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CLIMATE–CHEMICAL INTERACTIONS AND GREENHOUSE EFFECTS OF TRACE GASES

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ABSTRACT

A completely coupled one-dimensional radiative-convective (RC) and photochemical -diffusion (PC) model has been developed recently and used to study the climate-chemical interactions. The importance of radiative -chemical interactions within the troposphere and stratosphere has been examined in some detail. We find that increases of radiatively and/or chemically active trace gases such as CO_2, CH_4 and N_2O have both the direct effects on climate change by modifying the radiation balance through greenhouse effects and the indirect effects on climate change by changing the atmospheric O_3 profile through their interaction with chemical processes in the atmosphere. It is also found that the climatic effect of ozone depends strongly on its vertical distribution throughout the troposphere and stratosphere, as well as on its column amount in the atmosphere.

I. INTRODUCTION

Observations have shown that the concentrations of atmospheric CO_2 and other trace gases (OTG) such as CH_4, N_2O, and NOx have been increasing and the trend of increases will most likely continue. It has been suggested that human activities are the major contributions to these increases. Increases of these radiatively and/or chemically active trace gases may have implications for global climate both directly by modifying the earth's radiation budget through their absorption properties and indirectly by changing atmospheric O_3 which is both radiatively and chemically active through the chemical processes in the atmosphere [WMO, 1985]. The greenhouse effect of CO_2 has long since received the most emphasis as a mechanism for climate change, but now the problem concerning the greenhouse effects due to human activities has bordered in scope from the CO_2-climate problem to the trace-gas-climate problem. As many trace gases are both radiatively and chemically active, the climate effects of OTG are strongly governed by interactions between chemical, radiative and dynamic processes in the atmosphere. Complexity of the trace-gas-climate problem results also from the fact that the enhanced greenhouse effect due to the increasing CO_2 and the OTG will affect the temperature structure of the atmosphere and in turn the change in temperature profile will change the concentration distribution of many trace gases through their temperature dependence of the chemical reactions. A coupled radiation, chemistry and dynamics model is obviously needed for the trace-gas-climate studies.

II. MODEL DESCRIPTION AND CONTROL CASE

The model used in this study is a completely coupled one dimensional radiative-convective (RC) and photochemical-diffusive (PC) model which extends from 0 to 68 km with a uniform altitude resolution of 2 km. An accurate and efficient radiation transfer scheme, the exponential sum fitting of transmission function (ESFT) is used for the thermal radiation calculation [Shi, 1981], and a combination of the time-marching (TM) method and the Newton-Raphson (NR) iteration is used for finding the equilibrium temperature profile in the RC model [Shi, 1992a,b]. In the PC model, more than 50 species and 100 reactions are taken into account, including the Ox, HOx, NOx, BrOx, and ClOx chemistry. The chemical reaction rates and the photochemical absorption cross sections used in the calculations are taken from DeMore et al. [1990], and the incident solar flux as a function of wavelength is taken from Hudson et al. [1982]. The model leads to a control climate with the surface temperature of 288.15 K, the surface albedo of 0.102, the planet albedo of 0.316 and the ozone column amount of 0.344 atm-cm, which is a good representation for the present global mean climate.

The vertical temperature profile obtained from the coupled model is shown in fig. 1, along with the U.S. 1976 Standard Atmosphere. The agreement between them is quite good throughout the troposphere and stratosphere, with minor deviations near the tropopause and stratopause. This might mean that the radiative equilibrium

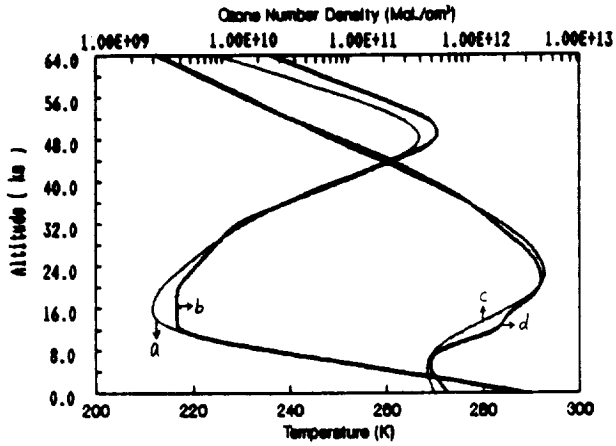

Fig. 1. Temperature (a) and ozone profile (c) obtained by the coupled RC-PC model, and the U.S. 1976 Standard Atmosphere (b and d).

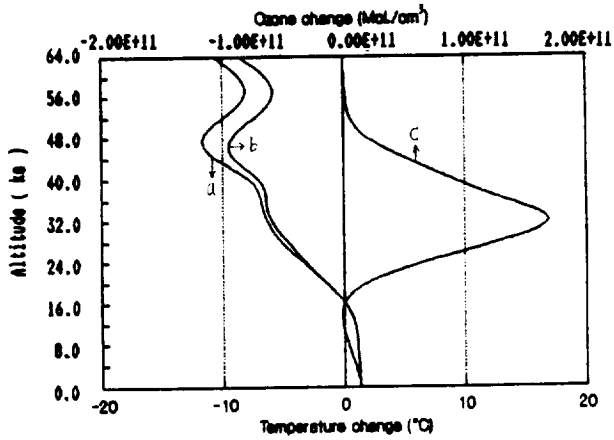

Fig. 2. Changes in temperature and ozone concentration for $2xCO_2$ (340 to 680 ppmv). a and b are the temperature changes from RC and RC-PC model, respectively, and c is the ozone concentration calculated from RC-PC model.

is not a good assumption in these regions. Fig. 1 also shows the ozone profile obtained from the coupled model, and from the U.S. 1976 Standard Atmosphere for comparison. It can be seen from the figure that the overall agreement between the two profiles is satisfactory, except for the troposphere where the model underestimated the ozone. This comes probably from the model's overestimating the reactive radicals HOx which catalyze ozone destruction in the troposphere.

III. RESULTS AND DISCUSSIONS

The results from several case studies on climate-chemical interaction will be presented here. These cases of perturbation in the atmospheric concentration of CO_2, CH_4 and N_2O will most likely occur in the next century.

1. $2xCO_2$ (340-680 ppmv)

CO_2 is a radiatively active gas. It governs the magnitude of stratospheric longwave emission and has significant impacts on stratospheric climate. As well known, the infrared emission to space by CO_2 and the absorption of solar radiation by stratospheric O_3 are primarily responsible for the radiative energy budget in the stratosphere. An increase in CO_2 concentration acts to cool the stratosphere while the troposphere warms. This change in temperature structure caused by increasing CO_2 affects the concentration of O_3 through the temperature dependence of chemical reaction related to O_3.

Fig. 2 shows the changes in the temperature and ozone profiles due to a doubled CO_2 concentration, obtained by the RC model and by the coupled RC-PC model, respectively. We can see from the figure that, the calculated maximum temperature change (-12.46^0C) occurs at 46 km and the calculated surface temperature increase is $+1.32^0$C by the RC model, and the corresponding values by the RC-PC coupled model are -10.30^0C and $+1.30^0$C, respectively. This suggests the incorporation of chemical process into RC mitigated somehow the stratospheric cooling and the surface warming due to a doubling CO_2. In fact, as the stratospheric temperature decreases, the temperature dependent ozone destruction reactions, $O+O_3 \rightarrow 2O_2$ and $NO+O_3 \rightarrow NO_2+O_2$, will be slowed down and the ozone production rate through the reaction, $O+O_2 \rightarrow O_3$, will increase. This results in a net increase in stratospheric ozone concentrations. The model calculations indicate, in general, a column O_3 increase of about 3.0% for a doubling of CO_2. This O_3 increase is mainly distributed over above 20km where the enhanced solar heating due to O_3 increase offseted, more or less, the stratospheric cooling due to CO_2 increase. It means that the chemical feedback gives a negative effect on the stratospheric temperature perturbation and the surface temperature change for the case of CO_2 doubling.

2. $2xCH_4$ (1.60-3.20 ppmv)

CH_4 is a radiatively active gas. Just like the radiative nature of CO_2. Increase of CH_4 will lead the temperature in the lower atmosphere to increase and the temperature in the upper atmosphere to decrease. By the RC model, the temperature changes at surface and at 56 km are 0.17^0C and -0.26^0C, respectively, for a doubling of CH_4 (Fig. 3.).

CH_4 is also a chemically active gas. Model result by the RC-PC for a doubling of CH_4 gives

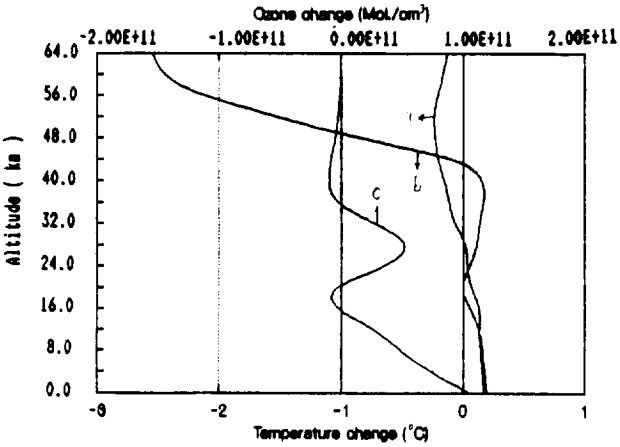

Fig. 3. As in Fig. 2, but for $2xCH_4$ (1.60 to 3.20 ppmv)

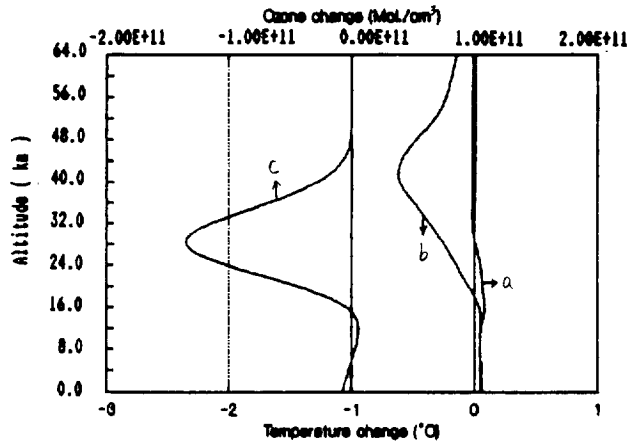

Fig. 4. As in Fig. 2, but for $1.2xN_2O$ (0.30 to 0.36 ppmv)

an increase of column ozone about 1.2% and the increase ranges from the surface to 36 km, as shown in the figure. We can find from the figure that there are two peaks in the increases of O_3. One is located near the ground, which is resulted from that the CH_4 oxidation produces O_3 through the CH_4-NOx-smog-reactions [Johnston, 1984]; the other is around 28 km, which is caused by the fact that CH_4 provided a sink for CLx and then led to slow down the O_3 destruction catalyzed by CLx.

Compared with the results by the RC, the temperature changes obtained by the RC-PC are 0.19^0C at the surface and -2.6^0C at 56 km, respectively, for a doubling CH_4. This means that the chemical feedback contributes a positive effect to the stratospheric temperature perturbation and the surface temperature change, as contrasted with the case of doubling CO_2.

The major sink of atmospheric CH_4 is its reaction with the hydroxyl radical OH while the CH_4 oxidation by OH is the main loss mechanism of OH in the free troposphere. So CH_4 and OH levels are coupled tightly. Increases in CH_4 lead to a decrease in OH and thereby to a further increase in CH_4 levels. With continued increases in CH_4 in the future, this positive feedback could become increasingly important.

3. $1.2xN_2O$ (0.30–0.36 ppmv)

N_2O is also a radiatively and chemically active gas. The results for $1.2xN_2O$ are shown in Fig. 4. In many respects, the radiative consequences due to $1.2xN_2O$ are similar to those of $2xCO_2$ and $2xCH_4$. Increase in the concentration of N_2O causes tropospheric heating and stratospheric cooling. Similar to the case of $2xCH_4$, the chemical feedback gives also a positive effect on the surface temperature change and the stratospheric temperature perturbation.

The chemical effects of increasing atmospheric N_2O arise from the reaction, $O(1D)+N_2O \rightarrow 2NO$, and this reaction provides the major source of odd nitrogen (NO_x) in the middle stratosphere from about 20 to 40 km. Consequently, an increases in atmospheric N_2O level will lead the stratospheric NO_x to increase and subsequently to reduce the stratospheric O_3 by catalytic destruction through the reactions, $NO+O_3 \rightarrow NO_2+O_2$ and $NO_2+O \rightarrow NO+O_2$. Decreased ozone heating will give a lower temperature in the stratosphere. On the other hand, the decreased stratospheric ozone will lead more solar radiation to reach the surface and cause the surface warmer.

4. O_3–climate problem

It is clear from the results mentioned above that in addition to their direct radiative effects, many of the greenhouse gases also have indirect radiative effects on climate through their interactions with atmospheric chemical processes. As a result of such interactions, the global distribution of ozone in the troposphere and stratosphere was changed.

Ozone plays an important dual role in affecting climate. While CO_2 and OTG are relatively well-mixed in the atmosphere, the climatic effect of ozone depends strongly on its vertical distribution throughout the troposphere and stratosphere, as well as its column amount in the atmosphere. The surface temperature changes calculated by the RC model are shown in Fig. 5 as a function of ozone percentage change and altitude where O_3 is changed. U. S. 1976 Standard Atmosphere is used in the calculations. It can be seen from the figure that changes in ozone in the middle-upper troposphere and lower stratosphere (below 22 km) are most effective in determining the change in surface temperature, with increased ozone leading to a

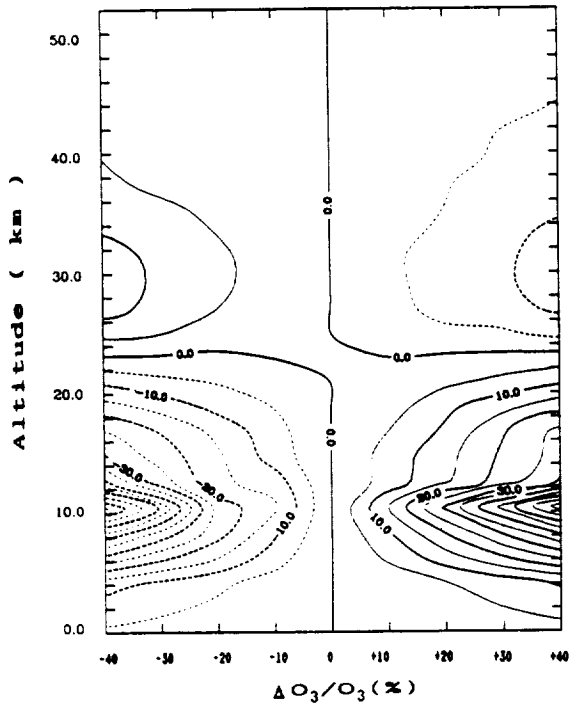

Fig. 5. Computed change in surface temperature ($x10^{-3}$ K) due to ozone change (%) as a function of altitude at which ozone is perturbed.

warmer surface (right part of the figure), and vice versa (left part of the figure). This is because the greenhouse effect is directly proportional to the temperature contrast between the level of emission and the levels where radiation is absorbed. This contrast is greatest near the tropopause and the climate sensitivity of ozone change reaches its maximum there. Above about 25 km, added ozone causes a small decrease in surface temperature (right part of the figure) because it absorbs extra solar radiation, effectively robbing the troposphere of direct solar energy that would otherwise warm the surface, and vice versa (left part of the figure).

In summary, ozone is a primary absorber of solar radiation in the stratosphere where it is directly responsible for the increase in temperature with altitude. Ozone is also an important absorber of infrared radiation. It is the balance between these radiative processes that determine the net effect of ozone on climate.

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A GENERAL CIRCULATION MODEL STUDY OF THE CLIMATIC EFFECT OF OBSERVED STRATOSPHERIC OZONE DEPLETION BETWEEN 1980 AND 1990

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1. Introduction

The TOMS and SAGE measurements show a significant reduction in the stratospheric ozone over the middle and high latitudes of both hemispheres between the years 1979 and 1991 (WMO, 1992). This change in ozone will effect both the solar and longwave radiation with climate implications. However, recent studies (Ramaswamy et al., 1992; WMO, 1992) indicate that the net effect depends not only on latitudes and seasons, but also on the response of the lower stratospheric temperature.

In this study we use a general circulation model (GCM) to calculate the climatic effect due to stratospheric ozone depletion and compare the effect with that due to observed increases of trace gases CO_2, CH_4, N_2O, and CFC's for the period 1980-1990. In the simulations, we use the observed changes in ozone derived from the TOMS data. The GCM used is a version of the NCAR community climate model referenced in Wang et al. (1991). For the present study we run the model in perpetual January and perpetual July modes in which the incoming solar radiation and climatological sea surface temperatures are held constant.

2. Model Experiments

Two sets of equilibrium experiments are conducted. In the first case (c1980), we use the atmospheric trace gas concentrations corresponding to 1980 observed conditions (Houghton et al., 1990; see Table 1). In c1980 the standard 23 level GCM ozone distribution, which is dependent on latitude, height and time of year, will be used to run both perpetual January and July simulations. The column ozone amounts calculated from the GCM ozone distribution correspond well to the TOMS distribution observed during the late 1970's and early 1980's.

The second experiment (c1990) uses the trace gas concentrations for 1990 along with a modified ozone distribution which reflects the observed changes in ozone between 1980 and 1990 from TOMS data. Figure 1 shows the time-latitude distribution of ozone change. The middle and high latitudes in both hemispheres show column ozone losses, while the tropics show a slight increase from February to July. The Antarctic ozone hole is evident, with over a 40% reduction in column ozone at the pole in October and November. Note that the TOMS data does not record ozone amount during polar night, so in these regions no change in column ozone is assumed.

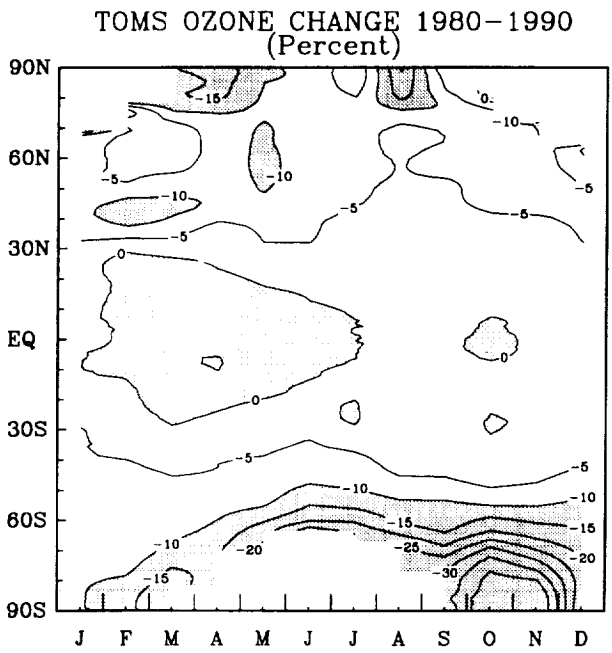

Figure 1. Latitude-time distribution of the percentage change in column ozone measured by TOMS between 1980 and 1990. Months are along the bottom axis. Shaded areas show ozone depletions of greater than 10% and net increases in ozone.

A particular problem for this study was the fact that although the TOMS data produces virtually complete horizonal coverage, it only shows the total column ozone. To derive a consistent ozone dataset for the model, with both horizontal and vertical structure, a procedure had to be devised to use the TOMS data to modify the original GCM ozone distribution. The ozone loss has been observed to occur primarily in the lower stratosphere using both SAGE data and limited ozonesondes (McCormick et al., 1991). To approximate this observed structure, a second-order polynomial was developed in which all of the change in the column ozone occurs in the lowest 11 km of the stratosphere, while retaining the total column ozone changes shown in Figure 1. A climatologically derived tropopause height is used for this calculation and for the subsequent radiative forcing calculations. Figure 2 shows the vertical cross-section of the percentage change in ozone mixing ratio used in the model experiments. In January a maximum ozone loss occurs between 10 and 16 km poleward of about 30 degrees. The structure is similar in July, but more diffuse in the northern hemisphere, while not extending to the pole in the south. These computed changes are applied to the model zonal ozone distribution and the model is run using 1990 trace gas amounts (c1990, see Table 1).

3. Radiative Forcing

We first compare the radiative forcing induced by changes in the trace gases concentrations. For these calculations a time mean equilibrium state taken from the last 90 days of a 300 day simulation from c1980 are used. Table 2 shows the change in the globally averaged radiative forcing for the troposphere-surface system between c1990 and c1980. The change of total radiative forcing, dominated by the longwave radiative forcing, is calculated to be 0.55 and 0.5 Wm^{-2} in January and July respectively. Ozone changes increase the solar flux into the troposphere while slightly reduce the longwave flux and results in a net warming of 0.14 and 0.08 Wm^{-2} for January and July respectively. These changes are comparable to the warming due to the increase of CFC's. Figure 3 shows the zonal distribution of change in radiative forcing, both for ozone and non-ozone gases. Changes in ozone forcing are dominated by the solar forcing changes, so the summer hemispheres show the largest ozone effect,

Table 1. Uniformly-mixed gas concentrations used in GCM experiments.

Experiment	CO_2	CH_4	N_2O	$CFCl_3$	CF_2Cl_2
c1980	337	1.57	0.30	0.16	0.27
c1990	354	1.72	0.31	0.28	0.48

Concentration unit is ppmv for CO_2, CH_4 and N_2O and ppbv for $CFCl_3$ and CF_2Cl_2.

Table 2. Changes in the global mean radiative forcing (Wm^{-2}) of the troposphere-surface system.

	January		July		January	July
	LW	SW	LW	SW	Total	Total
All gases	0.434	0.114	0.438	0.066	0.548	0.504
O_3	-0.040	0.175	-0.042	0.123	0.135	0.081
CFC's	0.117	0.0	0.119	0.0	0.117	0.119

LW refers to longwave radiation and SW to shortwave radiation. All gases include all trace gases listed in Table 1 and ozone. CFC's refers to $CFCl_3$ and CF_2Cl_2. The calculation used the January and July equilibrium states from case 1980.

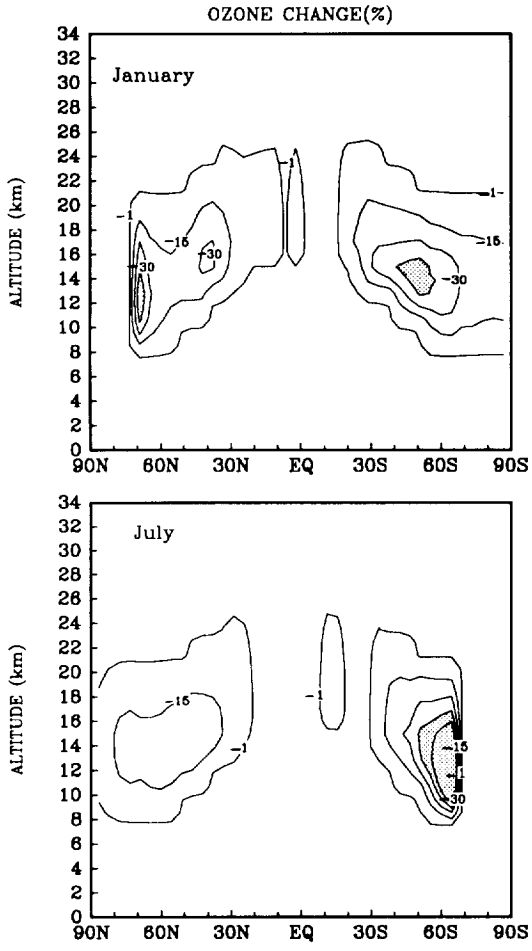

Figure 2. Latitude-height cross-section of the percentage reduction in ozone mixing ratio. The first contour is 1% and the subsequent contour interval is 15%. Shaded areas indicate greater than 45% ozone depletion.

at some latitudes being larger than the non-ozone forcing.

4. Equilibrium Temperature Response

The differences in the zonal mean temperature between c1980 and c1990 are shown in Figure 4. In January, the largest temperature change occurs at high latitudes and is the result of the dynamic response of the polar night stratosphere to the perturbed heating. The temperature is also reduced 1 to 2°C in the lower stratosphere south of 40°S, within the region of maximum ozone loss. The lower stratosphere in the tropics warms about 1°C. In July, there is a more general temperature reduction in the lower stratosphere of 0.5 to 1.0°C. In both months the troposphere is generally slightly warmer, which is consistent with the enhanced net radiative forcing shown in Table 1. Note that the small magnitude of the surface warming is also caused by the fixed sea surface temperature in the two experiments.

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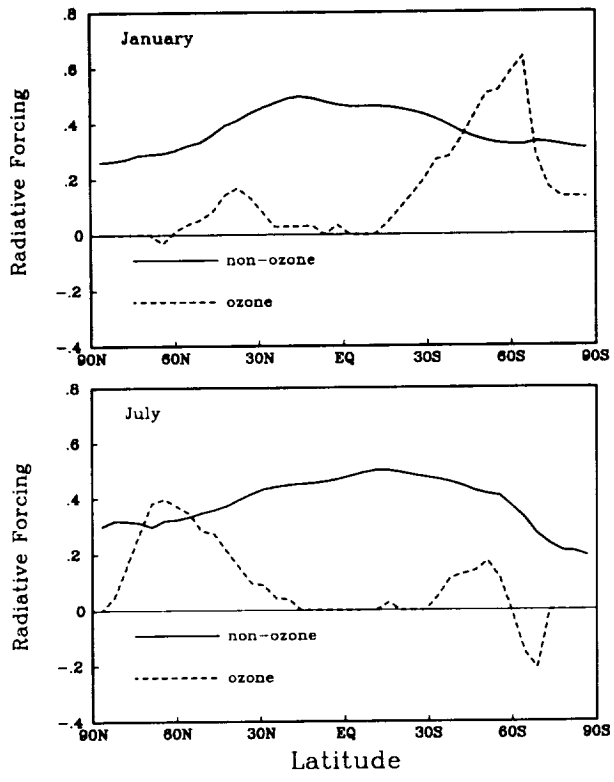

Figure 3. January and July total (both longwave and solar) radiative forcing (Wm^{-2}) due to the 1980 to 1990 increases in all non-ozone gases and that due to ozone changes.

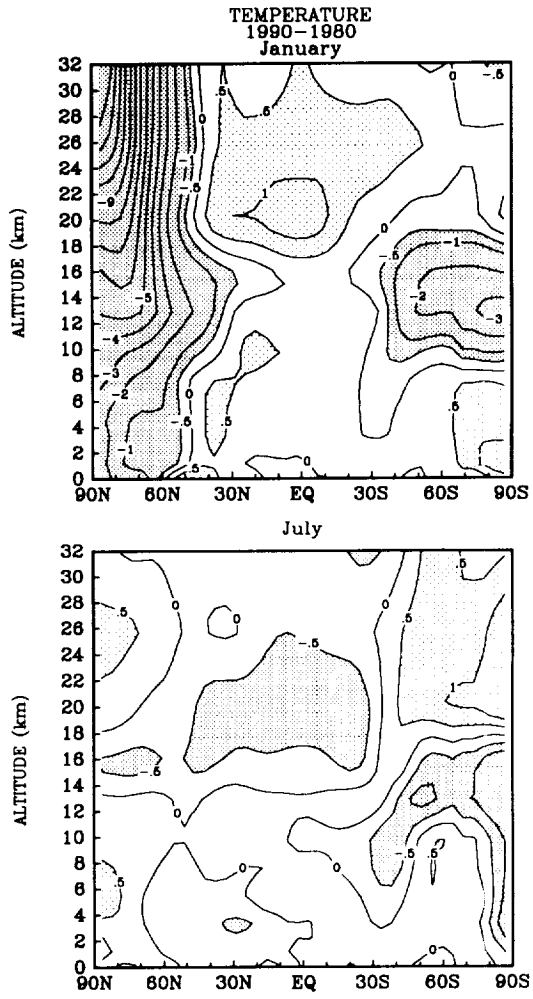

Figure 4. Latitude-height cross-section of the zonal temperature difference between c1990 and c1980 for January and July. Contour interval is every 1°C except for the addition of the -0.5°C and +0.5°C contours. Shaded areas show regions below -0.5°C and above +0.5°C.

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