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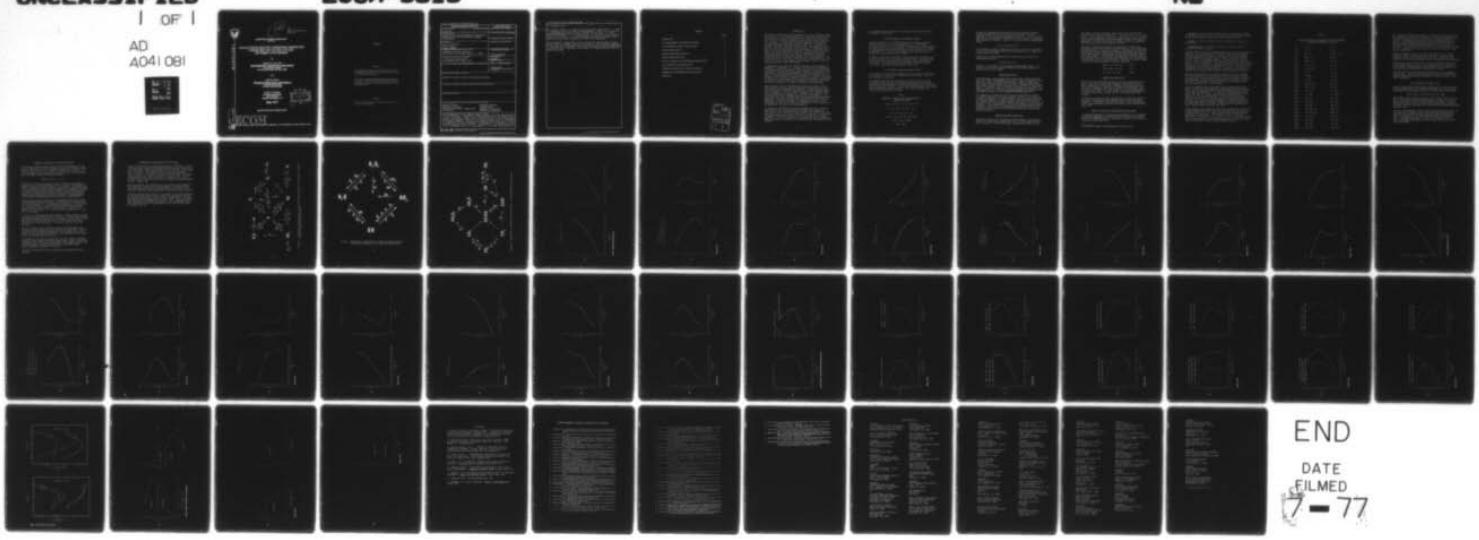

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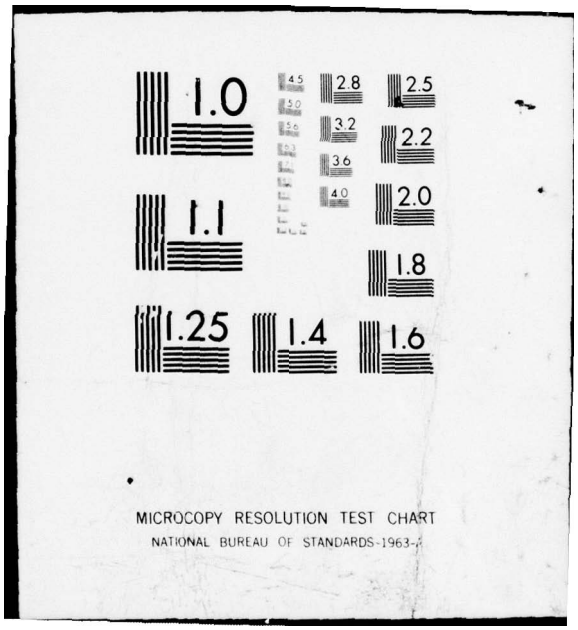
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RESEARCH AND DEVELOPMENT TECHNICAL REPORT
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CALCULATION OF SELECTED ATMOSPHERIC COMPOSITION PARAMETERS FOR THE MID-LATITUDE, SEPTEMBER STRATOSPHERE

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UNITED STATES ARMY ELECTRONICS COMMAND - FORT MONMOUTH, NEW JERSEY 07703

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Modeling the atmosphere

20. ABSTRACT *(Continue on reverse side if necessary and identify by block number)*

The stratosphere is a very complex, highly variable, critically important part of man's environment. Progress in gaining understanding of its characteristics, behavior, and potential modification is based principally on field measurements of necessary parameters. Because of the complexity and variability, a mechanism for assembling field data and supporting laboratory data in the context of the applicable physical laws is required. This mechanism is by numerical simulation of the stratosphere by use of a large computer. This "model" approximation to

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20. ABSTRACT (cont)

stratospheric behavior, in combining theory and actual measurements, is a powerful diagnostic tool in the study of the atmosphere. A model is very useful as an aid in developing both the general approach and the details of a field measurements program, and as an important tool in interpreting the experimental data of such a program. The interplay between measurements and model should produce the most effective approach to study of the stratosphere.

A few examples of calculational results from the ASL Numerical Model of Atmospheric Radiation (ANMAR), Composition and Dynamics, paralleling the conditions of the recent set of STRATCOM VI experiments, have been presented to demonstrate the range of results obtainable and the detail of treatment possible using the modeling approach.

CONTENTS

INTRODUCTION

The chemical composition of the stratosphere is an ever-changing, highly variable result of the complex interplay of solar-induced photodissociation processes, resultant photochemical and thermochemical reactions, and a variety of transport processes on space scales ranging from molecular to global. The magnitude or relative importance of any process in determining composition and atmospheric behavior is dependent on altitude, latitude, time-of-day, season-of-year, and other variables. Because of the great complexity and variability, the fundamental requirement for gaining understanding of the stratosphere is measurement of a number of critical atmospheric parameters, including particle densities of key trace constituents, solar flux and thermodynamic properties. These measurements in themselves, however, do not provide explicit information of the processes which produced the results observed. They allow prediction of the state or behavior of the stratosphere only in a statistical sense if large numbers of measurements, under a variety of time and space conditions, are obtained.

A supplemental (and necessary) parallel approach is the development of mathematical simulations, i.e., "models," of stratospheric composition and behavior based on the physical laws which apply. Field measurements and theoretical speculation are the usual modes of motivation for model development, and laboratory measurements provide the quantization. A model is modified as required by the accumulation of relevant field measurements; and ultimately the degree-of-validity of a model is determined by comparison of calculated results with actual atmospheric measurements. The relative validity establishes the degree of confidence that can be placed on the diagnostic or predictive capabilities of the model.

During the past 8 years, the Atmospheric Sciences Laboratory (ASL) at White Sands Missile Range, NM, has developed a large, complex program of balloon-based stratospheric measurements in cooperation with several other laboratories. One of the central themes of this STRATCOM (STRATospheric COMposition) program [1] is simultaneous measurement of sets of related composition, thermodynamic and radiative parameters. Such measurement of related parameters under the same conditions allows direct, rather than statistical, analysis and interpretation of relationships. Development, testing and validation of computer-based models of the atmosphere can also be more direct, and confidence in their capabilities is enhanced by relating to actual measurements.

To support and supplement this field measurements program, a chemical-kinetic model of the stratosphere [2], incorporating a parametric application of the vertical transport processes, has recently been made operational at ASL. The model is used as one of the tools in developing the overall measurements program, as well as in designing individual experiments. It will also be used as an exploratory tool in theoretical studies to extend the capability of interpreting the experimental results obtained.

This document presents examples of the kinds of calculational results obtainable from the model.

FIELD MEASUREMENTS AND ATMOSPHERIC MODELS

A recent operation in the STRATCOM atmospheric measurements program, STRATCOM VI [3] was held in late September 1975. The principal set of measurements was in the altitude range of 25-39 kilometers during a 34-hour period on 24 and 25 September. A supporting set of IR absorption measurements was made from an altitude of 31 kilometers at sunset, 26 September 1975, using a second balloon. Both flights were from Holloman Air Force Base, NM, 32° N latitude.

To parallel these field measurements, the calculations, from which the following sample results are taken, were made by use of photolytic data derived from solar conditions for the latitude, dates, and altitudes given above. The photodissociation coefficients were calculated by J. L. Collins, and are presented and discussed in [4].

THE ATMOSPHERIC CHEMICAL-KINETICS MODEL

The structure and calculational method used in the computer simulation of stratospheric composition is discussed in greater detail in [2]. A brief summary is given here.

The computer model uses the Gear method of solution of sets of stiff differential equations [5] to solve the coupled continuity equations describing the time dependence of the particle densities of 30 chemically reactive atmospheric species. (Oxygen and nitrogen molecules are included with suitable steady-state densities.) The atoms, molecules, and radicals treated are given in Table 1.

TABLE 1

ATMOSPHERIC CHEMICAL SPECIES CONSIDERED IN COMPUTATIONAL MODEL

O $O(^1D)$ $O(^1S)$ O_2 $O_2(^1\Delta)$ $O_2(^1\Sigma)$ O_3

N N_2 NO NO_2 N_2O NO_3 N_2O_5

H H_2 OH H_2O HO_2 H_2O_2

CO CO_2 CH_2 CH_3 CH_4 CHO $HCHO$

CH_3O CH_3O_2 CH_3OOH

HNO_2 HNO_3

A total of 34 photodissociative processes and 115 chemical reactions, coupling the densities of the species, are considered. These are tabulated in [2] and partially sketched here in Figs. 1, 2, and 3, which show families of atmospheric constituents and the processes of conversion from one species to another. Figure 1 is for part of the nitrogen/oxygen family:

$$O,\ O_2,\ O_3,\ NO,\ NO_2,\ N_2O.$$

In the diagram, a chemical species at the tail of an arrow is converted to the one at the head by interaction with any species, or a photon ($h\nu$) listed on the arrow shaft.

Figure 2 is a similar representation of some of the reactions of the oxygen/hydrogen family:

$$OH,\ H_2O,\ HO_2,\ H_2O_2$$

Figure 3 is a limited set for the carbon/hydrogen/oxygen species. All reactions are not shown in these diagrams, and reactions linking the families are not all included.

COMPUTATIONAL RESULTS

The calculations, which produced the selected sample results presented below, were fully time-dependent and diurnal. Many calculations of this type use a number of equilibrium relationships and/or a steady-state solar input, and are therefore less realistic. Although the calculations were diurnal, and the results obtained are available for any time (day, night, or transition), only the results for noontime are presented. The following figures are therefore the model-derived characteristics of the stratosphere at noon, in late September at latitudes near 32° N.

The categories of calculated results chosen for presentation include: (1) particle densities of selected important species, (2) mixing ratios of these constituents, (3) rates of several sets of important photolytic and chemical reactions, (4) total formation and removal rates of some key species, and (5) equivalent transport contribution to a few molecular densities. All are presented as a function of altitude between 10 and 50 kilometers. Other types of information can be derived from the model, and equivalent results for other latitudes and periods of the year are readily calculable.

COMPOSITION-PARTICLE DENSITIES

Figures 4a to 4k are the calculated altitude profiles of the particle densities of ground-state oxygen atoms O (^3P), excited oxygen molecules $O_2(^1\Delta)$, ozone (O_3), nitric oxide (NO), nitrogen dioxide (NO_2), nitric

acid (HNO_3), nitrous oxide (N_2O), water (H_2O), carbon monoxide (CO), hydroxyl radical (OH) and methane (CH_4). (Units are cm^{-3}.) All of these species are of fundamental importance in stratospheric chemical kinetics, and all have been measured in the stratosphere. However, the number of measurements is very limited except for ozone and water, and even with these two, variability and uncertainty are high.

Figures 4ℓ to 4p are calculated particle densities of some of the speculative species which have not yet been detected in the stratosphere, but whose presence is required by photolytic and chemical considerations. These species include electronically excited oxygen atoms $O(^1D)$, perhydroxyl radicals (HO_2), hydrogen peroxide (H_2O_2), formaldehyde (HCHO) and nitrous acid (HNO_2). In the absence of measurements, the only information available on such constituents comes from models and related theory. In particular, the $O(^1D)$ atoms and the HO_2 radicals are involved in some critically important atmospheric reactions, including:

$$O(^1D) + H_2O \rightarrow OH + OH \qquad (73)^*$$

$$O(^1D) + N_2O \rightarrow NO + NO \qquad (71)$$

$$HO_2 + OH \rightarrow O_2 + H_2O \qquad (132)$$

COMPOSITION-MIXING RATIOS

Figures 5a to 5p present the same data as 4a to 4p expressed as mixing ratios: the particle density of the species divided by the total particle density of all the species present in the volume considered. This ratio of numbers of particles is termed the volume mixing ratio. The ratio of the masses (mass mixing ratio) is also frequently used, particularly for water. In these figures, the abscissa designation 10^{-6} is one part per million (1 ppmv), and 10^{-9} is one part per billion (1 ppbv).

The figures provide no new basic information, but allow direct consideration of the fraction of the atmosphere that a given constituent represents. They emphasize the fact that species which play dominant roles in many atmospheric processes are present in only extremely small proportions.

RATES OF CHEMICAL AND PHOTODISSOCIATION REACTIONS

To understand atmospheric characteristics and behavior, it is necessary to know what processes are occurring and the role and relative importance of each under the range of conditions encountered. As indicated previously, the processes are:

*The reaction numbers used throughout are as used in [2].

Radiative - concerned with the transmission, scattering, absorption and emission of energy in the infrared, visible, and ultraviolet regions, and the thermodynamic consequences on atomic/molecular scales.

Dynamic - covering the range of particle motions from molecular to global scales.

Chemical Kinetic - treating the multitude of chemical reactions among the many species present.

A useful method of deriving and demonstrating the role of various chemical reactions is to calculate the rate at which a given reaction proceeds under the conditions considered. Figures 6a to 6n are the calculated altitude dependencies of the rates of a number of important reactions for the noon, 32° N, September situation. The reactions are listed in Table 2. Some of the reactions are grouped to allow direct comparison of certain critical relationships. Since the relevance or role of a specific reaction or set of reactions is dependent on the particular problem under consideration, the following discussion will not be in the context of stratospheric pollution, weapons effects, meteorology, or other specific present concerns, but will be limited to a few brief general remarks.

Figure 6a shows the rates for the most important atmospheric reaction: the initial production of oxygen atoms by photodissociation of oxygen molecules, which sets the stage for most of the stratospheric chemistry of consequence. It should be noted how rapidly this photolysis falls off below 25 kilometers altitude - the rate at 10 kilometers is less than one thousandth that at 25 kilometers.

Figure 6b represents the two principal modes of photodissociation of ozone. In reaction 5 the products are ground-state atoms and molecules, while reaction 7 produces electronically excited (and more reactive) atoms $O(^1D)$ and molecules $O_2(^1\Delta)$. Reaction 5 dominates below 30 kilometers, and reaction 7 above. Ozone formation (reactions 36 + 37) is shown in Fig. 6c; and the comparison of this three-body formation rate with the photolytic destruction (the sum of reactions 5 and 7) is presented in Fig. 6d. It should be noted that these reaction rates are very large compared with other atmospheric reactions and that they are almost equal at all altitudes, so that an equilibrium among O, O_2 and O_3 closely follows the solar flux input.

Three important ways in which the oxygen atoms (produced in Fig. 6a and brought somewhat into equilibrium in Fig. 6d) enter into the chemistry is demonstrated in Fig. 6e. These are three of the principal removal mechanisms of oxygen atoms. The removal of O and O_3 by the much-publicized NO_x "catalytic" cycle is shown in Fig. 6f. The $O+NO_2$ reaction (43) is much slower over this altitude range and is therefore the rate-controlling reaction for this "ozone depletion" process.

TABLE 2

REACTIONS SELECTED TO ILLUSTRATE ALTITUDE-DEPENDENCE OF RATES FOR FIGURES OF SECTION 3

(1)	$O_2 + h\nu$	→	$O + O$
(5)	$O_3 + h\nu$	→	$O + O_2$
(7)	$O_3 + h\nu$	→	$O(^1D) + O_2(^1\Delta)$
(11)	$NO_2 + h\nu$	→	$O + NO$
(32)	$HNO_3 + h\nu$	→	$OH + NO_2$
(36)	$O + O_2 + O_2$	→	$O_3 + O_2$
(37)	$O + O_2 + N_2$	→	$O_3 + N_2$
(38)	$O + O_3$	→	$O_2 + O_2$
(43)	$O + NO_2$	→	$O_2 + NO$
(50)	$O + HO_2$	→	$O_2 + OH$
(71)	$O(^1D) + N_2O$	→	$NO + NO$
(73)	$O(^1D) + H_2O$	→	$OH + OH$
(94)	$O_3 + NO$	→	$O_2 + NO_2$
(95)	$O_3 + NO$	→	$O_2 + NO_2^*$
(101)	$O_3 + OH$	→	$O_2 + HO_2$
(115)	$NO + NO_3$	→	$NO_2 + NO_2$
(117)	$NO + HO_2$	→	$OH + NO_2$
(119)	$NO + HO_2 + M$	→	$HNO_3 + M$
(123)	$OH + NO_2 + M$	→	$HNO_3 + M$
(126)	$H + O_2 + M$	→	$HO_2 + M$
(132)	$OH + HO_2$	→	$O_2 + H_2O$
(134)	$OH + CO$	→	$H + CO_2$
(135)	$OH + CH_4$	→	$H_2O + CH_3$

Also important in the NO/O_3 problem, and the utlimate controlling factor when combined with HNO_3 dynamic removal, is the three-body formation and photolytic loss of nitric acid in reactions 123 and 32 as plotted in Fig. 6g. The balance among formation, loss, and transport removal of HNO_3 will help establish the NO_2 density since it will remove NO_2 from the equilibrium concentration that the reactions in Fig. 6h tend to establish. These are the rates for formation of NO_2 by the $NO+O_3$ reaction and for its photodissociative destruction. Below 35 kilometers, these are very nearly equal.

Figure 6i compares the $NO+O_3$ rate with other important nitric oxide reactions, showing its dominance at all altitudes. Figure 6j demonstrates the role of the excited oxygen atoms $O(^1D)$ produced by ozone photolysis as the principal primary source of OH radicals and NO molecules.

Figure 6k gives the rates for three of the most important reactions of these hydroxyl radicals, while Fig. 6ℓ shows their role in destruction of the natural and anthropogenic carbon monoxide (CO) and methane (CH_4) that move into the stratosphere from the earth's surface.

Figure 6m shows the principal formation mechanisms of the perhydroxyl radical (HO_2). Figure 6n compares formation of nitric acid from NO and from NO_2 showing the potential importance of the OH/NO_2 process in the lower stratosphere.

TOTAL FORMATION AND REMOVAL RATES

Figures 7a and 7b show the calculated noontime, 32° N latitude, September altitude dependencies of the formation and removal rates, by all applicable chemical and photolytic processes, for eight minor constituents:

$$O(^3P),\ O(^1D),\ O_3,\ NO,\ NO_2,\ OH,\ HO_2 \text{ and } HNO_3$$

The formation rate is essentially equal to the removal rate for all but HNO_3 in this near-equilibrium noontime situation. Hence, for all but HNO_3 only one curve is seen for both formation and removal. This was implied for ozone in Fig. 6d since the entire production of ozone is by reactions 36 and 37, and almost all of the loss is by the photodissociation reactions 5 and 7.

Figure 7a also demonstrates that oxygen atom formation-removal rates are virtually equal to those of ozone in the altitude range considered. Similarly the NO and NO_2 pair have essentially equal production-loss rates. The hydroxyl radical (OH) formation-removal rates shown in Fig. 7b are approximately equal to those of the perhydroxyl radical (HO_2) in the lower stratosphere, but are a small factor larger in the upper stratosphere.

TRANSPORT CONTRIBUTION TO PARTICLE DENSITIES

The continuity equations used to describe the time dependencies of the particle densities (n_i) of the atmospheric chemical constituents contain chemical formation terms (F_i), chemical and photolytic loss terms (L_i), and a divergence term to describe the net gain or loss of the particles due to transport through the volume considered:

$$\frac{d}{dt}[n_i] = F_i - L_i - \nabla_p \bar{v}$$

The effect of the great variety of transport processes on atmospheric composition is very poorly understood, and is usually incorporated into models in somewhat artificial fashions. One purpose of the present model is to reverse this procedure, since the photolytic and chemical processes have been much better characterized, and derive mean characteristics of the vertical contribution to transport using atmospheric measurements combined with detailed chemical kinetic treatment. A preliminary part of this study is presented partially and briefly in Figs. 8a to 8e.

These figures show the regions of the stratosphere where the net result of the transport through the volume is an increase of the particle density (points plotted +) or a decrease (points plotted -). Alternatively, this can be described as a region where the chemical/photolytic loss rate is greater than the chemical formation rate (+), or where the formation rate is greater (-). The constituents considered are O_3, NO_2, N_2O, H_2O, and HNO_3.

In Fig. 8a, in the region above 30 kilometers altitude, removal of ozone by photolysis and reaction with NO, O, OH, etc., is faster than the three-body chemical formation, and ozone is transported into the region. Below the transition at 30 kilometers, there is a net formation and ozone transport is out from the region. The units of net loss or gain are $cm^{-3}\ sec^{-1}$.

For NO_2 (Fig. 8b), excess formation is above 17-19 kilometers, with deficient formation below and a net flow into the lower region. Nitrous oxide (N_2O) has a net chemical/photolytic loss throughout the stratosphere, and the equivalent contribution to the N_2O density by the upward flow is at the rates shown in Fig. 8c.

The reverse of this occurs for water (Fig. 8d) with chemical formation exceeding losses through the 10-50 kilometer range. Under the conditions considered, nitric acid losses dominate above 20 kilometers, with a flow upward and downward from the 10-20 kilometer region where there is excess formation, as indicated in Fig. 8e.

The results presented in Figs. 8a through 8e are exploratory and preliminary.

COMPARISON OF MEASUREMENTS AND CALCULATIONS

Results of some of the experimental measurements on STRATCOM VI reported in [3] have been plotted on the graphs of calculated particle densities and mixing ratios. These include ozone (Figs. 4c and 5c), nitrous oxide (Figs. 4g and 5g), carbon monoxide (Figs. 4j and 5j) and methane (Figs. 4k and 5k). It should be noted that since the model uses mean values as inputs, the results are plotted as smooth curves. The actual stratosphere can have sharp variations of a parameter over a small altitude interval, and an equivalent plot of measured altitude dependence could be a jagged line.

However, even with this consideration, the relatively good agreement of the computational results with the measurements for the given season, time, latitude, and altitude, gives some degree of confidence in the validity of the model and, consequently, in the other results calculated.

Similarly, some earlier results of J. G. Anderson, at the same latitude, for the short-lived highly reactive oxygen atoms and hydroxyl radicals are plotted comparatively in Figs. 4a and 4i, respectively. The oxygen atom results are for November 1974, 10:30 a.m. [6]. The length of the line indicates the experimental uncertainty. For the hydroxyl radical (OH), the dotted line is for measurements in July and the solid lines for January 1976 [7].

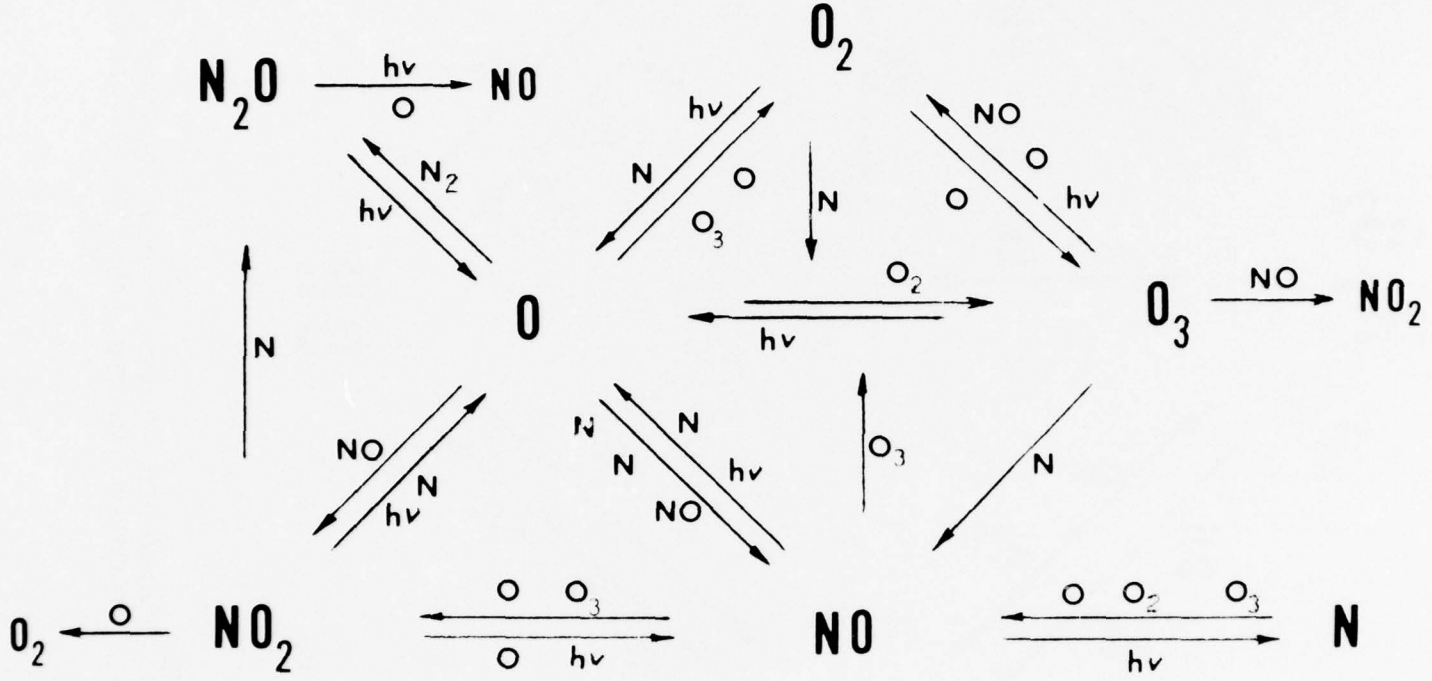


Figure 1. Diagrammatic representation of reactions among atmospheric constituents composed only of oxygen and nitrogen atoms.

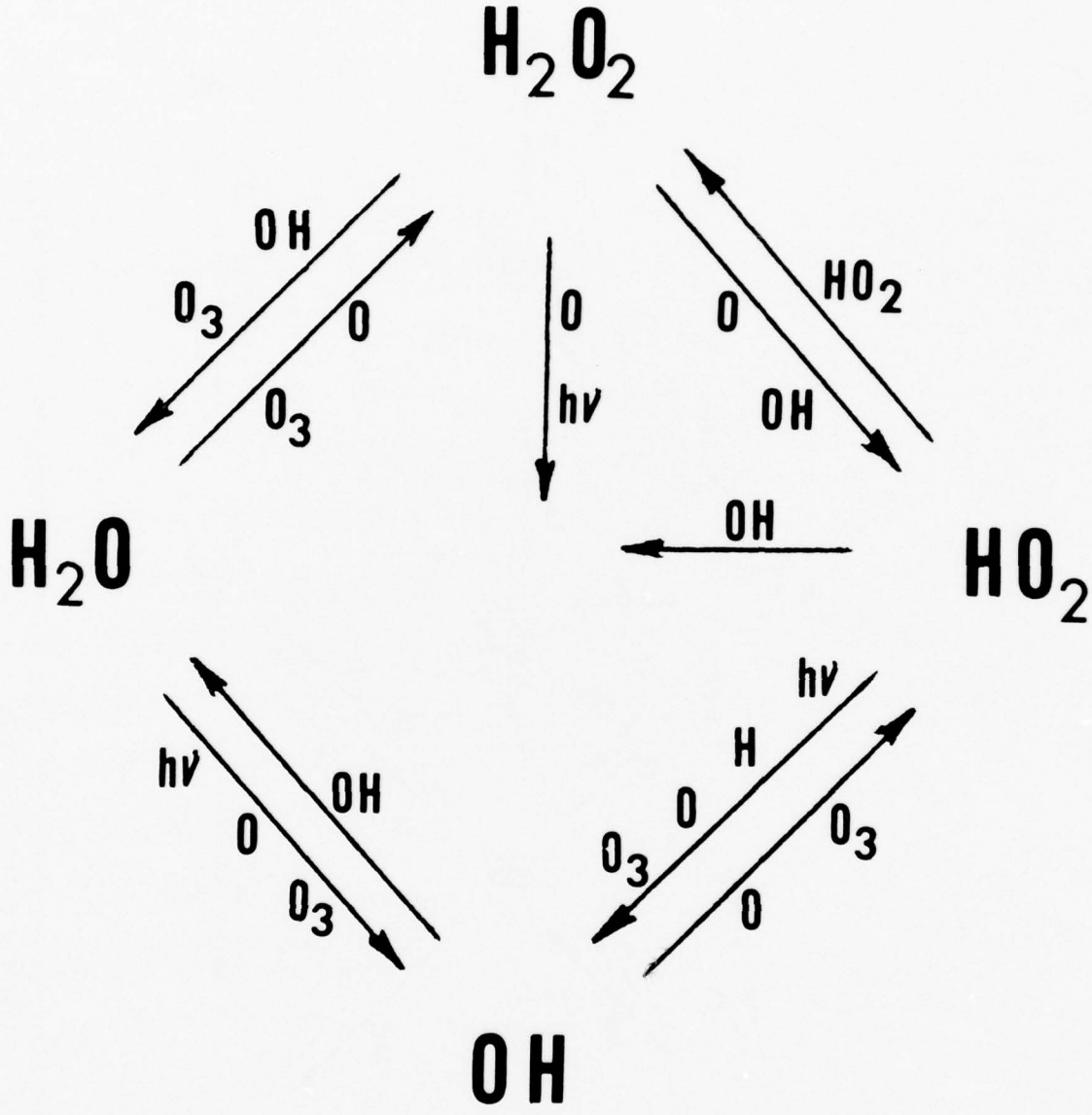


Figure 2. Diagrammatic representation of reactions among atmospheric constituents composed only of oxygen and hydrogen atoms.

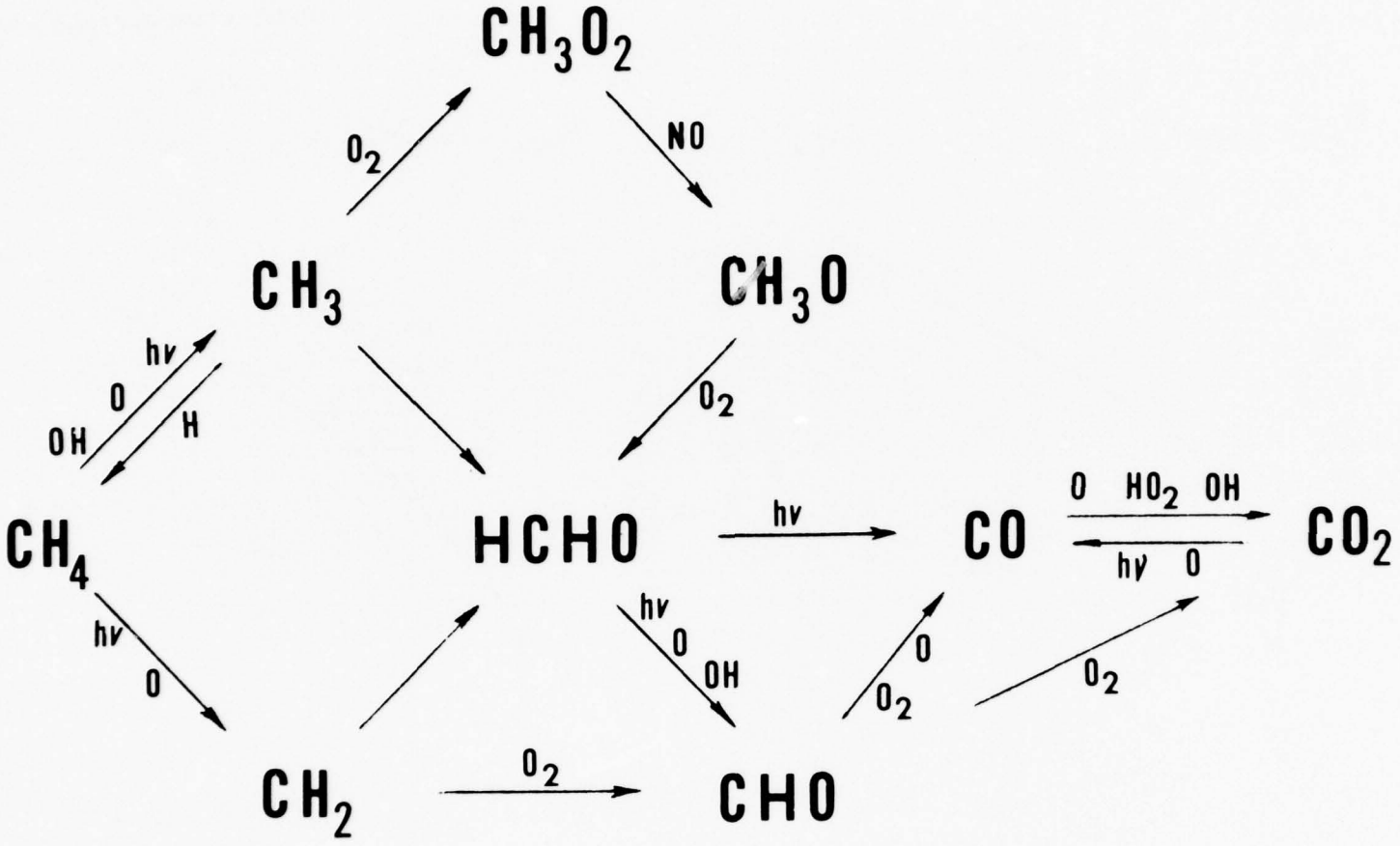


Figure 3. Diagrammatic representations of reactions of carbon/hydrogen/oxygen species.

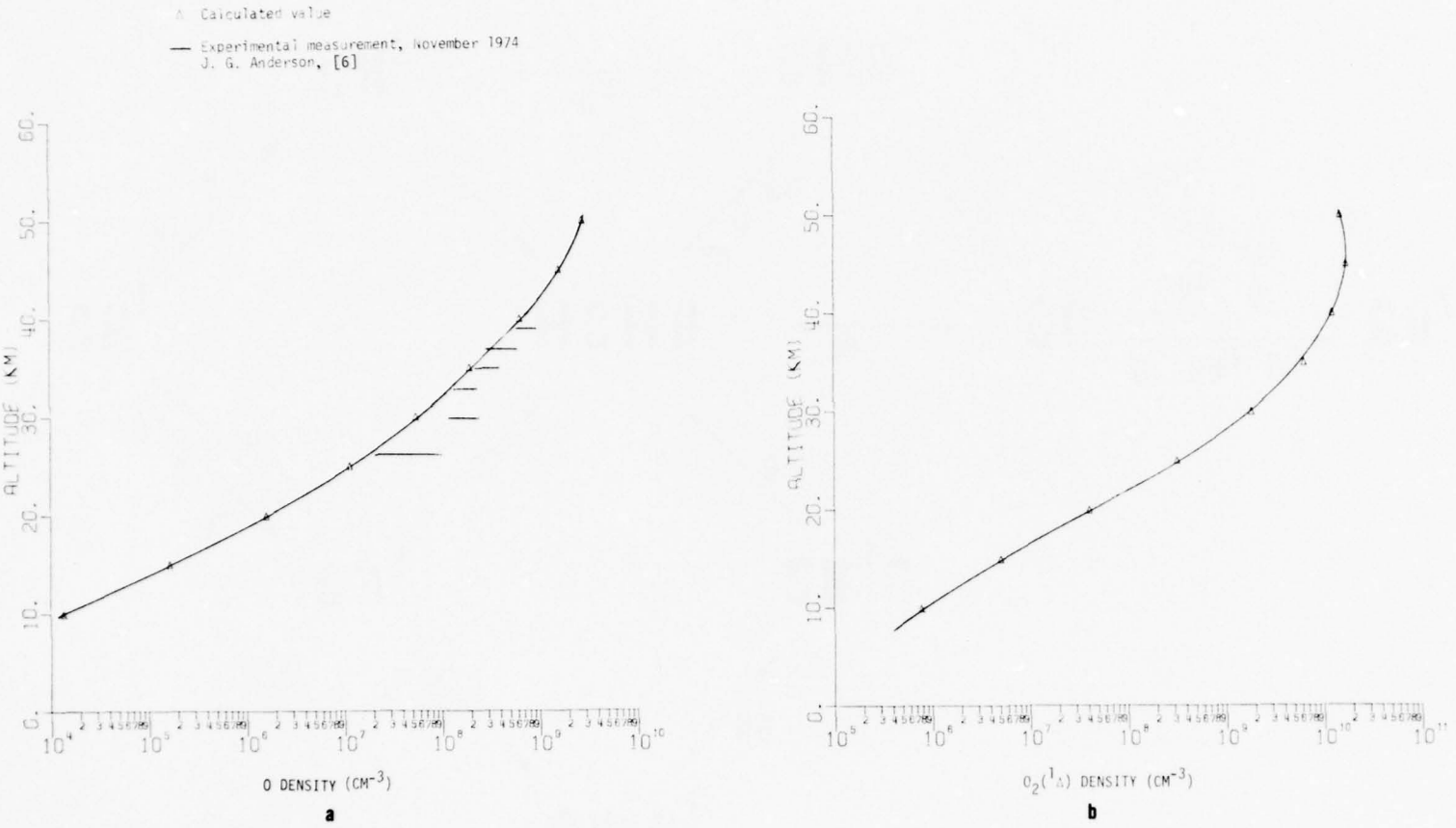


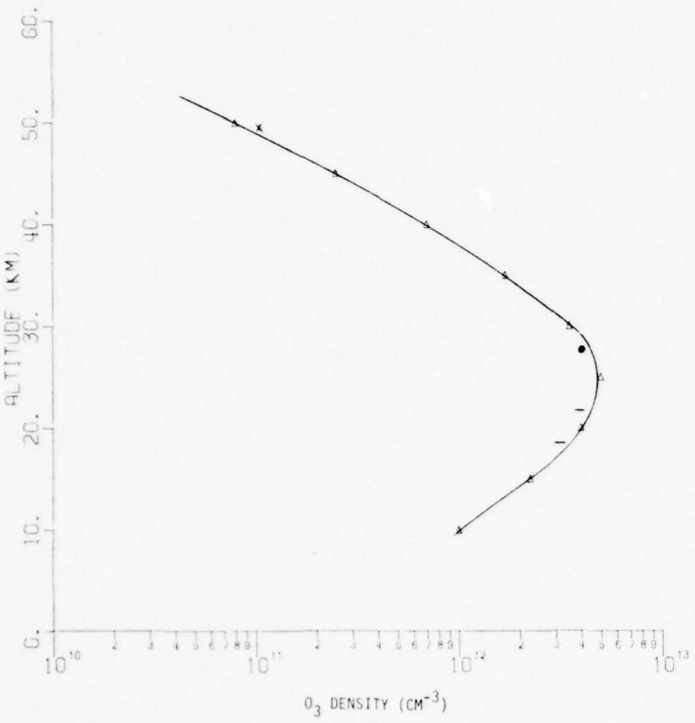
Figure 4. Composition-particle densities.

△ Calculated value

. Experimental measurement, STRATCOM VI

— Experimental measurement, Lowenstein, Sep 75 [8]

X Experimental measurement, Randhawa and Izquierdo, Sep 72 [9]



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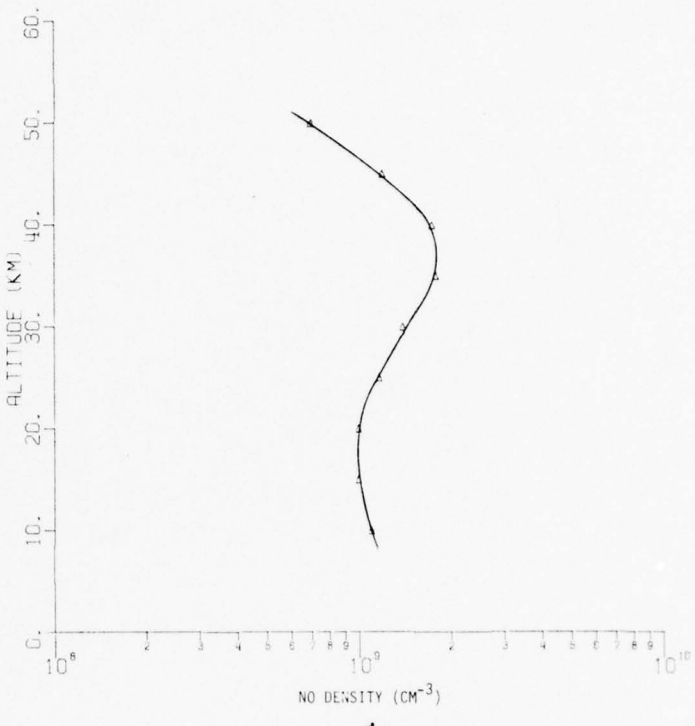

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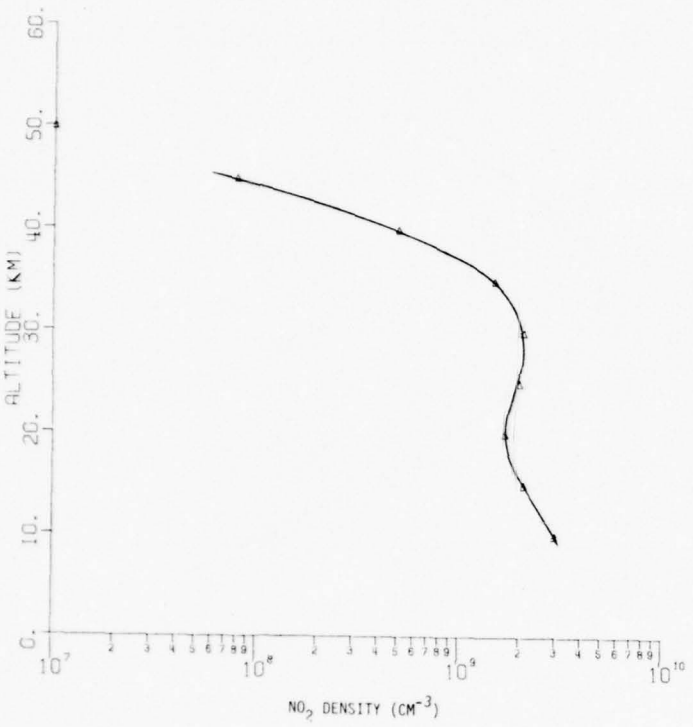

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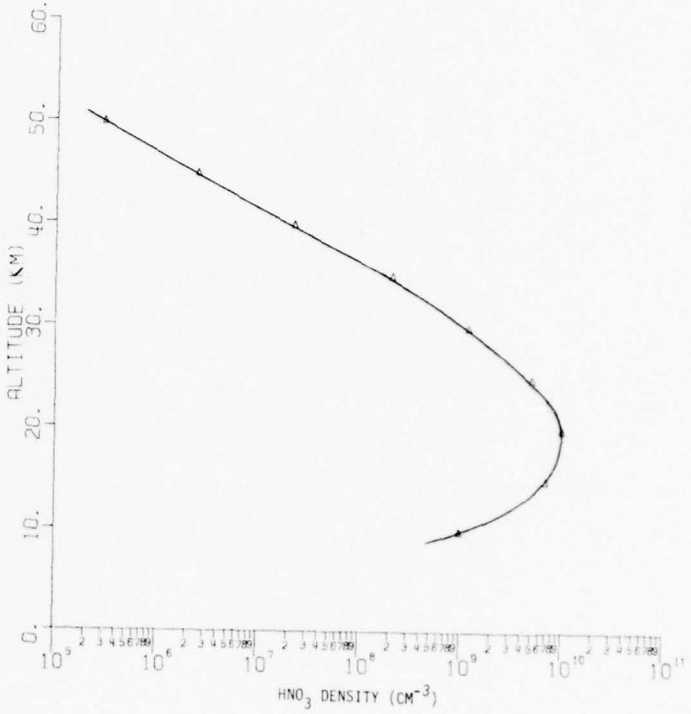

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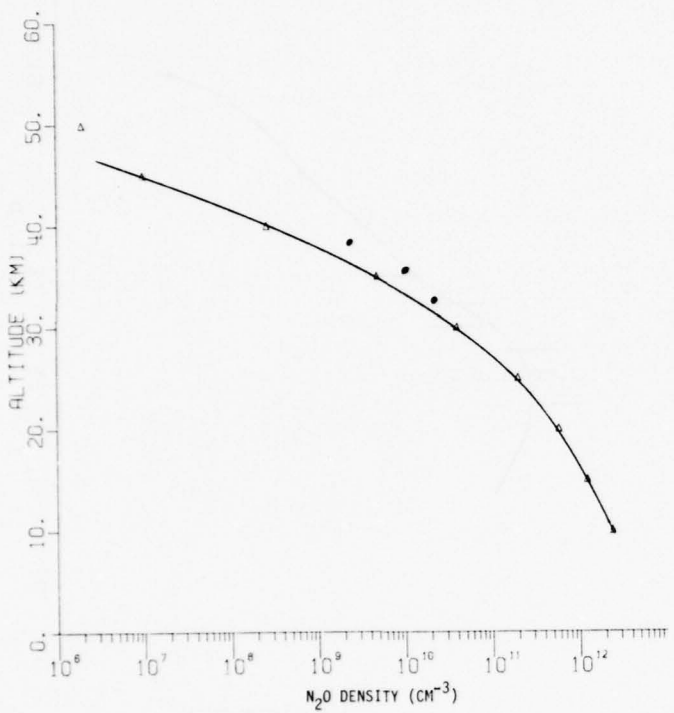
Figure 4. (cont)

△ Calculated value

. Experimental measurement, STRATCOM VI



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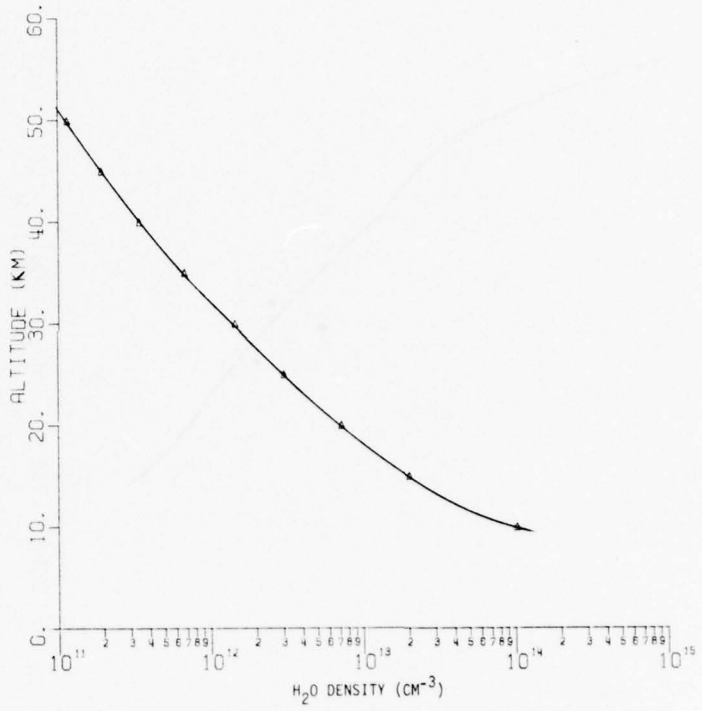

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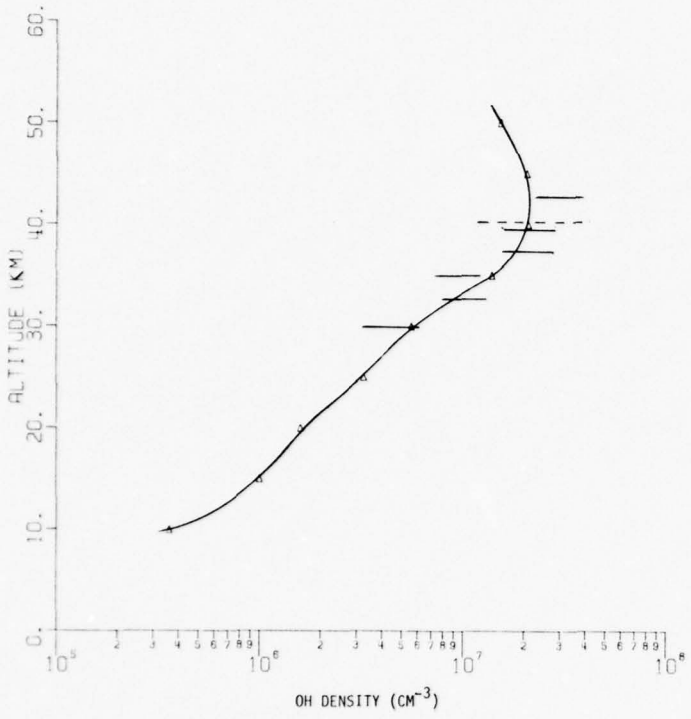
Figure 4. (cont)

Δ Calculated value

— Experimental measurement, January 1976
J. G. Anderson [7]

--- Experimental measurement, July 1975
J. G. Anderson [7]



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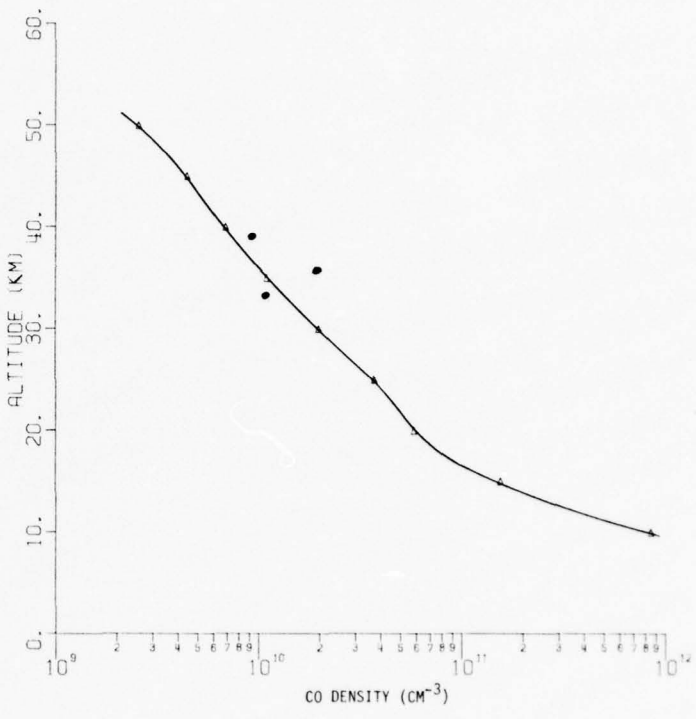
Δ Calculated value

. Experimental measurement, STRATCOM VI

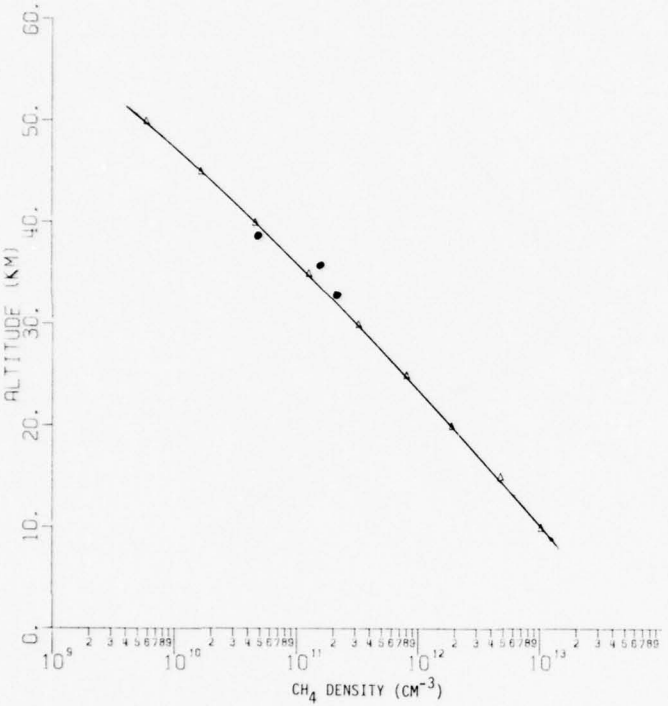


j

Figure 4. (cont)

△ Calculated value

. Experimental measurement, STRATCOM VI



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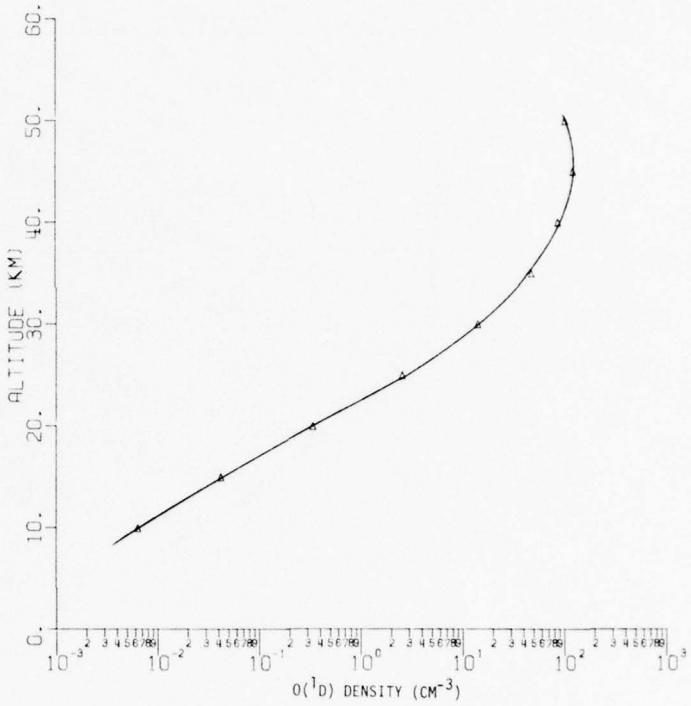

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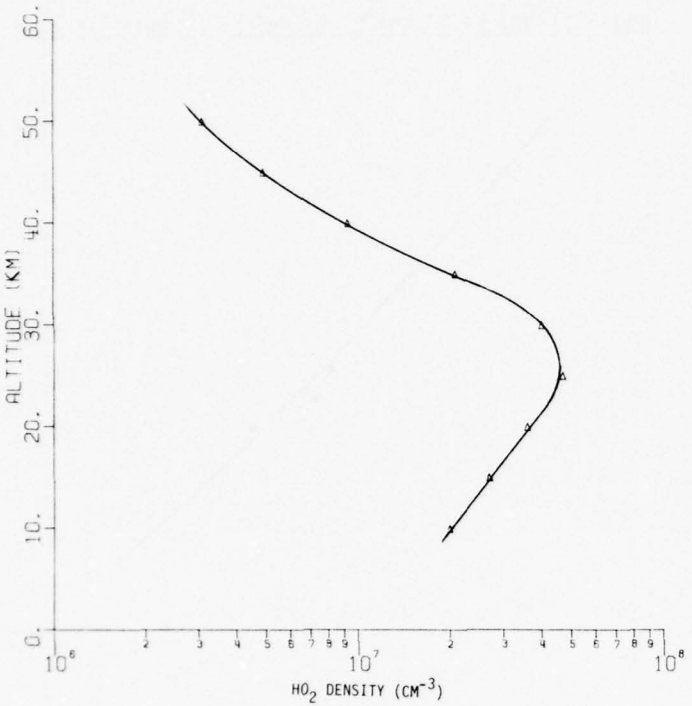
Figure 4. (cont)



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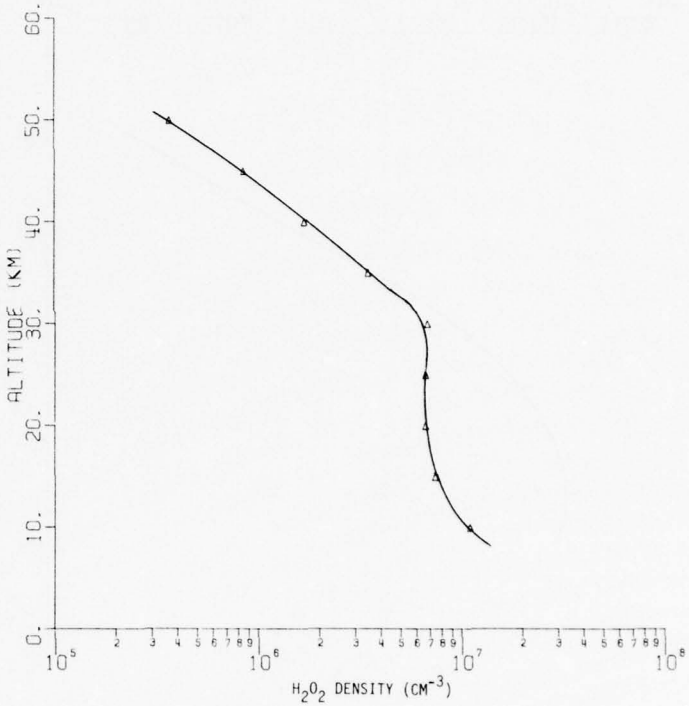

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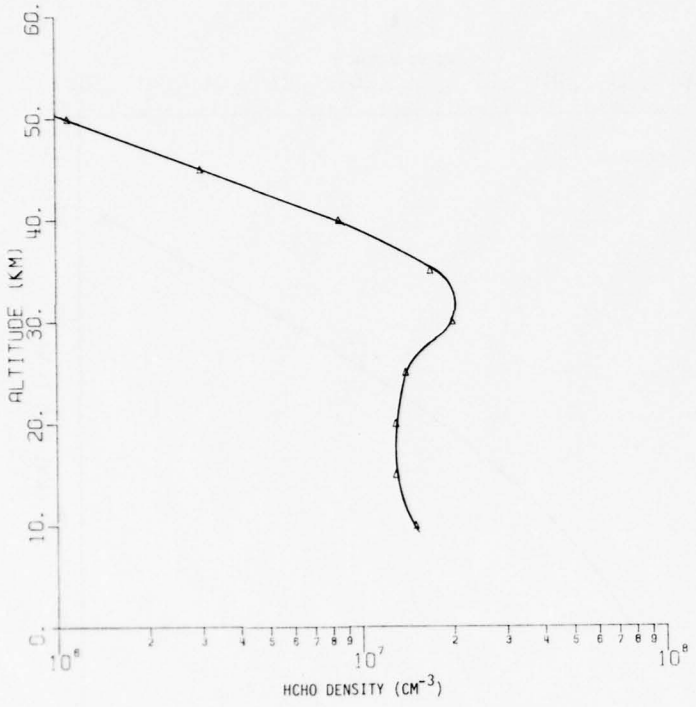

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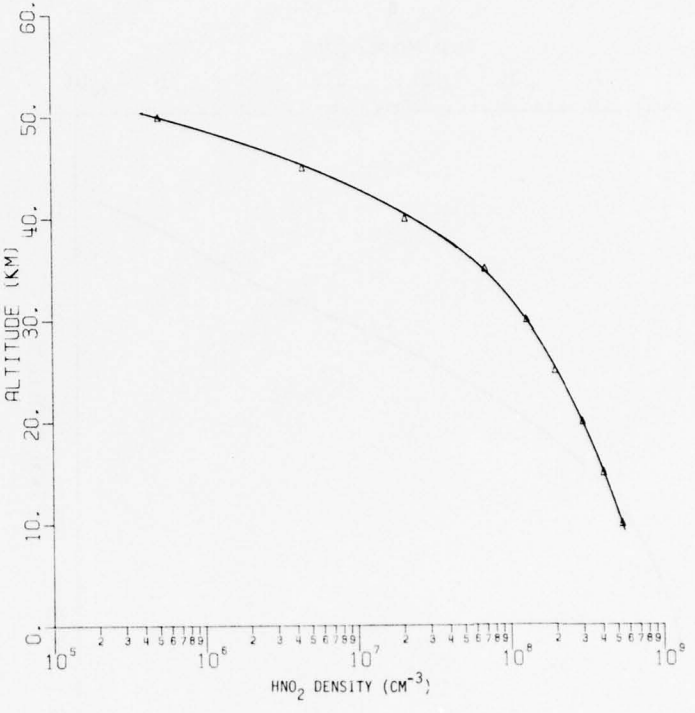

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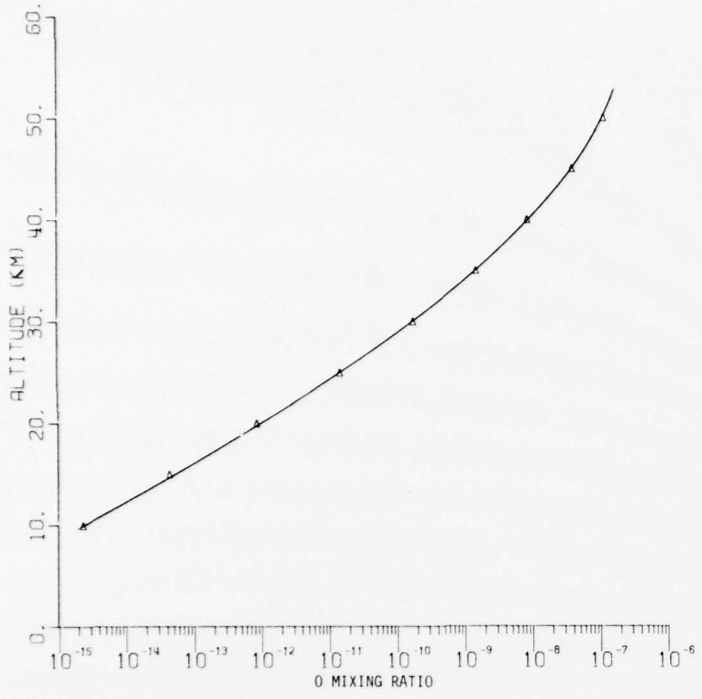

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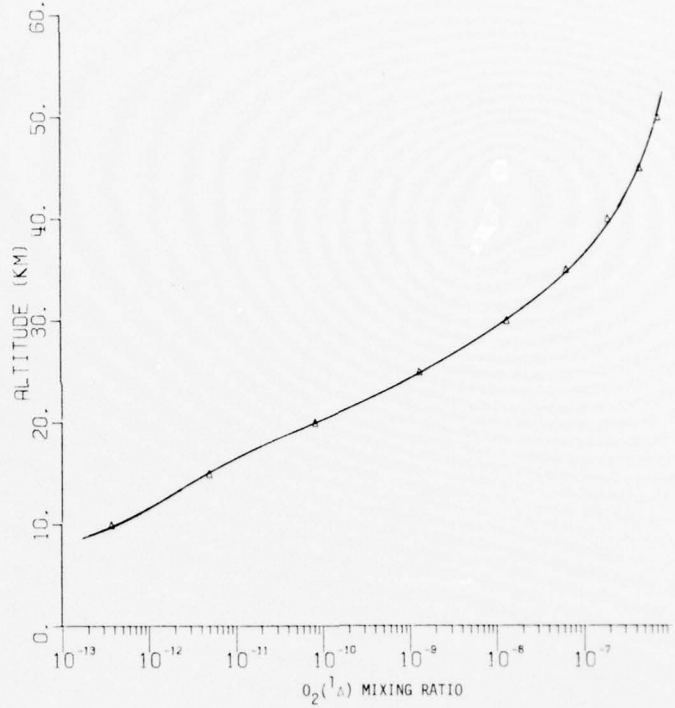


b

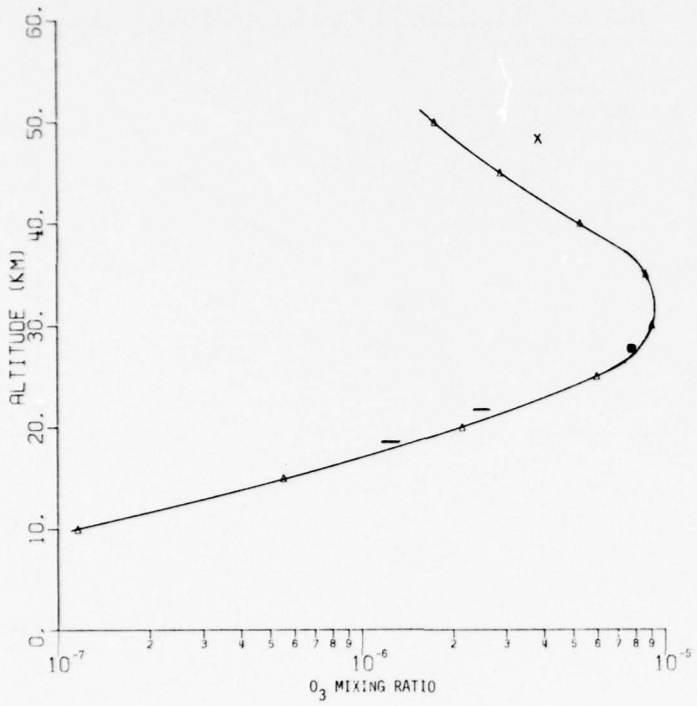
Figure 5. Composition-mixing ratios.

△ Calculated value

— Experimental measurement, Lowenstein, Sep 75 [8]

X Experimental measurement, Randhawa and Izquierdo, Sep 72 [9]

. Experimental measurement, STRATCOM VI



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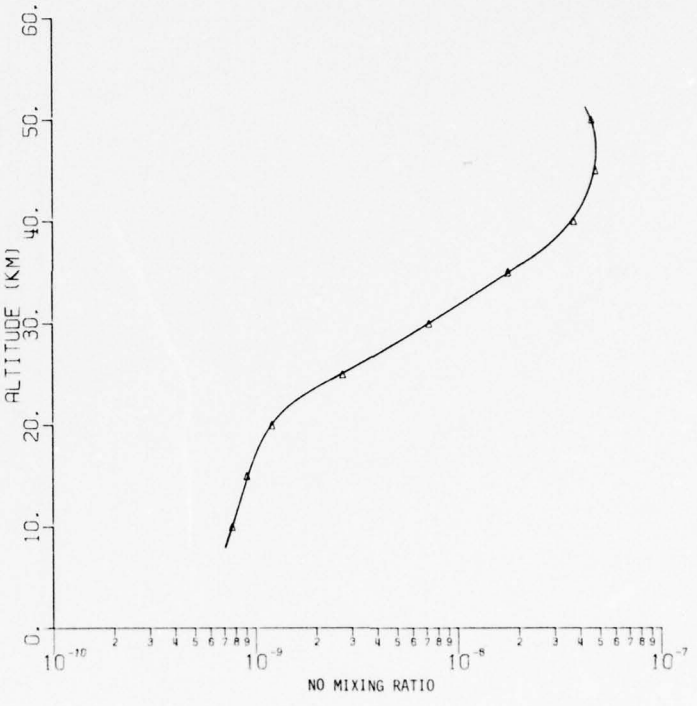

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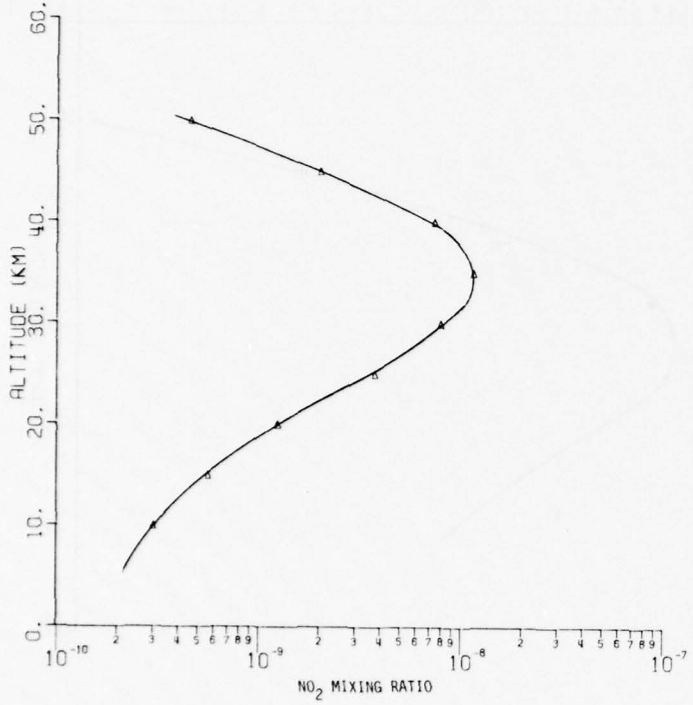

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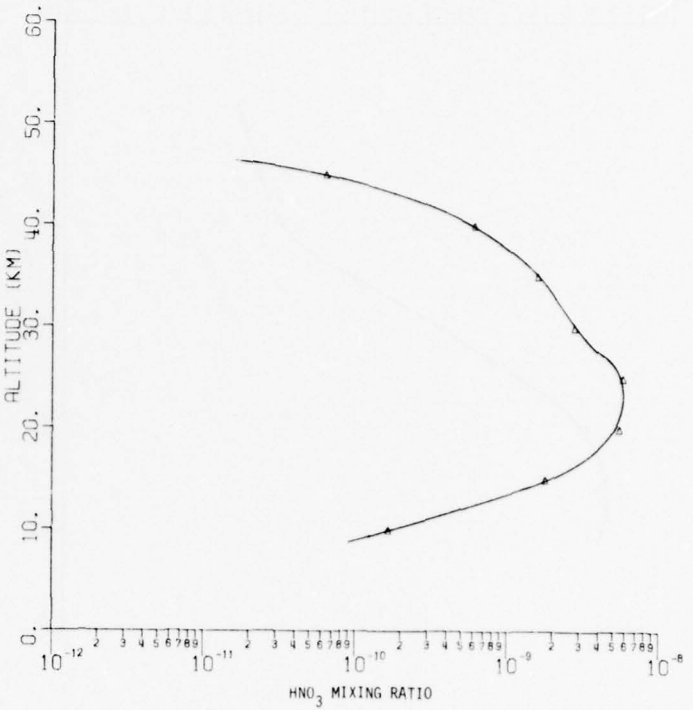

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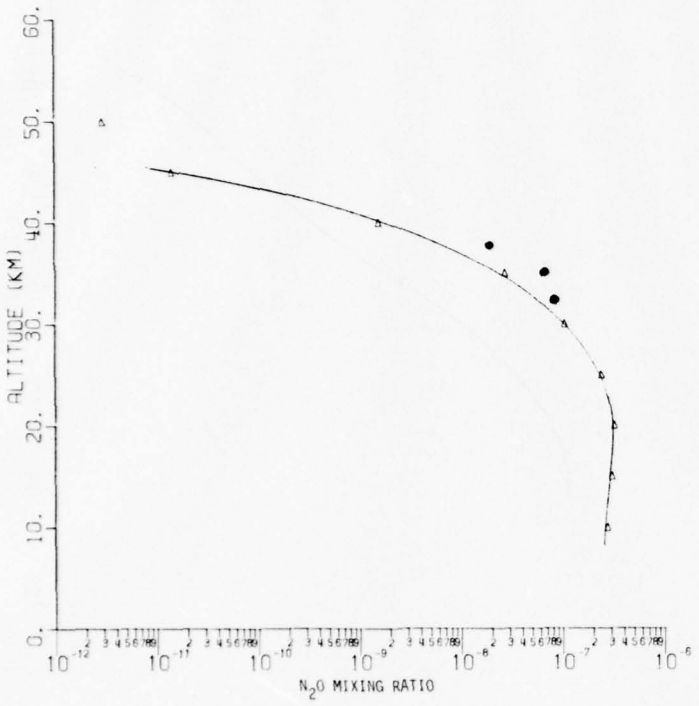
Figure 5. (cont)

Δ Calculated value

. Experimental measurement, STRATCOM VI



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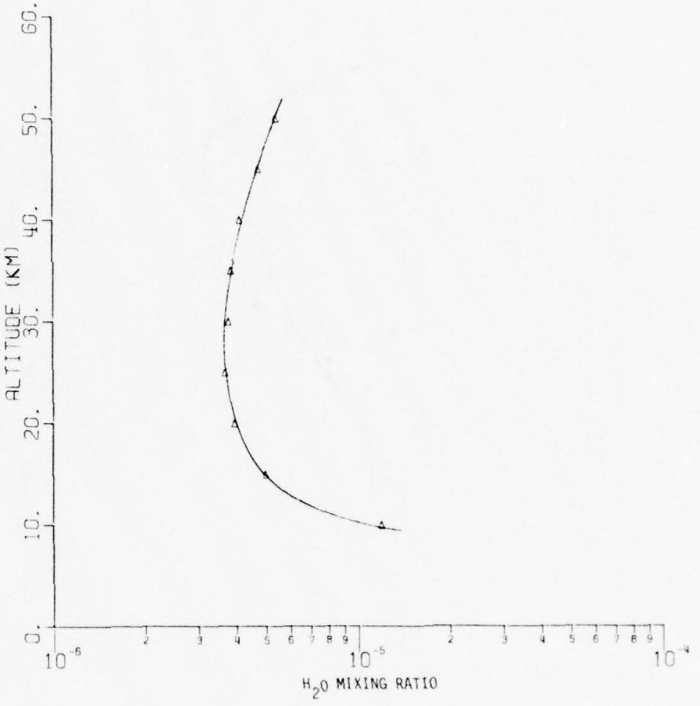

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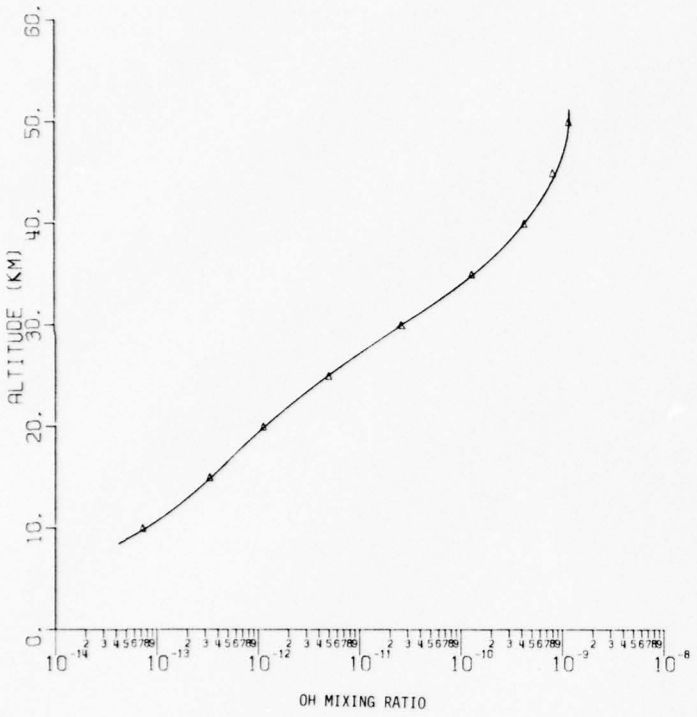
Figure 5. (cont)

Δ Calculated value

. Experimental measurement, STRATCOM VI



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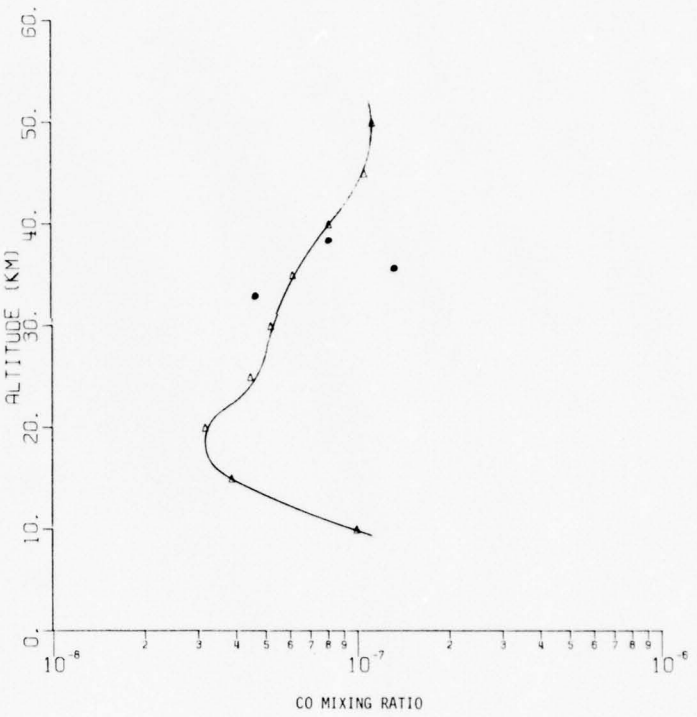

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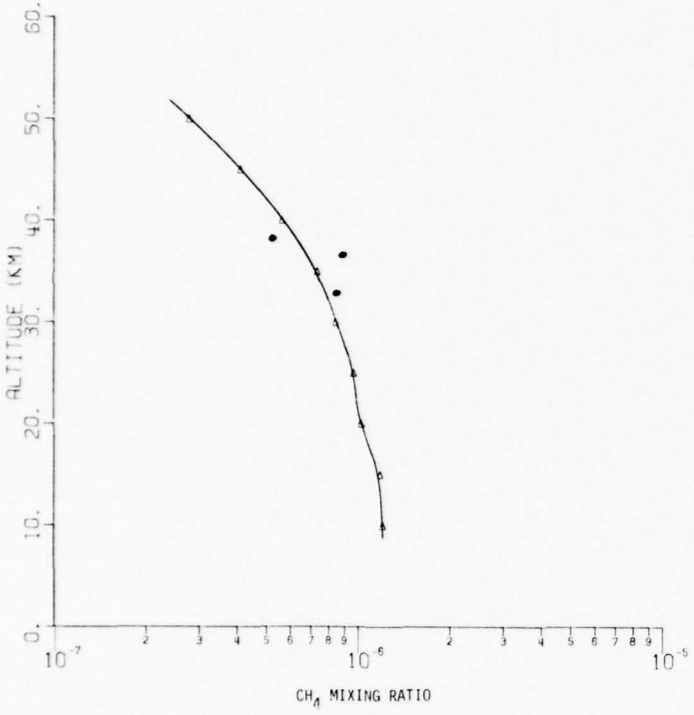
Figure 5. (cont)

△ Calculated value

. Experimental measurement, STRATCOM VI



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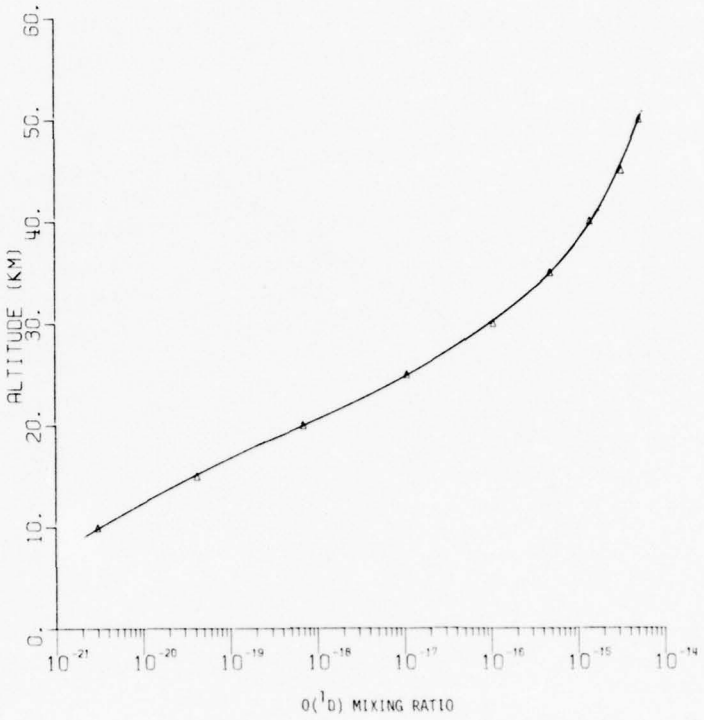

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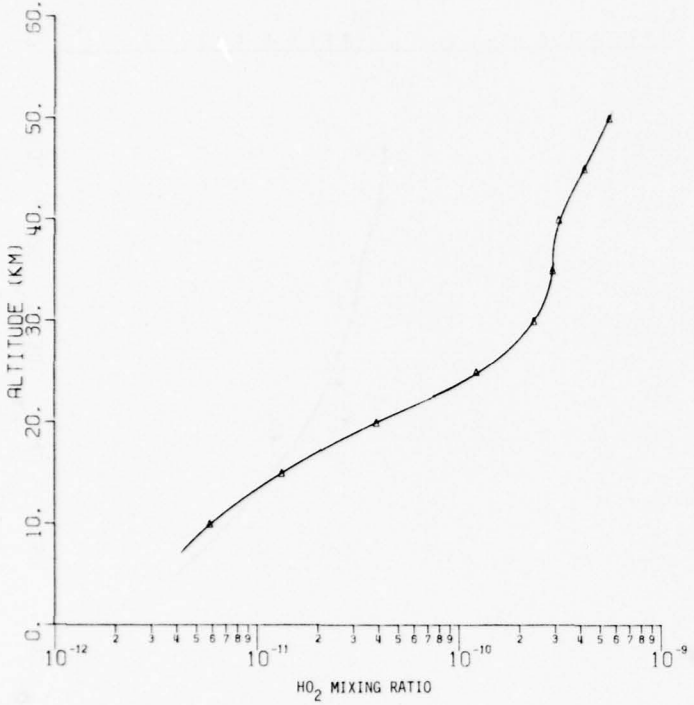
Figure 5. (cont)



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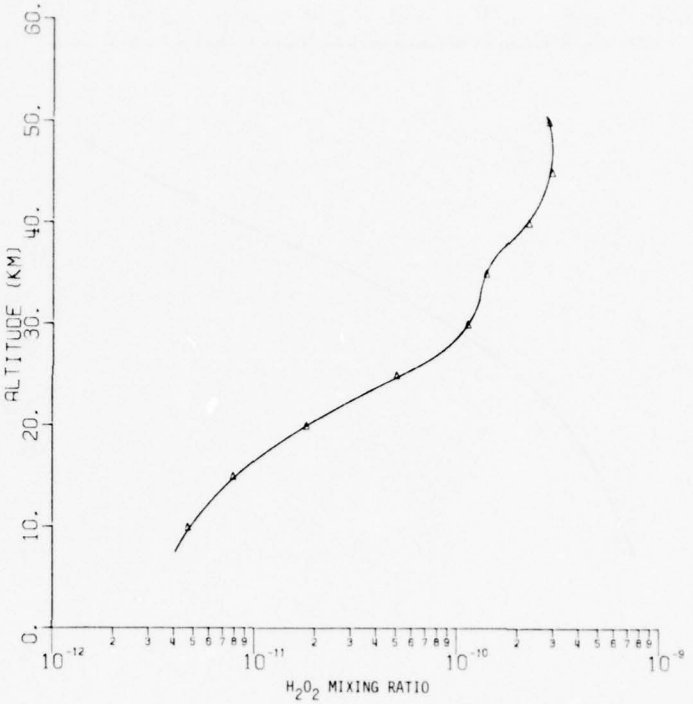

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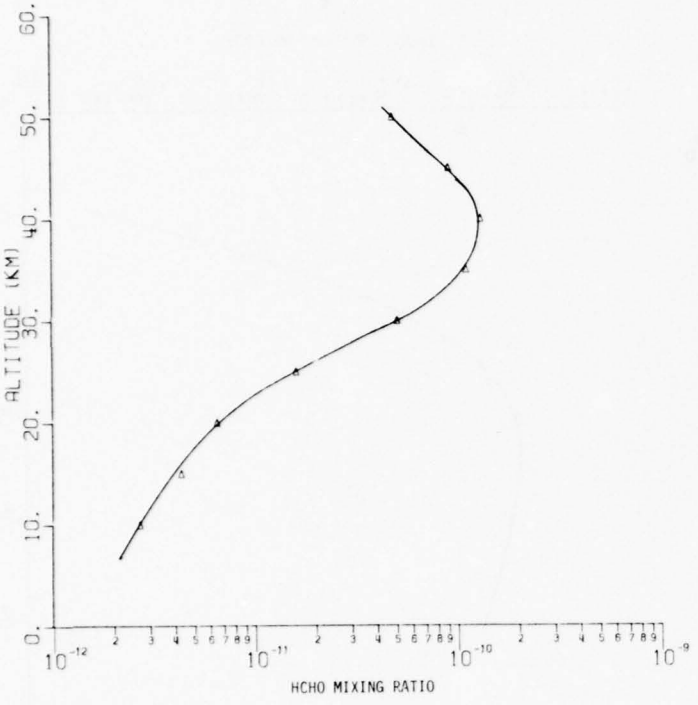

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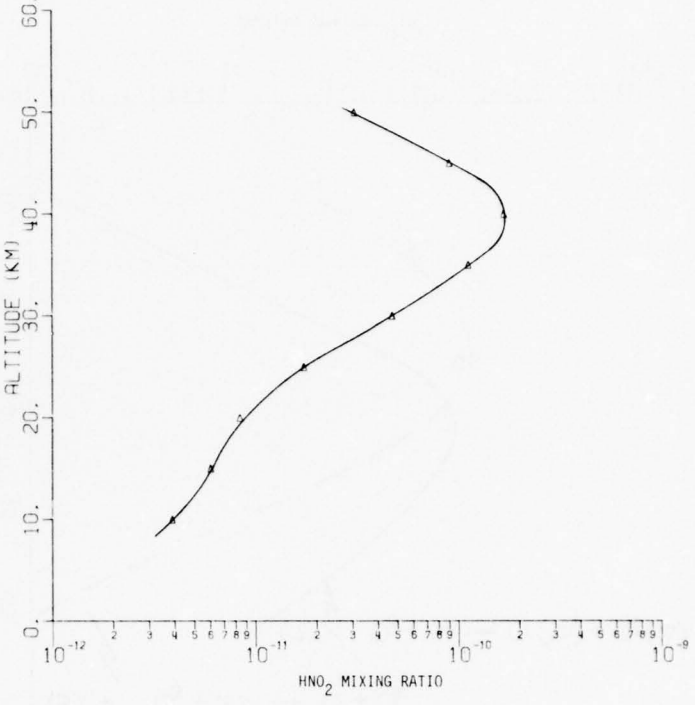

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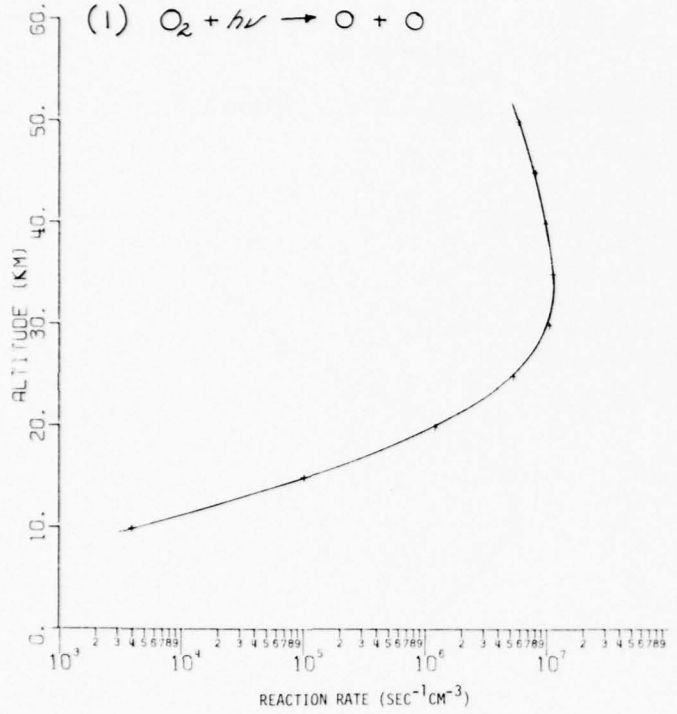

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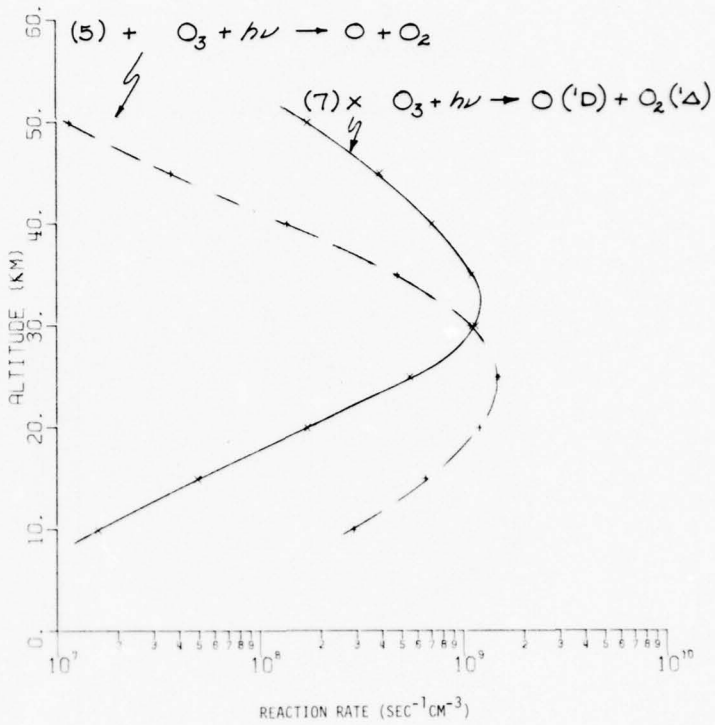
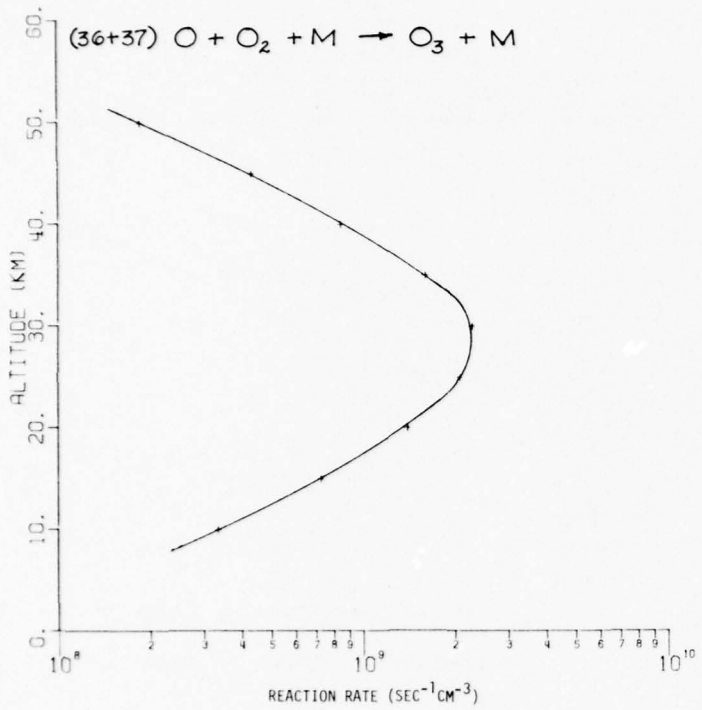


b

Figure 6. Rates of chemical and photodissociation reactions.



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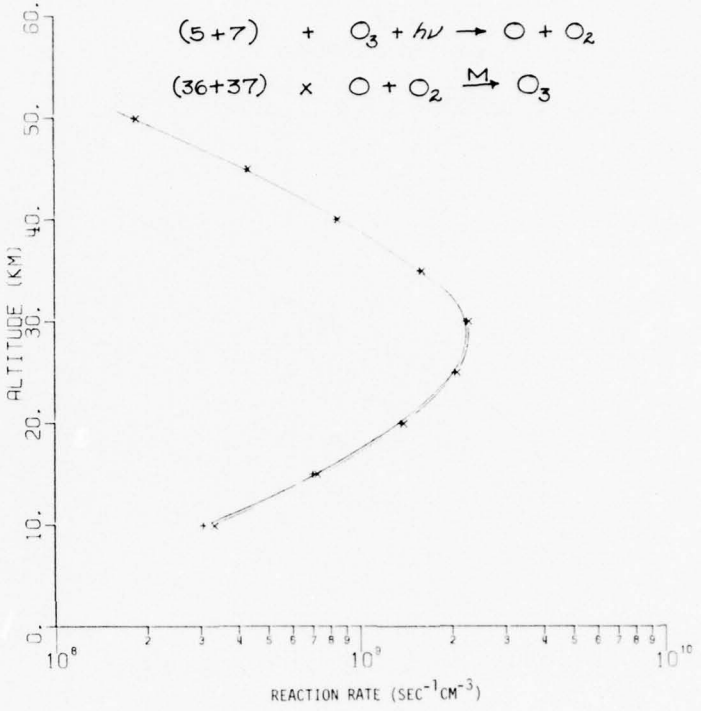

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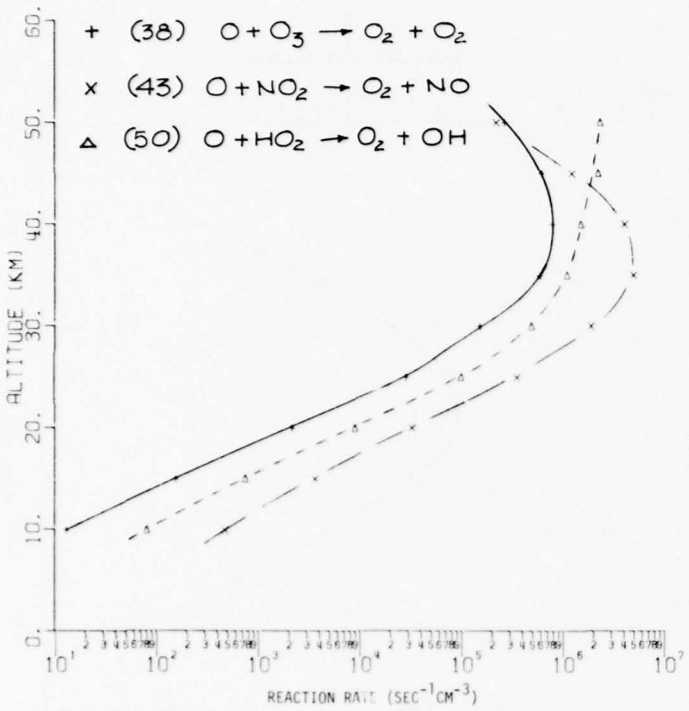
Figure 6. (cont)



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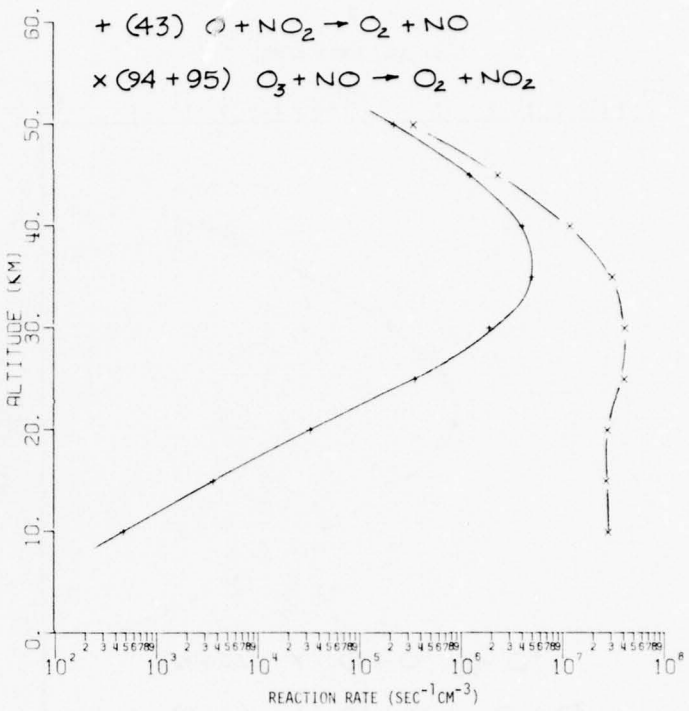

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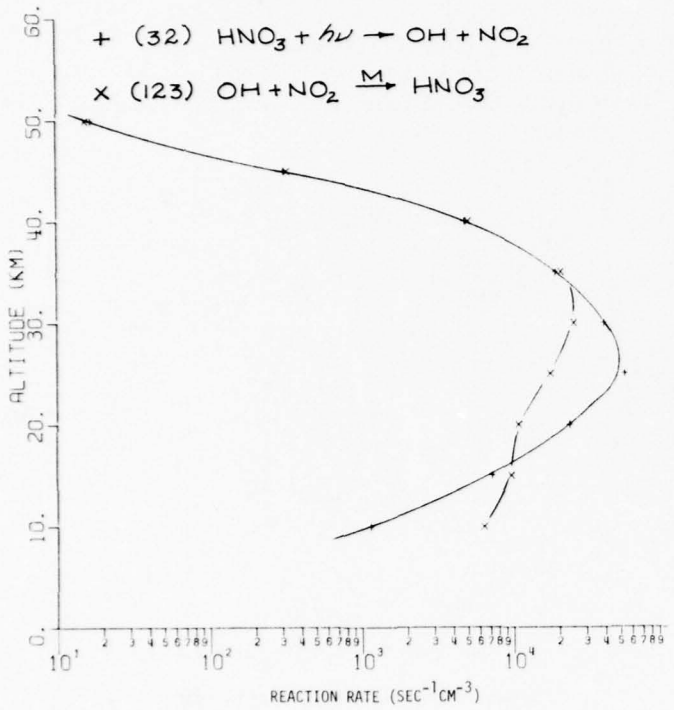
Figure 6. (cont)



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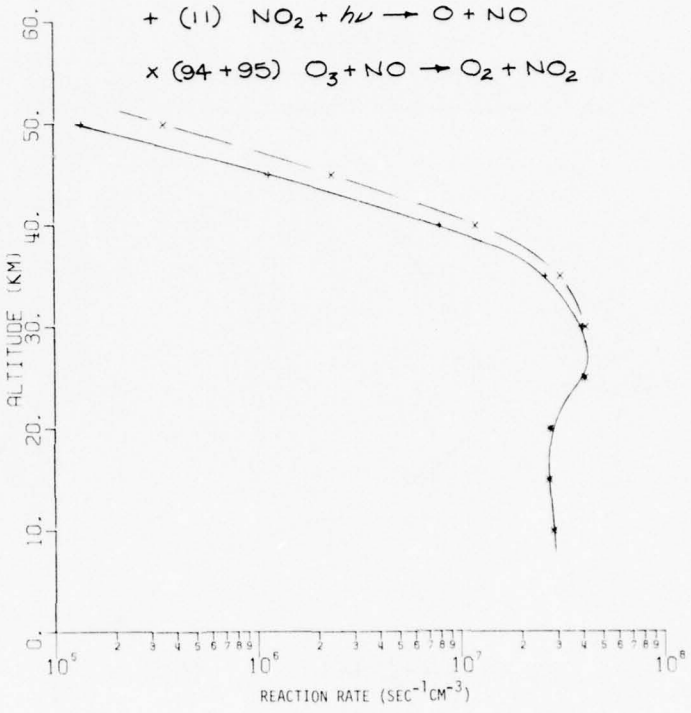

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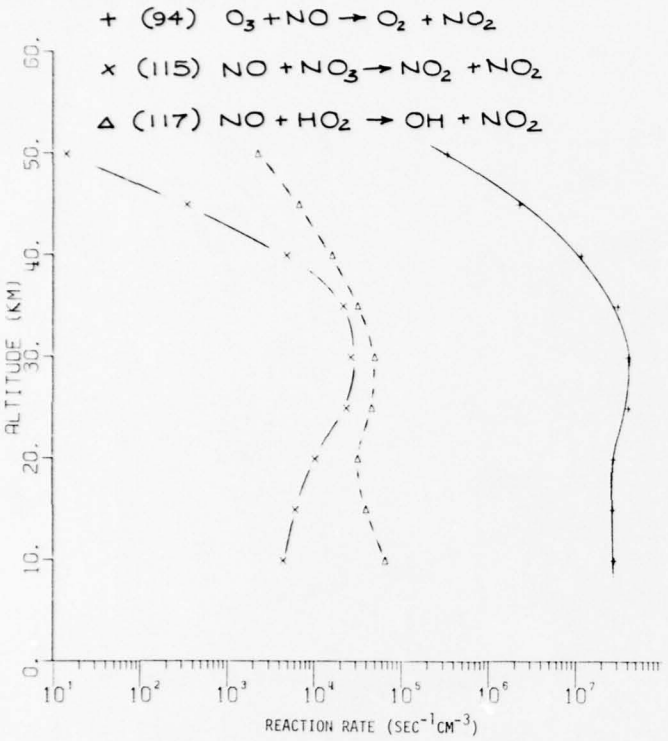
Figure 6. (cont)



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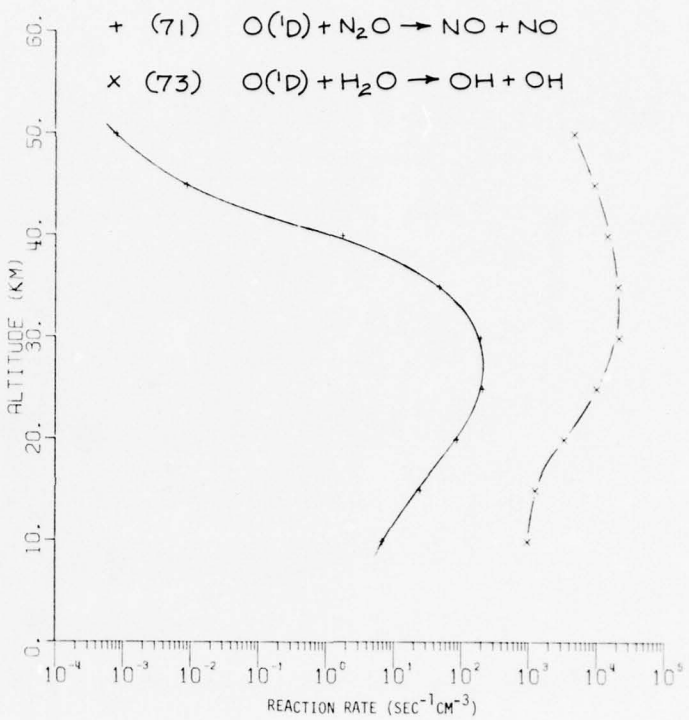

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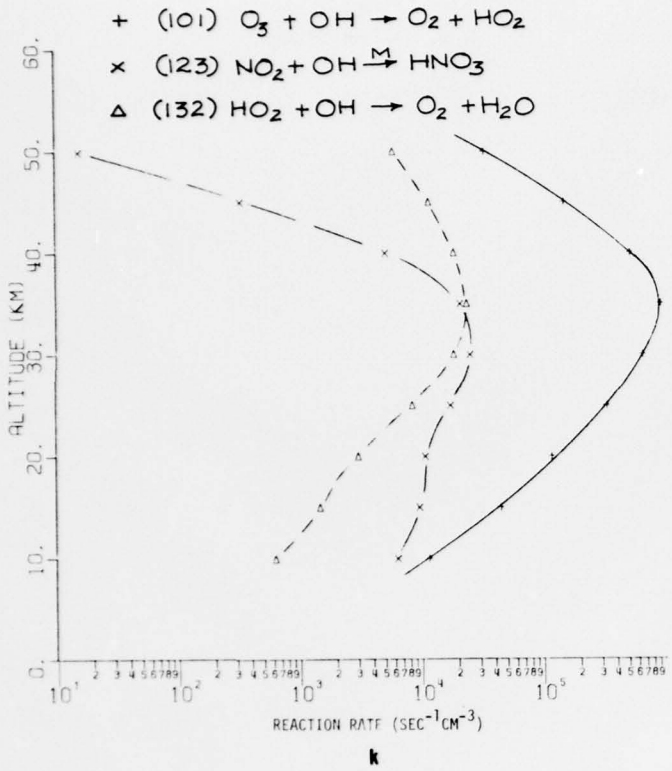

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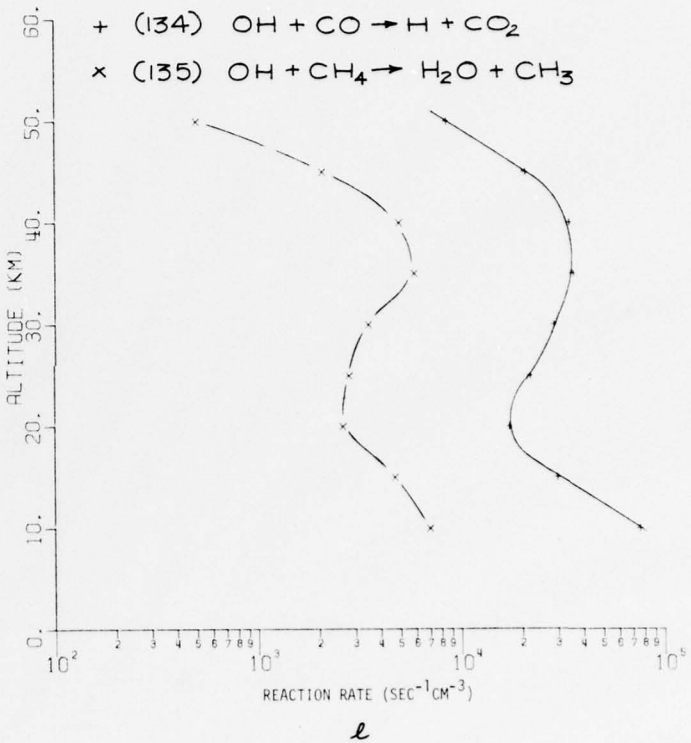

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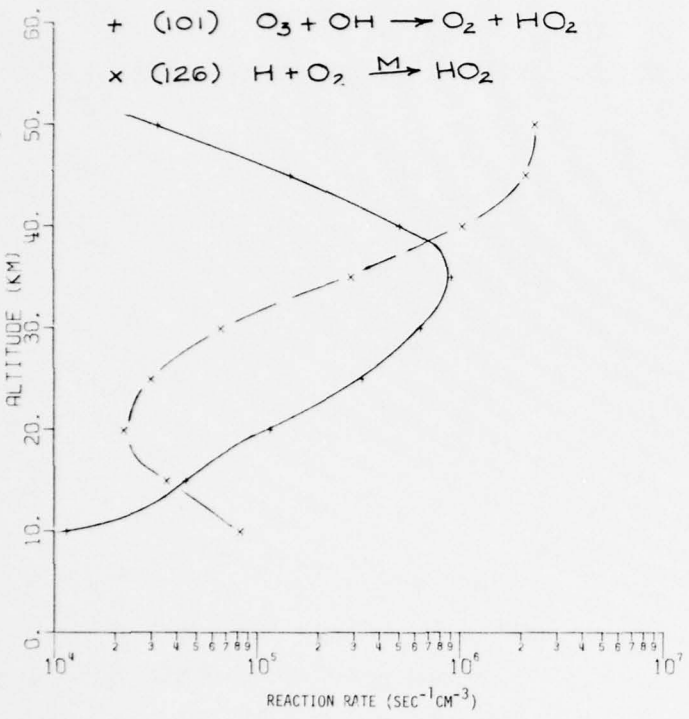

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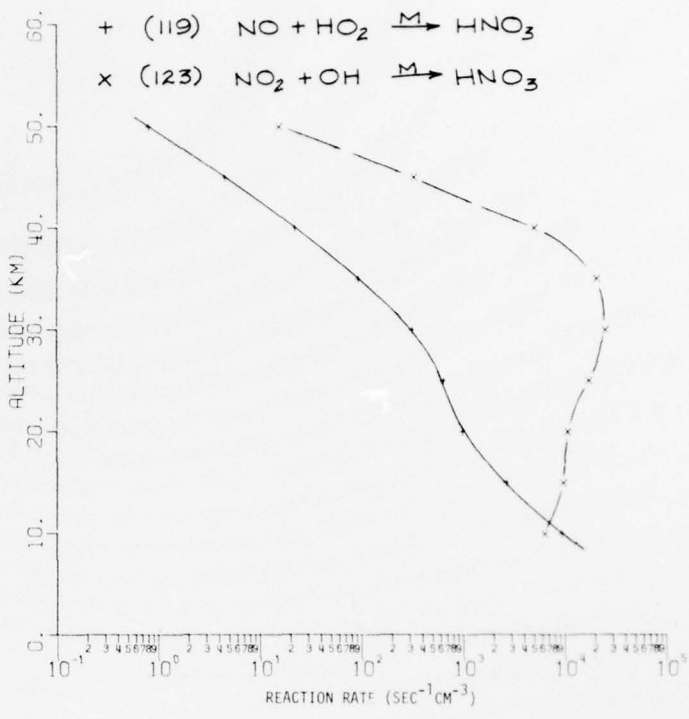

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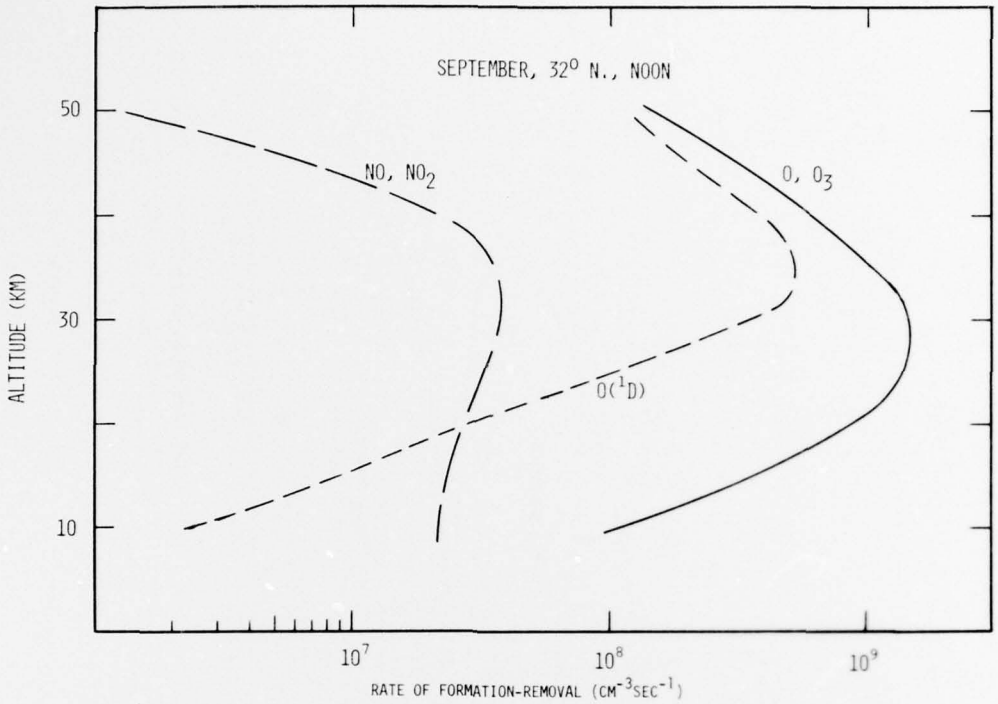

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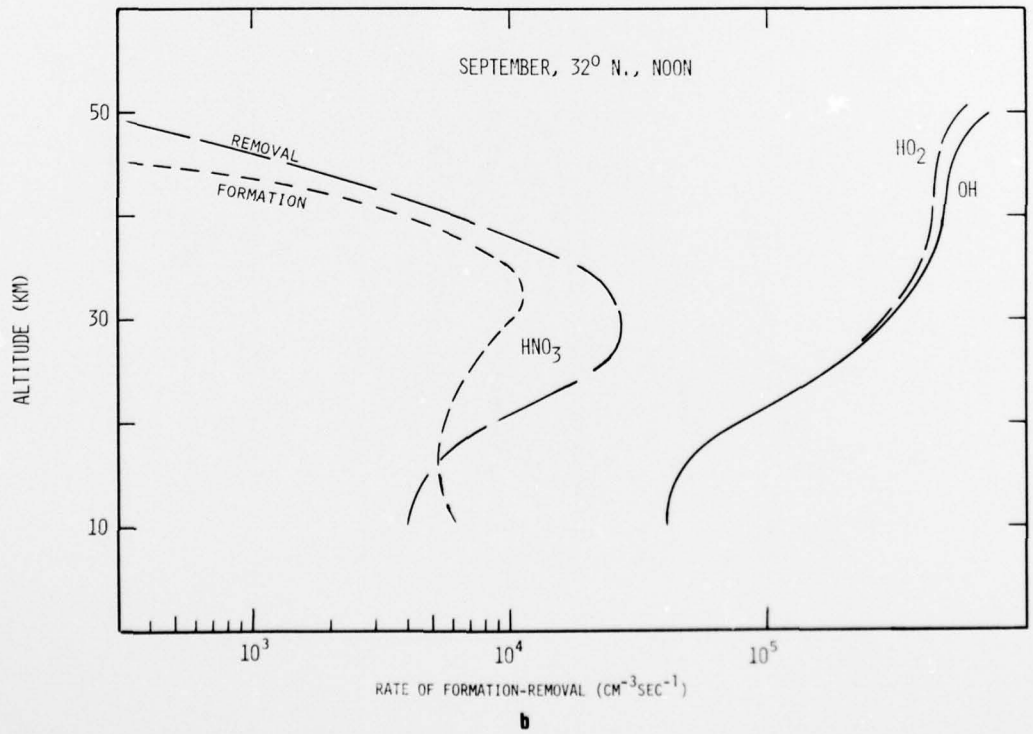
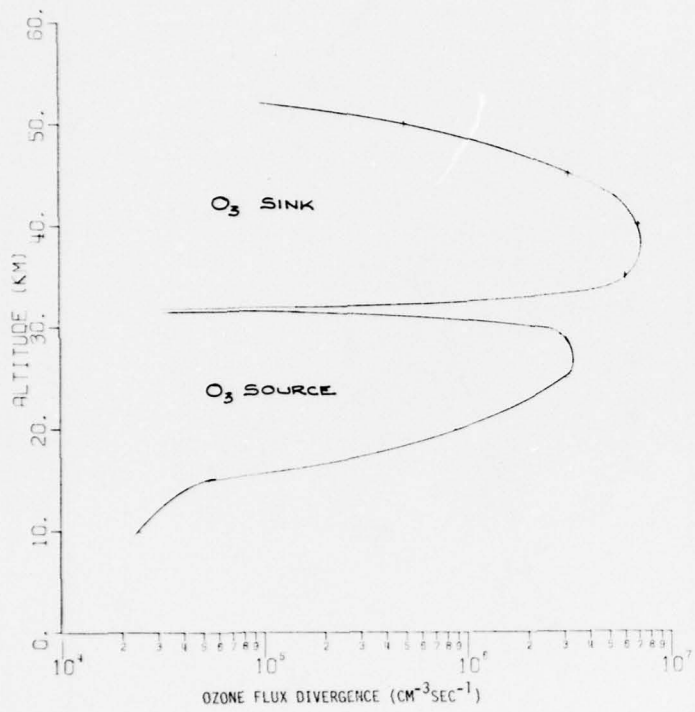


b

Figure 7. Total formation and removal rates.



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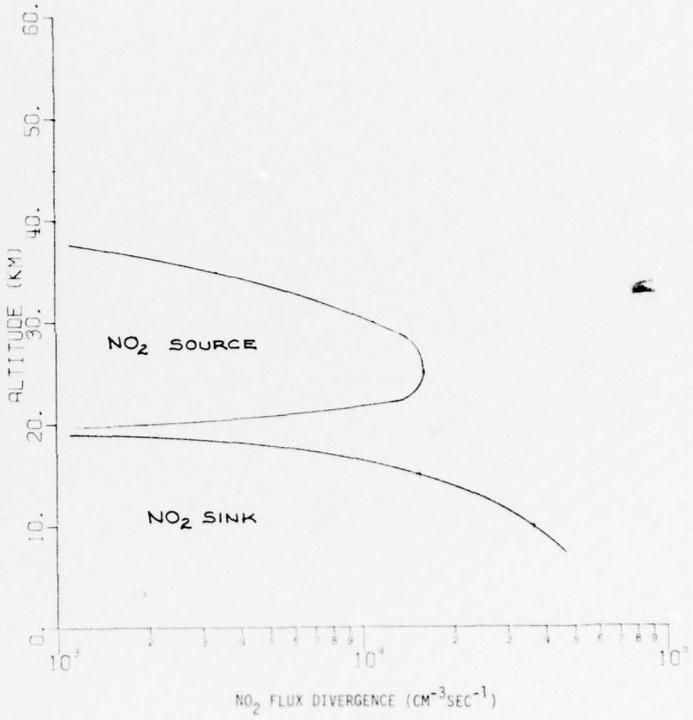
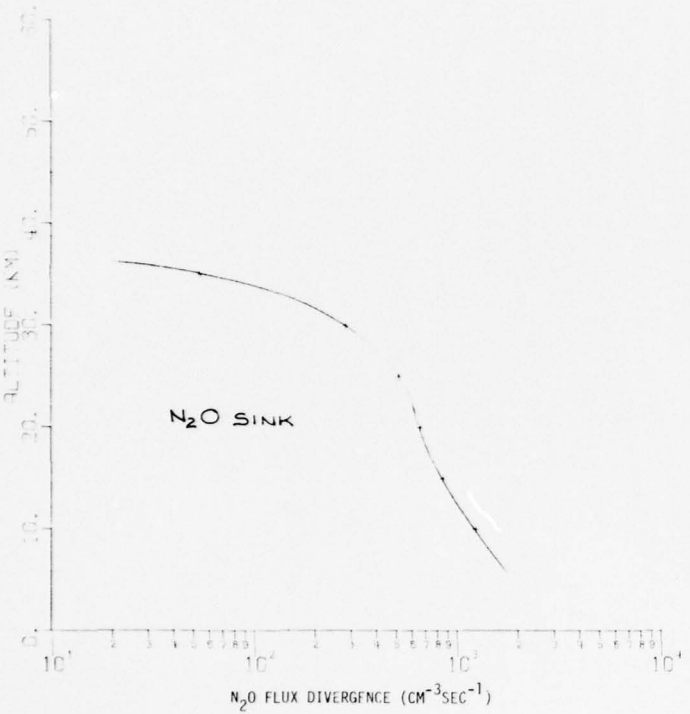


b

Figure 8. Transport contribution to particle densities.



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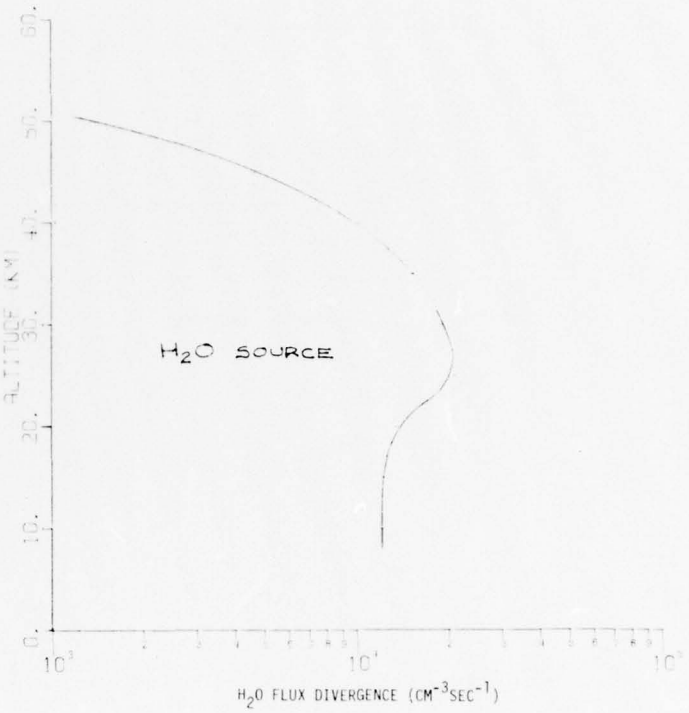

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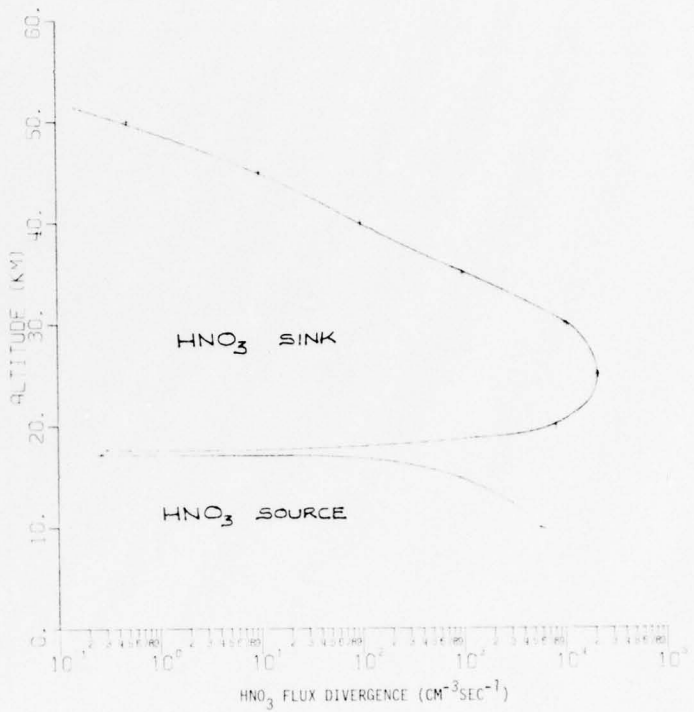
Figure 8. (cont)



e

Figure 8. (cont)

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