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ON THE POSSIBLE EFFECT OF NO<sub>x</sub> INJECTION IN THE STRATOSPHERE  
DUE TO PAST ATMOSPHERIC NUCLEAR WEAPONS TESTS

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ON THE POSSIBLE EFFECT OF NO<sub>x</sub> INJECTION IN THE STRATOSPHERE  
DUE TO PAST ATMOSPHERIC NUCLEAR WEAPONS TESTS\*

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Abstract

It has been pointed out that the atmospheric nuclear tests in the 1950's and early 1960's injected substantial amounts of NO<sub>x</sub> into the stratosphere. Estimates of the net amount of NO<sub>x</sub> injected and the possible effects on the O<sub>3</sub> distribution through the NO<sub>x</sub> catalytic cycle are made using a time-dependent, parameterized kinetics transport model. By introducing the estimated NO<sub>x</sub> injection per test chronologically, we obtain bounds for the possible observable perturbations of the O<sub>3</sub> distributions. Comparisons made with existing O<sub>3</sub> measurements may provide a direct test of the validity of the NO<sub>x</sub> catalytic cycle in the stratosphere.

I. Introduction

The possible effect of NO injection in the stratosphere by a fleet of supersonic transports has been studied by several investigators.<sup>(1,2)</sup> Of necessity, most of the results are based on model calculations. Recognizing the limitations of such theoretical models, one would like to validate the model-predicted results with some experimental observations. Foley and Ruderman<sup>(3)</sup> have pointed out that the atmospheric nuclear tests of the 1950's and early 1960's may have injected substantial amounts of NO into the stratosphere. It would follow that an examination of the ozone measurements of that period could possibly be used to estimate the significance of NO<sub>x</sub> catalytic destruction of O<sub>3</sub> in the stratosphere. The existence of this past inadvertent NO injection experiment is of great interest, provided that we can adequately analyze the experiment and properly assess the available data.

First we shall briefly examine the NO production due to a typical atmospheric test in the multi-megaton range. Then we shall consider the manner of injection from the series of Russian and American tests and construct a time-dependent NO source function. Given this source function we can carry out a time-dependent calculation to estimate the resulting globally averaged ozone perturbations based on a one-dimensional (vertical) transport model with coupled chemical kinetics. Starting with an unperturbed stratosphere, the time-dependent model was used to calculate the average vertical ozone column density from 1956 to 1972. We have at this time ignored all other possible perturbational effects on the ozone, such as the variations in solar flux associated with the sun spot cycle. Under the conditions assumed, it was found that at most a temporary decrease of less than 4% in the global annual mean would have been caused by the series of atmospheric nuclear tests. When this is compared to natural variations in

ozone, it is clear that until we can identify and properly consider all the other major influences it would be very difficult to directly correlate the effects of nuclear tests with globally averaged ozone observations.

II. NO Production

The major NO production mechanism seems to be the high-temperature chemical equilibrium among NO<sub>x</sub>, N<sub>2</sub>, O, and O<sub>2</sub>, and the subsequent quenching of this equilibrium state by the relatively fast cooling of the hot cloud. The rates of NO production and destruction become negligibly slow at some temperature above 1500°K. The value of this freeze-out temperature depends on the rate of cooling. The net NO produced then depends on the freeze-out temperature T<sub>0</sub>, the initial air mass M<sub>0</sub>, and the extent of uniform mixing in the cloud at freeze out. Although the initial average temperature of the cloud may be very high for multi-megaton yields, the system cools to 2000°K in about 20 to 30 sec and in another 10 sec or less the system is cooled below 1500°K.<sup>(4)</sup> Due to the relatively slow decrease in temperature in the early states of the cooling process, it is reasonable to assume that T<sub>0</sub> ≤ 2000°K. This is the temperature adopted by Foley and Ruderman. By assuming that the hot air contains one-third of the initial energy, and further assuming an entrainment factor of six and uniform mixing, they estimated the net NO production per test to be

$$N_{NO} \approx 2 \times 10^{32} Y_{MT}$$

where Y<sub>MT</sub> is the energy yield of the nuclear device measured in megatons. Of course, this number could be reduced by a factor as large as six, depending on the validity of the uniform mixing assumption. Later on Johnston et al.<sup>(5)</sup> recalculated the production term and explicitly included this factor of six:

$$N_{NO} \approx (0.17 - 1.0) \times 10^{32} Y_{MT}$$

In a recent study by Robert Gunton et al.<sup>(4)</sup> at Lockheed, this number was computed under a variety of situations. Based on their results, we may estimate

$$N_{NO} \approx 5 \times 10^{31} Y_{MT} \quad (1)$$

To a certain extent the computational results of the Lockheed calculations have been validated. Even in this calculation, uniform mixing of the entrained air is assumed, but the entrainment ratio at 2000°K is only in the range 2-4. We shall

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adopt Eq. (1) as the NO production number. It should be noted that the validity of using 2000°K as the freeze-out temperature, as used by Foley & Ruderman, and Johnston et al., has been verified by Gunton et al. Further cooling and entrainment of ambient air did not affect this number significantly. (4)

### III. Stratospheric NO Injection

The great interest in megaton-range tests lies with the fact that, for surface test above one megaton, the stabilized cloud is usually in the stratosphere. Assuming a pancake-shaped cloud, Foley and Ruderman<sup>(3)</sup> gave a parameterized fit to the cloud top  $H_T$ , cloud bottom  $H_B$ , and the radius R:

$$H_T = 21.64 Y_{MT}^{0.2} \text{ km}, \quad (2)$$

$$H_B = 13.41 Y_{MT}^{0.2} \text{ km}, \quad (3)$$

and

$$R \text{ (30 min old)} = 42.67 Y_{MT}^{0.5} \text{ km}. \quad (4)$$

It should be said that these formulae may not represent the best description of those quantities, but we believe they are as meaningful as other available methods for placing the clouds (see for example Seitz et al. (6)).

From Eqs. (1), (2), (3), and (4) we see that the initial NO concentration could be as high as

$$[NO]_0 = 1.06 \times 10^{12} Y_{MT}^{-0.2} \text{ molecules/cm}^3. \quad (5)$$

Based on pure chemical kinetics, this high concentration of NO will destroy almost all of the ozone in the cloud within seconds. But the cloud covers only a very small area. Furthermore, it will be transported around the globe rather rapidly (approximately once around every 3-4 weeks or sooner). If we assume no further expansion and dilution, then the relative area of the cloud and of the earth are approximately  $[1.11 \times 10^{-5}, 1.11 \times 10^{-4}, 5.55 \times 10^{-4}]$ , corresponding to 1, 10, and 50 megaton tests. Clearly, any chance of detection of this perturbation by a given ozone station would be small, and as a result of the period of circulation, the deviant measurements would be weeks apart. This means that it would be unlikely that this perturbation in the data from any particular ozone station could be statistically evident. More realistically, we note that once the cloud passed a particular station, it might be weeks before it was brought back by the zonal circulation process, and by that time the atmospheric diffusion process would have reduced the  $NO_x$  concentrations by factors of thousands. Let us consider only the eddy diffusion process. The vertical and horizontal eddy diffusion coefficients ( $K_z$ , and  $K_x = K_y$ ) are commonly estimated to be  $K_z = 10^4 \text{ cm}^2/\text{sec}$  and  $K_x = K_y = 5 \times 10^9 \text{ cm}^2/\text{sec}$ . Assuming the initial  $[NO]$  cloud to be a Gaussian with the standard deviations  $\sigma_z^2 = \frac{1}{2} (H_T - H_B)$  and  $\sigma_x^2 = \sigma_y^2 = R$ , then the growth in time of these parameters<sup>(7)</sup> is

$$[\sigma_z(t)]^2 = [\sigma_z^0]^2 + 2K_z t$$

and

$$[\sigma_x(t)]^2 = [\sigma_y(t)]^2 = [\sigma_x^0]^2 + 2K_x t,$$

where  $t$  is measured in seconds. Given the initial  $\sigma_z^0$ ,  $\sigma_x^0$ , and  $\sigma_y^0$  under consideration we see that for 7 days  $< t < 90$  days,

$$\sigma_z = \sigma_z^0, \text{ but } \sigma_x = \sigma_y = \sqrt{2K_x t}.$$

If we take this  $\sigma_x$  to be the size of the one-week-old cloud, then it still will not stay over any particular ozone station for more than a day or two at a time, and the cloud area is still only about 0.1% of the global surface area. Since very little precise information concerning the whereabouts of most of the stratosphere nuclear clouds is available, this irregularity of possible detectable perturbations and the irregularity in the daily measurement programs of many stations couple to make an analysis of the short-time perturbation from any single cloud a nearly impossible task. However, since the nuclear tests were carried out over a period of time and the total amount injected is indeed a large amount, one may hope to analyze globally averaged ozone data during this period (1960-1970) and possibly correlate any long-term effects with the nuclear events.

In view of this, we will use a global averaged NO injection function,

$$[NO] = \frac{N_{NO}}{(H_T - H_B) \times A} \text{ molecules/cm}^3,$$

where  $A$  is the global surface area. This reduces the source concentration, Eq. (5), by a factor of  $10^5$  or more. Because all of the tests were carried out in the northern hemisphere and because we prefer to overestimate rather than underestimate the globally averaged source rates, we will use  $A/2$  instead of  $A$ . This in effect constrains all of the injected NO to the northern hemisphere. In an effort to approximate the residence time of each nuclear cloud we have retained the initial stabilization heights, Eqs. (2) and (3). This will overestimate the local concentration of  $NO_x$  shortly after the test, and it will cause a transient overreaction of the global averaged  $O_3$  column to the NO<sub>x</sub> injection. This transient will have almost no effect on the yearly averaged results.

Given the dates of the nuclear tests  $t_i$ , the corresponding yield  $Y_i$ , and the cloud-stabilization altitudes, we can construct the time- and altitude-dependent source functions

$$S_{NO}(t, z) = \sum_{i=1}^I 2.36 \times 10^7 Y_i^{0.8} \delta(t - t_i) \times H(z - 13.41 Y_i^{0.2}) H(21.64 Y_i^{0.2} - z), \quad (6)$$

where  $\delta(\cdot)$  is the delta function and  $H(\cdot)$  is the unit step function. In order to simplify the numerical computation procedures slightly, the various test dates were grouped into weekly intervals. All the dates and yields are deduced from

unclassified literature. (6,8,9) This source function is now used in a time-dependent, one-dimensional (vertical) chemical kinetics transport model.

#### IV. One-Dimensional Kinetics Transport Model

Due to the periodicity of the dynamic variables in the angular directions, the time-dependent globally averaged vertical distribution of any minor chemical species  $c_i$  in the stratosphere may be described by the kinetics transport equation<sup>(10,11)</sup>

$$\frac{\partial c_i}{\partial t} = \frac{\partial}{\partial z} \left[ K_z \frac{\partial c_i}{\partial z} + \left( \frac{1}{T} \frac{\partial T}{\partial z} + \frac{1}{H} \right) c_i \right] + P(c) - L(c)c_i + S_i, \quad (7)$$

where

- $c_i$  = the  $i$ th minor chemical species,
- $K_z$  = the altitude-dependent mean vertical eddy diffusion coefficient,
- $T$  = the altitude-dependent average temperature,
- $H$  = the altitude-dependent average scale height factor,
- $z$  = the distance above earth surface,
- $P(c)$  = the production of  $c_i$  due to photochemical interaction of the other  $c_j$ 's,
- $L(c)c_i$  = the loss of  $c_i$  due to photochemical interaction, and
- $S_i$  = any other possible sources or sinks of  $c_i$ .

This equation is actually a simple generalization of the extreme cases of pure kinetics and pure dynamic transport. Nevertheless, it directly couples kinetics and transport to a significant degree, which is germane to the recognition of the stratosphere as the transition region between the transport-dominant troposphere and the kinetics-dominant mesosphere. Thus Eq. (7) can yield, and has yielded, valuable information about processes in the stratosphere. (1-2,10-14) We shall set the region of interest to be  $0 \text{ km} < z < 55 \text{ km}$ . The boundary conditions are constant flux at the upper boundary and fixed concentrations at the lower boundary. The eddy diffusion coefficients  $K_z$  are seasonal, globally averaged values based on Gudiksen et al.<sup>(15)</sup> The temperature and air density profiles are taken from the U.S. Standard Atmosphere (1962). In this report we shall not discuss in detail the numerical techniques used in solving Eq. (7). It has been reported elsewhere,<sup>(16)</sup> and an early written version is in the First Annual Report, DOT-CIAP Program from Lawrence Livermore Laboratory. It is sufficient to say that the numerical method is fully implicit, second-order accurate in  $z$ , and variable-order accurate in  $t$  (first through fifth order) and that the accuracy of the solution at every time step is internally checked by the program through a theoretically established error estimation procedure.

In this model we consider the species  $O(^1D)$ ,  $O(^3P)$ ,  $O_3$ ,  $NO$ ,  $NO_2$ ,  $N_2O$ ,  $HNO_3$ ,  $HO$ ,  $HO_2$ ,  $H_2O_2$ ,  $N$ , and  $H$ . The vertical distribution of  $H_2O$  and  $CH_4$  are interpreted from the literature and assumed fixed in time.<sup>(17,18)</sup> Furthermore,  $O(^1D)$ ,  $N$ , and  $H$  are assumed to be in instantaneous chemical equilibrium. The boundary conditions are estimated from the literature, as were those already cited and the tropospheric estimates of Levy.<sup>(19)</sup> The reactions considered are listed in Table 1, which is basically the list given by H. Johnston in CIAP monograph III, The Perturbed Stratosphere (1990). All the reaction rates are either from the LLL Atmospheric Data Library, (Edition 4)<sup>(20)</sup> or the list provided by H. Johnston. Figures 1, 2, and 3 give some of the computed profiles as compared to other theoretical or measured profiles. From these and other comparison results we believe that our model does provide a meaningful representation of the unperturbed stratosphere.

#### V. Results from the Nuclear NO Injection Experiment

Using the  $[NO]$  source function, Eq. (6), and starting from an unperturbed state as provided by the model, we carried through a time-dependent calculation from May 1956 to May 1973. We have carried the calculations back to 1956 to establish a proper initial state for the major injection in the 1961-1962 test series. Indeed, it was found that the early, relatively small tests of the 1950's have a residual effect as late as 1961, when the last series of tests began (Fig. 4). Figure 4 gives the monthly values of the percent of change in total ozone column from 1956 to 1968. Since we have not included the effect of the annual variation of the northern hemispheric average  $O_3$ , a direct month-by-month comparison to the data will not be meaningful. But a direct comparison with the yearly averaged value can be made. In Fig. 5 we have presented yearly averaged values and have compared them to the northern hemispheric  $O_3$  trend analysis of Johnston et al.<sup>(5)</sup> From Fig. 4, we see that in 1970 the effect of the nuclear tests has become very small, and since Johnston's analysis only covers the period 1960-1970, we decided to use 1970 as the reference point. Although the apparent agreement is striking, we believe that one should not overemphasize this point. The general trend and the fact that the predicted change is less than the change indicated in the ozone data is of real interest. Decreasing the  $NO$  production per test would decrease the magnitude of the dip in 1963, but the trend would remain the same. Since this predicted change due to nuclear tests is less than the total change in the data, one must, in order to ascertain this effect, remove all other cyclic influences to the extent that an aperiodic deviation of a few percent will be statistically significant. As a check on the reasonableness of our transport model we have computed the excess  $NO + NO_2$  in the stratosphere and have compared it with the Sr-90 and excess C-14 data from Johnston et al.<sup>(5)</sup> This is presented in Fig. 6. Because of the lack of a satisfactory reference point before tests began, we have normalized all the curves at 1969 and on the same semilog scale. The general agreement seems to validate the accuracy of the injection function, Eq. (6), and the residence time of our model.

Finally, in Fig. 7 we have the perturbed profiles corresponding to maximum decrease in  $O_3$  column. It is of interest to note that a depletion of  $O_3$  at high altitude will give more ultraviolet

Table 1 List of reactions included in the one-dimensional model

1. $O_2 + hv \rightarrow O + O$	$J_1$
2. $O_3 + hv \rightarrow O_2 + O$	$J_2$
3. $O_3 + hv \rightarrow O_2 + O(^1D)$	$J_3$
4. $O(^1D) + M \rightarrow M + O$	$3.2 \times 10^{-11}$
5. $O + O_2 + M \rightarrow O_3 + M$	$1.07 \times 10^{-34} e^{510/T}$
6. $O + O_3 \rightarrow O_2 + O_2$	$1.9 \times 10^{-11} e^{-2300/T}$
7. $NO_2 + hv \rightarrow NO + O$	$J_4$
8. $NO + O_3 \rightarrow NO_2 + O_2$	$9 \times 10^{-13} e^{-1200/T}$
9. $NO_2 + O \rightarrow NO + O_2$	$9.1 \times 10^{-12}$
10. $N_2O + hv \rightarrow N_2 + O(^1D)$	$J_5$
11. $N_2O + O(^1D) \rightarrow N_2 + O_2$	$6.6 \times 10^{-11}$
12. $N_2O + O(^1D) \rightarrow NO + NO$	$6.6 \times 10^{-11}$
13. $NO + hv \rightarrow N + O$	$J_6$
14. $N + O_2 \rightarrow NO + O$	$1.02 \times 10^{-14} T e^{-3130/T}$
15. $N + NO \rightarrow N_2 + O$	$5.1 \times 10^{-11} e^{-170/T}$
16. $N + NO_2 \rightarrow NO + NO$	$6 \times 10^{-12}$
17. $N_2 + O(^1D) + M \rightarrow N_2O + M$	$2.8 \times 10^{-36}$
18. $NO_2 + N \rightarrow N_2O + O$	$9 \times 10^{-12}$
19. $O(^1D) + H_2O \rightarrow HO + HO$	$2.8 \times 10^{-10}$
20. $O(^1D) + CH_4 \rightarrow HO + CH_3$	$2.4 \times 10^{-10}$
21. $HNO_3 + hv \rightarrow HO + NO_2$	$J_7$
22. $HO + O_3 \rightarrow HO_2 + O_2$	$1.3 \times 10^{-12} e^{-956/T}$
23. $HO + O \rightarrow O_2 + H$	$4.2 \times 10^{-11}$
24. $HO_2 + O_3 \rightarrow HO + O_2 + O_2$	$1.0 \times 10^{-12} e^{-1875/T}$
25. $HO_2 + O \rightarrow HO + O_2$	$1 \times 10^{-11}$
26. $H + O_2 + M \rightarrow HO_2 + M$	$1.1 \times 10^{-32} e^{407/T}$
27. $H + O_3 \rightarrow HO + O_2$	$2.6 \times 10^{-11}$
28. $HO_2 + HO_2 \rightarrow H_2O_2 + O_2$	$3 \times 10^{-11} e^{-500/T}$
29. $HO + HO_2 \rightarrow H_2O + O_2$	$2 \times 10^{-10}$
30. $HO + NO_2 + M \rightarrow HNO_3 + M$	$2 \times 10^{-11} [M]/(3 \times 10^{20} e^{-750/T} + 2 [M])$
31. $HO + HNO_3 \rightarrow H_2O + NO_3 + hv$ $\quad \quad \quad \rightarrow H_2O + NO + O_2$	$1.3 \times 10^{-13}$
32. $H_2O_2 + hv \rightarrow 2HO$	$J_8$
33. $H_2O_2 + HO \rightarrow H_2O + HO_2$	$1.7 \times 10^{-11} e^{-910/T}$

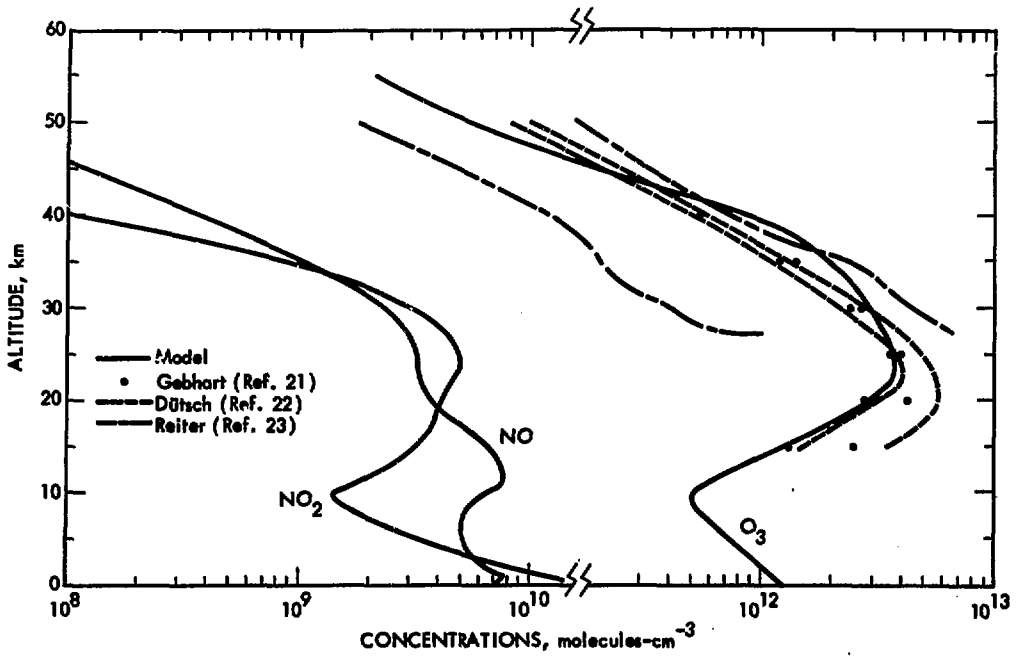


Fig. 1 Vertical distributions for  $\text{NO}_2$ ,  $\text{NO}$ , and  $\text{O}_3$ .

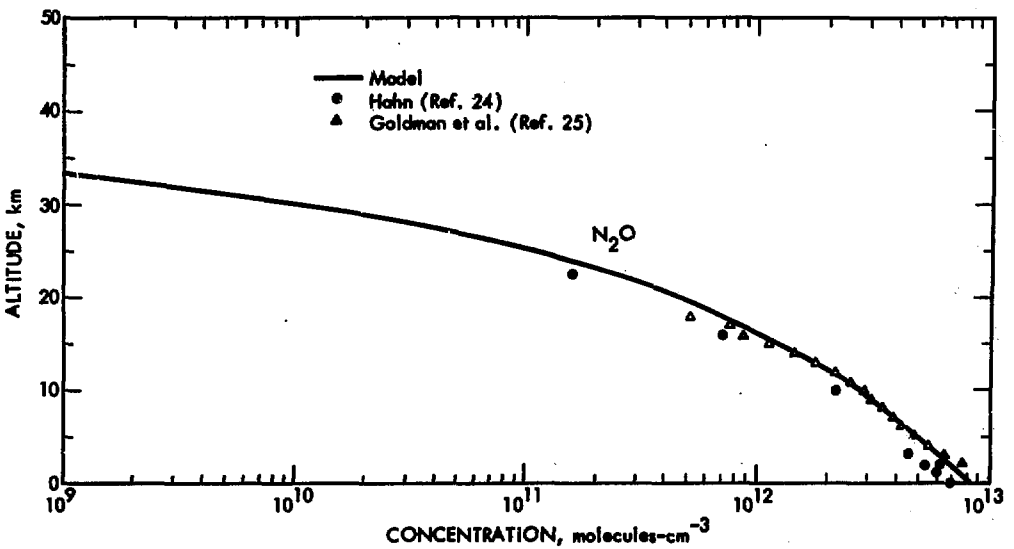


Fig. 2 Vertical distributions for  $\text{N}_2\text{O}$ .

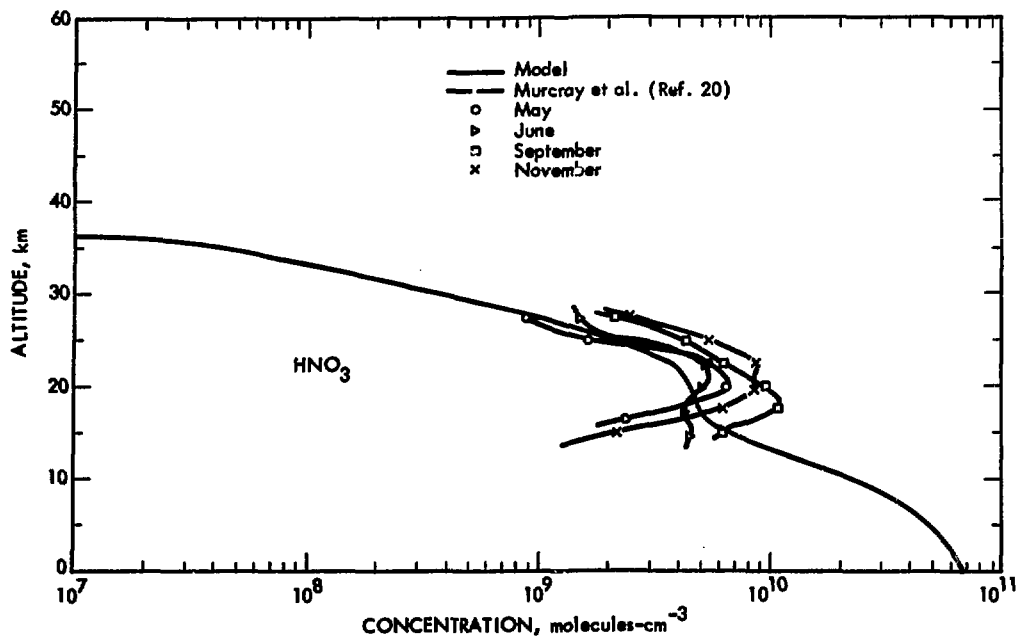


Fig. 3 Vertical distributions for HNO<sub>3</sub>.

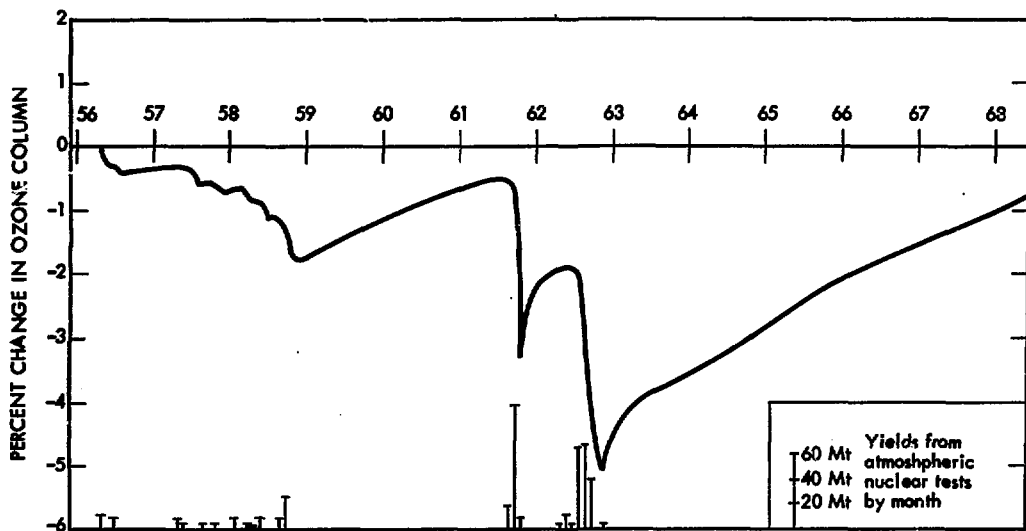


Fig. 4 Model-predicted percent changes in the northern hemispheric average ozone column due to past atmospheric nuclear tests.

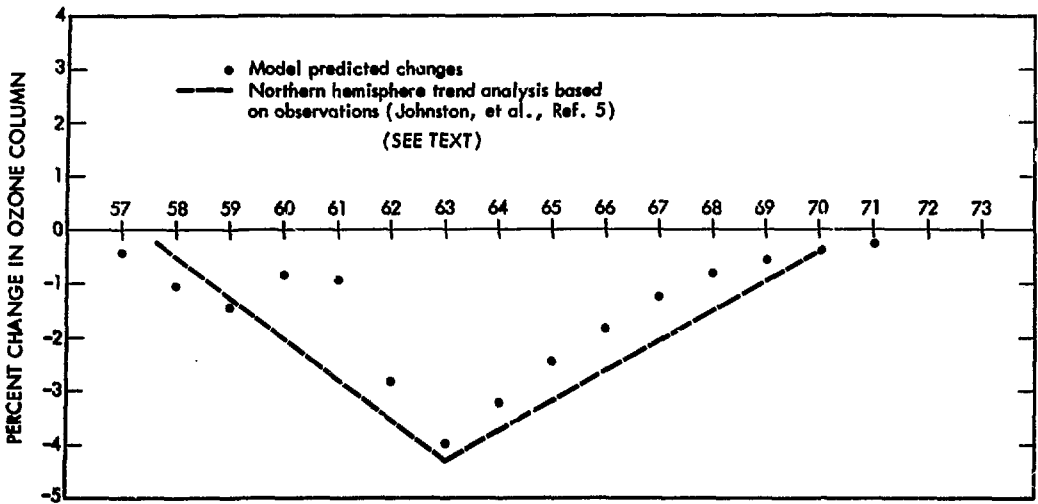


Fig. 5 A comparison of model-predicted changes in the yearly averaged northern hemispheric ozone column and trend analysis results based on observations.

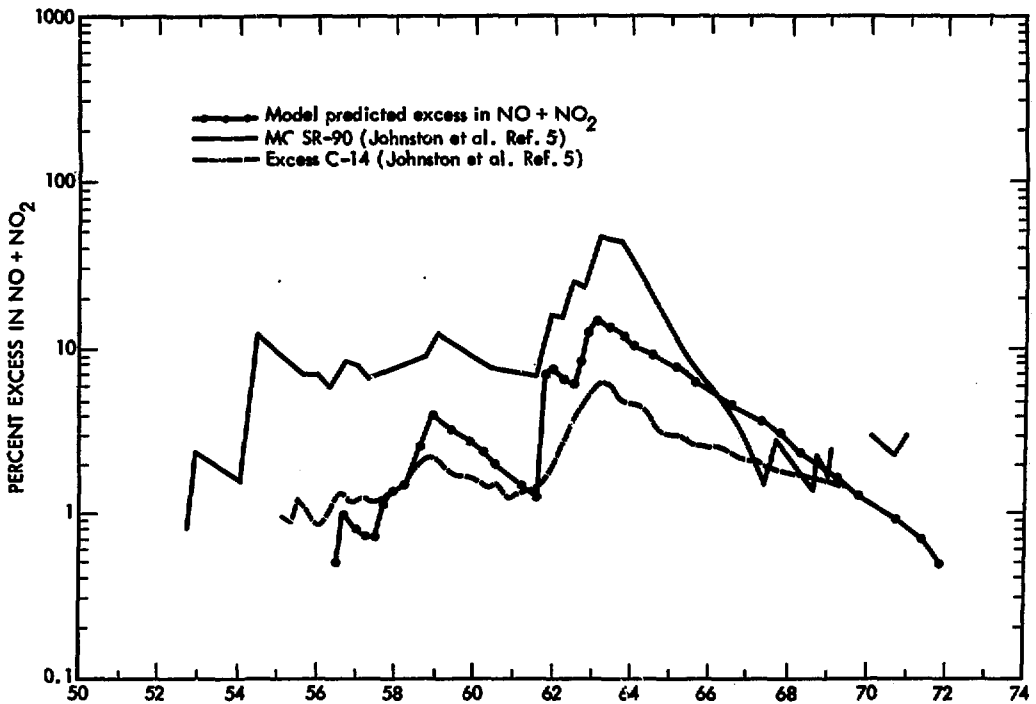


Fig. 6 A comparison of excess C-14, Sr-90, and model-predicted excess NO + NO<sub>2</sub> in the stratosphere from 1956 to 1970. The C-14 and Sr-90 curves have been normalized to the NO + NO<sub>2</sub> curve for 1969.



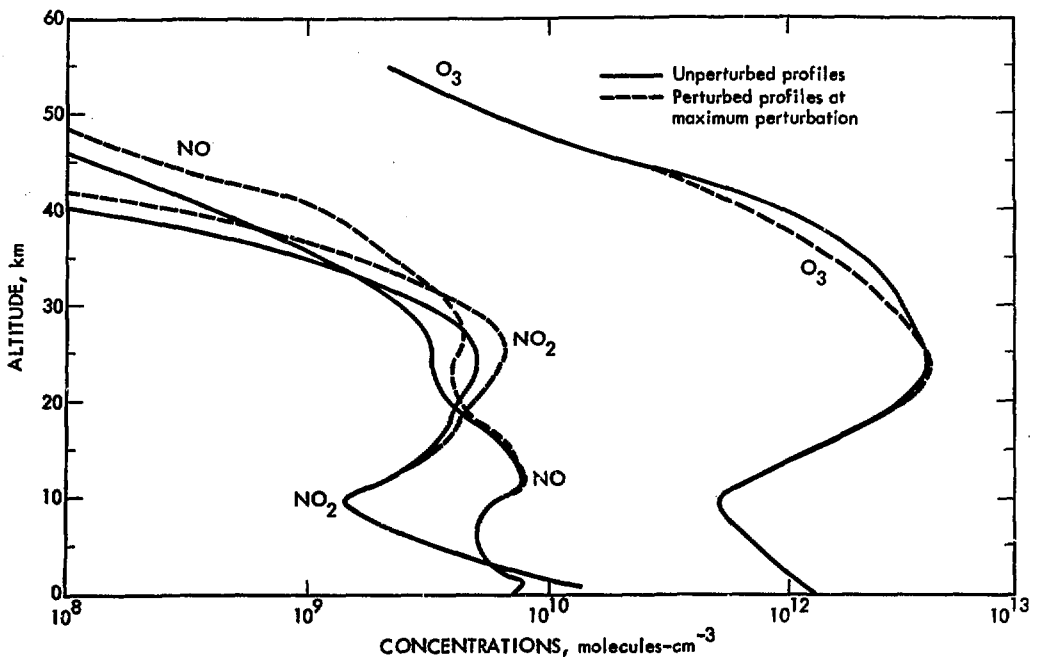


Fig. 7 Vertical distributions comparing unperturbed and perturbed profiles.

at lower altitude and hence more  $O_3$  production at this level. Unfortunately, this self-compensating effect does not seem to be fast enough to reduce the possible effect of NO injection from SST's significantly, especially if the  $NO_x$  injection is in the lower stratosphere.

#### VI. Conclusions and Comments

Our model calculation has indicated a maximum decrease of 4% in the northern hemispheric annual average ozone column from 1956-1972, and for the following reasons we believe this to be an overestimation. First we have not included diurnal or seasonal variations in solar intensities. The former will reduce the time duration for the effective  $NO_x$  catalytic reduction of  $O_3$  by almost one-half. Since most of the Soviet tests were done during the polar night, the injected NO would not be fully active until several months later, while in the meantime the net  $NO_x$  would be reduced by transport processes. This is especially important because the scaling law for the height of the stabilized cloud tends to overestimate for the large yield tests. Hence we may have overestimated the residence time of these events. Furthermore, by constraining all the injected NO to the northern hemisphere we get a modest overestimation factor. Of course, our NO production rate per test might be too low, but we believe that when all factors have been properly considered, even a factor of two in source rate will not lead to significantly different conclusions.

It should be pointed out that although the nuclear tests in the early 1960's produced an amount of NO comparable to the yearly production from SST's estimated in the literature, the net

effect on the stratosphere is quite different because of the nature of the injection process. The nuclear injection process is a transient perturbation and the net amount of  $O_3$  destruction due to NO, is actually decreasing with respect to time. But continuous injection from SST's will reach a new steady state, and the resulting NO distribution will have realized its maximum effect. It is this final state that is crucial climatologically. Consequently, our calculation on the possible effect of past nuclear tests does not answer the more important SST question. However, given the present results, it can be stated that the effects of the nuclear tests in the early 1960's are not inconsistent with the observed ozone behavior during the 1960's.

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