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EFFECTS OF NONEQUILIBRIUM PHENOMENA ON RADAR TRANSMISSION - NON--ETC(U)
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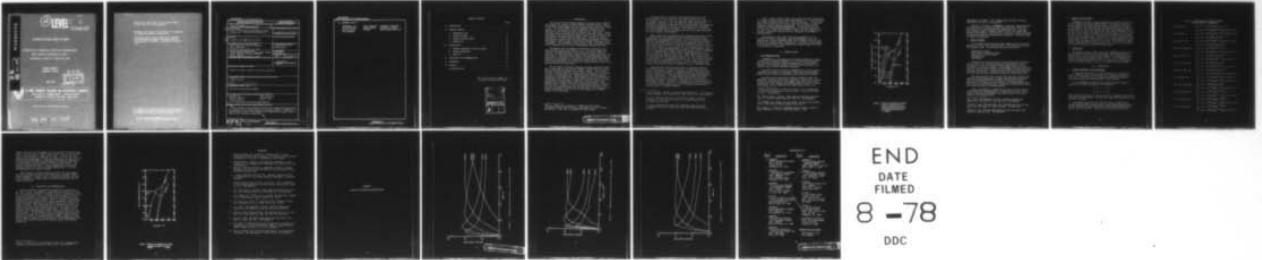
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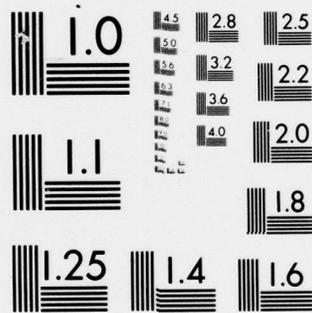
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TECHNICAL REPORT ARBRL-TR-02058

EFFECTS OF NONEQUILIBRIUM PHENOMENA
ON RADAR TRANSMISSION -
NONEQUILIBRIUM IONIZATION

Joseph Lacetera
Gaelen R. Daum

May 1978

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Nonequilibrium
Blackout

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I. INTRODUCTION

The Ballistic Missile Defense Advanced Technology Center (BMDATC) must address the prospect of their defensive systems having to operate in atmospheric environments which have been strongly modified by nuclear bursts. It has been shown¹ that the atmospheric properties most important to the operation of such systems are the free-electron densities and free-electron-density gradients in a radar path. Phenomenological models describing the operation of BMD radar systems in such environments use nonequilibrium chemical kinetics models to follow the deionization of the air along the radar path. Various reionization processes are accounted for, including the photodetachment of electrons from negative ions by radiant flux from the fireball (bomblight photodetachment). The assumption is made, however, that the fireball itself may be described by an LTE (Local Thermodynamic Equilibrium) model.

The general case of atmospheric deionization following a 5-MT burst has been described by Carney et al.¹ This report showed that negative ions formed exterior to the fireball may be an effective reservoir of charge from which electrons can be removed by photons from the fireball. The duration and extent of the photodetachment process depends on the conditions in the fireball, the absorptive and emissive properties of the heated air between the fireball and the radar path, and the photodetachment cross sections and concentrations of the various negative ions in the radar path.

Thus in order to accurately describe the photon spectrum incident upon a radar path far from the hot fireball, one must first accurately describe the photon sources in the fireball surface. This in turn requires a detailed knowledge of the interior structure of the fireball in terms of ion densities, gas temperatures and pressures, and the gradients of these quantities. These properties can be determined for temperatures above 8000 K by an atomic physics model which calculates the absorption and emission of radiation by positive atomic ions. For temperatures less than 8000 K, which occur in the region of space between the hot fireball interior and proposed radar paths, a chemical kinetics model is required to provide an accurate prediction of the molecular ion population densities relevant both to the absorptive and emissive properties of the air and to the transport of low-energy photons to the radar path.

¹Bruce W. Carney, E.L. Lortie, M. D. Kregel, and F.E. Niles, "Atmospheric Deionization Following a 5 MT Burst at 30 km Altitude," BRL Report No. 1671, Sep 73, Confidential (AD# 527883L).

Previous studies by Lacetera² have indicated that the total electron density in the fireball is relatively insensitive to the LTE assumption at very early times when only highly ionized atomic ions are present. However, it was also shown that when nonequilibrium population distributions among the atomic ions did exist, the absorptive and emissive properties of the hot gas were significantly different from those predicted by an LTE model. In particular it was shown that the actual fireball emission spectrum would not be correctly predicted by a code using the LTE assumption.

Other support for the hypothesis that nonequilibrium effects could be significant in this problem has been provided by Schleuter and Sappenfield³, and Sutherland and Zinn⁴. One of the primary purposes of Schleuter and Sappenfield's calculations was to provide information on late-time ionization relevant to the radar propagation studies. They showed that more than half of the thermal radiation from the fireball at times of the order of 1 sec, was above 3.2 ev in photon energy. This is sufficient to cause photodetachment of electrons from important negative ions. At temperatures of 4000 to 6000 K the opacity of air is primarily due to NO and from 1000 to 5000 K the principal emission from air is from the rotation-vibration bands of NO. Above 1000 K there is also a contribution from the electronic bands of NO. They calculated that NO would persist at a nonequilibrium density for a few seconds after the fireball cools to approximately 0.5 ev. In such a case an LTE calculation produces an uncertainty in absorption and a corresponding uncertainty in the amount of emission radiation in the 4-7 ev photon range.

Sutherland and Zinn⁴ have shown that for situations involving rapid cooling such as in nuclear fireballs, electron densities and negative ion densities remain in nonequilibrium for times significant in radar absorption problems, in temperature regions corresponding to potential radar paths. They have constructed a computational model of chemical kinetics in air, at temperatures between 300 K and 4000 K, including 1360 chemical reactions. They have also constructed an abbreviated 8-specie, 31-reaction set designed to be incorporated into fluid dynamics codes. This set is calibrated against the 1360-reaction set and gives good agreement with it for situations involving rapid cooling such as in a nuclear fireball.

²Joseph Lacetera, "Effects of Nonequilibrium Phenomena on X-Ray Transport in the Atmosphere (U)," BRL Report No. 1636, Feb 73, SRD. (AD #524945L)

³Warren A. Schleuter and Dale S. Sappenfield, "Effect of Nuclear Detonations at 35 and 45 km Altitude (U)," Mission Research Corp., DNA 2879F, Aug 1972, SRD.

⁴C. Dexter Sutherland and John Zinn, "Chemistry Computations for Irradiated Hot Air," Los Alamos Scientific Laboratory, LA-6055-MS, Aug 1975.

Figure 1 shows results of their calculations with the 8-specie model for systems starting at 2500 K and cooled abruptly to T, while subjected to a steady ionization source of $Q_Y = 10^{14}$ ion pairs/cc. The plotted values of N- and Ne are for 0.01 sec after the cool-down for three different fireball radiating temperatures. It can be seen that the extent of the nonequilibrium perturbation in a potential radar path (T = 500 K) is a function of fireball radiance, and that negative ions provide a substantial reservoir of electrons which can be freed by photodetachment.

The remainder of this report discusses the results of a non-equilibrium fireball calculation using the THUNDERBALL code⁵, and calculations of NO equilibration times using a chemical kinetics model based on the reaction rates of Reference 4. The results are described in terms of the effect of the nonequilibrium fireball upon the radiant flux incident on a radar path.

II. NUMERICAL MODEL

A. THE THUNDERBALL CODE

THUNDERBALL is a nonequilibrium, radiation-hydrodynamics, photon transport and atmospheric response code. Photon and mass transport are calculated in eulerian spherical geometry, and ion population densities are calculated directly from the interactions between the radiation field and the responding atmosphere.

The photon transport routine in THUNDERBALL has been verified in comparisons with other X-ray transport codes, and the hydrodynamics model has been verified by comparisons with calculations by RADFLO.

The code also has the option of using a parametric model for generating late-time nuclear blast conditions. This option, provided by Miller⁶, utilizes the Air Force Weapons Laboratory's one-kiloton nuclear blast code⁷ together with modified Sach's scaling⁸ to obtain thermodynamic initial conditions for large nuclear bursts in an

⁵Joseph Lacetera, Gaelen R. Dawn, and George J. Klem, "THUNDERBALL, the BRL Nonequilibrium Nuclear Fireball Code," BRL Report No. 1992, Jun 1977. (AD #A044531)

⁶M.S. Miller and L.J. Puckett, "Shock Induced Atmospheric Reionization (U)," BRL Memo Report 2440, Feb 1975, (S). (AD #B002709L)

⁷C.E. Needham, M.L. Havens, and C.S. Knauth, "Nuclear Blast Standard (1KT)," AFWL Technical Report, AFWLTR-73-55, Apr 73.

⁸P.A. Ellis, D.C. Sachs, F.H. Shelton, and J.F. Moulton, "Nuclear Blast Phenomena (U)," Vol 1, DASA 1200-1, Mar 71, SRD.

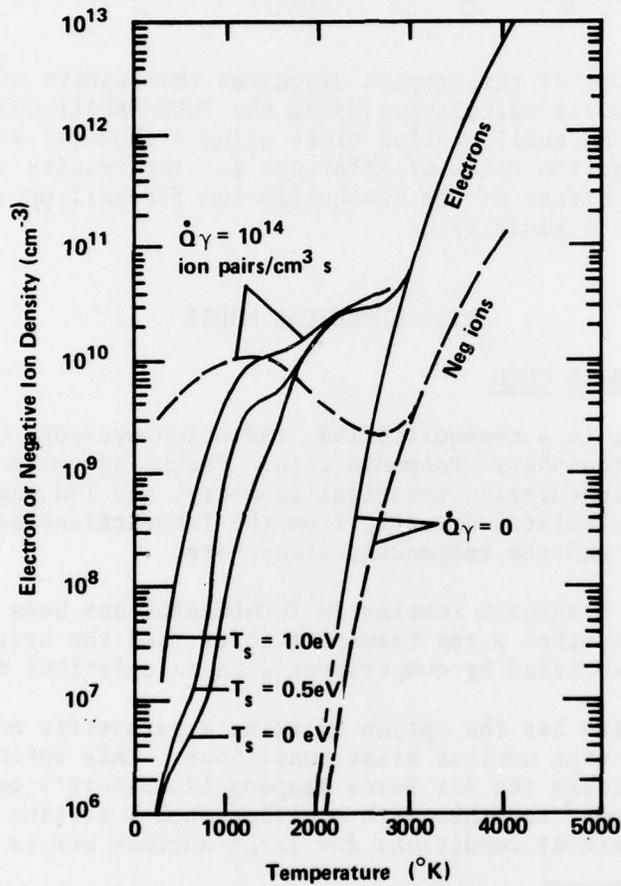


Figure 1. Electron and negative ion number densities calculated with the 8 -species lumped-parameter model, representing systems started at $T \geq 2500$ K.

inhomogeneous atmosphere. This scaling method provides a distinct improvement over Sach's scaling theory⁹.

The atomic ion model in THUNDERBALL is based on a photoionization cross section set calculated by Daum¹⁰ using the XSECT code¹¹ and collisional cross sections taken from experimental data. The code also includes a model of chemical kinetics in air for temperatures between 300 and 8000 K. The code will, at the user's option, calculate the atomic ion populations using the Saha equation, if the user wishes to assume LTE conditions.

B. ATOMIC ION MODEL

In the nonequilibrium mode this model computes atomic-ion specie populations directly from the interaction of the radiation field with the atmospheric gas. The important physical processes are:

- photoelectric effect
- auger effect/fluorescence effect
- ionization by electron impact
- bremsstrahlung
- scattering

Atomic and ionic photoionization cross sections were calculated with the XSECT code, which uses the dipole approximation for the calculation of cross sections and Hartree-Slater functions for the evaluation of the matrix elements. Good agreement with experiment and with more sophisticated calculations was obtained for those cases where comparison was possible. Length, velocity, and acceleration forms of the cross sections were calculated and generally agreed within 10%.

The photoionization cross sections were then used, employing the principle of detailed balance to compute inverse emissive reaction rates. Stimulated emission is included in the detailed balance equations for exactness, and the principles of detailed balance and mass action are used extensively to ensure that interrelations between the various processes are treated properly.

Kinetic equilibrium is assumed for the free electrons; however, excess energy from photoelectrons is accounted for in terms of collisional ionization and thermalization which increases the kinetic energy of the average particle.

⁹R.G. Sachs, "The Dependence of Blast on Ambient Pressure and Temperature," BRL Report No. 466, May 1944, (AD# AT139393).

¹⁰Gaelen R. Daum, "Photoionization Cross Sections for O, N, Al, and Their Ions," BRL Report No. 1894, Jun 1976. (AD #B012453L)

¹¹Gaelen R. Daum, "The BRL Photoionization Code Code XSECT," BRL Report No. 1869, Mar 1976. (AD #B010267L)

C. CHEMICAL KINETICS MODEL

THUNDERBALL includes a computational model of chemical kinetics in air for temperatures between 300 and 8000 K. The model is based on a minimum reaction set for NO production given by Sutherland and Zinn⁴. It is complete in the sense that for every reaction we also include the inverse process. The rate constants for these corresponding pairs of reactions are related to the equilibrium rate constant through $k_r = k_f / k_{eq}$. The set is also closed with respect to the seven species O, O₂, N, N₂, NO, NO₂, and O₃. Table I lists the reactions and their rate constants as a function of temperature. In table I, M is used to denote the total neutral number density. Other processes such as photodetachment, photo-dissociation, and photoionization are included in the atomic ion model; however, photodetachment of electrons from O₂⁻, and electron attachment to O₂ are included in the chemical kinetics model.

D. SSCHEM CODE

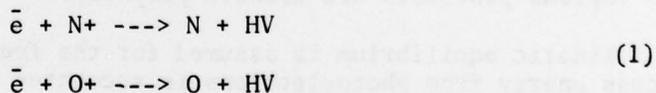
The chemical kinetics model used in the THUNDERBALL code has also been separately coded under the name SSCHEM. Given temperature and density, this code will calculate the relaxation to equilibrium of the various species from arbitrary initial values. It can therefore be used to calculate equilibration times for situations in which temperatures have changed abruptly, such as in a rapidly cooling fireball.

III. CALCULATIONS

A. FIREBALL CHEMISTRY AT 30-KM ALTITUDE

Calculations were made of a nuclear fireball at an altitude of 30 km. Chemical kinetics calculations were included beginning from T=1.0 sec. Calculations were also made using SSCHEM to determine equilibration times for the NO molecule at this altitude.

In the THUNDERBALL calculation the chemical kinetics model was coupled to the atomic-ion model via the reactions:



These reactions provided a source of N and O as a boundary condition for the chemical kinetics model. Equilibration times were seen to be on the order of a few msec.

The SSCHEM calculations also indicated that for temperatures between 3500 and 8000 K equilibration of the NO specie from arbitrary initial conditions occurs on the order of msec when temperature and density are held constant. These results indicate that for burst

Table I. Reaction Rates⁴ Used in Chemical Kinetics Model in Thunderball

| | |
|--|--|
| $N + O_2 \rightleftharpoons NO + O$ | $k_f = 2.3 \times 10^{-11} \exp(-4000/T) \text{ cm}^3 \text{ s}^{-1}$ |
| | $k_r = 6.3 \times 10^{-12} (T/300)^{-0.11} \exp(-20141/T) \text{ cm}^3 \text{ s}^{-1}$ |
| $O + N_2 \rightleftharpoons NO + N$ | $k_f = 2.0 \times 10^{-10} (T/300)^{0.05} \exp(-37986/T) \text{ cm}^3 \text{ s}^{-1}$ |
| | $k_r = 5.1 \times 10^{-11} \exp(-250/T) \text{ cm}^3 \text{ s}^{-1}$ |
| $O_2 + O_2 \rightleftharpoons O + O_3$ | $k_f = 3.8 \times 10^{-12} (T/300)^{0.44} \exp(-49407/T) \text{ cm}^3 \text{ s}^{-1}$ |
| | $k_r = 1.5 \times 10^{-11} \exp(-2240/T) \text{ cm}^3 \text{ s}^{-1}$ |
| $NO + O + M \rightleftharpoons NO_2 + M$ | $k_f = 1.0 \times 10^{-31} (T/300)^{-2.5} \text{ cm}^6 \text{ s}^{-1}$ |
| | $k_r = 2.6 \times 10^{-6} (T/300)^{-2.13} \exp(-36161/T) \text{ cm}^3 \text{ s}^{-1}$ |
| $NO_2 + O \rightleftharpoons NO + O_2$ | $k_f = 1.7 \times 10^{-11} \exp(-300/T) \text{ cm}^3 \text{ s}^{-1}$ |
| | $k_r = 3.4 \times 10^{-k2} (T/300)^{0.11} \exp(-23508/T) \text{ cm}^3 \text{ s}^{-1}$ |
| $O + O_2 + M \rightleftharpoons O_3 + M$ | $k_f = 1.1 \times 10^{-34} \exp(510/T) \text{ cm}^6 \text{ s}^{-1}$ |
| | $k_r = 2.3 \times 10^{-9} (T/300)^{0.04} \exp(-11693/T) \text{ cm}^3 \text{ s}^{-1}$ |
| $NO + O_3 \rightleftharpoons NO_2 + O_2$ | $k_f = 9.5 \times 10^{-13} \exp(-1300/T) \text{ cm}^3 \text{ s}^{-1}$ |
| | $k_r = 1.2 \times 10^{-12} (T/300)^{0.33} \exp(-25258/T) \text{ cm}^3 \text{ s}^{-1}$ |
| $N + O_3 \rightleftharpoons NO + O_2$ | $k_f = 3.1 \times 10^{-11} (T/300)^{0.50} \exp(-1200/T) \text{ cm}^3 \text{ s}^{-1}$ |
| | $k_r = 2.1 \times 10^{-12} (T/300)^{0.83} \exp(-64508/T) \text{ cm}^3 \text{ s}^{-1}$ |
| $NO + NO + O_2 \rightleftharpoons 2NO_2$ | $k_f = 6.6 \times 10^{-39} \exp(526/T) \text{ cm}^6 \text{ s}^{-1}$ |
| | $k_r = 7.3 \times 10^{-12} (T/300)^{-0.23} \exp(-947/T) \text{ cm}^3 \text{ s}^{-1}$ |
| $N + O + M \rightleftharpoons NO + M$ | $k_f = 1.1 \times 10^{-32} (T/300)^{-0.50} \text{ cm}^6 \text{ s}^{-1}$ |
| | $k_r = 1.5 \times 10^{-8} (T/300)^{-0.13} \exp(-75511/T) \text{ cm}^3 \text{ s}^{-1}$ |
| $N + N + M \rightleftharpoons N_2 + M$ | $k_f = 4.6 \times 10^{-33} (T/300)^{-1.70} \text{ cm}^6 \text{ s}^{-1}$ |
| | $k_r = 2.5 \times 10^{-8} (T/300)^{-1.29} \exp(-113246/T) \text{ cm}^3 \text{ s}^{-1}$ |
| $O + O + M \rightleftharpoons O_2 + M$ | $k_f = 3.0 \times 10^{-33} (T/300)^{-2.90} \text{ cm}^6 \text{ s}^{-1}$ |
| | $k_r = 1.6 \times 10^{-8} (T/300)^{-2.43} \exp(-59370/T) \text{ cm}^3 \text{ s}^{-1}$ |

altitudes of 30 km and lower the LTE approximation is valid for computing molecular species which contribute to the fireball emission spectrum.

B. SSCHEM CALCULATIONS

SSCHEM calculations were made for temperatures between 3000 and 8000 K, and densities corresponding to altitudes of 30, 60, and 90 km. Because we had not done fireball calculations at 60- and 90-km altitudes, it was not possible to estimate the actual time-dependent NO fraction that would occur as recombination (equation 1) led to the formation of N and O atoms in the fireball. Furthermore, the SSCHEM code in its present form is not valid at temperatures above 8000 K. For these reasons we initialized the SSCHEM calculations with a system consisting of N₂ and O₂ molecules for various initial temperatures. Temperature and density were then held constant while the time-dependent specie populations were monitored. Equilibration times, based on approach to steady-state values, are shown in Table II. Plots of the species populations as a function of time, starting from arbitrary initial conditions, are shown in the appendix.

| Altitude (km) | Particle Density (#/CC) | TEQ (EST) (sec) | TEQ (FIT) (sec) |
|------------------|----------------------------|--------------------|--------------------|
| 30 | 7.656E+17 | .001 | .0017 |
| 60 | 1.276E+16 | .100 | .102 |
| 90 | 1.312E+14 | 10.0 | 9.9 |

The values of TEQ (EST), in Table II, were estimated from the plots of the NO population relaxation shown in the appendix. If we assume that the equilibration times are inversely proportional to particle density, we can relate the three estimated equilibration times by multiplying TEQ by the density in each case. A best fit is attained by using C=1.3E15 in the equation

$$TEQ = C/M$$

where M is the particle density. The resultant equilibration times are listed as TEQ (FIT) in the table.

C. ANALYSIS

The SSCHEM calculations show that for intermediate altitudes (up to 90 km), the NO population can remain in nonequilibrium for significantly long times. Figure A.3 shows the NO population in 90-km air as a function of time starting from our arbitrarily chosen initial conditions. The order-of-magnitude overshoot of the final equilibrium value in the 5500 K case does not represent the actual worst case underestimate by the LTE assumption in a fireball calculation. However, we estimate from these plots that a worst case uncertainty in the NO population in the fireball surface caused by the LTE assumption could be an underestimate by at least an order of magnitude since recombination (equation 1) will also be very slow at these altitudes.

In LTE at these temperatures NO contributes approximately 30 percent of the total opacity while constituting a few percent of the total population. In this case the opacity per particle fraction normalized to total opacity can be written for NO as

$$O(\text{NO}) = .30/.01 = 30$$

and for all other species combined as

$$O(\text{M}) = .70/.99 = .707$$

The ratio of the non-LTE opacity to the LTE opacity can then be written as

$$\frac{.88*O(\text{M}) + .12*O(\text{NO})}{.99*O(\text{M}) + .01*O(\text{NO})} = \frac{.88*.707+.12*30}{.99*.707+.01*30} = 4.22$$

A factor of twelve underestimate in the contribution of NO to the total opacity therefore corresponds to approximately a factor of four underestimate of the total opacity in the photon energy range relevant to the photodetachment of electrons from negative ions. Because the fireball is optically thin at these times, this corresponds to a factor of four underestimate of the photodetaching flux incident upon the fireball.

An estimate was made of the effect of increasing by a factor of four the photodetachment rate predicted by a radar effects code such as WEPH V¹² at a temperature point corresponding to a possible radar path outside the fireball. As temperature decreases with increasing radius, the detachment rate for electrons from negative ions becomes

¹²W.S. Knapp, "A Fortran Code for the Calculation of Ionization and Absorption Due to Nuclear Detonations (WEPH V)," Tempo Report No. 71TMP-2, General Electric Co., Apr 71, DASA2884-1.

smaller relative to the attachment rate for electrons to neutral molecules. It can be seen in Figure 2 that at 500 K the ratio of negative ions to free electrons is approximately 1000. In such a case the negative ions constitute a substantial reservoir of electrons which can be freed by photodetachment. Using these populations a lumped parameter calculation was made based on negative ion densities and free electron densities typical of 500 K air outside the fireball. The results indicate that a factor-of-four underestimate of the photodetachment rate would lead to a maximum underestimate of the free-electron density by a factor of three.

The situation is further complicated by the fact that whenever free-electron densities are not near equilibrium, transport effects become important¹³. However, this is a higher order effect and need only be addressed if nonequilibrium free-electron densities are considered significant.

IV. CONCLUSIONS AND RECOMMENDATIONS

We have performed a nonequilibrium fireball calculation for an altitude of 30 km, and chemistry calculations for altitudes up to 90 km. We conclude from these calculations that for nuclear burst calculations at altitudes of 30 km or lower the LTE assumption provides an accurate description of the fireball chemistry and therefore of the radiant emission from the fireball. There is, however, a significant uncertainty in the absorptive and emissive properties of the fireball surface calculated using the LTE assumption for nuclear fireballs at higher altitudes. This uncertainty and its temporal extent will be greatest at an altitude of approximately 90 km (at higher altitudes fireball formation itself is not significant). The effect can be quantified by a nonequilibrium fireball chemistry calculation at the 90-km altitude. If the results of such a calculation were considered significant, a small set of nonequilibrium fireball chemistry calculations could be done to provide a basis on which to modify the radar effects codes to account for the deviation of the photodetachment rate from the LTE value as a function of altitude, yield, and radiating temperature.

¹³Kevin S. Fansler, "On the Free-Electron Density in a Nuclear-Blast Environment," BRL Memo Report No. 2688, Sep 76. (AD #B014240L)

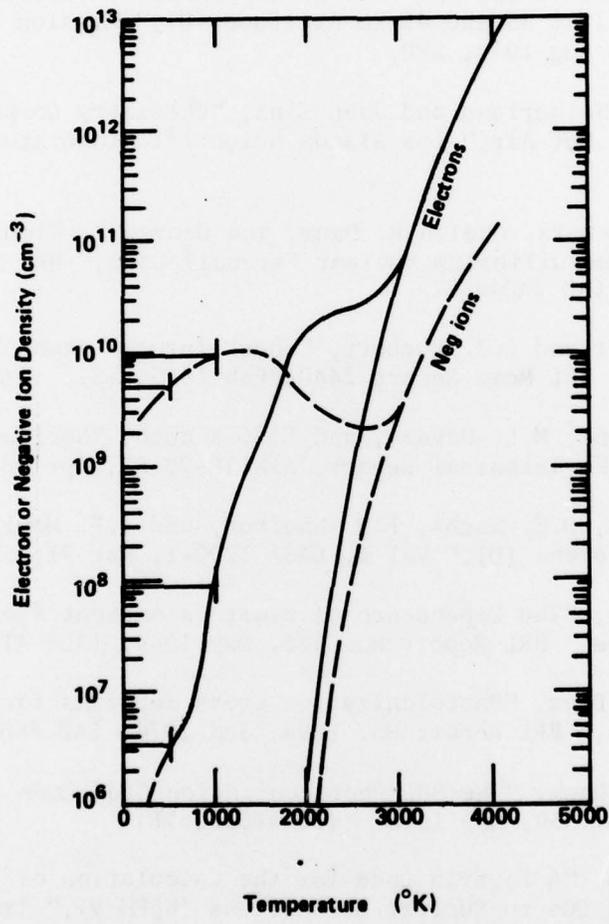


Figure 2. Electron and negative ion number densities calculated with the ^{s-} species model,

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APPENDIX
PLOTS OF NO POPULATION EQUILIBRATION

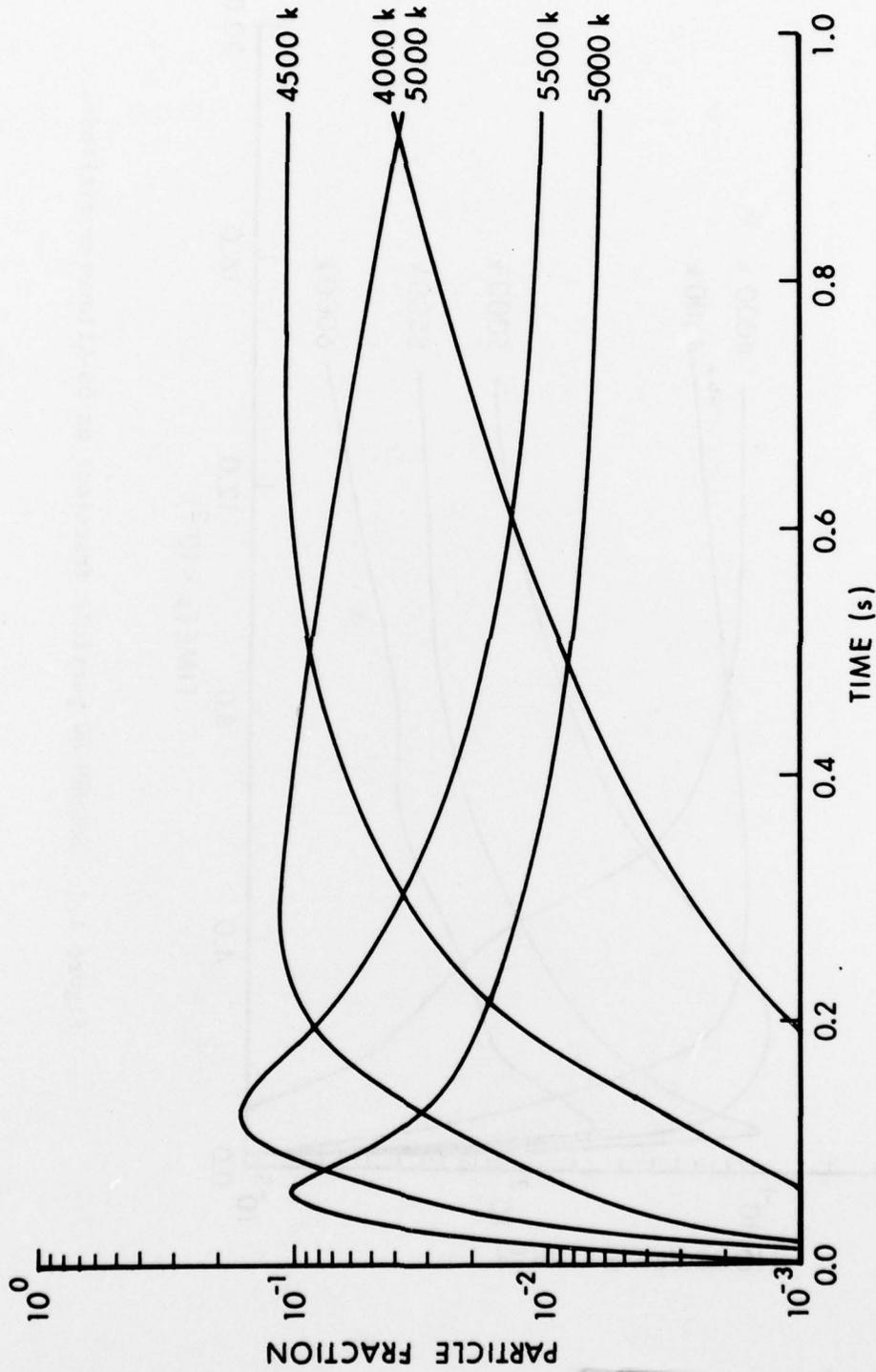


Figure A.1 SSCHENO particle fractions at 30-kilometer altitude.

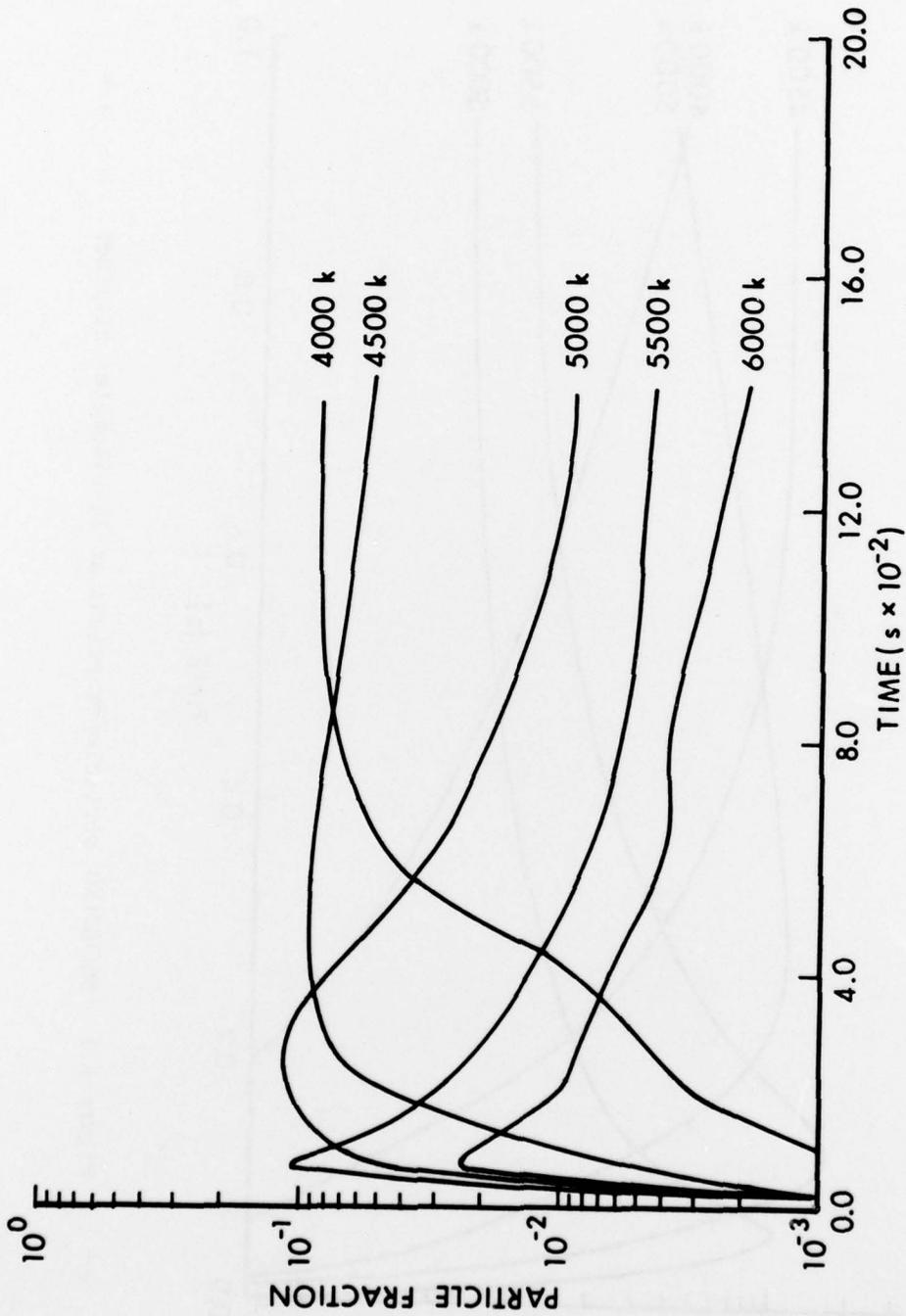


Figure A.2 SSChEM NO particle fractions at 60-kilometer altitude.

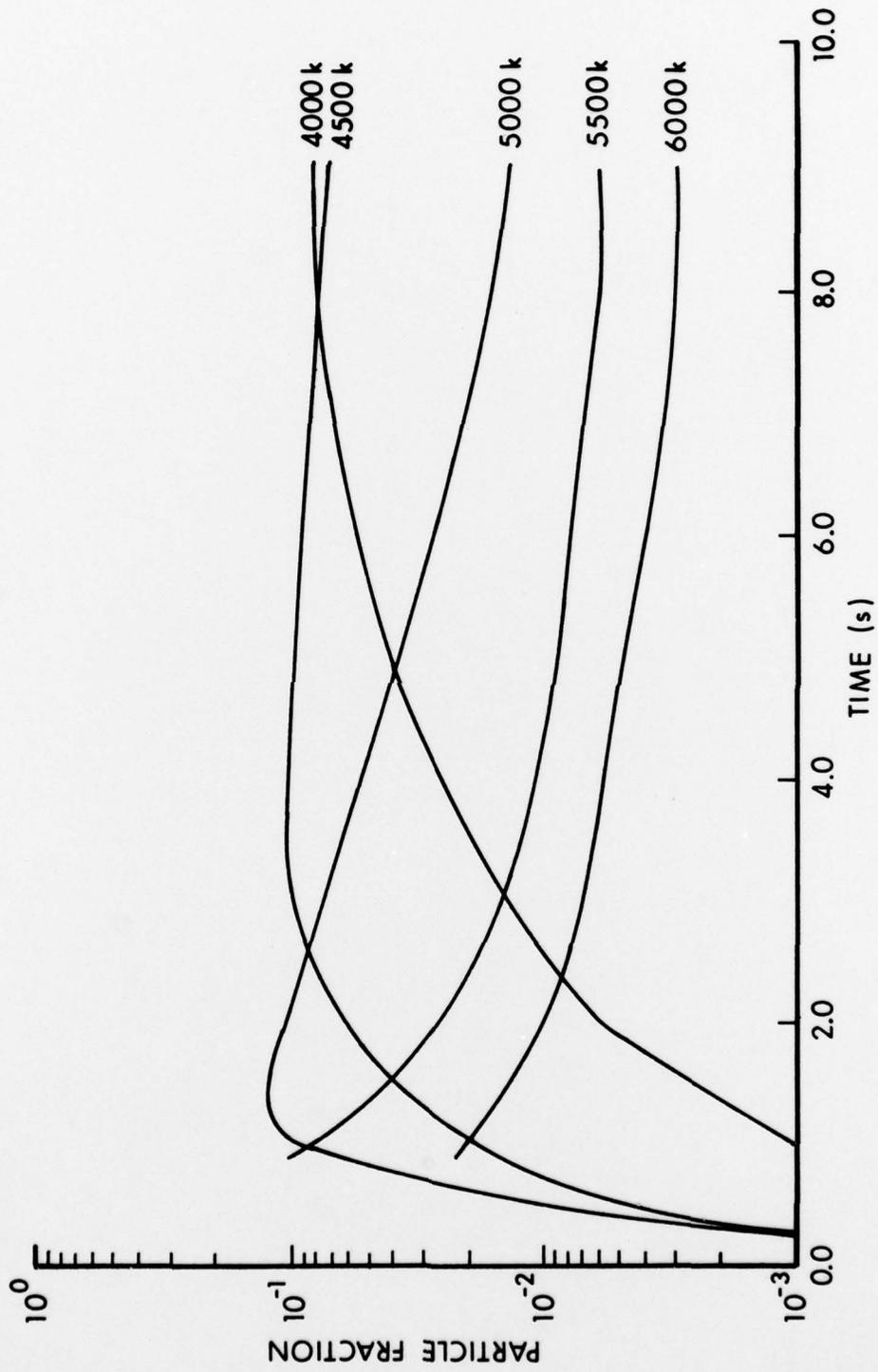


Figure A.3 SSCHEN NO particle fractions at 90-kilometer altitude.

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