DETERMINATION OF DIFFUSION COEFFICIENTS FROM OBSERVATIONS ON GRENADE GLOW CLOUDS

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Summary

Glows have been observed at Woomera when grenades ejected from Skylark rockets have been detonated in the altitude range 90–170 km. Using results obtained from these glows by a special scanning photometer located on the ground, an estimate has been made of the diffusion coefficient in the region 120–160 km. The theoretical model which is used to describe the behaviour of the explosion products incorporates the assumptions of molecular diffusion, a Gaussian distribution of particle density, and an optically thin cloud. The effects of consumption of the cloud particles are included in the model.

I. Introduction

Bates (1950) was the first to suggest that optical observations on contaminants ejected from rockets could be used to determine properties of the upper atmosphere. He discussed both the night-time and twilight phenomena which might be observed when sodium vapour is released. At night, the glow results from chemiluminescent reactions occurring between the contaminant and the ambient atmosphere; for a twilight release (in which, although the Sun is below the horizon for an observer on the ground, the cloud is still illuminated by the Sun's rays), the light emitted from the cloud is a result of resonant scattering of the Sun's rays by the contaminant particles. The observed rate of expansion of the glow cloud may be used to yield a value for the diffusion coefficient, from which density may be inferred. Marmo *et al.* (1960) released sodium under twilight conditions in the altitude range 80–120 km; they were mainly interested in radar observations of the artificial electron clouds so formed, but sufficient data were obtained to demonstrate the feasibility of experiments using optical techniques.

The importance of an appropriate theoretical model in the analysis of the data was clearly demonstrated by the work of Shklovskii and Kurt (1960). They obtained good records of a sunlit sodium cloud release at 430 km altitude; but, as Pressman and Marmo (1961) have shown, their analysis of the results was incorrect, leading to an error in the diffusion coefficient by a factor of four.

Over the last few years, grenades have been released from Skylark rockets fired at Woomera to determine wind and temperature (Groves *et al.* 1960). In the altitude range 90–170 km, contaminants produced by the grenade explosion give a persistent glow, which subtends an angle of about 2° at the ground. It is from observations on some of these glows that attempts to measure diffusion coefficients have been made. The glows whose observations are reported here were released

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under evening twilight conditions on March 5, 1962. The optical observations were made from the ground using a scanning photometer built for the purpose, whose construction is described below. This paper gives the first results obtained in these investigations. The most persistent glows were visible for over 5 min, although useful results could not be obtained for more than half this time. The variation in the altitude, and the times of observation of the glows by the scanning photometer are shown in Figure 1.

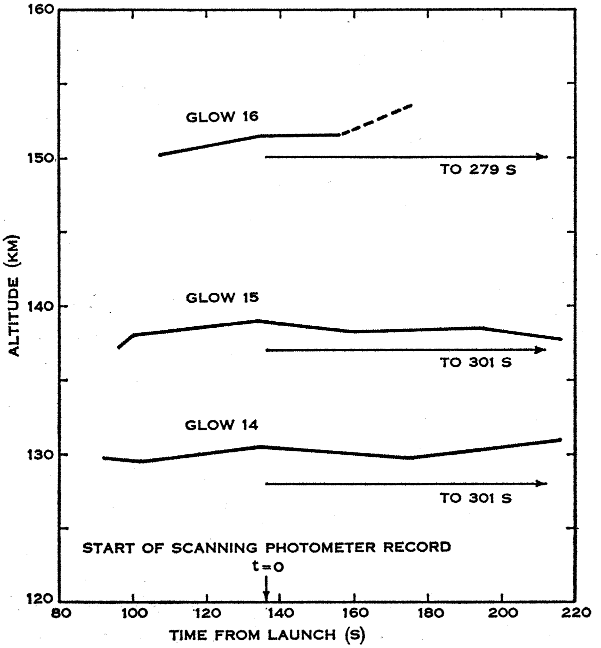

Fig. 1.—Altitude variation of glow clouds determined from photographic records.

The theoretical model is similar to that used by Pressman and Marmo (1961), with the addition of a factor to represent depletion of the contaminant particles.

II. Theory

The differential equation describing the behaviour of a diffusing, spherical glow cloud which is being depleted at a rate kn at a point where the contaminant particle number density is n is

$$\frac{\partial n}{\partial t}=D\left\{\frac{\partial^2 n}{\partial r^2}+\frac{2}{r}\frac{\partial n}{\partial r}\right\}-kn,$$

where D is the diffusion coefficient; see, for example, Crank (1956). The solution of this equation with the initial Gaussian distribution

$$n(\rho,0)=n_0\exp\{-\rho^2\}, \tag{1}$$

where $\rho = r/r_0$, r_0 being the initial effective radius of the cloud and n_0 the initial peak number density, is

$$n = n_0 b^3 \exp\{-b^2\rho^2 - kt\}, \tag{2}$$

where $b = (1+4Dt/r_0^2)^{-\frac{1}{2}}$ is a time-dependent dimensionless parameter.

In the following analysis, the zero for time is taken at the first observation on the cloud, and r_0 refers to this time. The effective radius of the cloud at the moment of the explosion (R) is calculated at the end of the analysis from the diffusion coefficient and r_0.

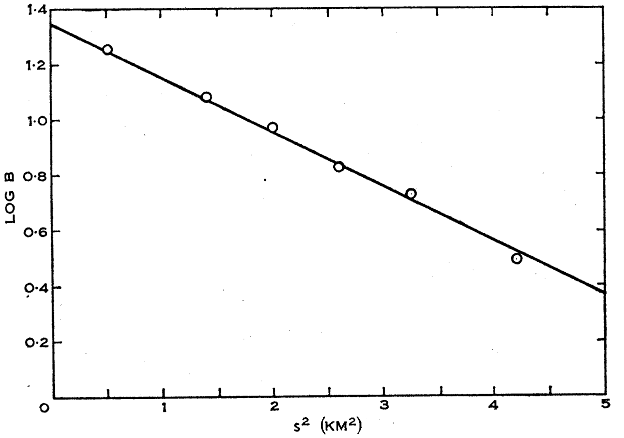

Fig. 2.—Radial brightness distribution of glow cloud. Glow 14 at $t = 96$ sec.

For an optically thin sunlit cloud, it can be shown (Lloyd 1963) that the brightness contour of the simple equivalent emitting surface of the cloud may be obtained by integrating equation (2) along the line of sight. At a distance s from the centre of the cloud the brightness B is given by

$$B = B_0 b^2 \exp\{-b^2\sigma^2 - kt\}, \tag{3}$$

where B_0 is the initial centre-point brightness and $\sigma = s/r_0$.

From equation (3) it is clear that the radial brightness distribution of the cloud is best examined by plotting the logarithm of the brightness against the square of the radius. Should the diffusion model be correct, these points will lie on a straight line. Such a plot is given in Figure 2 and shows that this is the case.

Experimental results on cloud brightness may be used to give two independent determinations of diffusion coefficient. The theoretical derivation of these two methods is given below. Method A is based on the radial distribution of brightness and Method B on the centre-point brightness.

Method A.—Taking the logarithm to base 10 of both sides of equation (3), differentiating with respect to s^2, and rearranging gives

$$-0\cdot 4343/[\partial \log B/\partial s^2] = r_0^2 + 4Dt. \tag{4}$$

A plot against time of the inverse of the gradient of log B against s^2 may thus be used to give the first estimate of D and r_0^2.

Method B.—The total light flux scattered by an optically thin sunlit cloud is given by twice the surface integral of the brightness of the equivalent simple emitter, if isotropic scattering is assumed. Integrating equation (3) gives the following expression for the total light flux

$$\begin{aligned} L &= 2\pi r_0^2 B_0 \exp\{-kt\} \\ &= 2\pi(r_0^2+4Dt)B_{\mathrm{max.}}, \end{aligned} \tag{5}$$

where $B_{\mathrm{max.}}$ is the peak brightness at time t (i.e. $B_{\mathrm{max.}} = B_0 b^2$), and r_0^2+4Dt has been determined in method A. From the rate of decay of the total light flux, k may be determined (step B_1).

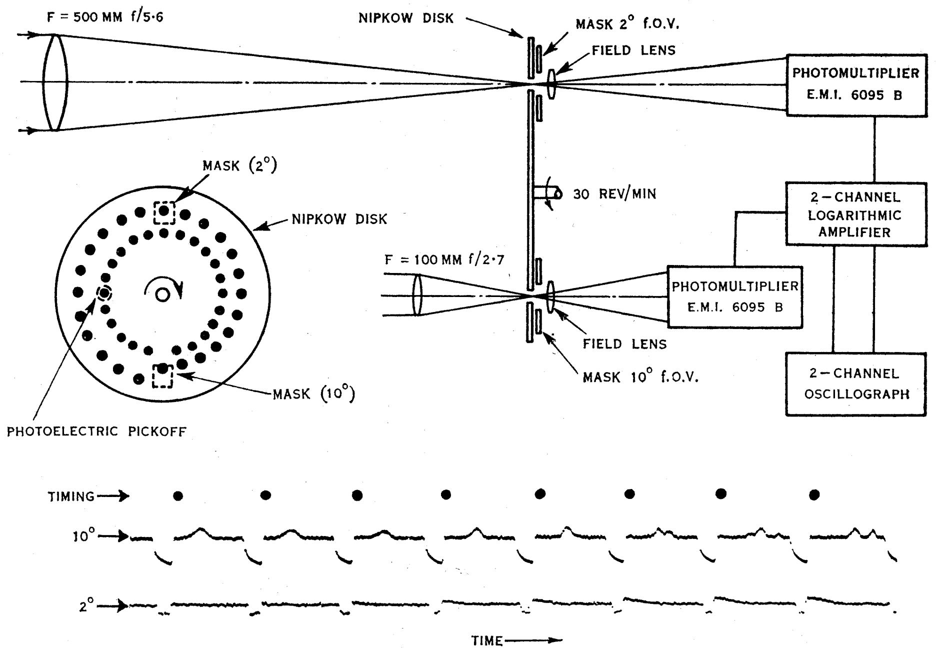

Fig. 3.—Scanning photometer and typical record.

The variation in peak brightness of the cloud with time is best shown by rewriting equation (3), with $\sigma = 0$, in the form

$$(B_0/B_{\mathrm{max.}})\exp\{-kt\} = 1+4Dt/r_0^2. \tag{6}$$

This shows that a plot of $(B_0/B_{\mathrm{max.}})\exp(-kt)$ against time will give a straight line whose gradient is $4D/r_0^2$ (step B_2).

Denoting by $B_{\sigma=1}$ the brightness of the cloud at $r = r_0$, and taking logarithms of both sides of equation (3) gives the expression

$$\log B_{\sigma=1} = \log B_{\mathrm{max.}} - 0\cdot4343/[1+4Dt/r_0^2]. \tag{7}$$

Using $4D/r_0^2$ obtained from step B_2, log $B_{\sigma=1}$ may be calculated. The corresponding value of r^2 ($= r_0^2$) can be read from the plots of log B against r^2 (step B_3). Steps B_2 and B_3 are combined to give a second estimate of D. This step completes method B.

III. Observations

Experience in the field of photographic photometry had demonstrated the difficulties involved in making accurate measurements of the brightness distribution of the glow clouds from photographs taken during a trial. It was to avoid these difficulties and to increase the sensitivity of the observations that a scanning photometer was built. A schematic diagram of this instrument and a typical record are shown in Figure 3.

The scanning photometer is a two-channel instrument, one channel having a field of view 10° square and the other 2° square. Both channels use the same Nipkow disk to scan the field of view. The disk, which rotates at 30 r.p.m., has a

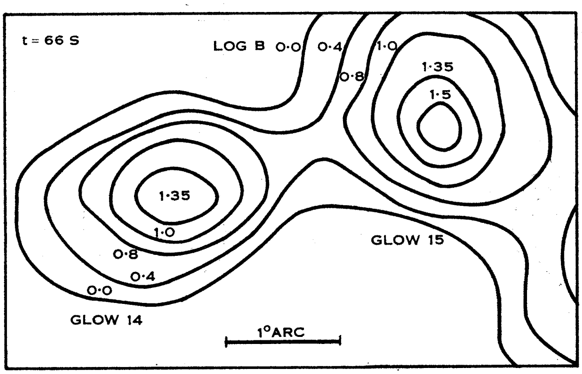

Fig. 4.—Isophots obtained from record of scanning photometer.

spiral of 31 holes spaced $2\pi/32$ radian apart. The blank position, where one further hole could have been placed, gives a measure of the dark current of the photomultipliers and identifies "frames". An inner ring of 31 holes, at constant radius, is detected photoelectrically to give timing and "line" identification.

The photomultipliers feed logarithmic amplifiers using electrometer tubes adjacent to the bases of the photomultipliers. These amplifiers operate over about 6 orders of magnitude, which is more than enough to cover the range from twilight glow cloud brightness down to below the night sky brightness.

The output of the amplifiers is displayed on a double-beam cathode-ray oscillograph which is photographed by a shutterless, 35-mm continuous film camera running at 6 in/s. The output of the timing photodetector fires a neon tube which is photographed alongside the cathode-ray tube traces (Fig. 3). In Figure 3 the blank "frame" position does not occur in the trace reproduced. Furthermore, since the trial in March 1962 the logarithmic amplifiers have been neutralized to improve their fall time.

The sensitivity of the photometer was such that the 10° field channel could detect the night sky; the other channel was somewhat less sensitive than this. Since the trial the EMI type 6095B photomultiplier in the 2° field channel has been replaced

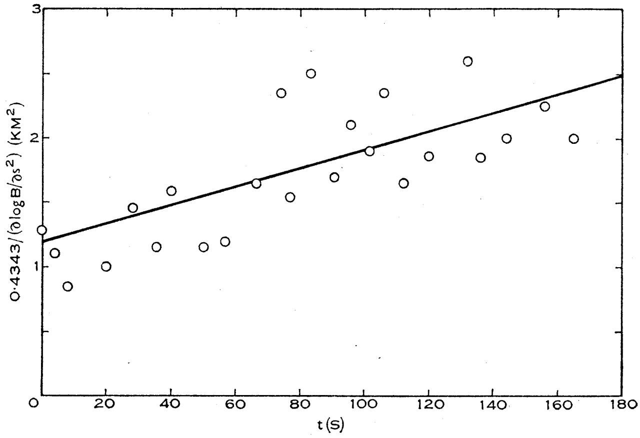

Fig. 5.—Gradient of cloud brightness (method A). Results for Glow 14: $D = 2 \cdot 3 \times 10^3$ m²sec⁻¹, $r_0 = 1 \cdot 09$ km.

by an EMI type 6097S, which has increased the sensitivity of this channel so that it can easily see the night sky.

At the time of the trial in March 1962, no equipment was available to calibrate the photometer absolutely in energy units. However, since only brightness ratios

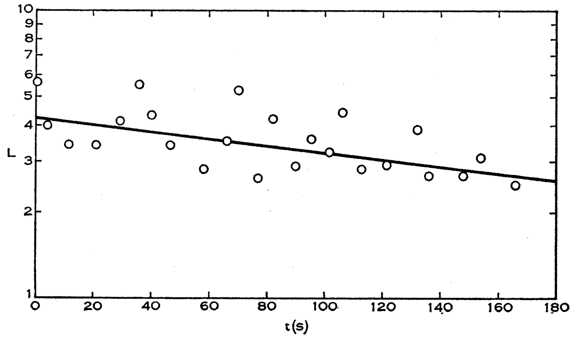

Fig. 6.—Variation with time of total light flux emitted (step B_1). Results for Glow 14, $k = 0 \cdot 0028$ sec⁻¹.

are involved in the calculation of diffusion coefficients it is sufficient to use an arbitrary unit for brightness.

IV. Results

Before data could be obtained from the records of the scanning photometer it was necessary to reconstruct from them the isophots for the glows; a part of one of these from the 10° field of view is reproduced in Figure 4.

As an example of the results obtained and an indication of the order of magnitude of their scatter, Figures 5–8 show the data points obtained for glow cloud number 14. The straight lines shown were fitted to the plotted points by the method of least

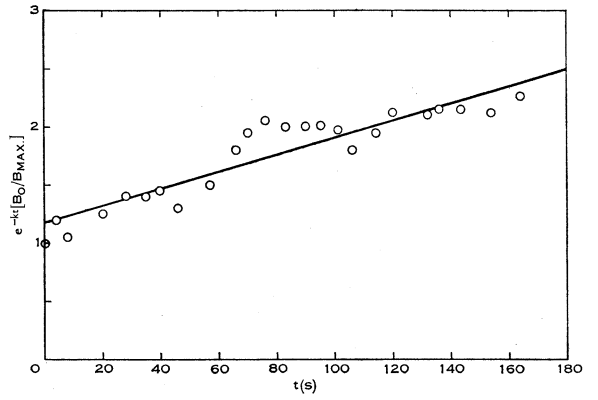

Fig. 7.—Peak brightness (step B_2). Results for Glow 14, $4D/r_0^2 = 0 \cdot 00596$ sec.

squares and, from these, the values of diffusion coefficient and initial effective radius were obtained (see Section II).

The steady decrease in the total flux emitted by the cloud (Fig. 6) is almost certainly due to depletion of the contaminant resulting from a second-order reaction

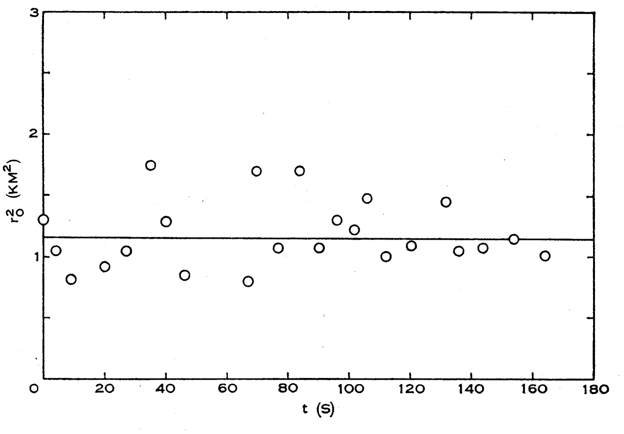

Fig. 8.—Initial effective radius (step B_3). Results for Glow 14, $r_0 = 1 \cdot 07$ km.

between it and the ambient atmosphere. When the number density of the contaminant is much less than that of the part of the atmosphere taking part in the reaction, and it is reasonable to suppose that this is the case here, the reaction is first-order and the rate constant k is equal to the product of the second-order rate constant and the number density of the reacting portion of the ambient atmosphere. Thus, knowledge of k and the chemical reaction allows one of these two factors to be determined once the other is known. The rate constant k for glow clouds 14, 15, and 16 was found to be 0·0028, 0·0017, and —0·0033 sec^{-1} respectively.

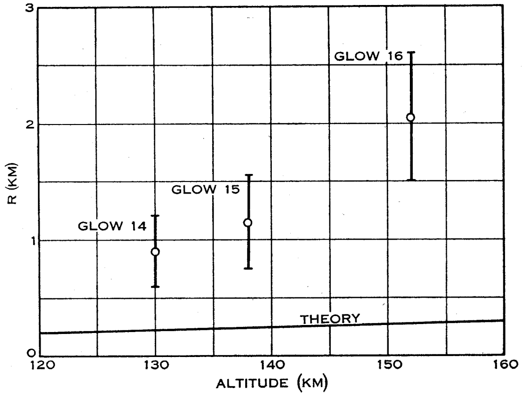

Fig. 9.—Effective radius at time of grenade explosion.

Spectrophotometric data obtained at the trial showed that most of the light emitted by the sunlit glow clouds falls in the molecular emission bands of AlO; thus AlO produced by the grenade explosion is the effective contaminant, and could

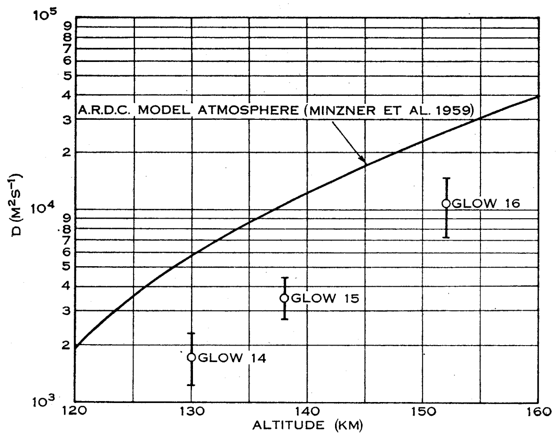

Fig. 10.—Diffusion coefficient.

occur in relatively large quantities since the "one-pound" grenades contain about 40% of powdered aluminium. However, the reaction which causes depletion of the AlO is not known and experiments are planned in an attempt to resolve this problem.

The initial effective radius corresponding to the first observation (r_0) can be converted to that at the time of explosion of the grenade (R) by assuming that the

Gaussian diffusion model is applicable over this time interval. It is found that

$$R^2 = r_0^2 - 4D\Delta t, \tag{8}$$

where Δt is the time between the explosion and first observation. The effective radius R for the clouds is plotted in Figure 9, where the standard error has been indicated. The solid line is the theoretical curve based on a yield of 86 g of gas with a molecular weight of 26. It assumes that the combustion products are in thermal and pressure equilibrium with the ambient atmosphere and that they have a radial Gaussian distribution (equation (1)), whose maximum number density is equal to the number density of the ambient atmosphere. The observed radii are much greater than the theoretical predictions, and the disparity increases with altitude.

Figure 10 shows the final results for the diffusion coefficient, with the standard error indicated. The results are the weighted means of the values given by methods A and B of Section II. Also shown is the theoretical value of the diffusion coefficient for the ambient atmosphere calculated from the expression given by Chapman and Cowling (1952), namely,

$$D = 1\cdot 019\,\frac{3}{8Nd^2}\left(\frac{kT}{\pi mM}\right)^{\frac{1}{2}}, \tag{9}$$

where d = molecular collision diameter,

m = mass of hydrogen atom,

M = molecular weight of the ambient atmosphere,

N = number density of ambient atmosphere,

T = temperature,

and (in this expression only) k denotes Boltzmann's constant. The A.R.D.C. model atmosphere (Minzner, Champion, and Pond 1959) has been used to supply the upper atmospheric parameters. The results may be converted to the interdiffusion of two gases of different molecular weights by replacing M by $2M_1M_2/(M_1+M_2)$ and d by d_{12}; this correction amounts to only a few per cent for the case under consideration. The experimental results are consistently lower than the theoretical curve, but show a similar altitude variation.

The standard error in the diffusion coefficient derived here is comparable with that quoted in the work of Shklovskii and Kurt (1960), but much less than obtained by Manring (1961), who observed the diffusion of a sodium vapour trail release. It is unfortunate that none of the glows were recorded below 120 km since it is in this region that there is a sharp drop in the theoretical diffusion coefficient; however, it is hoped to include observations on glows in the range 90–120 km at the next series of trials.

V. Acknowledgments

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VI. References

Bates, D. R. (1950).—*J. Geophys. Res.* **55**: 347–9.

Chapman, S., and Cowling, T. G. (1952).—"The Mathematical Theory of Non-uniform Gases." 2nd Ed. p. 198. (Cambridge Univ. Press.)

Crank, J. (1956).—"The Mathematics of Diffusion." 1st Ed. p. 124. (Oxford Univ. Press.)

Groves, G. V., *et al.* (1960).—"Space Research."—Proc. first Int. Space Sci. Symposium, Nice. (Ed. H. K. Bijl) p. 144. (North Holland Publishing Co.)

Lloyd, K. H. (1963).—Weapons Research Establishment. Tech. Note HSA 84. (in press.)

Manring, E. R. (1961).—*Aerospace Eng.* **20**: 8–9.

Marmo, F. F., Pressman, J., Manring, E. R., and Aschenbrand, L. (1960).—*Planet. Space Sci.* **2**: 174–86.

Minzner, R. A., Champion, K. S. W., and Pond, H. L. (1959).—The A.R.D.C. model atmosphere. Airforce Surveys in Geophys. No. 115 (G.R.D., U.S.A.F.).

Pressman, J., and Marmo, F. F. (1961).—*A.R.S. Journal* **31**: 819–21.

Shklovskii, I. S., and Kurt, V. G. (1960).—*A.R.S. Journal* **30**: 662–7.