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The Space Shuttle's Impact on the Stratosphere

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Launch of spacecraft using solid rocket motors leads to release of gaseous and particulate matter in the stratosphere. Concern over these emissions, particularly chlorine, goes back to the Climatic Impact Assessment Program (Hoshizaki, 1975). The buildup of these exhaust products and their perturbation to stratospheric ozone is followed with two- and three-dimensional atmospheric chemical transport models. Chlorine enhancements due to the current rate of shuttle launches is small, on average less than 0.6% above the current background. Other gases emitted from the solid rockets appear to have even smaller global effects, although the impact of particulate alumina remains uncertain.

1. INTRODUCTION

The launch of NASA's Space Shuttle and similar rockets injects chlorine compounds directly into the stratosphere, adding to the current burden of stratospheric chlorine. Depletion of the stratospheric ozone layer has been linked to increases in stratospheric chlorine compounds associated predominantly with chlorofluorocarbons (see recent assessments: *World Meteorological Organization (WMO)* [1986; 1990]; *Watson et al.*, [1988]). The purpose of this study is to determine the magnitude of the chlorine increases that might be caused by the Space Shuttle and to assess the overall impact on the chemistry and composition of the global stratosphere.

The solid rocket motors on the Space Shuttle and Titan IV launch vehicles use a solid fuel composed of ammonium perchlorate, aluminum and a polymer binder (P. D. Evanoff, Thiokol Corp., private communication, 1989). The exhaust consists primarily of gaseous HCl, carbon monoxide, water vapor, molecular nitrogen and aluminum oxide. The potential damage to the ozone layer by the Space Shuttle's solid rocket motors was recognized during the Climatic Impact Assessment Program (CIAP focussed on supersonic aircraft but also studied rocket plumes, see *Hoshizaki* [1975]). The last assessment of the Shuttle in terms of stratospheric ozone was more than a decade ago [*Potter*, 1978], and our understanding of stratospheric chemistry and modelling has evolved much since then.

The launch scenario considered here consists of nine Shuttles and six Titans per year; it is typical of the current schedule, but it may fall short of the frequency needed for major space projects. The chlorine from these launches is used as a stratospheric source of Cl_y (total inorganic chlorine: the sum of HCl, Cl, Cl₂, ClO, ClONO₂, and HOCl) in three

numerical models of the stratosphere. Calculations have been made with two two-dimensional (latitude by altitude) models with complete chemistry (AER model: *Ko et al.* [1985; 1989]; GSFC model: *Dougllass et al.* [1989; *Jackman et al.*, 1989a] and a three-dimensional model for chemical tracers (GISS model: *Prather et al.* [1990]).

In one numerical simulation, Cl_y enhancements in the middle stratosphere two days after a January launch of the Shuttle are still expected to be clumpy, but the exhaust plume is predicted to have spread over a region about 20° latitude by 30° longitude with an average increase of about 30 ppt (parts per 10¹²) or 2% above background. One month later the Shuttle plume is well mixed, and increases in Cl_y are less than 4 ppt throughout the stratosphere. The buildup of chlorine from these launches approaches a steady state limit after several years: on average, Cl_y would increase by about 10 ppt in the middle stratosphere of the northern hemisphere, less than 0.5% above current levels. Corresponding ozone depletions are predicted to be less than 0.2% locally, with smaller perturbations to the ozone column.

The profiles and amounts of chlorine injected from the solid rocket motors are summarized in section 2. The transient response to a single launch is shown in section 3, and model predictions of the steady state accumulation of Cl_y are described in section 4. The overall impact on stratospheric chemistry and ozone is discussed in section 5.

2. CHLORINE FROM ROCKET EXHAUST

Estimates of the amount and distribution of chlorine released from the launch of the NASA Space Shuttle and the Titan IV expendable launch vehicle are available from Thiokol (P. D. Evanoff, Thiokol Corp., private communication, 1989). In Table 1 we report the kilograms of chlorine (as Cl) released in 5 km vertical intervals for Shuttle launches from Cape Canaveral, Florida (nine per year) and for Titan IV launches from both Cape Canaveral (four per year) and Vandenberg Air Force Base, California (two per year). The chlorine is released as HCl, which we treat as Cl_y in the models.

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The total amount of chlorine released into the stratosphere (above 15 km) by the solid rocket motors is 725,000 kg (0.725 kilotons) per year, and can be compared with that associated with industrial halocarbons. The chemical industry's production of halocarbons exceeds 1250 kilotons of chlorine per year [WMO, 1990]. The release of chlorine during photochemical destruction of the chlorofluorocarbons (CFCs) occurs predominantly in the stratosphere, but happens slowly, on time scales of order 100 years, over the lifetime of the gas. The estimated annual source of stratospheric chlorine from the industrial halocarbons is about 300 kilotons of chlorine per year (AER model); the remainder of the annual emissions goes into the accumulating atmospheric burden of chlorinated halocarbons (about 600 kt/yr) or is destroyed in the troposphere. Thus, the launch schedule in Table 1 would add only about 0.25% to the current stratospheric source of Cl_y . These results are qualitatively similar to a conclusion reached 16 years ago [Cicerone and Stedman, 1974].

The release of chlorine from the 15 launches summarized in Table 1 was averaged over the year and put into the models as a continuous source of Cl_y every time step. The vertical distribution specified in Table 1 was used by the models. The latitudinal location of the two launch sites was included in all three models; but the longitudinal location could only be specified in the three-dimensional simulation.

3. TRANSIENT RESPONSE TO A SINGLE LAUNCH

One launch of the Space Shuttle injects a single, very large pulse of 68,000 kg of chlorine into the stratosphere. Although this amount of chlorine is inconsequential on a globally averaged scale, the greatly enhanced levels of Cl_y in the vicinity of the exhaust plume may lead to large ozone depletions over a spatially limited region. We examined the transient response of stratospheric Cl_y to a single Shuttle launch using the three-dimensional GISS model. The chlorine is released over Cape Canaveral (29°N, 80°W) by using one-ninth of the annual source given in Table 1 as an instantaneous source.

Simulations were initiated on January 1 and July 1, and continued for one month. Immediately following the launch, the exhaust plume will not be completely mixed over the

scales resolved by the model grid, and the calculations are intended to represent the average concentration of the resulting nonuniform distribution. Figures 1 and 2 summarize the instantaneous increase in Cl_y concentrations at 3.4 mbar (about 40 km altitude) as a function of latitude and longitude for days 2, 4, 8 and 30 following both the January and July initializations. Other levels in the upper stratosphere show similar effects, but lower altitudes in the stratosphere have smaller absolute enhancements of Cl_y (see Figures 3–5).

After two days (Figures 1a and 2a) the average increase in chlorine in the upper stratosphere, 50 ppt (January) and 70 ppt (July), is localized near the launch site with peak levels not resolved by the model grid (8° latitude by 10° longitude). After 8 days (Figures 1c and 2c) the maximum grid-averaged levels have decreased by factors of about four (January) and two (July), have moved away from the launch site in accord with the prevailing winds, and have spread over horizontal scales that are resolved by the model. For the January launch most of the added chlorine (concentrations greater than 1 ppt) resides in the region bounded by 20°N–50°N and 40°W–100°W. The summer stratosphere is less dispersive, and for the July launch most of the Cl_y is contained between 20°N–36°N and 150°E–140°W. Eight days after the January 1 launch (Figure 1c) the largest grid-averaged perturbation to Cl_y (as resolved by the model grid) is less than 10 ppt, about 0.4% above background levels. Eight days after the July launch (Figure 2c) the added Cl_y remains more concentrated: greater than 30 ppt (about 1%) above background over a smaller area.

One month after the Shuttle launch (Figures 1d and 2d), the added chlorine is predicted to have spread over most of the upper stratosphere in the northern hemisphere, and the plume is expected to have mixed thoroughly. The winter stratosphere is dispersive and Cl_y perturbations are less than 1 ppt everywhere. In summer the exhaust products remain predominantly over mid-latitudes (20°N–50°N) with perturbations still as large as 3 ppt. The globally averaged depletion of ozone associated with a single launch should be less than that caused by the steady state buildup of chlorine, as discussed below. Local destruction of ozone in the immediate vicinity of the rocket plume could be significantly larger and is not explicitly resolved in these global models.

4. STEADY STATE ACCUMULATION

The three models used the continuous source of stratospheric chlorine from the rocket launches as described in section 2. The ultimate removal for the injected stratospheric Cl_y is transport into the lower atmosphere (troposphere) where most inorganic chlorine species are soluble and therefore removed rapidly by rainfall and other processes. In these models this sink was applied by imposing either rapid loss for Cl_y below 10 km (two-dimensional models) or a negligibly small concentration of Cl_y below 1 km (three-dimensional model). Small differences in the application of this lower boundary condition do not affect the calculated stratospheric Cl_y distribution, because almost all of the chlorine transported into the troposphere is removed and cannot be recirculated back into the stratosphere.

These calculations were initiated and then continued for several model years until a steady state distribution was reached. In steady state, stratospheric Cl_y additions have built up until the amount of chlorine injected into the stratosphere equals that transported into the lower troposphere.

TABLE 1. Stratospheric Chlorine Released Annually by Shuttle and Titan IV Launches

Altitude, km	Kilograms of Chlorine (as Cl)		
	A	B	C
15–20	176,800	16,800	8,400
20–25	136,100	14,700	7,300
25–30	109,600	12,600	6,300
30–35	87,400	10,600	5,300
35–40	69,300	8,900	4,500
40–45	25,900	7,300	3,700
45–50	4,500	6,000	3,000

The launch scenario assumes nine Shuttles (A) and four Titans (B) from Cape Canaveral, Florida (29°N 80°W) and two Titans (C) from Vandenberg AFB, California (34°N 121°W).

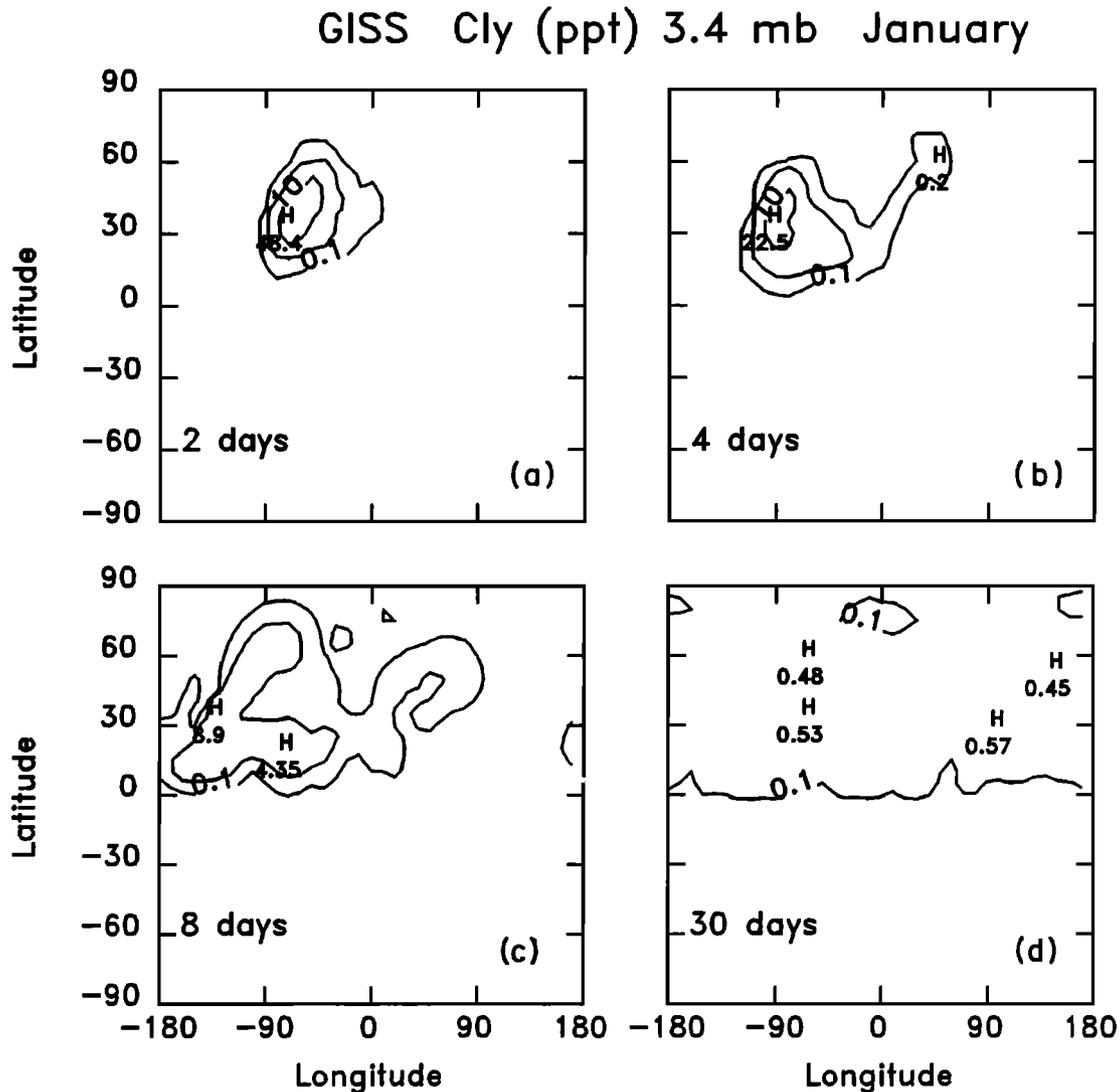


Fig. 1. Latitude by longitude contours of chlorine enhancements near 40 km altitude due to a single Shuttle launch on January 1. Results from the GISS model are shown for (a) 2, (b) 4, (c) 8, and (d) 30 days following the launch.

The computed addition to Cl_y (ppt) associated with the Shuttle / Titan launches is shown as monthly, zonal averages in Figures 3–5 for the GSFC, AER and GISS models, respectively. The magnitudes of the Cl_y increase is largest in northern mid-latitudes (30°N–50°N) near the source, and peaks in the upper stratosphere (30–45 km altitude) where the rocket emissions are largest on a molecule per molecule of air basis. Increased concentrations of Cl_y in the northern upper stratosphere range from 6 to 14 ppt in the GSFC model (Figure 3), 4 to 9 ppt in the AER model (Figure 4), and 6 to 12 ppt in the GISS model (Figure 5). The GSFC and GISS models predict a similar buildup of Cl_y; the AER model appears to have a more rapid stratospheric circulation [Jackman *et al.*, 1989b] that flushes out the chlorine more rapidly. In the lower stratosphere (15–25 km altitude) Cl_y concentrations are less enhanced: about 2 ppt at 15 km increasing to about 6 ppt at 25 km. Several years are required to transport substantial concentrations of the rocket source of Cl_y into the southern hemisphere stratosphere. Enhancements in the southern stratosphere range from less than 1 to as much

as 6 ppt, and are generally a factor of 2 or greater below the corresponding enhancements in the northern stratosphere.

During northern winter, all models show rapid northward mixing between 30°N and the Pole, with isolines that slant poleward and downward. This basic pattern shown in Figures 3a–5a is typical of other long-lived stratospheric tracers, both from model calculations [Jackman *et al.*, 1989b] and observations (see recent assessments: WMO [1986]). The pattern of Cl_y during northern summer, however, dramatically shows the impact of a localized stratospheric source in a season without substantial latitudinal mixing. The increases in Cl_y concentration in July (Figures 3b–5b) peak strongly at 30°N between 30 and 50 km altitude in all three models, with monthly and zonally averaged maxima of more than 12 ppt in the GSFC and GISS models, and 9 ppt in the AER model.

In order to determine the relative perturbation to Cl_y from the rocket launches, the two-dimensional models compared the calculated Cl_y enhancements (Figures 3–4) with the Cl_y calculated from all other sources: the CFCs, CCl₄, CHF₂Cl, CH₃CCl₃, CH₃Cl. Air enters the stratosphere containing a mix

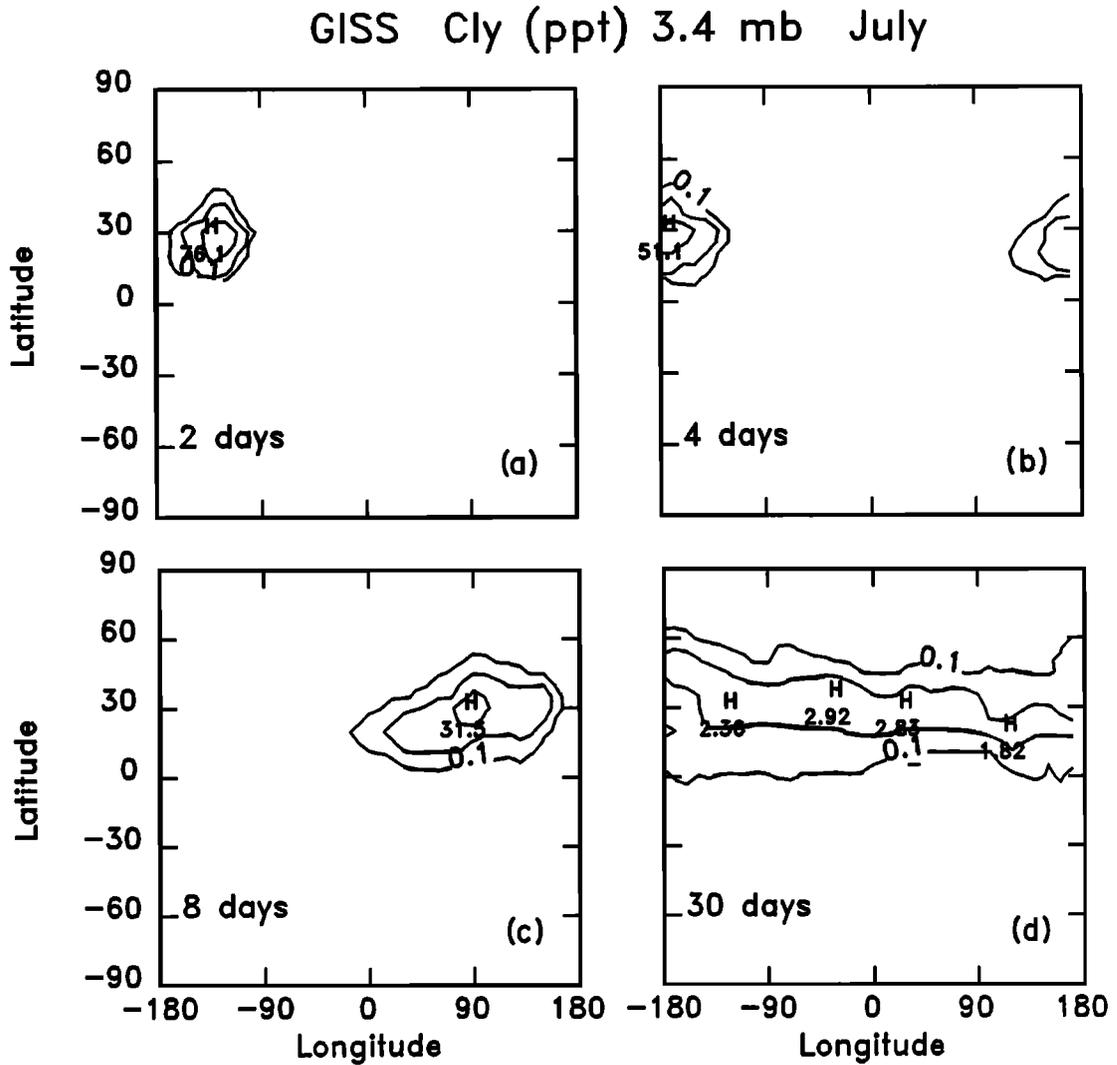


Fig. 2. Latitude by longitude contours of chlorine enhancements due to a single Shuttle launch on July 1. See Figure 1.

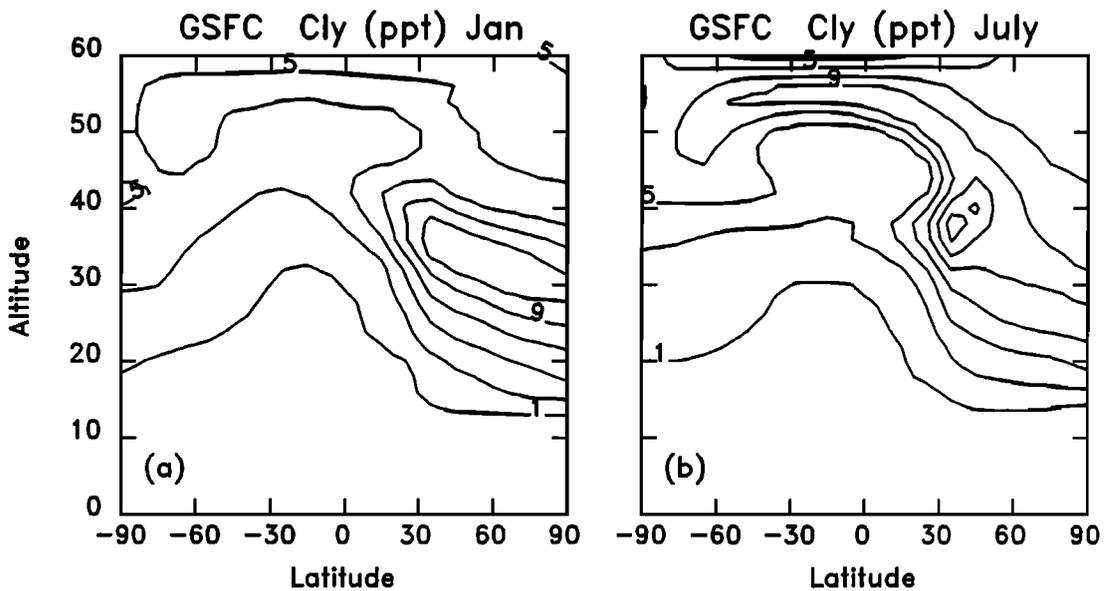


Fig. 3. Latitude by altitude (pressure) contours of enhanced chlorine (in ppt) from the GSFC model. The steady state buildup of chlorine as Cl_2 is caused by the launch of nine Shuttles and six Titan IV vehicles per year. The monthly and zonally averaged concentrations are shown for (a) January and (b) July.

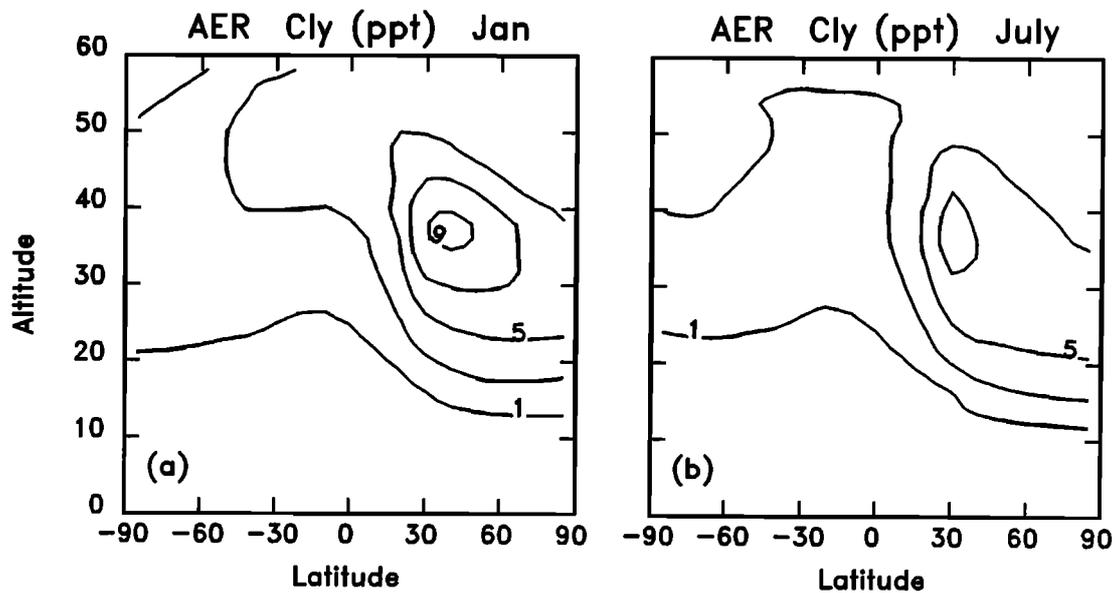


Fig. 4. Latitude by altitude (pressure) contours of enhanced chlorine (in ppt) from the AER model. See Figure 3.

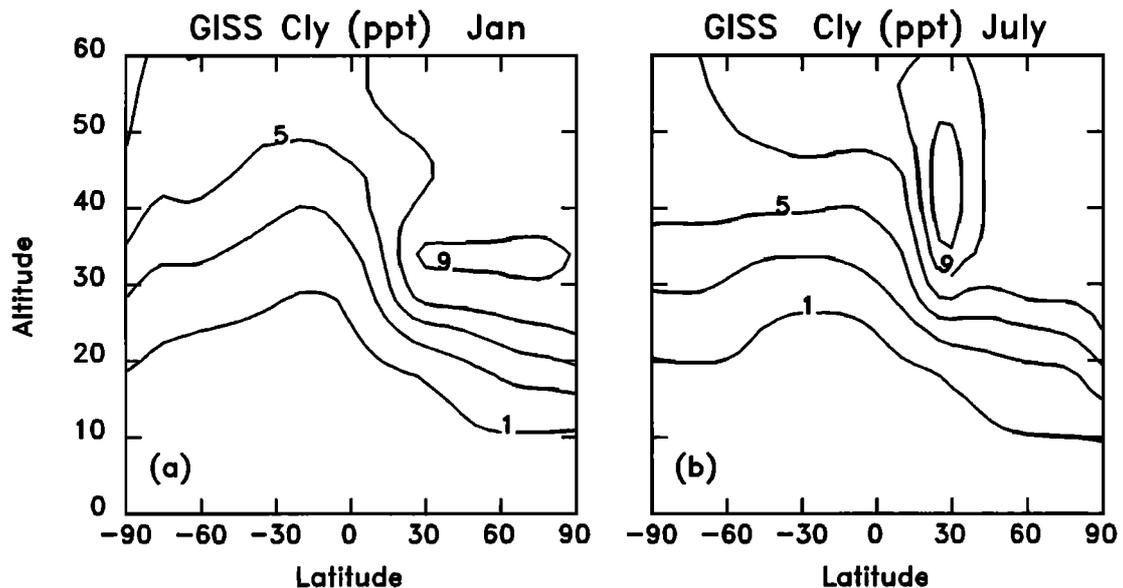


Fig. 5. Latitude by altitude (pressure) contours of enhanced chlorine (in ppt) from the GISS model. See Figure 3.

of these halocarbons representing the bulk tropospheric mixing ratios of these species and with negligible concentrations of the more soluble inorganic chlorine (Cl_y). In the lower stratosphere Cl_y concentrations are small because only a fraction of the CFCs have been photochemically destroyed, thereby releasing chlorine atoms. In the upper stratosphere where the halocarbon and CFC concentrations are greatly reduced, Cl_y concentrations approach their upper limit, currently about 3 ppb (parts per 10^9). The scenario for rocket launches in Table 1 leads to only modest perturbations in stratospheric Cl_y , as shown in Figures 6 (GSFC) and 7 (AER). The GSFC results predict increases ranging from 0.3 to 0.6% over the northern mid-latitudes; while the AER model gives smaller enhancements, 0.2 to 0.3%. In the southern

hemisphere, Cl_y increases are less than 0.2% for the GSFC model and 0.1% for the AER model.

Ozone perturbations at steady state associated with the source of Cl_y from rocket launches are shown in Figure 8 for the GSFC model. The largest depletions in ozone concentration, between 0.10% and 0.15%, occur in the upper stratosphere (30–45 km altitude) in the northern mid-latitudes. Losses elsewhere are much smaller with the exception of the corresponding locations in the southern hemisphere. The depletion of the total column abundance of ozone is likewise small, less than 0.1%. The photochemical model used here includes only gas phase, homogeneous chemical reactions; it does not account for the heterogeneous reactions occurring on polar stratospheric clouds that have been shown to be

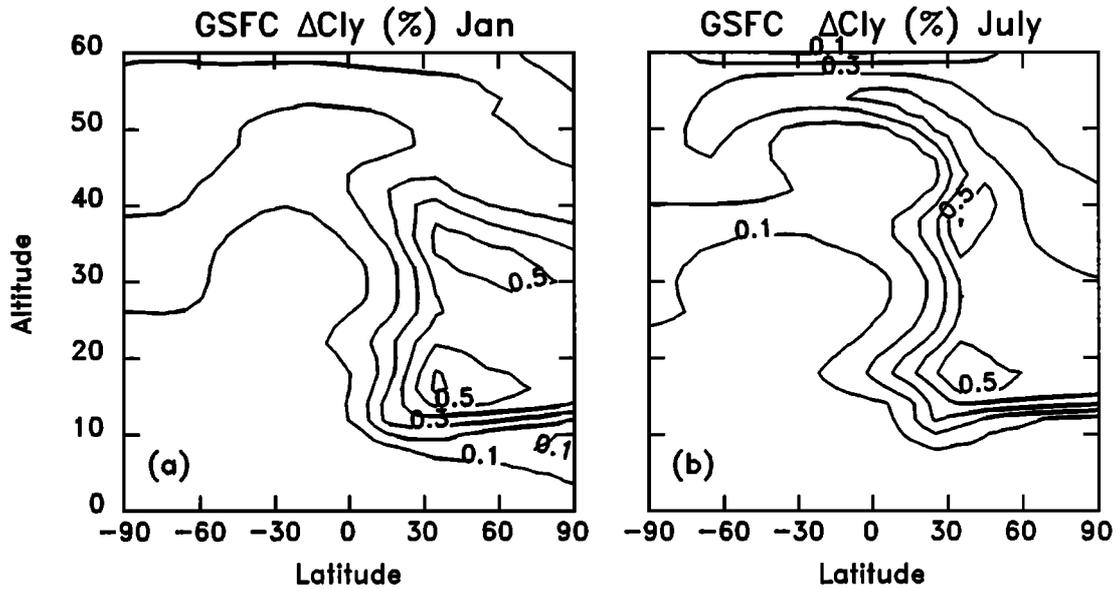


Fig. 6. Latitude by altitude (pressure) contours of the perturbation to background Cl_2 levels (%) from the GSFC model. See Figure 3.

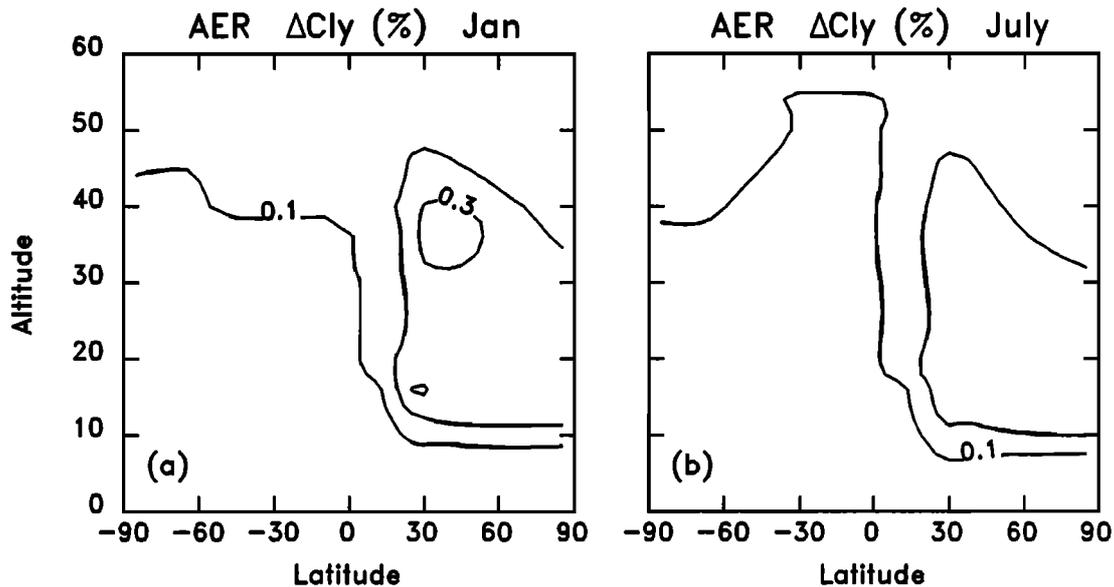


Fig. 7. Latitude by altitude (pressure) contours of the perturbation to background Cl_2 levels (%) from the AER model. See Figure 3.

responsible for the chlorine-catalyzed destruction of ozone in the lower stratosphere during polar winter (the Antarctic ozone hole). Nevertheless, the impact of Shuttle chlorine on further polar ozone loss can probably be constrained since the relative perturbations to Cl_2 in the lower stratosphere are less than 0.1% over Antarctica and 0.5% over the Arctic.

5. THE POTENTIAL FOR OZONE DEPLETION

Each launch of the Space Shuttle injects about 0.068 kt of chlorine into the stratosphere. This amount is small compared to the current background of stratospheric chlorine (3 ppb), which is generated from the photochemical destruction of industrial and natural halocarbons within the stratosphere at a

rate of about 300 kt/yr. Global stratospheric models are used here to assess the impact of the currently modest launch schedule on stratospheric chemistry and ozone depletion.

In the immediate exhaust plume of the Shuttle (about 40 m across in the upper stratosphere), concentrations of HCl are large, about 0.08 by volume. Even if this plume were mixed over a 10×10 km area, concentrations would still exceed 1000 ppb. However, a 100 km^2 area comprises less than 1/1,000,000 of the mid-latitude stratosphere, and the global or even regional effects of complete ozone destruction within this corridor would be inconsequential. Furthermore, the chlorine is released predominantly as HCl and would need some time to be chemically processed into more catalytically active forms

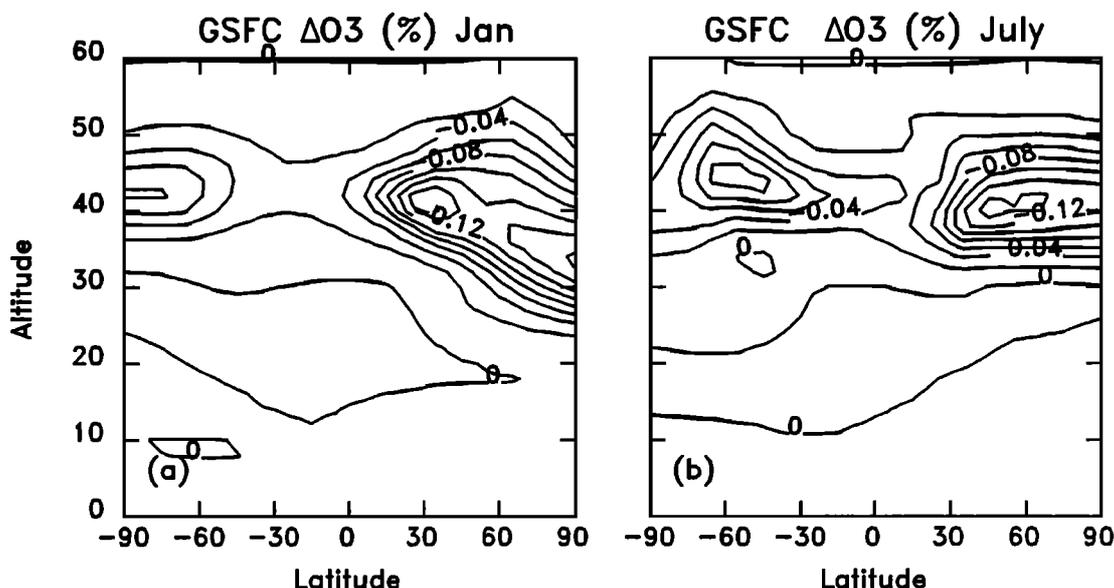


Fig. 8. Latitude by altitude (pressure) contours of the perturbation to ozone (%) from the GSFC model. See Figure 3.

(Cl or ClO). The path of the Shuttle is not vertically aligned, the corridor of exhaust gases is spread over lateral extent of more than a 1000 km in a day, and thus no local hole in column ozone could occur above the launch site.

This early stage following the launch is not adequately modeled in these calculations, but we believe that strict limits can be placed on the potential for global ozone destruction because the amount of chlorine injected is small compared with that contained in a 1000 km by 1000 km region (less than 0.2 ppb out of more than 2 ppb Cl_y). In order to have a significant impact on ozone globally, the plume must mix with the stratospheric environment and lead to significant perturbations over scales of at least 1000×1000 km.

In the few days following a launch, stratospheric winds will have stretched and dispersed the exhaust plume to scales greater than 1000 km. The average increase in stratospheric chlorine levels over such an area is modest, at most +5% within a 20° latitude by 20° longitude area. By the end of the month, these perturbations decrease rapidly to less than 0.2% above background levels as the chlorine is mixed laterally throughout the stratosphere.

A continuous series of rocket launches will lead to a buildup of chlorine in the stratosphere whose magnitude is governed by the frequency of launches and the rate of the stratosphere-troposphere circulation. For a scenario of nine Shuttle and six Titan IV launches per year, the accumulation of chlorine in the stratosphere is still modest, ranging from 0.2 to 0.6% over northern mid-latitudes and much less in the tropics and southern hemisphere. Corresponding ozone depletions are even smaller, less than 0.25% locally and less than 0.1% in the total column.

The addition of Cl_y to the winter polar stratosphere is of particular interest because of the role that chlorine plays in creating the Antarctic ozone hole; but the expected buildup at the poles represents only a small fractional increase to current levels. In the future, we might hope that stringent controls on industrial production of halocarbons, as envisaged by the Montreal Protocol, would reverse the current trend and lead to

lower background levels of Cl_y . Even the most optimistic controls over halocarbons would not lead to Cl_y concentrations less than 2 ppb much before the end of the twenty-first century.

The Shuttle and Titan IV solid rocket motors comprise the largest source of stratospheric chlorine that is expected from the current space fleet. The major launch vehicles from the United States and other space agencies use nonchlorinated (e.g., liquid) fuels or employ much smaller rockets. The U.S. strategic nuclear arsenal uses solid fuel containing chlorine, but in such launches most of the chlorine would be released below 15 km.

The exhaust from the solid rocket motors also contains other possible stratospheric pollutants. The major gaseous components are CO (24% by wt), HCl (21%), H_2O (10%), N_2 (9%), CO_2 (4%) and H_2 (2%). The perturbation to CO should be about the same in mixing ratio as that to Cl_y , and would not significantly affect the background levels of order 100 ppb. Clearly, the HCl represents the largest fractional perturbation to the background stratosphere; the remaining effluents should have negligible impact on the stratosphere.

Another principal exhaust product, particulate Al_2O_3 (30%), has the potential to perturb stratospheric chemistry. Most of the alumina is reported by Thiokol Corporation to form particles of radii greater than 1 micron; these fall out of the stratosphere more rapidly than the gaseous products which are removed by the circulation exchanging air between the stratosphere and troposphere (modeled here). The stratospheric abundance of larger alumina particles appears to be increasing and has been attributed to solid rocket motors as well as space debris [Radke *et al.*, 1982; Zolensky *et al.*, 1989].

Atmospheric measurements of Space Shuttle exhaust [Cofer *et al.*, 1987] indicate that about half of the mass of Al_2O_3 is in particles with radii less than 1 micron and thus might accumulate in a manner similar to Cl_y in these calculations. A simple scaling (i.e., Cofer *et al.* [1987] size distribution to a maximum of 10 ppt Cl_y in the lower stratosphere) gives an upper limit of 0.001 particles per cm^3 with radii greater than

0.1 micron as compared to background sulfate levels of order 1 per cm^3 . The corresponding surface area is $14 \times 10^{-12} \text{ cm}^2$ per cm^3 . These alumina particles may act as nuclei for ice deposition in the lower stratosphere [Turco et al., 1982]. Observational evidence [Cofer et al., 1984] and laboratory studies suggest that alumina particles react with the HCl in the exhaust to form chlorides and may provide active sites for Cl_2 - NO_x heterogeneous reactions in the lower stratosphere. Thus the buildup of small particles in the lower stratosphere may enhance both precipitation (dehydration or denitrification) and ozone destruction in the lower stratosphere. The role of particulates from solid rocket motors in the chemistry of the stratosphere is not well characterized today, and further research will be needed for a reliable assessment of future impacts.

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