

THEORETICAL MODELS FOR THE RADIANCE OF CONTAMINANT GLOW CLOUDS IN THE UPPER ATMOSPHERE

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Summary

A discussion is given of the time variation in radiance which would be observed across a diffusing glow cloud released in the upper atmosphere. A Gaussian distribution for the contaminant number density is used, and it is assumed that, when chemical reactions occur, either the contaminant density is everywhere very small, or an insignificant quantity of contaminant is consumed.

The cases of chemiluminescence and scattering of the Sun's rays are examined, and the effects on the observed radiance of absorption, re-emission, and scattering of radiation within the cloud are considered. The results may be used to interpret observations on upper atmosphere contaminant releases. It is found that if the cloud is optically thin the radiance distribution is Gaussian for both chemiluminescent and sunlit glow clouds, and the radiance is proportional to the number of cloud particles along the line of sight. Optically thick glow clouds will not be Gaussian, and it is inadmissible to use a method based on a Gaussian distribution in analysis of these clouds.

I. INTRODUCTION

This paper is concerned with the derivation of theoretical expressions for the radiance variation across diffusing, contaminant clouds released in the upper atmosphere. Marmo, Pressman, and Aschenbrand (1961) and Shklovskii and Kurt (1960) have used observations on the radiance of contaminant releases to find diffusion coefficients from which the upper atmospheric density is derived. However, these and other workers in the field have paid little attention to the theoretical derivation of cloud radiance from the number density of the cloud particles. In general an expression has been used for radiance which, as will be seen, is equivalent to assuming the cloud to be optically thin. This assumption is not always valid and can lead to serious errors in determining upper atmospheric properties.

The general properties of a spherically symmetric cloud which reacts with the ambient atmosphere are discussed in Section II. Section III contains the derivation of the radiance variation across chemiluminescent glow clouds for which the emission is a result of a chemical reaction of the contaminant with the ambient atmosphere. The number density of the diffusing cloud particles is assumed to be Gaussian, as is done by other workers, and both optically thin and optically thick clouds are treated. In Section IV, the same is done for sunlit glows; in this case, the emission results from scattering of sunlight that is incident on the cloud. Expressions for the total flux from each type of cloud are derived in Section V.

The quantities that describe the radiation field within the cloud, namely integrated intensity I , emission coefficient j , absorption coefficient κ , and albedo

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for single scattering $\tilde{\omega}_0$, are as defined by Chandrasekhar (1960). Following Chamberlain (1961), the integrated intensity from the cloud that is received by an observer will be named the surface radiance, R . The total flux escaping from the cloud as a whole will be denoted by P .

II. STATEMENT OF THE PROBLEM

The number density of the emitting particles in the diffusing cloud will be taken as (Johnson and Lloyd 1963, eqn. (2))

$$n(r, t) = \frac{N_0}{\pi^{3/2}(r_0^2 + 4Dt)^{3/2}} \exp\left(-\frac{r^2}{r_0^2 + 4Dt}\right), \quad (1)$$

where r = radial coordinate,
 D = diffusion coefficient,
 t = time,
 r_0 = initial effective radius,
 N_0 = total number of cloud particles.

Equation (1) is the solution of the spherically symmetric diffusion equation which neglects depletion or creation of contaminant particles, namely

$$\frac{\partial^2 n}{\partial r^2} + \frac{2}{r} \frac{\partial n}{\partial r} = \frac{1}{D} \frac{\partial n}{\partial t},$$

with the initial Gaussian radial distribution

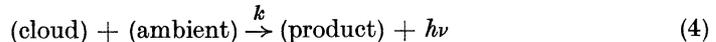
$$n = n(0, 0) \exp\{-r^2/r_0^2\}. \quad (2)$$

Introducing the dimensionless parameters $\rho = r/r_0$ and $b^2 = [1 + 4Dt/r_0^2]^{-1}$ simplifies the analysis, and equation (1) can be rewritten

$$n(\rho, b) = n(0, 0) b^3 \exp\{-b^2 \rho^2\}, \quad (3)$$

where $n(0, 0)$ is the initial peak number density.

Equation (3) assumes that there is no change in the total number of cloud particles with time. A reaction of the form



will result in a loss of cloud particles at a rate $kn_C n_A$, where k is the reaction rate constant, n_C is the number density of the reacting cloud particles, and n_A is the number density of the reacting portion of the ambient atmosphere. This loss means that the contaminant number density n given by equation (3) is not correct when reactions are occurring, although there are certain limiting cases for which simple solutions expressible in terms of n are obtainable.

- (i) *Case* $n_C \ll n_A$.—Since the absolute changes in n_C and n_A are of the same order of magnitude, the fractional change in n_A is very much less than the fractional change in n_C . Therefore the ambient particle density can be assumed to remain constant at its initial value n_{A0} . With this assumption, the relation between n_C and n is $n_C = n \exp(-kn_{A0}t)$, and the reaction rate is $kn n_{A0} \exp(-kn_{A0}t)$.

- (ii) *Case* $4D/r_0^2 \gg kn_A$.—The change in n_C resulting from reaction with the atmosphere is negligible compared with the change in n_C due to diffusion, and is therefore ignored. The cloud particle density is given by its reaction-free approximation, viz. n . Simple general expressions for n_A , and hence the reaction rate $kn_C n_A$, cannot be obtained. However, if changes in n_A resulting from diffusion only need be included, then n_A can be considered a (known) function of time. The reaction rate is now given by the product of n and some function of time. Another sub-case is $4D/r_0^2 \ll kn_C$, which implies that $n_C \gg n_A$, i.e. changes in n_A result almost entirely from reaction

with n_C . It can be shown that $n_A = n_{A0} \exp\left(-k \int_0^t n dt\right)$, and the reaction rate is therefore $kn n_{A0} \exp\left(-k \int_0^t n dt\right)$, which is not directly proportional to n .

The emission from the reaction given by (4) is proportional to the reaction rate $kn_C n_A$. Therefore, provided that either $n_C \ll n_A$ or chemical reaction effects are small (kn_A, kn_C both much less than $4D/r_0^2$), the emission is directly proportional to n and the solution of a diffusion equation, omitting reaction terms, can be used in calculating emission even when chemical reactions are present. The first case, $n_C \ll n_A$, seems to be the most important since no restriction is placed on kn_A . The fact that the total number of contaminant cloud particles, and therefore the total flux, may change significantly with time must not be overlooked; that is, when $n_C \ll n_A$ the total flux is proportional to $\exp(-kn_{A0} t)$.

III. CHEMILUMINESCENT CLOUDS

(a) *Simple Emission with No Absorption*

It will be assumed that the emission due to chemiluminescence by an element of volume dv of the cloud is given by

$$4\pi j_c n dv, \quad (5)$$

where j_c is the emission coefficient for chemiluminescence.

The dimensions of the cloud are much less than the distance of the cloud from the observer, so the surface radiance R is given by the line-of-sight integration

$$R = \int_{-\infty}^{\infty} j_c n dl. \quad (6)$$

Using n from equation (3), this gives

$$R = R_0 b^2 \exp(-b^2 \sigma^2), \quad (7)$$

where $R_0 = \pi^{\frac{1}{2}} j_c r_0 n(0,0)$ is the zero-time peak radiance for the cloud, $\alpha = Dt/r_0^2$ and $b^2 = (1+4\alpha)^{-1}$ are dimensionless time parameters, s is the coordinate perpendicular to the line of sight, and $\sigma = s/r_0$. The coordinate system is shown in Figure 1.

The time variation in radiance across the cloud, given by equation (7), is plotted in Figure 2. These curves reduce to a single curve if $(1/b^2)(R/R_0)$ is plotted

against $b^2\sigma^2$, as has been done in Figure 3. The value of R corresponding to any value of α , σ , and R_0 may be determined from this line when it is used in conjunction with the relation between α and b^2 .

(b) *Simple Emission and Absorption, with No Re-emission of Absorbed Radiation*

In this case some of the chemiluminescent flux is absorbed by the cloud. It will be assumed that the decrease in intensity as a result of absorption is given by Beer's Law,

$$dI = -\kappa n dh, \tag{8}$$

where κ is the absorption coefficient.

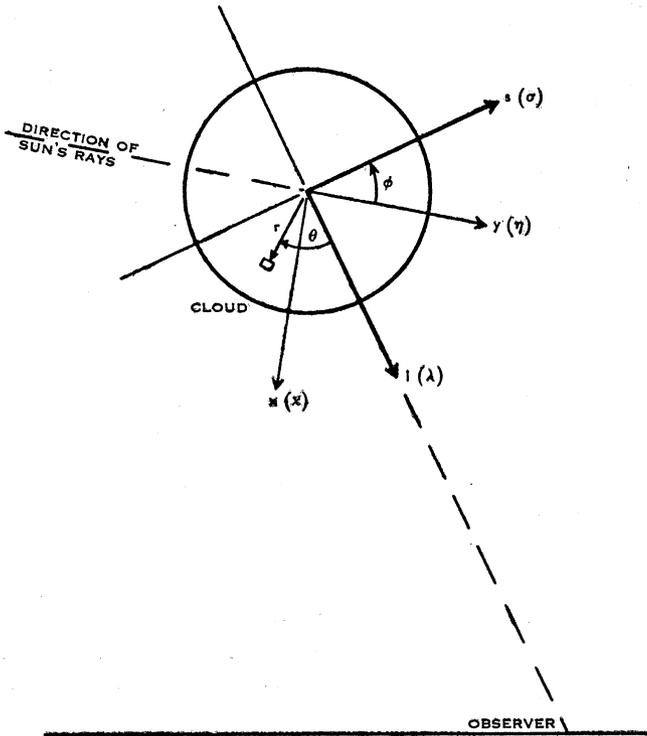


Fig. 1.—Coordinate system. The $z (\zeta)$ axis is perpendicular to the plane of the diagram.

Equation (6) is now replaced by the expression

$$R = \int_{-\infty}^{\infty} j_c n \exp\left(-\int_l^{\infty} \kappa n dh\right) dl. \tag{9}$$

The exponent in the integral represents absorption between the element at l and the observer.

By substituting

$$\tau = \int_l^\infty \kappa n \, dh,$$

where τ is the optical thickness of the cloud between l and infinity, equation (9) can be integrated to give the radiance as

$$R = (R_0/g)[1 - \exp\{-gb^2 e^{-b^2\sigma^2}\}], \tag{10}$$

where $g = (\kappa R_0/jc)$ is a measure of the total optical thickness of the cloud along a line of sight. This equation is illustrated in Figure 3.

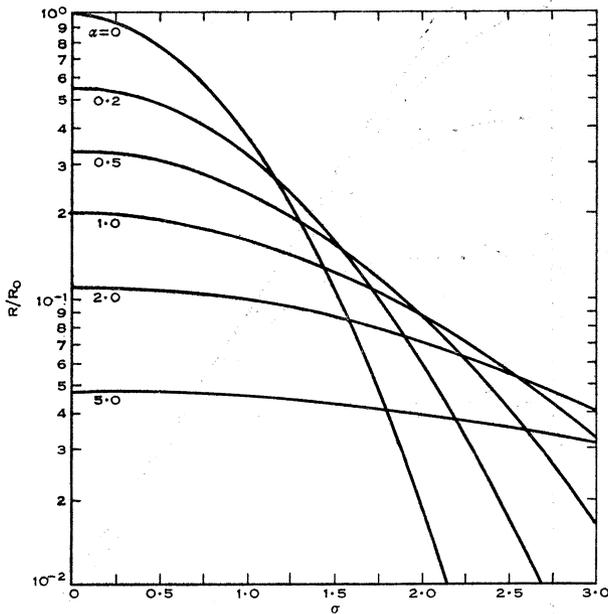


Fig. 2.—Variation with time of the radiance across a non-absorbing chemiluminescent cloud (equation (7)).

When the cloud is optically thin ($gb^2 \ll 1$), the exponential in equation (10) may be expanded giving, to the first order in g ,

$$R = R_0 b^2 \exp(-b^2\sigma^2) [1 - \frac{1}{2}gb^2 \exp(-b^2\sigma^2)]. \tag{11}$$

In the limit of great optical thickness ($gb^2 \gg 1$), equation (10) reduces to

$$R = \frac{R_0}{g} = \frac{jc}{\kappa}. \tag{12}$$

Plots of equation (10) with $g = 10$ are given in Figure 4 and compared with the radiance variation predicted by equations (7) and (11) for the case $\alpha = 0$. Note that, for optically thick clouds, equation (12) is valid at the centre, while equation (11) tends to the correct result at the edges, which are optically thin. Figure 5 shows

how the radiance of an optically thick cloud varies with absorption coefficient. The time variation of centre point radiance (R_{\max}) for clouds of different optical thickness is shown in Figure 6.

It is of interest to examine the error introduced by using the first-order optically thin approximation for an absorbing cloud (eqn. (11)). The maximum error in the

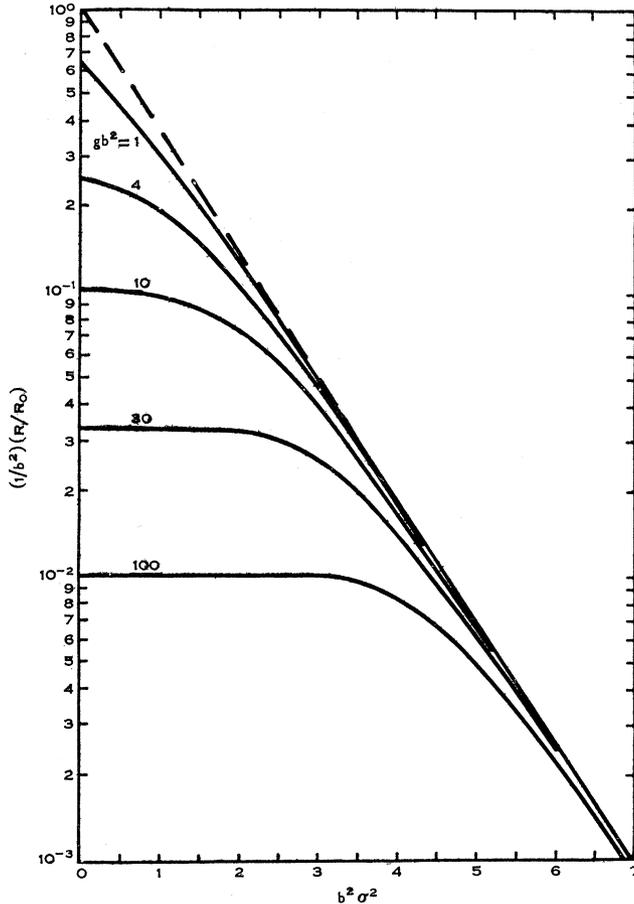


Fig. 3.—Variation of $(1/b^2)(R/R_0)$ with $b^2\sigma^2$ for chemiluminescent glow clouds.
 -- non-absorbing (equation (7)); — absorbing (equation (10)).

cloud radiance, which occurs at the centre of the cloud, is given in Table 1 as a percentage of its exact value for different values of gb^2 , which may be regarded as an optical thickness parameter.

(c) *Simple Emission and Absorption, with Re-emission of Absorbed Radiation*

In this section it is assumed that a fraction $\tilde{\omega}_0$ of the radiation absorbed by an element of the cloud is re-emitted isotropically; $\tilde{\omega}_0$ is called the albedo for single scattering. For perfect scattering $\tilde{\omega}_0 = 1$.

The emission j from an element of the cloud is the sum of the chemiluminescent emission, given by (5), and the re-emission of absorbed radiation (i.e. scattered radiation), thus:

$$j = j_c + j_s. \tag{13}$$

The emission due to scattering, j_s , is given by

$$j_s = \frac{\kappa \tilde{\omega}_0}{4\pi} \int_{\mathbf{r}' \neq \mathbf{r}} \frac{j(\mathbf{r}') n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|^2} \exp\left(-\int_{\mathbf{r}'}^{\mathbf{r}} \kappa n \, dh\right) dv'. \tag{14}$$

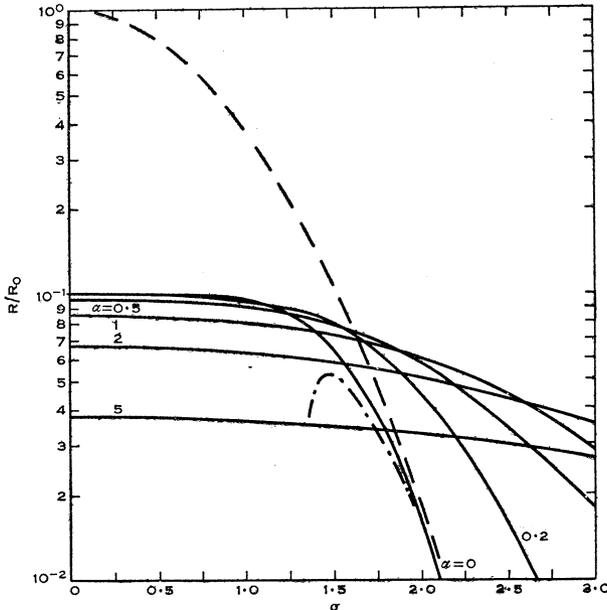


Fig. 4.—Variation with time of the radiance across an absorbing chemiluminescent cloud: — equation (10) for the case $g = 10$; - - - optically thin approximation (equation (11) with $g = 10$); - · - non-absorbing cloud (equation (7)).

Substitution of equation (14) into equation (13) gives a singular integral equation of the second kind in j . The equation does not admit of a general solution. However, if it is assumed that the cloud is optically thin and that, of the two contributions to emission from an element, chemiluminescence is by far the larger, a solution may be found for j by iteration. An outline of the steps made in solving equations (13) and (14), and in integrating equation (15) below, is given by Lloyd (1963).

Equation (9) relates j to the radiance of the cloud, that is,

$$R = \int_{-\infty}^{\infty} j n \left\{ 1 - \kappa \int_l^{\infty} n \, dh + \frac{\kappa^2}{2} \left(\int_l^{\infty} n \, dh \right)^2 \right\} dl, \tag{15}$$

where the exponential has been expanded to second order in κ since the glow cloud is assumed to be optically thin. Integration of equation (15) gives the variation in

radiance across an optically thin chemiluminescent glow cloud with time, including the effects of absorption and re-emission correct to the second order of small quantities in κ , as follows:

$$R/R_0 = b^2 \exp(-b^2\sigma^2) - \frac{1}{2}b^4 \exp(-2b^2\sigma^2)[g - (\tilde{\omega}_0 \kappa / j_c)R_0 K_1] + \frac{1}{4}gb^6 \exp(-3b^2\sigma^2)[\frac{2}{3}g - (\tilde{\omega}_0 \kappa / j_c)R_0\{K_1 + K_2\}], \quad (16)$$

where
$$K_1 = \int_0^1 \frac{\exp a^2 b^2 \sigma^2}{(2-a^2)^{\frac{1}{2}}} da, \quad (17)$$

and
$$K_2 = \int_0^1 \int_0^1 \frac{\exp\{-b^2\sigma^2(c^2-1)(1+a)^2(1+a^2)^{-1}\}}{(1+a^2)^{\frac{1}{2}}[3+(c^2-1)(1+a)^2(1+a^2)^{-1}]} da dc. \quad (18)$$

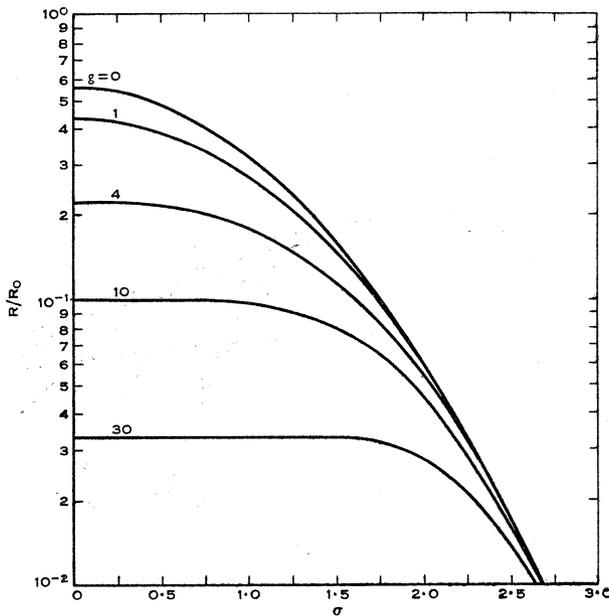


Fig. 5.—Variation with absorption coefficient of the radiance across an optically thick chemiluminescent cloud (equation (10) for the case $\alpha = 0.2$).

K_1 and K_2 are plotted in Figure 7. A plot of $\exp(b^2\sigma^2)$ is also given, so that the relative magnitudes of these three functions of $b^2\sigma^2$ may be compared.

IV. SUNLIT CLOUDS

It will be assumed in this section that the radiance of the diffusing cloud is due to isotropic scattering (or absorption and re-emission) of the Sun's incident rays. If I_∞ is the intensity of the Sun's radiation falling on the cloud, then the intensity of radiation at an element of the cloud whose coordinates are (x, y, z) is I_s , where

$$I_s = I_\infty \exp\left(-\int_{-\infty}^y \kappa n dy\right), \quad (19)$$

and y is the coordinate in the direction of the Sun's rays (see Fig. 1). The emission from this element is

$$\kappa \tilde{\omega}_0 I_s n dv. \tag{20}$$

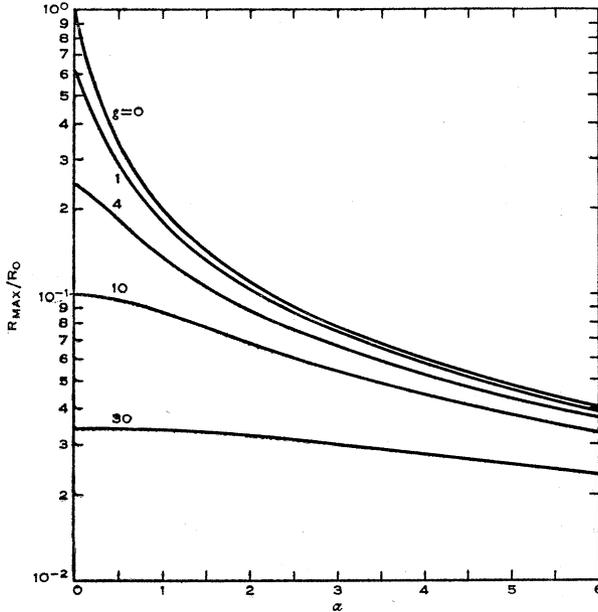


Fig. 6.—Variation of centre-point radiance with time for an absorbing chemiluminescent cloud (equation (10) with $\sigma = 0$).

TABLE I
ERROR IN FIRST-ORDER APPROXIMATION FOR CLOUD RADIANCE

gb^2	Error (%)
1.25	35
1.00	21
0.75	12
0.5	6
0.25	2
0.00	0

Secondary scattering will be neglected. With this assumption the radiance distribution across a sunlit glow cloud is given, from equations (9) and (19), and from (20), as

$$R = \frac{\tilde{\omega}_0 \kappa I_\infty R_0 b^2}{4\pi j_e \pi^{\frac{1}{2}}} e^{-b^2 \sigma^2} \int_{-\infty}^{\infty} e^{-\lambda^2} \exp\left[-\frac{1}{2}gb^2 e^{-b^2(\sigma^2 + \zeta^2)}\right] \times \left(\operatorname{erfc} \lambda + (e^{b^2 \sigma^2 \cos^2 \phi} e^{-\lambda \cos \phi (\lambda \cos \phi - 2b\sigma \sin \phi)} \{1 + \operatorname{erf}(\lambda \sin \phi + b\sigma \cos \phi)\})\right) d\lambda, \tag{21}$$

where ϕ is the angle between the direction of the Sun's rays and the normal to the observer's line-of-sight, and $\zeta = z/r_0$. The equation has been written in terms of the

peak radiance of the equivalent chemiluminescent cloud so that a comparison between the radiance profiles of chemiluminescent and sunlit glow clouds may be easily made. The factor $(\tilde{\omega}_0 \kappa I_\infty / 4\pi j_c)$ is the ratio of the emission by a given number of cloud particles for sunlit and chemiluminescent glow clouds.

It is not possible to obtain an analytic expression for the integral in equation (21). The equation may be integrated numerically, and Figure 8 presents the result for the case $\phi = 0$, $g = 10$, and $\alpha = 0.2$. The asymmetrical, crescent shape of the isophots results from a reduction of intensity of the incident Sun's rays by absorption as they pass through the cloud. Also shown in the figure are the isophots and the radiance

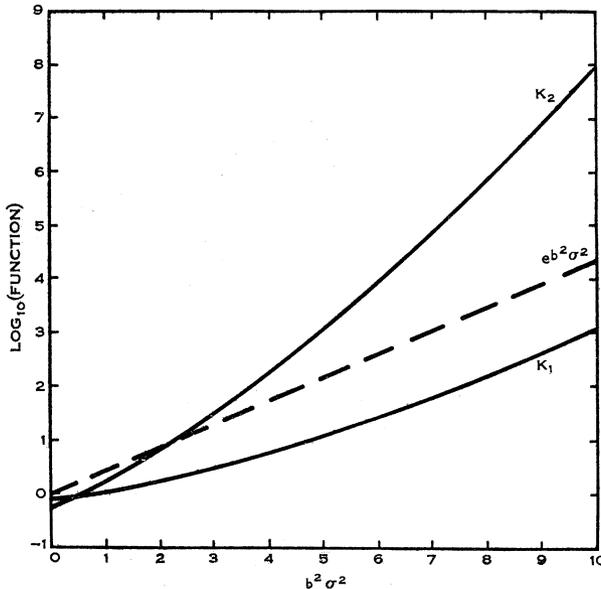


Fig. 7.—The functions K_1 and K_2 (equations (17) and (18)).

cross section for the equivalent chemiluminescent cloud (eqn. (10)); this has the same radiance gradient in the optically thin regions. The radiance contour for the sunlit cloud in Figure 8 is qualitatively similar to that observed by Groves (1963, Fig. 5) for a sunlit cloud produced by the release of 18 kg of explosive at 103 km altitude, i.e. a non-Gaussian radiance distribution with an asymmetric peak. This large contaminant release therefore produced an optically thick glow cloud.

An approximation for optically thin clouds is obtained by expanding the exponential in equation (21) and retaining terms to first order in g . This may be integrated to give

$$R = \frac{\tilde{\omega}_0 \kappa I_\infty}{4\pi j_c} R_0 b^2 \exp\{-b^2(\sigma^2 + \zeta^2)\} \left(1 - \frac{1}{2} g b^2 \exp\{-b^2(\sigma^2 + \zeta^2)\} \right) \times [1 + (\epsilon/\sqrt{2}) \sec \phi \exp(\epsilon^2 b^2 \sigma^2) \{1 + \operatorname{erf} \epsilon b \sigma\}], \tag{22}$$

where $\epsilon^2 = 2(1 + \sec^2 \phi)^{-1}$.

Isophots for an optically thin sunlit cloud, and the radiance contour in the direction of the Sun's rays for $\phi = 0$, $g = 10$, and $\alpha = 5$, are presented in Figure 9. Also shown in the figure are the isophots and radiance contour for the equivalent chemiluminescent glow cloud (eqn. (11) with $g = 10$, $\alpha = 5$). Note that the isophots for the sunlit glow cloud are very nearly circular. The circles are not concentric but are displaced in the direction of the Sun, the brighter isophots being displaced further. Comparison of the radiance contours for sunlit and chemiluminescent clouds shows that the two are very similar, and therefore optically thin sunlit clouds may be analysed to a first approximation using the equivalent expression for chemiluminescent clouds. This is, in fact, what other workers have done without having justified their action.

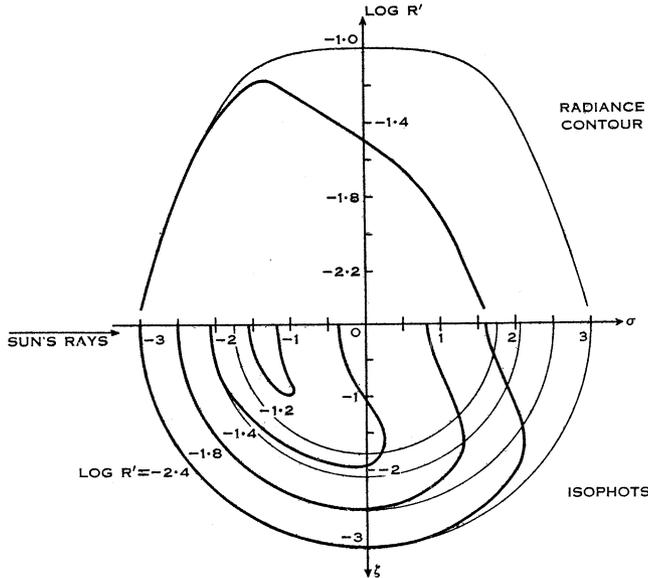


Fig. 8.—Optically thick sunlit cloud: — equation (21) with $R' = R[4\pi j_c / \kappa \omega_0 I_\infty R_0]$, $g = 10$, $\alpha = 0.2$, $\phi = 0^\circ$; - - - equivalent chemiluminescent cloud (equation (10) with $R' = R/R_0$, $g = 10$, $\alpha = 0.2$).

The maximum error introduced by the first-order approximation (eqn. (22)) is given by Table 1 with g replaced by $2g[1 + (1/\pi) \cot^{-1}(\sec \phi)]$.

V. TOTAL FLUX EMITTED BY THE CLOUD

For a glow whose emission is direction independent, the total flux emitted, P , is given by

$$P = 4\pi \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} R \, ds \, dz. \tag{23}$$

Since the chemiluminescent clouds considered are spherically symmetric, equation (23) may be applied to them. It may also be applied to optically thin sunlit clouds,

when the intensity of the Sun's incident rays is not decreased appreciably by scattering within the cloud.

For a non-absorbing chemiluminescent cloud, substitution of equation (7) into (23) shows that the total flux has the time-independent value

$$P_0 = 4\pi^2 r_0^2 R_0. \tag{24}$$

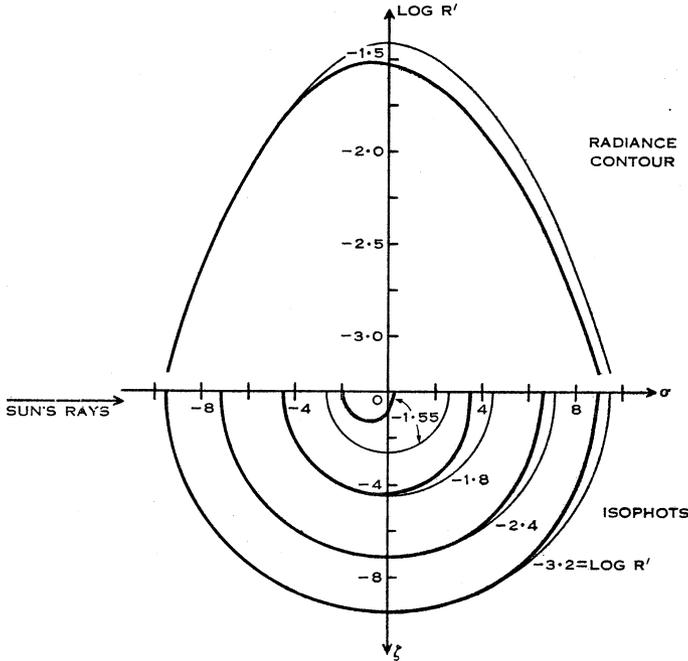


Fig. 9.—Optically thin sunlit cloud: — equation (22) with $R' = R[4\pi j_c / \kappa \tilde{\omega}_0 I_\infty R_0]$, $g = 10$, $\alpha = 5$, $\phi = 0^\circ$; — equivalent chemiluminescent glow (equation (11) with $R' = R/R_0$, $g = 10$, $\alpha = 5$).

For an absorbing chemiluminescent cloud of arbitrary optical thickness with no re-emission of absorbed radiation, represented by equation (10), the integration is effected by making the substitution

$$\xi = gb^2 e^{-b^2 \sigma^2}, \tag{25}$$

which reduces the integral to

$$P = \frac{P_0}{gb^2} \int_0^{gb^2} \frac{1 - e^{-\xi}}{\xi} d\xi. \tag{26}$$

This gives the total flux emitted by an absorbing chemiluminescent cloud of arbitrary optical thickness as

$$P = (P_0 / gb^2) [C + \ln gb^2 - \text{ei}(-gb^2)], \tag{27}$$

where C is Euler's constant (0.577 215...), and $\text{ei}(-x) = -\int_x^\infty \frac{e^{-\xi}}{\xi} d\xi$ is the

exponential integral (Lowan 1940). For an optically thin cloud ($gb^2 \ll 1$) equation (27) reduces to

$$P = P_0(1 - \frac{1}{4}gb^2). \tag{28}$$

Plots of equation (27) for different values of g are given in Figure 10.

It should be noted that the total flux emitted by an optically thick chemiluminescent cloud is not the same as the sum of the chemiluminescent fluxes emitted by all the elements of the cloud. However, as the cloud expands, the total amount of such self-absorption decreases, and the total flux emitted tends to the value for a non-absorbing cloud. It is seen that the time dependence of the total flux can give indications both of the optical thickness of the cloud and of the rate of the reactions, if any,

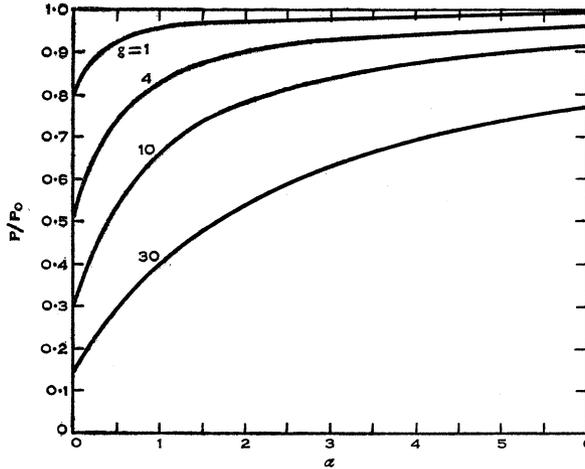


Fig. 10.—Variation with time of the total flux emitted by optically thick chemiluminescent clouds (equation (27)).

Note: For sunlit clouds $g \rightarrow 2g\{1 + (1/\pi) \cot^{-1}(\sec \phi)\}$

occurring within the cloud. For re-emission with $\tilde{\omega}_0 = 1$ (i.e. perfect scattering) there is no net absorption of radiation, and the total flux emitted is the same as for a non-absorbing cloud. Of course, the radiance variation across the cloud is not the same as in the non-absorbing case.

For the optically thin sunlit cloud which was considered in Section IV, the relevant integration is effected by changing to polar coordinates. The result obtained is

$$P = \left(\frac{\kappa \tilde{\omega}_0 I_\infty}{4\pi j_e} \right) P_0 \left[1 - \frac{gb^2}{2} \left\{ 1 + \frac{\cot^{-1}(\sec \phi)}{\pi} \right\} \right]. \tag{29}$$

Note the formal similarity of equation (29) to the optically thin approximation for chemiluminescent clouds (eqn. (28)); the effective optical thickness for an optically thin sunlit glow cloud is greater than for the equivalent optically thin chemiluminescent glow cloud by the factor $2\{1 + (1/\pi) \cot^{-1}(\sec \phi)\}$.

It is of interest to note that the sunlit sodium cloud observed at 430 km altitude by Shklovskii and Kurt (1960) behaved as predicted by equation (29), i.e. there is a rapid initial increase in total flux, tending to an asymptotic value at large time.

VI. DISCUSSION

The results of Sections III, IV, and V all ignore consumption of the contaminant particles. Care must be taken when the results in these sections are used that one of the restrictive conditions given in Section II is obeyed. In particular, when the contaminant particle density is small, all results in Sections III, IV, and V must be multiplied by the time-dependent factor $\exp(-kn_{A0}t)$ (see Section II). Even when the restrictive conditions are not satisfied, the results of Sections III, IV, and V will give an indication of the form of the results that may be expected for more general and more complex reaction mechanisms. In particular, the continual re-occurrence of dimensionless parameters (e.g. $b, g, \kappa\tilde{\omega}_0 I_\infty/4\pi j_c$) and the grouping of these parameters into expressions such as gb^2 and $b^2\exp(-b^2\sigma^2)$ should be noted; these parameters, and similar groupings, are likely to be present in a more general treatment.

Most of the results that have been derived above involve rather complex functions of the diffusion coefficient, which makes the use of such expressions in analysing results of observations on the radiance of glow clouds very difficult. A simple test for whether the cloud is non-absorbing, or optically very thin, is indicated by equation (7), and involves plotting $\log R$ against s^2 . Should the cloud be optically thin, i.e. the cloud radiance distribution be Gaussian, the points will lie on a straight line. An example of this is given by Johnson and Lloyd (1963, Fig. 2). It is, in practice, very difficult to determine whether a non-Gaussian cloud radiance distribution is due to optical thickness effects or to reaction mechanisms that are not of the simple type considered in Section II.

VII. ACKNOWLEDGMENTS

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