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The Influence of Ionization Events on Atmospheric Ozone

Arthur C. Aikin

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Goddard Space Flight Center
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Arthur C. Aikin

NASA/Goddard Space Flight Center
Laboratory for Planetary Atmospheres
Greenbelt, Maryland 20771

Abstract

Atmospheric ionization events can modify the concentration of neutral species in the stratosphere and mesosphere. In particular ozone is destroyed because of the production of significant quantities of odd nitrogen and hydrogen compounds which react photochemically to destroy ozone. Direct evidence of ozone depletion has come from data taken during and following two solar flares generating large fluxes of 10-100 Mev protons, which bombarded the polar stratosphere and mesosphere. Observations of ozone taken during X-ray emission by solar flares and energetic electron precipitation during aurorae indicates ozone destruction above 50 km by ionization produced odd hydrogen. Lightning is apparently a large contributor to the tropospheric odd nitrogen budget. Production of odd nitrogen in the stratosphere due to lightning from thunderstorms that penetrate the stratosphere has not been evaluated. Ion propulsion induced dumping of the inner proton radiation belt represents a human activity which may influence stratospheric NO_x.

Introduction

This paper reviews the changes in atmospheric ozone which occur as a result of ionization. With the exception of lightning, ionization results from solar activity and cosmic radiation. Solar flares produce orders of magnitude change in solar X-radiation which is deposited between 50 and 100 km. Large flares are accompanied by fluxes of 10 to 100 MeV protons. Both X-radiation and protons destroy ozone above 50 km by inducing changes in the HO_x content. In addition solar protons induce changes in the total amount of odd nitrogen below 50 km where NO_x is an important component for establishing the ozone distribution.

Cosmic radiation is the dominant source of odd nitrogen in the region near the tropopause. The ionization rate varies by a factor of 10 with geomagnetic latitude and a factor of two with sunspot cycle with the larger flux occurring at solar minimum.

The contribution of lightning to the tropospheric NO, NO_2, HNO_3 and N_2O budgets is still unclear. There are a number of factors which contribute to this uncertainty. The present situation of this rapidly changing research area will be reviewed.

Recently Curtis and Grebowsky (1979) have suggested that argon ions resulting from ion propulsion systems of manned orbital transfer vehicles can indice dumping of the inner proton radiation belt. Deposition of these 10 to 500 MeV protons can alter the NO_x distribution. This is the first example of a man-induced ionization event.

Geomagnetic activity induces auroral zone electron precipitation. Ionization results from the direct energy loss of the energetic electrons as well as the accompanying bremsstrahlung. Visible and ultraviolet photon fluxes associated with such precipitation are probably too low to change the HO_x or NO_x balance on any large scale.

Mechanisms for the production of odd nitrogen and hydrogen by ionization

It was first suggested by Dalgarno (1967) that ionization could lead to modification of the neutral upper atmosphere. Maeda and Aikin (1968) suggested that mesospheric ozone would be modified by production of atomic oxygen during auroral events. Ionization by cosmic radiation and the subsequent ion neutral reactions which lead to the formation of NO was proposed by Warneck (1972), Brasseur and Nicolet (1973) and in more detail by Nicolet (1975). The production of NO during solar flare proton events has been discussed by Crutzen et al., (1975), Frederick (1976), Reagan (1977) and Reid et al., (1978).

Odd nitrogen is produced by several processes following the creation of the primary ion pair. In addition to formation of N_2^+ by direct ionization secondary electrons resulting from ionization dissociate N_2 directly to produce 2 N atoms, Winters (1967). Atomic nitrogen can also result from the dissociative ionization of N_2, that is

$$N_2 + e \rightarrow N + N^+ + 2e$$

Nicolet (1975) has estimated that these processes lead to 1 nitrogen atom for each ion pair/cm^3sec. Once formed N^+ is transformed into NO by the processes

$$N^+ + O_2 \rightarrow NO^+ + O$$

The ion NO^+ reacts through a number of paths to produce $H^+(H_2O)_n$ ions and NO so that each N atom created results in one NO molecule.

The rate of production of NO by the neutral reaction

$$N + O_2 \rightarrow NO + O$$

depends on the state of the N atom. The formation rate of NO from $N(^2D)$ is 10^5 times that of ground state $N(^3S)$. While the population of $N(^2D)$ is high above 100 km quenching should severely deplete this excited atom in the stratosphere. Atomic nitrogen and nitric oxide can be destroyed by the process

$$N + NO \rightarrow N_2 + O$$

This reaction is negligible under most circumstances so that once formed NO is only destroyed by the catalytic cycle involving the formation of nitric acid, which is precipitated into the troposphere.

The quantity of NO generated per ion pair has been estimated. Nicolet (1975) used 1 NO per ion pair, Crutzen et al., (1975) adopted 1.5 NO molecules per ion pair for the August 4, 1972 PCA event. Frederick (1976) assumes 1.27 NO per ion pair. This figure is in agreement with calculated values by Porter et al., (1976). Most recently Fabian et al., (1979) have adopted a value of 2-2.5 NO molecules per ion pair based on experimental measurements of NO produced during an aurora and their two-dimensional model fit to the August 4, 1972 PCA observations. The same ozone data has been fitted with a value of 1.5 molecules/ion pair by Reagan et al., (1978) using a time-dependent photochemical model.

Jackman et al., (1979) estimate the upper limit as 2.68 NO/ion pair for 10 KeV electrons. This estimate includes the effect of N_2^+ which is lost in the stratosphere by charge exchange to form O_2^+. When the contribution from N_2^+ is omitted the upper limit for the stratosphere is 1.5.

Above 50 km odd hydrogen is more important in determining the ozone distribution than odd nitrogen. Ionization can lead to odd hydrogen in the following manner. A majority of the ion formed initially are N_2^+. These ions are lost by charge transfer to O_2. Once formed O_2^+ ions react rapidly with water to yield species of the form $H^+ \cdot (H_2O)_n$. In the process OH is created by

$$O_2^+ \cdot (H_2O) + H_2O \rightarrow H_3O^+ + OH + O_2$$

which occurs 20 percent of the time; the more likely reaction being

$$O_2^+ \cdot (H_2O) + H_2O \rightarrow H_3O^+ \cdot OH + O_2$$

This reaction is followed by

$$H_3O^+ \cdot OH + H_2O \rightarrow H_3O^+ \cdot (H_2O) + OH$$

Several positive ion-negative ion recombination processes are capable of producing an odd hydrogen including

$$H_3O^+ \cdot (H_2O)_n + NO_3^- \rightarrow NO_2 + OH + (H_2O)_{n+1}$$

$$H_3O^+ \cdot (H_2O)_n + CO_3^- \rightarrow CO_2 + OH + (H_2O)_{n+1}$$

$$H_3O^+ \cdot (H_2O)_n + NO_2^- \rightarrow NO_2 + H + (H_2O)_{n+1}$$

$$H_3O^+ \cdot (H_2O)_n + O_2^- \rightarrow O_2 + H + (H_2O)_{n+1}$$

The net production of OH and H depends on the species of negative ion present. It is usually assumed that each ion pair leads to the production of 2 odd hydrogens, Swider and Keneshea (1973). Odd hydrogen influences the O_3

distribution only above 50 km where ionization can significantly modify the odd hydrogen budget. Below 50 km the loss of ozone is controlled by odd nitrogen.

Observations of PCA Events

The decrease in ozone following the August 4, 1972 solar flare was observed by the Nimbus 4 BUV instrument, Heath et al., (1977). Data for both hemispheres is illustrated in figure 1 which plots total ozone above 4 mb as a function of time. The winter hemisphere is normally disturbed by planetary wave propagation. This masks to some extent the influence of proton precipitation. Thus while ozone recovers within 20 days in the summer hemisphere the winter hemisphere still exhibits ozone depletion after more than one month.

Figure 2 illustrates computational comparisons with ozone data. While most of the effort is devoted to study of the August 4, 1972 event below 60 km, ozone measurements by Weeks et al., (1972) for the November 2, 1969 event are given together with theoretical predictions for the sunset period, Swider et al., (1978). There is a reasonable fit for the limited data with the greatest difficulty occurring at 70 km where the odd hydrogen ozone interaction rates are the most uncertain. Curve set B is the result of time dependent computations which do not include eddy diffusion, Reagan et al., (1978). The poor agreement at 0.5 mb is attributed to incomplete odd hydrogen.

Another comparison for the August 4, 1972 event is shown in 2C. A two-dimensional model is employed by Fabian et al., (1979). It is claimed that calculations by Crutzen et al., (1975), solid curves, will only fit the data if 2.5 NO molecules are produced for each ion pair cm^3 sec.

On the basis of these calculations there appear to be major differences on the efficiency of creating NO from ionization. Further the same data can apparently be fit with NO production values which differ by a factor

of 1.7. There is difficulty in matching theory with experiment at altitudes above 50 km. This is probably due in large measure to the quality and paucity of data in this region. However, the exact efficiency figure for odd hydrogen production from ionization may be unknown at present. A better understanding of neutral species changes occurring in the ionization of air is required together with more accurate measurements of O_3 and other species during a PCA. Simultaneous measurement of NO, O_3, and OH should be attempted.

The August 4, 1972 proton event was the largest such event in 25 years. Bauer (1978) has made a comparison of NO production during proton events, the results of which are exhibited in figure 3.

Other Sources of Ionizing Radiation

Cosmic rays, solar flare X-radiation and electron precipitation are additional ionization sources. Cosmic radiation is always present and varies by about a factor of two with solar cycle reaching a maximum at solar sunspot cycle minimum, see for example, Heaps (1978). Ruderman and Clamberlain (1975) and Ruderman et al., (1976) have suggested that ozone variations with solar cycle can be explained on the basis of NO variation produced by cosmic ray modulation. The effect would be more pronounced at high geomagnetic latitudes since the flux is a factor of 10 greater than at the geomagnetic equator. In the sunlit atmosphere the production of nitric oxide by cosmic radiation dominates NO production by the reaction of $O(^1D)$ with N_2O only below 20 km.

Solar flares are characterized by large enhancements in hard X-radiation emitted from the flare region during 10 to 15 minutes. The X-ray energy spectrum observed for large flares such as the August 4, 1972 produces 10^6 ion pairs at 60 km. This radiation source will only affect ozone above 50 km and the effect will be limited due to the short duration of the X-ray

pulse. Aikin and Maeda (1978) have presented evidence that such an effect occurs based on the Nimbus 4 BUV ozone data.

Energetic electrons are precipitated into the atmosphere largely in the auroral zone and can influence ozone Maeda and Aikin (1968). The flux and energy spectrum are the determining factors in the altitude region where ozone changes occur. It is usually assumed that the energy spectrum does not contain enough high energy electrons to create significant ionization below 65 km and that the accompanying bremsstrahlung must account for any effect below that altitude. However, Imhof et al., (1977) have presented evidence for higher energy electrons and discussed their implications for O_3 depletion. Calculations of the influence of relativistic electron bremstrahlung throughout the stratosphere has been carried out by Thorne (1978). Simultaneous measurement of energetic electrons, X-radiation and ozone during an auroral event has been conducted by Hilsenrath et al., (1978). A 25 percent decrease in O_3 above 1 mb is reported. Observations during solar flares and electron precipitation events will be useful in studying the recovery phase of ozone depletion above 50 km.

Lightning

Although the debate on the production of NO_x from lightning has been ongoing for many years, see for example Reiter and Reiter (1958), Ferguson and Libby (1971), it is only recently that it has been introduced into the problem of the global troposphere NO_x budget and its relation to the stratosphere, Dubin (1975), Zipf and Dubin (1976), Noxon (1976), Griffing (1977), Chameides et al., (1977) and Chameides (1979). There is great uncertainty concerning formation efficiency for NO, NO_2, HNO_3, and N_2O. Calculations as well as measurements during simulated and natural lightning have been used to estimate these efficiencies. Formation of NO_2 has been observed during

a lightning storm and the estimated NO_2 production is 10^{17} NO_2 molecules per joule or 60 ev is required for 1 NO_2 molecule with 10^9 joules per lightning stroke. Based on laboratory measurements with a spark chamber Chameides et al., (1977) estimate $6x10^{16}$ NO_x molecules per joule. These figures are in agreement with calculations by Griffing (1977). Using shock dissociation of air approximation, Chameides (1979) has calculated the NO_x molecule/joule yield dependence on input energy in joules/meter. He concludes that lightning accounts for a large fraction of tropospheric nitrogen oxides.

This conclusion is based on an energy dissipation of 10^5 joule meter^{-1}. However, a recent survey of lightning energy estimates by Hill (1979) concludes that an upper limit of only 10^4 joules meter $^{-1}$ is dissipated per stroke. This limits lightning odd nitrogen production to 10% of the value quoted by Chameides (1979). Recent satellite optical data, Turman (1978) indicates that power and distribution of lightning can be mapped globally. If agreement is reached on the percentage of total power is represented by optical power, such satellite experiments can reduce the undertainty in lightning odd nitrogen production estimates.

The production of N_2O and CO has also been considered. Laboratory studies by Levine et al., (1979) give $7x10^{12}$ molecules/joule for N_2O and $1x10^{14}$ molecules/joule for CO. Theoretical studies have concentrated on the reaction

$$O^+ + N_2 + M \rightarrow N_2O^+ + M$$

$$N_2O^+ + O_2 \rightarrow O_2^+ + N_2O$$

for the production of N_2O during lightning Griffing (1976). This reaction must compete with

$$O^+ + O_2 \rightarrow O_2^+ + O$$

so that more work is needed in the area of N_2O production from ionization events. Since dissociation and chemical reactions at elevated temperatures within the shock is the major factor in determining the amount of fixed nitrogen, neutral reactions which may be endothermic must be considered for example,

$$O + N_2 + M \rightarrow N_2O + M$$

$$NO + NO \rightarrow N_2O + O$$

and

$$CO_2 + M \rightarrow CO + O + M$$

The production of CO by lightning and particle ionization has been considered by Green et al., (1973). It is concluded that if an upper limits of lightning production of $2x10^8$ tons is assumed than lightning represents a significant source of CO. The long lifetime of N_2O allows transport to the stratosphere. In this way lightning may directly affect stratospheric odd nitrogen.

Conclusions and Recommendations

The types of natural events which give rise to changes in atmospheric composition have been identified and the mechanisms for species production is understood in a qualitative manner. A quantative understanding is required if the influence of such events on stratospheric ozone is to be properly evaluated. There is uncertainty concerning the amount of odd nitrogen produced by proton impact on the atmosphere. The influence of photochemistry and dynamics on recovery from such events is poorly understood. Better laboratory data on important parameters is required. Future measurements of PCA should measure NO and its temporal variations as well as O_3.

The amount of ionization as a function of time and altitude is needed in order to quantify energetic electron precipitation and solar flare X-ray events. The transient nature of these events can be used to evaluate recovery

of the mesosphere and upper stratosphere following such events. Neutral and ionized species measurements in addition to ozone should be carried out during such events.

Since the global lightning distribution can be measured from orbit, Turman (1978) better estimates of global lightning production of odd nitrogen should be possible. Continued experiments and theoretical work on the efficiency for odd nitrogen production are required.

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Figure Captions

Fig. 1 Zonally averaged total ozone above the 4-mb level during July-August 1972. The solar proton event occurred on 4 August (day 217). (a) at 70°N, (b) at 70°S and (c) at 6-latitude bands, (70°S, 60°S, 0°, 60°N, 70°N and 80°N). The vertical bars for each data point in (a) and (b) indicate the standard deviations of all data obtained in the latitude band of that day. Maeda and Heath (1978).

Fig. 2 Comparisons of computed and actual ozone variations during the PCA events of A) 29 November 1969, B and C) 4 August 1972. In part A solid curves represent time-dependent calculations for (1) 2 November and (2) 4 November. X represents experimental data. Part B Experimental and calculated ozone variations for 4 August 1972, 77°N latitude. Part C comparison of theory and experiment for 4 August 1972 using two values of NO production rate -- Crutzen et al --- Fabian et al.

Fig. 3 The total number of ion pairs produced by different ionizing events, including galactic cosmic rays, PCA's and relativistic electron precipitation.

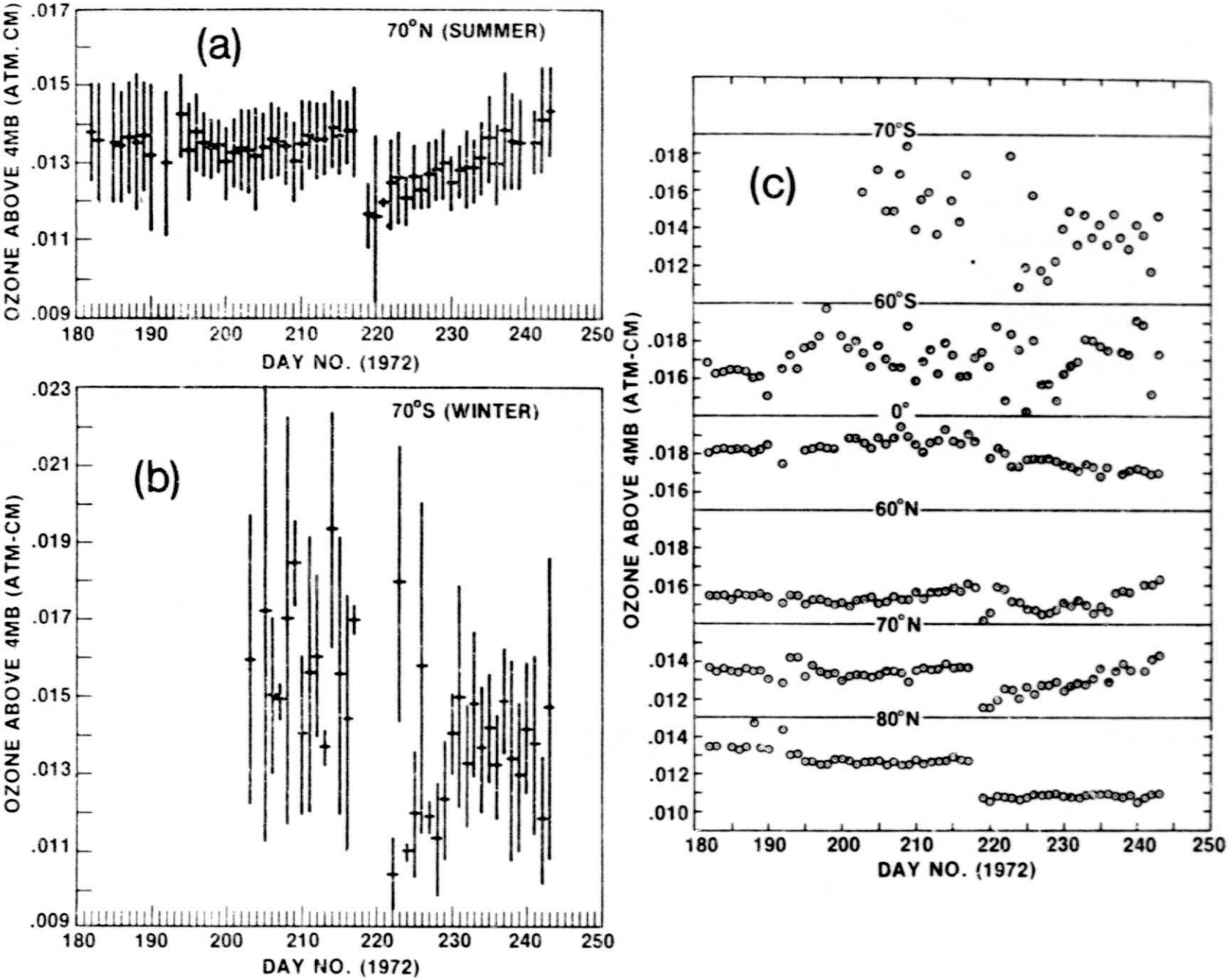
(a)
70°N (SUMMER)
OZONE ABOVE 4MB (ATM. CM)
.017
.015
.013
.011
.009
180
190
200
210
220
230
240
250
DAY NO. (1972)
(b)
70°S (WINTER)
OZONE ABOVE 4MB (ATM-CM)
.023
.021
.019
.017
.015
.013
.011
.009
180
190
200
210
220
230
240
250
DAY NO. (1972)
(c)
70°S
60°S
0°
60°N
70°N
80°N
OZONE ABOVE 4MB (ATM-CM)
.018
.016
.014
.012
.010
180
190
200
210
220
230
240
250
DAY NO. (1972)

Figure 1

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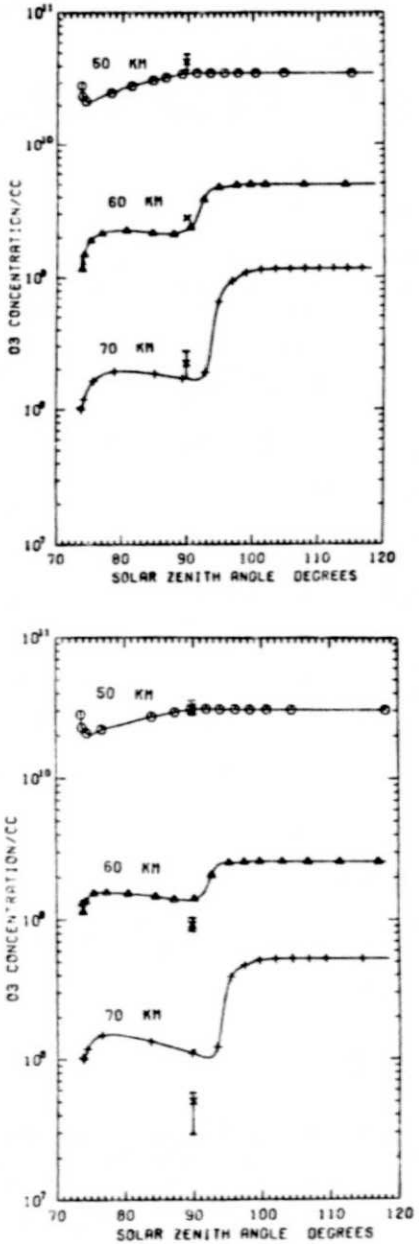

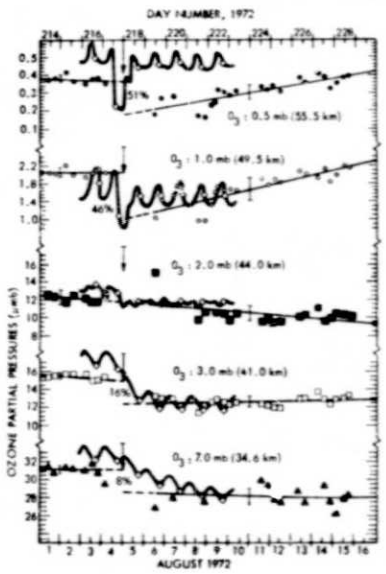

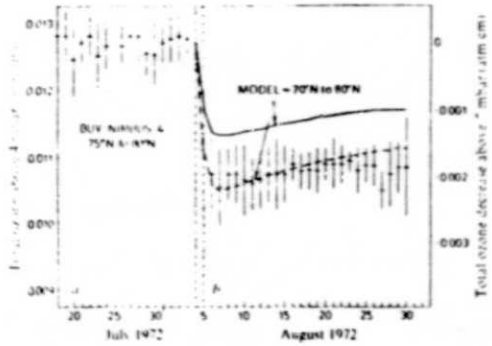

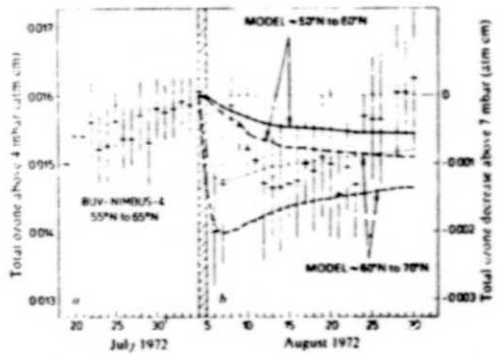

A B C

Figure 2

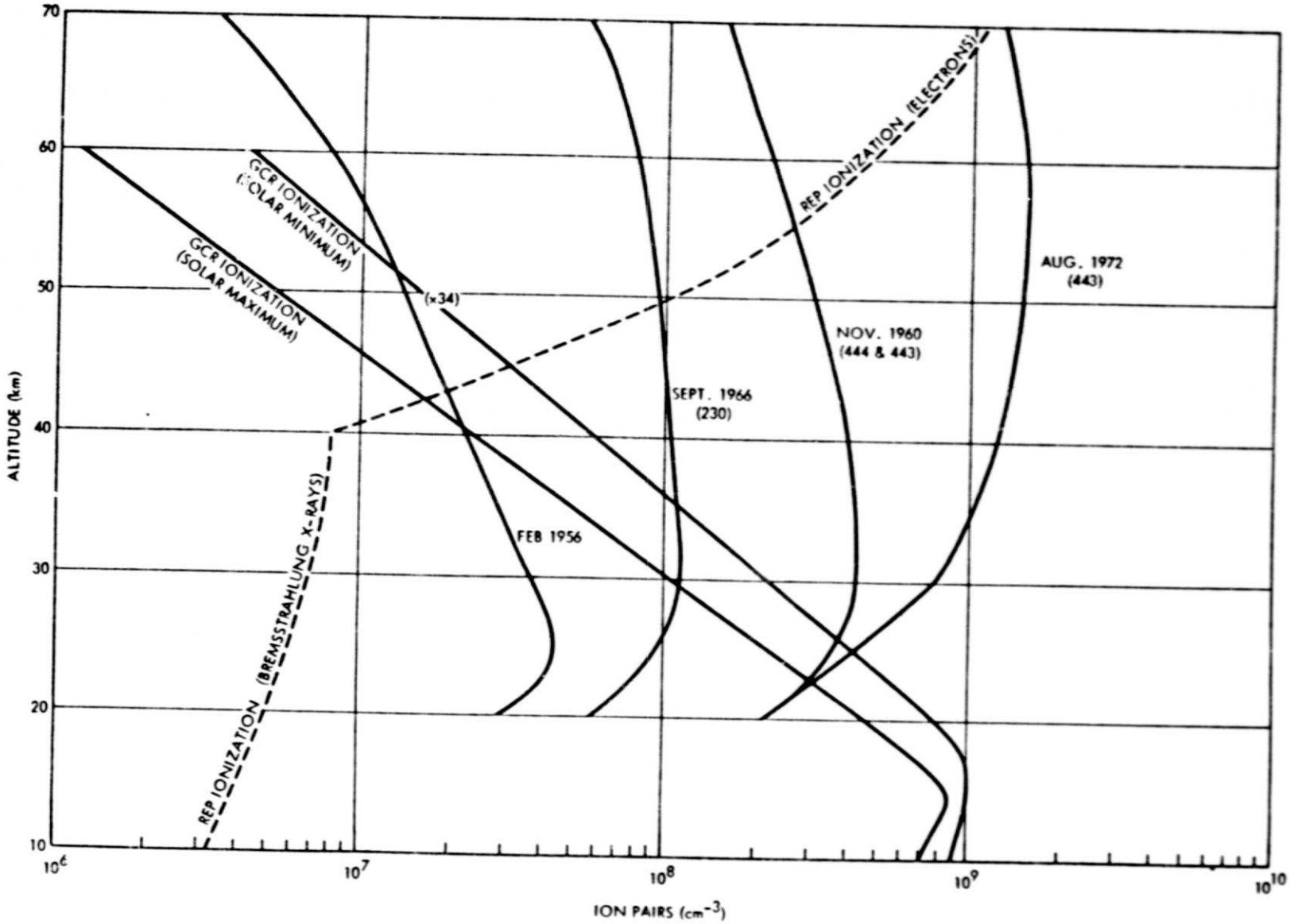
ALTITUDE (km)
70
60
50
40
30
20
10
10^6
10^7
10^8
10^9
10^{10}
ION PAIRS (cm^{-3})
GCR IONIZATION (SOLAR MINIMUM)
GCR IONIZATION (SOLAR MAXIMUM)
(×34)
REP IONIZATION (ELECTRONS)
REP IONIZATION (BREMSSTRAHLUNG X-RAYS)
FEB 1956
SEPT. 1966 (230)
NOV. 1960 (444 & 443)
AUG. 1972 (443)

Figure 3