Reprinted from Nature, Vol. 282, No. 5741, pp. 818-820, December 20/27 1979

Tropospheric ozone and climate

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Analysis[1] of the observed tropospheric ozone distribution has revealed that concentrations in the Northern Hemisphere were larger than in the Southern Hemisphere. This finding and the earlier identification of the necessary photochemical reaction scheme[2] has led Fishman and Crutzen[1] to suggest that an appreciably larger *in situ* photochemical source of tropospheric ozone may exist in the Northern Hemisphere than in the Southern Hemisphere and to speculate that anthropogenic activity might be responsible for a considerable fraction of the larger ozone concentrations in the Northern Hemisphere. In the present study, we examine the influence of tropospheric ozone on climate. Ozone is an optically active gas, which absorbs and emits terrestrial IR radiation in the 8–10-μm region and absorbs solar radiation in the UV and visible. Hence, a change in ozone concentrations will perturb the radiative energy budget of the Earth–atmosphere system which may in turn perturb the climate.

Table 1 O_3 and CO source strengths (in molecules $cm^{-2}\,s^{-1}$)

	Northern Hemisphere	Southern Hemisphere	Ref.
Carbon monoxide			
Industrial CO emissions	19×10^{10}	4×10^{10}	7
CO from methane oxidation	$4–8\times10^{10}$	$4–8\times10^{10}$	5
Ozone			
From *in situ* production	$12–32\times10^{10}$	$5–14\times10^{10}$	5
From stratospheric injection	$5–8\times10^{10}$	$3–4\times10^{10}$	8–10

The ozone–climate problem has been studied mainly by radiative–convective models (see review in ref. 3). The recent study by Ramanathan and Dickinson[4] revealed that a uniform percentage decrease of ozone in the troposphere has about the same net radiative cooling effect on the surface–troposphere system as the same percentage decrease of ozone in the stratosphere. This result is surprising as only about 10% of all ozone resides in the troposphere. The relatively strong sensitivity of the terrestrial radiative energy balance to a tropospheric ozone change is caused by several factors[4], the principal one being that the integrated 9.6-μm band opacity of ozone is a function of atmospheric pressure. More specifically the line halfwidths of the individual rotational lines of the 9.6-μm band are linearly proportional to pressure, at least below 30 km. Consequently, although only ~10% of ozone is present in the troposphere, the IR opacity of tropospheric ozone is nearly the same as that of stratospheric ozone.

Our study will focus on the following aspects of the tropospheric ozone–climate problem: (1) interhemispheric differences in radiative heating of the surface–troposphere system caused by observed interhemispheric ozone distribution differences as reported by Fishman and Crutzen[1]; and (2) the sensitivity of surface–troposphere radiative heating to tropospheric ozone changes. The effects are estimated as a function of latitude and season.

Several recent studies have hypothesised that ozone concentrations may have increased substantially by industrial activities in the Northern Hemisphere and will continue to grow as human activity increases[1,5,6]. Admittedly, except in highly polluted air, it is difficult to prove, by direct observations the *in situ* photochemical production of tropospheric ozone from the oxidation of carbon monoxide and hydrocarbons in the presence of nitric oxide, because of the slow production of ozone in background air[5]. Table 1 summarises some of the studies which suggest that the amount of *in situ* photochemical production of ozone may be considerably larger than injection from the stratosphere[5,7–10] and Table 2 presents data that show the asymmetry in both the CO and O_3 distributions between the hemispheres[5,7,11–13].

Note that there are several reports which either support or refute the importance of tropospheric ozone production[1,5,6,10,14–17]. An indication that tropospheric ozone production does indeed take place in the Northern Hemisphere remote troposphere has recently been obtained from an analysis of simultaneous observation of the concentrations of CO and O_3, generally showing a positive correlation between about 2 and 8 km in the atmosphere[11].

Of similar importance for the production of ozone in the troposphere is the presence of hydrocarbons or carbon monoxide together with nitric oxide. The anthropogenic inputs for both CO and NO are now estimated to be comparable to the natural sources[15–21]. As these emissions probably take place simultaneously, they are of special importance for tropospheric ozone production.

The indications support the view that the asymmetric distribution of ozone between the hemispheres may to a substantial degree be the result of human activities. However, there is no undisputable proof that tropospheric ozone concentrations have generally increased in the Northern Hemisphere over the past decades, as a sufficient long-term record of tropospheric ozone observations is not available.

We use the radiative transfer model described elsewhere[4] which extends vertically from the surface to 54 km. Eight latitude bands are considered from the Equator to 70°, equally spaced at 10° intervals. The model computes the vertical distribution of IR and solar fluxes, using observed Northern Hemisphere distributions of temperature, humidity, clouds at three levels, CO_2, CH_4, N_2O and O_3, and surface albedo as a function of latitude and month. The radiative fluxes computed from these observed distributions are referred to as unperturbed fluxes.

The perturbation calculations discussed here are brought about by prescribing varying amounts of ozone in the troposphere; the change in stratospheric temperatures from the observed value is computed from radiative equilibrium considerations. It was found, however, that the stratospheric temperature changes had a negligible influence on the present results. The distribution of IR and solar fluxes is computed next and the differences between the fluxes in the perturbed and unperturbed troposphere are computed. The flux difference at the tropopause denotes the change in the net radiative energy input to the surface–troposphere system. It is this quantity which is considered here.

The standard profile is based on the observed distribution of ozone concentrations up to 12 km in the Northern Hemisphere as a function of month and latitude. We used three perturbations to our 'standard' profile of tropospheric ozone: (1) the ozone concentrations below 12 km were prescribed according to their distribution in the Southern Hemisphere[1]; (2) ozone concen-

Table 2 Observed hemispheric asymmetry of integrated CO and O_3 in the troposphere

	Asymmetry (Northern/Southern Hemisphere)	Latitudinal domain	Ref.
Carbon monoxide	1.8	90° S–90° N	7
	1.4	53° S–67° N	11
Ozone	1.3	60° S–60° N	5
	1.4	53° S–53° N	11
	1.2	90° S–90° N	12
	1.3	30° S–30° N	13

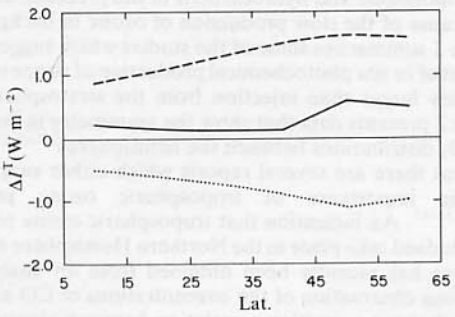

Fig. 1 Annual mean Earth–troposphere radiative heating (ΔI^T) resulting from tropospheric O_3 perturbations. Solid line, heating caused by interhemispheric O_3 differences; dotted line, heating caused by halving tropospheric O_3; dashed line, heating caused by doubling tropospheric O_3.

trations were arbitrarily halved at all levels from the ground to 12 km from the standard distributions; and (3) the standard ozone concentrations were arbitrarily doubled up to 12 km. The first case would simulate the present day heating difference between the two hemispheres solely because more ozone is observed in the Northern Hemisphere than in the Southern Hemisphere troposphere. The second case examines the consequences of the assumption that tropospheric ozone in the Northern Hemisphere has already doubled due to anthropogenic activities. The doubling of tropospheric ozone considered in the third case is a projection into future anthropogenic influence on this gas.

The latitudinal distributions of the annual average values of the surface–tropospheric heating for the three cases are shown in Fig. 1. Considering first the interhemispheric differences, the latitudinal distribution of the difference in radiative heating rates between the Northern and Southern Hemispheres is due mainly to the latitudinal distribution of corresponding ozone concentration differences. At high latitudes, there are also significant seasonal variations in the interhemispheric ozone differences which cause corresponding seasonal variations in interhemispheric heating rate differences (not shown in Fig. 1). For example, at 50°N we calculate interhemispheric heating rates of 0.61, 0.77, 0.48 and 0.35 W m^{-2} for winter, spring, summer and autumn, respectively. This seasonal variability is considerably less at low latitudes. However, more tropospheric ozone measurements in the Southern Hemisphere are needed before attaching too much importance to these seasonal variations. From Fig. 1, the hemispheric and annual mean value of the difference in the radiative heating rates between the Northern and Southern Hemispheres is about 0.3 W m^{-2}. This difference in radiative heating can be related to a corresponding difference in surface temperature, ΔT_s, by letting

$$\Delta T_s = \frac{\Delta I^T}{dI^T/dT_s} \quad (1)$$

where the numerator is the hemispheric average of the change in the energy input to the surface–troposphere system and the denominator is the climate sensitivity parameter which has been extensively discussed during the past decade or so. Values for this parameter[3,22] range between 0.75 and 2.5 W m^{-2} K^{-1}. We will choose the estimate derived by Ramanathan[23], dI^T/dT_s = 1.5 W m^{-2} K^{-1}, which is based on Wetherald and Manabe's[24] general circulation model climate sensitivity analysis. Introducing the difference of 0.3 W m^{-2} in heating between the hemispheres in equation (1), we obtain $\Delta T_s \approx 0.2$ K, that is, the Northern Hemisphere could be warmer than the Southern Hemisphere by about 0.2 K because of the larger amount of ozone in the Northern Hemisphere troposphere. Note that the Northern Hemisphere is indeed warmer than the Southern Hemisphere by abut 1.0 K (ref. 25). There are, of course, other inter-hemispheric differences, such as land and ocean distribution, cloud cover, snow and ice covered areas, and water vapour concentrations which together should be more important in explaining the observed temperature difference between the hemispheres than the differences in tropospheric ozone. Nevertheless, our calculations indicate that differences in ozone may contribute appreciably to the different observed mean temperatures. Applying equation (1) to the second and third cases shown in Fig. 1, we estimate that a halving of the tropospheric ozone concentrations may cool the surface by 0.5 K and that a doubling may warm it by 0.9 K.

We feel that considerations of man's alteration of the global climate must incorporate the possibility that tropospheric ozone concentrations may increase during the coming decades. Our calculations indicate that a doubling of the tropospheric ozone content, calculated by Logan *et al.*[6] to occur by the end of the next century, may increase surface temperatures by nearly 1 K. In a similar period of time a doubling of carbon dioxide in the atmosphere[26] will probably occur, which may result in a 2–3 K temperature increase[3]. A difference between these effects is that, as a result of shorter photochemical lifetimes, the heating resulting from increasing tropospheric ozone would be more concentrated in the Northern Hemisphere, whereas the heating by carbon dioxide would be distributed more evenly between the two hemispheres. It seems possible therefore, that the photochemical production of ozone in the troposphere may enhance the potential climatic consequences of future CO_2 increases by a substantial factor, in particular in the Northern Hemisphere.

The National Center for Atmospheric Research is sponsored by the NSF. Partial support for this research came from US Environment Protection Agency grant 804921-03 to Colorado State University.

Received 26 April; accepted 18 October 1979.

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Printed in Great Britain by Macmillan Production Ltd., Basingstoke, Hampshire