Production of NO by Galactic Cosmic Rays and Lightning

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As part of the ongoing development of the LLNL 2-D Stratospheric Transport-Kinetics Model, values for NO production rates due to galactic cosmic rays (GCRs) and lightning have been calculated. Inclusion of these sources of NO have been prompted by discrepancies between model results and satellite observations for NO, (N, NO, NO₂, NO₃, N₂O₅, HNO₂, HO₂NO₂, HNO₃, ClNO₃) in the lower stratosphere. These discrepancies, with model predicted mixing ratios being lower than observations, are not unique to the LLNL model (Ko et al, 1986; WMO, 1985), and suggest the need for sources of NO in addition to oxidation of N₂O by O(¹D).

The likelihood of GCRs as a source stratospheric odd nitrogen was noted in a brief report by Warneck (1972). Nicolet (1975) has given detailed NO production rates from this source as a function of altitude and geomagnetic latitude. Since the rate of GCRs entering the atmosphere is negatively modulated by solar activity, which varies over an 11 year cycle, values were given by Nicolet for both minimum and maximum NO production rates. Later authors have augmented this data by providing more accurate estimates of the production yield of NO per ion pair (Frederick, 1976; Porter et al, 1976; Jackman et al, 1979). These authors have recommended increasing Nicolet's estimate of one NO molecule per ion pair to 1.2-1.3 molecules per ion pair.

The suggestion of lightning as a source of NO has a long and rich history in the literature. Only a small fraction of this history will be discussed here, but much further detail and many more references are available from the works cited. Combining results from both a theoretical shock wave model and laboratory measurements, Chameides et al (1977) have estimated the production of NO_x by lightning to be about 6×10^{16} molecules $\cdot J^{-1}$ of energy discharged. They estimate about 2×10^9 J per flash, and about 400 flashes per second globally, yielding an approximate global dissipation rate of 8×10^{11} W.

Griffing (1977) estimates an NO production efficiency factor varying between 4×10^{16} and 6×10^{10} molecules $\cdot J^{-1}$, depending on the degree of ionization produced, and an average energy of about 3×10^9 J per per flash. Given the physical uncertainties involved, these estimates are extremely close to those of Chameides (1977)

Borucki and Chameides (1984) have provided a detailed review of existing data and literature on NO production rates by lightning. By including a consideration of the reduced energy in strokes subsequent to the first in a single flash, they have concluded the total flash energy is approximately 1.75 times the energy of the first return stroke. With this inclusion, they conclude that the energy per flash is 4×10^8 J with an uncertainty factor of 2.5. Using data from satellite observations, they estimate a global flashing rate of 100 flashes $\cdot s^{-1}$, a factor of four reduction from that estimated by Chameides et al (1977). These values for the global flash rate and energy per flash result in an estimate for the global dissipation rate ranging between 1.3×10^{10} and 12×10^{10} W. They estimate an efficiency factor for NO production of about 9×10^{16} molecules $\cdot J^{-1}$. With the lowered estimate for the flashing rate, this range would include the estimate of Chameides et al

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(1977) given above. Using the values given for the global dissipation rate and chemical yield, Borucki and Chameides obtain a global nitrogen fixation rate of $2.6 \times 10^9 \text{ kg N} \cdot \text{yr}^{-1}$. They caution, however that the actual value could range between $0.8 \times 10^9 \text{ kg N} \cdot \text{yr}^{-1}$ and $8.0 \times 10^9 \text{ kg N} \cdot \text{yr}^{-1}$.

Logan (1983) has also presented a review of tropospheric NO_x budgets containing a useful discussion of various sources of data for production of NO_x by lightning and the relevant uncertainties involved.

Ko et al (1986) have investigated the effect of including lightning produced NO_x in their model. Based on data including that reviewed above, they have utilized four separate source models. Their H2 and H4 models contribute sources of $2 \times 10^9 \text{ kg N} \cdot \text{yr}^{-1}$ and $4 \times 10^9 \text{ kg N} \cdot \text{yr}^{-1}$ uniformly distributed between 30°N and 30°S latitudes and between altitudes of 4 km to 15 km. These models correspond to source strengths of $1 \times 10^3 \text{ cm}^{-1} \cdot \text{s}^{-1}$ and $2 \times 10^3 \text{ cm}^{-1} \cdot \text{s}^{-1}$ respectively. Models L2 and L4 had the same respective source strengths, but were restricted to altitudes of 4 km to 11 km. All four models are well within the range for global source rates given by Borucki and Chameides (1984). The confinement of the sources to the tropical region was based both on geographical dependence of lightning flash frequency and consideration of the region where upward transport of NO_x across the tropopause would be expected.

Jackman et al (1980) have provided an intercomparison of source strength for NO, including production by GCRs and lightning. They note that GCRs produce NO mostly below 40 km, with lightning producing NO only below the tropopause. They also note that annual production of NO by GCRs integrated over latitudes above 50° varies between $1.1\text{--}1.6 \times 10^{33}$ molecules while production in this region by N_2O oxidation is 1.4×10^{33} . They also caution, however, that because on a global scale GCR production rates are at least an order of magnitude smaller than the N_2O oxidation source, transport processes can greatly affect the relative importance of different sources.

Jackman et al (1980) have also reviewed the literature on solar proton events (SPEs) as a source of NO at latitudes above 50° . Due to their inherently short-lived, episodic, and hence unpredictable nature, this source of NO has not been included in the LLNL model. SPEs are probably not important to annual NO_x budgets except for studies of these episodic events.

Method

Values for production of NO by galactic cosmic rays (GCRs) as a function of latitude and altitude were taken from Nicolet (1975). The data as published, were on several grids giving the production of NO as a function of altitude and geomagnetic latitude. The separate grids approximately corresponded to regions of low, middle, and high altitudes. Each such grid appears to have been chosen to show the variation in NO production rates with latitude and altitude using the minimum number of points adequate for the region covered.

To make the data more convenient to utilize, all of the data were first placed on the geomagnetic latitude grid originally used for middle altitudes. This grid was chosen because, being in a region where NO production was varying the fastest as a function

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of latitude, it was a superset of the other grids used. For the middle altitude region, specifically at altitudes of 10, 12.5, 15, 17.5, 20, 22.5, 25, 27.5, 30, 35, and 40 kilometers, data were given by Nicolet (1975) at geomagnetic latitudes of 0°, 10°, 15°, 20°, 25°, 30°, 35°, 40°, 45°, 50°, 55°, and 60°. For the interpolations described below, the 60° values were treated as being valid for specific points at 60° and 90°.

Because of a lack of other low altitude data, data given for 0 km to 10 km and latitudes greater than or equal to 44° during minimum NO production and for 5 km to 10 km and latitudes greater than or equal to 45° for average production of NO were used as a basis for extrapolating the NO production values below 10 km at all latitudes. Scale factors were derived from these data, relative to the production rate at 10 km. These scale factors, which are shown in Table 1, were then applied as a single relative profile at all latitudes.

In the "high altitude grid", data were given by Nicolet (1975) for altitudes of 35.0, 40.0, 45.0, 50.0, 55.0, and 60.0 kilometers at geomagnetic latitudes of 0°, 15°, 30°, 40°, 50°, 60°, and 70°. These production rates were linearly interpolated as a function of sine-latitude to the latitude grid for mid-altitude data given above.

The above process of producing NO production rate data on a uniform geometric grid was initially done for both minimum and maximum production rate data. Following collection and interpolation to a single grid, these data were averaged and differenced to convert to tables for average production and amplitude of variation (over a solar cycle). A scaling factor of 1.27 was then applied to these data, as recommended by Porter et al (1976) and Jackman et al (1979), to account for a higher production per ion pair than was used by Nicolet (1975). The resulting data are shown in Tables 2 and 3 as functions of altitude and sine of geomagnetic latitude.

To interpolate the average production rates (Table 2) to the (altitude, sine of geographic latitude) grid used by the LLNL 2-D model, the data were averaged around geographic latitude bands using 10 increments in longitude. For each of the 36 points in the zonal band, the corresponding geomagnetic latitude was calculated, following which an area weighted interpolation was made from the data in Table 2. The resulting rates on the LLNL 2-D model grid of NO production due to GCRs are shown in Table 4.

To include the production rate of NO by lightning, we used the H2 model described by Ko et al. (1986). This model uses a uniform production rate of $1000 \text{ molecules-cm}^{-3}\text{s}^{-1}$ between 30° latitude and between 4-15 km altitude, resulting in a total source of $2 \times 10^9 \text{ kg N-yr}^{-1}$. The total source strength with this source added to the GCR production is shown in Table 5.

Comparison with LIMS data and predictions from other models

With the combined NO production rates from GCRs and lightning included in the LLNL 2-D model, we compared our predicted NO_y mixing ratios with those from LIMS (Limb Infrared Monitor of the Stratosphere) data and other models (WMO, 1985). Graphs of NO_y mixing ratios versus latitude are shown in Figure 1 for pressure altitudes of 30 mb, 16 mb, and 3 mb. Although our predicted NO_y mixing ratios are lower than the LIMS data at 16 mb and 30 mb, our values at these pressures are generally higher and in better agreement with LIMS than are the corresponding mixing ratios from the other models

shown. At 3 mb, both our results and those of Garcia and Solomon are somewhat higher than the mixing ratios from LIMS. Further research is needed on the sensitivity of these results to changes in model transport processes.

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Table 1: NO production rate scale factors below 10 km relative to the 10 km production rate.

Alt	Scale
0.0	0.0992
1.0	0.1260
2.0	0.1603
3.0	0.2023
4.0	0.2672
5.0	0.3562
6.0	0.4589
7.0	0.5719
8.0	0.7226
9.0	0.8699

Table 2: Average (over a solar cycle) production of NO by galactic cosmic rays ($\text{cm}^{-3}\text{s}^{-1}$). Given on an altitude (km) by sin(geomagnetic latitude) grid.

km/lat	.0000	.1736	.2588	.3420	.4226	.5000	.5736	.6428	.7071	.7660	.8192	.8660
.00	1.68	1.80	1.96	2.19	2.42	2.63	2.82	3.17	3.45	3.67	3.74	4.13
1.00	2.12	2.28	2.48	2.77	3.06	3.32	3.57	4.01	4.37	4.64	4.73	5.21
2.00	2.68	2.89	3.14	3.51	3.88	4.21	4.52	5.08	5.53	5.88	5.99	6.61
3.00	3.37	3.63	3.94	4.41	4.88	5.29	5.68	6.38	6.96	7.38	7.53	8.31
4.00	4.44	4.78	5.19	5.80	6.42	6.96	7.48	8.40	9.15	9.71	9.90	10.92
5.00	5.88	6.33	6.88	7.69	8.50	9.23	9.91	11.13	12.12	12.87	13.12	14.47
6.00	7.58	8.16	8.86	9.91	10.96	11.89	12.76	14.34	15.62	16.58	16.90	18.65
7.00	9.44	10.17	11.04	12.35	13.66	14.82	15.91	17.87	19.47	20.66	21.06	23.24
8.00	11.93	12.85	13.95	15.60	17.25	18.72	20.10	22.58	24.59	26.11	26.61	29.37
9.00	14.36	15.47	16.79	18.78	20.77	22.54	24.19	27.18	29.61	31.43	32.04	35.35
10.00	16.51	17.78	19.30	21.59	23.88	25.91	27.81	31.24	34.04	36.13	36.83	40.64
12.50	17.78	19.05	20.45	22.10	24.64	28.07	33.34	34.48	38.29	42.16	44.01	49.91
15.00	13.34	14.60	15.49	17.15	19.43	21.97	25.08	29.53	33.46	37.47	40.01	47.63
17.50	9.52	10.16	10.80	12.06	13.46	15.62	18.10	21.65	25.27	29.21	31.81	39.62
20.00	6.35	6.60	6.86	7.62	8.76	10.16	11.62	14.35	17.27	20.51	23.18	29.46
22.50	3.43	3.56	3.94	4.57	5.33	6.22	7.11	8.76	10.80	13.21	15.75	21.21
25.00	2.16	2.29	2.41	2.79	3.17	3.94	4.25	5.40	7.05	8.83	10.67	14.73
27.50	1.27	1.27	1.40	1.65	2.03	2.41	2.73	3.56	4.64	5.78	6.92	10.16
30.00	1.27	1.27	1.27	1.27	1.27	1.40	1.65	2.29	2.98	3.68	4.51	6.86
35.00	.39	.42	.43	.53	.62	.71	.93	1.14	1.61	2.03	2.50	2.92
40.00	.19	.21	.22	.26	.31	.36	.46	.56	.79	1.01	1.23	1.42
45.00	.10	.11	.11	.14	.16	.18	.24	.29	.42	.53	.65	.75
50.00	.05	.06	.06	.08	.09	.10	.13	.15	.22	.29	.35	.41
55.00	.03	.03	.04	.04	.05	.05	.07	.09	.12	.15	.19	.22
60.00	.01	.02	.03	.03	.03	.03	.03	.04	.06	.09	.11	.13

Table 3: Amplitude of variation (over a solar cycle) production of NO by galactic cosmic rays (cm-3s-1). Given on an altitude (km) by sin(geomagnetic latitude) grid.

km/lat	.0000	.1736	.2588	.3420	.4226	.5000	.5736	.6428	.7071	.7660	.8192	.8660
.00	.00	.00	.00	.00	.00	.00	.12	.08	.10	.32	.39	.00
1.00	.00	.00	.00	.00	.00	.00	.15	.10	.13	.40	.49	.00
2.00	.00	.00	.00	.00	.00	.00	.19	.12	.17	.51	.62	.00
3.00	.00	.00	.00	.00	.00	.00	.23	.16	.21	.64	.78	.00
4.00	.00	.00	.00	.00	.00	.00	.31	.20	.27	.84	1.02	.00
5.00	.00	.00	.00	.00	.00	.00	.41	.27	.36	1.11	1.36	.00
6.00	.00	.00	.00	.00	.00	.00	.52	.35	.47	1.43	1.75	.00
7.00	.00	.00	.00	.00	.00	.00	.65	.44	.58	1.78	2.18	.00
8.00	.00	.00	.00	.00	.00	.00	.83	.55	.73	2.25	2.75	.00
9.00	.00	.00	.00	.00	.00	.00	.99	.66	.88	2.71	3.31	.00
10.00	.00	.00	.00	.00	.00	.00	1.14	.76	1.02	3.11	3.81	.00
12.50	.00	.00	.00	.00	.00	.00	-1.59	2.35	2.48	3.43	5.27	.00
15.00	.00	.00	.00	.00	.00	.00	1.97	2.60	2.73	3.17	5.72	.00
17.50	.00	.00	.00	.00	.00	.00	1.21	1.59	2.16	3.05	4.89	.00
20.00	.00	.00	.00	.00	.00	.00	1.21	1.40	1.78	2.48	3.49	.00
22.50	.00	.00	.00	.00	.00	.00	.76	.89	1.65	2.03	2.92	.00
25.00	.00	.00	.00	.00	.00	.00	.32	.44	.83	1.33	2.03	.00
27.50	.00	.00	.00	.00	.00	.00	.32	.38	.57	.95	1.21	.00
30.00	.00	.00	.00	.00	.00	.00	.25	.25	.32	.38	.70	.00
35.00	.00	.00	.00	.00	.00	.00	.00	.00	.20	.38	.72	1.02
40.00	.00	.00	.00	.00	.00	.00	.00	.00	.09	.17	.34	.48
45.00	.00	.00	.00	.00	.00	.00	.00	.00	.05	.09	.18	.25
50.00	.00	.00	.00	.00	.00	.00	.00	.00	.02	.04	.10	.14
55.00	.00	.00	.00	.00	.00	.00	.00	.00	.01	.03	.05	.08
60.00	.00	.00	.00	.00	.00	.00	.00	.00	.01	.01	.03	.04

Table 4: Average (over a solar cycle) production of NO by galactic cosmic rays (cm-3s-1). Given on the altitude (km) by sin(latitude) grid used by the LLNL 2-D model.

km/lat	-.985	-.945	-.885	-.800	-.675	-.500	-.300	-.100
1 1.5	5.91	5.90	5.64	5.31	4.59	3.78	3.02	2.61
2 4.5	12.70	12.68	12.12	11.41	9.86	8.12	6.49	5.60
3 7.5	26.30	26.27	25.11	23.64	20.43	16.82	13.43	11.59
4 10.5	42.49	42.44	40.41	37.81	32.53	26.40	20.99	18.15
5 13.5	49.00	48.91	45.63	41.39	34.11	25.88	19.96	17.26
6 16.5	42.82	42.72	38.85	34.20	26.11	18.77	13.93	11.91
7 19.5	31.50	31.41	28.02	23.91	17.22	11.69	8.53	7.34
8 22.5	21.21	21.13	18.27	14.96	10.01	6.43	4.47	3.66
9 25.5	13.82	13.76	11.74	9.43	6.01	3.65	2.55	2.10
10 285	8.84	8.80	7.35	5.86	3.57	2.16	1.51	1.30
11 315	5.68	5.65	4.74	3.78	2.28	1.38	1.06	1.01
12 345	3.31	3.31	2.94	2.39	1.56	.88	.59	.50
13 375	2.17	2.17	1.97	1.61	1.07	.59	.39	.31
14 405	1.36	1.35	1.23	1.01	.68	.37	.24	.20
15 435	.95	.95	.87	.71	.47	.26	.17	.14
16 465	.65	.65	.59	.48	.32	.18	.11	.09
17 495	.44	.44	.40	.33	.22	.12	.08	.06
18 525	.31	.31	.28	.23	.15	.09	.06	.05
km/lat	.100	.300	.500	.675	.800	.885	.945	.985
1 1.5	2.62	3.05	3.80	4.64	5.34	5.67	5.91	5.91
2 4.5	5.62	6.56	8.17	9.97	11.48	12.19	12.69	12.70
3 7.5	11.65	13.58	16.92	20.65	23.78	25.25	26.29	26.30
4 10.5	18.23	21.21	26.63	32.86	38.05	40.65	42.47	42.49
5 13.5	17.35	20.12	26.32	34.43	41.82	45.99	48.97	49.00
6 16.5	11.98	14.05	19.00	26.52	34.70	39.28	42.79	42.82
7 19.5	7.37	8.60	11.83	17.54	24.37	28.38	31.46	31.50
8 22.5	3.67	4.53	6.50	10.20	15.35	18.56	21.18	21.21
9 25.5	2.11	2.58	3.70	6.16	9.70	11.95	13.80	13.82
10 285	1.30	1.52	2.18	3.66	6.03	7.50	8.83	8.84
11 315	1.02	1.06	1.40	2.34	3.89	4.84	5.67	5.68
12 345	.51	.60	.90	1.60	2.45	2.97	3.31	3.31
13 375	.31	.39	.61	1.10	1.65	1.98	2.17	2.17
14 405	.20	.25	.38	.69	1.03	1.24	1.35	1.36
15 435	.14	.17	.27	.48	.72	.87	.95	.95
16 465	.09	.12	.18	.33	.49	.59	.65	.65
17 495	.06	.08	.12	.22	.33	.40	.44	.44
18 525	.05	.06	.09	.16	.24	.28	.31	.31

Table 5: Average (over a solar cycle) production of NO by galactic cosmic rays plus NO production by lightning (cm-3s-1). Given on the altitude (km) by sin(latitude) grid used by the LLNL 2-D model.

km/lat	-.985	-.945	-.885	-.800	-.675	-.500	-.300	-.100
1 1.5	5.91	5.90	5.64	5.31	4.59	3.78	3.02	2.61
2 4.5	12.70	12.68	12.12	11.41	9.86	363.68	673.15	672.26
3 7.5	26.30	26.27	25.11	23.64	20.43	550.15	1013.43	1011.59
4 10.5	42.49	42.44	40.41	37.81	32.53	559.74	1020.99	1018.15
5 13.5	49.00	48.91	45.63	41.39	34.11	559.22	1019.96	1017.26
6 16.5	42.82	42.72	38.85	34.20	26.11	18.77	13.93	11.91
7 19.5	31.50	31.41	29.02	23.91	17.22	11.69	8.53	7.34
8 22.5	21.21	21.13	18.27	14.96	10.01	6.43	4.47	3.66
9 25.5	13.82	13.76	11.74	9.43	6.01	3.65	2.55	2.10
10 285	8.84	8.80	7.35	5.86	3.57	2.16	1.51	1.30
11 315	5.68	5.65	4.74	3.78	2.28	1.38	1.06	1.01
12 345	3.31	3.31	2.94	2.39	1.56	.88	.59	.50
13 375	2.17	2.17	1.97	1.61	1.07	.59	.39	.31
14 405	1.36	1.35	1.23	1.01	.68	.37	.24	.20
15 435	.95	.95	.87	.71	.47	.26	.17	.14
16 465	.65	.65	.59	.48	.32	.18	.11	.09
17 495	.44	.44	.40	.33	.22	.12	.08	.06
18 525	.31	.31	.28	.23	.15	.09	.06	.05
km/lat	.100	.300	.500	.675	.800	.885	.945	.985
1 1.5	2.62	3.05	3.80	4.64	5.34	5.67	5.91	5.91
2 4.5	672.29	673.22	363.72	9.97	11.48	12.19	12.69	12.70
3 7.5	1011.65	1013.58	550.25	20.65	23.78	25.25	26.29	26.30
4 10.5	1018.23	1021.21	559.97	32.86	38.05	40.65	42.47	42.49
5 13.5	1017.35	1020.12	559.65	34.43	41.82	45.99	48.97	49.00
6 16.5	11.98	14.05	19.00	26.52	34.70	39.28	42.79	42.82
7 19.5	7.37	8.60	11.83	17.54	24.37	28.38	31.46	31.50
8 22.5	3.67	4.53	6.50	10.20	15.35	18.56	21.18	21.21
9 25.5	2.11	2.58	3.70	6.16	9.70	11.95	13.80	13.82
10 285	1.30	1.52	2.18	3.66	6.03	7.50	8.83	8.84
11 315	1.02	1.06	1.40	2.34	3.89	4.84	5.67	5.68
12 345	.51	.60	.90	1.60	2.45	2.97	3.31	3.31
13 375	.31	.39	.61	1.10	1.65	1.98	2.17	2.17
14 405	.20	.25	.38	.69	1.03	1.24	1.35	1.36
15 435	.14	.17	.27	.48	.72	.87	.95	.95
16 465	.09	.12	.18	.33	.49	.59	.65	.65
17 495	.06	.08	.12	.22	.33	.40	.44	.44
18 525	.05	.06	.09	.16	.24	.28	.31	.31

Figure 1. Model calculated mixing ratios of total odd nitrogen, NO_y , at 30, 16, and 3 mb as compared with LIMS data and several other models (adapted from WMO, 1985).