PROBLEM OF PHOTOCHEMICAL EQUILIBRIUM OF OZONE IN PLANETARY ATMOSPHERES: OZONE DISTRIBUTION IN THE LOWER ATMOSPHERE OF MARS

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Several investigators have applied techniques used for the study of the ozone distribution in the terrestrial atmosphere to the atmospheres of other planets. This paper shows that such techniques cannot be applied indiscriminately to calculate photochemical equilibrium distribution of ozone in planetary atmospheres. Limitations associated with some of the earlier treatments of photochemical equilibrium distributions of ozone in planetary atmospheres are discussed, and a technique having more universal application is presented. In addition, ozone concentration profiles for the Martian atmosphere based on the results of the Mariner 4 radio occultation experiment and the more recent results from Mariner 6 and Mariner 7 have been calculated using this approach.
Problem of Photochemical Equilibrium of Ozone in Planetary Atmospheres: Ozone Distribution in the Lower Atmosphere of Mars

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SUMMARY

Several investigators have applied techniques used for the study of the ozone distribution in the terrestrial atmosphere to the atmospheres of other planets. This paper shows that such techniques cannot be applied indiscriminately to calculate photochemical equilibrium distribution of ozone in planetary atmospheres. Limitations associated with some of the earlier treatments of photochemical equilibrium distributions of ozone in planetary atmospheres are discussed, and a technique having more universal application is presented. In addition, ozone concentration profiles for the Martian atmosphere based on the results of the Mariner 4 radio occultation experiment and the more recent results from Mariner 6 and Mariner 7 have been calculated using this approach.

INTRODUCTION

The basis for quantitative discussion of the photochemical production of atmospheric ozone is the assumption of photochemical equilibrium whereby the number of ozone molecules formed equals the number destroyed in unit time and unit volume. In most studies of equilibrium distributions of ozone, the analysis treats only the photochemical reactions which would occur in an atmosphere of atomic and molecular oxygen (0 and $O_2$) and ozone ($O_3$) in the presence of inert molecules (M). For this

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case, the basic photochemical reactions are:

\[ \text{O}_2 + h\nu \rightarrow \text{O} + \text{O} \quad \alpha_2 \quad \lambda \leq 2424 \, \text{Å} \]  
(1)

\[ \text{O} + \text{O}_2 + \text{M} \rightarrow \text{O}_3 + \text{M} \quad k_2 \]  
(2)

\[ \text{O} + \text{O}_3 \rightarrow \text{O}_2 + \text{O} \quad k_3 \]  
(3)

\[ \text{O}_3 + h\nu \rightarrow \text{O} + \text{O}_2 \quad \alpha_3 \quad \lambda \leq 11,800 \, \text{Å} \]  
(4)

\[ \text{O} + \text{O} + \text{M} \rightarrow \text{O}_2 + \text{M} \quad k_5 \]  
(5)

where \( \alpha \)'s represent the probability of dissociation of molecules and \( k \)'s represent the reaction rate constant. The third body, \( M \), conserves energy and momentum in the reaction. It is not necessary to include reactions involving excited molecules whose contributions to the equilibrium concentrations are negligibly small (ref. 1). The method of Wulf and Deming (refs. 2 & 3; see also Craig, ref. 4) for calculating equilibrium amounts of ozone is commonly used for analyses of the ozone distribution in the terrestrial atmosphere. This method utilizes reactions (1) through (4) only, the inclusion of reaction (5) is regarded to have a negligible effect on the numerical calculations for the altitudes of interest in the terrestrial atmosphere.

The model of ozone photochemistry developed by Wulf and Deming has been applied in several studies of the atmospheres of Mars and Venus. For example, some conclusions about the stratospheric circulation and wind patterns of Mars were derived by Paetzold (ref. 5) from his calculated vertical distribution of ozone. However, Marmo et al. (ref. 6) took exception to the use of this method for the study of the atmosphere of another planet and included reaction (5) in their calculations of the ozone distribution in the Martian atmosphere. Their results demonstrated the importance of the direct association of oxygen atoms by three-body collisions for the Martian atmosphere. In addition to a sharp reduction in the total amount of ozone in the atmosphere, significant differences in the shape of the ozone concentration
profile were obtained. The ozone concentration profile calculated by Marmo and his
coworkers increased monotonically with decreasing altitude to a maximum at the
surface of the planet, as opposed to the ozone-concentration maximum at about 40 km
predicted by Paetzold's analysis. Because of the importance of ozone in planetary
atmospheres, the system of photochemical reactions leading to the equilibrium
concentrations of ozone has been re-examined to better comprehend the implications
of this discrepancy.

Photochemical Scheme for Calculations of the
Vertical Distribution of Atmospheric Ozone

First consideration is given to the scheme of Wulf and Deming involving
reactions (1) through (4) which will henceforth be referred to as the 4-reaction
system. Let \( n_1, n_2, n_3, \) and \( n_m \) be the number densities of \( O, O_2, O_3, \) and \( M, \)
respectively. Then, under conditions of photochemical equilibrium, the rate of
change of the concentrations of the various compounds of oxygen will be given by:

\[
\frac{dn_1}{dt} = 2a_2n_2 + a_3n_3 - k_2n_1n_2n_m - k_3n_1n_3 = 0
\]  

(6)

\[
\frac{dn_2}{dt} = -a_2n_2 + a_3n_3 - k_2n_1n_2n_m + 2k_3n_1n_3 = 0
\]  

(7)

\[
\frac{dn_3}{dt} = -a_3n_3 + k_2n_1n_2n_m - k_3n_1n_3 = 0
\]  

(8)
These three equations are not independent since continuity of mass considerations require that

\[
\frac{dn_1}{dt} + 2\frac{dn_2}{dt} + 3\frac{dn_3}{dt} = 0 \quad .
\]  

(9)

By a linear combination of equations (6), (7), and (8), the following two independent equations are obtained:

\[
a_2 n_2 = k_3 n_1 n_3 \quad .
\]  

(10)

\[
a_2 n_2 + a_3 n_3 = k_2 n_1 n_2 n_m \quad .
\]  

(11)

When \( n_1 \) is eliminated from equations (10) and (11), we obtain the quadratic equation,

\[
\left( \frac{a_2 k_3}{n_3} \right)^2 + \left( \frac{a_2 k_3 n_2}{n_3} \right) n_3 - a_2 a_3 n_2^2 = 0 \quad .
\]  

(12)

In most previous studies, \( n_3 \) has been calculated from equation (12) as a function of altitude to give a vertical profile of ozone concentration, using an assumed vertical distribution for the equilibrium values of \( n_2 \). In the terrestrial atmosphere, this procedure has been well established for calculating the photochemical equilibrium distribution of ozone in the stratosphere, where most of the atmospheric ozone is known to exist. However, limited knowledge of the composition of the atmospheres of other planets prevents the application of such a simplified scheme as the 4-reaction system to studies of the ozone distribution in those planetary atmospheres. The earth's atmosphere, with oxygen as a major constituent, is decidedly different from the atmospheres of Mars and Venus, for example, which are now regarded to be composed mostly of carbon dioxide, containing only traces of oxygen. Thus, a technique which has been successful in studies of the observed temporal and spatial variations of the ozone distribution in our atmosphere should not be expected to apply with equal success to the atmospheres of other planets.

One aspect of the 4-reaction system that is particularly disturbing for studies of the ozone distribution is when equations (10) and (11) are solved to eliminate either \( n_2 \) or \( n_3 \). Then, the resulting quadratic equation in \( n_1 \),

\[
\left( \frac{k_2 k_3 n_m}{n_3} \right) n_1^2 - \left( \frac{a_2 k_3}{n_3} \right) n_1 - a_2 a_3 = 0
\]  

(13)
is independent of local concentrations of the other oxygen compounds. This implies that \( n_1, n_2, \) and \( n_3 \) are no longer in equilibrium, contradicting the assumption of equilibrium conditions that led to the derivation of equation (13). This contradiction would indicate that the 4-reaction system does not represent a physically stable condition. Then, the question arises: why has the 4-reaction system been successful in calculating photochemical equilibrium distributions in the Earth's stratosphere? This equation predicts that the number of oxygen atoms is determined only by the reaction rate constants, the local number density of all atmospheric molecules, and the probabilities of dissociation of oxygen and ozone molecules. In special cases, as the Earth's stratosphere, \( \alpha_2 \) and \( \alpha_3 \) can provide an indirect coupling with the columnar number densities of \( O_2 \) and \( O_3 \) above the given altitude which, in turn, would be approximately proportional to the local values of \( n_2 \) and \( n_3 \). The values of \( \alpha_2 \) and \( \alpha_3 \) are evaluated by integrating over wavelengths \( \lambda \) expressions of the form

\[
\alpha_2(z) = \int_0^{2424 \text{ Å}} \sigma_2(\lambda) \phi(z, \lambda) d\lambda
\]  
(14)

and

\[
\alpha_3(z) = \int_0^{11,800 \text{ Å}} \sigma_3(\lambda) \phi(z, \lambda) d\lambda
\]  
(15)

where \( \sigma_2 \) and \( \sigma_3 \) are, respectively, the absorption cross-sections of \( O_2 \) and \( O_3 \), and \( \phi \) is the solar flux at the altitude \( z \). For the case in which the only absorbing constituents of the atmosphere are \( O_2 \) and \( O_3 \), the solar flux at normal incidence can be evaluated from the expression

\[
\phi(z, \lambda) = \phi_\infty(\lambda) \exp \left( -\sigma_2(\lambda) \int_z^\infty n_2(z) dz - \sigma_3(\lambda) \int_z^\infty n_3(z) dz \right)
\]  
(16)
where $\phi_{\infty}$ is the solar flux at the "top" of the atmosphere. If the columnar number densities of $O_2$ and $O_3$ are large enough to affect the numerical value of the exponential factor in equation (16), as is true in the Earth's stratosphere, an indirect coupling, as described above, is possible. However, for columnar densities of $O_2$ and $O_3$ that are smaller than in the Earth's stratosphere, the coupling no longer exists and the contradiction is present. For example, the equilibrium number density of oxygen atoms calculated from equation (13) at 100 km in the terrestrial atmosphere would be approximately 2 orders of magnitude higher than $n_m$, the actual number density of all atmospheric molecules. The contradiction will also be present for planetary atmospheres in which $O_2$ and $O_3$ are no longer the major absorbers of the solar radiation. In that case, equation (16) would be rewritten with extra terms in the exponential factor which may be much larger than the terms that are related to absorption by $O_2$ and $O_3$ so that the coupling might again be destroyed. Since the atmospheres of Mars and Venus, for example, have oxygen present only as a trace constituent, it would be expected that the 4-reaction system is unsuitable for calculating equilibrium ozone concentrations in the atmosphere of these planets.

In order to assess the problem in more detail, another aspect of the 4-reaction system is considered. In this part of the analysis, an equation for conservation of mass is introduced,

$$n_1 + 2n_2 + 3n_3 = 2\tilde{n}_2$$  ,  \hspace{1cm} (17)

where $\tilde{n}_2$ is a constant which may be regarded as an original number density of oxygen molecules which, under photochemical action, has formed oxygen atoms, oxygen molecules, and ozone molecules. By combining equations (10), (11), and (17), an equation for the local ozone concentration is obtained:

$$n_3 = \frac{a_2(2\tilde{n}_2 - n_1)}{3a_2 + 2k_3n_1}$$  .  \hspace{1cm} (18)
Solution of this equation for the ozone number density requires a knowledge of the number density of oxygen atoms. By eliminating \(n_2\) and \(n_3\) from equations (10), (11), and (17), the following is obtained:

\[
\left( k_{23n} \right) n_1^3 - \left( a_2 k_2 + 2k_{23} \right) n_2 n_1^2 - \left( a_2 a_3 - 2a_2 k_3 n_2 \right) n_1 + 2a_2 a_3 n_2 = 0, \tag{19}
\]

which, at first glance, would appear to be coupled to the number of oxygen atoms available for establishing a condition of photochemical equilibrium. However, this equation can be factored in the form

\[
\left[ k_{23n} \right] n_1^2 - \left( a_2 k_2 \right) n_1 - a_2 a_3 \left[ 2n_2 - n_1 \right] = 0. \tag{20}
\]

This result again contradicts the assumption of photochemical equilibrium. A solution based on the first factor is identical with the solution for equation (13) which has already been determined to be contrary to the physical meaning of equilibrium concentrations of oxygen compounds. The solution based on the second factor, \(n_1 = 2n_2\), represents complete dissociation of the available oxygen compounds which, again, contradicts the assumption of photochemical equilibrium. Thus, a conservation of mass relation cannot be incorporated meaningfully into the solution of the 4-reaction system. In view of the foregoing discussion, it has been concluded that the 4-reaction system should not be applied without reservation for calculations of the ozone distribution of other planets and that investigations based on such calculations (see, for example, Paetzold (ref. 5) who studied the Martian atmosphere and Fabian and Libby (ref. 7) who studied the Cytherean atmosphere) should be reanalyzed within these limitations.
The system consisting of reactions (1) through (5) and, henceforth, referred to as the 5-reaction system is now considered. As before, the solution of the equations for the rate of change of \( n_1, n_2, \) and \( n_3 \) for equilibrium conditions leads to two independent equations,

\[
\alpha_2 n_2 = k_3 n_1 n_3 + k_5 n_1^2 n_m
\tag{21}
\]

and

\[
\alpha_2 n_2 + \alpha_3 n_3 = k_2 n_1 n_2 n_m + k_5 n_1^2 n_m
\tag{22}
\]

Equations (21) and (22), along with the conservation of mass relation represented by equation (17), are then available for solving for equilibrium number densities of the oxygen compounds. These equations may be combined to give the following expression for the number density of ozone:

\[
n_3 = \frac{k_2 k_5 n_1^3 n_m^2}{\alpha_2 \alpha_3 + \alpha_2 k_3 n_1 - k_2 k_3 n_1^2 n_m}
\tag{23}
\]

If \( n_2 \) and \( n_3 \) are eliminated from the equations describing the 5-reaction system, a cubic equation in \( n_1 \) results:

\[
\left( k_2 k_3 n_m - 3 k_2 k_5 n_m^2 - 2 k_3 k_5 n_m \right) n_1^3 - \left( \alpha_2 k_3 + 2 k_2 k_3 n_m \tilde{n}_2 + 2 \alpha_3 k_5 n_m \right) n_1^2 - \left( \alpha_2 \alpha_3 - 2 \alpha_2 k_3 \tilde{n}_2 \right) n_1 + 2 \alpha_2 \alpha_3 \tilde{n}_2 = 0
\tag{24}
\]
The solution of equation (24) is coupled to the available oxygen as well as the other photochemical parameters and, therefore, provides the coupling mechanism for the desired condition of equilibrium. Thus, the 5-reaction system is to be preferred since, unlike the 4-reaction system, it does not contain an implicit contradiction of the assumed state of equilibrium. Furthermore, inclusion of reaction (5) has the advantage of increasing the generality of the technique for use in exploratory investigations of photochemical equilibrium distributions of ozone in the atmosphere of other planets. Of course, the 5-reaction system is still not as general as a comprehensive analysis of all possible reactions such as the study of the photochemistry of a CO₂ atmosphere conducted by McElroy and Hunten (ref. 8). However it is pointed out that, for the lower atmosphere of Mars from the surface to about 60 km, McElroy and Hunten assumed, as has been assumed here, that the relative concentration of O₂ is constant with a mixing ratio of 10⁻³. While the 5-reaction system does not provide the amount of insight as a comprehensive study of the mechanisms which maintain this mixing ratio, it does not have intrinsic value for calculations of the ozone distribution in the lower atmosphere of Mars; and, therefore, such calculations have been performed based on the results of the Mariner 4 occultation experiment as well as the more recent results from Mariners 6 and 7.

Calculations of the Ozone Distribution in the Atmosphere of Mars

Calculations of the vertical distribution of ozone in the atmosphere of Mars based on the 4-reaction system and on the 5-reaction system have been reported by Paetzold (ref. 5) and Marmo et al. (ref. 6), respectively. On the basis of the previous discussion, the results of Marmo and his collaborators are more reliable. However, their atmospheric model has a surface pressure of 25 mb and partial pressures of 470 m STP for nitrogen, 55 m STP for carbon dioxide, and 70 cm STP for oxygen in accordance with the results of Kaplan et al. (ref. 9) and a temperature profile with a surface temperature of 250 K, a lapse rate of 2.9 K per km, and isothermal conditions above 30 km in accordance with the results of Schilling (ref. 10). This atmospheric model is now outdated; furthermore, solar radiation was limited to the
spectral region 1750 to 3000 Å in their calculations. For these reasons, it is now possible to provide better data of the ozone distribution of the atmosphere of Mars. Using an atmospheric model based on the results of Mariner 4 radio occultation experiment (ref. 11) and a computer program incorporating an iteration procedure to obtain self-consistent, steady-state distributions of ozone based on the 5-reaction system, calculations of the ozone content of the Martian atmosphere have been performed. In these calculations, no distinction has been made between O(1D) and O(3P) atoms as O(1D) are not present below about 50 km (ref. 8), the altitude interest of our major concern. The numerical data summarized in Table I were used for the calculations. The composition of the atmosphere was taken to be 99.9 percent carbon dioxide and 0.1 percent oxygen. Vertical profiles of O, O₂, and O₃ are shown in figure 1. Solid lines indicate calculated profiles based on the 5-reaction system and the dashed lines indicate profiles based on the 4-reaction system. The profiles for O graphically portray the problem regarding the 4-reaction system. For the 5-reaction system, O profiles increase with altitude to the point at about 65 km at which almost complete dissociation of O₂ has occurred (n₁≈2n₂). However, the O profile for the 4-reaction system increases at a much faster rate and exceeds the value of n₃, which is taken to be the number density of all atmospheric molecules at altitudes somewhat above ~60 km. For the 5-reaction system, the O₃ number density decreases monotonically with altitude and has an entirely different profile than the 4-reaction system. Thus, the maximum concentration of Martian ozone is expected at the surface of the planet. Although the significant differences in atmospheric models results in a surface number density of ozone that is over an order of magnitude smaller than that calculated by Marmo et al. (ref. 6), it is concurred that an ozone maximum exists at the surface of the planet. This is in disagreement with the results of Paetzold (ref. 5) which indicated an ozone maximum at an altitude of about 40 km, based on the 4-reaction system and an atmospheric model similar to that used by Marmo and his co-workers.

Additional calculations were performed to determine the effect of varying the parameters used in the model. None of the parameter variations yielded maximum concentrations of ozone at some altitude above the surface of the planet. The effect of lowering the content of carbon dioxide to 80 percent with 19.9 percent argon or nitrogen and 0.1 percent oxygen was to increase the ozone concentrations at all
altitudes by about 20 percent without appreciably altering the shape of the profile. When the oxygen content was increased to 1 percent with 99 percent carbon dioxide, the ozone concentrations were increased by a factor of about 2; a similar increase was found for an atmosphere of 80 percent carbon dioxide, 19 percent argon or nitrogen, and 1 percent oxygen. In addition, the effect of limiting the spectral flux to the 1750-3000 Å wavelength interval to match the calculations of Marmo et al. (ref. 6) was investigated. The resulting numerical values for the ozone number densities were about twice those obtained by using the solar flux from 1350-7100 Å. This discrepancy can be attributed mainly to the dissociation of ozone by radiation in the visible part of the solar spectrum.

Finally, atmospheric parameters obtained from Mariner 6 and 7 radio occultation data have been utilized for calculations of the ozone distribution in the Martian atmosphere. The results of these calculations are displayed in figure 2 as vertical profiles of $O$, $O_2$, and $O_3$ from the surface of Mars to an altitude of 35 km. The atmospheric parameters used in the calculations are those presented by Rasool et al. (ref. 12) for the Mariner 6 and Mariner 7 entry data which correspond to daytime conditions at low and high latitudes, respectively. Results obtained from Mariner 4 data are also displayed for comparison; it is noted that the Mariner 6 and 7 results are not substantially different, with the exception of the calculated ozone concentrations near the surface. Integrated total ozone amounts are calculated to be $1.4 \times 10^{-3}$, $7.7 \times 10^{-4}$, and $8.8 \times 10^{-4}$ cm STP for atmospheres corresponding, respectively, to Mariner 4, 6, and 7 results. It is pointed out, partially to give substance to the neglect of photochemical reactions involving $CO_2$ and $CO$ for calculations of equilibrium concentrations of ozone, that the calculated total ozone amounts are in excellent agreement with observations of the total ozone in the Martian atmosphere. The Mariner 6 and 7 ultraviolet spectrometer experiment detected ozone in the Martian atmosphere (ref. 13). Data from these measurements indicated a total ozone amount that was also about 3 orders of magnitude less than found on earth. These calculations, therefore, corroborate the measurements of ozone in the Martian atmosphere.


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Figure 1 - Comparative profiles of oxygen compounds calculated from the 4-reaction and the 5-reaction systems for a model atmosphere based on the results of the Mariner 4 occultation experiment.
Figure 2 - Comparative profiles of oxygen compounds calculated from the 5-reaction system for model atmospheres based on the results of the Mariner 4, 6, and 7 occultation experiments.
REFERENCES


"The aeronautical and space activities of the United States shall be conducted so as to contribute . . . to the expansion of human knowledge of phenomena in the atmosphere and space. The Administration shall provide for the widest practicable and appropriate dissemination of information concerning its activities and the results thereof."

—National Aeronautics and Space Act of 1958

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