Stratospheric Ozone Destruction by Aircraft-Induced Nitrogen Oxides

Three-dimensional simulations of ozone distributions are used to estimate ultraviolet radiation fluxes.

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During the last several years much concern has been voiced about the climatological impact of a fleet of operational supersonic transports (SST's) on the environment and ultimately on the quality of life in the biosphere as we know it today. Recent attention has focused on the problem suggested by Crutzen (1) and Johnston (2) of SSTinduced nitrogen oxides ($NO_x = NO +$ NO_2) catalytically reacting with oxygen allotropes to significantly reduce the total ozone (O_3) content of the atmosphere. Although O3 constitutes less than one-millionth of the mass of the atmosphere, it plays an important role in heating the stratosphere and in reducing the amount of solar ultraviolet (UV) radiation that reaches the ground. Thus, significant depletion of stratospheric O_3 would not only lead to a modification of stratospheric climate but could also induce undesirable biological effects at the surface.

The production of atmospheric O_3 depends upon the dissociation of molecular oxygen by sunlight to produce atomic oxygen, the first of the classical Chapman oxygen reactions (3). This dependence on solar radiation leads to maximum O_3 production rates in the tropical stratosphere at an altitude above 30 kilometers. However, the largest total O₃ amounts are found not at the tropical latitudes where they are produced but at high latitudes (4), particularly in the spring, with maximum concentrations occurring at an altitude of approximately 18 km. Transport processes are clearly important.

The fundamental problem in estimating the distribution of O_3 in the unperturbed stratosphere (that is, without the effects of aircraft operations) has been that the Chapman oxygen reactions predict too much O_3 by about a factor of 2. Recently, computations based on the use of simple vertical diffusion models in which the catalytic cycle of chemical reactions involving NO_x has been added to the Chapman set have verified that, if NO_x is present, the total O₃ content will be significantly reduced (5). However, the anomalous latitudinal and seasonal changes in the O₃ concentrations complicate a satisfactory quantitative verification of the results from these one-dimensional vertical diffusion models. In view of the obvious importance of atmospheric transport processes for the distribution of O_3 , computations utilizing two- or three-dimensional models are clearly preferable.

Aircraft engines produce NO_x . This is of particular concern when aircraft fly in the stratosphere because of the relatively long time periods required for the net removal of NO_x at these altitudes. Such long residence times are primarily due to the inherent vertical thermal stability of the stratosphere. The principal removal mechanism for NO_x in the stratosphere appears to involve its conversion to hydrogen nitrate (HNO_3) with subsequent transport into the troposphere and rainout as nitric acid. Thus, in order to estimate the destructive effect of NO_x on O_3 , one must account not only for the chemical balance between the NO_x species and HNO₃ but also variations in their seasonal and spatial distributions resulting from large-scale transport processes.

Model Description

In this article we report the first major results from a three-dimensional dynamical-chemical model applied to the SST-NO_x problem, the essential details of which have been described by Cunnold et al. (6). Briefly, all model variables are represented along 26 horizontal surfaces equally spaced in units of the logarithm of atmospheric pressure between the ground and approximately 72 km. On each surface, horizontal variations are allowed through global expansions in two-dimensional spherical harmonic series with the smallest resolutions approximately equivalent, at mid-latitudes, to a horizontal grid spacing of 2000 km in the longitudinal direction and 2900 km latitudinally. The dynamical and thermodynamical aspects of the model are treated through the use of the quasi-geostrophic balance set of equations (7) to which representations for large-scale heating, surface orography, and effects due to small-scale diffusion and surface drag have been added. This system has the advantage of being integrable with time steps of an hour, whereas more elaborate general circulation models require much shorter time steps.

The chemical reactions contained in the model consist of the Chapman oxygen reactions

$$O_2 + h\nu \rightarrow 2O$$

$$O + O_2 + M \rightarrow O_3 + M$$

$$O_3 + h\nu \rightarrow O_2 + O$$

$$O + O_3 \rightarrow 2O_2$$

in which M is any background molecule, coupled with three NO_x reactions

$$NO + O_3 \rightarrow NO_2 + O_2$$
$$NO_2 + O \rightarrow NO + O_2$$
$$NO_3 + h\nu \rightarrow NO + O$$

where hv is a photon. In the current version of our model, in addition to atmospheric winds and temperature, only the O_3 distribution is predicted. Thus all other species distributions must be specified or determined diagnostically. This is particularly troublesome for NO_{x} since its observed distributions are not well known and yet, in order to assess the influence of SST-produced NO_x , we must determine realistic values both for the unperturbed and for the perturbed situations. As we shall describe below, the results of two-dimensional model calculations are used to specify this quantity.

Although our model is global, it is convenient to assume that the two hemi-

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spheres are geographically similar. This is done primarily because of the uncertainties in the tropospheric heating data for the Southern Hemisphere but it also has the advantage that the approach to a statistically stationary behavior of O_3 , in the presence of a pronounced annual cycle at a fixed latitude, can be examined at intervals of 6 months rather than 12 months. Thus, our model Southern Hemisphere represents Northern Hemisphere conditions with a 6-month time lag. With this approach, realistic interactions between the hemispheres have been retained.

Time integrations for both the unperturbed and the perturbed cases were carried forward for a period of 3 years in order to obtain quasi-equilibrium states. Statistical climatological results were then compiled from data obtained in year 3. In each case the initial dynamical configuration consisted of an atmosphere in a state of rest. The only difference between the unperturbed and perturbed conditions was the prescribed NO_x distributions to be described below.

Model Dynamical Climatology

The dynamical results of the two calculations (unperturbed and perturbed cases) are quite similar, and we will not attempt to differentiate between them here. Climatological winter and summer mean zonal winds obtained from the model results exhibit the essential large-scale characteristics of atmospheric motion. For example, a winter-hemisphere upper tropospheric westerly jet with maximum wind speeds of 40 meters per second is found at an altitude of 10 km in the subtropics (although about 15° too far toward the equator). In the upper part of the stratosphere, a westerly wind belt exists in winter and easterlies prevail during the summer. The model winter-summer meridional circulation pattern is shown schematically in Fig. 1 and is based on meridional stream function data. Here each hemisphere exhibits a tropospheric three-cell circulation pattern that rather abruptly changes to two cells in the stratosphere. The direct circulation cell of the winter stratosphere



Fig. 1. Mean winter-summer meridional circulation patterns averaged over the last season of unperturbed model year 3. The schematic representation is based on meridional stream function data. is particularly dominant and indeed extends from about 25°S to 35°N, covering nearly all of the tropical latitudes. This results in a large stratospheric mass transport across the equator from the summer hemisphere to the winter hemisphere in accord with observational evidence and provides a satisfactory basic meridional flow field for the initial poleward transport of stratospheric O_3 away from the equatorial production regions.

Input of NO_x and O_3 Distributions

Since our model calculations do not explicitly predict NO_x distributions, we are forced to provide these values from independent sources. Specifically, because NO is diagnostically related to NO₂, we need only provide temporal and spatial distributions for NO₂. In the calculation for the unperturbed stratosphere we have adopted NO₂ values as predicted by a two-dimensional model calculation of Hesstvedt (8). These NO_2 values lie within the range of uncertainty (about a factor of 2) of the limited number of measured values of NO_2 , and we therefore consider them adequate as a first estimate. This adopted NO₂ distribution, at its midsummer and midwinter extremes, is shown in Fig. 2a. We generated seasonal changes in NO₂ by applying a simple annual sinusoidal variation to the data shown.

Using this NO₂ profile, we carried out a 3-year integration of the complete model to simulate the unperturbed stratosphere. As a basis for comparison with our model O₃ results, Fig. 3a shows the total columnar O₃ values observed by Dütsch (4) as a function of latitude



Fig. 2. (a) The unperturbed solstitial NO_2 distribution (in parts per billion by volume) according to Hesstvedt (8). (b) The perturbation in NO_2 during Northern Hemisphere summer.

and season for the Northern Hemisphere. A similar picture, averaged over year 3 of the unperturbed model results, is contained in Fig. 3b. The model has correctly simulated the gross features of both the seasonal and latitudinal variations in total O_3 , although the resulting spring maximum generated by the model in the polar regions is smaller than the observed value, reaching just over 400 Dobson units (9) compared to more than 440 observed.

A second 3-year integration was carried out to simulate the stratosphere as it might appear when perturbed by an additional source of NO_x due to SST operations. To calculate the distribution of this additional NO_x , and thus to be able to obtain the additional NO₂ values required as input for the three-dimensional model, we have designed a twodimensional transport model specifically for this purpose (10). This model incorporates an enlarged set of chemical reactions considered important for the production and destruction of NO_x species in the stratosphere and requires more than 10 years of integration to reach a state of quasi-equilibrium. This transport model was not available in time to provide NO_x values for the unperturbed stratosphere calculations, and therefore its use here is limited to providing the additional NO2 concentrations resulting from a fleet of SST's. Although there is a great deal of uncertainty about the number and type of SST aircraft that will fly in the stratosphere, we have assumed, as a reasonable upper limit, an NO_x emission rate of 1.8×10^6 metric tons per year, to be released in a corridor centered at 45°N and an altitude of 20 km. The corridor extends 15° in latitude and 1 km in height. Although some may consider this emission rate high [Broderick et al. (11) estimate a "first guess" upper limit of 0.9×10^6 metric tons per year for a fleet of SST's in 1990], it is equivalent to the upper bound rate projected in Grobecker (12) for 1997 but represents only 20 percent of the upper limit projected for 2010. The results of the two-dimensional calculation for the NO₂ perturbation are shown in Fig. 2b (Northern Hemisphere summer). A tenfold increase in NO₂ occurred in the region of injection, but at higher altitudes (above 27 km) the increase amounted to approximately 20 percent in the Northern Hemisphere and 10 percent in the Southern Hemisphere. These perturbations were added to the NO2 values assumed for the unper-11 APRIL 1975

turbed stratosphere, and, as before, we generated seasonal changes by assuming a sinusoidal annual variation. The NO₂ distribution to be used for the perturbed stratospheric O_3 calculation was thus produced.

As expected, the effect of this increased amount of NO_2 in the perturbed stratosphere, as obtained from the two-dimensional calculations, can be seen in the O_3 distributions simulated by the three-dimensional model. Figure 4 shows the seasonally averaged distributions of total columnar O_3 as a function of latitude calculated for both the perturbed stratosphere (solid lines) and the unperturbed stratosphere (dashed lines). On a global scale the perturbed stratospheric O_3 depletion amounts to about 12 percent. However,





Fig. 3 (left). Columnar O_3 distribution (in Dobson units) in the Northern Hemisphere as a function of season and latitude: (a) as determined by Dütsch (4) from observations and (b) as predicted during year 3

in our unperturbed model. Fig. 4 (right). Seasonally averaged distributions of columnar O_8 (in Dobson units) as a function of latitude predicted for the unperturbed (dashed lines) and perturbed (solid lines) stratospheres.



Fig. 5. Daily mean surface UV fluxes at four wavelengths (clear sky) as predicted for (a) January and (b) July with the use of model O_3 data in a multiple scattering radiative flux program carried out at the University of California, Los Angeles (13).

the model shows that about 16 percent of the Northern Hemisphere O_3 will be destroyed. Despite the absence of any direct NO_x injection in the Southern Hemisphere, the calculations indicate that 8 percent of the total O_3 will be destroyed there. In addition, we see that the mid-latitude injection of NO_x apparently has a filtering effect on the northward transport of O_3 through this region so that there is a larger (by about a factor of 2) reduction of O_3 at high northern latitudes than at low latitudes.

Surface UV Flux Estimates

We are concerned not only with the destruction of O_3 in the stratosphere but also with the effect this destruction has on UV radiation reaching the ground. In order to obtain a quantitative estimate of UV fluxes at the ground, we submitted the model total O₃ distributions for January and July to Dr. S. V. Venkateswaran, who used this data in a multiple scattering program (13) to compute daily mean surface UV flux values at four wavelengths under clear sky conditions. The values for both the unperturbed and perturbed stratosphere are shown in Fig. 5. An increase in UV flux at the ground due to O₃ depletion in the perturbed stratosphere occurs at all latitudes and for both months shown, and the shorter wavelengths exhibit a greater sensitivity to O₃ changes than the longer wavelengths. The largest absolute UV changes are associated with the Northern Hemisphere during July, as we would expect from the O₃ differences shown in Fig. 4 and the latitudinal position of the sun. The differences indicate that the July mid-latitude UV radiation levels in the Northern Hemisphere under perturbed conditions are roughly equivalent to present-day unperturbed levels some 15° of latitude to the south. At the same time, Southern Hemisphere mid-latitude UV radiation levels change very little. During January, however, mid-latitude UV fluxes in the Southern Hemisphere are comparable to an equatorward latitudinal shift of about 7°. At tropical latitudes (30°N to 30°S), radiational differences between the perturbed and unperturbed cases are nearly symmetrical about the subsolar point in January but exhibit a definite increase to the north during July.

As an aid to assessing the potential biological effects of increased UV radiation, we have prepared curves of ap-



Fig. 6. Approximate daily mean erythemal doses predicted from the data of Fig. 5 (clear sky conditions).

proximate erythemal dosage (sunburnproducing effect on Caucasian skin) from the flux data of Fig. 5. If $F(\lambda)$ represents the solar UV flux per unit wavelength arriving at the ground, then the erythemal dose D at a given latitude and time of year can be represented by

$$D=\int_{\lambda}E(\lambda)\ F(\lambda)\ d\lambda$$

where $E(\lambda)$ denotes an erythemal efficiency function (14). The results of the numerical integration of this equation for July and January conditions are shown in Fig. 6. Here again, the greatest differences between the calculations for the perturbed and unperturbed states occur during July in the Northern Hemisphere. The erythemal dose at 30°N, for example, for the perturbed state is about 30 percent greater than that for the unperturbed state, whereas the corresponding difference at 60°N amounts to 58 percent. These increases in erythemal dosage are in reasonable agreement with those estimated by use of the "rule of thumb" that the percentage reduction in columnar O_3 should be multiplied by a factor of 2 to obtain the percentage increase in erythemal dosage (8, p. 1598).

Summary and Model Limitations

The results of our model simulations show that, for a continuous NO_x injection rate of 1.8×10^6 metric tons per year from a hypothetical fleet of SST's flying at an altitude of 20 km in the mid-latitudes of the Northern Hemisphere, a significant depletion of about 12 percent in total stratospheric O_3 would be realized. This depletion is particularly evident in the Northern Hemisphere (Fig. 4) in which, on an annual basis, we estimate that 16 percent of the existing O_3 would be destroyed. In addition, interhemispheric transport of the aircraft-injected NO_x would produce at least 8 percent O_3 depletion annually in the Southern Hemisphere even though no aircraft were assumed to fly there. Estimates of the resulting percentage increases in UV flux arriving at the ground, in the form of approximate erythermal doses, amount to about twice the percentage decreases in total O_3 (Fig. 6).

The NO_x injection rate assumed in our model calculations (1.8×10^6) metric tons per year at 20 km) was chosen to most nearly represent the effect of a large fleet (about 500 aircraft) of the now canceled American SST's (Boeing 2707) which might have existed near the year 2000 (12). However, if, in fact, only SST's of the current Anglo-French Concorde and Russion Tupolev 144 prototypes, which are smaller and fly at lower, less harmful altitudes (~ 17 km) than the proposed American SST's, are built, we estimate that it would require a worldwide fleet of a few thousand aircraft to attain an effective injection rate as large as the one used in our model simulation. Clearly, the nature and proliferation of the global SST fleet of the future must be limited if we intend to maintain the environment as we know it today.

Although the data presented here represent only the first results of a continuing investigation, realistic O₃ distributions on a planetary scale have been simulated. Of course, as with any atmospheric model, a number of accompanying uncertainties serve to obscure verification of the results. Of particular importance to the calculations presented here is the limitation associated with prescribing time-dependent NO₂ distributions from the results of two-dimensional calculations and thus accepting the uncertainties inherent in parameterized transport processes. In addition, because the calculation of NO_x distributions in the two-dimensional transport model is accomplished independently from the three-dimensional Q₃ simulation, any chemical feedbacks due to the perturbed O₃ distributions are not contained in the NO_x results. However, for an assumed O₃ perturbation of 10 percent, our twodimensional model calculations indicate that the resulting NO₂ changes amount to only a few percent and suggest that the O3 chemical feedbacks that have been neglected will not significantly alter our results. Moreover, because model

SCIENCE, VOL. 188

limitations are essentially the same for both the unperturbed and perturbed stratospheric calculations, it can be assumed that the differences between the results for the two cases will be more reliable than their absolute values. Future refinements in the model structure are being planned. These include primarily increased model resolution and incorporation of an enlarged chemical package, so that NO_x distributions can be predicted explicitly. This will permit a greater measure of confidence in the climatological results and should lead to a more detailed picture of the distribution of O_3 , both in the natural stratosphere and in a hypothetically perturbed stratosphere.

References and Notes

- P. Crutzen, J. Geophys. Res. 76, 7311 (1971).
 H. Johnston, Science 173, 517 (1971).
 S. Chapman, Mem. R. Meteorol. Soc. 3, 103
- (1930). 4. For example, see H. Dütsch, Advan. Geophys. 15, 219 (1971).
- 15, 219 (1971). 5. For example, see M. McElroy, S. Wofsy, J.
 - Penner, J. McConnell [J. Atmos. Sci. 31, 287 (1974)] or P. Crutzen [Can. J. Chem. 52,

- (19/4)] or P. Crutzen [Can. J. Chem. 52, 1569 (1974)].
 D. Cunnold, F. Alyea, N. Phillips, R. Prinn, J. Atmos. Sci. 32, 170 (1975).
 E. Lorenz, Tellus 12, 364 (1960).
 E. Hessvedt, Can. J. Chem. 52, 1592 (1974).
 One Dotson unit (= 10⁻³ cm) is the thickness of the pure O. layer that would be obtained of the pure O_3 layer that would be obtained if all the O_3 in the vertical column were concentrated at normal temperature and pressure
- See R. Prinn, F. Alyea, D. Cunnold, A. Katz, in The Second International Conference on the Environmental Impact of Aerospace Operations in the High Atmosphere (American Meteorological Society-American Institute of Aeronautics and Astronautics, San Diego, Calif., 8 to 10 July 1974) (American Meteor-

ological Society, Boston, 1974), pp. 180-186. A. Broderick, J. English, A. Forney, in American Institute of Aeronautics and Astro-nautics-American Meteorological Society In-11. A. ternational Conference on the Environmental Impact of Aerospace Operations in the High Atmosphere (Denver, 11 to 13 June 1973) [Am. Inst. Aeronaut. Astronaut. Pap. No. 73-508 (1973)].

- 12. Data Data quoted by A. Grobecker [Acta Astronaut. 1, 179 (1974)] from calculations by J. M. English and A. J. Broderick (Cliquoted by 3. M. English and A. J. Broderick (Ch-matic Impact Assessment Program Mono-graph II, U.S. Department of Transportation, Washington, D.C., in press). N. Sundararaman, D. St. John, S. Venkate-waran in preparties
- 13. swaran, in preparation.
- 14. The erythemal efficiency function used here was taken from P. Cutchis [Science 184, 13] (1974), figure 8]. This research was supported as part of the
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Technology Observed: Attitudes of a Warv Public

Supportive of science yet guarded about technology, the public is uneasy about future technological developments.

Todd R. La Porte and Daniel Metlay

The relatively recent prominence given to issues concerning the environment, notably the debate on supersonic transport, and to the so-called energy crisis reflects a growing uneasiness about technological matters among a generally acquiescent public. There no longer appears to be a broad consensus on the automatic benefits of technological development; its consequences are increasingly perceived as problematical. This new situation could affect both scientists and engineers in terms of the legitimacy accorded their work, the limits within which they may do it, and the level of resources made available for it. For even though a direct relationship between public attitudes and the way decision-makers behave is difficult to establish, the public's mood does create boundaries within which officials generally act.

This article presents findings concerning the public's attitudes toward tech-11 APRIL 1975

nology and science which suggest that considerable refinement of our past generalizations is necessary. Evidence suggests that (i) the public makes a distinction in their evaluations of the outcomes of scientific work and technological work; (ii) the public's reaction to the impact of technology upon society is one of wariness and some skepticism; (iii) the public applies a rather wide range of sometimes contradictory values to its evaluation of technology; (iv) the public has a distrust of the institutions associated with decisionmaking in technical policy areas; and (v) a clear element of political ideology is present in the evaluations of technology made by an important segment of the public.

Only recently has there been sufficient evidence concerning potential public uneasiness about science and technology to stimulate systematic attempts to gauge prevailing opinion on these matters. Most commentaries on these attitudes have been largely impressionistic. They note that the "golden age" of science and technology has passed. They agree that the widespread conviction about the inevitable benefits to come from scientific advance (a conviction pointed to as early as 1830 by de Tocqueville as imprinted on the American genius) has been severely eroded. Edward Shils sums up the case (1):

Whereas it was once believed that every new technological possibility was automatically and inevitably beneficial, the great achievements in outer space [among others] have helped to dim the light once cast by technological progress. . . . Science, engineering and technology have all become amalgamated into a single entity which is conceived as a source of damage and costly waste. The research workers, engineers, military men, industrialists, and politicians are seen as homogeneous groups with each section pursuing its own advantage at the expense of the rest of society.

This slackening in public approval has been attributed to a number of factors. Robert Morrison, for example, cites the distrust of the way power holders manipulate the world; the concern over maldistribution of resources; anxiety about the ethical implications of further technological advances in some areas of medicine and the biological sciences; and growing awareness that much scientific research lacks social relevance (2). The picture of the public mind presented in such commentaries

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