are related primarily to the variations in trace gas concentrations. Solar cycle variations alone induce very little gradient with season or latitude. This implies that global measurements of total ozone showing such gradients are indicative of effects expected to be induced by from CFC's and other trace gases, and perhaps providing a "signature" for trace gas influence on ozone.

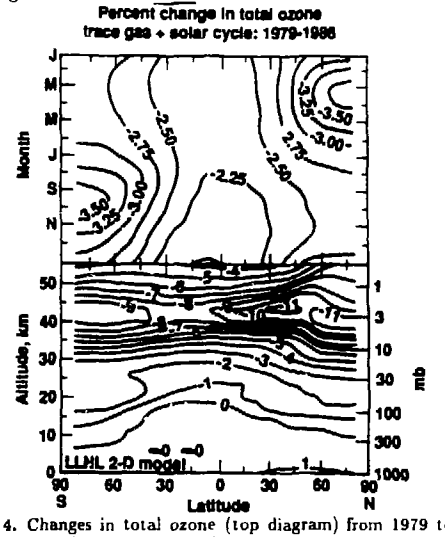

Fig. 4. Changes in total ozone (top diagram) from 1979 to 1986 for trace gases only, and changes in local ozone vs. latitude and altitude (bottom diagram) in January for trace gas only scenario.

Similarly, the largest percentage changes in ozone as a function of latitude and altitude occur in the upper stratosphere, near 3 mb, and show a gradient with the largest changes at higher latitudes and minimum changes in the tropics. While the maximum solar effect also occurs near 3 mb, the calculated gradient with latitude is small. Therefore, measured latitudinal gradients in ozone change near 3 mb would be primarily indicative of CFC and other trace gas effects.

These calculated changes in stratospheric ozone also compare extremely well with available Umkehr data at mid-northern latitudes for layer five through eight (J. DeLuisi, private communication, 1988). There are indications, however, that the model may underestimate decreases in ozone in the lower

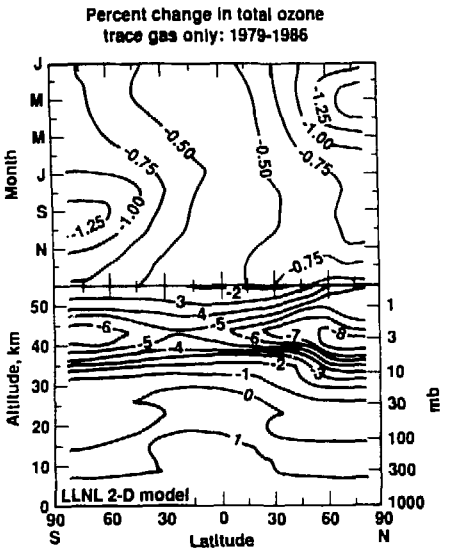

Fig. 5. Changes in total ozone (top diagram) from 1979 to 1986 for trace gases only, and changes in local ozone vs. latitude and altitude (bottom diagram) in January for trace gas only scenario for trace gases plus solar cycle scenario.

stratosphere as implied by measuremeasurements near 20 km from both Umkehr and ozonesonde data.

ACKNOWLEDGMENTS

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