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Variation in the Deposition Velocity of a Reactive Gas

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Abstract

The flux of a gas to the surface arising from absorption (or other chemical and physical processes) is commonly represented by the product of the concentration of the gas at some height z and a velocity, called the deposition velocity, v_d. Since the concentration changes with height, the deposition velocity must also change. Furthermore since the flux of the gas to the surface and its uptake there depends on the surface fluxes of momentum and heat, v_d must also be made a function of these variables too. If we follow Chamberlain (1966) and represent the extra surface resistence in terms of B, the Stanton number of Owen and Thomson (1963), which Thom (1972) expressed in terms of the surface friction velocity u_*, then v_d can be determined as a function of wind speed and temperature gradient.

The Analysis

For any rough surface, there is less resistence to the air to surface transfer of momentum than to the transfer of gaseous material or heat because momentum can be taken up through the action of pressure forces whereas the others depend entirely on molecular conduction. The difference in the resistences has been expressed in terms of the dimensionless reciprocal Stanton number B^{-1} introduced by Owen and Thomson (1963), defined by :

$$v_d(z) = \frac{u_*^2}{u(z)} \left[1 + \frac{u_*}{u(z)} B^{-1} \right]^{-1} \qquad (1)$$

Experiments carried out by Chamberlain (1966), Rider (1954) and Thom (1972) strongly suggest that B varies very little over a wide range of vegetated surface-types. B does vary, however, with the friction velocity u_*. Thom finds best agreement with a relationship of the form:

$$B^{-1} = a\, u_*^{1/3} \qquad (2)$$

in which a is purely a function of the character of the gas being deposited. For example a(Thorium B) / a(water vapour) = 1.6, according to Chamberlain (1966). The results presented in this paper are for a = 10 (when u_* is in m s^{-1}), which is appropriate for Thorium B, a fairly reactive gas used by Chamberlain. Figure 1 suggests that for other materials in which the coefficient a might range from 0 to 40 the value of v_d would change from roughly $2v_d$(ThB) to $\frac{1}{2}v_d$(ThB).

Although B^{-1} appears to be very insensitive to the surface, typified by the roughness length z_o, the deposition velocity $v_d(z)$ depends much more sensitively on z_o through the dependence of u_* and $u(z)$ on z_o, by which v_d is expressed (eqn (1)).

This dependence is shown in Figure 2 for a given geostrophic wind ($G = 10 \text{m s}^{-1}$). The Figure also shows how relatively insensitive v_d at a height of 1 metre is to the surface sensible heat flux H, at least on the unstable side when $H \geqslant 0$.

To calculate v_d, for a given value of the parameter a and a specified z_o, values of u_* and $u(z)$ have to be calculated. It will be assumed that $u(2)$, the wind speed at a height of 2 metres, is available together with a temperature difference between 1 metre and 8 metres.

The following Monin–Obukhov similarity relations are assumed to be valid within the surface layer of the boundary layer:

$$\frac{du}{dz} = \frac{u_*}{kz} \varphi_m\left(\frac{z}{|L|}\right) \tag{3}$$

where $L = -\dfrac{\rho c_p T u_*^3}{k g H}$

H = the surface sensible heat flux $\equiv \rho c_p u_* T_*$ (Wm^{-2})

T = temperature ($^{\circ}$K)

ρ = air density

c_p = specific heat at constant pressure

g = acceleration due to gravity

k = von Karman's constant, taken to be 0.4

Ignoring small changes in temperature and their effect on ρc_p ($\rho c_p \cong 1305 - 4.3(T-273)$) we may write

$$L \cong 29.358 \frac{u_*^2}{T_*} \tag{4}$$

Following Dyer and Bradley (1982) on the unstable side (L negative):

$$\varphi_m = \left(1 + 28 \frac{z}{|L|}\right)^{-1/4} \qquad \text{for} \quad 0 < \frac{z}{|L|} < 4 \tag{5a}$$

equation (3) may be integrated to give:

$$u(z) = \frac{u_*}{k}\left\{ \ln\left[\frac{s-1}{s+1} \cdot \frac{s_o+1}{s_o-1}\right] + 2\tan^{-1} s - 2\tan^{-1} s_o \right\} \tag{6a}$$

where $s = \left(1 + 28 \frac{z}{|L|}\right)^{1/4}$

and $s_o = \left(1 + 28 \frac{z_o}{|L|}\right)^{1/4}$

On the stable side we take Webb's (1970) form:

$$\varphi_m = \left(1 + 5.2 \frac{z}{L}\right) \qquad \text{for} \quad 0 < \frac{z}{L} < 1 \tag{5b}$$

which yields

$$u(z) = \frac{u_*}{k}\left\{ \ln \frac{z}{z_o} + \frac{5.2}{L}(z - z_o) \right\} \tag{6b}$$

Similarly

$$\frac{d\theta}{dz} = \frac{T_*}{k} \varphi_H\left(\frac{z}{|L|}\right) \tag{7}$$

On the unstable side, we again use Dyer and Bradley's results:

$$\varphi_H\left(\frac{z}{|L|}\right) = \left(1 + 14 \frac{z}{|L|}\right)^{-1/2} \qquad \text{for} \quad 0 < \frac{z}{|L|} < 4 \tag{8a}$$

whereas on the stable side, following Webb:

$$\varphi_H\left(\frac{z}{L}\right) = \left(1 + 5.2 \frac{z}{L}\right) \qquad \text{for} \quad 0 < \frac{z}{L} < 1 \tag{8b}$$

The temperature difference between 1 and 8 metres is therefore

$$\Delta\theta = \frac{T_*}{k} \ln\left[\frac{r_8 - 1}{r_8 + 1} \cdot \frac{r_1 + 1}{r_1 - 1}\right] \qquad \text{for } L < 0 \qquad (9a)$$

where $r_8 = \left(1 + 14 \times \frac{8}{|L|}\right)^{1/2}$

$r_1 = \left(1 + 14 \times \frac{1}{|L|}\right)^{1/2}$

or

$$\Delta\theta = \frac{T_*}{k}\left[\ln 8 + 5.2\ (8-1)/L\right] \approx T_*\left(5.2 + \frac{91}{L}\right) \qquad \text{for } L \geqslant 0 \qquad (9b)$$

Thus for given u_* and T_*, equations (6) and (9) enable values of u(2) and $\Delta\theta$ to be determined. It is clearly easier to go from u_* and T_* to u(2) and $\Delta\theta$ than the reverse since L cannot readily be expressed in terms of u(2) and $\Delta\theta$. Whilst this is fine for the construction of graphs it has to be recognised that it is less convenient for numerical determinations of v_d from the measured variables u(2) and $\Delta\theta$. Here some form of iteration is required in which informed guesses for u_* and T_* are made, these are used to form L and then L is used to derive better estimates of u_* and T_* in terms of u(2) and $\Delta\theta$, a revised value of L, even better estimates of u_* and T_*, and so on until steady values are reached.

No matter the magnitude of H, for small enough z the $\varphi \approx 1$. Thus if $\varphi_{gas} \equiv \varphi_H$, $\varphi_{gas} = 1$ for small enough z. If q(z) is the concentration of the gas at height z, if E is the downward flux of the gas and z_q is the equivalent of z_o, then

$$q(z) - q(0) = \frac{E}{k\rho u_*} \ln\frac{z}{z_q}$$

where E by definition is given by $E = \rho v_d(z)[q(z) - q(0)]$

Thus $\dfrac{u_*}{v_d(z)} = \dfrac{1}{k}\ln\dfrac{z}{z_q}$ for small enough z.

Inserting this into equation (1) and assuming for the same small z that $\dfrac{\bar{u}(z)}{u_*} = \dfrac{1}{k}\ln\dfrac{z}{z_o}$ it follows that

$$B^{-1} = \frac{1}{k} \ln\frac{z_o}{z_q} \qquad \text{independent of z.} \qquad (10)$$

or

$$z_q = z_o \exp(-kB^{-1}) = z_o \exp(-aku_*^{1/3}) \qquad (11)$$

For more general z:

$$v_d(z) = \frac{ku_*}{\int_{z_q}^{z} \frac{1}{z}\varphi_H\left(\frac{z}{|L|}\right)dz} \qquad (12)$$

which by generalising equations (9) can be seen to take the forms:

$$v_d(z) = \frac{ku_*}{\ln\left[\frac{r-1}{r+1} \cdot \frac{r_q+1}{r_q-1}\right]} \qquad \text{for } L < 0 \qquad (13a)$$

where $r = \left(1 + 14\frac{z}{|L|}\right)^{1/2}$

$r_q = \left(1 + 14\frac{z_q}{|L|}\right)^{1/2}$

and

$$v_d(z) = \frac{ku_*}{\ln\frac{z}{z_q} + \frac{5.2}{L}(z - z_q)} \qquad \text{for } L \geqslant 0 \qquad (13b)$$

where z_q is given by equation (11).

Results

For specified z_o and parameter a, equations (6),(9),(11) and (13) can be solved to give $u(2)$, $\Delta\theta$, z_q and $v_d(z)$ in terms of u_*, T_* and z. The results are displayed in Figures (1) - (4). Figure (1) shows the effect of parameter a on v_d at 1 metre in moderately unstable conditions. Figure (2) shows the sensitivity of v_d at 1 metre to z_o and to the surface sensible heat flux H for a given geostrop wind. If a polluted airmass passes over heterogeneous terrain in which z_o varies by two orders of magnitude, say, then $v_d(1)$ might vary by a factor of 3 according to this Figure. The variation of the deposition per unit area would probably be less than this because $q(1)$ would tend to decrease over areas of high $v_d(1)$ and vice versa.

Figure (3) shows two rather typical $v_d(z)$ profiles in unstable and stable conditions. In unstable conditions v_d varies only by some 20% above 1 metre, and most of this variation occurs below 10 metres. In stable conditions a much greater variation is seen, reflecting the larger variation in concentration due to stabilisation.

Figure (4) is a nomogram for estimating v_d at 1 metre in terms of $u(2)$ and $\Delta\theta$ for z_o = 1 cm. The parameter a is taken to be 10. In stable conditions v_d depends almost entirely on $u(2)$:

$$v_d \approx 2.8\, u(2) \qquad (\text{u is in m s}^{-1},\ v_d \text{ in cm s}^{-1}) \qquad (14)$$

whereas in unstable conditions v_d increases significantly with increasing $\Delta\theta$. Contours of heat flux H and Pasquill Stability P are also shown (Pasquill, 1974). In near neutral and unstable conditions H depends principally on $\Delta\theta$, varying only slowly with $u(2)$, whereas in stable conditions ($P > 5$) H depends mainly on $u(2)$ and hardly at all on $\Delta\theta$. On the stable side, two curves are given for each stability value of P to indicate the present uncertainty in the validity of a simple assumption that $P = P(u, \Delta\theta)$, and the range of values implied by very limited data (Smith, unpublished).

The important conclusion is one that has been known for a long time but is often "conveniently forgotten", namely that v_d is not only a function of height z but also depends quite critically on the state of the surface layer and chiefly on u and $\Delta\theta$.

Nevertheless the picture is a little simpler when we consider the deposition rate of a gas being emitted from a source a long way upstream. Ignoring for a moment the response of the total depth of the cloud of gas to wind speed, there is a tendency for the increase in v_d as wind speed increases to be balanced by a corresponding decrease in local concentration due to dilution effects at source. The deposition rate per unit surface area is by definition the product of the two and therefore depends only weakly on the state of the air, provided this is virtually constant. If the depth of the cloud increases significantly with increasin wind speed then the deposition rate will decrease and more material will travel to longer range. However on many occasions, perhaps even the majority of occasions, vertical mixing will be controlled by factors other than wind speed (e.g. by an overhead inversion), and this means that as a first approximation it may be sufficient in long range transport models to ignore the influence of u and on deposition rates unless "extreme" stability conditions (A and G) are encountered.

Surface Resistance

Many gases experience a further resistance to their take-up at the surface. Sulphur dioxide is one of these. The extra resistance r_s reflects the apparent fact that such gases are absorbed most readily within the stomata on leaves, although other factors may also play a role. The magnitude of r_s and its dependence on the state of the soil and the vegetation has been broadly estimated in studies of the surface energy balance and the surface heat flux (Wang, forthcoming publication) and it may be argued that the same values should apply to gases like sulphur dioxide. Except at night, or within an hour of sunrise or sunset, the following rough values have been inferred from the energy balance studies:

		r_s ($s\ m^{-1}$)
Oct. - March :	soil contains moisture	80
	no rain for over 10 days	400
April- Sept. :	soil contains moisture	50
	soil dry	2000

The value of the dry deposition velocity v_d' for such gases is then given by:

$$\frac{1}{v_d'} = r_s + \frac{1}{v_d}$$

Data collected at Barton Aerodrome in 1973 by the CEGB (Sugden et al.,1976) will be used to test the applicability of such a correction. The important data from their Tables II and III are reproduced below together with inferences concerning the sensible heat flux H, r_s (expressed in units of $s\ cm^{-1}$), and the implied v_d'. Sugden et al.'s observed values of v_d' are deduced from measurements of SO_2 concentration at two levels. The accuracy of such estimates is probably not very high and some anomolies are apparent in the data : e.g. on June 12th, three consecutive estimates of v_d' in periods in which the meteorology and other relevant factors changed very little were 0.74 $cm\ s^{-1}$, 2.29 and 1.02.

The "observed" v_d' have a mean value of 0.95 $cm\ s^{-1}$, and the calculated v_d' have a mean 0.89 $cm\ s^{-1}$ (in reasonably good accord), whereas the uncorrected v_d have a mean 2.17 $cm\ s^{-1}$ which is much too high. The correlation coefficient between $v'_{d\ calc.}$ and the $v'_{d\ obs.}$ is 0.61, which in view of the uncertainty in the input data is quite reasonable. Figure 5a shows a plot of the calculated v_d against the observed v_d', showing that without the r_s-correction the agreement is quite poor, all the v_d' being significantly larger than the v_d. Figure 5b on the otherhand shows better overall agreement, albeit still with quite a lot of scatter. Obviously with such limited data no claim can be made that the method has general validity, but at least the results are promising.

Table : BARTON data on SO_2 Deposition Velocities

Date	Julian day No.	Time	Cloud	u_1	u_2(approx)	u_{10}(approx)	T	$T_{dew\ pt.}$	r_s	H (estim.)	v_d calc	v_d' calc.	v_d' obs.
2.3.73	61	12	6	4.5	5.2	5.8	8.3	4	0.8	28	2.1	0.78	1.57
		13	5	4.9	5.7	6.4	9.8	4.5	0.8	12	2.3	0.81	1.42
		14	8	5.2	5.9	6.7	9.6	4	0.8	0	2.3	0.81	1.17
		15	8	4.2	4.8	5.4	9.5	5	0.8	0	1.9	0.75	0.50
13.3.73	72	12	8	3.4	3.9	4.4	6.2	-1	0.8	25	1.7	0.72	0.42
		14	8	2.3	2.6	3.0	6.4	1	0.8	25	1.3	0.64	0.27
		15	8	1.8	2.1	2.3	6.5	0	0.8	6	1.0	0.56	0.36
23.3.73	82	12	2	3.9	4.5	5.1	13.5	0	4.0	164	2.1	0.22	0.21
26.4.73	116	15	5	4.5	5.1	5.8	13	5	0.5	19	2.1	1.02	1.64
15.5.73	135	15	7	1.7	1.9	2.1	13.8	0	0.5	41	1.2	0.75	0.56
		16	7	1.3	1.5	1.6	14	3	0.5	28	1.0	0.67	0.46
17.5.73	137	14	7	7.8	9.0	10.1	17.4	-2	0.5	0	3.3	1.25	0.93
		15	6	7.9	9.1	10.2	17.1	-4	0.5	0	3.3	1.25	1.28
		16	7	8.7	10.0	11.3	15.5	-1	0.5	0	3.5	1.27	1.14
12.6.73	163	12	7	5.6	6.4	7.2	16.7	11	0.5	20	2.4	1.09	1.20
		13	7	5.7	6.6	7.4	16.9	11	0.5	14	2.5	1.11	0.74
		14	7	6.1	7.1	7.9	19.1	9	0.5	0	2.7	1.15	2.29
		15	7	5.5	6.4	7.1	17.7	11	0.5	0	2.4	1.09	1.02

Units: cloud in oktas (eighths), u in $m\ s^{-1}$, T in ^{o}C, r_s in $s\ cm^{-1}$, H in $W\ m^{-2}$, v_d in $cm\ s^{-1}$

Conclusions

These may be summarised as follows:

(i) the deposition velocity may be assessed using Monin–Obukhov similarity theory and the inverse Stanton number (B^{-1}) concept used by Chamberlain (1966).

(ii) B^{-1}, according to Thom (1972), can be written as $au_*^{\frac{1}{3}}$ where a is virtually independent of the nature of the vegetated surface and depends only on the character of the gas. Parameter a has to be determined experimentally.

(iii) v_d varies rather slowly with heat flux H and with the parameter a, but varies significantly with height z, especially in stable conditions.

(iv) except in extreme stability conditions, v_d is almost proportional to wind speed whereas the deposition rate per unit area is often independent of u and $\Delta\theta$ provided they are almost constant along the path of the cloud.

(v) Many gases of practical importance (like sulphur dioxide) do not find vegetated surfaces a perfect sink, and an extra surface resistance r_s has to be incorporated. The simple scheme presented for estimating r_s considerably improves the agreement between calculated and limited observed deposition velocities.

(vi) Over vegetated surfaces, the incorporation of r_s reduces the deposition velocity to values typically in the range 0.5 – 1.4 cm s^{-1}. This is very close to the range of values expected over sea surfaces (see Figure 2) where $z_o = 10^{-5} - 10^{-4}$ and $r_s=0$. Thus v_d' appears to be rather insensitive to the nature of the underlying surface, at least those commonly encountered in and around Europe.

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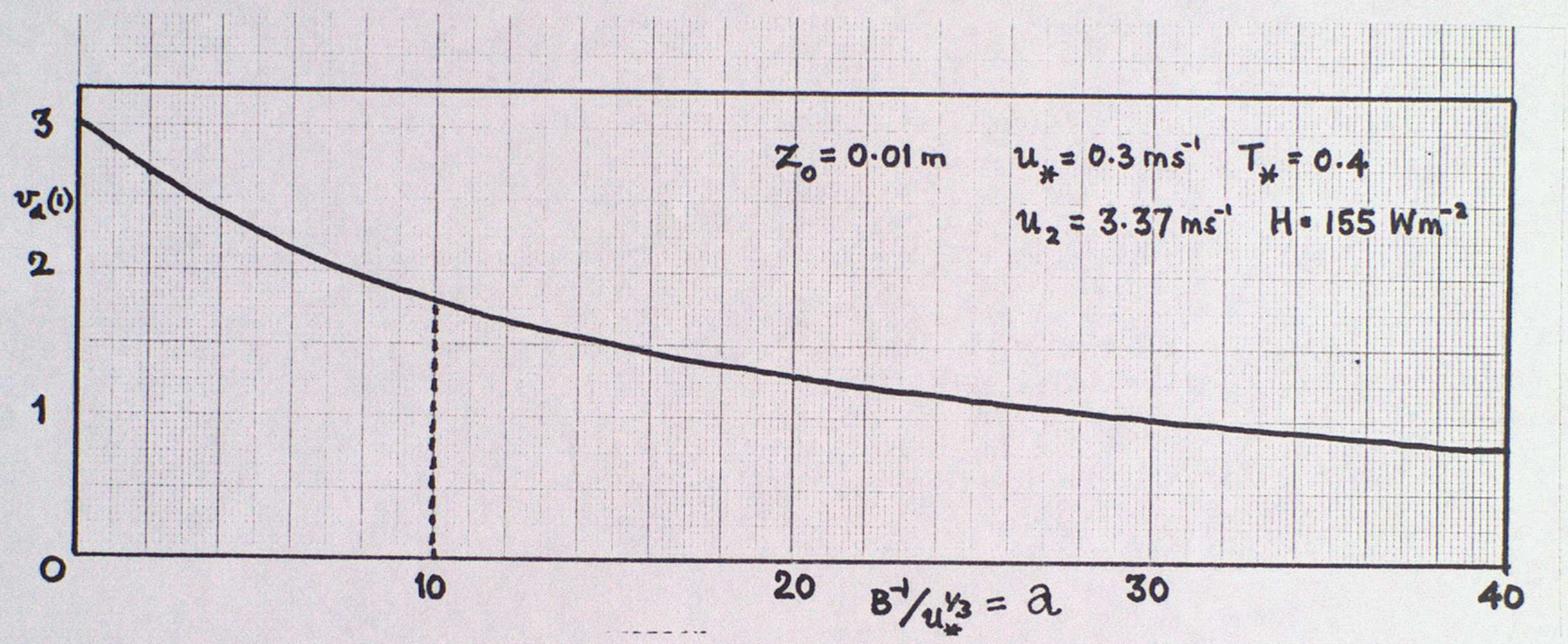


Figure 1. The variation of the deposition velocity v_d with the parameter a that appears in Thom's formulation in the inverse Stanton number B^{-1}.

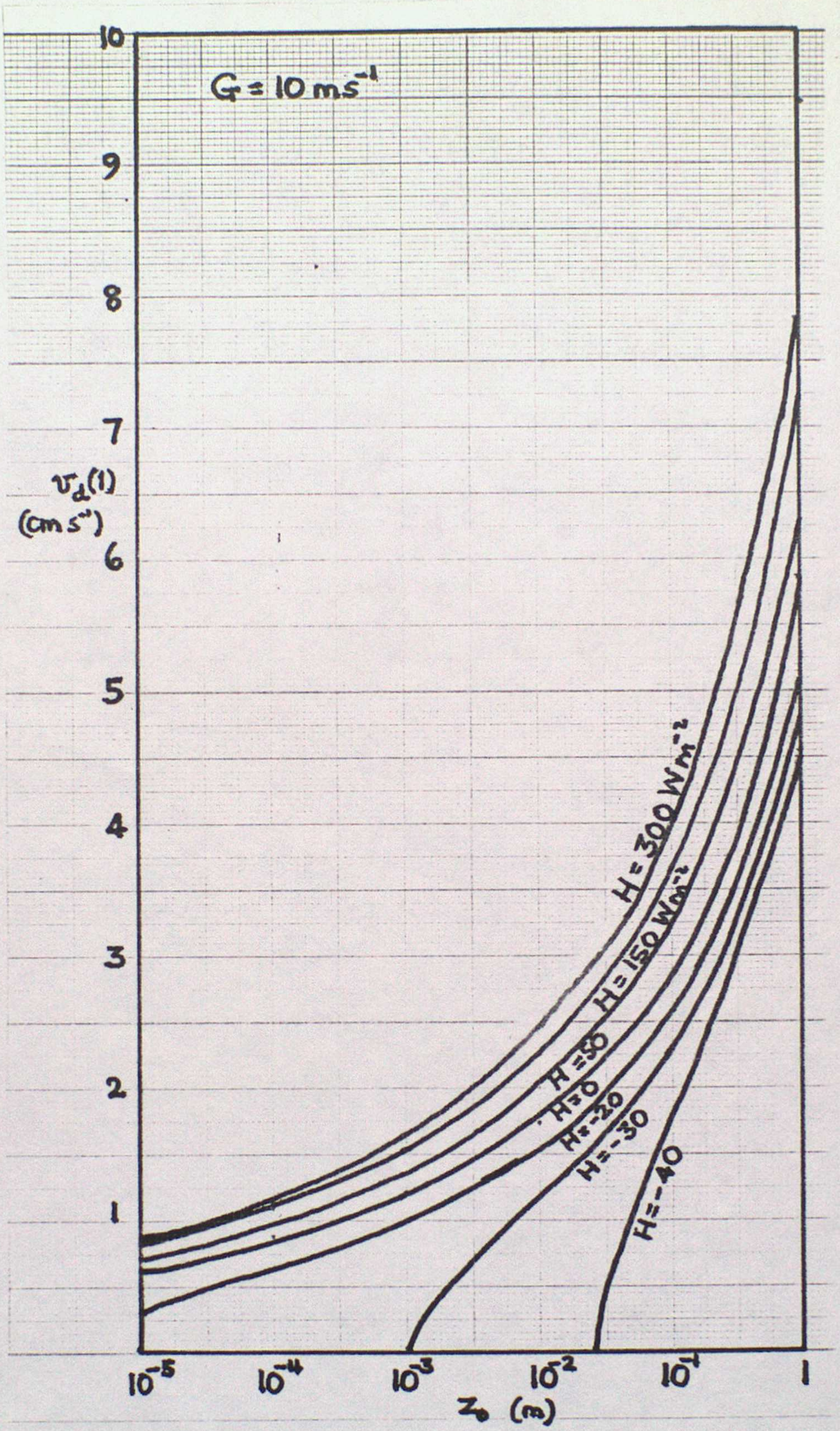


Figure 2. Variation in the deposition velocity v_d with surface roughness and heat flux H for a specified geostrophic wind speed G.

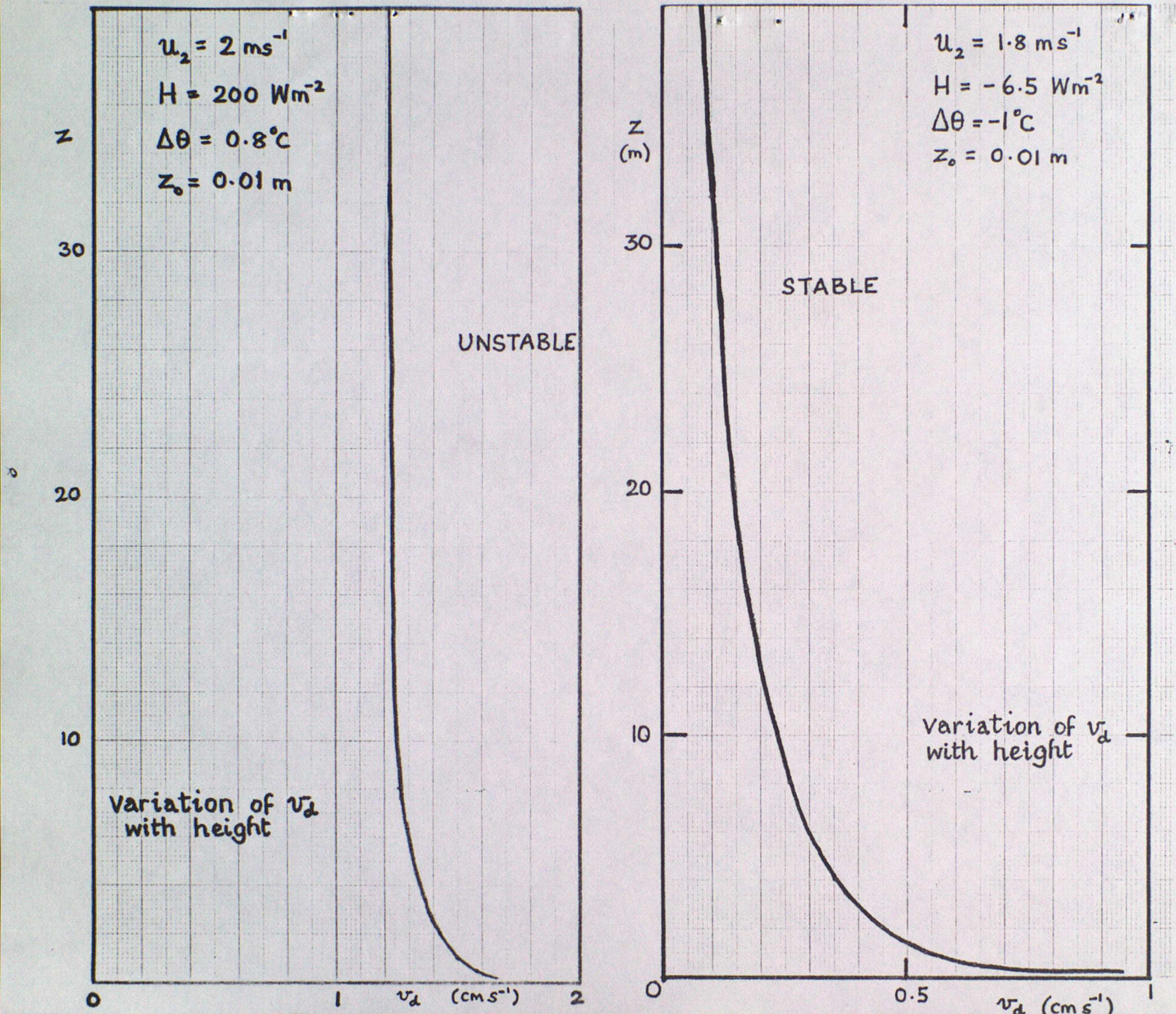


Figure 3.
Typical profiles of v_d in unstable and stable conditions.

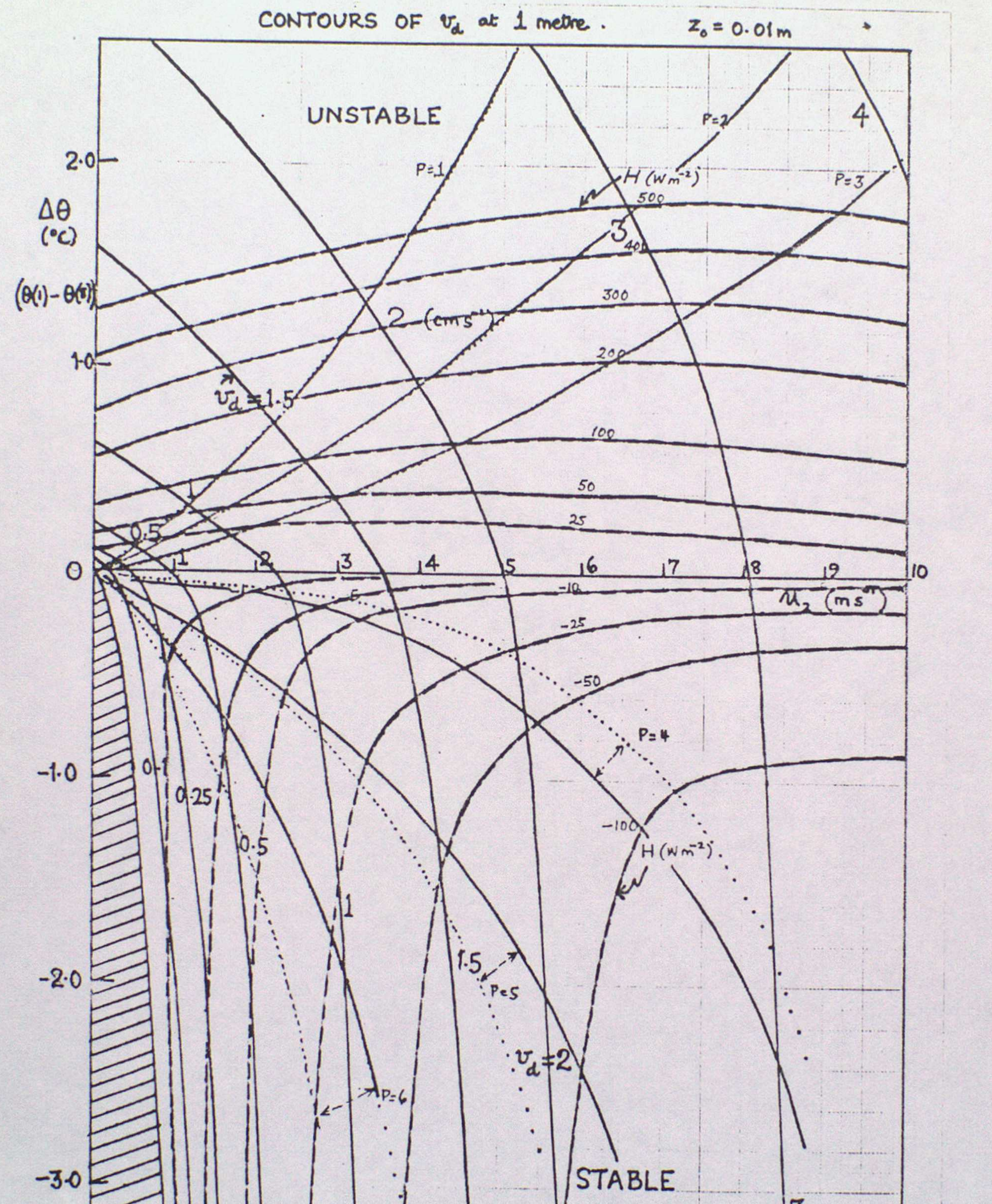
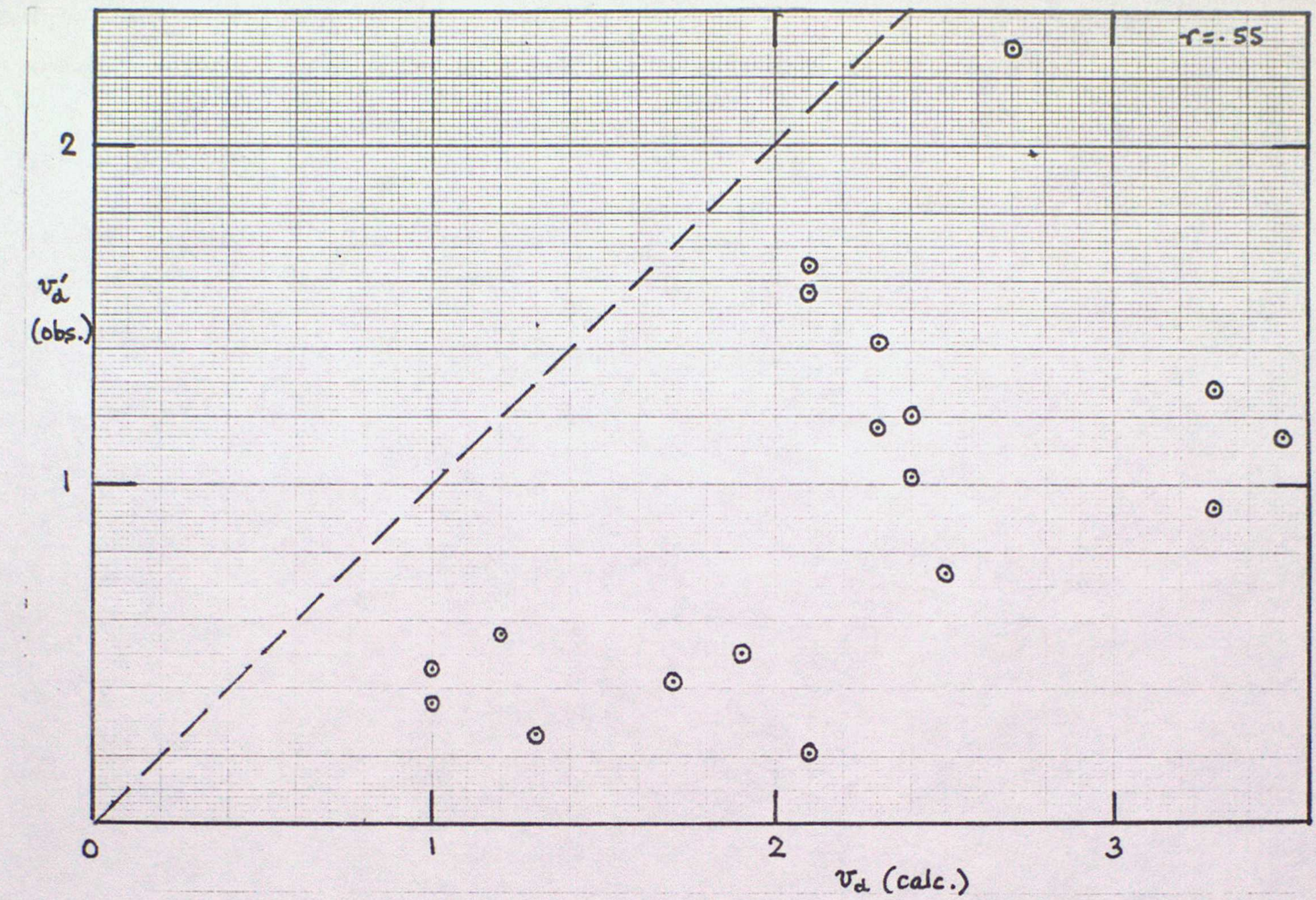


Figure 4. Contour lines of deposition velocity v_d, heat flux H, and Pasquill Stability P, for z_o = 0.01m and parameter a = 10.



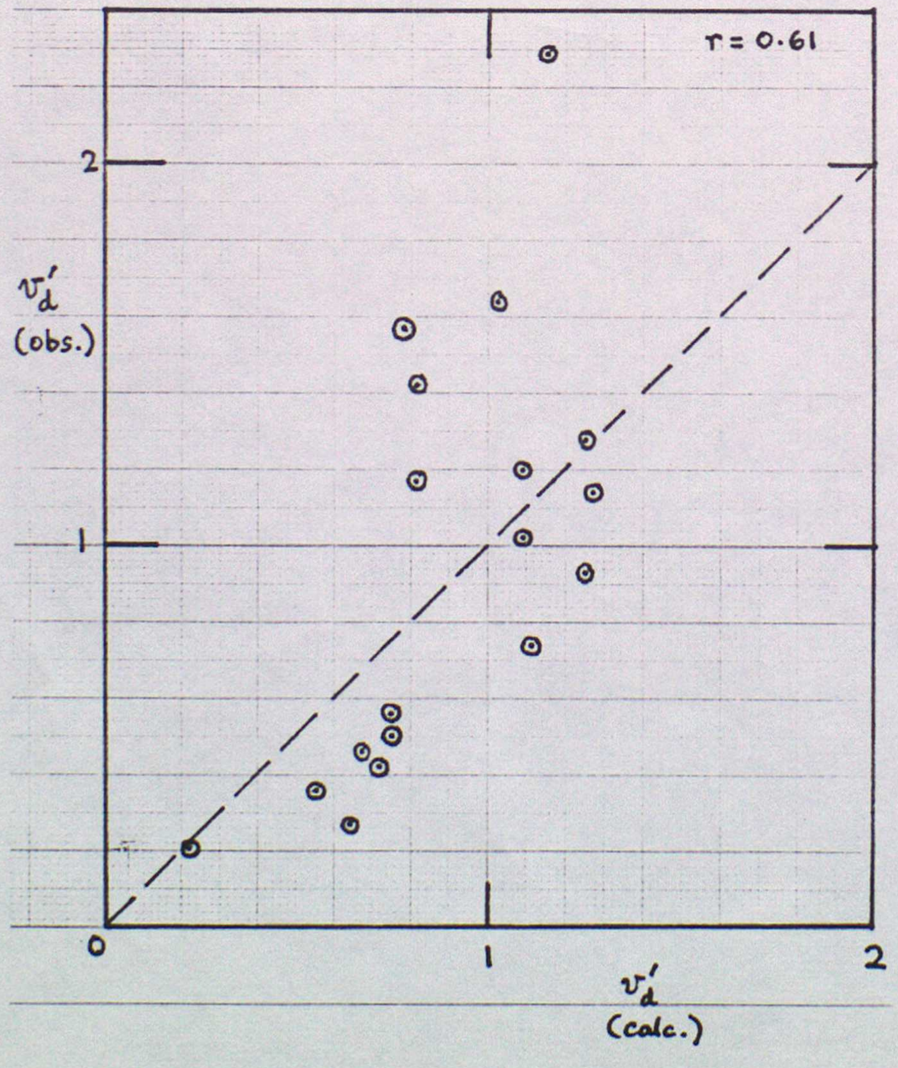


Figure 5a. A comparison of uncorrected v_d (calculated) and "observed" deposition velocities taken at Barton (Sugden et al.1976). The calculated values are clearly too large.

Figure 5b. The calculated values are now corrected by incorporation of the surface resistance r_s. Agreement with the "observed" is considerably improved although errors in the input data still give rise to significant scatter.