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**A MIXING LENGTH MODEL FOR GAS
TRANSFER AT THE AIR-WATER INTERFACE**

by

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ABSTRACT

Heat and mass fluxes between a moving fluid and its boundaries are generally limited by the resistance to transfer in the thin viscous sub-layer adjacent to the boundary. A mixing length model is constructed which yields a simple way of modelling these fluxes for any roughness Reynolds number and Schmidt number. The model is tested against laboratory data in small and large tanks for wide ranges of roughness Reynolds number and Schmidt number. The effects of density gradients in the fluid and waviness on the boundary are discussed in the context of the natural air-water interface.

RÉSUMÉ

Les flux de chaleur et de masse entre un fluide en mouvement et ses limites sont en général limités par la résistance au transfert dans la mince sous-couche visqueuse adjacente à la limite. On a construit un modèle de la longueur de mélange qui fournit un moyen simple de modéliser ces flux pour tout nombre de rugosité de Reynolds et de Schmidt. Le modèle est comparé à des données de laboratoire dans des essais dans des bassins petits et grands pour de vastes gammes de nombres de rugosité de Reynolds et de Schmidt. Les effets de gradient de densité dans le fluide et d'ondulation à la limite sont analysés dans le contexte de l'interface naturelle air-eau.

MANAGEMENT PERSPECTIVE

This paper proposes a new model for the transfer of gases at the air water interface. Understanding this process and being able to compute transfers of gases such as water vapour, sulphur dioxide and ammonia is essential for modelling evaporation losses, significant in the prairies. Sulphur dioxide, as well as falling in acidic rain, can be absorbed directly in large lakes and seas.

This research therefore should improve atmospheric models of gas and vapour exchange and will be significant for a number of uses including industrial gas exchanges such as ammonia.

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PERSPECTIVE DE GESTION

La présente communication propose un nouveau modèle pour le transfert des gaz à l'interface air-eau. Il faut comprendre ce processus et pouvoir calculer les transferts de gaz tels que la vapeur d'eau, le dioxyde de soufre et l'ammoniac pour modéliser les pertes par évaporation, importantes dans les Prairies. Le dioxyde de soufre, comme la pluie acide, peut être absorbé directement dans les grands lacs et les mers.

La présente recherche devrait donc contribuer à améliorer les modèles atmosphériques des échanges de gaz et de vapeurs et sera importante pour différentes applications comme les échanges de gaz industriels tels l'ammoniac.

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1.0 INTRODUCTION

The flux of gases across an air water interface is driven by a concentration difference. The partitioning of the concentration difference between liquid and gas phases depends on the resistance presented by each phase. Further, within each boundary layer the resistance to gas flux varies markedly with distance from the boundary. A phenomenological description of the way in which these resistances behave is at the heart of the problem of parameterizing gas transfer across air-water interfaces.

A model appropriate to gas transfer at a rigid boundary contains viscous and diffusive sublayers on both sides of the interface as well as the outer boundary layers in which turbulence reduces the resistance dramatically. Clearly the largest concentration difference occurs in the layer in which the largest resistance to flux occurs. Generally these are the diffusive sublayers on one or both sides of the interface. When the resistance is larger above the interface the flux of gas is said to be under gas phase control; when it is smaller the flux is under liquid phase control. Most of the environmentally common gases fall clearly into one or the other camp. Liss (1983) lists H_2O , SO_2 , SO_3 and NH_3 as being under gas phase control; while O_2 , N_2 , CO_2 , CO , CH_4 and the inert gases are under liquid phase control. Because the molecular diffusivity of gases in liquids is much lower than in the gas phase, gas fluxes are under liquid phase control unless the gas has high aqueous solubility or chemical reactivity.

For gases under gas phase control the problem of estimating gas fluxes, given an air-water concentration difference, amounts to determining the gas transfer coefficient, C_g

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$$C_g = - \frac{F}{\rho(G_z - G_s)(U_z - U_s)} \quad (1)$$

where the gas flux F is positive upwards and is independent of height; ρ is the air density, U is the mean wind speed, G the mean gas concentration and the subscripts s and z refer to the surface and any other height in the constant flux and stress layer. The vertical coordinate z is positive upwards with its origin at the mean surface.

Under the usual assumptions of steady state and horizontal homogeneity, the gas transfer coefficient can depend only on other non-dimensional parameters.

$$C_g = f\left(\frac{Z}{Z_0}, R_*, Sc, \zeta, n\right) \quad (2)$$

where Z_0 is the virtual origin of the outer logarithmic velocity profile or the roughness length; $R_* = Z_0 u_* / \nu$ is the roughness Reynolds number, u_* is the friction velocity (ρu_*^2 is the surface stress), ν is the kinematic viscosity; $Sc = \nu/D$ is the Schmidt number, D is the molecular diffusivity of the gas; ζ is the Monin-Obukhov (1954) stability index and n represents the effect of the mobility of the surface.

The functional dependence (2) is the gas transfer law. This paper is devoted to the development of a semi-empirical gas transfer law for gases under gas phase control. As explained above, this entails a description of the boundary layers above the water surface. Various attempts to establish a gas transfer law have been given by Sverdrup (1937), Sheppard (1958), Kader and Yaglom (1972), Yaglom and Kader (1974), Brutsaert (1975), Deacon (1977), Liu et al (1979). In all cases the boundary layer is modelled using semi-empirical theories applicable to one or both asymptotes of aerodynamically smooth ($R_* = 0.1$) or rough ($R_* > 2.2$) flow. Much of the time natural water surfaces are transitional, i.e., neither completely

aerodynamically smooth or fully rough. Figure 1 illustrates this for typical oceanic conditions. The relationship between R_* and U_{10} shown is based on the average dependence of the aerodynamic drag coefficient on wind speed given by Garratt (1977). In general, the open ocean is aerodynamically transitional when the wind speed is between 2.8 m/s and 7.5 m/s. Clearly a boundary layer model which treats a transitional surface as the intersection of the smooth and rough asymptotes will often be inappropriate. Furthermore, it is common practice in modelling gas transfer to treat the water surface as a flat plate. The most obvious characteristic of a wind-disturbed water surface is the presence of waves, i.e., mobile roughnesses. In this paper we attempt to rectify both these deficiencies by describing a gas transfer law which is appropriate for any value of roughness Reynolds number R_* and which incorporates the effects of surface waves.

2.0 EFFECTIVE ROUGHNESS

The large density difference between air and water suggests that, from the point of view of the air flow, the water surface behaves much like a solid wall; i.e., the air flow does not appreciably deform the water surface. Of course, deriving its energy from the air flow, the water moves vertically and horizontally and its deformations grow and propagate. However, it is still reasonable to apply the "solid wall analogy" if, near the surface, the time scale of development of the turbulence $\sim (dU/dZ)^{-1}$ is small compared to the time scale of evolution of the surface relief (a few wave periods). Riley et al. (1982) have demonstrated that this is indeed the case. Thus it is sufficient, at least for gas transfer calculations of gases under gas phase control, to characterize the surface by an "effective roughness".

It is well known that wavy water surfaces are generally aerodynamically smoother than a snapshot of the surface texture would suggest. This has led to the hypothesis that the very short waves, which are in equilibrium with the wind, are the roughness elements.

Dimensional arguments then yield Charnock's (1955) relation $Z_0 \sim u_*^2/g$. Kitaigorodskii and Volkov (1965), Kitaigorodskii (1968) and Donelan (1982) have demonstrated by argument and observation that all components of the wave spectrum contribute to the effective roughness, but that the contribution of the longer waves to the total form drag is reduced by their greater phase speed in the wind direction. Kitaigorodskii and Volkov (1965) have shown that the effective roughness height h of the waves is given by:

$$h = \left[\int_0^{\infty} \phi(\omega) \exp \{-2k g / (u_* \omega)\} d\omega \right]^{1/2} \quad (3)$$

On the assumption of a wave spectrum $\phi(\omega)$ with a saturated rear face, h has two asymptotic regions depending on the stage of wave development $\omega_p u_* / g$:

$$\frac{\omega_p u_*}{g} \rightarrow \infty ; h = \sigma = \left[\int_0^{\infty} \phi(\omega) d\omega \right]^{1/2} \quad (4)$$

$$\frac{\omega_p u_*}{g} \rightarrow 0 ; h = \beta \frac{u_*^2}{g} \quad (5)$$

The early stages of wave development or the short fetches characteristic of laboratory wave tanks correspond to (4). This is directly analogous to rough flow over a solid wall in which roughness length and height of roughness elements are directly related. Figure 2 (from Kitaigorodskii, 1968) demonstrates this conclusively for data from two laboratory experiments. The final stages of wave development or the long fetches characteristic of open ocean conditions correspond to (5) which is, in effect, Charnock's relation. Most open ocean estimates of surface roughness are in agreement with (5); (see, for example, Garratt, 1977).

Thus, the effective roughness of an aerodynamically rough water surface may be calculated from the wind speed and the wave

spectrum using the method of Kitaigorodskii (1968) or Donelan (1982). When the flow is not rough the ratio of the roughness length Z_0 to the effective height of the roughness elements h depends on R_* (Monin and Yaglom, 1971). Thus once the ratio of Z_0 to h (3) has been determined for rough flow, Z_0 , u_* and R_* may be determined iteratively for other conditions (Fig. 28 of Monin and Yaglom, 1971).

3.0 A MIXING LENGTH MODEL

Having determined the roughness length Z_0 for a given water surface, what can be said about the gas transfer coefficient C_g or, equivalently, the virtual origin of the gas concentration profile or gas roughness length Z_g ? Most of the resistance to gas transfer occurs in the very thin viscous sub-layer near the surface. Therefore a description is required of the average diffusive properties of the turbulence near the interface. For all its faults and oversimplifying assumptions Prandtl's mixing length hypothesis seems to lead to velocity distributions (e.g., Reichardt, 1951, Van Driest, 1956), which have been verified experimentally for smooth flow. Other mixing length models (e.g., Rotta, 1962) are appropriate for rough flow. Recently Riley et al. (1982) have suggested a mixing length model which is suitable for all roughness Reynolds numbers:

$$\ell = \ell_s + \kappa Z [1 - \exp(-Zu_*/13\nu)]^2 \quad (6)$$

In which the surface value of the mixing length ℓ_s is dependent on R_* and is zero only for smooth flow. The momentum balance for the constant stress layer

$$\left(\nu + \ell^2 \frac{dU}{dZ}\right) \frac{dU}{dZ} = u_*^2 = C_d U^2 \quad (7)$$

may be integrated for various values of ℓ_s , and thus Z_0 may be determined from the fully logarithmic outer boundary layer. These

values, when coupled with Nikuradse's pipe flow measurements (Monin and Yaglom, 1971, Fig. 28), yield the dependence of the surface mixing length λ_s on the sand grain roughness height h_0 for various R_* values (Fig. 3). From zero at smooth flow λ_s increases to about 3% of h_0 in transitional flow and decreases again to a constant value of about 1.4% when the flow is fully rough. This is in accord with the view that as R_* increases the viscous sub-layer thins exposing the roughness elements until flow separation from the crests leaves the fluid in the troughs relatively undisturbed so that only about the top half of the roughnesses is exposed to the stream. The mixing length at the surface λ_s is proportional to the height of the roughness elements above the viscous sub-layer or the quiescent trough zone, whichever is the deeper.

It is interesting to note that the constant ratio between λ_s and h_0 for very rough flow is equivalent to Levich's (1962) hypothesis that the turbulent momentum diffusivity $\nu_t (= \lambda^2 \cdot dU/dy)$ is proportional to $u_* h_0$ near the surface but above the quiescent trough zone.

4.0 MIXING LENGTH FOR GAS FLUX

In a constant gas flux layer the profile of gas concentration is given by:

$$\rho(D + D_t) \frac{dG}{dZ} = -F = \rho u_* g_* \quad (8)$$

where the turbulent gas diffusivity $D_t = \lambda_g^2 \cdot dU/dZ$.

How does λ_g differ from λ ? That is, can we find, by argument and experiment, a relationship between the mixing length for momentum and that for mass transfer? In rough flow some of the surface momentum transfer is effected by the action of pressure forces on the roughness elements. Gas transfer, on the other hand, occurs entirely

by diffusion. However the intense turbulence produced by flow separation will disturb the diffusive boundary layer and, in places, it will be swept away entirely. In our mixing length model this corresponds to a non-zero surface value of the gas transfer mixing length λ_{gs} . Depending on the Schmidt number Sc , the viscous and diffusive boundary layers will have different thicknesses. Thus the effectiveness of the surface turbulence in penetrating these sub-layers will depend on Sc .

$$\lambda_{gs}/\lambda_s = m Sc^n \quad (9)$$

For identical sub-layer thickness (i.e., $Sc = 1$) λ_{gs} should be less than λ_s ($m < 1$), because pressure gradients and turbulent diffusion both contribute to $(v_t)_s$ whereas only turbulent diffusion determines $(D_t)_s$. We must also allow for the fact that in the logarithmic layer, the turbulent Schmidt number $Sc_t (= v_t/D_t)$ may be different from unity. Thus the mixing length for gas transfer is:

$$\lambda_g = m Sc^n \lambda_s + Sc_t^{-1/2} \kappa Z [1 - \exp(-Zu_*/13v)]^2 \quad (10)$$

The coefficients m and n and the turbulent Schmidt number Sc_t must be determined by experiment. The laboratory wave tank experiments of Möller and Schumann (1970) cover a suitably wide range of Schmidt number. Deacon (1977) has argued convincingly that their estimates of u_* are anomalously low; we agree, and in the following have used Deacon's corrected results of the Möller and Schumann data. From u_* and U we obtain Z_0 and R_* and thence λ_s (Riley et al. 1982). Integration of (7) with (6) and (8) with (10) yields C_g and C_d . By adjusting m and n in (9) we obtain the best fit to the data (Fig. 4) with $m = 0.54$, $n = -0.39$. The value of Sc_t used was 0.85, which was the average value given by Kader and Yaglom (1972) for the turbulent Prandtl number Pr_t from a review of 22 experiments. Kader

and Yaglom point out that Pr_t cannot be a function of Pr and must be independent of Z in the logarithmic layer. The same comments apply to the Schmidt numbers.

The data reported by Möller and Schumann cover a wide range of Sc but are limited to a single value of R_* ($= 5.3$). Liss's (1973) laboratory measurements of evaporation ($Sc = 0.6$) in a wind-water tunnel cover a wide range of R_* from nearly smooth to nearly rough $0.27 < R_* < 2.2$. (Here again the corrections to u_* , recommended by Deacon, have been used). Figure 5 compares calculated transfer velocities $V[= - F/\rho(G_z - G_s) = C_g(U_z - U_s)]$ with Liss's observations.

5.0 LARGE TANKS AND FIELD EXPERIMENTS

The experiments of Möller and Schumann and of Liss were performed in very small laboratory tanks with fetches of 3 m and 4.5 m respectively. The small and slow moving waves so generated would behave essentially like a solid wall to the air flow. It behooves us therefore to examine data in which the surface more closely resembles natural wind disturbed water surfaces. Such data may be obtained in very large tanks (e.g., Resch and Selva, 1979 and Mangarella, et al., 1971), and in field experiments (e.g., Large and Pond, 1982).

Resch and Selva (1979) present profiles of wind speed, temperature and humidity under weakly stable conditions. There are eight[#] profiles, at various fetches from 2.6 m to 37.8 m, from which values of Z_0 , Z_θ , Z_q and u_* have been abstracted. Normalized values of Z_θ and Z_q versus Z_0 are shown in Fig. 6, wherein calculated values of Z_g versus Z_0 have been graphed for various Sc . The data of Resch and Selva cover the roughness Reynolds number range corresponding to transitional flow. To supplement these we have added ten cases from Mangarella et al. (1971). Although the data are scattered the trend is in agreement with the calculations and the

The humidity profile at station 8 has not been used since it contains no logarithmic region.

values of Z_q ($Sc = 0.6$) are higher than those of Z_θ ($Pr = 0.7$). The data cover the R_* range of $0.25 < R_* < 154$.

For comparison with the recent field measurements of Large and Pond (1982), we have computed the transfer coefficients C_d and C_q versus wind speed (Fig. 7). The values of the drag coefficient C_d are derived from the smooth flow condition ($R_* = 0.137$), at low wind speeds, patched to Garratt's (1977) linear dependence from open ocean observations. The corresponding values of the evaporation coefficient C_q are derived from integrating (8) with $Sc_t = 0.85$. This suggests that, within the normal scatter (typically 20%) of field observations, the neutral coefficients of drag and evaporation will be indistinguishable, provided that Sc_t remains close to 0.85. The measurements of Large and Pond (1982) are in general agreement with this: compare their Figs. 8 and 11.

6.0 TURBULENT DIFFUSIVITIES AND STABILITY EFFECTS

In a neutral boundary layer the existence of logarithmic profiles of wind speed and a contaminant requires that the appropriate turbulent Prandtl (or Schmidt) number be independent of height (Kader and Yaglom, 1972). In the absence of a buoyancy gradient the source of all turbulent fluctuations is mechanical, so that all contaminants will have the same value of Sc_t (or Pr_t). The present best estimate of Sc_t under these conditions is 0.85, which is the value of Pr_t deduced from 22 experiments by Kader and Yaglom.

Under diabatic conditions the logarithmic layer vanishes and with it the requirement that Sc_t be height independent. This has been clearly demonstrated by Businger et al. (1971) from detailed measurements of fluxes and profiles of temperature and velocity over flat land. The turbulent Prandtl number is equal to the ratio of the non-dimensional temperature gradient ϕ_θ to the non-dimensional wind shear ϕ_u and is strongly stability dependent. Large and Pond (1982), in correcting their measured C_θ (heat transfer coefficient) values to

the neutral equivalents, assumed that $Pr_t = 1.0$. In fact, over the stability range covered by most of their data ($-0.4 < \zeta < 0.1$), Pr_t (Businger et al., 1971) varies by about the same amount as the adjusted values of C_θ shown in their Fig. 9. Had they accounted for the variation of Pr_t with ζ , the values of C_θ adjusted to neutral would have shown no pronounced stability dependence. However, since strong winds (high R_*) are generally associated with nearly neutral conditions, there will be a tendency for higher transfer coefficients to occur near $\zeta=0$.

The turbulent Prandtl number dependence shown by Businger et al. (1971) is in agreement with the behaviour deduced from a mixing length concept, in which the turbulent Prandtl number is equivalent to the square of the ratio of the momentum transfer mixing length to the heat transfer mixing length $(\lambda/\lambda_\theta)^2$. The mixing length represents the vertical distance over which a displaced fluid element can travel before losing its identity. Under neutral conditions, this length is smaller for momentum than for a scalar contaminant because momentum is transferred by pressure gradients as well as by direct mixing. This implies that Pr_t and Sc_t are less than unity under neutral conditions as observed. With increasing instability, the vertical transfers are enhanced, thereby increasing both λ and λ_θ ; but the additional transfer of momentum by pressure gradients keeps λ from increasing as quickly as λ_θ . Thus the turbulent Prandtl number decreases further. On the other hand, a buoyancy stabilized boundary layer restricts vertical transfers so that the mixing lengths are reduced. Displaced fluid tends to overshoot and move towards its original level until its buoyancy difference has been smeared out. Since momentum is transferred more rapidly while the fluid is away from its "equilibrium" position, λ decreases more slowly than λ_θ and consequently Pr_t increases with increasing stability as observed.

It is therefore clear that the stability dependence of Sc_t must be included in estimating gas transfer from water surfaces. Thus under diabatic conditions the mixing lengths λ and λ_g become:

$$\lambda = \lambda_s + \kappa Z \phi_u^{-1} [1 - \exp(-Zu_*/13\nu)]^2 \quad (11)$$

$$\lambda_g = 0.54 Sc^{-0.39} \lambda_s + Sc_t^{-1/2} \kappa Z \phi_u^{-1} [1 - \exp(-Zu_*/13\nu)]^2 \quad (12)$$

where ϕ_u and Sc_t are obtained from observations. The empirical relations of Businger et al. (1971) are based on the most comprehensive set of flux-profile observations in the atmospheric boundary layer to date. However, they reported a 33% difference between drag plate and sonic anemometer stress estimates. They used the (lower) sonic anemometer results in their analysis. Wieringa (1980) has argued that flow distortion around the anemometer would cause the stress to be underestimated. Wyngaard et al. (1982) have attempted to refute Wieringa's argument, but Wieringa (1982) remains undaunted. Whatever the reason, the fact remains that the anomalous results of Businger et al. ($\kappa = 0.35$, $Pr_t = 0.74$ and the drag plate-sonic anemometer difference) are brought into line with laboratory results ($\kappa = 0.4$, $Pr_t = 0.85$) if we take it that the friction velocity and heat flux were underestimated by Businger et al. by almost 14%. Appropriate corrections to their empirical relationships yield:

$$\phi_u = \begin{cases} (1 - 17\zeta)^{-1/4} & ; \zeta < 0 \\ 1 + 5.4\zeta & ; \zeta > 0 \end{cases} \quad (13)$$

$$Sc_t = Pr_t = \frac{\phi_\theta}{\phi_u} = \begin{cases} \frac{0.85 (1 - 17\zeta)^{1/4}}{(1 - 10\zeta)^{1/2}} & ; \zeta < 0 \\ \frac{0.85 + 6.2\zeta}{1 + 5.4\zeta} & ; \zeta > 0 \end{cases} \quad (14)$$

7.0 WAVE EFFECTS

The fundamental difference between atmospheric boundary layers over land and water derives from the scale and the mobility of

the surface roughnesses. We have already discussed the pronounced effect on the roughness caused by the speeds of the various wave components. The scale of the roughnesses varies from sizes on the order of the depth of the viscous sub-layer to the depth of the entire constant stress layer. Turbulence in the interior of the boundary layer is generated by shear production and buoyancy; while, near the surface, flow separation from the steep (generally small) roughnesses enhances the surface turbulence. Flow over the less steep (generally larger) waves may be too organized to contribute much to the diffusive effects of the turbulence. These wave-coherent disturbances may support some of the stress but not contribute to the eddy viscosity.

In the context of Miles' (1957, 1959) theory of wave generation by wind, Stewart (1961, 1967) has discussed some effects of wave coherent disturbances on the boundary layer over water. In particular, he pointed out (Stewart, 1961) that if, near the surface, the wave-coherent disturbances carry a significant portion of the stress, then the turbulent stress will increase with height. In this theory the wave-coherent stress vanishes above the "critical height" of the longest waves. Therefore, stress estimates from profile measurements will increase with height away from the surface, as more and more of the stress is carried by the turbulence. Furthermore, estimates of heat and mass transfer, based on stress measurements above the critical height, will be high since near the surface the turbulence is less intense than would be inferred from the measured stress.

Under conditions of moderate wave amplification, when the wave-coherent disturbances might be significant, the critical height is typically of the order of 1 cm. So that it seems unlikely that the highly idealized theory of Miles would be a suitable description under these conditions and, in any case, field observations of the effect discussed by Stewart are well nigh impossible. However recent observations, under strong wave forcing (both generation and attenuation) in the large laboratory tank at the Canada Centre for Inland Waters, show wave-coherent fluctuations far above the critical

height. Most of these measurements of velocity components and surface elevation (u' , w' and η) are yet to be analysed, but the results available to date are sufficient to provide an estimate of the importance of wave-coherent fluctuations in estimating gas transfer.

The amplitudes of the wave-coherent parts of u' and w' are proportional to wave slope and relative wind speed and they decay exponentially with height. The phase between them is roughly 110° . The wave-coherent stress τ_w is thus:

$$\tau_w \approx \rho \int_0^\infty (U_z - g/\omega)^2 \exp(-2\omega^2 Z/g) \left(\frac{\omega^2}{g}\right)^2 \phi(\omega) d\omega \sin(20^\circ) \quad (15)$$

The wave-coherent stress decreases with height and therefore the velocity profile will become steeper relative to its logarithmic form, while the heat and mass transfer profiles will become less steep. Observations of wind speed and temperature profiles over Lake Ontario (Donelan et al. 1974) exhibit this behaviour as, to a lesser extent, do the Arabian Sea observations of Badgley et al. (1972).

In order to investigate the effects of the wave-coherent stress on gas transfer, (15) was used to calculate C_d and C_g for various stages of wave development. In general, the ratio C_g/C_d decreases as τ_w increases. Under typical field conditions the effects are small (about 10%); however in the early stages of wave development or with waves advancing against the wind, much reduced C_g/C_d values are to be expected. Large and Pond (1982) report a significant increase in C_d with little change in C_g during an episode in which the wind turned rapidly to face the waves it had created. Kitaigorodskii and Volkov (1965) have graphed C_g/C_d and C_d/C_d versus R_* from four different sets of profile observations (their Fig. 2). Apart from the short fetch observations of Takahashi (1958), the ratio of transfer coefficients is close to and a little above unity, in agreement with the calculations above leading to

Fig. 7. The wave effects at short fetch in Takahashi's data would tend to reduce the ratio and increase its variability.

It is clear that precise parameterization of gas transfer depends on knowing not only the effective surface roughness and thermal stability, but also the way in which energy and momentum are transferred through the boundary layer to the wavy wall. There still does not exist a wholly satisfactory theory of wind-wave interaction supported by experimental data, and it would appear that this is a necessary building block in parameterizing gas transfer above natural water surfaces.

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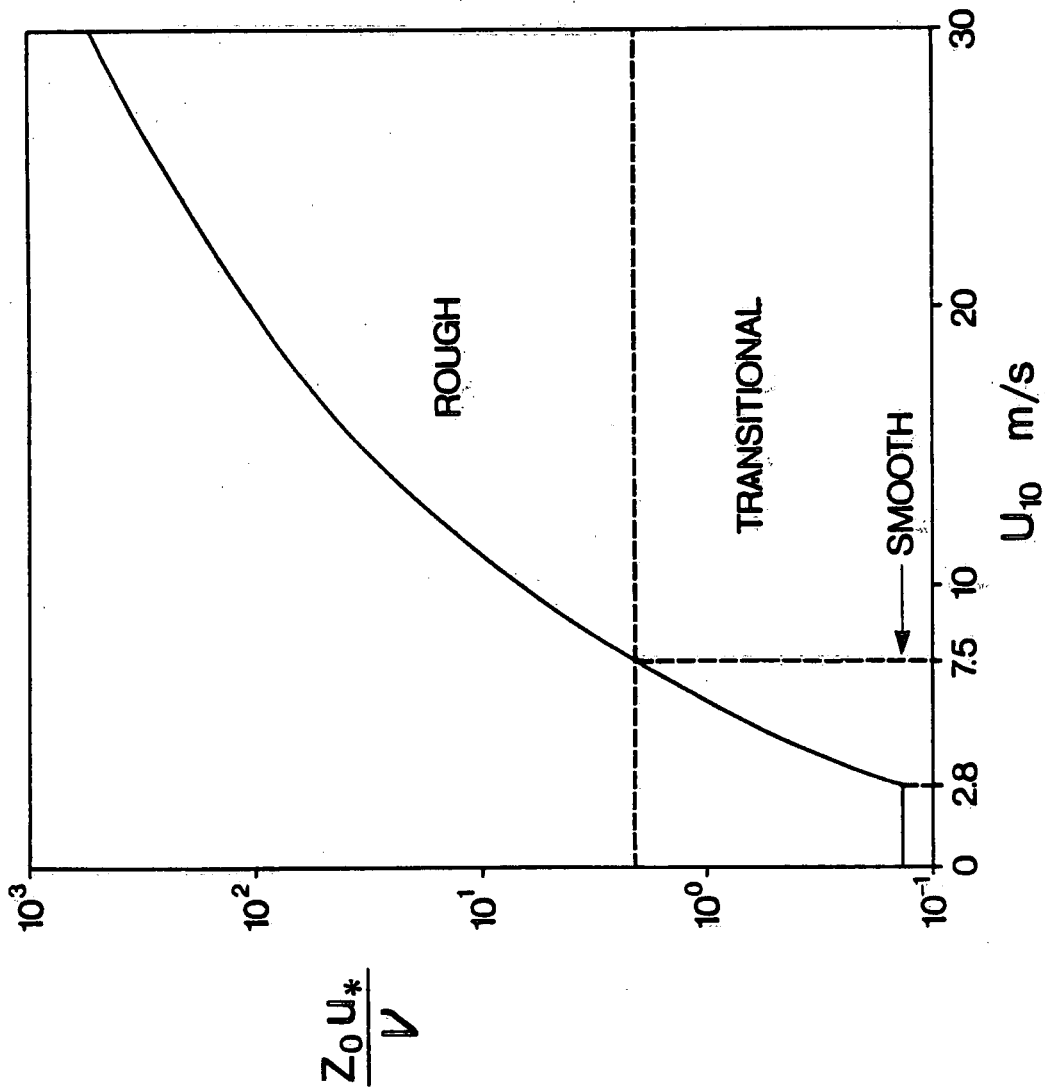
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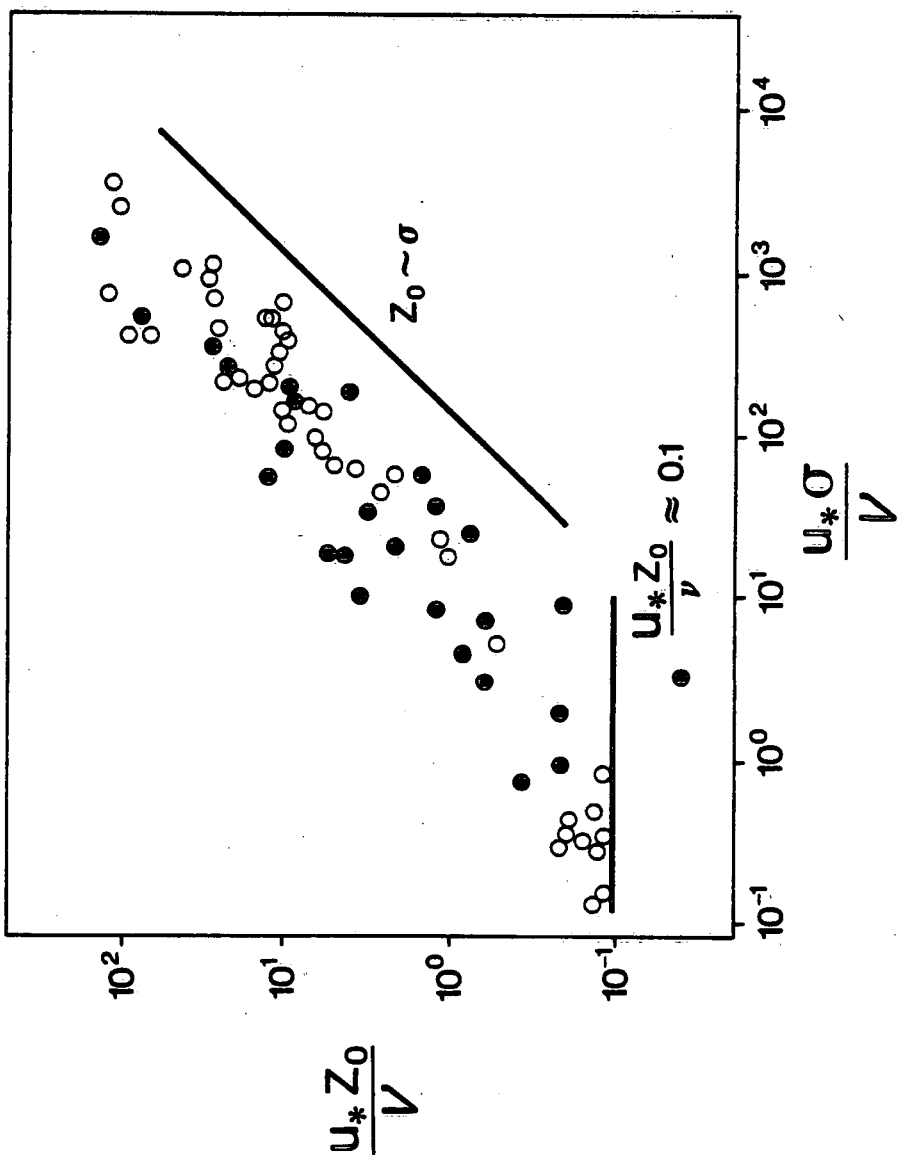
FIGURE CAPTIONS

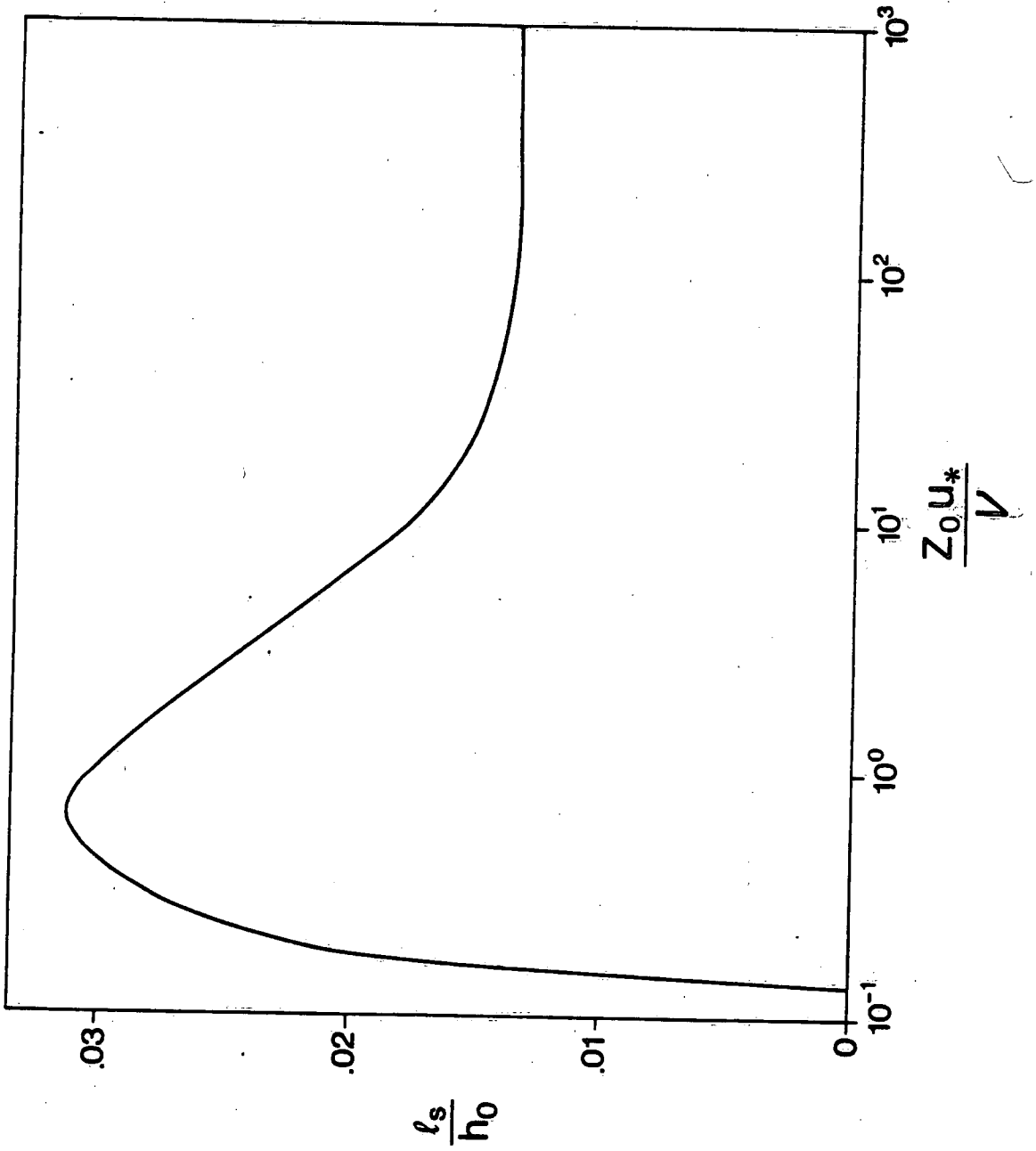
1. Roughness Reynolds number versus mean wind speed at 10 m. The values of $R_* = Z_0 u_* / \nu$ are derived from Garratt's (1977) empirical relation between drag coefficient and wind speed.
2. Roughness Reynolds number versus normalized rms surface elevation from two different laboratory experiments (solid and open circles). The figure has been redrawn from Kitaigorodskii (1968).
3. The ratio of surface mixing length λ_s to height of sand grain roughness h_0 versus roughness Reynolds number.
4. Normalized transfer velocity versus Schmidt number. The seven data points are taken from the laboratory experiments of Möller and Schumann (1970). The curve is derived from the mixing length model presented in this paper.
5. Comparison of model calculations with the laboratory observations of Liss (1973) of the transfer velocity of water vapour. The vertical lines extend over the model results for no surface drift velocity U_s (top) to $U_s = 0.03 U_z$.
6. Normalized gas roughness length versus roughness Reynolds number for various molecular Schmidt numbers. The curves are from the mixing length model in which the turbulent Schmidt number is set to 0.85. The data points are from the profiles of Resch and Selva (1979) for temperature (solid triangles) and humidity (solid circles) and of Mangarella et al. (1971) (open symbols).

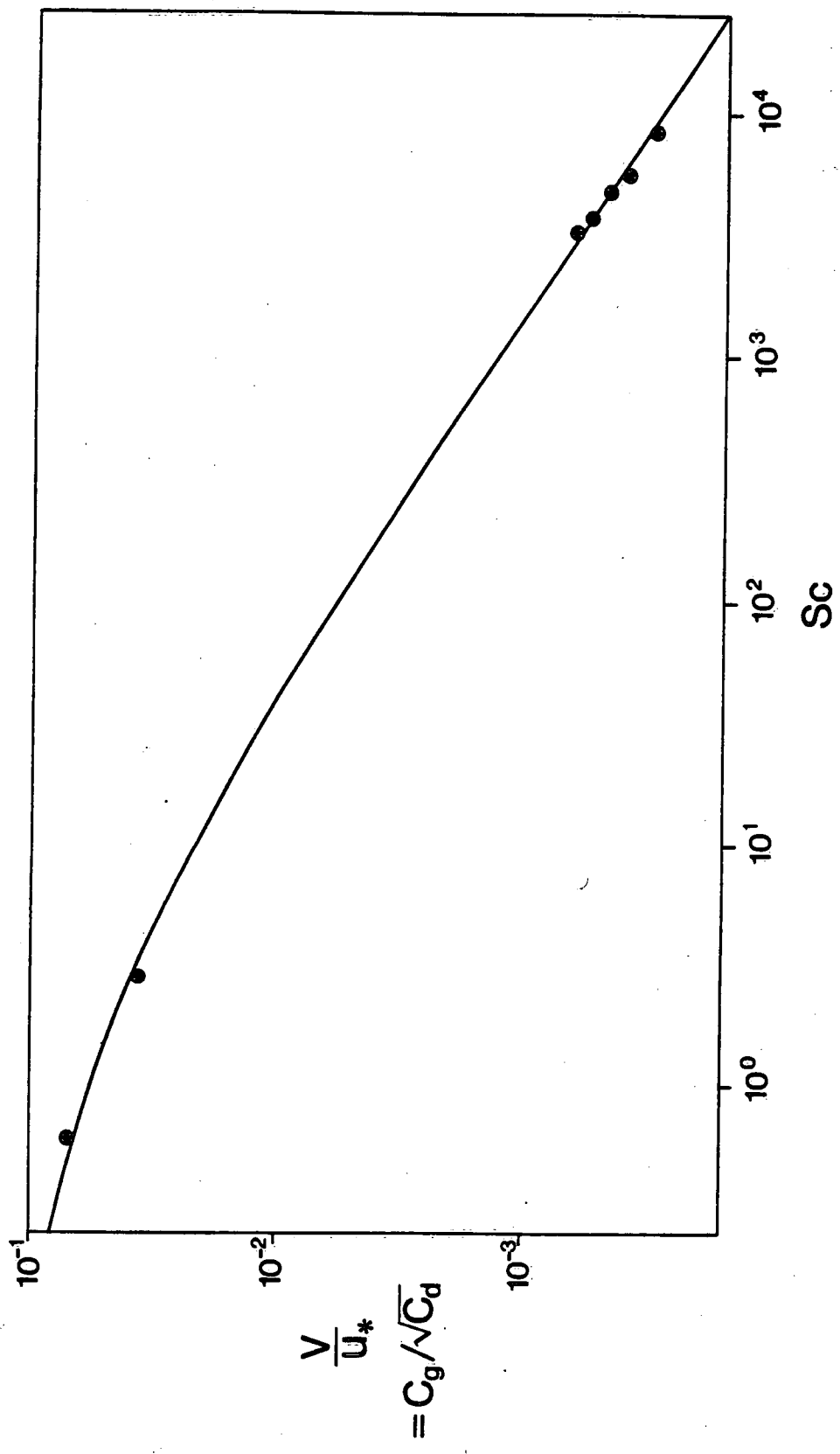
7. A comparison of moisture C_q and drag C_d transfer coefficients for typical open ocean conditions. The smooth flow values of C_d below wind speeds of 2.8 m/sec are patched to the empirical relationship of Garratt (1977). The C_q values are computed from the mixing length model.



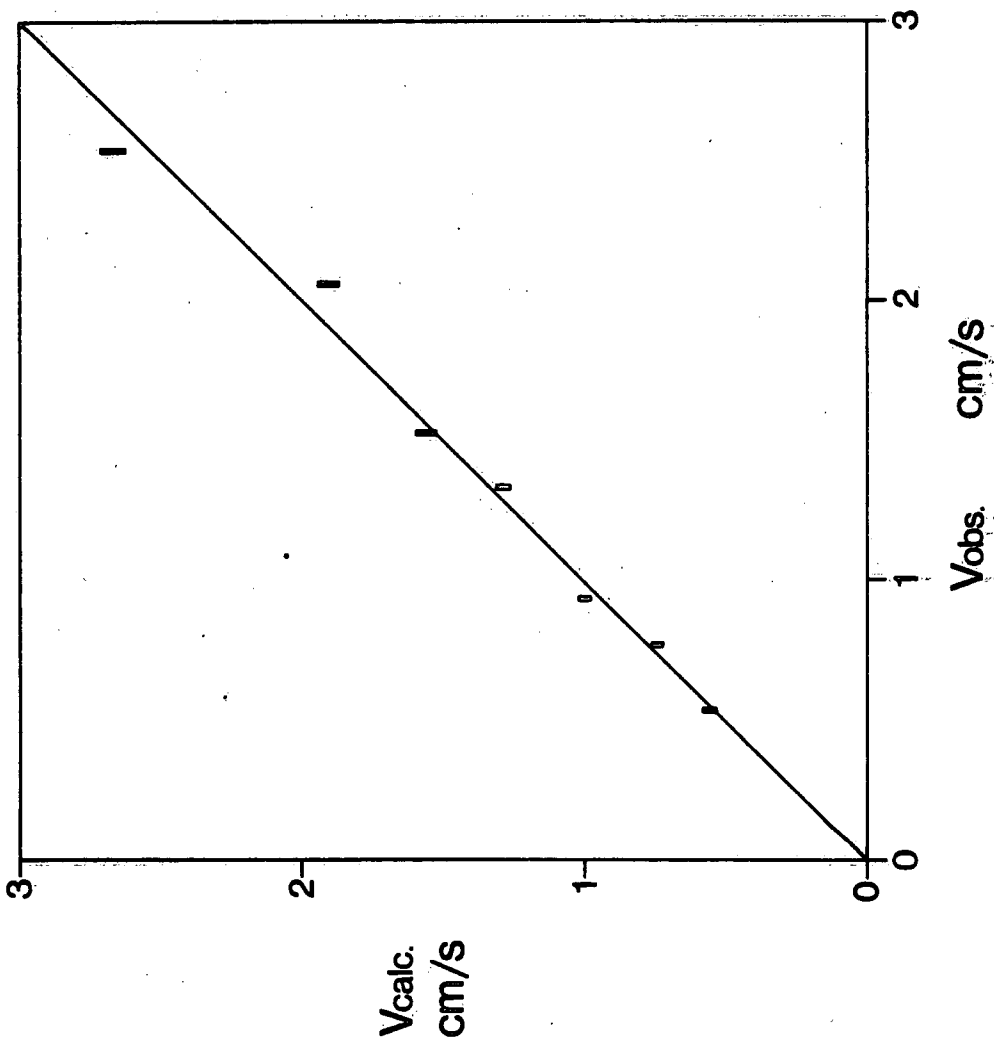
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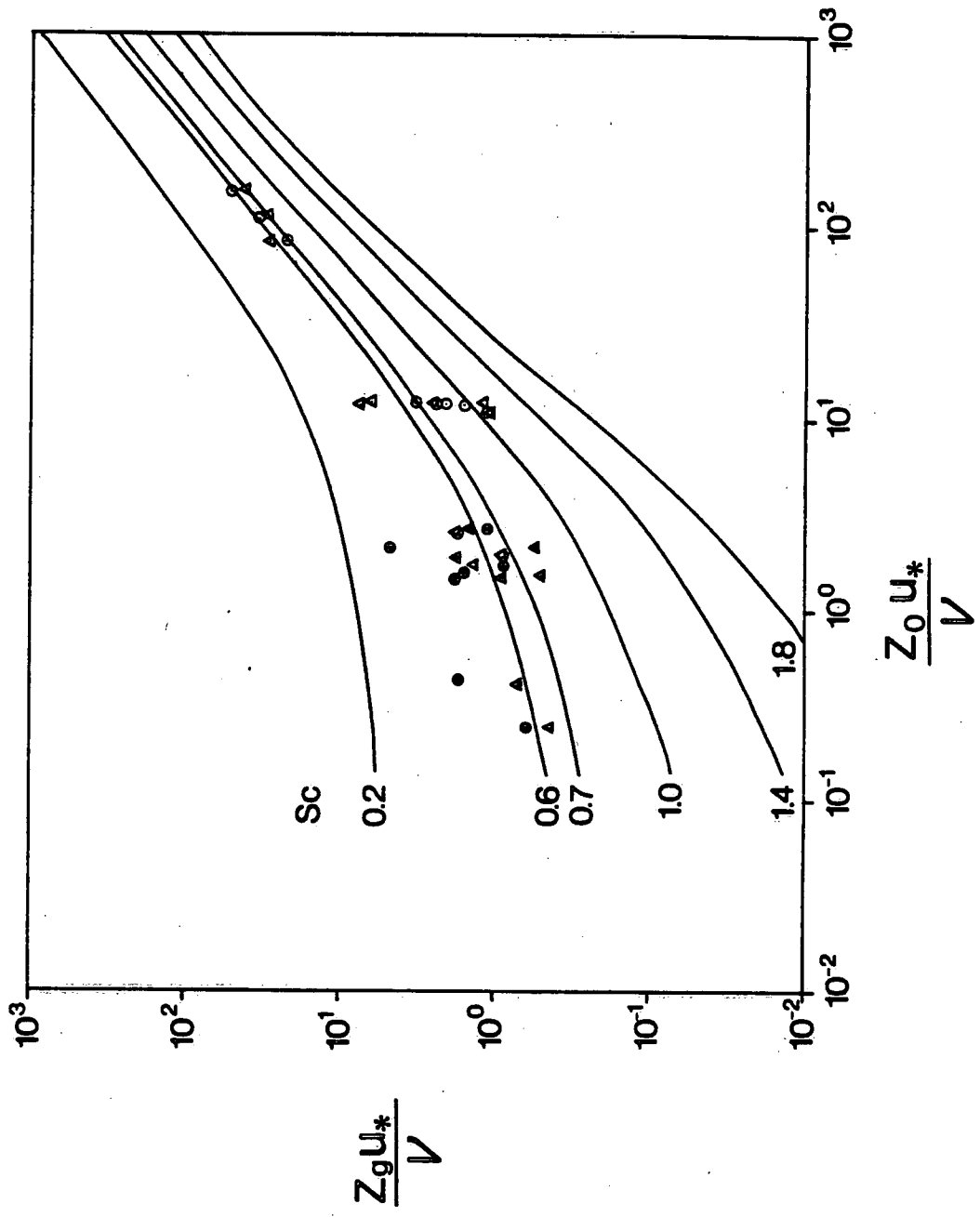






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