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## AERONOMICA ACTA

A - N° 134 - 1974

On the production of nitric oxide by cosmic rays  
in the mesosphere and stratosphere

by

M. NICOLET

B E L G I S C H   I N S T I T U U T   V O O R   R U I M T E - A E R O N O M I E

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## FOREWORD

"On the production of nitric oxide by cosmic rays in the mesosphere and stratosphere" is reproduced as Scientific Report of the Ionospheric Research Laboratory of the Pennsylvania State University and will be published in Planetary and Space Science.

## AVANT-PROPOS

L'article intitulé "On the production of nitric oxide by cosmic rays in the mesosphere and stratosphere" est également publié comme Scientific Report of the Ionospheric Research Laboratory of the Pennsylvania State University et sera publié dans la revue Planetary and Space Science.

## VOORWOORD

"On the production of nitric oxide by cosmic rays in the mesosphere and stratosphere" zal gepubliceerd worden in Scientific Report of the Ionospheric Research Laboratory of the Pennsylvania State University en in Planetary and Space Science.

## VORWORT

Diese Arbeit "On the production of nitric oxide by cosmic rays in the mesosphere and stratosphere" wird in Scientific Report of the Ionosphere Research Laboratory of the Pennsylvania State University und in Planetary and Space Science herausgegeben werden.

# ON THE PRODUCTION OF NITRIC OXIDE BY COSMIC RAYS IN THE MESOSPHERE AND STRATOSPHERE

by

M. NICOLET

## *Abstract*

Nitric oxide is formed in the atmosphere through the ionization and dissociation of molecular nitrogen by galactic cosmic rays. One NO molecule is formed for each ion pair produced by cosmic ray ionization.

The height-integrated input (day and night) to the lower stratosphere is of the order of  $6 \times 10^7$  NO molecules  $\text{cm}^{-2} \text{sec}^{-1}$  in the auroral zone (geomagnetic latitude  $\Phi \geq 60^\circ$ ) during the minimum of the sunspot cycle and  $4 \times 10^7$  NO molecules  $\text{cm}^{-2} \text{sec}^{-1}$  in the subauroral belt and auroral region ( $\Phi \geq 45^\circ$ ) at the maximum of solar activity. The tropical production is less than  $10^7$  NO molecules  $\text{cm}^{-2} \text{sec}^{-1}$  above 17 km and at the equator the production is only  $3 \times 10^6$  NO molecules  $\text{cm}^{-2} \text{sec}^{-1}$ .

## *Résumé*

L'oxyde nitrique est formé dans l'atmosphère terrestre à la suite de l'ionisation et de la dissociation de l'azote moléculaire par le rayonnement cosmique galactique. Une molécule NO est formée pour chaque paire d'ions produite par l'ionisation du rayonnement cosmique.

La production totale qui a lieu jour et nuit dans la stratosphère inférieure est de  $6 \times 10^7$  molécules NO  $\text{cm}^{-2} \text{sec}^{-1}$  pour toute la zone aurorale (latitudes géomagnétiques supérieures à  $60^\circ$ ) au cours d'un minimum d'activité solaire. Elle atteint au cours d'un maximum d'activité solaire  $4 \times 10^7$  molécules NO  $\text{cm}^{-2} \text{sec}^{-1}$  dans la ceinture subaurorale et la région aurorale ( $\Phi \geq 45^\circ$ ). La production tropicale est inférieure à  $10^7$  molécules NO  $\text{cm}^{-2} \text{sec}^{-1}$  au-dessus de 17 km alors qu'à l'équateur elle descend jusqu'à  $3 \times 10^6$  molécules NO  $\text{cm}^{-2} \text{sec}^{-1}$ .

### *Samenvatting*

Stikstofoxyde wordt in de atmosfeer gevormd door ionisatie en dissociatie van moleculaire stikstof door de kosmische straling van galactische oorsprong. Eén molekule NO wordt gevormd voor elk ionenpaar dat ontstaat tijdens ionisatie door de kosmische straling.

De totale productie die zowel overdag als tijdens de nacht plaats heeft in de lagere stratosfeer bedraagt voor het auroral gebied (geomagnetische breedte  $\Phi$  groter dan  $60^\circ$ ) ongeveer  $6 \times 10^7$  NO molekulen  $\text{cm}^{-2} \text{sec}^{-1}$  tijdens het minimum van de zonnecyclus. Zij bedraagt tijdens het maximum van de zonneactiviteit  $4 \times 10^7$  molekulen  $\text{cm}^{-2} \text{sec}^{-1}$  in het auroral gebied en in de sub-auroral gordel ( $\Phi \geq 45^\circ$ ). De productie in de tropen is kleiner dan  $10^7$  molekulen NO  $\text{cm}^{-2} \text{sec}^{-1}$  boven 17 km en aan de evenaar bedraagt zij slechts  $3 \times 10^6$  NO molekulen  $\text{cm}^{-2} \text{sec}^{-1}$ .

### *Zusammenfassung*

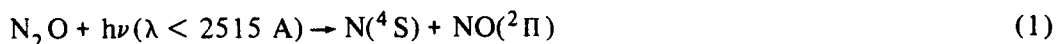
Stickstoffoxid wird in der Atmosphäre durch Ionisation und Dissoziation von Molekularstickstoff bei galactischen kosmischen Strahlungen gebildet. Eine NO Molekul wird gebildet für jedes Ionenpaar, das durch kosmische Strahlung Ionisation produziert ist.

Die totale Produktion (Tag und Nacht) in der untere Stratosphäre ist  $6 \times 10^7$  NO Molekulen  $\text{cm}^{-2} \text{sec}^{-1}$  für die ganze Polarlichtzone (geomagnetische Breite  $\Phi$  grösser als  $60^\circ$ ) während ein Minimum der Sonnenaktivität. Sie erreicht  $4 \times 10^7$  NO Molekulen  $\text{cm}^{-2} \text{sec}^{-1}$  in der Unterpolarlichtzone und in der Polarlichtzone ( $\Phi \geq 45^\circ$ ) während ein Maximum der Sonnenaktivität. Die tropische Produktion ist kleiner als  $10^7$  NO Molekulen  $\text{cm}^{-2} \text{sec}^{-1}$  oberhalb 17 km und am Äquator ist die Produktion nur  $3 \times 10^6$  NO Molekulen  $\text{cm}^{-2} \text{sec}^{-1}$ .

## 1. INTRODUCTION

Oxides of nitrogen formed in the thermosphere by reactions involving charged species (Nicolet, 1965b; Norton and Barth, 1970; Strobel *et al.*, 1970; Nicolet, 1970a; Strobel, 1971a, b; 1972) cannot penetrate down into the stratosphere (Brasseur and Nicolet, 1973). The photodissociation of nitric oxide (Strobel 1972; Brasseur and Nicolet, 1973) is more important in the mesosphere than its transport rate and the normal presence of nitrogen oxides in the stratosphere must be due to their formation in that region.

Among the various possible processes leading to the formation of nitrogen oxides in the stratosphere, we may consider as the principal process the decomposition of nitrous oxide (Bates and Hays, 1967; Crutzen, 1970, 1971; Nicolet, 1970b, 1971; McElroy and McConnell, 1971; Nicolet and Vergison, 1971; Nicolet and Peetermans, 1972). However, direct photodissociation such as



is aeronomically impossible (Nicolet and Peetermans, 1972) and the exclusive primary dissociation process of  $\text{N}_2\text{O}$  yields oxygen atoms. But, as introduced by Nicolet (1970b, 1971), nitric oxide is only generated by the reaction



The  $\text{O}(^1\text{D})$  atoms are formed by the photodissociation of stratospheric ozone. The calculated vertical distribution of  $\text{N}_2\text{O}$  in the stratosphere depends strongly on the values adopted for the eddy diffusion coefficient (Nicolet and Vergison, 1971; McElroy and McConnell, 1971; Crutzen, 1971; Nicolet and Peetermans, 1972).

The oxidation of  $\text{NH}_3$  has also been proposed as a dominant stratospheric source of  $\text{NO}_x$  (McConnell, 1973; McConnell and McElroy, 1973). It requires a  $\text{NH}_3$  mixing ratio of the order of at least  $10^{-9}$  in the lower stratosphere in order to play a role in the production

of nitric oxide molecules (Fig. 10, Nicolet, 1973; Fig. 8, 1974). Since  $\text{NH}_3$  has not been observed in the stratosphere such a source is probably not important. According to Crutzen, (1974),  $\text{NH}_3$  can be removed efficiently by heterogeneous processes in the troposphere. A recent analysis of the solar spectrum by Kaplan (1973) shows that the mixing ratio could be much smaller than  $8 \times 10^{-11}$ . However, recent measurements by Georgii and Muller (1974) lead to mixing ratios as low as  $5 \times 10^{-10}$  to  $1 \times 10^{-9}$  in the polar maritime air and to about  $5 \times 10^{-9}$  over the continent. In any case, an accurate analysis of this problem is still required.

Finally, recent independent studies (Warneck, 1972; Nicolet and Peetermans, 1972) have proposed the production of nitric oxide by cosmic rays acting on  $\text{N}_2$ . Brasseur and Nicolet (1973) have indicated that the production of nitric oxide molecules is of the order of one molecule per ion pair produced by cosmic rays. The production of nitric oxide by cosmic rays is also a nighttime process and is related to the geomagnetic latitude. An attempt will be made here to estimate the world-wide mesospheric and stratospheric production of nitric oxide by the process.

## 2. IONIZATION AND DISSOCIATION BY COSMIC RAYS

Ionization due to cosmic rays (Dalgarno, 1967) is produced mostly by secondary electrons ejected by heavy particles. The experimental results of Rapp *et al.* (1965) give the cross-sections for dissociative ionization of molecules by electron impact from the threshold up to 1000 eV (Fig. 1). The dissociative ionization cross-section rises from threshold to a maximum near 100 eV and decreases with increasing energy. The cross-section for the dissociative ionization process  $\text{N}_2 \rightarrow \text{N} + \text{N}^+$  corresponds to about 20% of the total ionization cross-section for electron energies greater than 100 eV and to about 25% for  $\text{O}_2 \rightarrow \text{O} + \text{O}^+$  under the same conditions. Thus, the ratios of  $\text{N}_2^+/\text{N}^+$  and  $\text{O}_2^+/\text{O}^+$  ion production are 4 and 3, respectively.

The production of *molecular* nitrogen ions in the air is, therefore,  $\frac{4}{5} \times 0.8 = 0.64$  of an ion pair and the production of *atomic* nitrogen ions is  $\frac{4}{5} \times 0.2 = 0.16$  in the same condi-

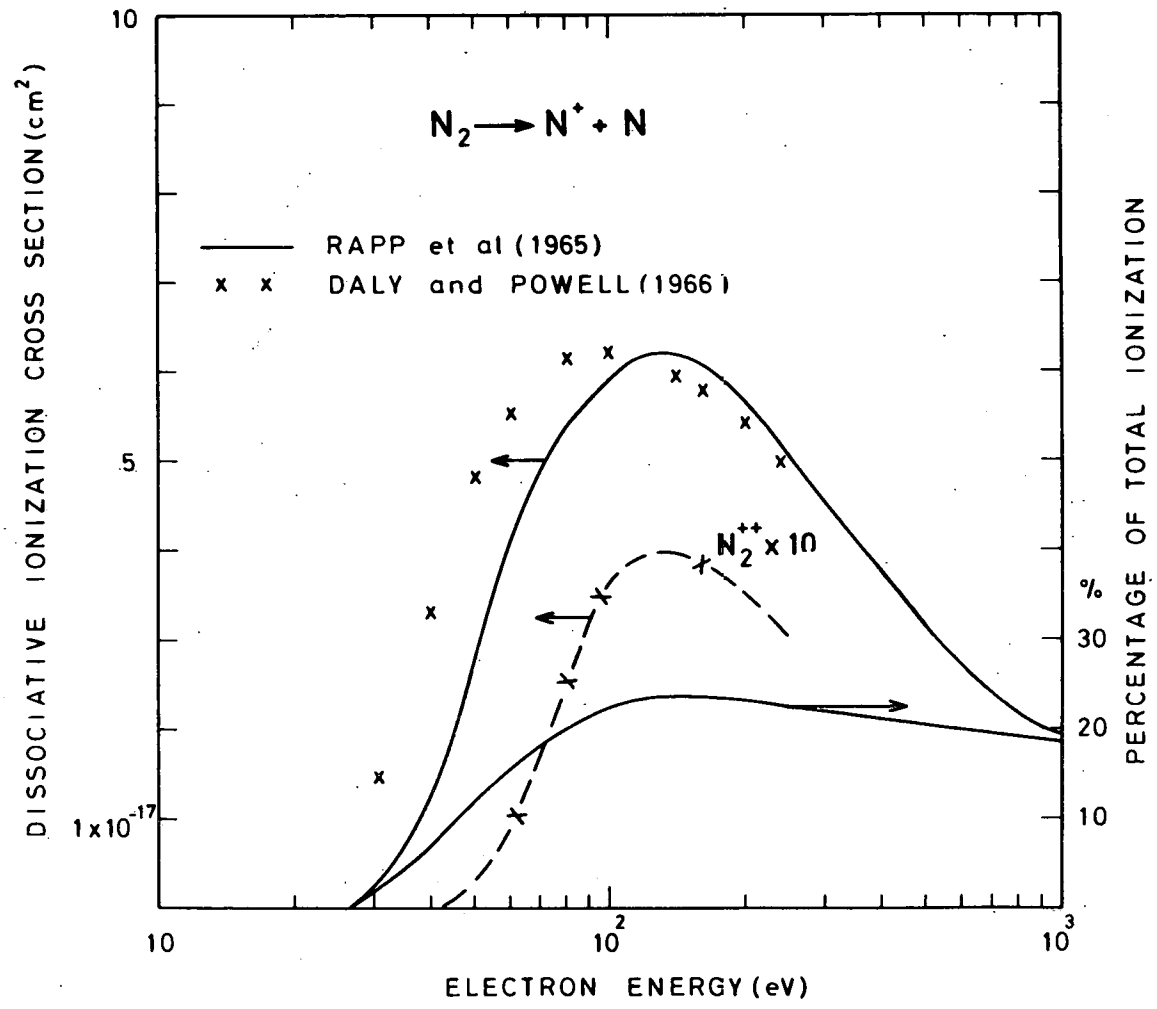
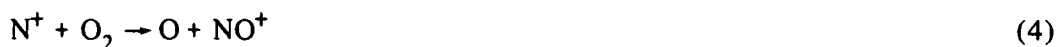


Fig. 1.- Dissociative ionization cross-section of molecular nitrogen versus electron energy.

tions. Assuming that the  $N^+$  ions (Dalgarno, 1967) are immediately converted by a charge transfer reaction into neutral nitrogen atoms



or indirectly transformed through the reaction



the production of nitrogen atoms through the dissociative ionization process must be  $0.16 \times 2 = 0.32$  nitrogen atoms per ion pair produced by cosmic rays.

The total dissociation cross-section has been measured (fig. 2) by Winters (1966). The average cross-section for electron energies between 30 and 300 eV is of the order of  $1.75 \times 10^{-16} \text{ cm}^2$ , corresponding to about three times the average dissociative ionization cross-section ( $5 \times 10^{-17} \text{ cm}^2$ ) in the same energy range. Thus, the production of nitrogen atoms by direct dissociation is  $0.32 \times 2 = 0.64$  per ion pair produced by cosmic rays. The total yield of nitrogen atoms produced by dissociation and dissociative ionization of  $N_2$  is therefore approximately *one* (0.96) nitrogen atom for *every* ion pair produced by cosmic rays. The accuracy of this ratio can be improved when more precise experimental data are obtained. A possible effect of  $O^+$  ions will not change the approximate value of 1.00 adopted for the production of nitrogen atoms. The total yield of odd nitrogen would, however, increase above one nitrogen atom for one ion pair produced by cosmic rays if a reaction such as  $O_2^+ + N_2 \rightarrow NO^+ + NO$  were accepted at stratospheric levels.

### 3. REACTION OF NITROGEN ATOMS

The dissociative ionization and dissociation of  $N_2$  and the dissociative recombination of molecular ions may lead to a production of nitrogen atoms in the normal  $^4S$  state and in the excited states  $^2D$  and  $^2P$ . Since the life time of the excited state  $^2D$  is long ( $9.4 \times 10^4$  sec), a direct reaction between  $N(^2D)$  and  $O_2$  must be considered. Thus, after its pro-



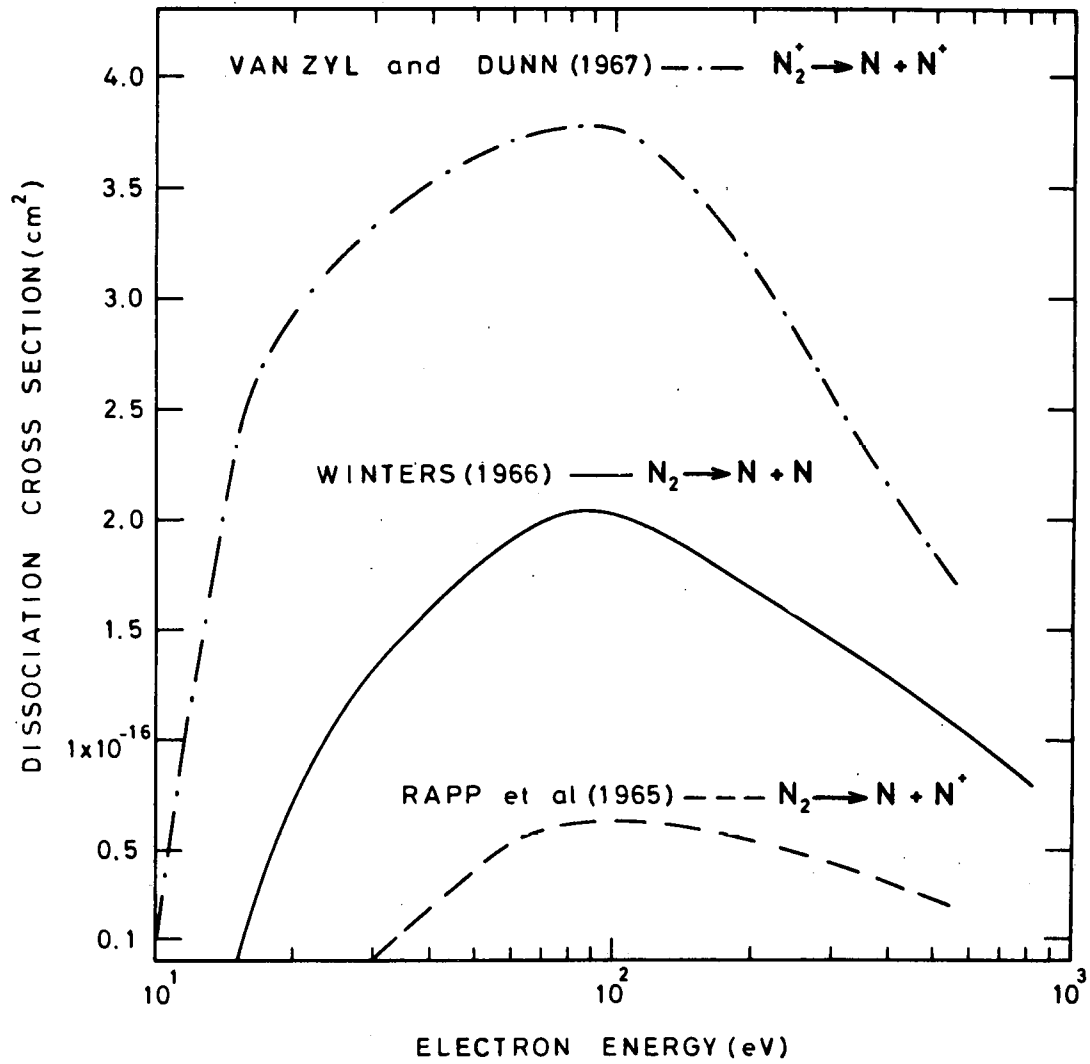


Fig. 2.- Dissociation cross-section of  $N_2$  and  $N_2^+$  versus electron energy.

duction, atomic nitrogen reacts as follows



with a rate coefficient (Becker *et al.*, 1969)

$$b_7 = 7.5 \times 10^{-15} T e^{-3000/T} \text{ cm}^3 \text{ sec}^{-1} \quad (6)$$

or



with a rate coefficient (Lin and Kaufman, 1971; Slanger *et al.*, 1971)

$$b_7^* = 6 \times 10^{-12} \text{ cm}^3 \text{ sec}^{-1} \quad (8)$$

Of the various aeronomic processes leading to the union of odd nitrogen atoms, only the following reaction (Nicolet, 1965a) can play a role



with a rate coefficient known within a factor of two

$$b_6 = 1.5 \times 10^{-12} T^{1/2} \text{ cm}^3 \text{ sec}^{-1} \quad (10)$$

Its role can be neglected in the normal processes of NO formation by (5) and (7); with an excess of nitrogen atoms, reaction (9) must be considered. In a perturbed atmosphere, the reaction  $\text{N} + \text{NO}_2 \rightarrow \text{NO} + \text{NO}$ ,  $\text{N}_2\text{O} + \text{O}$  and  $\text{N}_2 + \text{O}_2$  should be considered for a complete analysis of NO production.

#### 4. THE COSMIC RAY VARIATION

The galactic cosmic radiation, which is essentially isotropic as observed on the Earth, is modulated by the interplanetary magnetic field and hence its effect on NO production is related to solar activity. Since its intensity depends on the geomagnetic field, there is an ionization or dissociation effect which varies strongly with latitude. Table I gives the production of ion pairs, nitrogen atoms or nitric oxide molecules between 85 km and 35 km for a solar activity cycle. The minimum value is reached near the maximum of the solar activity and the maximum value corresponds with minimum solar activity. There is a solar activity effect which is greater than a factor of 2 at high latitudes. In fact, the variation with solar activity depends on the geomagnetic regions. Fig. 3a and 3b show the geomagnetic regions which are defined as follows, using geographic coordinates of  $\varphi_0 = 78^\circ 5' \text{ N}$  and  $\lambda_0 = 69^\circ \text{ W}$  for the north geomagnetic pole (Nicolet, 1959):

- (1) The two *Auroral Regions* for each hemisphere cover the polar regions down to  $60^\circ$  geomagnetic latitude.
- (2) The two *Subauroral Belts* cover the regions between  $60^\circ$  and  $45^\circ$  geomagnetic latitudes
- (3) The *Minauroral Belt* covers the regions between geomagnetic latitudes  $45^\circ \text{ N}$  and  $\text{S}$ .
- (4) The *Equatorial Region* is that part of the minauroral belt with a geomagnetic latitude of less than  $20^\circ \text{ N}$  and  $\text{S}$ .

The production is almost constant in the equatorial region (see Fig. 4 and Table I); the latitudinal and solar activity effects are very small. In the other part of the minauroral belt ( $20^\circ < \Phi < 45^\circ$ ) there is a rapid increase of the NO production with latitude with smaller differences due to the solar activity effect. In the subauroral belt ( $45^\circ \leq \Phi \leq 60^\circ$ ) the production rate of nitric oxide almost reaches its maximum and the variation with solar activity is particularly large at the border of the auroral zone ( $\Phi = 60^\circ$ ). In the auroral zone ( $\Phi > 60^\circ$ ) the production rate of nitric oxide is constant for the same solar activity conditions but varies strongly with solar activity. The values covering the solar maximum (1958) and solar minimum (1965) lead to the minimum and maximum production of nitric molecules in the auroral regions and in a part of the subauroral belt ( $\Phi < 60^\circ$ ) for high solar activity conditions. A knee in the curves for 20 km (Fig. 4) occurs at  $\Phi < 60^\circ$  at solar

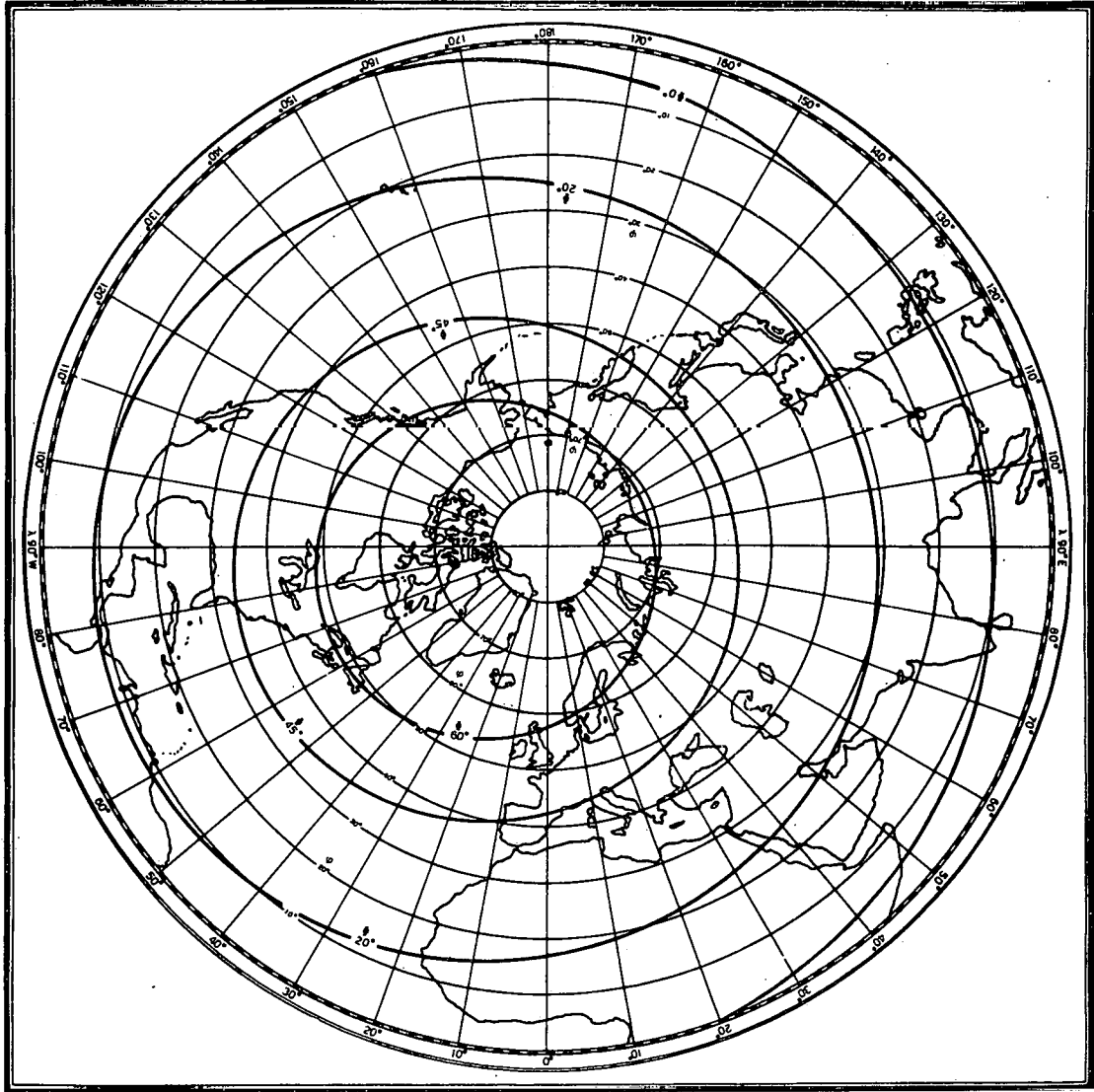


Fig. 3a.- Geomagnetic regions in the northern hemisphere.

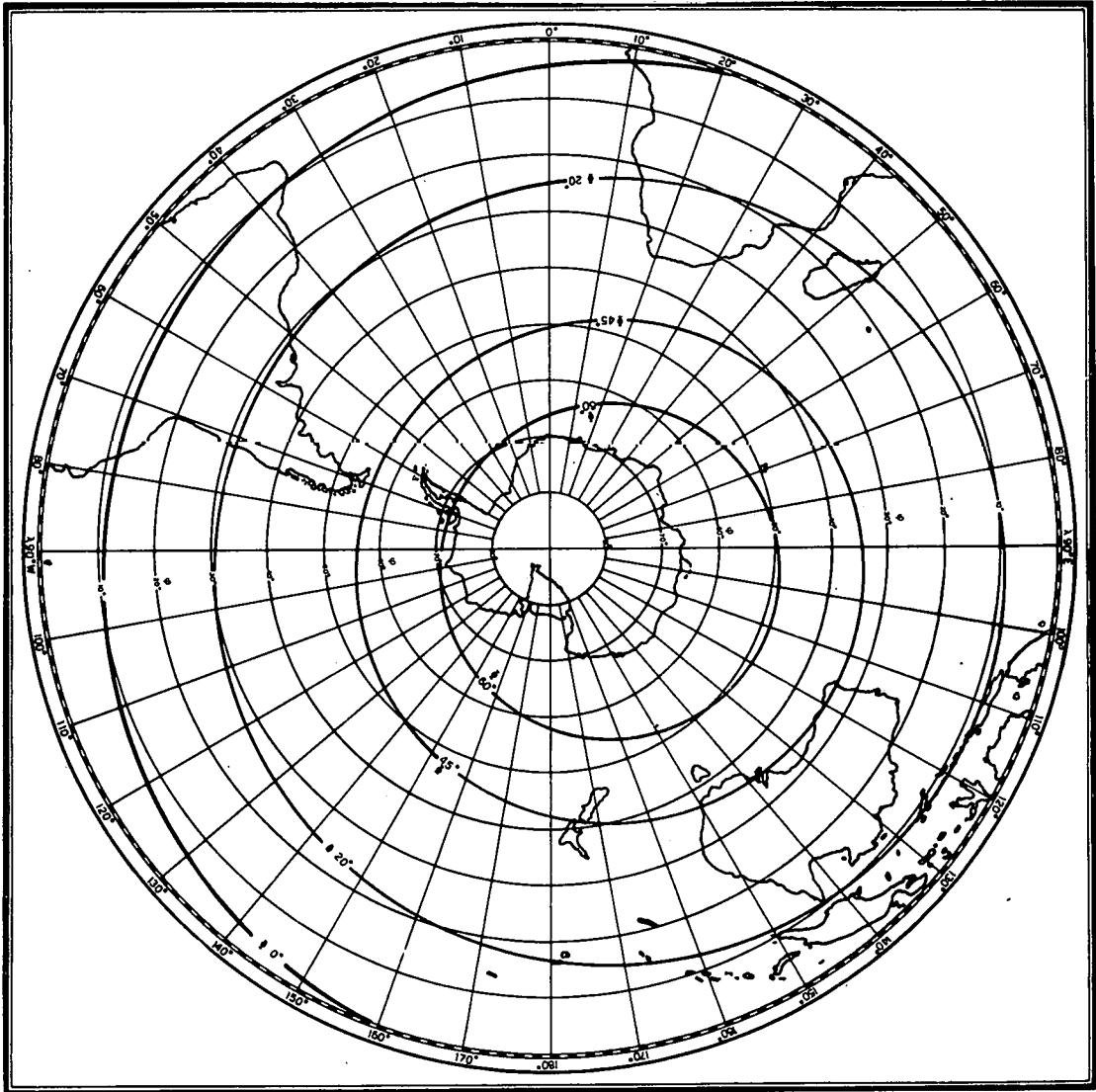


Fig. 3b.- Geomagnetic regions in the southern hemisphere.

TABLE I.- Cosmic ray production of nitric oxide [(or ion pair or atomic nitrogen) ( $\text{cm}^{-3} \text{sec}^{-1}$ )] in the mesosphere and upper stratosphere.

Altitude (km)	Latitude $\Phi$			
	$0^\circ$	$15^\circ$	$30^\circ$	$40^\circ$
85	$(3.3 \pm 0.1) \times 10^{-4}$	$(3.6 \pm 0.1) \times 10^{-4}$	$(6.0 \pm 0.3) \times 10^{-4}$	$(9.6 \pm 0.4) \times 10^{-4}$
80	$(8.0 \pm 0.2) \times 10^{-4}$	$(8.8 \pm 0.2) \times 10^{-4}$	$(1.5 \pm 0.1) \times 10^{-3}$	$(2.3 \pm 0.1) \times 10^{-3}$
75	$(1.8 \pm 0.05) \times 10^{-3}$	$(2.0 \pm 0.05) \times 10^{-3}$	$(3.3 \pm 0.2) \times 10^{-3}$	$(5.2 \pm 0.3) \times 10^{-3}$
70	$(3.7 \pm 0.1) \times 10^{-3}$	$(4.1 \pm 0.1) \times 10^{-3}$	$(6.9 \pm 0.4) \times 10^{-3}$	$(1.1 \pm 0.1) \times 10^{-2}$
65	$(7.2 \pm 0.2) \times 10^{-3}$	$(8.0 \pm 0.2) \times 10^{-3}$	$(1.3 \pm 0.1) \times 10^{-2}$	$(2.1 \pm 0.1) \times 10^{-2}$
60	$(1.3 \pm 0.05) \times 10^{-2}$	$(1.5 \pm 0.05) \times 10^{-2}$	$(2.4 \pm 0.2) \times 10^{-2}$	$(3.4 \pm 0.2) \times 10^{-2}$
55	$(2.3 \pm 0.05) \times 10^{-2}$	$(2.6 \pm 0.15) \times 10^{-2}$	$(4.3 \pm 0.2) \times 10^{-2}$	$(6.8 \pm 0.3) \times 10^{-2}$
50	$(4.3 \pm 0.1) \times 10^{-2}$	$(4.7 \pm 0.1) \times 10^{-2}$	$(7.8 \pm 0.5) \times 10^{-2}$	$(1.2 \pm 0.1) \times 10^{-1}$
45	$(7.9 \pm 0.2) \times 10^{-2}$	$(8.7 \pm 0.2) \times 10^{-2}$	$(1.4 \pm 0.1) \times 10^{-1}$	$(2.3 \pm 0.1) \times 10^{-1}$
40	$(1.5 \pm 0.5) \times 10^{-1}$	$(1.7 \pm 0.05) \times 10^{-1}$	$(2.8 \pm 0.1) \times 10^{-1}$	$(4.4 \pm 0.2) \times 10^{-1}$
35	$(3.1 \pm 0.1) \times 10^{-1}$	$(3.4 \pm 0.1) \times 10^{-1}$	$(5.6 \pm 0.3) \times 10^{-1}$	$(9.0 \pm 0.4) \times 10^{-1}$

TABLE I. cont.

Altitude (km)	$\Phi = 50^{\circ}$			$\Phi = 60^{\circ}$		
	Min	Mean	Max	Min	Mean	Max
85	$1.4 \times 10^{-3}$	$1.7 \times 10^{-3}$	$2.0 \times 10^{-3}$	$1.6 \times 10^{-3}$	$1.8 \times 10^{-3}$	$3.3 \times 10^{-3}$
80	$3.5 \times 10^{-3}$	$4.2 \times 10^{-3}$	$4.0 \times 10^{-3}$	$3.9 \times 10^{-3}$	$4.4 \times 10^{-3}$	$8.0 \times 10^{-3}$
75	$7.8 \times 10^{-3}$	$9.3 \times 10^{-3}$	$1.1 \times 10^{-2}$	$8.8 \times 10^{-3}$	$9.8 \times 10^{-3}$	$1.8 \times 10^{-2}$
70	$1.6 \times 10^{-2}$	$2.0 \times 10^{-2}$	$2.3 \times 10^{-2}$	$1.8 \times 10^{-2}$	$2.1 \times 10^{-2}$	$3.8 \times 10^{-2}$
65	$3.2 \times 10^{-2}$	$3.8 \times 10^{-2}$	$4.5 \times 10^{-2}$	$3.6 \times 10^{-2}$	$4.0 \times 10^{-2}$	$7.3 \times 10^{-2}$
60	$5.8 \times 10^{-2}$	$7.0 \times 10^{-2}$	$8.2 \times 10^{-2}$	$6.5 \times 10^{-2}$	$7.3 \times 10^{-2}$	$1.3 \times 10^{-1}$
55	$1.0 \times 10^{-1}$	$1.2 \times 10^{-1}$	$1.4 \times 10^{-1}$	$1.1 \times 10^{-1}$	$1.3 \times 10^{-1}$	$2.3 \times 10^{-1}$
50	$1.9 \times 10^{-1}$	$2.2 \times 10^{-1}$	$2.6 \times 10^{-1}$	$2.1 \times 10^{-1}$	$2.4 \times 10^{-1}$	$4.3 \times 10^{-1}$
45	$3.5 \times 10^{-1}$	$4.1 \times 10^{-1}$	$4.9 \times 10^{-1}$	$3.9 \times 10^{-1}$	$4.4 \times 10^{-1}$	$7.9 \times 10^{-1}$
40	$6.6 \times 10^{-1}$	$7.9 \times 10^{-1}$	$9.3 \times 10^{-1}$	$7.4 \times 10^{-1}$	$8.3 \times 10^{-1}$	1.5
35	1.3	1.6	1.9	1.5	1.7	3.1

Altitude (km)	$\Phi = 70^{\circ}$		
	Min	Mean	Max
85	$1.6 \times 10^{-3}$	$2.8 \times 10^{-3}$	$4.0 \times 10^{-3}$
80	$3.9 \times 10^{-3}$	$6.8 \times 10^{-3}$	$9.6 \times 10^{-3}$
75	$8.8 \times 10^{-3}$	$1.5 \times 10^{-2}$	$2.2 \times 10^{-2}$
70	$1.8 \times 10^{-2}$	$3.2 \times 10^{-2}$	$4.5 \times 10^{-2}$
65	$3.6 \times 10^{-2}$	$6.2 \times 10^{-2}$	$8.8 \times 10^{-2}$
60	$6.5 \times 10^{-2}$	$1.1 \times 10^{-1}$	$1.6 \times 10^{-1}$
55	$1.1 \times 10^{-1}$	$2.0 \times 10^{-1}$	$2.8 \times 10^{-1}$
50	$2.1 \times 10^{-1}$	$3.7 \times 10^{-1}$	$5.2 \times 10^{-1}$
45	$3.9 \times 10^{-1}$	$6.7 \times 10^{-1}$	$9.5 \times 10^{-1}$
40	$7.4 \times 10^{-1}$	1.3	1.8
35	1.5	2.6	3.7

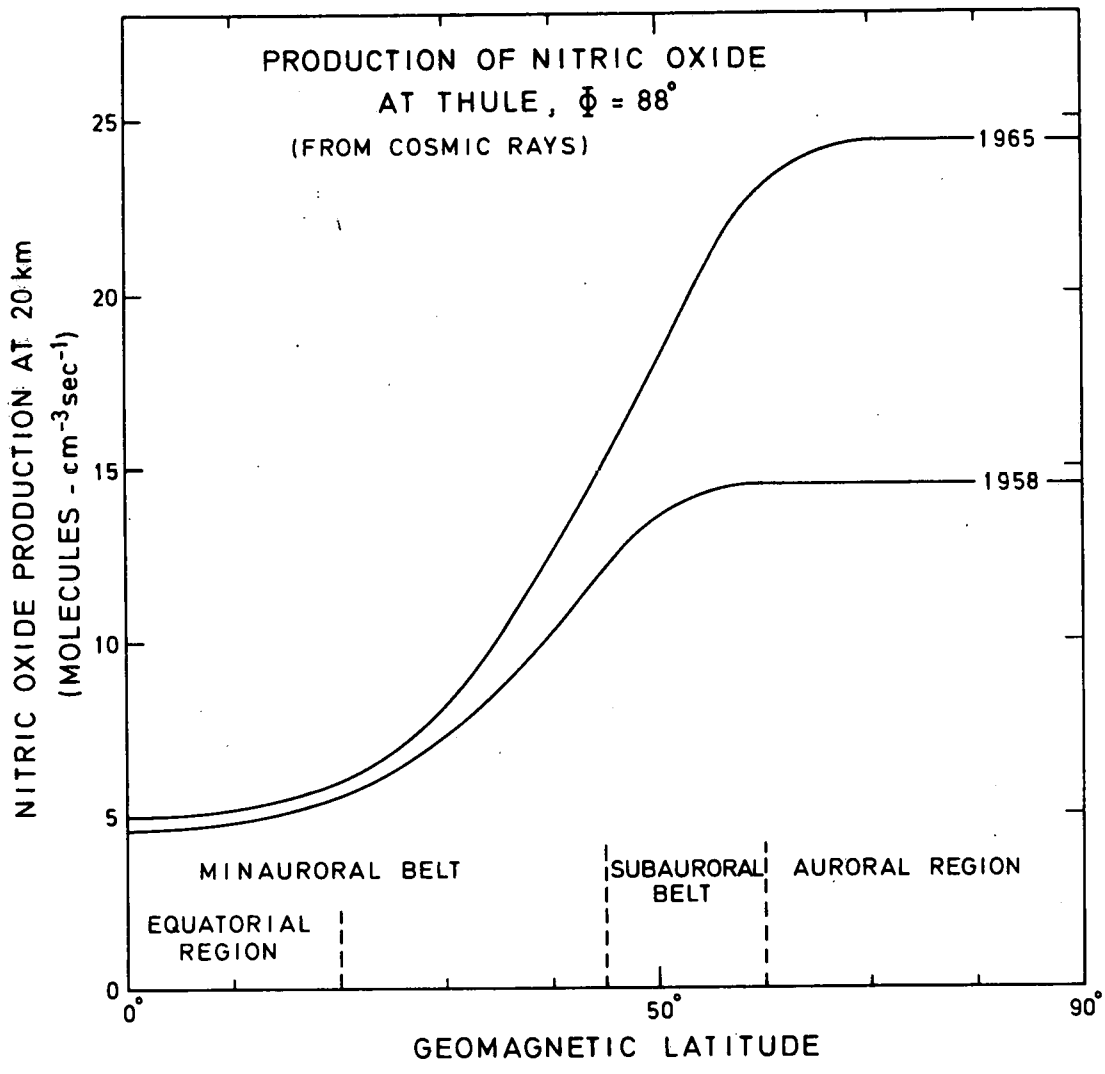


Fig. 4.- Nitric oxide production at 20 km by cosmic rays versus latitude for conditions of solar activity maximum (1958) and minimum (1965). Cosmic ray data deduced from Neher (1961, 1967).



maximum and at  $\Phi > 60^\circ$  at solar minimum. An example of the variation in the auroral region is given in Fig. 5 for two solar cycles from 1955 to 1970. The production of nitric oxide molecules at 20 km varies from a minimum of about  $15 \text{ molecules cm}^{-3} \text{ sec}^{-1}$  to a maximum of about  $25 \text{ molecules cm}^{-3} \text{ sec}^{-1}$ .

### 5. THE VERTICAL DISTRIBUTION OF THE PRODUCTION OF NITRIC OXIDE IN THE STRATOSPHERE

From measurements of cosmic rays in the stratosphere at high latitudes covering several solar maxima and minima (Neher, 1971) it is possible to deduce the maximum and minimum production rates of nitric oxide in the entire auroral region. Fig. 6 is an illustration of various NO production rates. The vertical distributions at different solar minima (1954, 1965) and at different solar maxima (1958, 1969) are not too different, respectively. The variation of the nitric oxide production with altitude is particularly large above the production peak which is at  $12.5 \text{ km} \pm 1 \text{ km}$  depending on the latitude and on the solar cycle. Below the peak, the results (Fig. 6) on cosmic ray ionization obtained by George (1970) on January 10-11, 1968 at constant North geomagnetic latitude  $44^\circ \text{ N}$  (near the beginning of the subauroral belt) do not differ very much from the value for the auroral region ( $\Phi > 60^\circ$ ) near a maximum of solar activity. The following values can be used at  $\Phi \geq 44^\circ$  for the minimum production of nitric oxide molecules.

Altitude (km)	0	1	2	3	4	5	6
NO ( $\text{cm}^{-3} \text{ sec}^{-1}$ )	2.6	3.3	4.2	5.3	7.0	9.1	12.0
Altitude (km)	7	8	9	10	11	12	12.5
NO ( $\text{cm}^{-3} \text{ sec}^{-1}$ )	15.8	19.5	23.1	26.2	29.3	30.6	30.8

There is, therefore, significant stratospheric production of NO molecules (day and night) in the region of  $\Phi \geq 45^\circ$ . Another detailed observation of the cosmic ray ionization (Neher, 1967) at Bismark,  $\Phi = 56^\circ$ , in 1965 from June 28 to August 4 leads to values of nitric

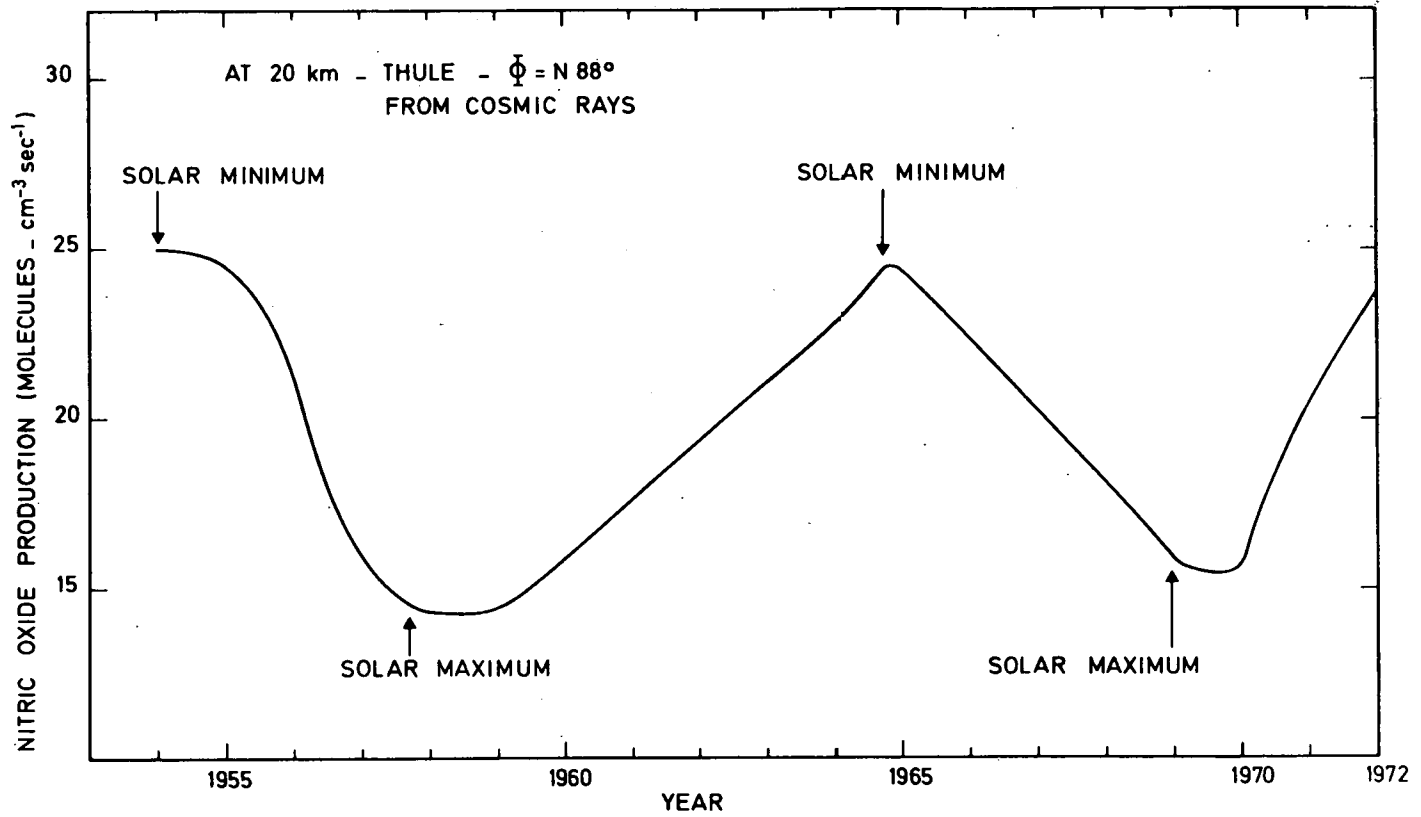


Fig. 5.- Nitric oxide production at 20 km in the northern auroral region at Thule  $\Phi = 88^\circ$ . Cosmic ray data deduced from Neher (1971).

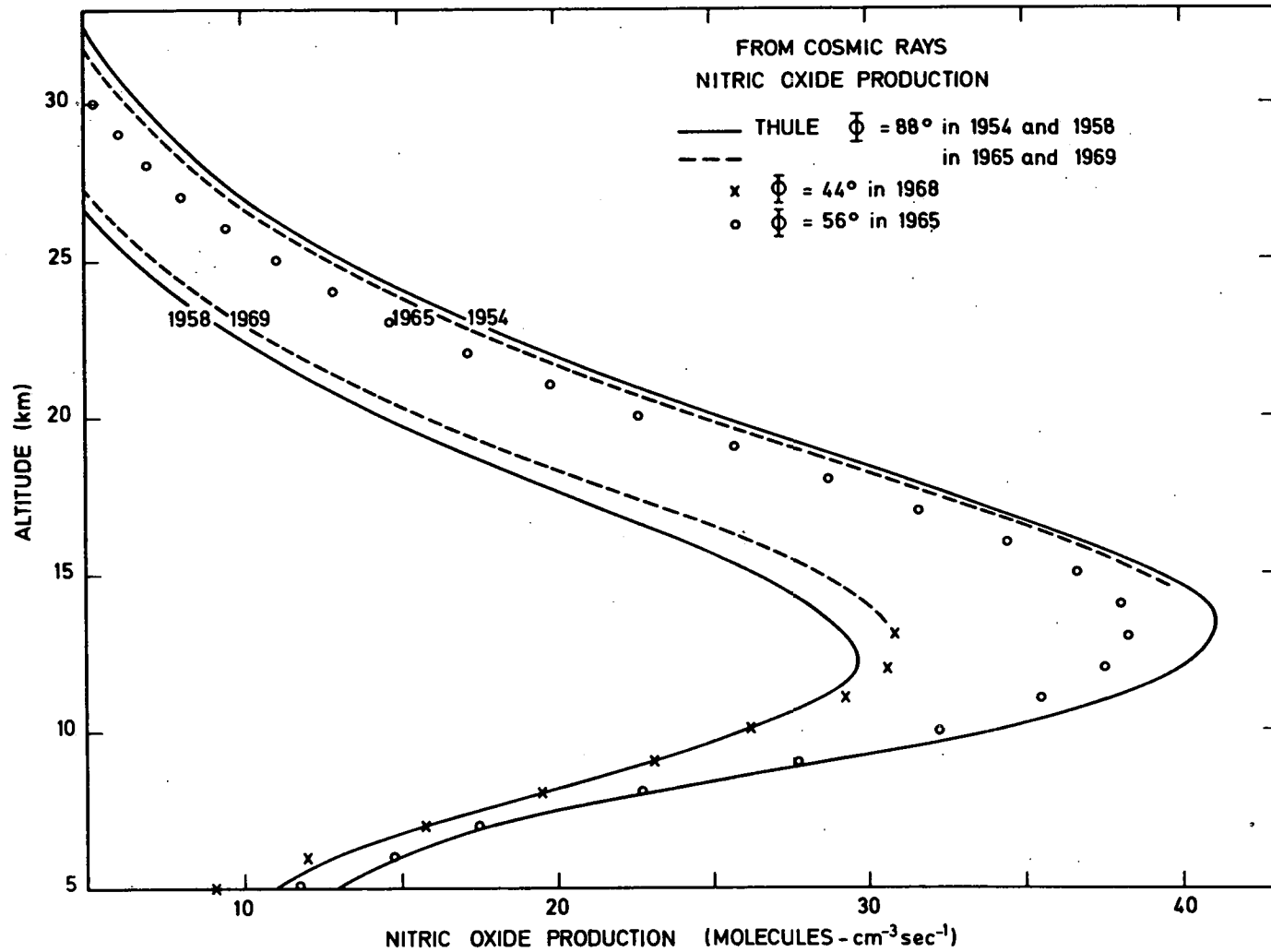


Fig. 6.- Production of nitric oxide at high geomagnetic latitudes during various solar cycles. 1954 and 1965 are minimum solar activity; 1958 and 1969 are maximum solar activity. Cosmic ray data from Neher (1971).  $\Phi = 44^\circ$  in 1968 are from measurements made by George (1970) and  $\Phi = 56^\circ$  by Neher (1967) at Bismark  $\Phi = 56^\circ$  and  $\varphi = 47^\circ$ .

oxide production (Fig. 6) which are not too different from the observations made at Thule ( $\Phi = 88^\circ$ ), particularly below the production peak. Considering the values observed at  $\Phi = 44^\circ$  for maximum solar activity and at  $\Phi = 56^\circ$  for minimum solar activity, the following average values for the production of nitric molecules can be adopted for the upper troposphere and lower stratosphere at  $\Phi \geq 45^\circ$

Altitude (km)	5	6	7	8	9	10
NO ( $\text{cm}^{-3} \text{ sec}^{-1}$ )	$10.4 \pm 1.3$	$13.4 \pm 1.4$	$16.7 \pm 1$	$21.1 \pm 1.5$	$25.4 \pm 2$	$29.2 \pm 3$

A latitude survey of the cosmic ray ionization (Neher, 1967) was made during July 1965 corresponding to a minimum of solar activity which was not too different from 1954 in regards to the production of nitric oxide. Table II gives the values for all latitudes from 10 km to 30 km. The production peak is near 12.5 km ; the variation of the peak altitude can be followed in Fig. 7 and the maximum is reached with a production of 40 NO molecules  $\text{cm}^{-3} \text{ sec}^{-1}$ . As far as the absolute cosmic ray ionization during maximum solar conditions is concerned, there is no systematic survey in the equatorial region. From observations made in July 1958 (Neher, 1961) we obtained the values which are given in Table III and illustrated in Fig. 8. They represent a low minimum for nitric oxide production. From the production peak ( $\sim 12.5$  km) up to 30 km, there is an extension of the auroral region ( $\Phi \geq 60^\circ$ ) (where the NO production is maximum) into the subauroral belt to geomagnetic latitudes  $\Phi \cong 50^\circ$  and  $\Phi \cong 55^\circ$ , respectively. Below the production peak, there is an extension into the whole subauroral belt ( $\Phi \geq 45^\circ$ ). This characteristic (last column of Table III) referred to as the "knee" (Neher, 1961) corresponds at a constant height to the latitude beyond which as one proceeds to higher latitudes, little or no change occurs.

The position of the knee increases with altitude. During maximum solar conditions, the knee occurs at a geomagnetic latitude of about  $40^\circ$  at sea level, at  $\Phi = 44^\circ$  (see Table III) at an altitude of 10 km and at  $\Phi = 56^\circ$  at an altitude of 30 km. During minimum solar conditions, the knee occurs at higher latitudes, for example, at  $\Phi = 52^\circ$  at an altitude of 10 km, at  $\Phi = 55^\circ$  for the production peak at  $12.5 \pm 1.0$  km and  $\Phi = 60^\circ$  at an altitude of 20 km.

If we compare (Fig. 9) the vertical distributions of the nitric oxide production during conditions of minimum and maximum solar activity we can see that there are several regions

TABLE II. Maximum production of nitric oxide molecules ( $\text{cm}^{-3} \text{sec}^{-1}$ )

Altitude (km)	Latitude $\Phi$						
	$0^\circ$	$10^\circ$	$15^\circ$	$20^\circ$	$25^\circ$	$30^\circ$	$35^\circ$
10	(13)	(14)	15.2	17.0	18.8	20.4	22.8
12.5	(14)	(15)	16.1	17.4	19.4	22.1	25.0
15	(10.5)	(11.5)	12.2	13.5	15.3	17.3	21.3
17.5	(7.5)	(8.0)	8.5	9.5	10.6	12.3	15.2
20	(5.0)	(5.2)	5.4	6.0	6.9	8.0	10.1
22.5	(2.7)	(2.8)	3.1	3.6	4.2	4.9	6.2
25	(1.7)	(1.8)	1.9	2.2	2.5	3.1	3.6
27.5	(1.0)	(1.0)	1.1	1.3	1.6	1.9	2.4
30	< 1	< 1	< 1	< 1	< 1	1.1	1.5

Altitude (km)	Latitude $\Phi$				
	$40^\circ$	$45^\circ$	$50^\circ$	$55^\circ$	$\geq 60^\circ$
10	25.2	27.6	30.9	32.0	32.0
12.5	29.0	32.1	35.9	38.8	39.3
15	25.3	28.5	32.0	36.0	37.5
17.5	18.3	21.6	25.4	28.9	31.2
20	12.4	15.0	18.1	21.0	23.2
22.5	7.6	9.8	12.0	14.7	16.7
25	4.6	6.2	8.0	10.0	11.6
27.5	3.1	4.1	5.3	6.4	8.0
30	2.0	2.6	3.2	4.1	5.4

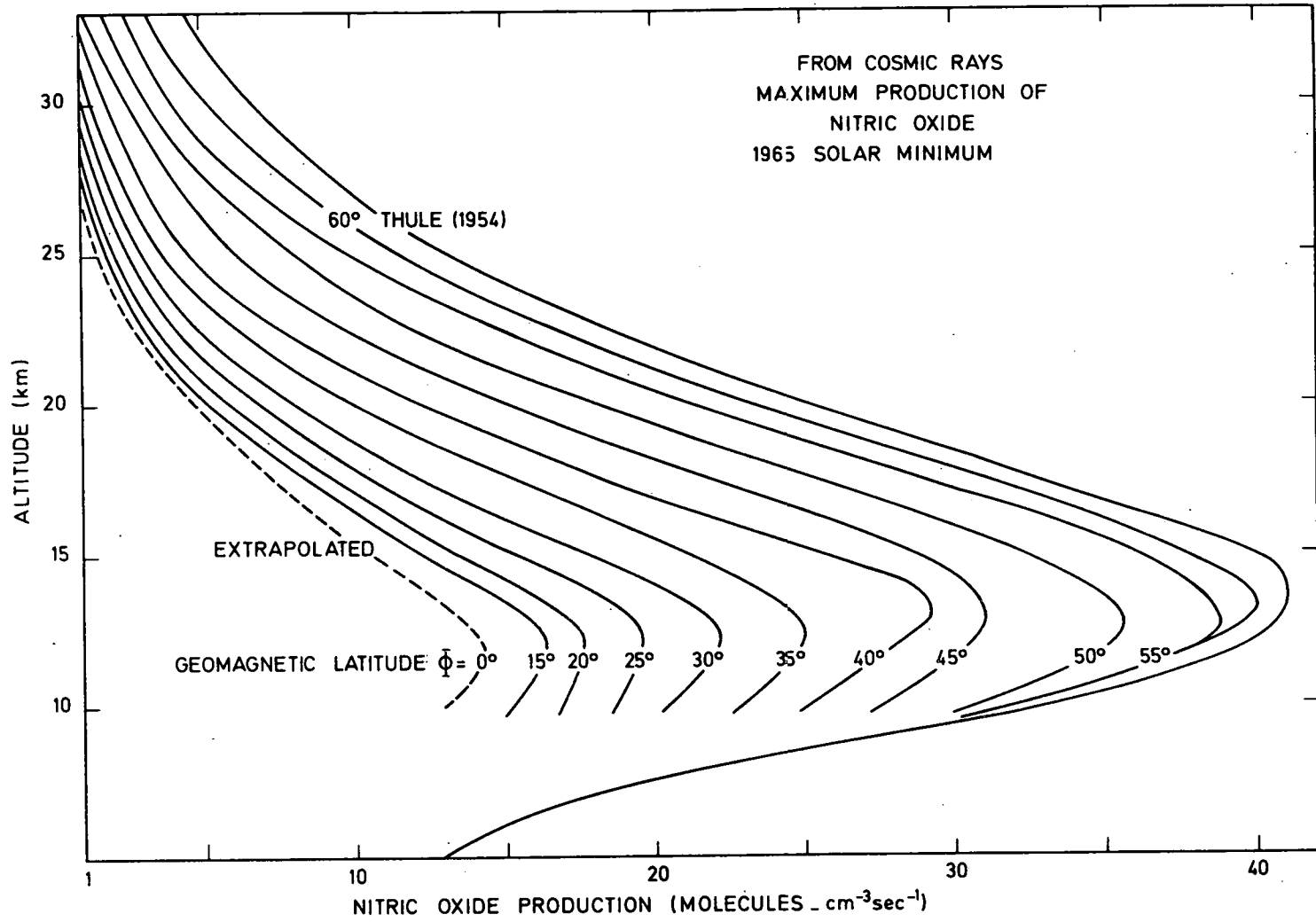


Fig. 7.- Production of nitric oxide during a minimum of solar activity. Cosmic ray data deduced from Neher (1967, 1971).

TABLE III. Minimum production of nitric oxide molecules ( $\text{cm}^{-3} \text{sec}^{-1}$ )

Altitude (km)	Latitude $\Phi$					Knee
	35	40	45	50	55-90 <sup>0</sup>	
10	21.0	24.0	26.0	26.0	26.0	44 <sup>0</sup>
12.5	27.5	25.3	28.2	30.5	30.5	49
15	18.2	21.2	24.2	27.0	27.0	49
17.5	13.3	15.8	18.2	20.6	21.2	51
20	8.2	10.2	12.2	14.2	15.5	53
22.5	5.0	6.2	7.2	8.8	10.1	55
25	3.1	3.9	4.9	5.9	6.8	55
27.5	1.9	2.5	3.2	3.8	4.5	56
30	1.1	1.6	2.1	2.6	3.0	56

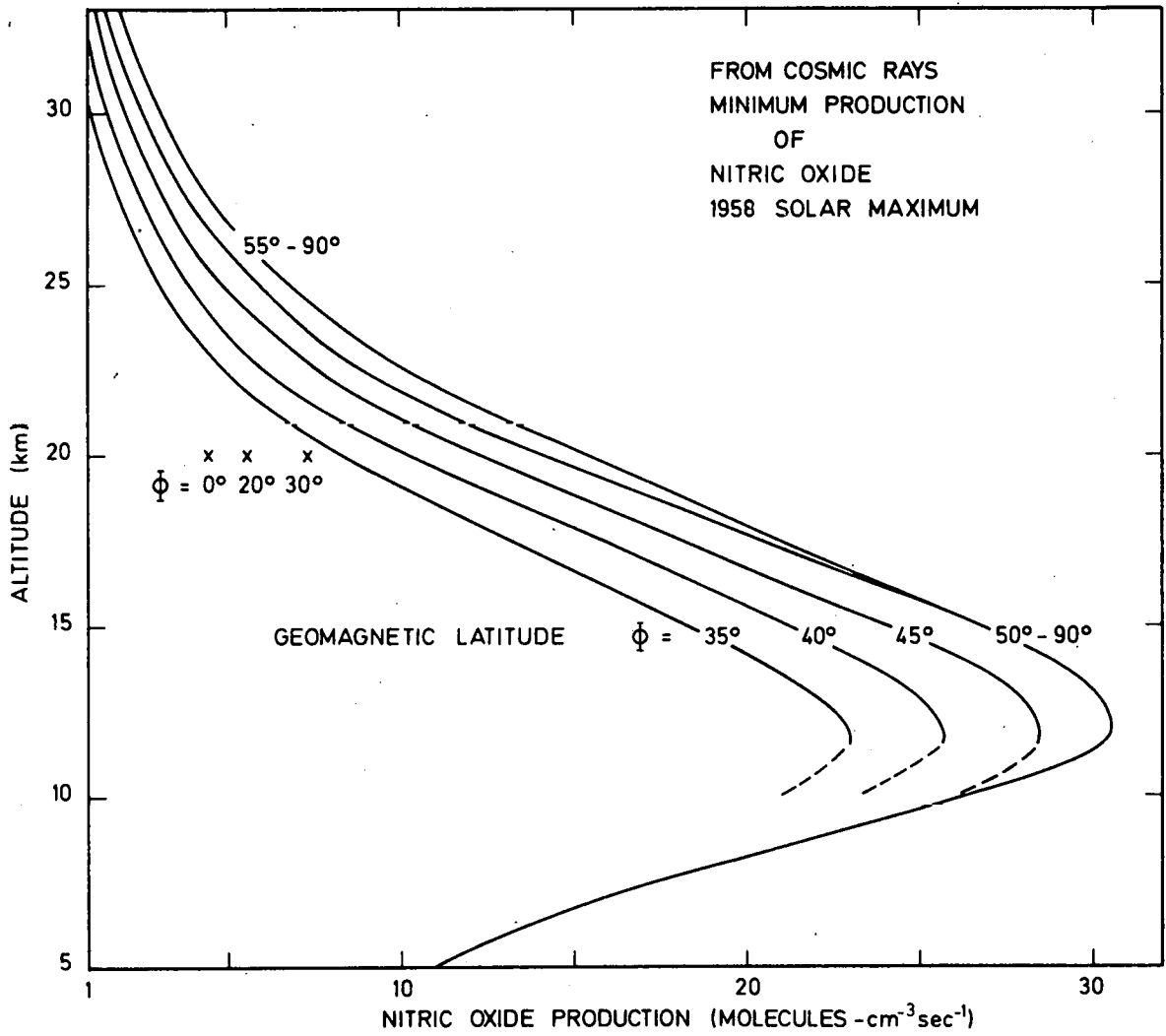


Fig. 8.- Production of nitric oxide during a maximum of solar activity. Cosmic ray data deduced from Neher (1961).



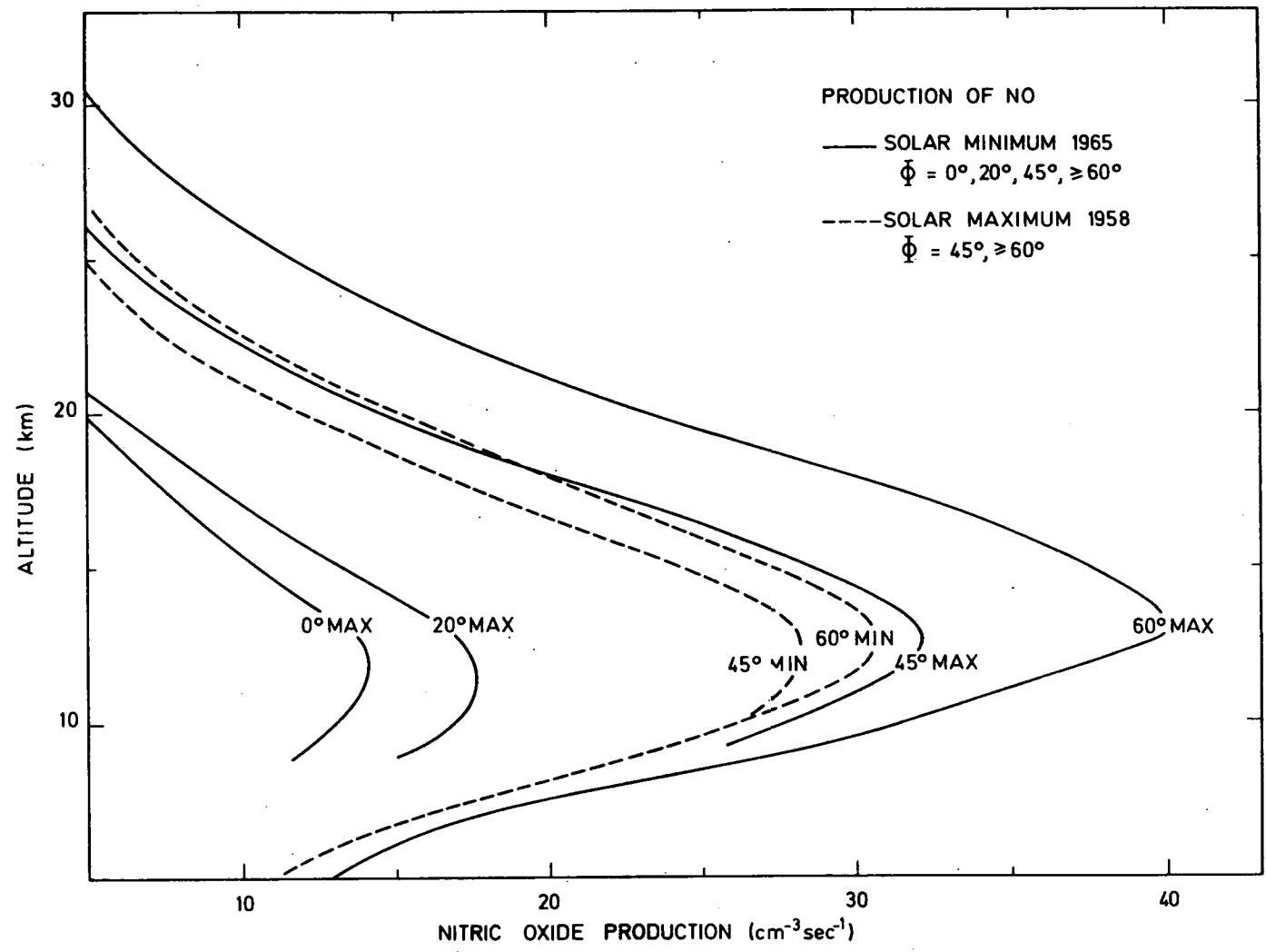


Fig. 9.- Comparison of the production of nitric oxide molecules for minimum and maximum solar activities in the equatorial region ( $\Phi \leq 20^\circ$ ), in the subauroral belt  $45^\circ \leq \Phi \leq 60^\circ$  and in the auroral region ( $\Phi \geq 60^\circ$ ).

to be considered. In the equatorial region ( $\Phi < 20^\circ$ ), the maximum production of nitric oxide is less than  $17.5 \text{ molecules cm}^{-3} \text{ sec}^{-1}$  at the peak. In the minauroral belt the increase is such that the minimum peak production is about  $28 \text{ NO molecules cm}^{-3} \text{ sec}^{-1}$ . It is clear that in the subauroral belt and the auroral region ( $\Phi > 45^\circ$ ) this NO formation rate is important ;  $35 \pm 5 \text{ molecules cm}^{-3} \text{ sec}^{-1}$  at the peak which represent an important source at lower stratospheric levels and may, therefore, be comparable with or larger than other sources since cosmic ray ionization also is present at night.

The ionization and dissociation of molecular nitrogen by cosmic rays provide (Fig. 10) a height-integrated input to the stratosphere of  $6 \times 10^7 \text{ NO molecules cm}^{-2} \text{ sec}^{-1}$  (day and night) in the auroral zone ( $\Phi \geq 60^\circ$ ) during minimum solar activity conditions and  $4 \times 10^7 \text{ NO molecules cm}^{-2} \text{ sec}^{-1}$  in the subauroral belt and auroral region ( $\Phi \geq 45^\circ$ ) during maximum solar activity conditions. The extension of the polar tropopause to geomagnetic latitudes of  $35^\circ$  inside of the minauroral belt leads (Fig. 10) to a NO production greater than  $2 \times 10^7 \text{ molecules cm}^{-2} \text{ sec}^{-1}$ . As far as the equatorial and tropical stratosphere is concerned, the production is less than  $10^7 \text{ NO molecules cm}^{-2} \text{ sec}^{-1}$  since the tropopause reaches an altitude of 17 km. At the tropopause break, the stratospheric production may increase by a large factor according to the synoptic situation. At mean latitudes it is well known that the tropopause height is very variable with no special concentration near the mean value.

## 6. CONCLUDING REMARKS

It may be concluded that the production of nitric oxide molecules by the ionization and dissociation of molecular nitrogen in the mesosphere and stratosphere is negligible in the mesosphere (NO production less  $1 \text{ cm}^{-3} \text{ sec}^{-1}$ ) but increases to a maximum in the lower stratosphere in the polar regions. The distribution of the ionization and dissociation of  $\text{N}_2$  by cosmic rays implies that the NO production is greater than  $10^7 \text{ molecules cm}^{-2} \text{ sec}^{-1}$  throughout the lower stratosphere for geomagnetic latitudes  $\Phi \geq 45^\circ$  (Fig. 10 from  $2 \times 10^7$  to  $6 \times 10^7 \text{ cm}^{-2} \text{ sec}^{-1}$ ). In the equatorial region  $\Phi < 20^\circ$  the NO production is small,  $3 \times 10^6$

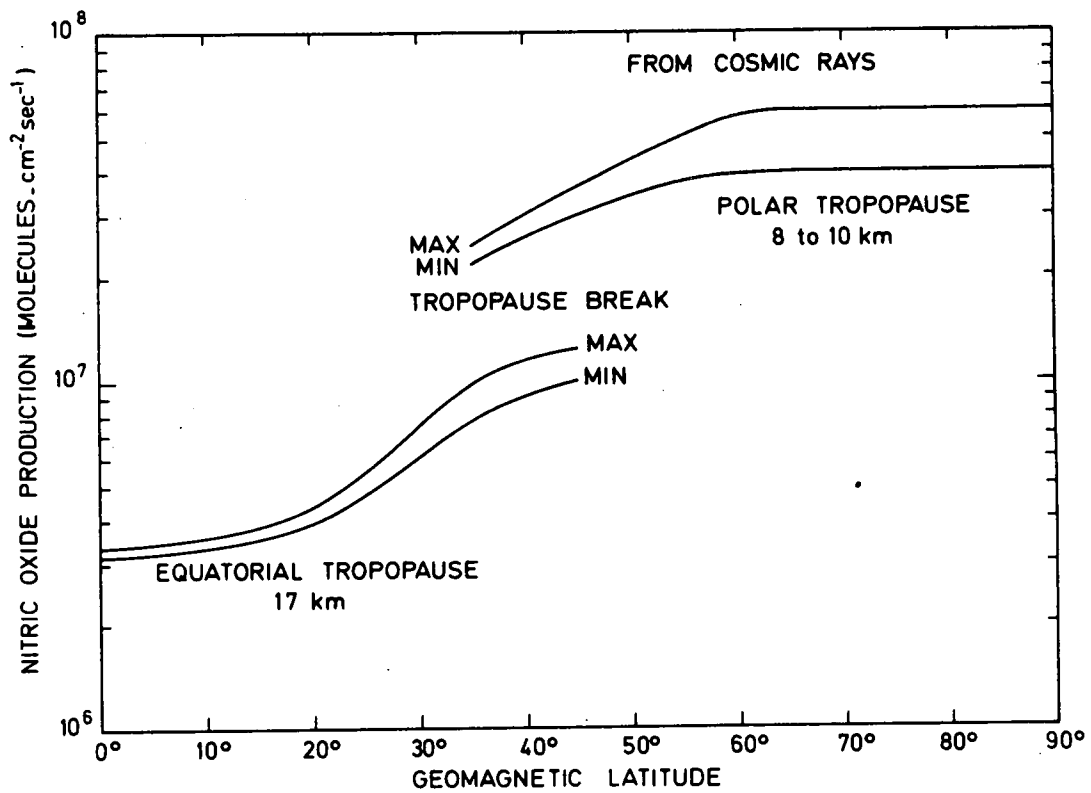


Fig. 10.- Production in the stratosphere of nitric oxide molecules ( $\text{cm}^{-2} \text{sec}^{-1}$ ) versus geomagnetic latitude for conditions of minimum ( $\text{NO}_{\text{max}}$ ) and maximum ( $\text{NO}_{\text{min}}$ ) solar activity.

molecules  $\text{cm}^{-2} \text{sec}^{-1}$ .

If the vertical distribution of the production of nitric oxide by cosmic rays (Fig. 11) and by the reaction of  $\text{O}(^1\text{D})$  with  $\text{N}_2\text{O}$  are compared it can be seen that production by cosmic rays is important below 20 km between  $45^\circ$  latitude and the pole. Since cosmic rays lead to a daytime and nighttime NO production, the process is particularly important during the winter at high latitudes. It is necessary that production of about  $5 \times 10^7$  NO molecules  $\text{cm}^{-2} \text{sec}^{-1}$  be introduced in the computation of stratospheric models and that correct lower boundary conditions be used.

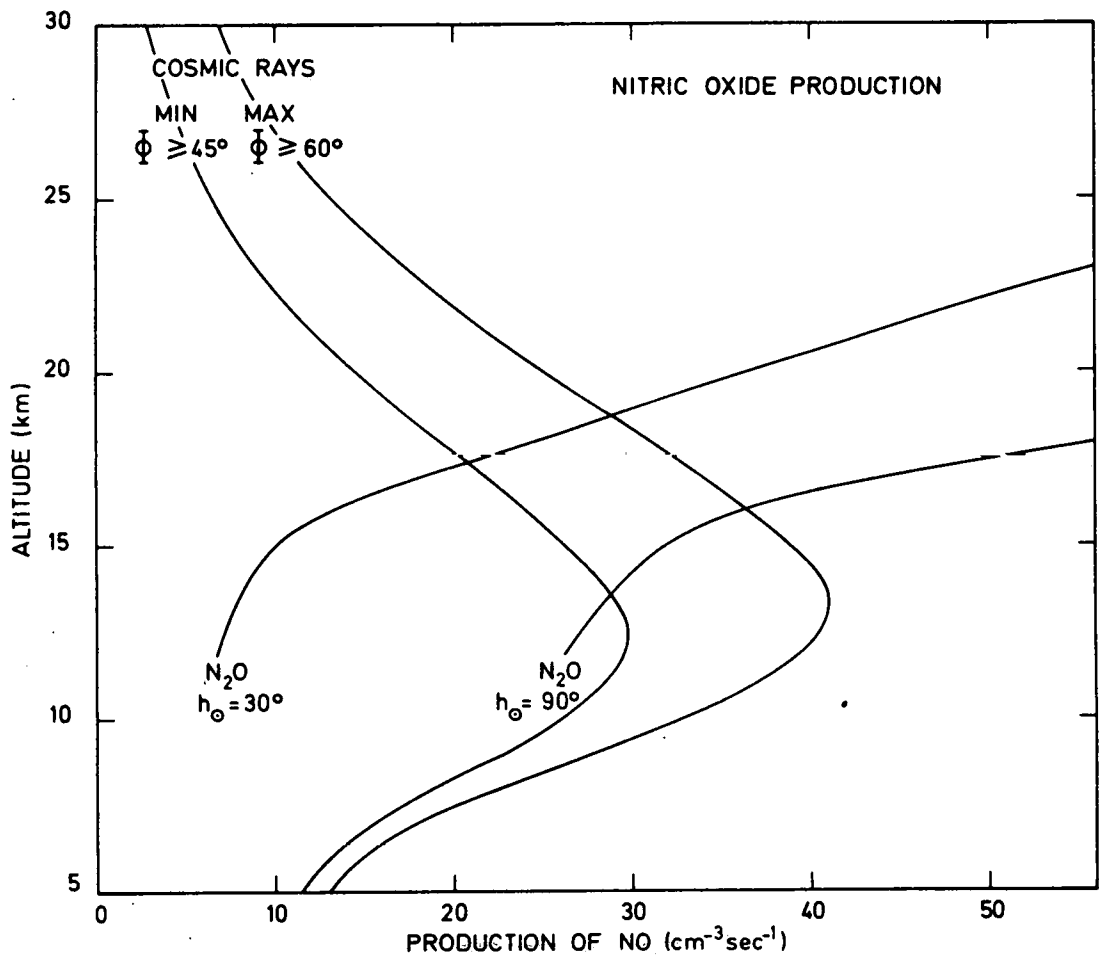


Fig. 11.- Comparison of the production of nitric oxide molecules in the lower stratosphere by cosmic ray ionization and dissociation of  $N_2$  and by oxydation of  $N_2O$ . The "cosmic ray curves" are given for maximum at minimum conditions at geomagnetic latitudes  $\Phi \geq 60^\circ$  and  $\Phi \geq 45^\circ$ , respectively. The " $N_2O$  curves" are given for an overhead sun  $h_\odot = 90^\circ$  and an average height  $h_\odot = 30^\circ$ .

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