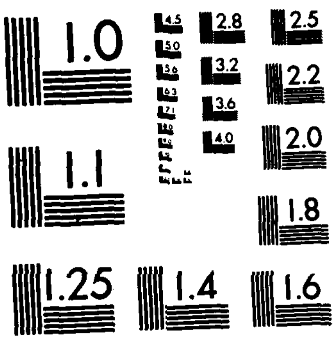
AD-A134 516 NATURAL AND ANTHROPOGENIC SOURCES OF OXIDES OF NITROGEN (NOX) FOR THE TROPOSPHERE(U) INSTITUTE FOR DEFENSE ANALYSES ALEXANDRIA VA E BAUER JAN 82 IDA-P-1619 1/1

UNCLASSIFIED FAA/EE-82-7 DTFA01-81-C-10011 F/G 4/1 NL



MICROCOPY RESOLUTION TEST CHART
NATIONAL BUREAU OF STANDARDS-1963-A

(12)

AD-A134 516

U.S. Department
of Transportation
Federal Aviation Administration

Natural and Anthropogenic Sources of Oxides of Nitrogen (NO_X) for the Troposphere

Office of Environment
and Energy
Washington, D.C. 20591

Ernest Bauer | January 1982 | FAA-EE-82-7

The work reported in this document was conducted under Contract No. DT-FA01-81C-10011 for the Department of Transportation. The publication of this IDA Report does not indicate endorsement by the Department of Transportation, nor should the contents be construed as reflecting the official position of that agency.

Technical Report Documentation Page

1. Report No. FAA-EE-82-7	2. Government Accession No. AD - A154516	3. Recipient's Catalog No.
4. Title and Subtitle Natural and Anthropogenic Sources of Oxides of Nitrogen (NO_x) for the Troposphere		5. Report Date January 1982
		6. Performing Organization Code
7. Author(s) Ernest Bauer		8. Performing Organization Report No. IDA Paper P-1619
9. Performing Organization Name and Address Institute for Defense Analyses 1801 N. Beauregard Street Alexandria, Virginia 22311		10. Work Unit No. (TRAIS)
		11. Contract or Grant No. DTFA01-81-C-10011
12. Sponsoring Agency Name and Address Department of Transportation Federal Aviation Administration Office of Environment and Energy Washington, D.C. 20591		13. Type of Report and Period Covered
		14. Sponsoring Agency Code Final
15. Supplementary Notes		

16. Abstract

This paper lists all known sources of oxides of nitrogen (NO_x) in the free troposphere to enable evaluation to be made of the significance of aircraft injections of NO_x for tropospheric photochemistry. Sources considered include the combustion of fossil fuels and biomass, lightning (summarizing the results of a parallel study by Kowalczyk and Bauer), transport from the stratosphere, and cosmic ray ionization. The results are provided in a form convenient for two-dimensional computation with extension to three-dimensional application. Data apply to 1975, with a projection to 1990.

17. Key Words Atmospheric pollution, Sources of oxides of nitrogen, NO_x, Aircraft, Fossil fuel combustion, Biomass combustion	18. Distribution Statement Document is available to the public through the National Technical Information Service, Springfield, VA 22151

19. Security Classif. (of this report)	20. Security Classif. (of this page)	21. No. of Pages	22. Price
Unclassified	Unclassified	52	

Form DOT F 1700.7 (8-72) Reproduction of completed page authorized

ABSTRACT

This paper lists all known sources of oxides of nitrogen (NO_x) in the free troposphere to enable evaluation to be made of the significance of aircraft injections of NO_x for tropospheric photochemistry. Sources considered include the combustion of fossil fuels and biomass, lightning (summarizing the results of a parallel study by Kowalczyk and Bauer), transport from the stratosphere, and cosmic ray ionization. The results are provided in a form convenient for two-dimensional computation with extension to three-dimensional application. Data apply to 1975, with a projection to 1990.

ACKNOWLEDGMENTS

I wish to thank the following people who provided information, data, and critical comments: Drs. A.C. Aikin (NASA-GSFC), F. Albini, R. Brandt, and C. Chandler (U.S. Forest Service), P.J. Crutzen* (Max-Planck Institut f. Chemie), M.L. Kowalczyk* (IDA), J.S. Levine (NASA-LaRC), H. Levy II* (NOAA-GFDL), S.C. Liu* (NOAA-ERL), J.A. Logan* (Harvard), R.C. Oliver (IDA) and R.W. Stewart (NASA-GSFC).

*Reviewers.

CONTENTS

TABLES

FIGURE

I. INTRODUCTION AND SUMMARY

Current and projected aircraft fleets inject large amounts of oxides of nitrogen, NO_x ($NO + NO_2$) into the atmosphere during cruise. For purposes of regulation, it is important to evaluate the significance of this source relative to other atmospheric sources of NO_x, both natural and anthropogenic, to determine whether limitations on aircraft exhaust emissions may be called for. The purpose of this compilation is to provide input data for two-dimensional model calculations of tropospheric photochemistry. Some guidance for extension to three dimensions is given in Appendix A.

The significance to atmospheric chemistry of a given source depends not only on its magnitude, but also on its injection height and location, and on local meteorological conditions, which affect its subsequent dispersal and removal. Thus, a given amount of NO_x injected near the tropopause in the upwelling Hadley Cell in the tropics, which may remain in the atmosphere for several months, will have a much greater effect on atmospheric photochemistry than an equal amount of NO_x from a combustion source injected near the surface, which will be lost within a few days. The discussion of Liu et al. (1980) suggests that past and current aircraft injections of NO_x may have significantly changed the current level of ozone in the Northern Hemisphere at mid-latitudes in the upper troposphere.

This paper lists NO_x sources in the troposphere and lowest stratosphere, supplementing an earlier "Catalog of Perturbations to Stratospheric Ozone, 1955-1975" [Bauer (1978a)] which considers neither tropospheric sources of NO_x, as such, nor steady sources of NO_x. Also not included here is a detailed discussion of lightning as a source, which is the subject of a separate report [Kowalczyk and Bauer (1981)].

All tropospheric sources of odd nitrogen are expressed as $NO_x = NO + NO_2$, but in considering transport from the stratosphere we list the flux of NO_y (= $NO_x + NO_3 + 2\ N_2O_5 + HNO_3 + HNO_2 + ClONO_2$ + PAN, i.e., total odd nitrogen); NO_x represents 5 to 20 percent of the total flux of odd nitrogen from the stratosphere [Levy et al. (1980)]. See Logan et al. (1981) for current review of observed NO, NO_2, and HNO_3 profiles in the troposphere.

In this summary, source strengths are listed in Tg N ^{12} g N, whereas in the following sections source strengths are sometimes listed in Tg NO_2. There is no inconsistency in this, but the user is cautioned to make the appropriate division of odd nitrogen into its separate chemical constituents.*

The principal sources of atmospheric NO_x are listed in Table 1, which presents source strengths, the heights and latitudes of injection, and where a particular topic is discussed, either in this paper or elsewhere.

Tables 2 through 6 give the best estimate of total injection of NO_x from all sources into the troposphere as a function of latitude and altitude, for use in a two-dimensional model. (See Appendix A, Table A.1, for a global, i.e. three-dimensional distribution).

*Note that in the EPA literature, emissions are typically quoted as mass of NO_x, which presumably is the sum of the mass of NO and the mass of NO_2. Pollutants are typically emitted as NO and then transformed in part to NO_2, so that if one quotes emissions on a mass basis there is a certain ambiguity in the number of molecules emitted. In this paper, NO_x mass emission rates are interpreted as NO_2.

TABLE 1. ATMOSPHERIC SOURCES OF ODD NITROGEN: 2-DIMENSIONAL REPRESENTATION

Source	Injection Rate (Tg N/yr)	Mean Injection Height (km)	Latitudinal Range of Injection	For Details See:
Aircraft	0.15 (1975) 0.53 (1990)	6-16 km	Northern Hemisphere, mid/high latitudes, see Tables 19,10	Section 6
Fossil fuel combustion	19.0 (1975) 27.0 (1990)	Ground level Ground level	Northern Hemisphere, mid-latitude, see Table 12	Section 2
Biomass burning				Section 3
Forest fires	1.7	1-2 km	Tropics	
Other	3.3 (1975) 3.8 (1990)	Ground level Ground level		
Lightning	5.7	7-12 km and lower	Tropics, land only	Section 5
Transport from stratosphere	0.5*	Tropopause	See Table 24	Section 8
Cosmic rays	0.06	Upper troposphere		Section 7
Exhalation from soils	~10.0	Ground level		Section 4A
NH_3 Decomposition	<8.0	Throughout troposphere		Section 4C; Crutzen (1979)

*Total odd nitrogen, of which 5-20% is NO_x.

TABLE 2. TOTAL ANNUAL INJECTION RATE OF NO_x INTO THE TROPOSPHERE FROM MISCELLANEOUS NEAR-SURFACE SOURCES FOR USE IN A 2-DIMENSIONAL MODEL
[Tg N/yr = 10^{12}g N/yr]

Altitude (km)	Southern Latitudes				Northern Latitudes						
	40–30	30–20	20–10	10–0	0–10	10–20	20–30	30–40	40–50	50–60	60–70
	Fossil Fuels and Biomass Burning (1975)										
Ground Level		0.68	0.77	1.06	1.05	0.97	0.66	4.40	7.60	3.80	0.92
	Fossil Fuels and Biomass Burning (Projected to 1990)										
Ground Level		1.28	1.38	1.72	1.71	1.62	1.26	5.35	9.45	5.21	1.41
	Exhalation from Soils										
Ground Level		0.02	0.05	0.06	0.07	0.04	0.08	1.1	1.5	1.6	2.1
	Forest Wildfires										
1-2 km	0.003	0.16	0.20	0.35	0.35	0.31	0.15	0.04	0.04	0.07	0.04

Notes:
The number of digits is not significant.
If applied to a three-dimensional model, note that all these sources occur essentially over land only.

TABLE 3. ANNUAL NO_x INJECTION RATE FROM LIGHTNING: 2-DIMENSIONAL DISTRIBUTION

10^{12} gN/yr-km

ALTITUDE km \ LATITUDE	-60 to -50	-50 to -40	-40 to -30	-30 to -20	-20 to -10	-10 to 0	0 to 10	10 to 20	20 to 30	30 to 40	40 to 50	50 to 60
14–15				0.020	0.039	0.049	0.045	0.033	0.030			
13–14				0.024	0.046	0.057	0.057	0.039	0.035			
12–13				0.027	0.052	0.066	0.060	0.045	0.041			
11–12	—	0.001	0.006	0.032	0.062	0.079	0.072	0.053	0.049	0.032	0.012	0.001
10–11	—	0.002	0.007	0.036	0.071	0.089	0.081	0.060	0.055	0.037	0.014	0.002
9–10	—	0.002	0.008	0.016	0.028	0.031	0.030	0.024	0.024	0.042	0.016	0.002
8–9	—	0.002	0.009	0.018	0.031	0.035	0.034	0.027	0.027	0.047	0.018	0.002
7–8	—	0.003	0.010	0.021	0.035	0.040	0.038	0.031	0.030	0.053	0.020	0.003
6–7	—	0.003	0.011	0.023	0.039	0.044	0.043	0.034	0.034	0.060	0.034	0.005
5–6	—	0.003	0.013	0.026	0.044	0.049	0.048	0.038	0.038	0.067	0.038	0.006
4–5	—	0.004	0.014	0.029	0.049	0.055	0.053	0.043	0.042	0.075	0.042	0.006
3–4	—	0.004	0.015	0.032	0.054	0.061	0.059	0.048	0.046	0.083	0.046	0.007
2–3	—	0.005	0.017	0.035	0.060	0.068	0.065	0.053	0.051	0.092	0.051	0.007
1–2	—	0.005	0.019	0.039	0.066	0.075	0.072	0.058	0.057	0.101	0.057	0.008
0–1	—	0.006	0.021	0.042	0.073	0.083	0.079	0.064	0.063	0.112	0.063	0.009

TABLE 4A. TOTAL ANNUAL INJECTION RATE OF NO_x INTO THE ATMOSPHERE FROM AIRCRAFT FOR USE IN A 2-DIMENSIONAL MODEL (1975 Data)
(Tg N/yr-km = 10^{12}g N/yr-km)

Altitude (km)	Southern Latitudes							Northern Latitudes						
	S. Pole–60	60–50	50–40	40–30	30–20	20–10	10–0	0–10	10–20	20–30	30–40	40–50	50–60	60–N. Pole
14–13	0	0	1.18E-9	1.87E-6	4.16E-9	3.89E-6	8.39E-7	2.94E-6	1.01E-5	1.71E-5	6.51E-5	6.26E-5	5.32E-6	8.94E-7
13–12	0	3.22E-10	1.88E-6	4.16E-5	3.44E-5	3.34E-5	2.12E-5	3.53E-5	1.53E-4	2.81E-4	8.42E-4	5.41E-4	6.96E-5	5.02E-6
12–11	0	8.57E-8	1.06E-5	2.56E-4	3.95E-4	5.59E-4	5.41E-4	6.17E-4	1.25E-3	3.83E-3	7.96E-3	8.00E-3	5.65E-3	1.08L-3
11–10	0	7.60E-7	3.80E-5	6.48E-4	7.27E-4	8.48E-4	7.39E-4	7.93E-4	1.82E-3	5.56E-3	1.69E-2	1.40E-2	6.23E-3	1.08E-3
10–9	0	1.20E-6	2.37E-5	4.23E-4	5.53E-4	6.81E-4	7.27E-4	7.48E-4	1.26E-3	4.44E-3	1.23E-2	1.23E-2	6.87E-3	1.26E-3
9–8	0	1.58E-6	1.83E-5	2.74E-4	2.39E-4	1.78E-4	2.07E-4	2.00E-4	5.08E-4	1.41E-3	5.65E-3	4.68E-3	1.04E-3	1.49E-4
8–7	0	9.24E-7	5.02E-6	9.58E-5	9.15E-5	5.29E-5	7.33E-5	8.34E-5	2.25E-4	5.93E-4	1.54E-3	1.52E-3	3.83E-4	7.96E-5
7–6	0	9.24E-7	5.02E-6	9.58E-5	9.15E-5	5.29E-5	7.33E-5	8.34E-5	2.25E-4	5.93E-4	1.54E-3	1.52E-3	3.83E-4	7.96E-5

TABLE 4B. TOTAL ANNUAL INJECTION RATE OF NO_x INTO THE ATMOSPHERE FROM AIRCRAFT FOR USE IN A 2-DIMENSIONAL MODEL (1990 Base Case) (Tg N/yr-km = 10^{12}g N/yr-km)

Altitude (km)	Southern Latitudes S. Pole–60	60–50	50–40	40–30	30–20	20–10	10–0	Northern Latitudes 0–10	10–20	20–30	30–40	40–50	50–60	60–N. Pole
19	0	0	0	1.10E-6	1.26E-5	1.40E-5	3.10E-5	3.04E-5	3.22E-5	8.72E-5	1.62E-4	4.47E-4	7.05E-4	2.70E-4
18	0	0	0	5.38E-6	2.22E-5	2.96E-5	6.66E-5	6.44E-5	7.93E-5	3.13E-4	3.86E-4	1.03E-3	1.54E-3	3.98E-4
17	0	0	0	8.45E-6	2.64E-5	4.13E-5	7.93E-5	7.63E-5	1.01E-4	3.50E-4	5.08E-4	1.32E-3	1.95E-3	4.89E-4
16	0	0	0	2.92E-6	1.88E-5	2.06E-5	5.65E-5	5.44E-5	6.08E-5	1.50E-4	2.45E-4	7.87E-4	1.26E-3	3.25E-4
15	0	0	0	9.36E-6	0	2.47E-5	0	0	2.84E-5	2.20E-4	3.22E-4	3.95E-5	3.98E-4	7.72E-5
14	0	0	3.25E-9	1.64E-5	9.91E-6	5.93E-5	2.60E-5	3.25E-5	8.76E-5	2.89E-4	5.84E-4	8.88E-4	7.05E-4	1.92E-4
13	0	2.71E-7	1.79E-5	2.36E-4	1.81E-4	2.13E-4	1.74E-4	2.34E-4	6.78E-4	1.64E-3	5.78E-3	4.77E-7	1.56E-3	2.52E-4
12	0	2.05E-6	8.63E-5	1.19E-3	1.67E-3	2.21E-3	2.07E-3	2.57E-3	5.41E-3	1.32E-2	3.34E-2	3.10E-2	1.55E-2	2.80E-3
11	0	4.83E-6	1.60E-4	2.29E-3	2.65E-3	2.93E-3	2.65E-3	3.40E-3	7.96E-3	1.93E-2	6.02E-2	3.10E-2	2.01E-2	2.50E-3
10	0	5.63E-6	6.99E-5	1.29E-3	1.83E-3	2.19E-3	2.33E-3	2.83E-3	4.96E-3	1.28E-2	3.22E-2	3.47E-2	1.80E-2	2.75E-3
9	0	7.24E-6	5.93E-5	9.03E-6	7.75E-4	6.17E-4	7.30E-4	9.64E-4	2.19E-2	5.35E-3	1.79E-2	1.68E-2	2.44E-3	5.81E-4
8	0	4.65E-6	2.30E-5	3.89E-4	3.56E-4	2.64E-4	3.22E-4	4.53E-4	1.03E-3	2.48E-3	7.51E-3	7.33E-3	2.23E-3	3.22E-4
7	0	4.65E-6	2.30E-5	3.89E-4	3.56E-4	2.64E-4	3.22E-4	4.53E-4	1.03E-3	2.48E-3	7.51E-3	7.33E-3	2.23E-3	3.22E-4
6														

TABLE 5. TOTAL ANNUAL INJECTION RATE OF ODD NITROGEN (NO_y) INTO THE TROPOSPHERE FROM THE STRATOSPHERE FOR USE IN A 2-DIMENSIONAL MODEL ($Tg\ N/yr = 10^{12}\ N/yr$)

Altitude	Southern Latitudes									Northern Latitudes								
(km)	90-80	80-70	70-60	60-50	50-40	40-30	30-20	20-10	10-0	0-10	10-20	20-30	30-40	40-50	50-60	60-70	70-80	80-90
16							0.050	0.041	0.024	0.033	0.049	0.075						
13				0.012	0.019	0.038							0.075	0.033	0.012			
9	0.0025	0.0065	0.007													0.095	0.009	1.003

TABLE 6. NO_x PRODUCTION DUE TO GALACTIC COSMIC RAY IONIZATION IN THE TROPOSPHERE
[After Heaps, Appendix E, Bauer (1978a)]
(Units: Tg N/yr)

	In both polar caps (geomagnetic latitudes > 60°, uniformly distributed between 5 and 10 km altitude)	At low geomagnetic (geomagnetic latitudes < 60°, uniformly distributed between 5 and 15 km altitude)
At Solar Minimum (1976, 1987, ...)	0.024	0.048
At Solar Maximum (1969, 1980, 1991, ...)	0.014	0.043

It must be stressed that all emissions other than those due to aircraft and surface fossil fuel combustion are exceedingly uncertain. I cannot provide error bounds, but suggest an overall factor of variation of 2 to 3 as a starting point for discussion.

II. THE FOSSIL FUEL COMBUSTION SOURCE OF NO_x

A. INTRODUCTION

When fossil fuels are burned, NO_x is produced, partly from nitrogen compounds in the fuel and, also, to some extent, as a result of heating air to temperatures above 2000 to 2500 K. Current figures for U.S. NO_x emissions are relatively detailed and adequate [see, e.g., Benkovitz (1980)], but when one asks for global figures, in particular for projections to 1990, there are large uncertainties. Here I use reasonably current and authoritative figures for energy use, but the projections have been changing over the last few years, predicting lower growth rates as the increase in prices of fossil fuels has led to a reduction in consumption. This source of uncertainty must be borne in mind if one wishes to use the present quantitative estimates.

First I list U.S. energy use for 1975 and projections to 1990, and then provide available data and estimates for the rest of the world. Then I present estimates of NO_x emission indexes for the various major fuels and finally estimate worldwide NO_x emissions due to fossil fuel combustion.

B. U.S. ENERGY USE

Table 7 lists U.S. energy use for 1978 and several projections for 1990. These projections come from the Energy Information Administration of the U.S. Department of Energy (EIA) and from EXXON (1980). Reference to Table 7 shows that anticipated changes between 1978 and 1990 are not very large (0.89 percent mean annual increase, compounded over the period), and that the estimates for U.S. energy use listed by EXXON (1980) are comparable with the other sources used by EIA (1980), as follows:

- Data Resources Inc., "Energy Review," Autumn 1980
- Bankers Trust Company, "U.S. Energy and Capital Forecast, 1980-1990," Summer 1980
- Policy and Evaluation, U.S. Department of Energy, "Reducing U.S. Oil Vulnerability," November 1980

TABLE 7. U.S. ENERGY USE (1978) AND PROJECTIONS FOR 1990
[in Q/yr][a]
[Source: EIA (1980), p. 107]

Energy Source	1978	1990; From EXXON (1980)	Three Others Used by EIA
Oil	37.8	34.5	30.7 to 31.5
Gas	20.4	17.8	20.1 to 20.7
Coal	14.1	20.9	24.5 to 25.8
Other[b]	6.1	14.0	12.3 to 13.1
Total	78.4	87.2	87.6 to 91.1

[a] $Q = 10^{15}$

[b] Nuclear, hydroelectric, or solar energy, i.e., does not produce any NO_x.

C. WORLDWIDE FOSSIL FUEL USE

The results discussed below are derived by the same methodology as the U.S. figures, but are less reliable. Table 8 lists world energy use and Table 9 lists the worldwide distribution of energy demand. These figures come from EXXON (1980), the most current and authoritative source I have found. The projected worldwide rate of increase of energy use, 2.8 percent per annum, is significantly greater than the rate of increase projected for U.S. energy use.

TABLE 8. WORLD ANNUAL ENERGY USE (1975) AND PROJECTION TO 1990 (in Q/yr) [Source: EXXON (1980)]

Energy Source	1975	1990
Oil	110	137
Gas	42	72
Coal	66	97
Other	22	55
Total	240	361

TABLE 9. GLOBAL DISTRIBUTION OF ENERGY DEMAND, 1975 AND 1990 [Source: EXXON (1980) Chart 3, Figures in Percent of Total]

Country/Group	Latitude Range	1975	1990
U.S.	30°-50°N	29	23
Canada	45°-50°N	4	3
Europe	40°-60°N	19	18
Japan	30°-45°N	6	5
Centrally Planned Economies (USSR, China, E. Europe)	65°-35°N	29	30
Other	30°N-30°S	12	21

D. NO_x EMISSION INDEXES

Table 10 lists emission indexes for different fossil fuels, and Table 11 compares the U.S. NO_x emissions predicted from the results of Tables 7 and 10 with the DOE 1975 figures and median projections of Pechan (1978). Since the upper-bound estimates of emission indexes from Table 10 are consistent with Pechan's projections, these upper-bound values are used here.

TABLE 10. NO_x EMISSION INDEXES FOR DIFFERENT FOSSIL FUELS

A. Emission Indexes [Source: Böttger et al. (1980) values in [ ] from NAPCA (1970)]

Coal: 3.0 - 9.0 g NO_2/kg [3.6 - 8.9]

Natural Gas: 2.0 - 9.9 g NO_2/m^3 [1.9 - 6.3]

Oil: 4.9 - 9.8 g NO_2/kg [2.1 - 40]

B. Energy Equivalence [Source: NRC (1979), p. xxxviii]

1 Q = 10^{15} BTU = 0.5 Mbpd oil equivalent*

1 Q corresponds to:

44.3 million short tons coal = 4.02×10^{10} kg

0.979 trillion ft^3 natural gas = 2.77×10^{10} m^3

181 million barrels oil (assume 140 kg/barrel) = 2.53×10^{10} kg

C. Thus, emission index, in Tg NO_2/Q:

Coal	0.12 - 0.36
Natural Gas	0.055 - 0.27
Oil	0.12 - 0.24

*Mbpd = million barrels per day.

TABLE 11. U.S. NO_x EMISSIONS: COMPARISON OF TWO ESTIMATES

Year	1975	1985	1990
Total Annual Energy Use (Q) [Pechan (1978)]	77.8	94.6	108.6
Fractional Distribution by Source[a]			
Coal	0.158	0.212	0.240
Natural Gas	0.282	0.228	0.204
Oil	0.505	0.430	0.396
Total Fossil Fuel	0.945	0.870	0.840
NO_x Emissions[b] (Tg NO_2)/yr)			
Coal	1.5 - 4.5	2.4 - 7.2	2.8 - 8.3
Natural Gas	1.2 - 6.0	1.2 - 5.9	1.4 - 6.8
Oil	4.7 - 9.4	4.9 - 9.8	5.2 - 10.3
Total	7.4 - 19.9	8.5 - 22.9	9.4 - 25.4
Total NO_x Emissions from Pechan (Tg NO_2/yr)	21.8	23.5	25.1

[a]Linear extrapolation and interpolation from Table 7.

[b]Uses present source strengths and emission index ranges from Table 10, item C.

TABLE 12. LATITUDINAL DISTRIBUTION OF GLOBAL EMISSIONS, OF NO_x FROM FOSSIL FUELS 1975 AND 1990

Latitude	1975	1990
60-70°N	0.045	0.050
50-60°N	0.195	0.190
40-50°N	0.400	0.352
30-40°N	0.230	0.198
20-30°N	0.020	0.035
10-20°N	0.020	0.035
0-10°N	0.020	0.035
0-10°S	0,020	0.035
10-20°S	0.020	0.035
20-30°S	0.020	0.035
Total	0.990	1.00

E. WORLDWIDE NO_x EMISSIONS

Combining the energy use figures of Table 8 and the upper-bound emission index values of Table 10 yields total NO_x emissions of 61.7 Tg NO_2 (18.8 Tg N) in 1975, and 87.5 Tg NO_2 (26.6 Tg N) in 1990. Assuming that the breakdown of energy use between "coal," "gas," "oil," and "other" is uniform over the globe at any given time, and ignoring the possible effects of air-pollution controls in reducing NO_x production as well as the possible effect of higher-temperature (more efficient) combustion in increasing NO_x production, the global distribution of NO_x emissions will be the same as the global distribution of energy demand of Table 9, giving the latitudinal distribution of Table 12.

Regarding uncertainties, the self-consistency of the various estimates of U.S. fuel use in 1975-1978 is no better than 10 percent. Projections surely differ by more than this for the

U.S.A. and become more uncertain as one goes to worldwide fuel use estimates and emission index figures. I do not know how to quantify these uncertainties.

III. BIOMASS BURNING AS A SOURCE OF ATMOSPHERIC NO_x

A. INTRODUCTION

There are several different sources of biomass burning, namely:

- Forest wild fires*
- Forest burning associated with shifting agriculture in the tropics
- Bushland and savanna burning
- Deforestation by burning due to population increase
- Wood burning for fuel

Each of these sources produces some NO_x, mainly from nitrogen compounds in the wood, leaves, and bark, but with some contribution from the heating of air to temperatures above 2000 to 2500 K. To estimate the overall significance of these individual sources of NO_x, one must know the total mass burned, the emission index (EI), and the effective height of injection of the NO_x.

Table 13 presents several estimates for various components of the global biomass burned; the relatively good agreement between the different estimates does not necessarily mean that the "truth" lies within the range listed. Table 14 presents statistics and estimates on forest fires and Table 15 presents estimates on wood burning for fuel. Forest fires are generally more energetic than the other sources, so that they have a higher effective injection height, and thus residence time, for the NO_x produced. The problem of injection height is discussed

---

*Forest fires not associated with human activity.

in Subsection C, after a discussion in Subsection B of emission indexes, which are taken to be the same for all the sources considered here.

Note that, in contrast to fossil fuel combustion which occurs predominantly at mid to high latitudes in the Northern Hemisphere, most biomass is burned in the tropics.

TABLE 13. BIOMASS BURNED PER YEAR[a] (UNITS: 10^3 Tg CO_2/yr)[b]

A. Total Estimate

- 5.5, Bolin (1977); Wong (1978)[c]
- 11 (8 to 15), Seiler and Crutzen (1980)
- 11.5, Logan et al. (1981)
- *10.6, My estimate for 1975, 11.3 for 1990

B. Breakdown

Forest Fires

- 3.9 (3.2 to 5.6), Seiler and Crutzen (1980),[d]
- *4.4, My estimate derived from Table 14

Shifting agriculture

- *2.8 (1.5 to 4.1), Seiler and Crutzen (1980), p. 220

Deforestation due to population increase

- *1.2 (0.9 to 1.5), Seiler and Crutzen (1980), p. 225

Fuel wood burning

- 0.5 ± 0.3, Bolin (1977)
- 1.7 (1.6 to 1.8), Crutzen et al. (1979); Seiler and Crutzen (1980)
- *2.2 (1.5 to 2.9), My estimate from Table 15 for 1975; 2.9 (2.0-3.9) for 1990

[a]*Denotes that this estimate is the one used in this paper.

[b]I use the conventional conversion that 1 g CO_2 corresponds to 0.606 g dry matter or 0.273 g carbon.

[c]Woodwell et al. (1978) suggest that this estimate may be too small by as much as a factor of two, but Fahnestock (1979) and Wong (1979) suggest that it may be an upper bound.

[d]Seiler and Crutzen (1980) give no explicit discussion of forest fires in the tropics. They give an estimate of 2.4 to 3.8 for burning in bushland and savannas, which seems much too high compared with the estimate of Table 14 and therefore is not included here.

TABLE 14. GLOBAL DISTRIBUTION OF FOREST FIRES ON BASIS OF BIOMASS BURNED
(Source: private communication from Dr. Craig Chandler, Head, Fire Research, U.S. Forest Service, February 1981)

A. Amount of Wood Burned per Hectare

Tropical forest clearing	120 tons/ha
African range burning	5 tons/ha
All others	40 tons/ha

B. Annual Extent of Forest Fires

	Area (10^6 ha)	Wood burned (10^6 ton)
U.S.A.	1.8[a]	72[a]
Canada	1.1[a]	44[a]
Australia	0.36[a]	14[a]
West Europe	0.08[a]	3[a]
Spain	0.4[a]	16[a]
Rest of Mediterranean	0.8[b]	32[b]
USSR	3.0[b]	120[b]
Latin America	10.1[b]	1212[b]
Asia	7.8[b]	936[b]
Africa	1.7[b]	204[b]
Africa-range burning	1.7[b]	9[b]
Totals	28.8	2660

C. Normalized Global Distribution of Biomass Burning Due to Forest Fire

	Latitude	Normalized Fraction
North America	(30-60°N)	0.044
Latin America	(20°N-30°S)	0.455
West Europe	(40-60°N)	0.001
USSR	(50-70°N)	0.045
Mediterranean	(30-50°N)	0.017
Asia	(10°S-30°N)	0.352
Africa	(20°N-10°S)	0.080
Australia	(10-40°S)	0.006
Total		1.000

a "firm number"
b "estimate"

TABLE 15. GLOBAL USE OF WOOD AS FUEL

A. Estimates of Total Burning (in 10^{15} g CO_2 yr)

0.5 ± 0.3, Bolin (1977)

1.7 (1.6 to 1.8), Crutzen et al. (1979)

2.0, Seiler and Crutzen (1980), p. 230

*2.2 (1.5 to 2.9), My estimate for 1975 (see B below)

*2.9 (2.0 to 3.9), My estimate for 1990

B. The Model

- Annual mean world consumption of fuel wood, 1963-1974 is 1.18 x 10^9 m^3 of roundwood - FAO (1976)
- Assume a mean density 1.48 m^3/tonne (primate communication from Dr. R. Brandt, Head, International Forestry, U.S. Forest Service, December 1981)
- Assume use of firewood is proportional to population, which increases at 2% per annum [World Almanac and Book of Facts (1980), p. 513ff]
- Assume estimates may be low by a factor of 2 due to underreporting [Seiler and Crutzen (1980), p. 228ff]

C. Global Distribution of Wood Burning [FAO (1976)]

North and Central America	0.043
South America	0.163
Europe	0.035
USSR	0.069
Africa	0.239
Asia	0.445
Australasia	0.005

*Denotes that this estimate is the one used in this paper.

B. EMISSION INDEX (EI)

Various measurements of the EI in different cases of biomass burning situations are listed in Table 6. Considering the difficulties associated with the measurements and the inherent variation between the different cases considered--see, e.g., Crutzen et al. (1979)--the overall agreement of measurements of Table 6 is remarkably close. For all the applications considered here, I adopt a mean value from Table 6 of 4.2 g NO_2/kg fuel burned. Note that, while most forest fires occur in the tropics, there are as yet no data on the EI from tropical forest fires. [Observations in Brazil have been made by National Center for Atmospheric Research (NCAR) but they have not yet been analyzed or reported].

Comparison with Section 2 shows that the EI for biomass burning is smaller by a factor 3 to 4 than that due to fossil fuel combustion. Presumably, this is because the combustion temperature in most biomass burning is relatively low so that most of the NO_x must originate from oxidation of the fixed nitrogen in plant tissue rather than from heating of the air to temperatures above 2000 to 2500 K, as in the case of lightning. However, reference to the summary of Cook et al. (1978) who review "prescribed burns" in Oregon and Washington (see item e of Table 16), gives a somewhat lower EI than is adopted here. Prescribed (controlled) burns involve letting the trees dry out before burning them, so that the net temperature generated will tend to be somewhat higher than in a wild fire. This may lead to production of more N_2 and less NO_x from fixed nitrogen in the plant material, but more NO_x by heating air.

C. HEIGHT OF POLLUTANT INJECTION DUE TO FOREST FIRES

Substantial forest fires send smoke clouds well into the free troposphere, above the planetary boundary layer. Thus, the contribution of NO_x injections from forest fires to the

TABLE 16. NO_x EMISSIONS FROM FOREST FIRES

	Volume Mixing Ratio $[V(NO_x)/V(CO_2)]$	Mass Emission Index (gm NO_2/kg fuel)	Experiment	Reference
a.	$(1.2 \text{ to } 2.3) \times 10^{-3}$	2 to 4	Open burning of agricultural waste (California)	Darley et al. (1966)
b.	$(0.3 \text{ to } 1.2) \times 10^{-3}$	0.5 to 2	Open burning of landscape refuse (Ohio)	Gerstle and Kemnitz (1967)
c.	6.5×10^{-3}	11.2	Pine slash burning (Washington State)	Malte (1975)
d.	$(0.6 \text{ to } 1.0) \times 10^{-3}$	1.0 to 1.7	Airborne measurements of prescribed burns in Australia	Evans et al. (1977)
e.	$(0.6 \text{ to } 1.8) \times 10^{-3})$	1 to 3	Best estimate for prescribed burns	Cook et al. (1978)

global NO_x burden will tend to be larger than it would be if the injection occurred at the surface because of the increase in effective atmospheric residence time.

For the energetics, one may estimate a mean energy output of a forest fire of 8500 Btu/lb of wood burned, or 2×10^7 J/kg. The distribution of this energy is roughly 50 percent latent heat of vaporization of water, 25 percent radiation, and 25 percent convection (C. Chandler, private communication). Thus, for a 10 ha (25 acre) forest fire, with a mass loading of 40 t/ha (mid-latitude) or 120 t/ha (tropical), the total energy output is, respectively, 3 or 24 TJ, (1 TJ = 10^{12} Joule) i.e., 0.75 or 6 kt TNT equivalent.

The cloud rise height is a function of the energy output of a fire. Thus, for example, the "Meteotron," a very large array of kerosene burners (in Southern France), which dissipates energy at a rate of 1000 MW over a typical burning time of 20 min, so that it generates 1.8 TJ, sends its plume to altitudes of 1 to 2 km [Church et al. (1980)]. On an overall basis, Chandler reports that in the U.S. the average forest fire plume rises to 4000 to 6000 ft, with heights of up to 43,000 ft (13 km) reported. For more detail, see Fig. 6 in Taylor et al. (1973) which provides estimates for the plume rise height as a function of fuel burned per acre in a large controlled burn. See also Cook et al. (1978) who report on plume rise and transport on p. 89 ff. Some further reports are given by Evans et al. (1977) and Westberg et al. (1981). Note that all of these data refer to the U.S. or to Australia rather than to the tropics, which are a much larger source of NO_x.

In the U.S., at least, the combustion of a forest fire is non-uniform, with vigorous activity every afternoon alternating with long periods of smoldering. Most of the burning occurs during the afternoon bursts, and thus the plume associated with most of the NO_x and CO_2 emissions penetrates well above the boundary layer [Albini (1981)].

From this data base I conclude that the energy of a substantial (multi-acre) forest fire is so large that the cloud rises well into the free troposphere. Accordingly, it is postulated that the NO_x injection is in the 1- to 2-km altitude range. This is a very tentative conclusion, since most forest fire injection of NO_x occurs in the tropics, while most data apply to mid-latitudes.

By contrast, all the other sources of biomass combustion considered here will be much less energetic as individual events, and thus are combined with fossil fuel combustion (other than aviation) in providing a near-surface injection.

D. ANNUAL NO_x INJECTION RATE DUE TO BIOMASS BURNING

From Table 13 the annual combustion rate corresponds to 2.7×10^{15} g of dry matter/yr due to forest fires, 1.3×10^{15} g of dry matter/yr due to fuel wood burning (in 1975: 1.8×10^{15} g of dry matter/yr in 1990), and 2.4×10^{15} g of dry matter/yr due to other sources. Except for fuel wood burning, I reduce the figure for the amount of material that is actually burned at high temperature, producing NO_x, rather than just charred logs, by a factor of 2 [see, e.g. Seiler and Crutzen (1980) for a more detailed discussion of this].

Subsection B provides an EI of 4.2 g NO_2/kg fuel burned, giving an annual atmospheric injection rate of 5.7 Tg NO_2/yr (1.7 Tg N/yr) due to forest fires, which is injected in the 1 to 2 km altitude range. The other sources of biomass burning give a ground-level injection of 10.7 Tg NO_2/yr (3.3 Tg N/yr) in 1975 and 12.5 Tg NO_2/yr (3.8 Tg) N/yr) in 1990.

Regarding the latitude distribution of injections, I shall assume that all sources of biomass follow the distribution of forest fire injections, (see item C in Table 14) and thus Table 17 gives the latitude distribution of NO_x injections due to biomass burning. Table 17 also gives the maximum land elevation above mean sea level, h_M. The altitude range of injection of NO_x due to forest fires should be taken as (1/2 h_M + 1 km) to (1/2 h_M + 2 km), if the altitude resolution of the model is such that 1/2 h_M is not effectively zero.

TABLE 17. NORMALIZED GLOBAL DISTRIBUTION OF NO_x INJECTIONS DUE TO FOREST FIRES FOR USE IN A 2-DIMENSIONAL MODEL

Latitude	Fraction of Biomass Burned	Maximum Land Elevation Above Mean Sea Level, h_M (m)
>70°N	-	-
60-70°N	0.022	<200
50-60°N	0.040	<200
40-50°N	0.023	<1000
30-40°N	0.023	<1000
20-30°N	0.087	<300
10-20°N	0.180	<300
EQ-10°N	0.204	<300
10°S-EQ	0.208	<300
20-10°S	0.118	<300
30-20°S	0.093	<300
40-30°S	0.002	<500
<40°S	-	-

Notes:

1. Results compiled from information presented in Table 14.
2. The height range of NO_x injections should be taken as $(1/2\ h_M + 1\ km)$ to $(1/2\ h_M + 2\ km)$.
3. Depending on the complete code used, it may be appropriate to ignore h_M.

IV. SOME OTHER TERRESTRIAL SOURCES OF NO_x

A number of other terrestrial sources of NO_x have been suggested; they will be discussed here in turn.

A. BACTERIAL PRODUCTION OF NO_x IN SOILS

From experiments conducted by placing an open-ended box on a patch of ground and measuring the rate of increase in NO_x, Galbally and Roy (1978) have suggested that NO_x exhalations from soil may account for a source of 10 Tg N/yr. This figure seems rather high, but a quite different set of experiments by Lipschultz et al. (1981) using cultures of nitrifying bacteria gives a comparable estimate (15 Tg N/yr) as an upper bound. Thus I suggest a tentative source strength of this order should be considered. [Note that this is much lower than the estimate of 166 Tg N/yr of Robinson and Robbins (1968)]. The latitude distribution may be assumed to be proportional to land surface area.

B. OXIDATION OF AMMONIA FROM FERTILIZER VOLATILIZATION AND FROM DECOMPOSITION OF ANIMAL AND OTHER ORGANIC WASTES UNDER ALKALINE CONDITIONS

Global fertilizer production is great (66 Tg N/yr in 1978), and is increasing at perhaps 3.7 percent compounded annually [Foster (1980)], so that it would be 102 Tg N/yr by 1990. Possibly up to 10 percent of this fertilizer is volatilized as NH_3. Some of this will rain out directly. Some will form aerosols (ammonium chloride, nitrate, or sulfate) which rain out and some NO_x will be formed. Levine et al. (1980, and work in progress) have discussed the problem of atmospheric ammonia recently. At present, it is not feasible to consider this as a quantifiable source of tropospheric NO_x: Crutzen (1979) has estimated an upper bound of 8 Tg N/yr, while Logan et al. (1981) suggest that oxidation of NH_3 could provide a sink of odd nitrogen for $[NO_x] > 60$ ppt.

C. NITRITE PHOTOLYSIS IN TROPICAL OCEANS

Zafiriou and McFarland (1981) estimated the rate of solar photolysis of nitrite ions in the central equatorial Pacific, and found that it is a small source, perhaps 0.05 Tg N/yr if it occurs over 20 percent of the surface of the globe. It is mentioned here mainly to indicate the potential importance of oceanic sources of NO_x.

D. CHEMI-DENITRIFICATION IN ACIDIC SWAMPS AND SOILS

This process has been suggested, but not quantified by the National Research Council (1978), p. 276.

V. NO_x PRODUCTION BY LIGHTNING

Lightning discharges heat large amounts of air to temperatures above 2000 to 2500 K and so provide a significant source of atmospheric NO_x. A number of workers have made estimates of the global production of NO_x due to lightning, ranging from 2 to 80 Tg N/yr. In a detailed review of this topic, Kowalczyk and Bauer (1981) estimate a net production rate of 5.7 Tg N/yr, with an estimated uncertainty range of 2 to 20 Tg N/yr.

Most lightning occurs in the tropics and at mid-latitudes over land. Lightning discharges generally occur at altitudes between 5 and 8 km, but the effective injection altitude of NO_x is not necessarily the same as the discharge altitude because of strong updrafts and downdrafts which may transport thunderclouds up to the vicinity of the local tropopause.

One should distinguish between cloud-to-cloud discharges (which are often not adequately reported in ground-based observations, but which provide perhaps 30 percent of the NO_x source on a season/latitude averaged basis) and cloud-to-ground discharges. For cloud-to-cloud discharges the injection is postulated to be near the tropopause, while for cloud-to-ground discharges the injection will be uniformly distributed between the discharge altitude, corrected somewhat for cloud rise, and the surface.

Table 18 gives some details of the assumed NO_x production per flash, of the global lightning frequency (flashes/sec), of the partitioning between cloud-to-cloud and cloud-to-ground discharges, and of the assumed injection height of NO_x as a function of latitude. Table 19 summarizes the results of Kowalczyk

TABLE 18. SUMMARY OF LIGHTNING PARAMETERS AND NO_x INJECTION RATE [KOWALCZYK AND BAUER (1981)]

NO_x production by ground flashes	10^{26} NO_x/flash	
Global lightning frequency, cloud-to-ground	60 flashes/second	
Global lightning frequency, cloud-to-cloud and cloud-to-ground	300 flashes/second	
NO_x production, cloud-to-ground	3.8 Tg N/yr	
NO_x production, cloud-to-cloud and cloud-to-ground	5.7 Tg N/yr	
Partitioning between cloud and ground flashes	See Table 19	
Effective injection height	Cloud-to-Cloud	Cloud-to-Ground
Tropical regions between ± 30° latitude	10 to 15 km	0 to 10 km
Mid-latitude regions ±(30 to 60)°	7 to 12 km	0 to 7 km
2-D global distribution of NO_x - see Table 19		

and Bauer (1981) insofar as the latitude/altitude distribution of NO_x is concerned. Note that while there are significantly more cloud-to-cloud than cloud-to-ground discharges, more NO_x is produced by cloud-to-ground than by cloud-to-cloud discharges because these are more energetic.

TABLE 19. SUMMARY OF THE LIGHTNING INJECTION STUDY OF KOWALCZYK AND BAUER (1981)

	Latitude												Global Total
	-60 to -50	-50 to -40	-40 to -30	-30 to -20	-20 to -10	-10 to 0	0 to 10	10 to 20	20 to 30	30 to 40	40 to 50	50 to 60	
Annual average lightning rate, flashes/sec	0.1	1.2	6.9	22.3	42.2	52.3	49.1	37.7	33.2	36.1	16.7	2.1	300.0
Lightning fraction within clouds, f_c to ground, f_g	0.67 0.33	0.72 0.28	0.78 0.22	0.83 0.17	0.85 0.15	0.86 0.14	0.86 0.14	0.85 0.15	0.83 0.17	0.78 0.22	0.72 0.28	0.67 0.33	
NO_x injection rate, 10^9gN/yr/km 10 - 15/km 0 - 10/km				27.8 28.5	54.4 48.0	66.9 54.5	63.4 51.6	47.2 41.6	41.0 42.0				
7 - 12/km 0 - 7/km	0.1 0.4	1.3 3.6	7.7 15.4							4.19 84.4	16.8 49.7	2.1 7.4	
NO_x column, 10^{12}gN/yr within cloud- to-ground Total	0.01 0.00 0.01	0.01 0.03 0.04	0.04 0.11 0.14	0.14 0.28 0.42	0.27 0.48 0.75	0.33 0.54 0.87	0.31 0.52 0.83	0.23 0.42 0.65	0.21 0.42 0.63	0.21 0.59 0.80	0.08 0.35 0.43	0.01 0.05 0.06	1.9 3.8 5.7

VI. AIRCRAFT SOURCES OF NO_x

A listing of aircraft sources of NO_x for 1975 is given in Table 20 as a function of altitude and latitude. The data come from Arthur D. Little (1976), corrected as indicated by Oliver et al. (1977), p. 2-30 ff, for the fact that NO_x emissions of CF-6 engines are roughly equal to those of JT9D engines. Overall aircraft operations have been increasing very rapidly since jet transport aircraft came into widespread use in the 1960s, but the rate of increase is declining (see, e.g., Interavia, October 1981, pp. 1013-1019). International Civil Aviation Organization (ICAO) figures on passenger travel per year show a mean annual rate of increase of 9.4 percent per annum compounded, from 1960 to 1975.

Comparable NO_x injection figures projected to a 1990 "base case" are listed in Table 21. These figures are based on 1976 work and correspond to an 9 percent annual rate of increase from 1975 on.

Note that in Tables 20 and 21 no injections are listed below 6 km, because no detailed emission estimates are available for this region.

TABLE 20. 1975 WORLDWIDE AIRCRAFT NO_x EMISSIONS
Total Emissions for all Aircraft (kg NO_2/yr-km)
[Source: Arthur D. Little (1976) modified as indicated in text]

Latitude	Altitude (km)						
	6-8	8-9	9-10	10-11	11-12	12-13	13-14
60 to N Pole	5.23E 05	4.90E 05	4.15E 06	3.55E 06	3.56E 06	1.65E 04	2.94E 03
50 to 60	2.52E 06	3.42E 06	2.26E 07	2.05E 07	1.86E 07	2.29E 05	1.75E 04
40 to 50	9.99E 06	1.54E 07	4.03E 07	4.60E 07	2.63E 07	1.78E 06	2.06E 05
30 to 40	1.12E 07	1.86E 07	4.05E 07	5.55E 07	2.62E 07	2.77E 06	2.14E 05
20 to 30	3.89E 06	4.63E 06	1.46E 07	1.83E 07	1.26E 07	9.24E 05	5.64E 04
10 to 20	1.48E 06	1.67E 06	4.13E 06	5.98E 06	4.11E 06	5.03E 05	3.33E 04
0 to 10	5.48E 05	6.59E 05	2.46E 06	2.61E 06	2.03E 06	1.16E 05	9.66E 03
-10 to 0	4.82E 05	6.82E 05	2.39E 06	2.43E 06	1.78E 06	6.97E 04	2.76E 03
-20 to -10	3.47E 05	5.84E 05	2.24E 06	2.79E 06	1.84E 06	1.10E 05	1.28E 04
-30 to -20	6.01E 05	7.87E 05	1.82E 06	2.39E 06	1.30E 06	1.13E 05	1.37E 04
-40 to -30	6.29E 05	9.01E 05	1.39E 06	2.13E 06	8.42E 05	1.37E 05	6.15E 03
-50 to -40	3.30E 04	6.01E 04	7.79E 04	1.25E 05	3.49E 04	6.20E 03	3.89E 00
-60 to -50	6.08E 03	5.20E 03	3.95E 03	2.50E 03	2.82E 02	1.06E 00	0.0
-60 to S Pole	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Total of all NO_x = 4.855E 08 (kg/yr)

TABLE 21. 1990 WORLDWIDE AIRCRAFT NO_x BASE CASE, ADJUSTED[a,b] (kg NO_2/yr-km)

Latitude	Altitude (km)												
	6-8	8-9	9-10	10-11	11-12	12-13	13-14	14-15	15-16	16-17	17-18	18-19	Total
N 60+	2.11E6	1.91E6	9.03E6	8.23E6	9.20E6	8.29E5	6.31E5	2.54E5	1.07E6	1.61E6	1.31E6	8.89E5	3.707E7
50-	1.47E7	8.02E6	5.92E7	6.60E7	5.11E7	5.14E6	2.32E6	1.31E6	4.13E6	6.43E6	5.06E6	2.32E6	2.257E8
40-	4.82E7	5.51E7	1.14E8	1.76E8	1.02E8	1.57E7	2.92E6	1.30E5	2.59E6	4.33E6	3.39E6	1.47E6	5.270E8
30-	4.94E7	5.89E7	1.06E8	1.98E8	1.10E8	1.90E7	1.92E6	1.06E6	8.06E5	1.67E6	1.27E6	5.34E5	5.486E8
20-	1.63E7	1.76E7	4.22E7	6.36E7	4.33E7	5.41E6	9.51E5	7.24E5	4.95E5	1.15E6	1.03E6	2.87E5	1.930E8
10-	6.80E6	7.21E6	1.63E7	2.62E7	1.78E7	2.23E6	2.88E5	9.33E4	2.00E5	3.32E5	2.61E5	1.06E5	7.780E7
0-	2.97E6	3.17E6	9.31E6	1.12E7	8.44E6	7.69E5	1.07E5	0	1.79E5	2.51E5	2.12E5	1.00E5	3.671E7
0-	2.11E6	2.40E6	7.68E6	8.73E6	6.82E6	5.72E5	8.56E4	0	1.86E5	2.61E5	2.19E5	1.02E5	2.917E7
10-	1.74E6	2.03E6	7.21E6	9.65E6	7.26E6	7.01E5	1.95E5	8.14E4	6.78E4	1.36E5	9.73E4	4.61E4	2.921E7
20-	2.33E6	2.55E6	6.01E6	8.73E6	5.49E6	5.95E5	3.26E4	0	6.18E4	8.68E4	7.31E4	4.15E4	2.600E7
30-	2.56E6	2.97E6	4.23E6	7.54E6	3.93E6	7.77E5	5.38E4	3.08E4	9.61E3	2.78E4	1.77E4	3.63E3	2.215E7
40-	1.51E5	1.95E5	2.30E5	5.25E5	2.84E5	5.89E4	1.07E1	0	0	0	0	0	1.444E6
50-	3.06E4	2.38E4	1.87E4	1.58E4	6.73E3	8.93E2	0	0	0	0	0	0	9.652E4
S 60+	0	0	0	0	0	0	0	0	0	0	0	0	0
Total	1.494E8	1.621E8	3.814E8	5.844E8	3.656E8	5.178E7	9.504E6	4.854E6	9.795E6	1.628E7	1.294E7	5.899E6	1.755E9

Reference: Arthur D. Little (1976), p. E-16, and R.C. Oliver, private communication.

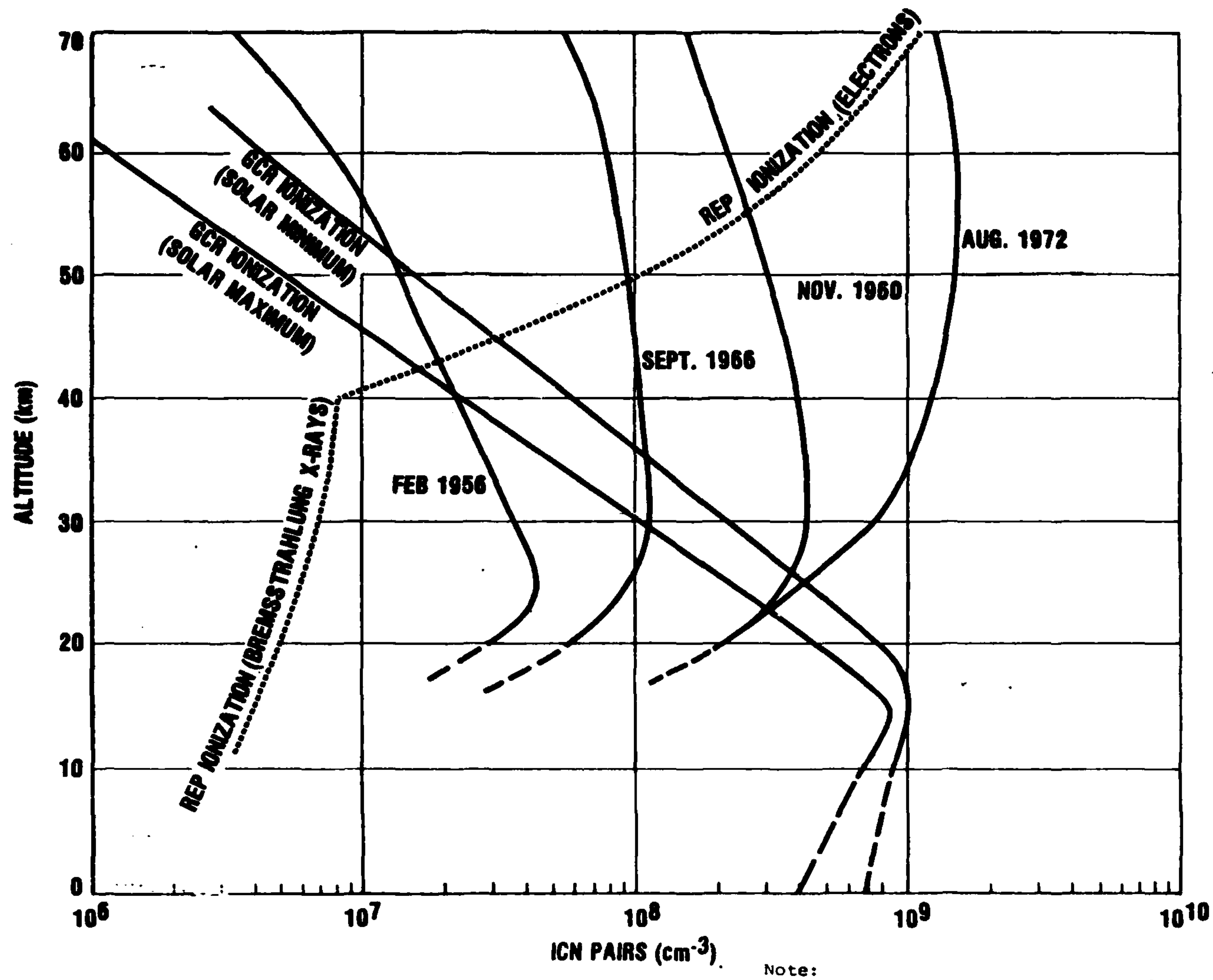
[a]To make all "CF-6" aircraft have same emission indices as "JT-9D" aircraft. See Oliver et al. (1977).

[b]To distribute SST emissions in the 15-18 km band more closely to prior estimates. See Oliver et al. (1977).

VII. NO_x PRODUCTION DUE TO COSMIC RAY IONIZATION

Cosmic rays produce a certain amount of ionization in the upper atmosphere, and as a result of the ionization-deionization chemistry each ion pair produces on the average 1.2 to 1.5 NO molecules; we use a value of 1.3 NO molecules/ion pair. Most of this ionization occurs in the stratosphere, but a certain amount of NO_x is produced in the troposphere. The total effect is not very large: here I present some simple estimates, based largely on the analysis of M. Heaps [see Bauer (1978a), especially Appendix E].

Figure 1 shows the altitude profile of ionization due to solar and galactic cosmic rays. Solar cosmic rays are associated with solar flares and are emitted in relatively infrequent bursts of relatively low energy, so that there is not very much ionization due to this source below 20 to 30 km altitude. The ionization profiles of some of the largest "solar proton events" observed between 1950 and 1975 are shown in the figure, and their tropospheric effects are evidently negligible. Galactic cosmic rays provide a steadier and more energetic source of ionizing radiation, so that the effect goes down to much lower altitudes (see Fig. 1). The effect of galactic cosmic rays depends on the solar (sunspot) cycle. During periods of high solar activity the geomagnetic field is strong and cosmic rays are deflected away from the earth, giving a minimum in ionization at sunspot maximum. Bauer (1978a), especially Appendix E there, contains a relatively detailed discussion of this whole field.



SOURCE: Bauer 1978a.

10-6-81-2

Note:
GCR = galactic cosmic rays -- of importance here.
REP = relativistic electron precipitation
Dated events are large solar proton events (SPFs).
The effects of REPs and SPEs on tropospheric NO_x are negligible.

FIGURE 1. Cosmic Ray Ionization Profiles in the Atmosphere

Table 22 lists the total number of ion pairs in the stratosphere due to galactic cosmic rays; from Fig. 1 we find the ratio of ionization in the troposphere to that in the stratosphere is 0.61 at solar (sunspot) minimum and 0.70 at solar sunspot maximum. Scaling with latitude, as in the stratosphere, we find the estimate for tropospheric NO_x injection due to galactic cosmic rays to be as shown in Table 23.

TABLE 22. NO_x PRODUCTION DUE TO GALACTIC COSMIC RAY IONIZATION IN THE STRATOSPHERE
[After Heaps, Appendix E, Bauer (1978a)]
(Units: Tg N/yr)

	In both polar caps (geomagnetic latitudes > 60°, height > 10 km)	At low geomagnetic (geomagnetic latitudes < 60°, heights > 15 km)
At Solar Minimum (1976, 1987, ...)	0.039	0.079
At Solar Maximum (1969, 1980, 1991, ...)	0.021	0.060

TABLE 23. NO_x PRODUCTION DUE TO GALACTIC COSMIC RAY IONIZATION IN THE TROPOSPHERE
[After Heaps, Appendix E, Bauer (1978a)]
(Units: Tg N/yr)

	In both polar caps (geomagnetic latitudes > 60°, uniformly distributed between 5 and 10 km altitude	At low geomagnetic (geomagnetic latitudes < 60°, uniformly distributed between 5 and 15 km altitude
At Solar Minimum (1976, 1987, ...)	0.024	0.048
At Solar Maximum (1969, 1980, 1991, ...)	0.014	0.043

VIII. DOWNWARD TRANSPORT OF ODD NITROGEN FROM THE STRATOSPHERE

A source of tropospheric odd nitrogen, which may be important for tropospheric photochemistry, is provided by downward transport from the stratosphere where NO_x is produced, mainly by reaction of N_2O with $O(^1D)$. Levy et al. (1980) have estimated a stratospheric source strength of NO_y (total odd nitrogen = $NO_x + NO_3 + 2\ N_2O_5 + HNO_2 + HNO_3 + HNO_4 + ClONO_2$ + PAN) of 0.52 to 1.0 Tg N/yr. Kley et al. (1981) estimate the ratio NO_x/NO_y = 0.05 to 0.2 at the tropopause. See also Liu et al. (1980) and Fishman (1981).

In a model calculation, Hameed et al. (1981) suggest a source strength for NO_y of 0.5 Tg N/yr, with the latitude distribution given in Table 24. Note that a marked seasonal variation may be expected [see Noxon et al. (1979)], and that the model of Hameed et al. (1981) assumes that in none of the 10^o latitude bands used is there net upward transport of NO_y due to the upwelling Hadley cell, etc., circulation. I do not dispute the estimate of Hameed et al. (1981), but the user should verify the current best estimate for this source before starting on a major calculation.

Note that there are some other high-altitude sources of NO_x in addition to the reaction of N_2O with $O(^1D)$. The most important is the stratospheric source strength due to galactic cosmic rays which is in the range 0.08 to 0.12 Tg N/yr at solar maximum/minimum, respectively. There are also some mesospheric sources of NO_x due to auroral ionization [see, e.g., Bauer (1978b)], due to meteoroid reentry heating [Park and Menees (1978)], and due to production of NO by reaction of $O(^1D)$ with

TABLE 24. LATITUDE DISTRIBUTION OF THE STRATOSPHERIC NO_x SOURCE [Source: Hameed et al. (1981)]

Latitude	Fraction of Injection
80-90°N	0.006
70-80°N	0.018
60-70°N	0.019
50-60°N	0.024
40-50°N	0.066
30-40°N	0.150
20-30°N	0.150
10-20°N	0.098
0-10°N	0.066
0-10°S	0.048
10-20°S	0.082
20-30°S	0.100
30-40°S	0.076
40-50°S	0.038
50-60°S	0.024
60-70°S	0.014
70-80°S	0.013
80-90°S	0.005
Total	0.997

N_2O resulting from electron impact due to cosmic ray events [SPE and REP, see Prasad and Zipf (1981)]. However, it is unlikely that NO_x produced above 50 km will affect the flux of NO_y through the tropopause to a significant extent.

REFERENCES

Albini, F. A., U.S. Forest Service, private communication, 24 April 1981.

Anderson, G. E. et al., "Processes Influencing the Concentration of Nitrogen Oxides in the Lower Troposphere," SAI Report EF78-31R3, 1978.

Bauer, E., "A Catalog of Perturbing Influences on Stratospheric Ozone, 1955-1975," IDA Report No. FAA-EQ-78-20, September 1978a. (See also J. Geophys. Res., 84, 6929, 1979.

Bauer, E., "Matters Arising: Non-biogenic Fixed Nitrogen in Antarctic Surface Waters," Nature, 276, 96, 1978b.

Benkovitz, C. M., "MAP3S Source Emissions Inventory Progress Report," Brookhaven National Laboratory Report BNL-51378, December, 1980.

Boettger, A., D. H. Ehhalt, and G. Gravenhorst, "Atmospheric Cycles of Nitrogen and Ammonia," Report Juel-1558, Kernforschungsanlage Juelich GmbH (ISSN 0366-0885), 2nd Edition, September 1980.

Bolin, B., "Changes of Land Biota and Their Importance for the Carbon Cycle," Science, 196, 613, 1977.

Chandler, C., Chief of Fire Research, U.S. Forest Service, private communication, 24 February 1981.

Church, C. R., J. T. Snow, and J. Dessens, "Intense Atmospheric Vortices Associated with a 1000 MW Fire," Bull. A. Meteorol. Soc., 61, 682, 1980.

Cook, J. D., J. H. Himel, and R. H. Moyer, "Impact of Forestry Burning Upon Air Quality," (GEOMET, Inc.) EPA 910/9-78-052, available through NTIS as PB-290472, October 1978.

Crutzen, P. J., "The Role of NO and NO_2 in the Chemistry of the Troposphere and Stratosphere," Ann. Rev. Earth and Planet. Science, 7, 443, 1979.

Crutzen, P. J., L. E. Heidt, J. P. Krasnec, W. H. Pollock, W. Seitler, "Biomass Burning as a Source of Atmospheric Gases $CO, H_2, N_2O, NO, CH_3Cl$ and COS," Nature, 282, 253, November 1979.

Crutzen, P. J., I.S.A. Isaksen, and G. C. Reid, "Solar Proton Events: Stratospheric Sources of Nitric Oxide," Science, 189, 457, 1975.

Darley, E. F., F. R. Burleson, E. H. Mateer, J. T. Middleton and V. Posteri, "Contribution of Burning of Agricultural Wastes to Photochemical Air Pollution," J. Air. Poll. Control Assoc., 11, 685, 1966.

Delwiche, C. C. and G. E. Likens, "Biological Response to Fossil Fuel Combustion Products," p. 73 ff in W. Stumm, Ed., Global Chemical Cycles and Their Alterations, Berlin: Dahlemkonferenzen, 1977.

EIA (Energy Information Administration) "1980 Annual Report to Congress, Volume 3, Forecasts" Report DoE/EIA-0173(80)/3, Vol. 3 of 3, March 1981 (See p. 107).

EPA (Environmental Protection Agency) "Mobile Source Emission Factors," Report EPA-400/9-78-005, March 1978.

Evans, L. F., I.A. Weeks, A. J. Eccleston and D. R. Packham, "Photochemical Ozone in Smoke from Prescribed Burning of Forests," Environ. Sci. Technology, 11, 896, 1977.

EXXON Corporation, "World Energy Outlook," December 1980.

FAO, Yearbook of Forest Products 1963-1974, Food and Agricultural Organization, Rome, 1976.

Fishman, J., "The Distribution of NO_x and the Production of Ozone: Comments on 'On the Origin of Tropospheric Ozone,'" by Liu et al. ms, NASA/LaRC, May 1981.

Foster, R. S., "Nitrogen (Ammonia)," Mineral Facts and Problems, Bureau of Mines Bulletin 671, U.S. Department of Interior, 1980.

Galbally, I. E. and C. R. Roy, "Loss of Fixed Nitrogen from Soils by NO Exhalation," Nature, 275, 734, 1978.

Gerstle, R. W. and D. A. Kemnitz, "Atmospheric Emissions from Open Burning," J. Air Poll. Control Assoc., 17, 324, 1967.

Hall, D. O., "Plants..as a Source of Renewable Resources," Nature, 278, 114 (1979).

Hameed, S., O. G. Paidoussis, and R. W. Stewart, "Implications of Natural Sources for the Latitudinal Gradients of NO_y in the Unpolluted Troposphere," Geophys. Res. Ltrs., 8, 591, 1981.

Kotaki, M., I. Kuriki, C. Katoh, and H. Sugiuchi, "Global Distribution of Thunderstorm Activity," to be submitted to J. Radio Research Labs., Japan, ms. 1981.

Kelly, T. J., D. H. Stedman, J. A. Ritter, and R. B. Harvey, "Measurements of Oxides of Nitrogen and Nitric Acid in Clean Air," J. Geophys. Res., 85, 7417, 1980.

Kley, D., J. W. Drummond, M. McFarland, and S. C. Liu, "Tropospheric Profiles of NO_x," J. Geophys. Res., 86, 3153, 1981.

Kowalczyk, M. L., and E. Bauer, "Lightning as a Source of NO_x in the Troposphere," IDA P-1590, December, 1981.

Levine, J. S., T. R. Augustsson, and J. M. Hoell, "The Vertical Distribution of Tropospheric Ammonia," Geophys. Res. Ltrs., 7, 317, 1980.

Levy, H., II, J. D. Mahlman, and W. J. Moxim, "A Stratospheric Source of Reactive Nitrogen in the Unpolluted Troposphere," Geophys. Res. Ltrs., 7, 441, 1980.

Lipschultz, F., O. C. Zafiriou, S. C. Wofsy, M. B. McElroy, F. W. Valois, S. W. Watson, "Production of NO and N_2O by Soil Nitrifying Bacteria," Nature, 294, 641, 1981.

Little, Arthur D., Inc., "Stratospheric Emissions Due to Current and Projected Aircraft Operations," C77327-10 (draft report to FAA), August 1976.

Liu, S. C., D. Kley, M. McFarland, J. D. Mahlman, and H. Levy II, "On the Origin of Tropospheric Ozone," J. Geophys. Res., 85, 7546, 1980.

Logan, J. A., M. J. Prather, S. C. Wofsy, and M. B. McElroy, "Tropospheric Chemistry: A Global Perspective," J. Geophys. Res., 86, 7210, 1981.

Malte, P. C., Washington State University Bulletin 339 (Pullman, Washington 1975), as quoted in Crutzen, et al., 1979.

NAPCA, "Control Techniques for Nitrogen Oxides From Stationary Sources," quoted from K. Wark and C. F. Warner, Air Pollution, Its Origin and Control, 2nd Edition, Report AP-67, Department of HEW, 1970, Harper and Row, New York 1981.

National Research Council, "Energy in Transition, 1985-2010," National Academy of Sciences, 1979.

National Research Council, "Nitrates: An Environmental Assessment," (see p. 276) National Academy of Sciences, 1978.

Noxon, J. F., E. C. Whipple, and R. S. Hyde, "Stratospheric NO_2, 1. Observational Method and Behavior at Mid-Latitude," J. Geophys. Res., 84, 5047, 1979.

Oliver, R. C., E. Bauer, H. Hidalgo, K. A. Gardner, W. Wasylkiwskyj, "Aircraft Emissions: Potential Effects on Ozone and Climate: A Review and Progress Report," IDA P-1207, Federal Aviation Administration Report FAA-EQ-77-3, March 1977.

Park, C., and G. P. Menees, "Odd Nitrogen Production by Meteoroids," J. Geophys. Res., 83, 4029, 1978.

Pechan, E. H., "An Air Emissions Analysis of Energy Projections for the Annual Report to Congress," Analysis Memorandum DoE;/ EIA-0102/16 (AM/IA/78-18, September 1978).

Pozdena, R., Forecasts of Aircraft Activity by Altitude, World Region and Aircraft Type, Stanford Research Institute, FAA-AVP-76-18, November 1976.

Prasad, S. S., and E. C. Zipf, "Atmospheric Nitrous Oxide Produced by Solar Protons and Relativistic Electrons," Nature, 291, 564, 1981.

Ralston, C. W., "Where has all the Carbon Gone?", Science, 204, 1345, 1979.

Robinson, E. and R. C. Robbins, Sources, Abundance, and Fate of Gaseous Atmospheric Pollutants," Stanford Research Institute, Menlo Park, California, 1968.

Seiler, W., and P. J. Crutzen, "Estimates of Gross and Net Fluxes of Carbon Between the Biosphere and the Atmosphere From Biomass Burning," Climatic Change, 2, 207, 1980.

Taylor, R. J., S. T. Evans, N. K. King, E. T. Stephens, D. R. Packham, and R. G. Vines, "Convective Activity Above a Large-Scale Bushfire," J. Appl. Meteorol., 12, 1144, 1973.

Turman, B. N., and B. C. Edgar, Global Lightning Distributions at Dawn and Dusk, Aerospace Corporation, Space Sciences Laboratory Report SSL-80(5639)-1, to appear in J. Geophys. Res., December 1980.

Westberg, H., K. Sexton, D. Flygt, "Hydrocarbon Production and Photochemical Ozone Formation in Forest Burn Plumes," J. Air Poll. Control Assoc., 31, 661, 1981.

Wong, C. S., "Atmospheric Input of Carbon Dioxide from Burning Wood," Science, 200, 197 (1978), see also comments by G. R. Fahnestock and C. S. Wong, Ibid 204, 209, 1979.

Woodwell, G. M., R. H. Whittaker, W. A. Reiners, G. E. Likens, C. C. Delwiche, and D. B. Botkin, "The Biota and the World Carbon Budget," Science, 199, 141, January 1978.

World Almanac and Book of Facts, Newspaper Enterprises Association, 1980.

Zafiriou, O. C. and M. McFarland, "NO from Nitrite Photolysis in the Central Equatorial Pacific," J. Geophys. Res., 86, 3173, 1981.

APPENDIX A

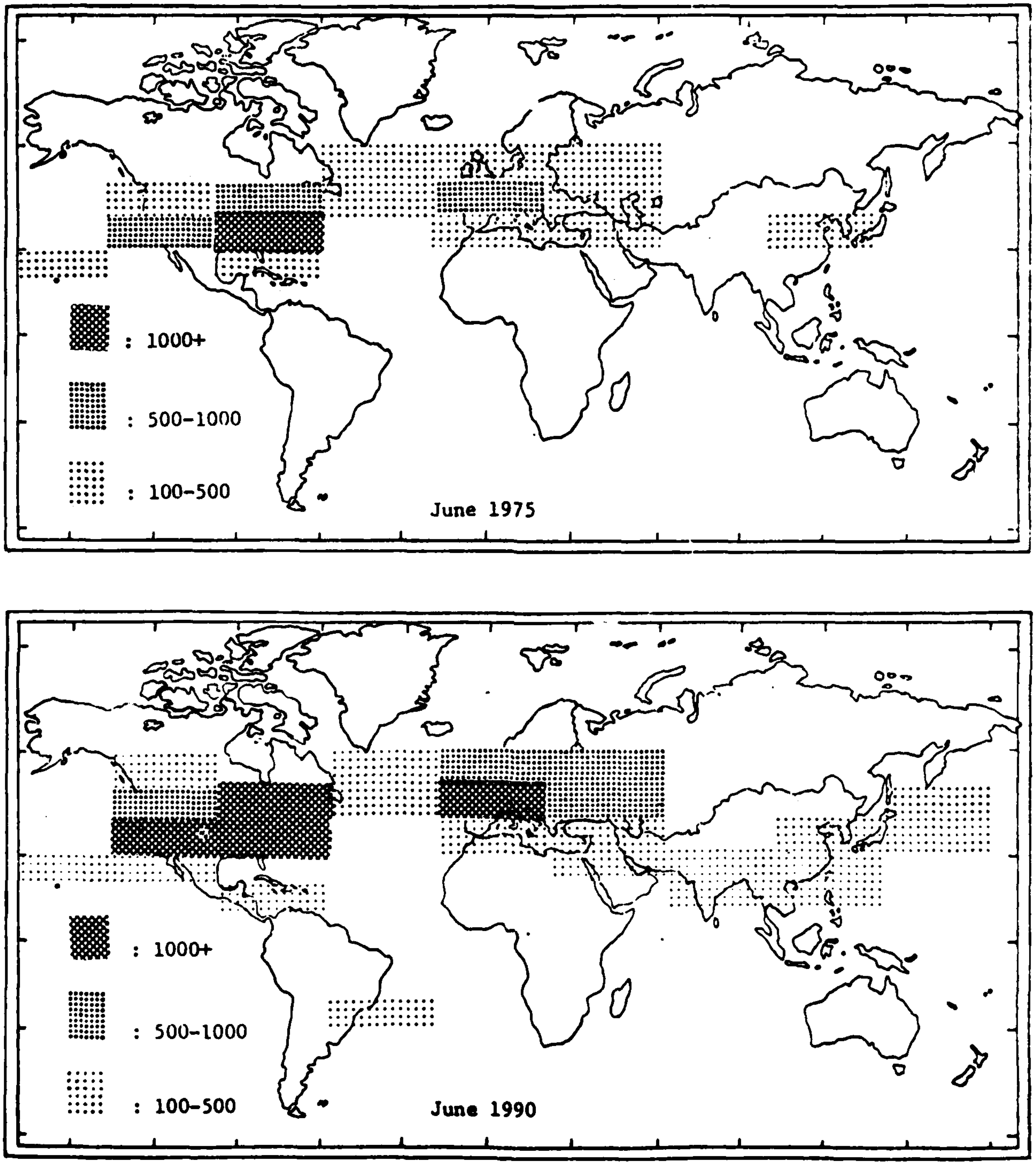
TABLE A-1. TENTATIVE LONGITUDINAL DISTRIBUTION OF NO_x INPUTS OF TABLE 1

Source	Latitude Range	Longitudinal Weighting
Aircraft[a]	50°N-60°N	Uniform over 60°E-60°W
	30°N-50°N	Uniform over 60°E-135°W
	Below 30°N	Uniform distribution
Fossil Fuel	Above 50°N	Uniform over 50°E-5°W
	40°N-50°N	0.5 uniformly distributed over 70°W-120°W 0.2 uniformly distributed over 15°E-5°W 0.15 uniformly distributed over 50°E-5°W 0.1 uniformly distributed over 100°E-120°E 0.05 uniformly distributed over 150°E-170°E
	30°N-40°N	0.6 uniformly distributed over 70°W-120°W 0.35 uniformly distributed over 100°E-120°E 0.05 uniformly distributed over 150°E-170°E
	Below 30°N [b]	0.5 uniformly distributed over 45°E-15°W 0.28 uniformly distributed over 40°W-110°W 0.22 uniformly distributed over 45°E-120°E
Biomass Burning		0.5 uniformly distributed over 45°E-15°W 0.28 uniformly distributed over 40°W-110°W 0.22 uniformly distributed over 45°E-120°E
Lightning	See Turman and Edgar (1980) or Kotaki et al. (1981).	
Transport from stratosphere		Uniform distribution
Cosmic Rays		Uniform distribution[c]
Exhalation from soils		Assume distribution uniform over land area

[a] For more details, see Fig. A-1.

[b] Perhaps 5 percent of total emissions in Australasia.

[c] Some enhancement associated with South Atlantic anomaly.



Note: The 1990 forecast in this figure is based on the Base case.

FIGURE A.1. Air Traffic Distributions, 1975 and Projected for 1990. Flight hours per day. (Pozdena, 1976)

END

FILMED

1-83

DTIC