

Production of Odd Nitrogen in the Stratosphere and Mesosphere: An Intercomparison of Source Strengths

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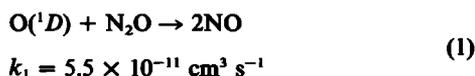
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This study intercompares several sources of odd nitrogen (ON) in the stratosphere and mesosphere (middle atmosphere) so as to place galactic cosmic rays (GCRs), nuclear explosions, lightning, solar proton events (SPEs), relativistic electron precipitation, meteors, and the downward diffusion of NO from the thermosphere in their proper context relative to the oxidation of nitrous oxide. Use of published measurements of O₃ and N₂O show that the source of ON owing to the reaction of O(¹D) + N₂O peaks between 25 and 35 km and is by far the largest source in an annually, globally averaged sense with a magnitude of 4.5×10^{34} molecules yr⁻¹. At solar minimum the GCRs add about the same amount of ON as N₂O oxidation (1.7×10^{33} molecules yr⁻¹) for geographic latitudes greater than 50°. Nuclear explosions in 1961 and 1962 added 1.1 and 2.2×10^{34} NO molecules, respectively, to the total global ON content. SPEs produced more ON at latitudes above 50° than did N₂O oxidation for the years 1958, 1959, 1960, and 1972. Analysis of available measurements shows the downward flux of NO from the thermosphere to be between 8 and 33% of the integrated N₂O source. Large variations in measurements of ON should be expected at geographic latitudes above 50° where the highly variable SPE source of ON is substantial. Because of the long lifetime of ON in the middle atmosphere, significant variations in its content may also be expected to occur at mid-latitudes.

INTRODUCTION

The odd nitrogen (including N, NO, NO₂, NO₃, N₂O₅, and HNO₃) budget of the stratosphere and mesosphere has received considerable attention owing to its relevance in the ozone balance [Crutzen, 1970, 1971; Johnston, 1971; Brasseur and Nicolet, 1973; McConnell and McElroy, 1973; Crutzen et al., 1975; Johnston et al., 1979] and in the formation of the ionospheric D region [Bates, 1952; Nicolet, 1955, 1960; Strobel et al., 1970; Strobel, 1971]. Because the major nitrogen containing gas in the atmosphere, N₂, has no optically allowed transitions connecting the ground and repulsive states it is extremely transparent to radiation. Although predissociation occurs in the thermosphere at wavelengths between 80 and 100 nm [Strobel et al., 1976], there is no photodissociation of N₂ in the middle atmosphere here defined as the stratosphere and mesosphere. Production of ON thus occurs via a variety of somewhat less obvious mechanisms.

Stratospheric ON is produced mainly from nitrous oxide (N₂O), which is produced in the biospheric nitrogen cycle and transported up from the earth's surface [Bates and Hays, 1967; Nicolet and Peetermans, 1972]. In the stratosphere, N₂O is photodissociated to produce N₂ but also reacts with O(¹D) atoms produced in photolysis of O₃ to produce NO [Nicolet, 1971; Crutzen, 1970, 1971; McElroy and McConnell, 1971].



This and all other reaction rate coefficients are taken from Hudson [1977] unless otherwise noted. Figure 1 illustrates the altitude regions where this ON source, as well as several others, are efficient. Although each ON source operates over a larger altitude range than indicated in Figure 1, only that altitude region where the source has at least 10% of its peak magnitude is included. The large and relatively constant source from nitrous oxide is used throughout this paper as a basis for

comparison of the other sources which are generally more sporadic and frequently limited in spatial extent.

These other sources include the dissociation of N₂ and ion chemistry associated with solar proton events, SPEs [Crutzen et al., 1975; Frederick, 1976], galactic cosmic rays, GCRs [Warneck, 1972; Nicolet, 1975a], and relativistic electron precipitations, REPs [Thorne, 1977]. As indicated in Figure 1, a hard spectrum SPE will influence the entire middle atmosphere, from 15 up to 85 km. The GCRs produce ON mostly below 40 km while REPs are influential as an ON source above ~60 km. Nuclear explosions, affecting the ~7-30 km regime, cause sufficiently high temperatures that thermal decomposition of N₂ and O₂ occurs followed by the formation of NO [Johnston et al., 1973; Foley and Ruderman, 1973; Gilmore, 1975]. Lightning strokes produce ON in a similar manner [Griffing, 1977; Chameides et al., 1977], however, they form odd nitrogen only below the tropopause where it is efficiently removed by rainfall. Ablating meteoroids are a constant source of ON through high-temperature reactions of air in their continuum wake [Park and Menees, 1978] but their major ON contribution is above 80 km. Thermospheric ion chemistry is an efficient source of nitric oxide and leads to a downward flux at the mesopause [Strobel et al., 1970; Strobel, 1971; Rusch, 1973]. This has important mesospheric consequences but is efficiently screened from the stratosphere by NO photolysis.

This work intercompares several of the ON sources in the middle atmosphere with the goal of placing the SPEs, GCRs, REPs, nuclear explosions, lightning, meteors, and downward flux from the thermosphere in their proper context in relation to the ambient source associated with N₂O. In doing this we use available atmospheric measurements whenever possible. The SPEs are of particular interest since much study has been devoted to the unusually large event of August 1972, but little has been done toward examining the relevance of this ON production over long time periods. To provide a realistic assessment of this source, we estimate the total ON produced by SPEs per year for the period 1954-1979.

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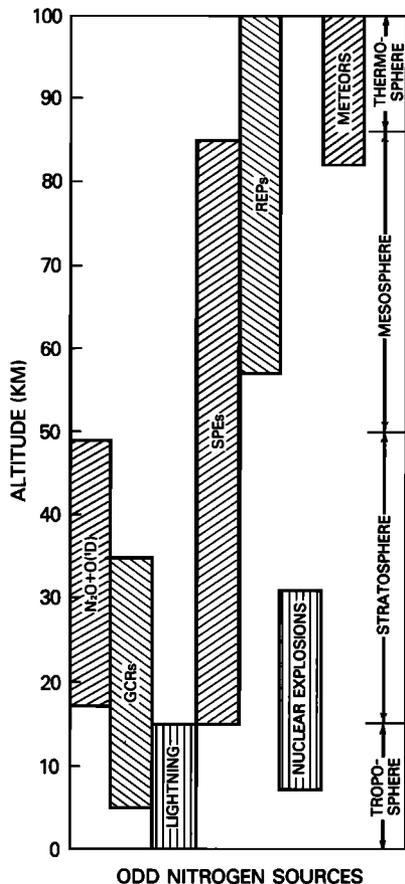


Fig. 1. Altitude region of influence from several odd nitrogen sources. These sources include the oxidation of nitrous oxide ($N_2O + O(^1D)$), galactic cosmic rays (GCRs), lightning, meteors, solar proton events (SPEs), nuclear explosions, and relativistic electron precipitations (REPs).

ODD NITROGEN SOURCE FROM N_2O OXIDATION

Derivation of the odd nitrogen source from N_2O oxidation requires a knowledge of N_2O and $O(^1D)$ as a function of season, latitude, and altitude. To do this we use the method of instantaneous rates described by Johnston and Whitten [1975] which is based on empirically determined N_2O , and we compute $O(^1D)$ from measured solar fluxes, cross sections, quantum yields, and ozone profiles. Altitude dependent profiles of N_2O at several latitudes have been measured by Fabian *et al.* [1979a] and by Schmeltekopf *et al.* [1977] and Goldan *et al.* [1980]. The magnitude of the N_2O densities given in these three papers agree fairly well in the 40 to 45° latitude range. Since the data of Schmeltekopf *et al.* [1977] and Goldan *et al.* [1980] are self-consistent, we have used these papers to derive the N_2O profiles.

Seasonal data are only available at a few middle latitudes, while complete latitudinal coverage is available only in the fall. We thus had to make several assumptions to extrapolate the available data base. The following are the assumptions:

1. 0°–10° latitude; use the fall data at 5° of Goldan *et al.* [1980] and assume no seasonal change.
2. 10°–30° latitude; use the spring data at 9° of Goldan *et al.* [1980] and Schmeltekopf *et al.* [1977] and assume no seasonal change.
3. 30°–50° latitude; use seasonal data from all three sources quoted above. Winter and spring profiles were so similar that they were assumed identical. Summer and fall pro-

files were similar and both larger than the winter-spring profile.

4. 50°–70° latitude; use spring data at 63° and summer data at 51° from Schmeltekopf *et al.* [1977] and assume winter the same as spring and fall the same as summer.

5. 70°–90° latitude; use summer data of Goldan *et al.* [1980] at 78° and 90° and assume no seasonal change.

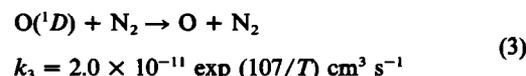
6. Above altitude of highest data assume the N_2O mixing ratio decreases with a constant scale height. Although details of the assumed extrapolation are important to eddy coefficient determination [see Johnston *et al.*, 1979], they have little effect on column integrated odd nitrogen production.

Johnston *et al.* [1979] have also recently calculated instantaneous NO production rates from N_2O oxidation. They established N_2O profiles as a function of latitude and season which are similar to ours up to an altitude of 35 km except at 60° latitude. We used the data taken at Saskatchewan (51°N) in August 1975 [Schmeltekopf *et al.*, 1977], which Johnston *et al.* [1979] rejected as inconsistent to establish the 60° N_2O profile. Our N_2O mixing-ratio contours thus differ qualitatively with those of Johnston *et al.* [1979] by having a rather sharp dip around 60° latitude. It is not clear which is correct but for the purpose of globally integrated source comparisons in this paper the differences are insignificant.

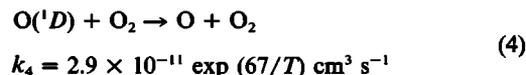
The other crucial factor in obtaining odd-nitrogen production rates is the $O(^1D)$ concentration. We used a calculation involving the multiple scattering of sunlight [Herman, 1979], diurnal averaging (Rundel [1977] for latitudes below 60°, and J. R. Herman, private communication, 1979, for latitudes at and above 60°), solar irradiance [Nicolet, 1975b], O_3 seasonal profiles [Dütsch, 1974; London *et al.*, 1977], O_3 absorption cross sections [Ackerman, 1971], quantum yields from Nicolet [1975b] for the photolysis reaction



and the $O(^1D)$ quenching reactions



and



to derive the $O(^1D)$ profiles.

Using the N_2O and $O(^1D)$ profiles, we derive diurnally averaged NO production rate profiles for the four seasons at the five latitudes of 0°, 20°, 40°, 60°, and 80°. Figures 2 and 3 present the final results. Peak production in spring-fall is at 30 km and latitude 0° with a rate of 370 NO molecules $\text{cm}^{-3} \text{ s}^{-1}$. In the winter-spring results, production ceases poleward of 60°, and a peak of 325 NO molecules $\text{cm}^{-3} \text{ s}^{-1}$ occurs at latitude 0°.

There is a large uncertainty associated with the results, especially at 60° and above due to the necessity of using an incomplete data set to establish a two-dimensional production rate distribution. Clearly, more experiments should be undertaken in the high and low latitude regions to provide better seasonal N_2O profile information.

ON SOURCE FROM PARTICLE PRECIPITATION

Both solar and galactic cosmic rays deposit much of their energy in the middle atmosphere. The more energetic galactic cosmic rays (GCRs) penetrate lower and deposit most of their

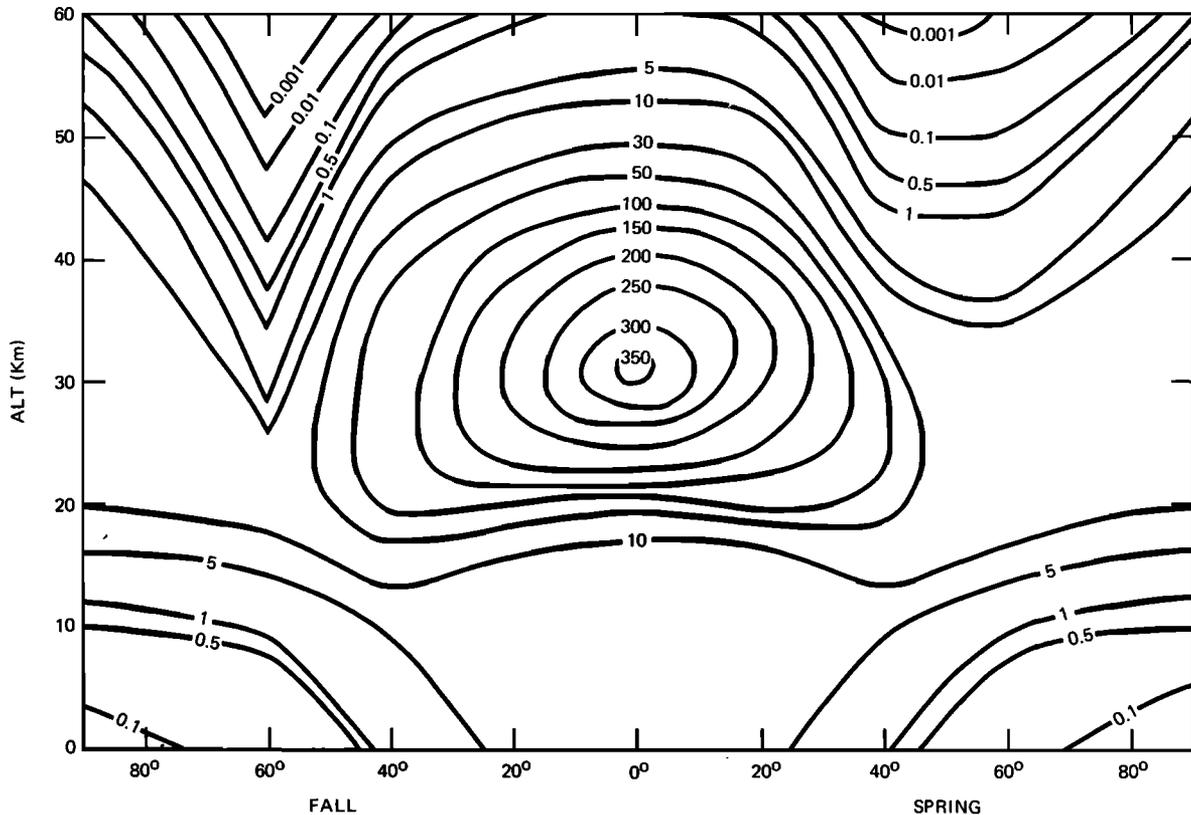


Fig. 2. Instantaneous nitric oxide production rate contours ($\text{cm}^{-3} \text{s}^{-1}$) from the oxidation of nitrous oxide for fall and spring.

energy below about 30 km. The solar particles consist primarily of protons in the MeV region which occur sporadically and deposit most of their energy between 20 and 100 km. These sporadic events are frequently referred to as solar-proton events (SPEs) or as polar cap absorptions (PCAs). Nicolet [1975a] calculated the amount of ON produced by GCRs in the stratosphere and mesosphere. He assumed that one NO molecule was produced per ion pair. Several papers [Porter *et al.*, 1976; Frederick, 1976; Jackman *et al.*, 1979] have since recommended 1.2–1.3 NO molecules per ion pair as more likely, and we have therefore used Nicolet's [1975a] production rates multiplied by 1.25.

The effects of individual solar proton events (SPEs) on the atmosphere have been studied in detail [Reid, 1961; Weeks *et al.*, 1972; Swider and Keneshea, 1973; Crutzen *et al.*, 1975; Frederick, 1976; Heath *et al.*, 1977; Reagan *et al.*, 1978; Fabian *et al.*, 1979b]. Both the total energy flux and the hardness of the spectrum are important in determining the effects of SPEs on the atmospheric ON content. High-energy protons ($E \geq 30$ MeV) penetrate into the stratosphere while lower energies influence only the mesosphere. The lifetime of ON below the stratopause is significantly longer than at higher altitudes where dissociation of nitric oxide followed by recombination is efficient. Hence, for a given total energy input, the ON column abundance will show a greater increase for a harder energy spectrum. The appendix describes the information available and how we used that data to determine ON production rates from all of the known SPEs from 1954 through 1979.

We have used the information in the appendix to compute the instantaneous maximum ON production rate for three separate events: July 12, 1961 ($0.06 \text{ ergs cm}^{-2} \text{ s}^{-1}$); February 25, 1969 ($0.04 \text{ ergs cm}^{-2} \text{ s}^{-1}$); and August 4, 1972 (7 ergs cm^{-2}

s^{-1}). The duration of production rates of this magnitude varies considerably and could be a few hours to a couple of days. Figure 4 compares these instantaneous rates with the GCR source above 60° geomagnetic latitude at solar maximum and minimum and also with the production from N_2O at both the 60° and 80° geographic latitudes. The production of NO from N_2O , as computed by Johnston *et al.* [1979] at 60° summer, is included as well. The SPE production rate falls off rapidly below 60° geomagnetic latitude due to the geomagnetic cutoff, so that SPEs directly affect only the area of the earth above about 50° geographic latitude. The July 12, 1961, event illustrates a moderate SPE with a relatively soft energy spectrum (i.e., a predominance of low-energy protons). Its energy is deposited mainly in the mesosphere. The February 25, 1969, event is also a moderate one but with a harder spectrum of proton energies. It thus penetrates to stratospheric altitudes where the loss processes for ON are quite slow. The production rates from this very small event are seen to be comparable to the $\text{O}(^1D) + \text{N}_2\text{O}$ source of ON at high latitudes for the duration of the event. Events of this magnitude can make a significant contribution to the annual high-latitude ON budget. The extreme case is the August 4, 1972, SPE which is also shown in Figure 4 and has been discussed in some detail by Crutzen *et al.* [1975], Heath *et al.* [1977], and Reagan *et al.* [1978].

We have summed the contribution of the individual SPEs in annual intervals for the years from 1954 through 1979 by a method described in the appendix. Since these SPEs directly influence only high latitudes, the total annual production rates of ON from SPEs, GCRs, and N_2O in the stratosphere and mesosphere have been integrated over latitudes above 50° and are shown in Figure 5. As can be seen, the contributions due

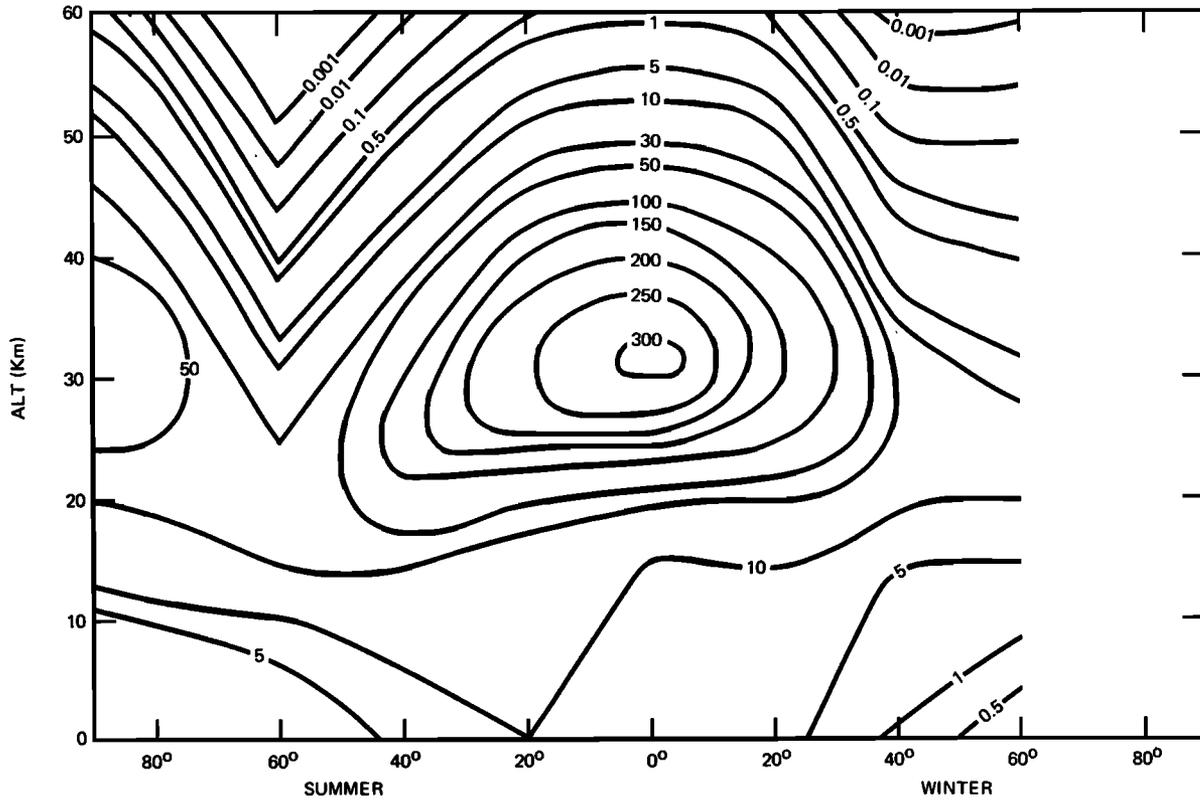


Fig. 3. Instantaneous nitric oxide production rate contours ($\text{cm}^{-3} \text{s}^{-1}$) from the oxidation of nitrous oxide for summer and winter.

to GCRs and N_2O oxidation are comparable and relatively constant, the GCR source varying from 1.6×10^{33} NO molecules yr^{-1} at solar minimum down to 1.1×10^{33} NO molecules yr^{-1} at solar maximum with the N_2O oxidation source at 1.4×10^{33} NO molecules yr^{-1} . The SPE contribution is highly variable, being entirely negligible in some years while being the major source above 50° latitude in other years.

Also shown in Figure 5 are the sources due to N_2O oxidation in the mesosphere and the source due to SPEs in the stratosphere. It is clear that in most years the SPE source of ON will dominate both the mesospheric NO production due to N_2O oxidation, 7.6×10^{30} NO molecules yr^{-1} , and the GCR source of NO which is concentrated mainly below about 40 km. On the average the SPE source of NO in the stratosphere

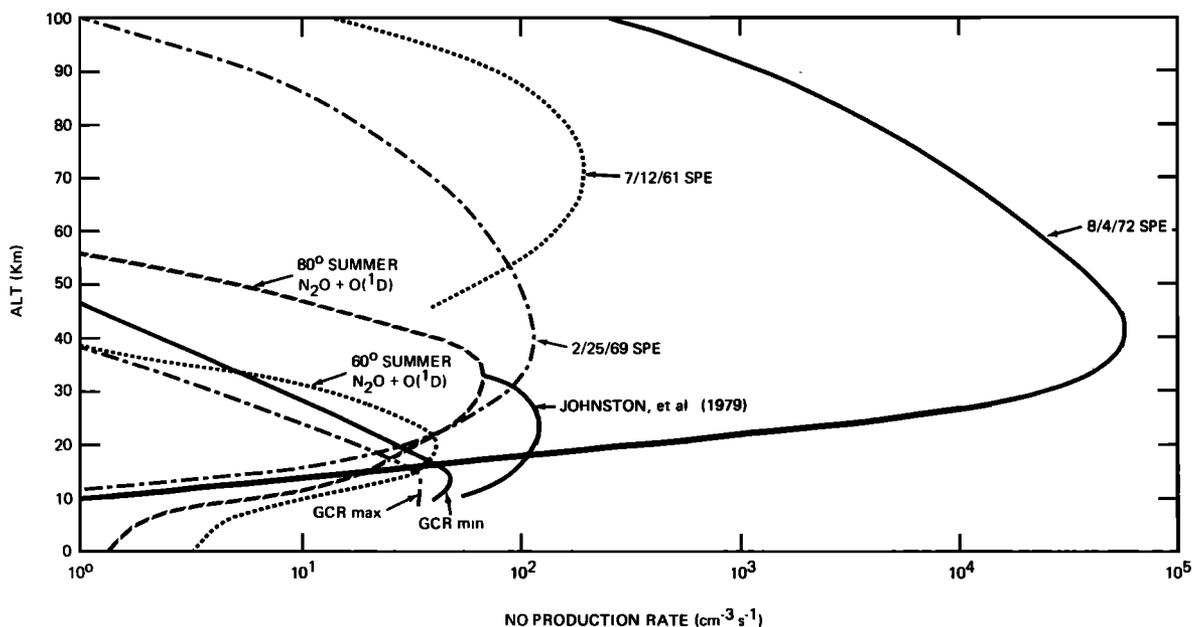


Fig. 4. Instantaneous nitric oxide production rates for GCRs (at solar maximum and minimum), SPEs (July 12, 1961, February 25, 1969, August 4, 1972), and oxidation of nitrous oxide (60° summer, 80° summer and *Johnston et al.* [1979] at 60° summer).

is roughly 20% of the total SPE annual NO source, but this contribution can vary from 6 to 60%. This stratospheric source in most years is smaller than that due to both N₂O and GCRs, these sources being primarily stratospheric.

Several caveats should be given concerning the N₂O oxidation source. If the 60° latitude measurements of *Schmeltekopf et al.* [1977] are unrepresentative then that source should be somewhat larger than shown in Figure 5. A rough calculation using *Johnston et al.* [1979] gives a N₂O source about twice our N₂O source plotted in Figure 5. Also, the N₂O source is shown as a constant throughout the 1954–1979 period. The N₂O source flux has probably been changing throughout this period [e.g., *Sze and Rice*, 1976; *Liu et al.*, 1976; *Crutzen*, 1976; *McElroy et al.*, 1976; *Johnston*, 1977; *CAST*, 1977; *Liu et al.*, 1977], but because of the expected long lifetime of N₂O (~100 yrs) these changes should not yet have appeared in the N₂O concentration and hence the ON production rate. The N₂O source may also show a solar cycle variation if upper stratospheric ozone has such a variation. Whether or not ozone varies significantly with solar activity is still an open question [e.g., *London and Reber*, 1979; *Angell and Korshover*, 1976;

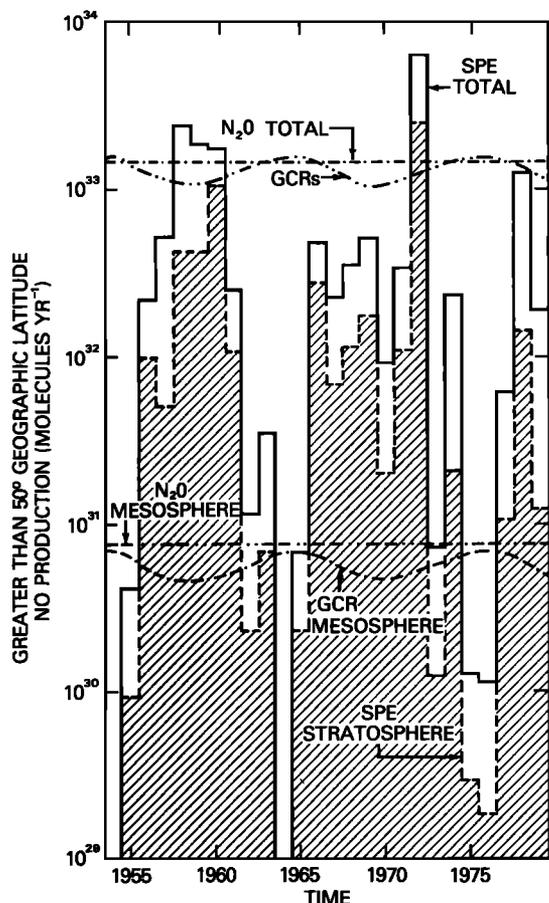


Fig. 5. Annual nitric oxide production rates for geographic latitudes above 50°. The solid line and dashed line histograms represent the solar proton event source in the stratosphere and mesosphere together and in the stratosphere alone, respectively. The dash-dot and double dash-double dot straight lines represent the oxidation of nitrous oxide source in the stratosphere and mesosphere together and in the mesosphere alone, respectively, while the dash-double dot and double dash-dot sinusoidal lines exhibit the galactic cosmic ray source in the stratosphere and mesosphere together and in the mesosphere alone, respectively.

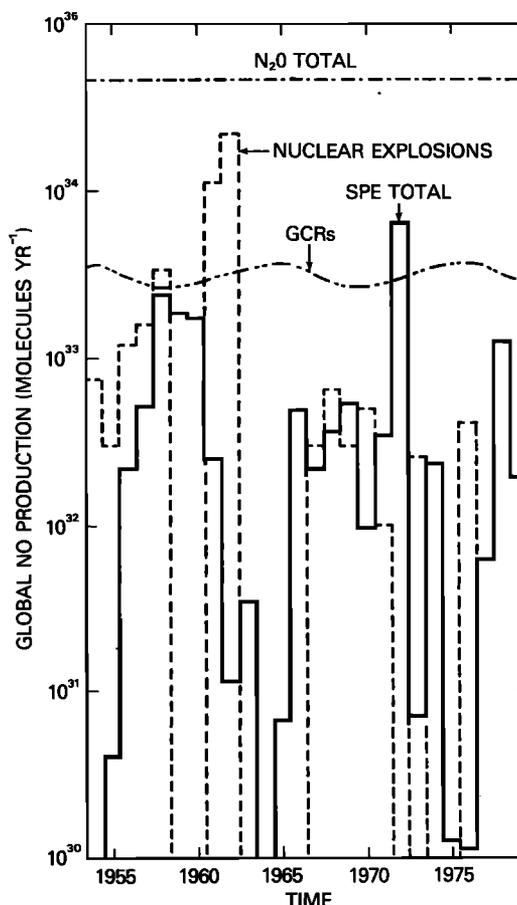


Fig. 6. Global annual stratosphere and mesosphere nitric oxide production rates. The solid lines and dashed line histograms represent the solar proton event and nuclear explosion sources, respectively. The dash-dot straight line and dash-double dot sinusoidal line exhibit the oxidation of nitrous oxide and the galactic cosmic ray sources, respectively.

Paetzold, 1973]. Thus for the purposes of the present comparison a constant N₂O oxidation source should suffice.

Horizontal transport can have a significant effect on ON due to its long lifetime in the stratosphere. This transport would redistribute the ON from SPEs formed in the polar cap regions down to the mid-latitudes [*Frederick*, 1976]. For this reason, Figure 6 shows a comparison of the ON sources on a global annual scale. In this context the ON sources from SPEs and GCRs are at least an order of magnitude smaller than the N₂O oxidation source.

Thorne [1980] has estimated the production rate of ON in the mesosphere due to REPs to be 1.4×10^{33} to 1.4×10^{34} molecules yr⁻¹. While the upper limit of this rate is higher than all other ON sources above 50° latitude, 99.8% of this ON production occurs above 50 km. If one accepts these numbers, the REPs are a significant, if not the dominant, ON source in the mesosphere.

OTHER ON SOURCES

Bauer [1979] has estimated the magnitude of stratospheric injections of ON due to past atmospheric nuclear explosions. The ON injection rate per year was calculated from these data and Figure 6 compares this with the global annual SPE, GCR, and N₂O source of ON. The stratospheric and mesospheric N₂O oxidation source of 4.5×10^{34} NO molecules yr⁻¹ is by

TABLE 1. Listing and Comparison of ON Sources in the Stratosphere and Mesosphere

Source	Area	Description	Strato- spheric (S) Source in ON Molecules, yr ⁻¹	Meso- spheric (M) Source in ON Molecules, yr ⁻¹	S and M Source in ON Molecules, yr ⁻¹
N ₂ O + O(¹ D)	Global	average	4.5 + 34	5.3 + 32	4.5 + 34
N ₂ O + O(¹ D)	>50°	average	1.4 + 33	7.6 + 30	1.4 + 33
GCRs	Global	solar maximum	2.7 + 33	1.2 + 31	2.7 + 33
GCRs	Global	solar minimum	3.7 + 33	1.6 + 31	3.7 + 33
GCRs	>50°	solar maximum	1.1 + 33	4.7 + 30	1.1 + 33
GCRs	>50°	solar minimum	1.6 + 33	7.0 + 30	1.6 + 33
REPs	Global	maximum	2.7 + 31	1.4 + 34	1.4 + 34
	>50°				
REPs	Global	minimum	2.7 + 30	1.4 + 33	1.4 + 33
	>50°				
Down flux from thermo- sphere	Global	maximum		1.5 + 34	1.5 + 34
Down flux from thermo- sphere	Global	minimum		3.7 + 33	3.7 + 33
Meteors	Global	average		6.3 + 32	6.3 + 32
Lightning	Global	average			
Nuclear explosions	Global	maximum 1962	2.2 + 34		2.2 + 34
SPEs	Global	maximum 1972	2.5 + 33	3.9 + 33	6.4 + 33
	>50°				

far the largest. The total atmospheric N₂O oxidation source of 4.7×10^{34} NO molecules yr⁻¹ compares quite favorably with the 4.5×10^{34} NO molecules yr⁻¹ calculated by *Johnston et al.* [1979]. The other ON sources are at least 1 order of magnitude lower except in the years 1961 and 1962 when a large number of nuclear tests were conducted and 1.1 and 2.2×10^{34} NO molecules were added to the total global ON content.

Lightning is another source of ON, however; this production occurs in the troposphere where ON production is least efficient in affecting the middle atmosphere ON budget because of down drafts and rainout/washout processes. Thus this production rate of 1.6×10^{36} ON molecules yr⁻¹ [*Chameides et al.*, 1977], which could in principal dominate the total annual production rate, adds little ON to the middle atmosphere. Lightning may produce N₂O [*Griffing*, 1977; *Levine et al.*, 1979], and thereby influence the ambient chemical source of NO.

The source of ON produced in the wake of large meteors could dominate all others in the middle atmosphere. *Park* [1978] estimated the production due to the Tunguska meteor of 1908 at 3.8×10^{35} molecules of NO between 10 and 50 km. In ordinary years, however, most of this production occurs above the mesopause with only about 6.3×10^{32} molecules being produced annually in the mesosphere. The N and NO produced above 86 km (7.7×10^{33} molecules yr⁻¹) could add to the downward flux of NO from the thermosphere.

NET NO FLUX INTO THE MESOSPHERE

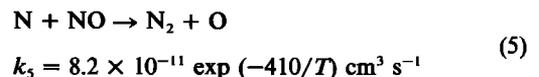
Past theoretical studies indicate that downward transport of thermospheric nitric oxide provides a significant source of ON for the mesosphere. Values of the downward flux at the mesopause, based on model calculations, range from 1.5×10^7 cm⁻² s⁻¹ [*McConnell and McElroy*, 1973] to a maximum of 5.0×10^8 cm⁻² s⁻¹ [*Strobel*, 1971]. If these values represent global averages, then the NO supplied to the mesosphere lies between 2.4×10^{33} and 8.0×10^{34} molecules yr⁻¹. Figure 6 indicates that fluxes near the upper limit would constitute the major source of middle atmospheric ON. In any case, above

45–50 km the ON source associated with nitrous oxide is small so that transport from the thermosphere likely supplies much of the ambient nitric oxide in the mesosphere. Note, however, that production of odd nitrogen in SPEs and REPs can be a locally significant, although sporadic, source at high latitudes.

Available measurements of nitric oxide allow an estimate of the downward flux from the thermosphere as follows. Over the altitude range 50–86 km, the integrated chemical loss of ON is much greater than the production [*Strobel*, 1971; *Brasseur and Nicolet*, 1973]. Thus to an acceptable approximation the net flux into the region is balanced by in situ loss:

$$-\phi_{\text{ON}} = -\phi(z = 86) + \phi(z = 50) = \int_{z=50}^{86} dz' L$$

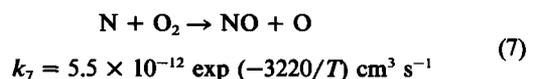
where ϕ_{ON} is the NO flux measured positive upwards and L is the loss rate due to



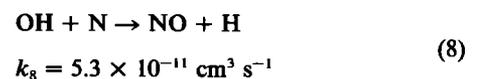
The only source of nitrogen atoms in the mesosphere, aside from sporadic energetic particles and meteors, is the predissociation of NO



Some of the N atoms produced in (6) reform NO in



and



The reaction rate coefficient k_8 is taken from *Hampson and Garvin* [1978]. The reaction of atomic nitrogen with ozone is now thought to proceed so slowly as to be negligible (L. J.

Stief, personal communication, 1979). The ON loss rate is then

$$L = 2k_5 [\text{NO}][\text{N}] = \frac{2k_5 J_6 [\text{NO}]^2}{k_5[\text{NO}] + k_7[\text{O}_2] + k_8[\text{OH}]}$$

The O₂ model used in the calculation is derived from the *U.S. Standard Atmosphere* [1976] with the OH profile being taken from *Herman* [1979]. The NO dissociation rate, *J*₆, is that of *Frederick and Hudson* [1979] averaged over a diurnal cycle at 40°N. Separate values of the loss rate and net flux were computed for summer and winter.

Two nitric oxide models have been constructed based on the few available measurements so as to span the likely range of variability. Model I represents a small NO content and is based on the data of *Meira* [1971] above 80 km, where a factor of 2 correction required for the updated gamma band emission rate factor is included [*Oran et al.*, 1975]. Below 60 km the profile is based on the data of *Mason and Horvath* [1977] and *Horvath and Mason* [1978]. Model I predicts a small upward flux at the stratopause with the NO mixing ratio decreasing from ~8 × 10⁻⁹ by volume at 50 km to ~7 × 10⁻⁹ at 65 km. The steep mixing ratio increase above 80 km provides a substantial downward flux here. Model II, representing a large NO content, is based on the *Meira* [1971] profile above 80 km and has a constant mixing ratio of 2 × 10⁻⁸ to the stratopause. In this case a large flux flows from the thermosphere to the mesosphere but the flux at the stratopause is zero.

Use of Models I and II each with the values of the NO dissociation rates for summer and winter produce the following. For summer

Model I
 $-\phi_{\text{ON}} = 3.6 \times 10^7 \text{ cm}^{-2} \text{ s}^{-1}$

Model II
 $-\phi_{\text{ON}} = 9.4 \times 10^7 \text{ cm}^{-2} \text{ s}^{-1}$

and for winter

Model I
 $-\phi_{\text{ON}} = 2.3 \times 10^7 \text{ cm}^{-2} \text{ s}^{-1}$

Model II
 $-\phi_{\text{ON}} = 5.6 \times 10^7 \text{ cm}^{-2} \text{ s}^{-1}$

Inclusion of (8) in our calculations results in about a 10%–15% decrease in the NO flux required from the thermosphere. In all cases the results based on the NO models are bracketed by past theoretical predictions. If we adopt the Model I winter and Model II summer results as the extremes of a globally averaged downward flux at 85 km, then the middle atmosphere receives between 3.7 × 10³³ and 1.5 × 10³⁴ NO molecules yr⁻¹ from the thermosphere. These values are between 8% and 33% of the source associated with N₂O and are larger than the annual production due to GCRs and, in most years, SPEs. We note that the enhanced nitric oxide content of the lower thermospheric auroral zones [*Rusch and Barth*, 1975] may lead to enhanced fluxes at high latitudes which cannot be estimated with the steady state, globally averaged approach used above.

CONCLUSIONS

A comparison of the annual production rates from several ON sources appears in Table 1. Where appropriate, the average annual production rate is given. When there is a range of ON production between solar maximum and minimum, the

extreme values are listed. For REPs the values shown represent the uncertainty range given by *Thorne* [1980]. The range for the source of ON from the thermosphere was calculated in this work. The results for nuclear explosions and SPEs are for the years of largest annual production since 1954. For SPEs the minimum production is insignificant but greater than zero. In a global sense the oxidation of nitrous oxide exceeds any other stratospheric ON source by 1 order of magnitude or more. However, this process is much less efficient in the mesosphere. Only 1.2% of the global production of ON due to N₂O oxidation occurs above 50 km and, under this circumstance, the downward flux from the thermosphere, REPs, and SPEs become the major mesospheric sources of ON.

In select years, the stratospheric production of ON due to SPEs can be comparable to that of N₂O oxidation above latitude 50°. Hence, high latitude ON likely shows a substantial year-to-year variability related to solar activity. Since ON has a long lifetime in the stratosphere, this variability should also appear at middle latitudes due to the action of horizontal transport. In the mesosphere the three largest sources of ON are linked to solar activity. The downward flux of nitric oxide at the mesopause should reflect variations in the thermosphere which are known to occur [*Rusch and Barth*, 1975] while the occurrences of REPs and SPEs are sporadic. The temporal behavior of these mesospheric sources suggests a large global scale variability in the ON abundance above 50 km both in terms of the yearly average and on time scales comparable to the chemical lifetime.

APPENDIX: TREATMENT OF SPEs

In this appendix we express the spectrum of all SPEs in the same analytic form and quantify the ON injection rate from SPEs.

The energy loss *E*_L by an isotropic flux Φ(*E*) in cm⁻² sr⁻¹ MeV⁻¹ of protons with kinetic energy *E* in the *i*th slab of a plane parallel atmosphere is

$$E_{L_i} = \int_0^\infty \Phi(E) E_{L_i}(E) dE \quad \text{MeV cm}^{-2} \text{ s}^{-1} \quad (\text{A1})$$

where *E*_L(*E*) is the energy loss by a monoenergetic flux of protons of energy *E*. This integral equation requires the use of the well-known range energy relation [*Whaling*, 1958; *Sternheimer*, 1959; *Green and Peterson*, 1968]. For this work the range data of *Sternheimer* [1959] were fit by using

$$R(E) = A \left(\frac{E}{1 \text{ MeV}} \right)^B \quad \text{gm cm}^{-2} \quad (\text{A2})$$

with *A* = 2.71 × 10⁻³ and *B* = 1.72 for energies from 1 up to 1550 MeV and with *A* = 0.834 and *B* = 0.94 for energies greater than 1550 MeV. These fits are good to within 16% at energies up to 10⁵ MeV and to within 5% at most energies. We assume isotropically directed incident particle fluxes. The energy range from 1–10⁴ MeV is divided into 60 logarithmic intervals while the pitch angle range 0–π/2 radians is split into 35 equal intervals.

The atmosphere from 0–110 km was divided into slabs of 2 km thickness. The energy loss *E*_L(θ, *E*) in the *i*th slab from a proton with pitch angle θ and kinetic energy *E* could then be calculated from

$$E_{L_i}(\theta, E) = E - \left\{ -\frac{\Delta Z_i}{A} \sec \theta + E^B \right\}^{1/B} \text{ MeV} \quad (\text{A3})$$

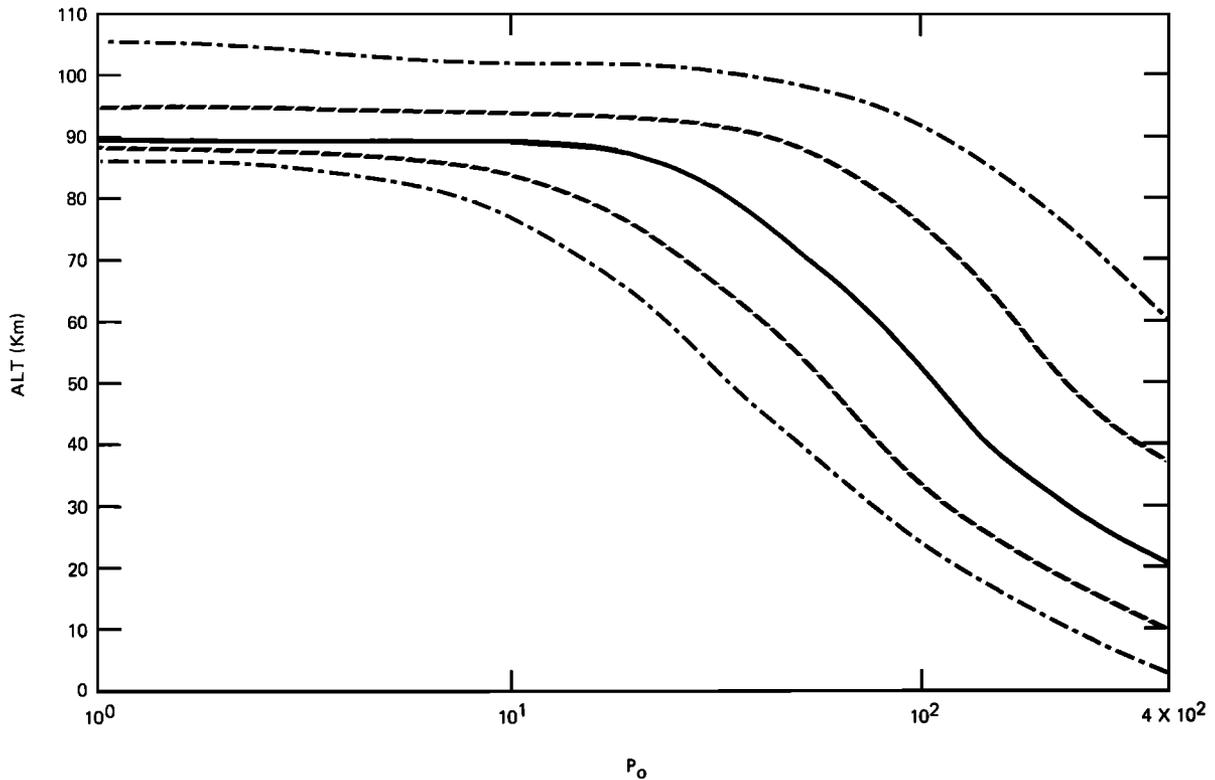


Fig. A1. The maximum intensity, J_p , and e folding rigidity, P_0 , for SPEs in the solar cycle from 1956 to 1967 are given.

where ΔZ_i is the atmospheric mass in gm cm^{-2} contained in layer i . Finally, the energy loss $E_{Li}(E)$ by an isotropic flux of monoenergetic protons over the upper hemisphere is found by integrating $E_{Li}(\theta, E)$

$$E_{Li}(E) = \int_0^{2\pi} \int_0^{\pi/2} \cos \theta E_{Li}(\theta, E) \sin \theta d\theta d\phi \quad \text{MeV sr} \quad (\text{A4})$$

where ϕ is the azimuthal angle and θ the polar angle.

The N, N^+ , O, O^+ , and ion pairs produced in the i th slab can be calculated by using the energy loss per specie given in Porter *et al.* [1976]. In large SPEs enough N atoms are created to cause (5) to be quite important (e.g., August 1972 SPEs) as a loss for ON species. Here, we ignore this loss and assume that 1.25 NO molecules are produced per ion pair [Jackman *et al.*, 1979].

Previous studies have used several different analytic forms to characterize the energy spectra of SPEs [Freier and Webber, 1963; Zmuda and Potemra, 1972]. Here we choose a differential spectrum which is exponential in rigidity

$$\Phi(P) = J_p \exp(-P(E)/P_0) \text{ cm}^{-2} \text{ s}^{-1} \text{ sr}^{-1} \text{ MeV}^{-1} \quad (\text{A5})$$

where $P(E) = pc/q = (E^2 + 2Em_0c^2)^{1/2}$ with p = the momentum, c = the speed of light, m_0 = the mass of a proton, E = the kinetic energy of the proton, and $q = 1$ for the charge on protons. We also assume that the e folding rigidity, P_0 , does not change during the event. The intensity is assumed to be time-dependent with an exponential rise of $\exp(t/T_1)$, where t is the time in hours from the initiation of the event up to the peak intensity, J_p , at time T_{max} . The intensity fall off has an $\exp(-t/T_2)$ shape where t here is the time in hours from T_{max} up to T_D , the duration of the event. Use of this time-dependent spectral form requires the six parameters P_0 , J_p , T_1 , T_2 , T_{max} , and T_D .

Through this form many SPE proton spectra from the years 1954–1979 were characterized. Figure A1 relates J_p and P_0 for a solar cycle from 1956 through 1967. Generally, the lower the value of J_p the higher the e folding rigidity P_0 . The altitude of ON production is important in determining the total net ON actually added to the atmosphere. Figure A2 presents the altitude of energy deposition as a function of the e folding rigidity P_0 . The solid line shows the altitude of maximum NO production, the dashed lines indicate the altitudes of one half the maximum production and the dash-dot lines represent the altitudes of one tenth the maximum production. It was found at all values of P_0 that 70%–80% of the total NO production was between the one-half maximum values and that 97%–99% of the total NO production was between the one-tenth maximum value points.

Data to obtain the six parameters (P_0 , J_p , T_1 , T_2 , T_{max} , and T_D) needed to characterize each SPE were taken from several sources. Svestka and Simon [1975] present data on 352 events that have occurred in the time frame from 1955 through 1969. For each event they give the total duration time, T_D , and the Smart and Shea [1971] index numbers which are an indirect measure of the intensity and rigidity of the event. Information from the Solar-Geophysical Data was taken for SPEs in the time frame from May 1967 through August 1972. Freier and Webber [1963] provided J_p and P_0 information for SPEs occurring from 1956 through 1961. Bailey [1964] gave intensity, T_{max} , and T_D information. Zmuda and Potemra [1972] gave peak spectral information which could easily be converted to give the P_0 and J_p needed in (A5). Detailed information about SPEs was available, using the proton flux data from the Goddard Space Flight Center experiment on the IMP 7 (1973–1977) and IMP 8 (1978 and 1979) satellites (R. E. McGuire, T. T. von Rosenvinge, and F. B. McDonald, private communication, 1979). These data were used to give values for all six

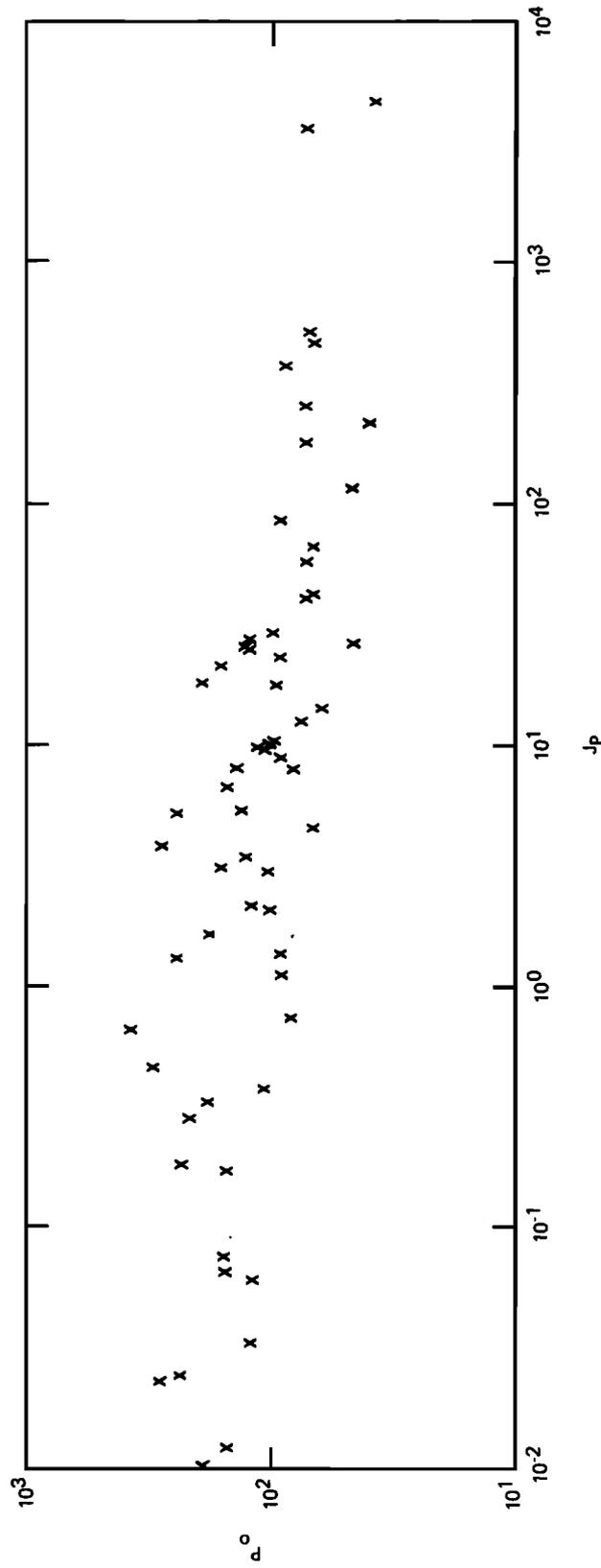


Fig. A2. Altitude range of influence for SPEs with proton spectra of ϵ folding rigidity, P_0 . Dashed lines and dash-dot lines represent one-half and one-tenth maximum ionization-rate altitudes, respectively, while the solid line represents the altitudes of maximum ionization rate.

parameters needed to characterize the time dependency of each SPE proton spectrum.

Some time integral information is available about proton fluxes of the SPEs. King [1974] provides this information for several SPEs occurring in the time period from July 1966 through August 1972. These data were used to derive a J_p and P_0 for a time integral form of (A5). We obtain further integral information by using ion pair production profiles provided in Bauer [1978] for the SPEs occurring in February 1956, November 1960, September 1966, and August 1972.

Where information was not available concerning each of the six time-dependent spectrum parameters, the parameters from known SPEs, which had similar characteristics, including the Smart and Shea index, duration of the event, and the peak intensity of protons with energies greater than 10 MeV, were used to infer the remaining unknown parameters. There is a large uncertainty in this approach for several reasons, three of which are (1) the e folding rigidity, P_0 , is not constant throughout the event; (2) the time dependency cannot always be described appropriately with an exponential increase and decrease; and (3) SPEs with similar Smart and Shea indices, time duration, and/or peak intensity of protons with energies >10 MeV may not have the same time-dependent spectrum parameters, in general. The data are quite good for SPEs that have occurred since October 1972, thus a more exact approach may be used in future work to describe the time-dependent NO production rate.

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