SIMULATION OF CHEMILUMINESCENT REACTION OF NITRIC OXIDE WITH ATOMIC OXYGEN IN A SUPERSONIC LOW DENSITY WIND TUNNEL - PART 2

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ARO, Inc.

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FOREWORD

This report presents results of the second series of wind tunnel tests of an atmospheric probe model. The project is a joint effort of the Air Force Cambridge Research Laboratories (AFCRL), Arnold Engineering Development Center (AEDC), and Mithras, Inc., Cambridge, Massachusetts. The investigation is being conducted by D. Golomb and F. P. DelGreco of AFCRL, R. E. Good of Mithras, Inc., and J. A. van der Bliek and R. A. Cassanova of AEDC under Program Element 62405424, Project 7635.

The results of tests presented were obtained by ARO, Inc. (a subsidiary of Sverdrup & Parcel and Associates, Inc.), contract operator of the AEDC, Air Force Systems Command (AFSC), Arnold Air Force Station, Tennessee, under Contract AF 40(600)-1200. The tests were conducted from March 14 to April 29, 1966, under ARO Project No. SB0609, and the manuscript was submitted for publication on November 18, 1966.

This technical report has been reviewed and is approved.

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ABSTRACT

Low density wind tunnel tests were carried out at Mach 2 and 3 to simulate the release of nitric oxide in the upper atmosphere from a sounding rocket. The supersonic flow contained atomic oxygen, and the wall-less chemiluminescent reaction of O with NO was investigated. Reaction rate constants thus obtained were up to 5 orders of magnitude higher than previously obtained laboratory rate constants. This is an extension of the work reported in AEDC-TR-66-105.

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Illustrations

NOMENCLATURE

E	Exposure
h	Planck's constant
I	Intensity of transmitted light
k	Rate constant
M	Mach number
$\dot{M}$	Mass flow rate
n	Particle density
p	Pressure
Re	Reynolds number
T	Temperature
T_R	Relative transmission of film negative
t	Time
u	Velocity
α	Degree of dissociation for oxygen
λ	Mean free path
ν	Frequency of emitted light
Φ	Photon emission rate per unit volume at stagnation point

SUBSCRIPTS

j	Jet
NO	Nitric oxide
o	Stagnation conditions
∞	Free-stream conditions

SUPERSCRIPTS

'	Stagnation conditions behind normal shock
*	Excited state

SECTION I
INTRODUCTION

A new method of determining atomic oxygen (O) profiles in the altitude range of from 90 to 140 km has been developed by Golomb et al., Ref. 1. Nitric oxide (NO) was released at a known rate from a rocket at night. The radiation in the mixing zone, the so-called headglow, was analyzed using ground-based cameras. Extremely high radiation intensity was observed, which was incompatible with the laboratory rate constant of the reaction $NO + O \rightarrow NO_2 + h\nu$. Also, the aerodynamic model used in the data reduction was unrealistic. Wind tunnel tests (reported in Ref. 2) were carried out to provide a calibration of the headglow, and based on these results Golomb and Good, Ref. 3, re-evaluated the flight data.

This report deals with wind tunnel tests carried out in 1966. The work will be continued with wind tunnel tests in early 1967.

The purpose of this series of tests was to obtain additional data for $M_\infty \simeq 3$ at $T_0 = 290$, 550, and 700°K and to extend the range of tests to include $M_\infty = 2$ at $T_0 = 290$°K. The latter condition resulted in reaction zone pressures closer to those of previous laboratory tests.

SECTION II
CURRENT STATUS AND EXTENT OF TESTS

The results and analysis of experiments carried out at AEDC have been reported in Refs. 2, 4, 5, and 6. The early rocket flights, Ref. 1, and their re-evaluation, Ref. 3, are concerned with a model with two side orifices. The atomic oxygen concentrations of Ref. 3, based on wind tunnel calibration, are reproduced in Fig. 1. For comparison, the total particle concentration from the U. S. Standard Atmosphere, 1962[1] and the measured molecular oxygen (O_2) concentrations reported in Ref. 7 are also given in this figure.

With a model having one forward facing orifice, an effective rate constant for the reaction of NO and O was determined in the wind tunnel and reported by DelGreco et al., Ref. 4. The photon emission rate

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[1]U. S. Standard Atmosphere, 1962, U. S. Government Printing Office, December 1962.

constant as determined from photographs from the headglow in the wind tunnel was

$$k = \frac{\Phi}{[NO]\,[O]} = 2.4 \times 10^{-12}\ cm^3\ molecule^{-1}\ sec^{-1}$$

as compared to the rate constant

$$k = 6.4 \times 10^{-17}\ cm^3\ molecule^{-1}\ sec^{-1}$$

measured by Fontijn et al. in a chemical flow tube, Ref. 8.

The rocket flights reported by Golomb et al., Ref. 1, produced a glow with photon emission rates in line with the wind tunnel data. Spindler, Ref. 9, reported also unexpectedly high photon emission rates from NO released by rockets; however, no direct comparison of k for this test could be made because of a lack of aerodynamic data of the rocket flow pattern.

The use of the supersonic wind tunnel for chemical reaction studies offers the unique advantage of producing wall-less reactions heretofore only produced by molecular beams at much lower pressures and constituent concentrations. However, the mixing process in the present aerodynamic configuration needs further investigation before an explanation of the observed reaction rates can be made.

SECTION III
LOW DENSITY WIND TUNNEL

The tests were conducted in the Aerospace Research Chamber (ARC (8V)) of the Aerospace Environmental Facility at AEDC. The chamber, operated as a low density wind tunnel, is shown in Fig. 2. The chamber is 8 ft in diameter and 16 ft long. The nozzles are mounted on a removable dished head.

The pumping equipment of the chamber consists of a 6-in. oil diffusion pump in series with a 60-liter/sec mechanical pump and cryopumping surfaces. The major pumping action during a tunnel run is provided by a series of radially positioned cryopanels cooled with gaseous helium (GHe) at temperatures from 15 to 20°K with a total area of 240 ft^2. The GHe is cooled in a continuously operating 1-kw refrigerator. The cylindrical, perforated precooler and the chamber end panel are cooled by liquid nitrogen (LN_2) at 77°K. The cylindrical part of the chamber is provided with an LN_2 liner which serves as a heat shield for the helium cryopump.

The nozzles, shown in Fig. 2, were cooled with LN_2 to reduce nozzle wall boundary-layer thickness. During this test, Mach 3 and 2 nozzles were used.

The nozzle gas supply system for the test was similar to the system used in Ref. 2. The N_2 and O_2 were fed to the gas heater from standard bottles, via standard flowmeters. The gas was passed through two separate tubular gas heaters into a heated plenum chamber. The O_2 was passed through a quartz manifold where three microwave cavities dissociated part of it (Fig. 2). These cavities were powered by three 80-w microwave generators (2.5 kMc). The O_2 heater upstream of the cavities was not turned on when the power supplies were operating. The plenum chamber temperature was held at the desired reservoir temperature.

The operating range for the tests was selected so that the anticipated flight trajectory could be approximately duplicated. The ranges selected were:

P_o = 100 to 400 μ Hg
T_o = 290 to 700°K
p_∞ = 1 to 24 μ Hg
T_∞ = 85 to 230°K
Re_∞/cm = 2.9 to 53
M_∞ = 1.99 to 3.49
λ_∞ = 0.165 to 1.60 cm
u_∞ = 500 to 1000 m/sec
Uniform core diameter = 15 to 30 cm

The atomic oxygen concentration in the test section was determined as described in Ref. 2. The percent of oxygen dissociation in the test section is given as a function of settling chamber pressure in Fig. 3.

SECTION IV
MODELS AND PROCEDURE

The model used to simulate the release of NO from the rocket test vehicle into the atmosphere is shown in Fig. 4. The model had an orifice at the apex of a cone and is similar to the rocket vehicle which was flown after the present tests were completed. The model was connected via a supply line to gas bottles with a mixture of NO and N_2. The pressure inside the model, measured with a strain-gage pressure

transducer, was regulated with a valve near the bottles. Model pressures (p_{0_j}) ranged from 1 to 5 atm. By varying the mixture of NO and N_2 in the model the same headglow geometry (p_{0_j} and p_{0_∞} constant) could be maintained while varying the concentration of NO in the reaction region.

The overall light output of the headglow was measured with photomultiplier (PM) tubes mounted on two of the chamber Plexiglas® ports.

The headglow was photographed with 35-mm Kodak® Plus-X film with exposure times of 1/8 to 1 sec and lens openings of from f/1.9 to f/2.8.

The film and PM were calibrated with a light box which was mounted at the position of the headglow. This light box, in turn, was calibrated against a standard source at AFCRL. To provide a complete calibration over the range of exposures in the tests, a calibrated step wedge and the calibrated light box were photographed on each roll of film. A typical calibration curve is shown in Fig. 5. The transmission of the film negative relative to the light box photo transmission was obtained from microdensitometer traces of the photographic negatives, and the corresponding exposure was read from the calibration curve.

For the rate constant calculations, the volume intensity at the stagnation point was determined from the projected exposures on the negatives. The observed projected exposure (photons/cm^2-sec) from the photographs can be related to the specific radial intensity (photons/cm^3-sec) for the axially symmetric geometry by an Abel integral inversion. The inversion from surface brightness to radial intensity is described in Ref. 2. The film response dropped sharply at a wavelength of 0.68μ. Since the spectrum of the glow obtained by Fontijn et al., Ref. 8, extends well beyond 0.68μ, the measured value of the intensity was multiplied by a factor of 2.5 to account for the film response.

SECTION V
EXPERIMENTAL RESULTS

A typical photograph of the headglow is shown in Fig. 6. The sketch in Fig. 7 indicates the general flow characteristics. A spherical shell-like area around the contact surface between the two gas streams is the reaction zone.

With this configuration, the following investigations were carried out:

A. Spectrometric measurements in the visible regime of the headglow. These were obtained by Dr. F. P. DelGreco of AFCRL, and no results are reported here.

B. Initial infrared spectrometric measurements were carried out by Dr. A. T. Stair et al. of AFCRL. These runs were of an exploratory nature, but demonstrated the use of the wind tunnel for investigations of the rotational-vibrational structure of gases. Typical cases covered are:

Wind Tunnel Flow	Flow from Model	Reaction of Interest
O_2, N_2, O	N_2, NO	O + NO
O_2, N_2, O	CO	O + CO
N_2, N	NO	N + NO
N_2, N	CO	N + CO
N_2, N	O_2	N + O_2

Atomic nitrogen was produced in the same manner as atomic oxygen.

C. A total of 103 runs were made to measure the radiation attributable to the reaction of NO and O. The major results of this series are discussed here.

The overall radiation output of the headglow is plotted versus the ratio of NO pressure to total pressure in the model in Figs. 8 and 9. These figures represent the typical behavior. On the basis of limited results, it was stated in Ref. 2 that the photomultiplier reading was proportional to the NO concentration, as would be expected from the second-order reaction of NO + O. Beyond a "saturation" point, a further increase in p_{NO} does not produce a further net radiation increase. Although this "saturation" effect is confirmed by the present experiments, the decrease of the PM reading with decreasing p_{NO} was far more rapid than expected. The low values of p_{NO}/p_{o_j} were obtained by pre-mixing NO and N_2 in separate bottles. One possible explanation of the above behavior would be the presence of impurities, reacting with NO, in the nitrogen bottles, which would reduce the effective NO concentration. Chemical analysis after the test on similar bottles indicated that the impurities were not sufficient to produce the observed fast decrease of radiation. A series of tests with high purity NO and N_2 is required to eliminate this possibility completely.

The right-hand sides of Figs. 8 and 9 give typical photon production rates per unit volume as obtained from photographs versus the ratio of [NO]/[O]. Note that for the data in these figures only [NO] was varied. The value of [O] and the headglow geometry are constant.

The rate constant, k, is defined by:

$$k = \frac{\Phi}{1/2[NO]\ 1/2[O]}$$

where Φ is the photon production rate per unit volume at the brightest point, near the stagnation point. Half the values of the concentrations of O and NO on either side of the contact surface were used as a first approximation to the actual concentrations at the point of maximum light intensity.

Lines of constant k are drawn in Figs. 8 and 9. All values of k, determined from photographs, are summarized in Fig. 10.

In considering this figure, the following must be noted:

A. There was no distinct trend for points obtained with constant value of [NO]/[O]. Therefore, in this figure, all photon rates are plotted indiscriminately.

B. The M = 2 nozzle was installed to provide data at a higher stagnation (reaction) pressure, but this nozzle had a very poor exit velocity profile at the higher pressures. Since only stagnation point data are plotted here, this effect is expected to be minor, but the data in the high pressure regime ($p_O' > 100\,\mu$ Hg) need confirmation with an improved nozzle.

Allowing for the above uncertainties, it seems nevertheless evident that the production of excited NO_2 molecules is highly quenched as the pressure in the reaction zone is increased. For comparison, the reaction rates obtained in chemical flow tubes at higher pressures, Kaufman, Ref. 10, and Fontijn et al., Ref. 8, are shown.

Kistiakowski and Volpi obtained a value of $k = 1.6 \times 10^{-11}$ for $NO + O \rightarrow NO_2$ with a mass spectrometer in their approximation to a "stirred" reaction vessel, Ref. 11. If that value were valid over the complete pressure range of Fig. 10, it would constitute the maximum value of $NO + O \rightarrow NO_2^* \rightarrow NO2 + h\nu$; that is, the case in which all the formed NO_2 molecules are excited and assuming that in the deactivation process only one photon is emitted.

SECTION VI
CONCLUDING REMARKS

The present series of tests and the tests reported in Ref. 2 have simulated the AFCRL high altitude rocket flights, and correlations have been developed for analyses of flight data, Refs. 3 and 5. Several differences between rate constants observed in the wind tunnel and the rate constants observed in the flow tube need further exploration. The data presented here indicate that there is a strong effect of pressure on the reaction.

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APPENDIX
ILLUSTRATIONS

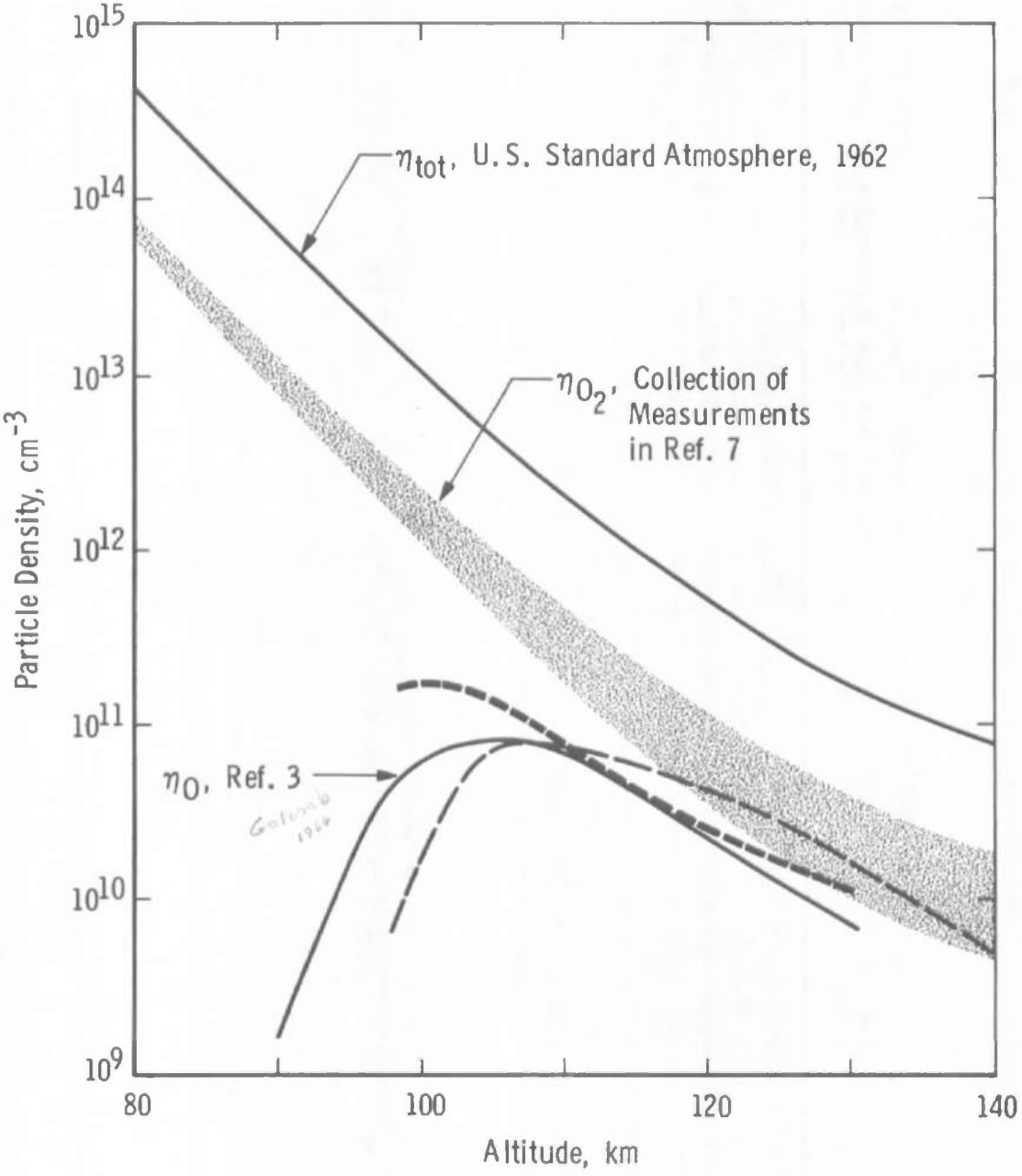

Fig. 1 Summary of Atomic and Molecular Oxygen Measurements

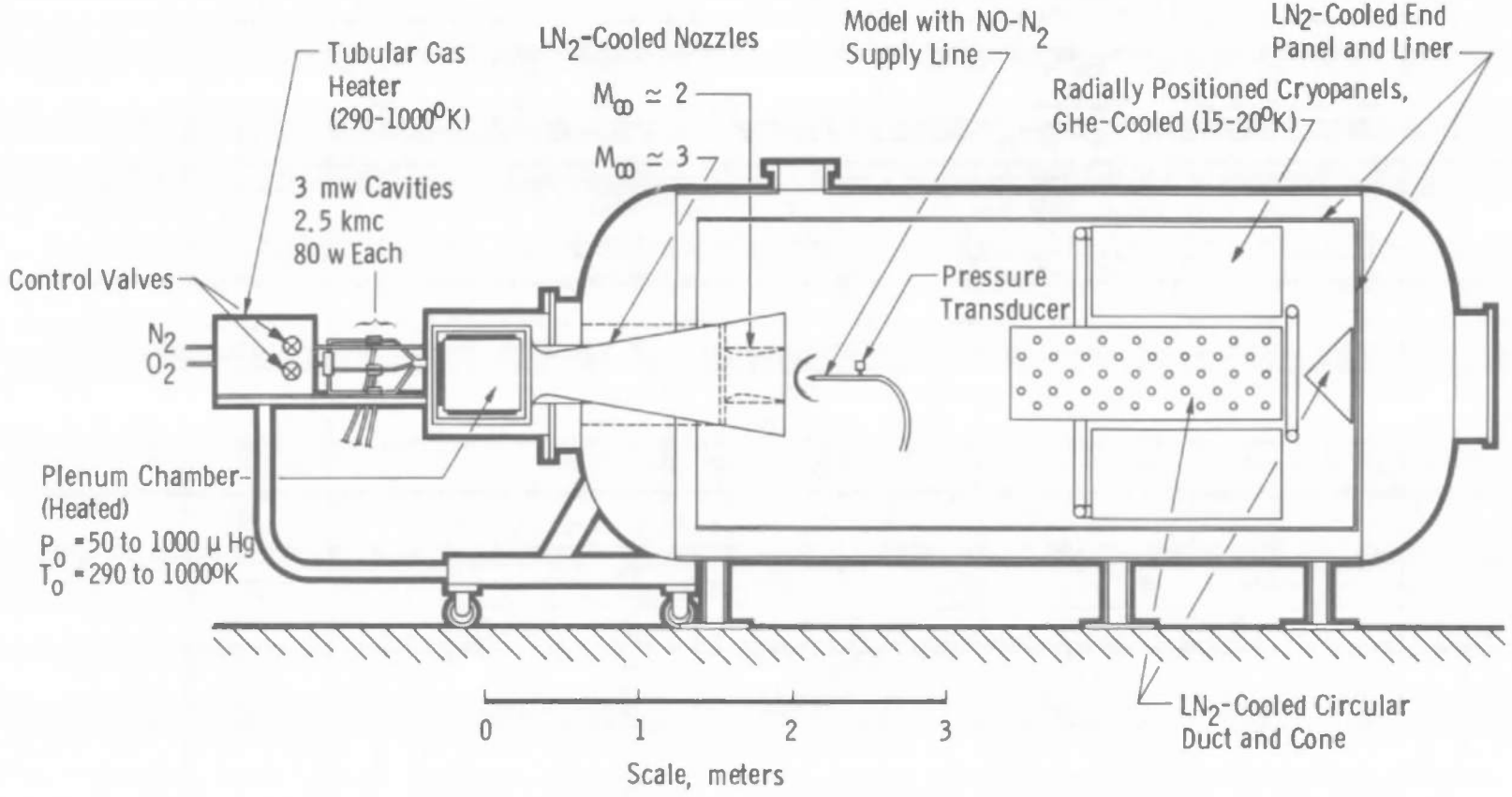

Fig. 2 Schematic of Low Density Tunnel (Aerospace Research Chamber (8V))

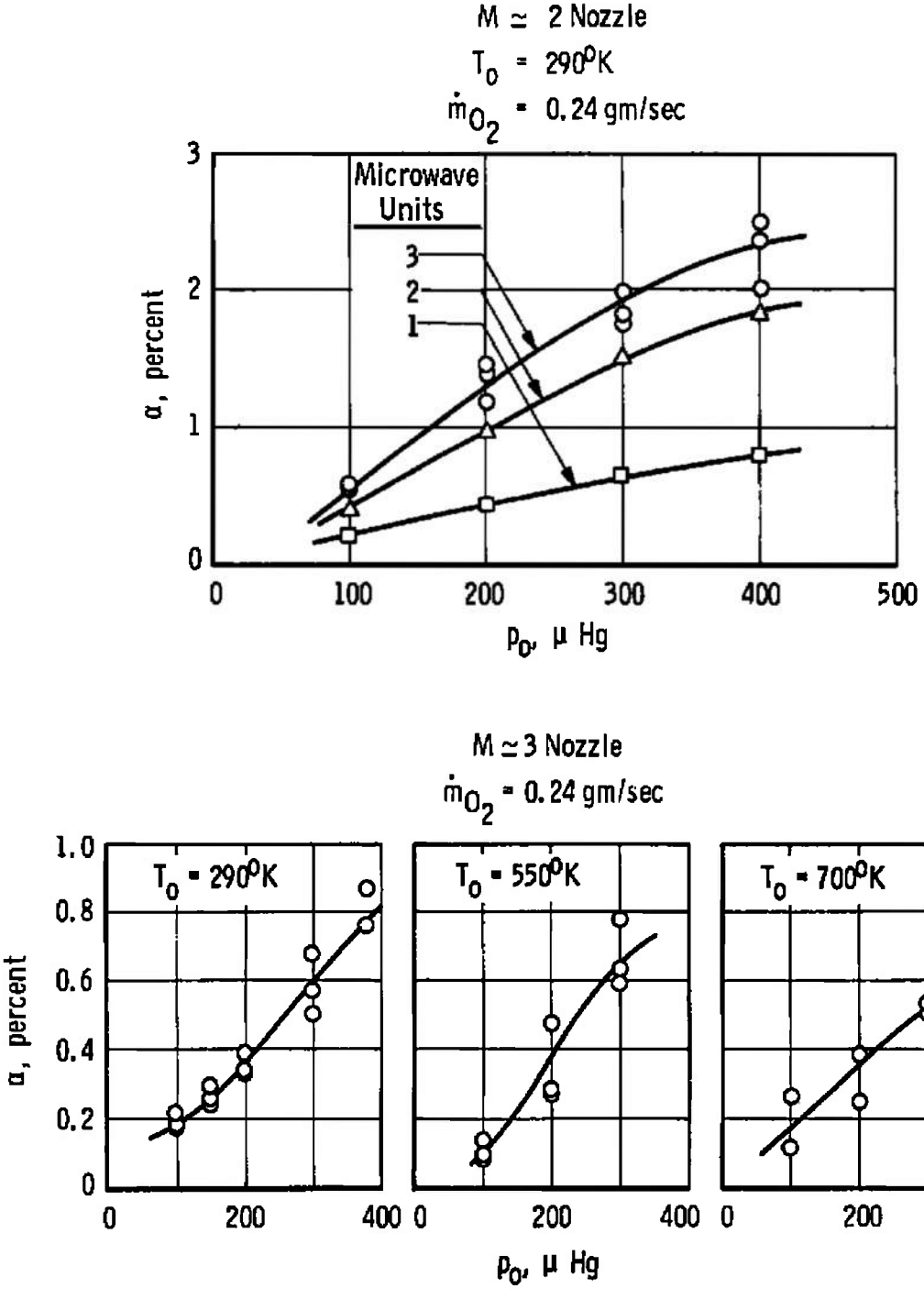

Fig. 3 Atomic Oxygen Calibration Results

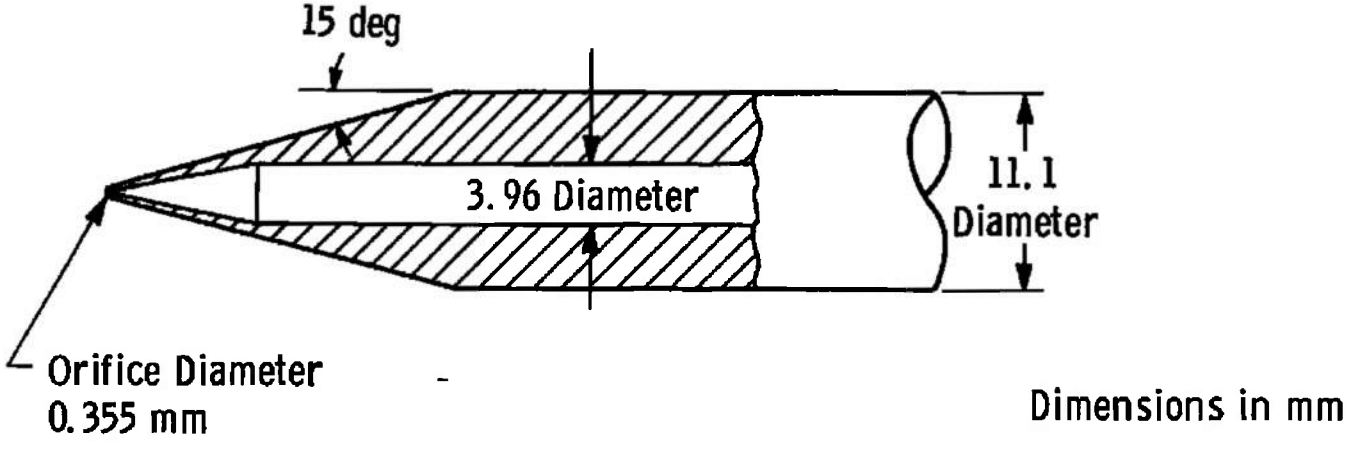

Fig. 4 Geometry of Test Model

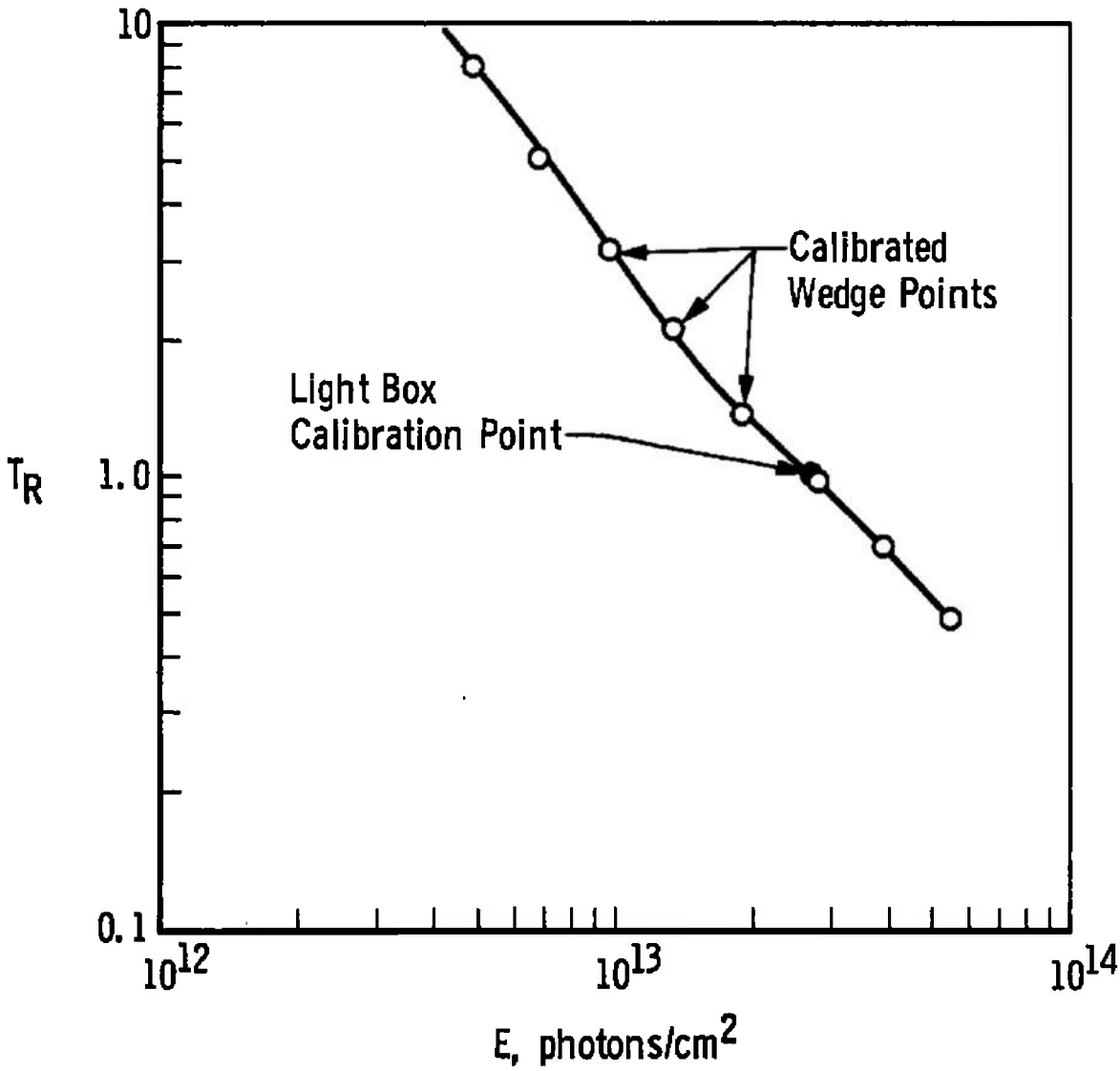

Fig. 5 Film Negative Calibration

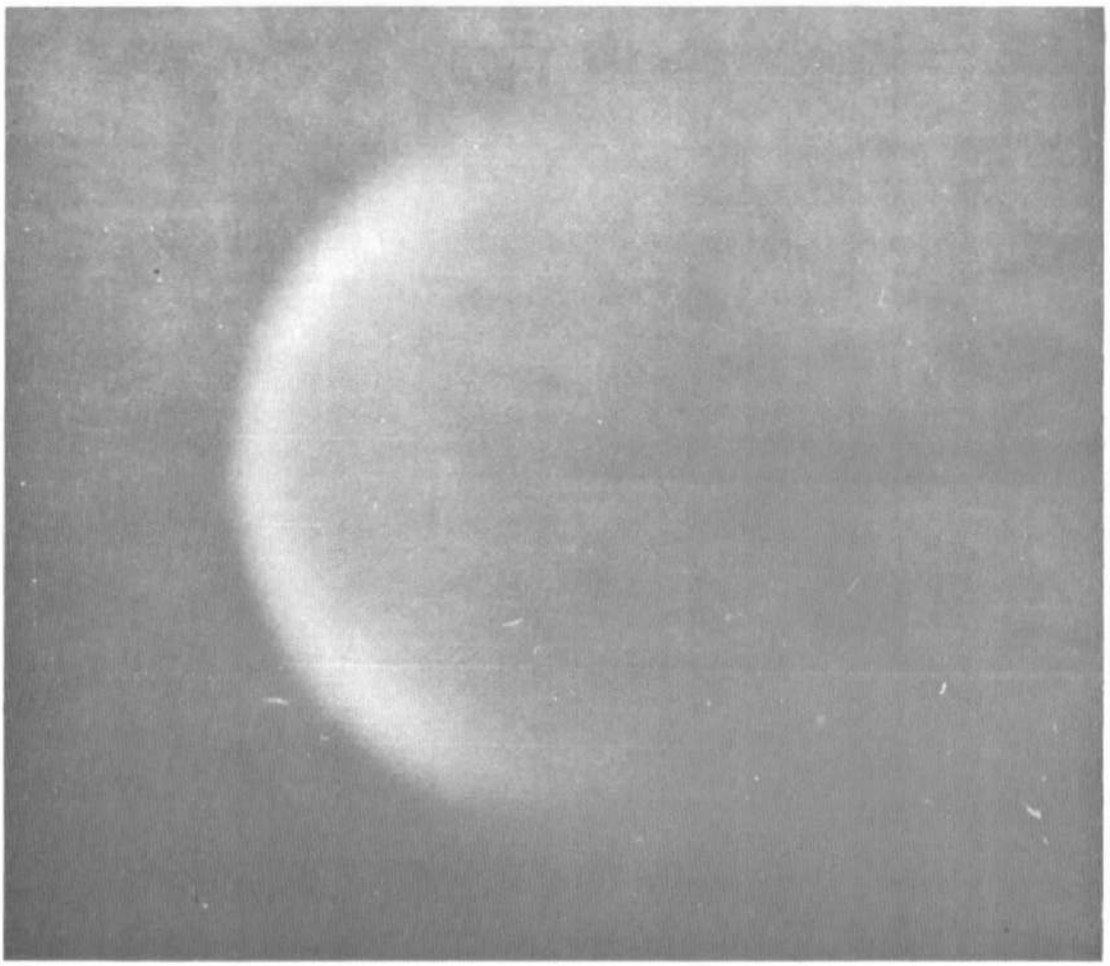

Fig. 6 Typical Photograph of Headglow

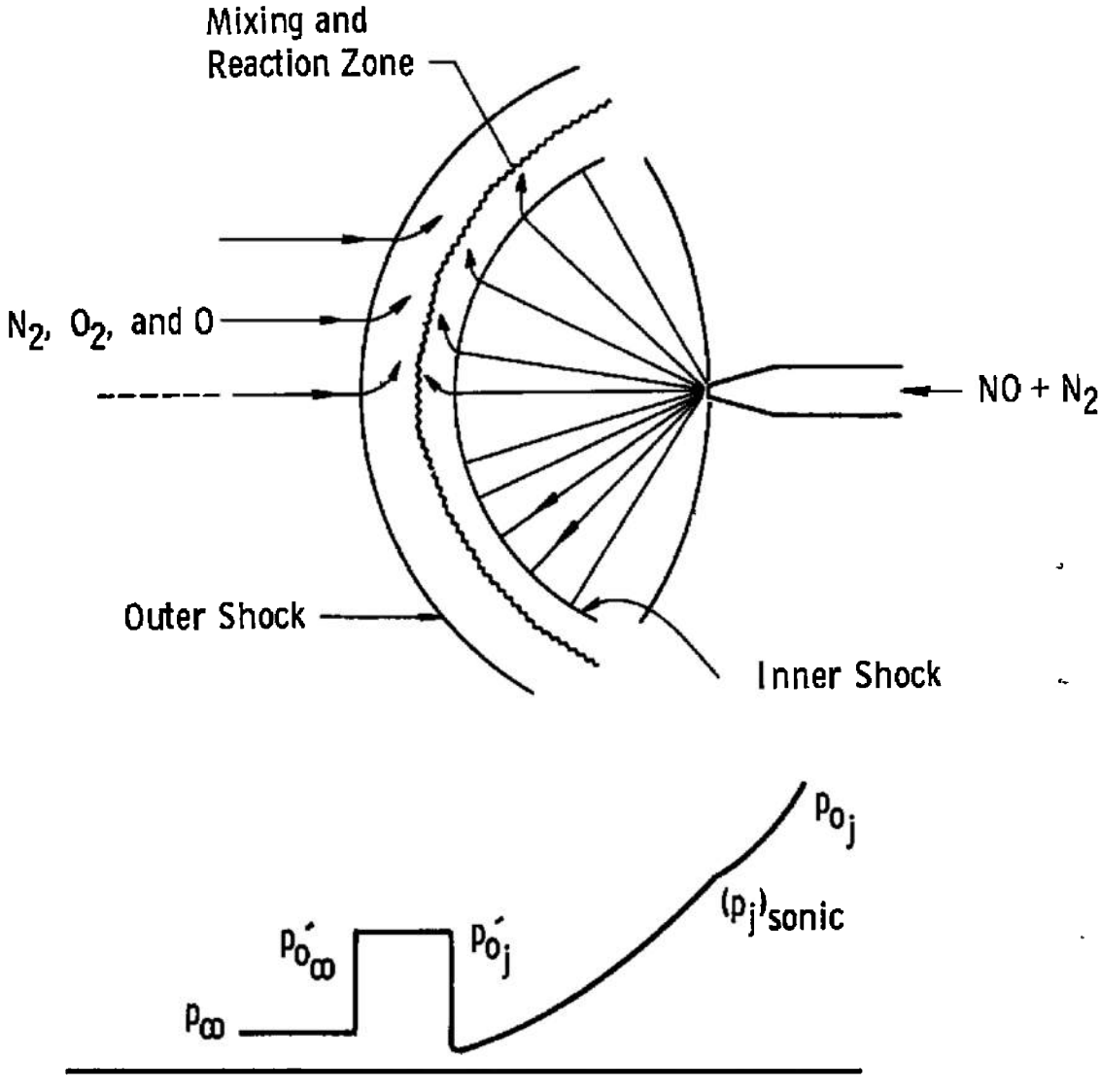

Fig. 7 Sketch of Headglow Flow Field

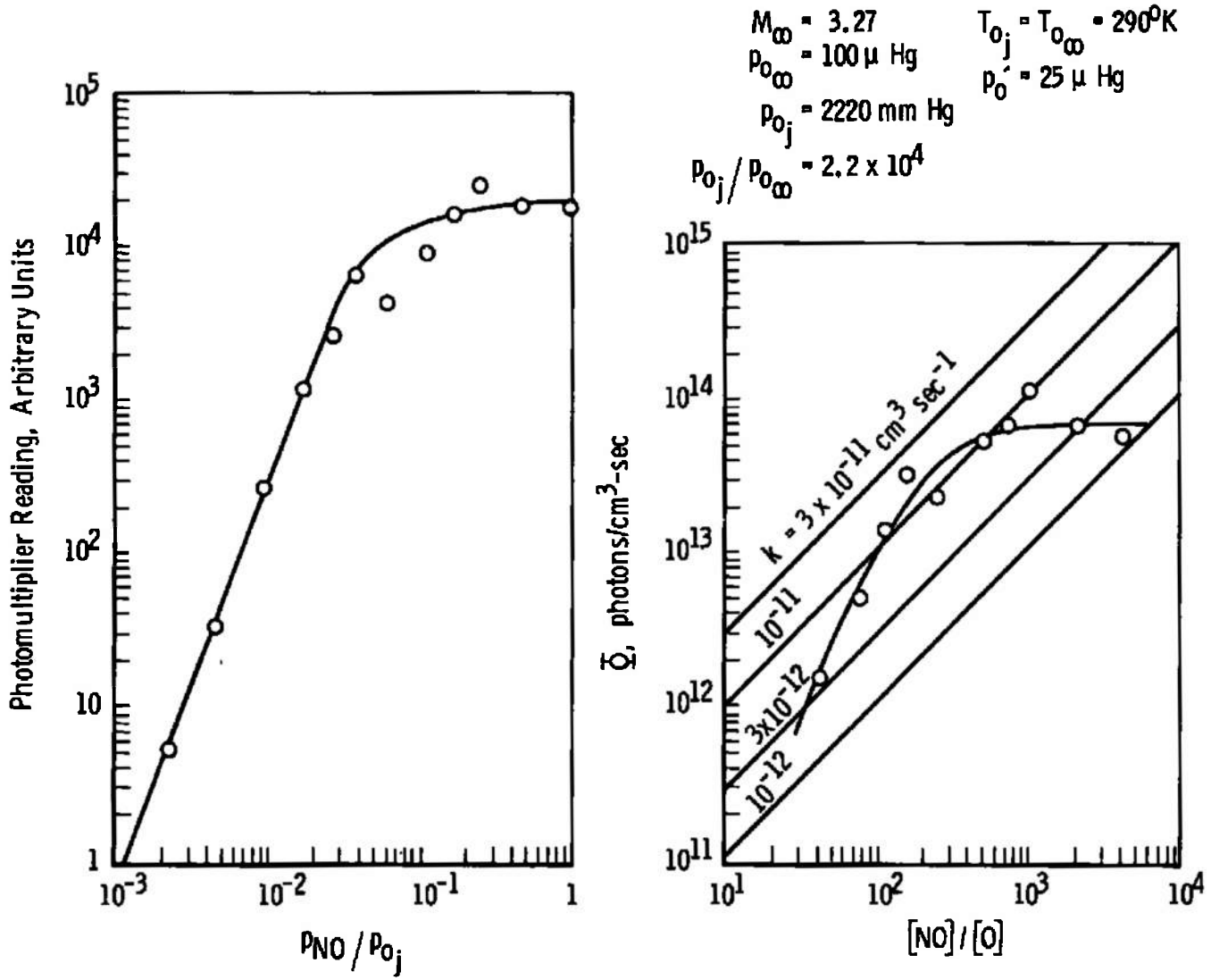

Fig. 8 Typical Variation of Headglow Radiation with NO Concentration in Forward Jet, p_o' = 25 μ Hg

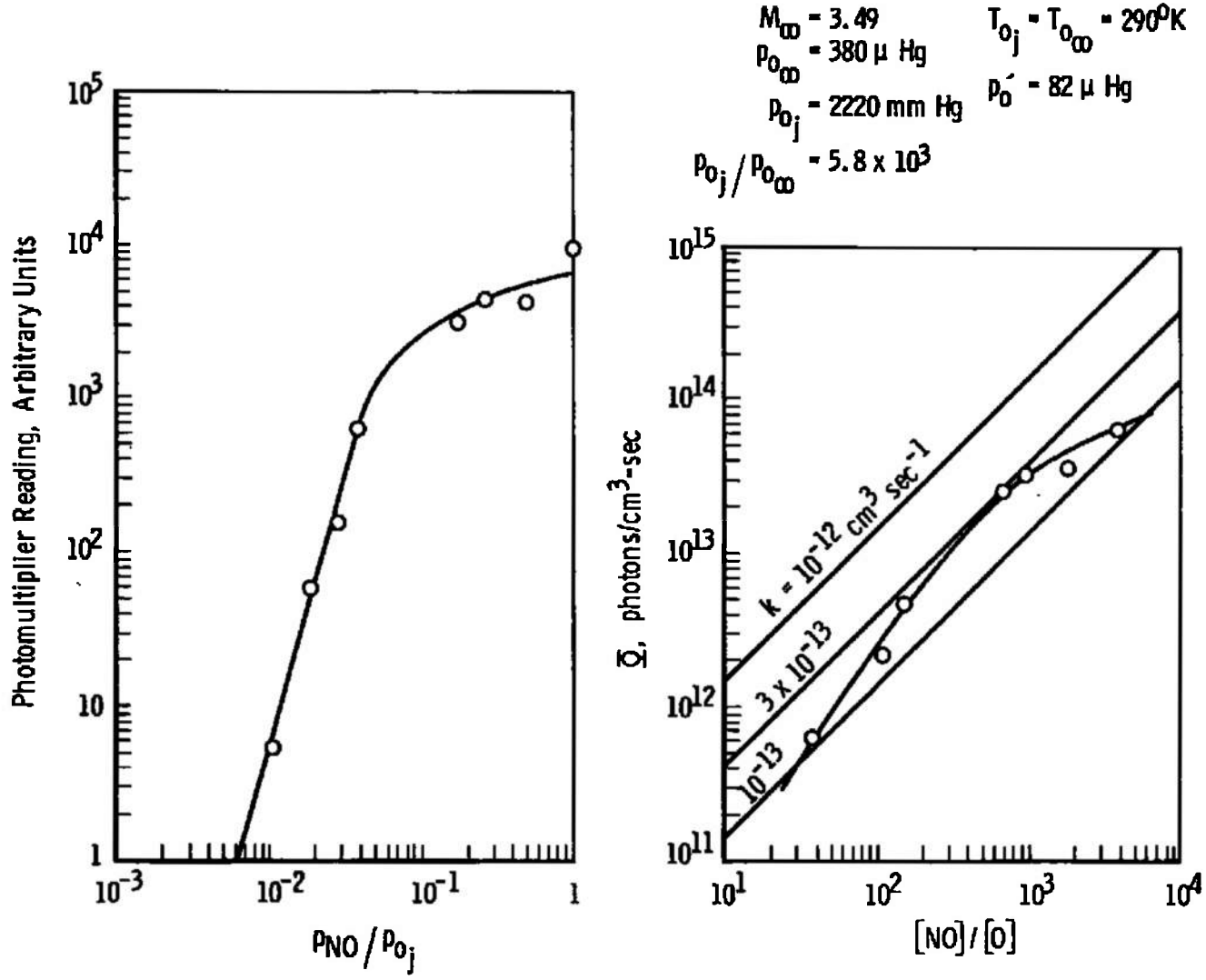

Fig. 9 Typical Variation of Headglow Radiation with NO Concentration in Forward Jet, $p_o' \approx 82$ μ Hg

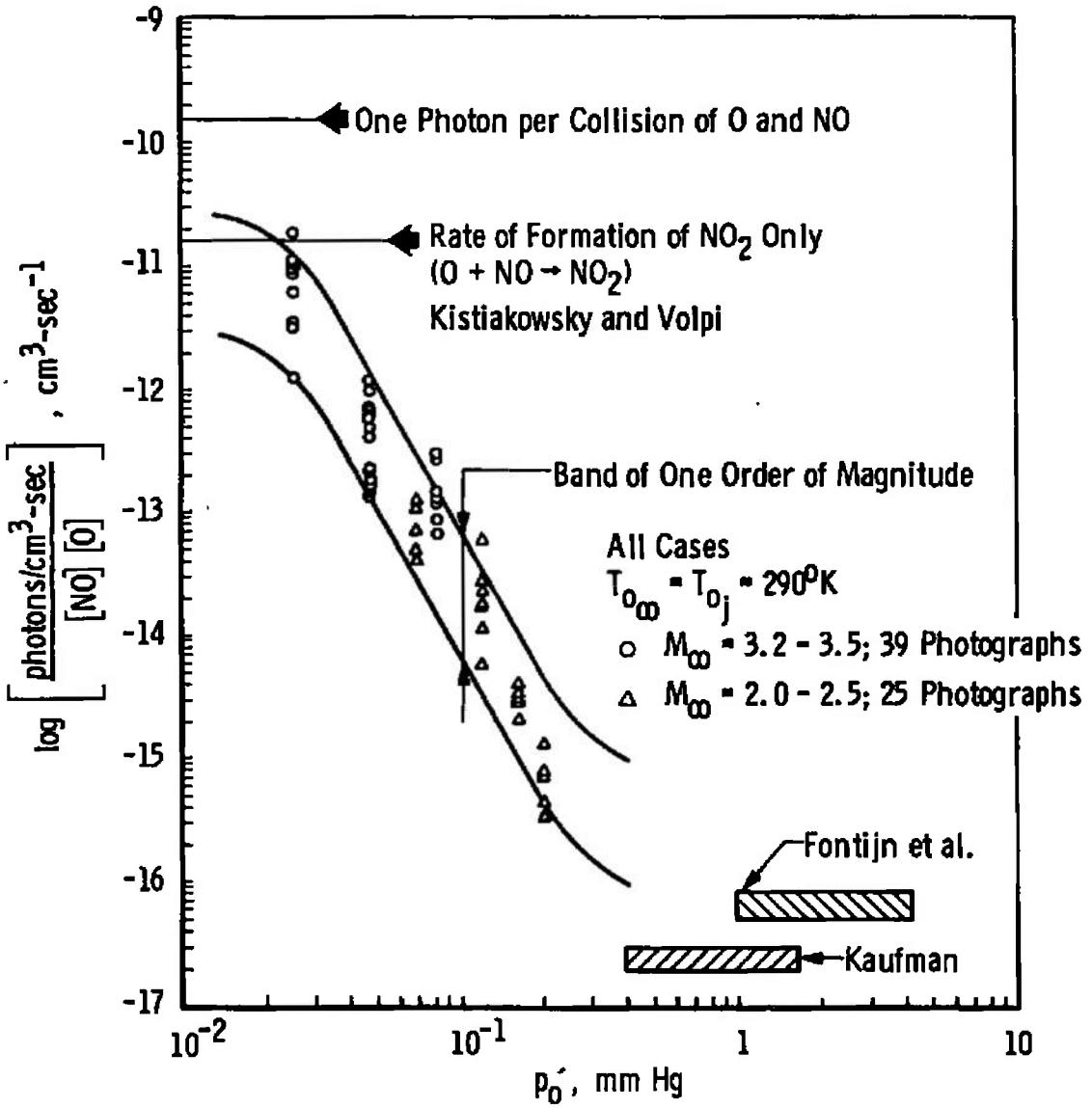

Fig. 10 Summary of Photon Production Rates at Stagnation Point, Determined from Photographs

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