Modeling man's influence on stratospheric ozone

Complex mathematical constructs are being developed to predict the ozone-depleting effect of a large fleet of high-altitude flying supersonic aircraft

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Man's activities might lead to a significant diminution of stratospheric ozone. For some years the principal concern was with the effects of emissions from aircraft flying in the upper atmosphere. When plans to build the U.S. Boeing SST, which was designed to cruise at 20 km, were dropped—at least partially for environmental reasons—concern shifted to the potential environmental hazards of the more imminent Anglo-French Concordes (17 km cruise altitude) and the U.S.S.R.'s Tupolev 144, and since then to any potential follow-on fleets of stratospheric aircraft.

More recently, worry over the effects of aviation on stratospheric ozone has abated considerably, and greater and more continuing concern has focused on the effects of the release into the atmosphere of CFMs (chlorofluoromethanes, also called halocarbons and fluorocarbons, and typified by the Freons) as the result of their use as refrigerants and as propellants in aerosol spray cans.

The postulated destructive effects of either aircraft or CFM emissions on stratospheric ozone are based essentially on the work of modelers.

The modelers use mathematical models of chemical kinetics and atmospheric transport to show that either the nitrogen oxides in the aircraft exhaust at stratospheric altitudes, or the chlorine atoms freed from the CFMs that survive ground-level release will interact with and destroy significant quantities of ozone molecules in the stratosphere.

No one has yet shown by direct measurement that stratospheric aircraft flight leads to ozone destruction. According to the modelers, the fleets of stratospheric aircraft would have to be considerably larger than the few Concordes flying today for a statistically significant ozone change to be produced and detected. The expected ozone changes would be small compared to the natural variability of stratospheric ozone.

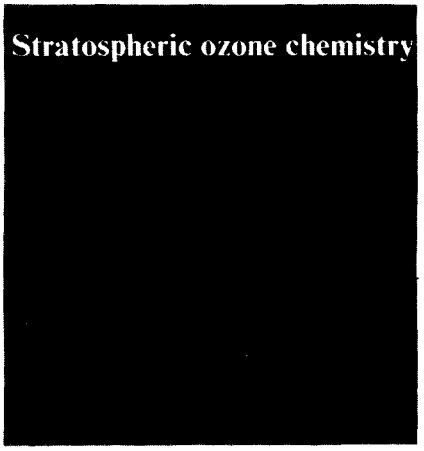
In effect, the modelers have been prophesying possible calamities and urging remedial measures while, at the same time, saying there is no short-term hope of verifying their predictions short of allowing the calamities to occur.

This being the case, it is a matter of great interest to know the "track record" the modelers have established in predicting ozone destruction because of man's activities. I propose to trace through a sequence of predictions of the effects on the ozone region of exhaust emissions from stratospheric aircraft, ignoring the CFM problem with which I am less familiar.

The stratospheric ozone layer plays two physically related roles of direct concern to man: its climatic role and its ultraviolet shielding role.

Climatic role, uv shielding

The climatic pattern that results from ozone absorption of solar ultraviolet (uv) radiation at high altitudes is one of relative atmospheric instability in the lower, tropospheric regime where temperatures tend to decrease with height capped by relative stability in the upper, stratospheric regime where temperatures increase with height.



Of more immediate concern to mankind than possible climatic change, is the possible reduction in uv shielding by the ozone layer. It is widely believed that skin cancer in man—especially fair-skinned man—is caused by exposure to excessive uv radiation in the same wavelengths that produce sunburn (erythema).

According to the National Academy of Sciences (NAS) report (see additional reading), ". . . a reasonable estimate of the percentage increase in skin cancer . . . may be made by assuming that a 1% decrease of stratospheric ozone will cause roughly a 2% increase in skin cancer."

In recent years it has been pointed out, particularly by Harold Johnston, that earlier accepted reaction schemes are not adequate to balance out the ozone formation rate. Johnston and Paul Crutzen have both shown that catalytic ozone-destroying reactions involving the oxides of nitrogen (NO_x) could provide an adequate balance between worldwide ozone formation and destruction.

Once it is accepted that NO_x may be the key to determining the natural balance of stratospheric ozone, it is then natural to raise questions about the effect of NO_x from aircraft exhausts. This is what Johnston and Crutzen and, later, others did. They realized that aircraft exhausts, like automobile exhausts, contain NO_x and that aircraft flying in the stratosphere would inject the NO_x at altitudes near the natural ozone peak.

Johnston calculated the annual total amount of NO_x from a proposed fleet of 500 Boeing SSTs and found it to be about the same amount as might be expected to be present in the natural stratosphere. And then he sounded the alarm! If man's activities might double the natural amount of NO_x in the stratosphere, and NO_x is the key constituent in balancing global ozone production and destruction, what will be the new equilibrium level of ozone?

When Johnston did his initial cal-

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culations he did find substantial ozone reductions. Crutzen found correspondingly large reductions for sufficiently large fleets of Concordes. Concerns like these were the stimulus that led to the initiation of DOT's Climatic Impact Assessment Program (CIAP).

U.S. findings

The DOT/CIAP *Report of Findings* was issued in December 1974, and was followed by the NAS' *Environmental Impact of Stratospheric Flight* in 1975. The two reports agreed qualitatively that stratospheric fleets of aircraft would cause ozone destruction, but quantitatively there was considerable difference.

Since only rarely do any two studies use the same input parameters for a problem, all results presented here have been linearly scaled to a standard level of 1.8×19^9 kg/y NO_2 injected into the stratosphere at 20 km, nominally representing the average global input of a projected fleet of 500 U.S. Boeing SSTs. On this basis, CIAP found approximately 12% ozone reduction globally and the NAS, 16%.

Since publication of the DOT/CIAP and NAS reports, work has continued and the estimated effects owing to aircraft have continued to change.

Evolution of calculations

The majority of the modeling studies that played a role in the DOT and NAS assessments of the aircraft threat to ozone were presented in DOT's CIAP Monograph 3, *The Stratosphere Perturbed by Propulsion Effluents*. I will draw upon the work cited there plus two reports (SCEP and SMIC) that antedated the CIAP study.

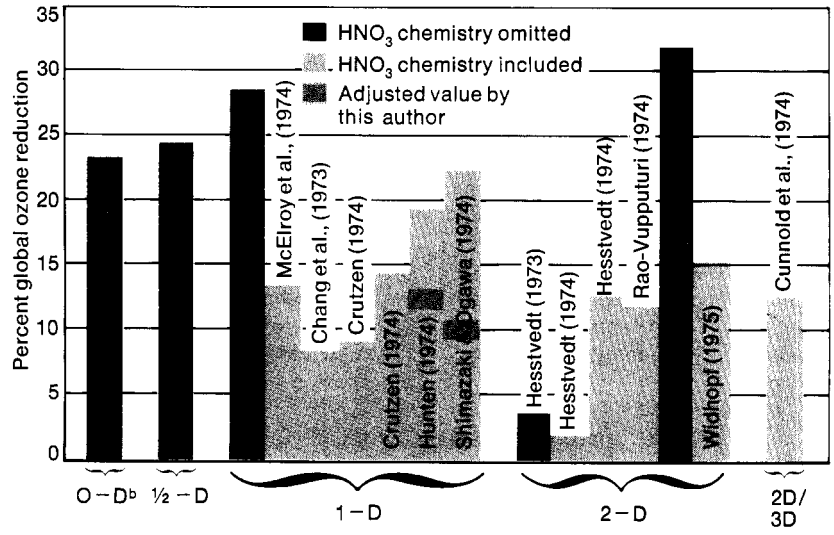
The 1970 *Study of Critical Environmental Problems* (*SCEP*) presented an assessment of the then-recognized global environmental problems attributable to man. It addressed, specifically, the impact of stratospheric flight by supersonic transports and concluded that NO_x emissions were of no importance.

Still, hedging slightly, the authors did recommend a monitoring program for the lower stratosphere to include NO_x as well as water vapor, cloudiness, oxides of sulfur, hydrocarbons and particles.

According to SCEP (using prescribed emission parameters), the 500 SSTs would emit 2.63×10^9 kg/y of NO leading to a world average con-

FIGURE 1

Ozone-reduction predictions[a]



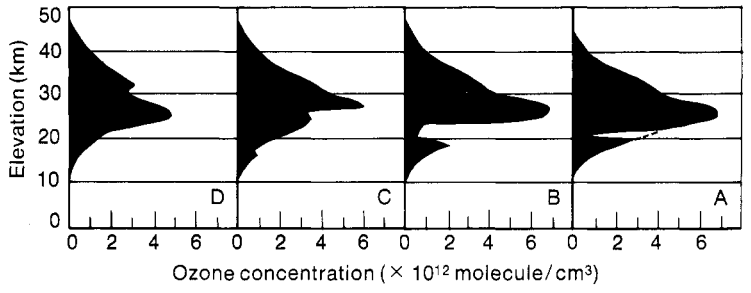
[a] 1.8×10^9 kg/y NO_2 injected at 20 Km
[b] indicates type of transport model used

Author (Publication date)	Ozone reduction (%)	Nitric acid included yes/no	Model dimensions	Time dependent/steady state	Notes
Johnston (1971)	23	No	0-D	Steady state	No ozone diffusion, assumed uniform mixing of NO_x in specified band
Crutzen (1972)	23.6	No	½-D	Steady state	Ozone diffusion, NO quasidiffusion
Chang et al. (1973)	28	No	1-D	Steady state	Full diffusion, error in photolytic rate calculation procedure accounts for high value
McElroy et al. (1974)	13 (Without temperature feedback)	Yes	1-D	Steady state	11% with temperature feedback
Chang et al. (1973)	8	Yes	1-D	Steady state	
Crutzen (1974)	8.5	Yes	1-D	Steady state	
Crutzen (1974)	14	Yes	1-D	Steady state	$10 \times K_z$
Hunten (1974)	19	Yes	1-D	Steady state	2-km adjustment
Shimazaki & Ogawa (1974)	22	Yes	1-D	Time dependent	Injection between 30–60 °N and 6000 km path N.Y. to Paris
Hesstvedt (1973)	3	No	2-D	Steady state	8 × Gudiksen et al. diffusion
Hesstvedt (1974)	2	Yes	2-D	Steady state	8 × Gudiksen et al. diffusion injection at 50°
Hesstvedt (1974)	12.3	Yes	2-D	Steady state	Luther diffusion, injection at 50°
Rao-Vupputuri (1974)	9.4	Yes	2-D	Steady state	Reed & German diffusion, temperature-dynamics—chemistry feedback
Widhopf & Taylor (1974)	32	No	2-D	Steady state	Luther diffusion, 50° injection
Widhopf (1975)	16	Yes	2-D	Steady state	Luther diffusion, 50° injection
Cunnold et al. (1974)	12	Yes (?)[c]	2D/3D	Time dependent	Gudiksen diffusion, 40–50° injection, HNO_3 in 2-D model, which set inputs to 3-D model.

[c] Indicates ambiguity in running "complete" chemistry in the auxiliary 2-D models, but incomplete chemistry in the 3-D model.

FIGURE 2

Johnston's calculations of the effect of injecting NO_x at 20 km



centration of 6.8 ppb, with a possible peak concentration ten times greater. Apparently unaware of the possible NO_x catalytic cycle, the SCEP authors did not regard this NO buildup as significant.

The 1971 *Study of Man's Impact on Climate* (*SMIC*) was instituted "to review SCEP findings critically to point to global environmental problems that were slighted or overlooked, . . ." The SMIC authors returned to the question of the effects on ozone of the oxides of nitrogen emitted in the stratosphere.

They noted that NO_x participates in the chemical process for stratospheric ozone and they described the now well-known catalytic cycle involving NO and NO_2 with the destruction of ozone, but did not draw any conclusions as to the amount of ozone destruction that would be caused by the injection of NO_x in the stratosphere by the high-flying aircraft.

Figure 1 is the road map guiding the reader through the evolution of calculated ozone-reduction results. On the chart, the abscissa indicates the kind of model used: 0-D (D for dimension) means no explicit transport; ½-D means *ozone* transport modeled in the vertical, but not other species; 1-D means vertical transport of all species; 2-D means 1-D plus north-south (meridional) transport; and 3-D means 2-D plus east-west (zonal) transport. Partial hatching on two of the bars indicates that I have adjusted the author's result to show comparability with other results except for a unique assumption made by the author. Full hatching shows the instances in which nitric acid chemistry was omitted from the models and indicates that possible ozone reduction tends to double because of the omission.

Simpler models

Harold Johnston at the University of California, Berkeley, in 1971 tested the effect of introducing the SCEP amount of NO_x, reduced by the factor 0.35 (based on the engine manufacturer's recommendation).

He computed two general cases: case 1 where the NO_x is distributed uniformly worldwide in the stratosphere in the horizontal sense but stratified vertically in a prescribed fashion; and case 2 where the NO_x achieves a ten-times higher concentration corresponding to a latitudinal corridor enhancement effect, and is vertically stratified as in case 1. He assumed a two-year residence time for the injected material as had been done by SCEP.

Johnston's results for the uniform horizontal distribution throughout the world with four different patterns of vertical distribution are shown in Figure 2. In case A he assumed that all the NO_x stayed in a 1-km band between 20–21 km, and found 3% ozone reduction. The greatest reduction occurred in case C where the injected NO_x was assumed to spread over the band 17–25 km. It led to the often-quoted Johnston result of 23% ozone reduction.

In his worst-case calculations where he followed the SCEP lead and assumed a ten-time enhancement in the "corridor" where the SSTs would fly, Johnston computed 50% ozone reduction.

Paul Crutzen, now at NSF's National Center for Atmospheric Research (NCAR), introduced explicit time-dependent vertical diffusion calculations into the equations used to compute the ozone concentration.

Crutzen's results were reported in February 1972. Like Johnston, he made various assumptions about the vertical distribution of the injected NO_x, but treated the vertical diffusion of ozone explicitly. (Because ozone formation and diffusion were modeled but the NO_x distribution was imposed, I denote Crutzen's modeling effort ½-D).

Crutzen noted that nitric acid formation could be an important process for removing NO_x from the stratosphere but, like Johnson, did not take it into account in making his calculations.

Both Johnston and Crutzen had to make various assumptions about the background amount of NO_x present in the "natural" stratosphere. For Crutzen's case closest to Johnston's, Crutzen found 23.6% ozone reduction, remarkably close to Johnston's 23%!

All the models described hereafter do explicitly include transport, in one, two, or three dimensions, of the reacting species.

One-dimensional models

Julius Chang and his colleagues at the Lawrence Livermore Laboratory published, in 1973, one of the earliest sets of results for ozone reduction, using full vertical diffusion of all species. Their adopted eddy diffusion profile had a minimum near 30 km.

The chemical reaction set was narrowly limited to the Chapman cycle for ozone formation and destruction plus the NO_x catalytic cycle; nitric acid reactions were not considered. As can be seen in Figure 1, their result was among the highest shown in this review—28%.

In a later paper, also published in 1973, this time by Chang, Hindmarsh, and Madsen of Lawrence Livermore Laboratory (see the bar labeled Chang et al., 1973 in Figure 1) the authors calculated an approximate 8% ozone decrease with what appeared to be the same model. This large shift in effects was explained in a 1974 review by Chang and Johnston. They pointed out that not only was nitric acid's role ignored in the early paper, but also the photolysis coefficients used in the early paper were not properly recomputed as the calculated ozone was reduced.

Michael McElroy and colleagues at Harvard University used a simple one-dimensional eddy-diffusion model that included nitric acid chemistry. Their eddy diffusion profile differed from that of the Livermore group.

Since the stratospheric modelers have so often referred to different rationales leading to different choices of parameters, it is instructive to look at a sampling of the many profiles of eddy diffusion coefficients they have used.

For the works reviewed here (Figure 3), McElroy's minimum value of the eddy diffusion coefficient is apparently the lowest in value and occurs at the lowest altitude (about 16–18 km). Chang's minimum value is about twice McElroy's and occurs at near twice the altitude. Hunten's profile at the mini-

mum strongly resembles McElroy's.

The vertical diffusion profiles are indicators of how well ozone will diffuse downward from its formation region at higher altitudes; how well the injected NO_x will diffuse upward to mix with and destroy ozone; and how well the nitric acid will diffuse downward out of the stratosphere to serve as a sink for NO_x.

McElroy and coworkers obtained results for both a fixed-temperature case and one in which atmospheric temperatures were allowed to change as a result of computed changes in the amount of ozone and the consequent changes in energy absorption. The changed temperatures altered chemical reaction rates. By interpolation of the published results I found 13% ozone reduction as the "fixed-temperature" result to compare with the other studies in Figure 1 and 11% as the "radiative equilibrium temperature" result.

Crutzen, in 1974, published two new sets of results using 1-D models. By using some 60 reactions, now permitting vertical diffusion of all constituents, he found a decrease of 8.5% in ozone.

Later, in 1974, in *Ambio,* Crutzen revised his results, this time reporting approximately 14% ozone reduction. This newer answer comes from using larger vertical diffusion coefficients as shown in Figure 3.

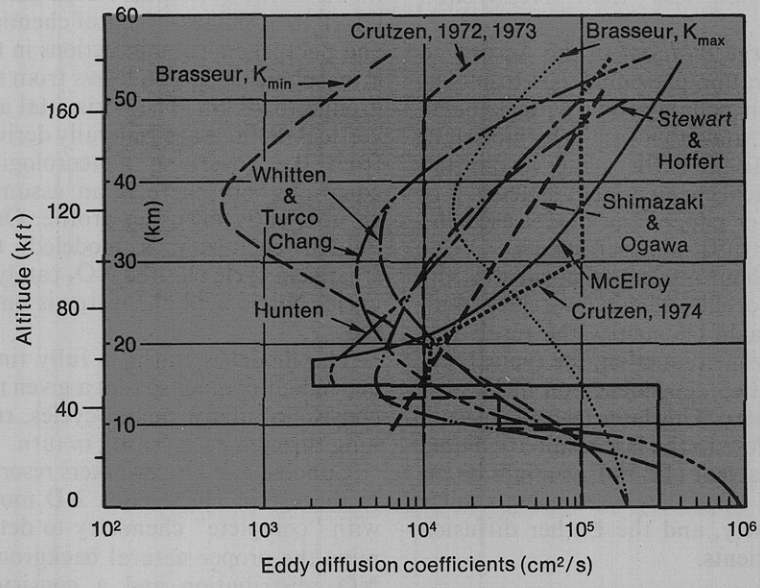
In the key region between 20–30 km, the new Crutzen values of eddy diffusion coefficient are very close to the McElroy values and perhaps this accounts for their similar answers (14% and 13%, respectively).

Donald Hunten of Kitt Peak National Observatory used a composite model in which he computed the vertical mixing of NO_x, using his own choice of eddy diffusion profile, and then used the McElroy model to determine the relationship between ozone reduction and NO_x mixing ratio. Where McElroy had computed 13% ozone reduction, Hunten found 19%. The reason for the more drastic Hunten result was his assumption that aircraft altitudes ought to be adjusted to an effective altitude based on relative height above the tropopause. For instance, to determine the impact of SST flights at 20 km, Hunten used an effective altitude of 22 km.

Tatsuo Shimazaki and Toshihiro Ogawa, in 1974, reported on their 1-D results at the IAGA-IAMAP meeting in Melbourne, Australia. Unlike the steady-state models discussed above, this model is an example of the time-dependent solution to the ozone reduction caused by injection of NO_x.

FIGURE 3

Eddy diffusion coefficient profiles used to predict ozone depletion



The model has a very complete set of chemical reactions.

The results appear to be equivalent to a 22% global reduction, unexpectedly high when compared to other 1-D results with comparable chemistry and transport. Figure 1 shows that this is the greatest ozone reduction obtained with any of the 1-D models that included nitric acid chemistry.

Was there something unique in the Shimazaki and Ogawa treatment? In fact there was. Unlike the other modelers, Shimazaki and Ogawa assumed that the injected NO_x would be confined to the typical SST flight corridor between New York and Paris (6000 km long), 30°–60° North latitude and 3 km thick.

In this way, they achieved an initial injection concentration of NO_x two-and-one-half times greater than that of Julius Chang, for instance, and this would go far to account for their more alarming prediction of ozone reduction.

Two-dimensional models

Eigil Hesstvedt of the University of Oslo was a pioneer among those who participated in the development of 2-D models portraying the interactions of stratospheric chemistry and transport. In order to introduce the meridional motions, he used two-dimensional eddy diffusion coefficients developed by Gudiksen and colleagues. But he found that the Gudiksen coefficients did not lead to a satisfactory representation of the natural ozone. Drastic changes were required. He ended by multiplying the Gudiksen coefficients by 8, and doubling his original values of vertical wind speed.

Making calculations with the adjusted transport coefficients and omitting nitric acid chemistry, Hesstvedt, in a 1973 paper, found ozone reductions equivalent to 3%.

In a follow-up paper in 1974, Hesstvedt used much the same model as above but now added reactions related to the formation and destruction of nitric acid and permitted transport of the NO_x-related spcies. This time Hesstvedt reported a global average ozone reduction of about 2%.

Also in 1974 at the IAGA/IAMAP conference, Hesstvedt reported yet another set of calculations of ozone reduction, using diffusion coefficients developed by Fred Luther of Lawrence Livermore Laboratory. With the reduced rate of transport provided by the Luther coefficients, Hesstvedt now found 12.3% ozone reduction, which brought his results in line with the others.

Krishna Rao-Vupputuri of the Atmospheric Environment Service, Canada, reported calculations with a 2-D model in which the mean meridional circulation is generated from the meteorological equations of motion and there is thermal feedback involving both the transport and the chemical reaction rates. The model solves the equations for zonal momentum and heat balance as well as the continuity equations for the atmospheric trace species.

Rao-Vupputuri used 2-D eddy diffusion coefficients developed by Reed and German that are typically

large—often by as much as a factor of 2—than the Luther coefficients used by Hesstvedt. Rao-Vupputuri's calculated ozone reduction (9.4%) was somewhat less than Hesstvedt's 12.3%.

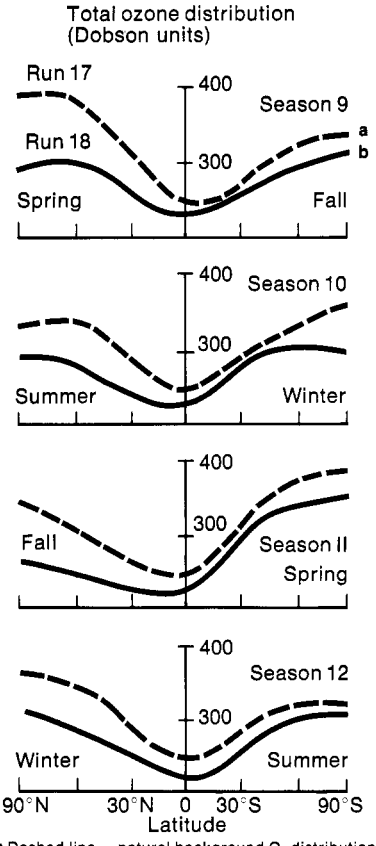
George Widhopf of the Aerospace Corporation presented 2-D results in 1974 in collaboration with Thomas Taylor, and other results by himself in 1975. In the earlier paper he omitted nitric acid chemistry but included it in the later paper. The model used the Luther diffusion coefficients.

Without nitric acid chemistry, the global ozone reduction was 32%. With nitric acid chemistry, the result was 16% ozone reduction, the typical factor-of-two dependence on nitric acid chemistry. This later result is reasonably close to the last result attributed to Hesstvedt (12.3%), as might be expected since they were using similar chemistry, and the Luther diffusion coefficients.

Three-dimensional model

Derek Cunnold and his coworkers at MIT, *Fred Alyea, Norman Phillips,* and *Ronald Prinn,* have published several papers concerning stratospheric modeling with the MIT three-dimensional general circulation model (GCM) and ancillary two-dimensional models.

FIGURE 4
Results from the MIT 3-D model of the atmosphere



[a] Dashed line = natural background O_3 distribution
[b] Solid line = perturbed distribution

The MIT GCM was the only 3-D model of the atmosphere used during CIAP to calculate effects of chemical and photochemical interactions in the atmosphere. It has 26 levels from the ground to 70 km. The horizontal and vertical motions are generally derived from the governing meteorological equations, but there is an assumed vertical eddy diffusion profile. Only limited chemistry is modeled: the Chapman cycle plus the NO_x catalytic cycle. Nitric acid chemistry is omitted.

The model is run in a fully time-dependent manner so that a given run consists of many annual cycles, running through each season in turn.

Cunnold and his coworkers resorted to the use of Hesstvedt's 2-D model with "complete" chemistry to determine the proper natural background NO_x distribution and a consistent distribution for the natural ozone in the 3-D model. They then used a different 2-D model of their own with "complete" chemistry to determine the NO_x distribution caused by the addition of the standard NO_x injection from the 500-SST fleet. This new disturbed NO_x distribution was then introduced into the 3-D model to determine the change in ozone distribution.

Figure 4 shows the seasonal and latitudinal distribution (averaging over longitudes) of the total ozone in the course of a simulated year as redetermined by the MIT 3-D model. The net global ozone reduction was found to be 12%. This result is labeled 2D/3D in Figure 1 to acknowledge the device of running part of the problem with 2-D models and part with the 3-D model.

Assessment

Figure 1 summarizes the results presented in this article. The column labeled "time dependent/steady state" indicates a distinction among modeling techniques not emphasized in the preceding text. Briefly, steady-state modelers use some numerical "relaxation" technique to develop the equilibrium state of the atmosphere corresponding to a particular or average state of the atmosphere, whereas the time-dependent modelers seek a set of answers that evolve in time as the days, months, or seasons unfold.

Various reports in the literature have been reviewed. The object was to learn whether there was a reasoned path from the earliest conclusions of no effect, through a possible 50% reduction of ozone, to the consensus generally reached in the U.S. by the end of 1974 of 8–15% global ozone reduction that might be caused by the uniform global introduction of 1.8×10^9 kg/y NO_2 at 20 km from high-flying aircraft.

My conclusion is that out of the tangled jungle of special cases that were studied in this review, and the different assumptions that were made by the various authors, a reasoned path can be cleared. Discrepancies can generally be, and often have been, reconciled. The body of evidence does generally point in a consistent direction.

One-, two-, and three-dimensional modelers using different numerical methods, using time-dependent or steady-state calculations, using various assumptions about eddy diffusion coefficients, various boundary conditions, various choices of prescribed or predicted motions, various choices of inclusion or exclusion of feedback among temperature, dynamics, and chemistry, and various patterns of NO_x injection did manage to obtain results that, by and large, can be sensibly related to each other.

Additional reading

CIAP Monograph 3. The stratosphere perturbed by propulsion effluents, Department of Transportation, DOT-TST-75-53, Washington, D.C. (1975).

Grobecker, A. J., Coroniti, S. C., Cannon, R. H., Jr., Report of findings, the effects of stratospheric pollution by aircraft, Department of Transportation DOT-TST-75-50 (1974).

National Academy of Sciences. Environmental impact of stratospheric flight: biological and climatic effects of aircraft emissions in the stratosphere (1975).

SCEP. Report of the study of critical environmental problems, In *Man's Impact on the Global Environment,* MIT Press, Cambridge, Mass., 1970.

SMIC. Report of the study of man's impact on climate, in *Inadvertent Climate Modification,* MIT Press, Cambridge, Mass., 1971.

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